Institut für Tierwissenschaften

Assessment of different innovative packaging strategies to maintain the quality of fresh produce

Dissertation

zur Erlangung des Grades

Doktorin der Ingenieurwissenschaften

(Dr.-Ing.)

der Landwirtschaftlichen Fakultät

der Rheinischen Friedrich-Wilhelms-Universität Bonn

von

Imke Marie Korte

aus

Minden

Bonn 2023

Referentin: Prof. Dr. Judith Kreyenschmidt

Korreferent: Prof. Dr. Ralf Pude

Fachnahes Mitglied: Prof. Dr. Margit Schulze

Tag der mündlichen Prüfung: 13. Juli 2023

Angefertigt mit Genehmigung der Landwirtschaftlichen Fakultät
der Rheinischen Friedrich-Wilhelms-Universität Bonn

Meiner Familie

Wir müssen Ausdauer und vor allem Vertrauen in uns selbst haben. Wir müssen glauben, dass wir begabt sind und dass wir etwas erreichen können.

Marie Skłodowska Curie (1867-1934)
Physikerin und Chemikerin

Abstract

Assessment of different innovative packaging strategies to maintain the quality of fresh produce

The objective of this thesis was to assess the influence of different innovative packaging strategies on typical quality parameters of fresh produce and thus to keep and even improve the resource efficiency in food supply chains. First, the status quo and current practical use of bio-based polymers were analyzed; in terms of fundamental packaging functions and industrial applicability. Additionally, plant extracts as active packaging agents (capable of prolonging the food shelf life) were considered, and the adoption potential of packaging based on polymers from renewable resources was discussed from a bioeconomic perspective. The practical trials comprised studies to analyze the impact of innovative packaging strategies on quality changes of fresh cherry tomatoes as well as emulsion-type sausage. The use of both bio-based materials and fossil-based materials (multi-layers with reduced material thickness and a recyclable mono-layer) was assessed. Next, product-specific quality parameters and general packaging characteristics were investigated. Furthermore, the antimicrobial activity of coatings under development, containing a mineral binder based on liquid potassium silicate, non-conserved acrylate polymers, and non-conserved styrene-acrylate polymers, was analyzed.

Literature research showed that numerous (bio-based) materials are under development that provide significant opportunities to current fossil-based packaging in terms of sustainability and biocompatibility. Nevertheless, their use in industrial applications is often restricted due to various factors, such as poor commercial availability, insufficient material properties, high costs, and lower performance in fundamental packaging functions, which negatively impact shelf life, and thus the amount of food waste. The conducted product studies with tomatoes showed that the moisture absorption of the materials in particular had an influence on quality changes. Groundwood pulp and sugar cane showed promising preserving characteristics of fresh cherry tomatoes and pointed to the advantages of using this kind of bio-based packaging material, even from an economical and sustainable perspective. The product studies with emulsion-type sausage showed that sufficient barrier properties of both under and upper foil are crucial in keeping product quality during storage. So far, the practical implementation of fossil-based multi-layers with reduced material thickness and recyclable mono-layers is still challenging and remains highly case-dependent and may represent a promising alternative for replacing conventionally used multi-layer packaging. The analyzed organic coatings showed high antimicrobial activity against several microorganisms. These findings underline the potential of these coatings as a new antimicrobial material for different applications, such as antimicrobial packaging and antimicrobial food contact surfaces, to increase the safety and quality of perishable products and reduce waste.

Kurzfassung

Bewertung verschiedener innovativer Verpackungsstrategien zur Aufrechterhaltung der Qualität von Frischprodukten

Ziel der vorliegenden Arbeit war es, den Einfluss verschiedener innovativer Verpackungsstrategien auf die Qualität von Frischprodukten zu bewerten und damit die Ressourceneffizienz in Lebensmittel Supply Chains zu verbessern. Zunächst wurde der Status quo zu Verpackungsmaterialien aus biobasierten Polymeren und deren aktuelle industrielle Nutzung im Hinblick auf grundlegende Verpackungsfunktionen analysiert. Zudem wurden Pflanzenextrakte als aktive Additive von Verpackungen, die die Haltbarkeit von Lebensmitteln verlängern können, betrachtet und das Adaptionspotenzial von Verpackungen auf Basis von Polymeren aus nachwachsenden Rohstoffen aus bioökonomischer Sicht diskutiert. In (Produkt-)Studien wurde der Einfluss innovativer Verpackungsmaterialien auf Qualitätsveränderungen von frischen Kirschtomaten sowie Brühwurst untersucht. Dabei wurde der Einsatz von biobasierten Verpackungsmaterialien und fossilbasierten Multilayern mit reduzierter Materialstärke und recyclefähigen Monofolien bewertet. Neben produktspezifischen Qualitätsparametern wurden allgemeine Verpackungseigenschaften bestimmt. Des Weiteren wurden sich in der Entwicklung befindliche antimikrobiell wirksame Beschichtungen auf Basis einer Wasserglas-Mischung als mineralischem Bindemittel, nicht konservierte Acrylate und Styrol-Acrylate mit funktionalem mineralischem Füllstoff untersucht.

Aus der Literaturrecherche ging hervor, dass zahlreiche (biobasierte) Materialien in der Entwicklung sind, die derzeitigen fossilbasierten Verpackungen in Punkto Nachhaltigkeit und Biokompatibilität überlegen sind. Dennoch ist ihre industrielle Anwendung bisher unter anderem aufgrund mangelnder Verfügbarkeit, nicht ausreichender Werkstoffeigenschaften, hoher Kosten und verminderter Barriereeigenschaften, die sich negativ auf die Haltbarkeit und damit auf die Menge an Lebensmittelverlusten auswirken, oft limitiert. In den durchgeführten Produktstudien mit Tomaten zeigte sich, dass insbesondere die Feuchtigkeitsaufnahme der Materialien einen Einfluss auf die Qualitätsveränderungen darstellte. Die Verwendung von Holzschliff und Zuckerrohr führte zu einem verzögerten Qualitätsverlust und wies zudem Vorteile in Wirtschaftlichkeit und Nachhaltigkeit auf. Aus den Produktstudien mit Brühwurst ging hervor, dass ausreichende Barriereeigenschaften sowohl der Unter- als auch Oberfolie entscheidend zum Erhalt der Produktqualität sind. Bisher ist der praktische Einsatz von fossilbasierten Multilayern mit reduzierter Materialstärke und recycelfähigen Monofolien noch herausfordernd und bedarf weiterer Entwicklungen. Abhängig vom Einsatzzweck stellen die genannten Materialien eine vielversprechende Alternative zum Einsatz herkömmlicher Verpackungsmaterialien dar. Die untersuchten organischen Beschichtungen zeigten eine hohe antimikrobielle Aktivität gegen verschiedene Mikroorganismen. Diese Ergebnisse unterstreichen das Potenzial der Beschichtungen für antimikrobielle Verpackungen und Oberflächen, um die Sicherheit und Qualität verderblicher Produkte zu erhöhen und Ausschüsse zu reduzieren.

Contents

1 Introduction

1.1 Food waste and spoilage process of fresh produce

Food waste and loss describe a major concern in the food supply chain that takes all involved stakeholders into consideration (Bhat & Jõudu, 2019). There is an increasing concern about the amount of food waste in Europe (United Nations, 2015). The United Nations set a target of halving the actual amount of global food waste per capita at retail and consumer levels and reducing food losses along production and food supply chains as part of their sustainable development goals for 2030 (goal 12.3 of the UN General Assembly) (United Nations, 2015). Wasted food means that not only the products themselves are lost; it has a significant impact on the use of natural resources along the entire supply chain and the environment (Rossaint & Kreyenschmidt, 2014; Scherhaufer et al., 2018; United Nations, 2015). If food waste occurs, the overall negative environmental impact rises with every step in the supply chain due to more used resources (Heller et al., 2019; Matthews et al., 2021; Scherhaufer et al., 2018). Fresh produce products with a relatively short shelf life, like fresh meat, tend to be the most wasteful products (Mena et al., 2011; Rossaint & Kreyenschmidt, 2014). Among different food groups, fruits and vegetables are identified as the food groups that generate the highest overall amount of food loss and waste (Caldeira et al., 2019). Wasting of fresh produce is often caused by wrong handling, leading to product spoilage before the best-before date or sell-by date is reached, and short selling times, which cause throwing away not sold products (Kreyenschmidt et al., 2013; Mena et al., 2014). The food waste of meat at about 23 % is relatively low compared to other products such as fruit (41 %), vegetables (46 %), and fish (51 %) (Caldeira et al., 2019), but the environmental impact is considerably higher for meat than for plant-based food due to higher environmental burdens along the supply chain (Cooreman-Algoed et al., 2022; Molina-Besch et al., 2019; Petrovic et al., 2015).

The spoilage of fresh meat and meat products is mainly caused by microbial growth and metabolism; and thereby determines the short shelf life (Czerwiński et al., 2021; Dave & Ghaly, 2011; Gill, 1983; Huis in't Veld, 1996). As meat and meat products are characterized by a matrix of high concentrations of proteins, moisture, and fats, a combined effect of microbial and endogenous enzymes (proteases and lipases) occurs, causing food deterioration and promoting lipid and protein oxidation (Bekhit et al., 2021; Comi, 2016). Microbial activity results in major deteriorative changes, which are perceived organoleptically by the consumer in odor changes, the release of metabolites, and the formation of slime on the surface of meat and meat products. Furthermore, biochemical processes become obvious by lipid oxidation and color changes, whereas autolytic enzymatic mechanisms change the appearance of meat (products) (Dave & Ghaly, 2011; Huis in't Veld, 1996). The length of shelf life is next to product and process factors, mostly influenced by environmental factors that are referred to storage conditions such as temperature, light, moisture, gas atmosphere, and packaging of meat and meat products (Kreyenschmidt & Ibald, 2012).

In fresh plant-based products, a large number of natural ripening and aging processes lead to changes even after harvesting, which influence the composition of the ingredients as well as sensory properties (Ščetar et al., 2010). Therefore, fruits and vegetables can often only be kept for a few days and belong to the group of easily perishable products (Dannehl et al., 2008). The quality or freshness loss of fruits and vegetables is caused by biochemical and physical factors or by the growth of molds (Ahvenainen, 1996; Barry-Ryan & O'Beirne, 1998; Krämer & Prange, 2017). Depending on the type, fruits and vegetables have numerous natural protective factors that protect them from microbial spoilage for a certain period of time (Krämer & Prange, 2017). Biochemical changes are primarily caused by respiration and ripening processes, while transpiration in the fruit tissue leads to dehydration (water loss) and rapid aging after harvest depending on external factors like temperature, humidity, and packaging conditions (Bertin, 2018; Buendía−Moreno et al., 2020; Chakraverty & Singh, 2016; Kalamaki et al., 2012; Opara et al., 2012). Transpiration is one of the most important factors reducing the physiological and economic value of fruits and vegetables (Fischer & Glomb, 2015; Robertson, 2012). The water loss is responsible for most of the wilting and shriveling, weight loss, and negative texture changes of fruits and vegetables. The softening during tomato storage is caused by enzymatic degradation of cell walls and results in product water losses as well as observable sensory changes of the food (Bertin, 2018; Buendía−Moreno et al., 2020; Chakraverty & Singh, 2016; Kalamaki et al., 2012; Opara et al., 2012). As the water content decreases, the products become soft and flabby due to a reduction in hydrostatic tissue pressure and thus lose their bite and juiciness (Kader & Barrett, 2005; Robertson, 2012).

In order to reduce food loss and waste, the selection of a suitable and appropriate packaging solution is a key factor related to food safety and the high-quality properties of fresh produce (Dannehl et al., 2008; Korte et al., 2021; Wikström et al., 2019). The insurance of high quality, safety, and a shelf life along the entire supply chain can be achieved by slowing down microbial growth, reducing spoilage reactions like respiration rates and enzymatic browning, as well as preventing the products from transport damages (Corrado et al., 2017; Marsh & Bugusu, 2007; Opara & Mditshwa, 2013). Depending on packaging structure and material, the ability for adhesion and persistence of microorganisms on surfaces can affect contamination as well as the shelf life (Patrignani et al., 2016). Thus, an appropriate packaging ensures a safe and extended shelf life of foods along the supply chain as well as prevents possible waste of the food product prior to final consumption (Accorsi, 2019; Aggarwal & Langowski, 2020; Coelho et al., 2020; Kreyenschmidt et al., 2013; Sharma & Ghoshal, 2018; Wikström et al., 2019; Youssef & El-Sayed, 2018).

When considering fruits and vegetables food loss contributes most to the total emissions of the supply chain, followed by the packaging, while transportation and farm production account for a small proportion (Qin & Horvath, 2022). Thus, the environmental impact for a variety of food products (especially energy consumption during agriculture, processing, logistics, use, end of life) is much higher than that of the packaging (Heller et al., 2019; Matthews et al., 2021; Scherhaufer et al., 2018). Kan & Miller (2022) stated that the environmental impact of plastic packaging accounts for less than 10 %, whereas more than 75 % are attributed to the food item itself.

Reducing packaging is important, but in terms of sustainability, it must fulfill its duty of protection. Therefore, it is critical to find a balance between the environmental impact of the package itself, on the one hand, and the impact originating from the potential loss of the packaged product, on the other. Otherwise, the supply chain will be less sustainable overall (Heller et al., 2019; Pauer et al., 2020; Scherhaufer et al., 2018; Verghese et al., 2015; Williams & Wikström, 2011). Significant factors that need to be considered when choosing a packaging solution are appearance, color, lipid stability, nutritive value, and palatability (texture, flavor, aroma) (McMillin, 2017).

1.2 The role of packaging related to product quality and sustainability

The use of food packaging is one of the most important ways to maintain product quality and guarantee its shelf life. Regarding product quality and shelf life, the material, with its specific characteristics and barrier properties, is of crucial importance. Depending on the product and the primary function of the packaging, there are numerous requirements for the packaging materials. A distinction between general packaging functions and the requirements that the different actors along the supply chain have can be made (Figure 1.1). The main functions of food packaging are containment, protection and preservation, communication, marketing, and convenience (with a special focus on protection) (Aggarwal & Langowski, 2020; Coles, 2003; Sharma & Ghoshal, 2018; Youssef & El-Sayed, 2018). These include the pure protection of the product from external factors as well as from shocks and vibrations during transport (ensuring damage-free transportation) and the protection in form of maintaining the product quality and extending the shelf life of foods (Coles, 2003; Pathare & Opara, 2014). The requirements can go beyond the protective function and include points such as legislation, process capability, cost-effectiveness, sustainability, and logistics referring to product, distribution, consumer, and market needs and wants (Coles, 2003).

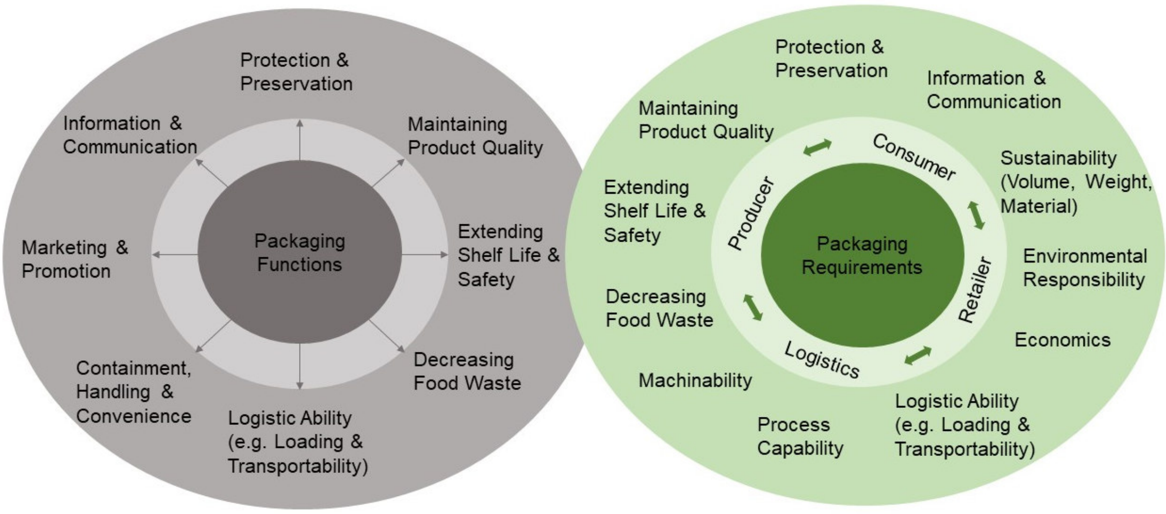


Figure 1.1 Functions and requirements of food packaging in general. Based on Aggarwal & Langowski (2020); Coles (2003); Sharma & Ghoshal (2018); Youssef & El-Sayed (2018).

Up to now, various fossil-based polymers applied in complex multi-layer packaging are mainly used in the food sectors. As these offer effective and well-optimized solutions to maintain food safety and quality, achieve optimal and prolonged shelf life, and minimize food waste (Accorsi, 2019; Aggarwal & Langowski, 2020; Coelho et al., 2020; Kreyenschmidt et al., 2013; Matthews et al., 2021; Sharma & Ghoshal, 2018). Polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyethylene terephthalate (PET) are commonly used plastics among fossil-based polymers (Robertson, 2016). These polymers meet the requirements of many foods attributed to: mechanical (impact and tear strength, stiffness, flexibility), thermal and physical properties; especially certain barrier functions like gas, water vapor permeability, and aroma transfer (Table 1.1); that can be easily adjusted to a broad variety of different food types to make their use superior to many bio-based polymers (Barlow & Morgan, 2013; Matthews et al., 2021; Sharma & Ghoshal, 2018). Furthermore, these polymers can be easily manufactured into different packaging shapes. Especially for perishable products like meat and meat products the barrier properties of fossil-based plastic packaging are advantageous for maintaining product quality and safety (Lee et al., 2008).

As high barrier layers, ethylene vinyl alcohol (EVOH) copolymers are widely used in multi-layer packaging materials. These exhibit a very low permeability of gases (like O_2 and CO_2) and organic vapors (Gavara et al., 2016; López-Rubio, 2011). Especially the barrier against oxygen is a key factor for food quality and safety; notably in modified atmosphere packaging (MAP) (Barlow & Morgan, 2013); by slowing down food respiration rates, reducing microbial growth, and inhibiting enzymatic spoilage and/or autolysis (McMillin, 2017; Pellissery et al., 2020). Therefore, MAP materials typically consist of multiple joined material layers comprising low-density polyethylene (PE-LD), linear low-density polyethylene (PE-LLD), polyamide (PA), PP, EVOH, and PET (Barukčić et al., 2020; Kargwal et al., 2020; Seier et al., 2022; Walker et al., 2020). Furthermore, fossil-based plastics are favored because of their lightweight nature and low costs (Ellen MacArthur Foundation and McKinsey & Company, 2016; Matthews et al., 2021).

In addition to plastics, paper, solid and corrugated cardboard are often used as materials in the packaging sector. Especially for fruits and vegetables, (corrugated) cardboard is a preferred packaging material due to its good protective function and good mechanical properties (high strength) (Abejón et al., 2020; Pathare & Opara, 2014). Major material properties of the main packaging materials plastic, and paper and cardboard are listed in (Table 1.1). In packaging applications, these are often used in combination with each other in order to best exploit their functional properties (Coles, 2003).

Table 1.1 Major material properties of plastics, and paper and cardboard (Coles, 2003).

Plastics	Paper & Cardboard
• Low-density materials with a wide range of physical and optical properties • Wide range of barrier properties • Permeable to gases and vapors to varying degrees • Usually have low stiffness • Tensile and tear strengths are variable • Can be transparent • Functional over a wide range of temperatures depending on plastic type • Flexible and can be creased in certain cases	• Low-density materials • Poor barriers to light, liquids, gases, and vapors (without coating, lamination or wrapping)) • Good stiffness • Can be grease resistant • Absorbent to liquids and moisture vapor • Can be creased, folded, and glued • Tear easily • Not brittle (but not so high in tensile as metal) • Excellent substrates for inexpensive printing

Currently, European legislation and regulations are forcing companies to eliminate or reduce conventional plastic in packaging materials (COM/2018/028 final) (Matthews et al., 2021). Safe disposal and recycling of materials often remain challenging. The reasons are poor management and enforcement, regulatory disparities, lack of infrastructure, and high costs of waste recycling systems (Taleb & Al Farooque, 2021). Moreover, the multi-layer structure makes plastic waste one of the most complex material mixtures from a recycling perspective (Ragaert et al., 2017). Recycling of these materials is accompanied by either high costs, technical difficulties regarding the separation process of the different polymers, or the inability to recycle mixed polymers (Dilkes-Hoffman et al., 2018; Matthews et al., 2021).

To deal with the issue of the adverse impact of fossil-based plastics on human health and the environment, there are different strategies for developing more sustainable packaging which still offer the necessary moisture or oxygen barriers to preserve the product quality and to reduce the environmental impact of the packaging itself. These include reducing packaging materials by decreasing the thickness, reducing the number of layers, using easily recyclable materials and applying bio-based and/or biodegradable materials (Pro Carton, 2010; Soro et al., 2021; Teck Kim et al., 2014). Reducing the environmental impact of the material can be best achieved by minimization of used materials (thinner layers) that retain mechanical and barrier properties rather than emphasizing end-of-life issues (such as recycling or disposal) (Barlow & Morgan, 2013). Furthermore, material properties of simple mono-layer materials can be improved by design of the packaging itself or using active packaging solutions (Schumann & Schmid, 2018; Soro et al., 2021; Yildirim et al., 2018).

Next to the production of bio-based plastics, the production of packaging materials from renewable raw materials has also gained attention; this is due to the fact, that wood can be counted among the renewable raw materials compared to fossil-based raw materials but compared to other renewable raw materials such as corn and hemp, it is a raw material that only grows slowly. Due to the heavy burden on forests caused by increasing deforestation, the production of paper and cardboard from rapidly renewable raw materials is becoming increasingly interesting (Bajpai, 2012). Renewable raw materials such as bamboo, corn, hemp, or sugar cane now play an important role in replacing wood-based products. They are used as fibrous materials in papermaking and as composite materials. The fiber properties, such as thickness, length, and density, which guarantee the later stability of the paper or cardboard, play an important role in the suitability for use as a packaging material. For this reason, fiber-rich plants such as miscanthus or bamboo are considered as promising, universally applicable fibers (Dungani et al., 2014).

Due to a growing world population, food production is also increasing, which leads to an increase in the production of agricultural residues. Agricultural residues are produced as a by-product of biomass-rich plants (Dungani et al., 2014) and represent the biomass that occurs during crop production and is not later consumed or processed. This includes cereal straw and husks, soybean, corn or potato stubble, or banana leaves in tropical growing regions. Some of the residues remain on the field to enrich the soil with nutrients, and a large part has to be removed from the field and cannot be used for energy (Hakeem, 2014). The use of agricultural residues as fibers for packaging is therefore an approach to completing the biomass cycle of agricultural products. The main advantage of natural fibers is their very light and biodegradable nature. The low weight reduces transport costs. The main problem with fibers is that they can absorb a lot of moisture. However, a primary function of most food packaging is to keep moisture away from the packaged product. The fibers then usually have to be wetted with chemical additives in order to adjust their surface properties (Verma et al., 2012).

Depending on the nature of the product, product deterioration mechanisms, including chemical breakdown, biochemical changes, and microbiological spoilage process, as well as product shelf life requirements (Coles, 2003), there are different packaging requirements, especially for the barrier properties of the packaging material. Therefore, materials characterized by a low gas and water vapor permeability are required to prolong the shelf life of meat and meat products, as the exclusion of oxygen migration in both directions through the packaging is important (Barlow & Morgan, 2013; Lee et al., 2008; Matthews et al., 2021; Sharma & Ghoshal, 2018). Oxygen migration and water infiltration into the packaging leads to an undesirable change in color and taste and, in the worst case, to increased growth of aerobic, pathogenic microorganisms. Likewise, the loss of water vapor through the packaging can lead to undesirable drying out and a consequent change in the textural properties (Detzel et al., 2018; Lee et al., 2008). In MAP, often used for meat and meat products, the gaseous environment within the packaging headspace is replaced with a desired, specific gaseous atmosphere to reduce microbial growth and inhibit enzymatic spoilage and/or autolysis as well as physical and chemical degradation extending the shelf life by up to several weeks or even months (Herbert et al., 2013; Herbert & Kreyenschmidt, 2015; McMillin, 2017; Opara et al., 2019; Pellissery et al., 2020; Seier et al., 2022; Torrieri, 2015).

In contrast to these requirements for meat and meat products, in the packaging of fruits and vegetables, gas exchange, and therefore a certain permeability to water vapor and O_2 is desirable to maintain the product quality (Detzel et al., 2018; Lee et al., 2008). This is due to the characteristic of a completely gas-tight packaging where respiratory gases produced cannot be discharged (Matche, 2005). Sufficient material O_2 permeability ensures that, depending on the product, at least 2 - 5 % O_2 is present in the packaging during the entire storage period (Detzel et al., 2018), avoiding taste and color changes as well as anaerobic conditions within the packaging that causes fermentation metabolism of plant-based products (Agrartechnik Bornim, 2005). The relative humidity influencing microbial spoilage inside the packaging depends, among other things, on the water vapor permeability of the packaging itself (Almenar et al., 2010; Matche, 2005). If the products are exposed to changing temperatures, the formation of condensation is expected if the packaging is not sufficiently permeable to water vapor. However, the packaging of fruits and vegetables should not be too permeable to water vapor. Otherwise, plant-based foods tend to dry out quickly (Agrartechnik Bornim, 2005; Buchner, 1999).

With regard to the mechanical properties, the requirements of the packaging machines play a decisive role in addition to the product requirements (European Commission, 2018). The mechanical properties of a packaging material describe how it reacts to external forces. Various forces are evaluated, distinguishing between the direction, origin, and time of the impacted force (Lee et al., 2008). In order to protect plant-based foods from mechanical damage, the packaging material must have a certain tear and tensile strength, as well as compressive strength and elasticity (Agrartechnik Bornim, 2005). The physical protective function against mechanical damage and external contamination is important for most plant-based food packaging as bruises for example often only become visible at a later stage of ripeness (Sousa-Gallagher et al., 2016; van Linden et al., 2008). If packaged fruits and vegetables are stored in a cool place, the mechanical stability of the packaging must be maintained, even at low temperatures (Detzel et al., 2018).

Due to ambitious sustainability goals given by legislation, plastic packaging, in particular, is the focus of public and political discussion and requirements. Current challenges that producers, industry, and retailers are still facing are closing material cycles, saving packaging, and making it more sustainable (Seyring et al., 2020). Figure 1.2 shows approaches to increase the sustainability and resource efficiency of packaging while keeping in mind that a change in packaging should preserve product quality. Otherwise, the supply chain will be less sustainable.

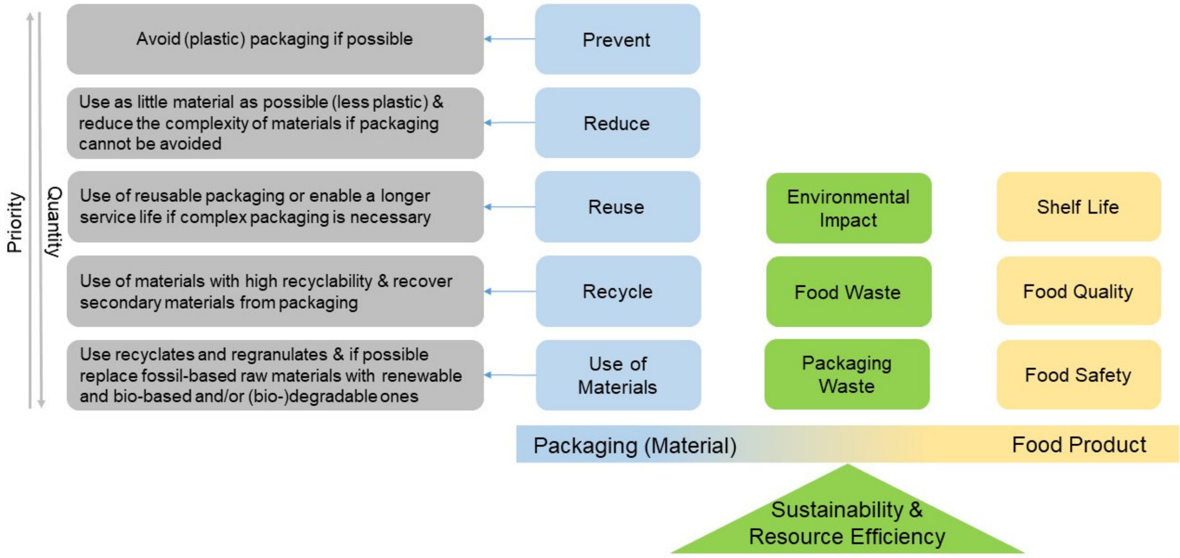


Figure 1.2 Approaches to increase the sustainability and resource efficiency of packaging. Based on Kreyenschmidt (2019) and Seyring et al. (2020).

Ensuring the quality of food and safety is one of the greatest challenges of the present food industry and, at the same time, one of the most important aspects when choosing appropriate packaging (Czerwiński et al., 2021). For this purpose, the functional properties of the packaging must fulfill the requirements of the specific food (Guillard et al., 2018). Therefore, research focuses on improving the characteristics of bio-based packaging materials, in particular mechanical, thermal, and physical properties focusing on maintaining food safety and shelf life and reducing food waste (Matthews et al., 2021). Different studies show a limited but growing number of natural polymers used as films and coatings applied for food packaging (Asgher et al., 2020; Biscarat et al., 2015). Although bio-based materials provide significant opportunities in terms of sustainability and biocompatibility and a broad interest from the food industry for implementation, until now, their use in industrial applications is often restricted due to poor commercial availability, lack of efficient production processes, and lower performance in fundamental packaging functions. Challenges such as relatively poor thermal, mechanical, and rheological properties; higher costs; lack of compatibility with the processing and recycling systems currently available; or perceived environmental issues of natural polymers must be overcome. In addition, the barrier properties of natural polymers, especially the moisture barrier properties due to the hydrophilic nature of these polymers, are detrimental to existing packaging materials (Aggarwal & Langowski, 2020; Cazón et al., 2017; Korte et al., 2021; Kumar et al., 2017; Thakur et al., 2018).

Microbiological contamination of food has led to the search for solutions to slow down the growth of microorganisms in food. To improve shelf life and to reduce food waste, new technologies, such as nanotechnology and active packaging, are promising strategies as these protect foods in physical, chemical, sensory, and microbiological ways (Czerwiński et al., 2021; Ilg & Kreyenschmidt, 2012; Wikström et al., 2019; Youssef & El-Sayed, 2018). The application of antibacterial polymers in food packaging can help to fulfill these requirements (Appendini & Hotchkiss, 2002; Czerwiński et al., 2021; Peelman et al., 2014). Antimicrobial systems target the control or reduction of microbial growth, which often results in the extension of the lag phase or a reduced growth rate in the exponential phase (Coma, 2008; Lavoine et al., 2014). Thus, these reactions achieve a longer shelf life, among other things (Barlow & Morgan, 2013; Matthews et al., 2021). The integration of antimicrobial agents in packaging can be realized by direct incorporation of the antimicrobial into the packaging material or by coating the packaging material with antimicrobial agents (Appendini & Hotchkiss, 2002; Cooksey, 2005; Han, 2003). To counteract non-satisfactory packaging characteristics of bio-based materials, research and developments include active packaging based on bioactive polymers and composites obtained from renewable resources. There are several developments in this field, but up to now, widespread use in the market is still missing. Additionally, adding active ingredients to packaging materials and/or using functionalized polymers lead to improved basic barrier properties (Korte et al., 2021).

Even though bio-based and biodegradable polymers, as well as the continued use of agricultural and industrial by-products and waste flows as raw materials, represent a growing field in creating sustainable packaging materials in the last years, mostly driven by political requirements, up to now a limited number of bio-based (polymer) materials are commercially available as food packaging. Comprehensive approaches considering sustainable packaging materials themselves, providing a significant advantage in terms of environmental impact, in relation to reducing food waste by maintaining shelf life, quality and safety of fresh produce are lacking. The effect of these packaging materials on the quality loss and shelf life of fresh produce like meat products and vegetables in comparison to commonly used fossil- or wood-based materials is often not considered, even though the packaging characteristics are known to have an impact. When pursuing approaches to improve material characteristics by antimicrobial systems, it is not known if these materials are still active over the broad spectrum of environmental and product factors existing in the food industry. The application of coatings either based on a mineral binder or the use of a mineral mixture as a functional extender, due to surface-active properties of minerals, as a coating in antimicrobial packaging to increase food safety and shelf life has not been investigated yet.

1.3 Research questions and outline of the thesis

The main objective of this thesis is the assessment of different innovative packaging strategies to maintain product characteristics and quality of different fresh produce, and thus to keep and even improve the resource efficiency of food processing and the packaging material itself. The innovative packaging strategies range from bio-based to more recyclable fossil-based packaging materials for use in supply chains of fresh produce. Furthermore, the potential and antimicrobial effectiveness of a novel antimicrobial active compound applied as a coating for active packaging materials is assessed.

For this purpose, the following research questions are considered:

- How is the status quo on research and practical use of bio-based packaging materials focusing on plant-derived polymers combined with plant-derived additives to maintain product quality and thereby prevent and reduce food waste? (Chapter 2)
- What is the effect of different bio-based packaging materials in comparison to conventionally used materials on product quality and shelf life of plant-based products? (Chapters 2, 3)
- Is it possible to substitute conventional multi-layered APET/PE foils with more sustainable packaging materials keeping product quality and safety of animal-based products and reducing the amount of packaging waste? (Chapters 2, 4)
- Are novel antimicrobial active compounds applied as surface coatings able to reduce the microbial count of spoilage and pathogenic bacteria, and how is it influenced by product, process, and environmental factors? (Chapters 2, 5)

In the first part (chapter 2) of the thesis, the status quo and practical use of bio-based polymers is discussed. The focus is laid on the use of renewable resources and biomass waste as raw materials for polymer production. The current practical use of bio-based polymers is considered, especially in terms of their applicability related to fundamental packaging functions, as these directly influence food quality and safety, the length of shelf life, and thus the amount of food waste. Additionally, this part of the thesis looks at plant extracts as active packaging agents that are capable to prolong the food shelf life, and their promising use by being incorporated into packaging materials. Finally, the adoption potential of packaging based on polymers from renewable resources is discussed from a bioeconomy perspective.

In chapter 3, the effect of bio-based packaging materials on quality loss and shelf life of fresh cherry tomatoes is determined. For this purpose, cherry tomatoes are packaged in different bio-based packaging materials from renewable resources comprising groundwood pulp, sugar cane, bamboo, cellulose, grass paper and polylactic acid and in reference materials (corrugated cardboard and rPET) and stored for 20 days under dynamic temperature conditions. The influence of packaging materials during storage was analyzed based on typical quality parameters. Besides the quality loss the packaging materials itself were investigated for surface bacterial counts before and after storage. Moreover, general material parameters were determined for selected (non-plastic) packaging materials.

The next chapter (chapter 4) focuses on the determination of possible substitution of conventional used multi-layer foils APET/PE with more sustainable packaging materials differing in foil thickness and combination, covering multi- and mono-layers to an innovative paper compound while keeping product quality of emulsion-type sausage and reducing the amount of packaging waste. In storage trials, the gas composition inside the packages was analyzed to identify gas permeability and defects of the applied materials. Microbiological parameters were analyzed, and instrumental color measurements were conducted to investigate the effect on product quality and shelf life.

In chapter 5, novel antimicrobial active compounds incorporated in different percentages in silicates and acrylic polymers are investigated for their effectiveness in reducing the microbial count of gram-positive and gram-negative spoilage and pathogenic bacteria. The focus is laid on the application as surface coatings acting as a coating for active packaging materials in general. The studies focused on the influence of selected factors of product, process, and environment, which are characteristic of perishable products, especially in chilled (meat) supply chains. The influence of selected specific gram-positive and gram-negative organisms, contact time, initial bacteria concentration, temperature, pH value, and presence of food components on the antimicrobial activity were determined. The results are used to evaluate the potential use of the investigated coatings as packaging coating for active packaging materials.

References

Abejón, R., Bala, A., Vázquez-Rowe, I., Aldaco, R., & Fullana-i-Palmer, P. (2020). When plastic packaging should be preferred: Life cycle analysis of packages for fruit and vegetable distribution in the Spanish peninsular market. *Resources, Conservation and Recycling*, *155*, 104666. https://doi.org/10.1016/j.resconrec.2019.104666.

Accorsi, R. (2019). A support-design procedure for sustainable food product-packaging systems. In *Sustainable Food Supply Chains* (pp. 61–81). Elsevier. https://doi.org/10.1016/B978-0-12-813411-5.00005-3.

Aggarwal, A., & Langowski, H.-C. (2020). Packaging Functions and Their Role in Technical Development of Food Packaging Systems: Functional Equivalence in Yoghurt Packaging. *Procedia CIRP*, *90*, 405–410. https://doi.org/10.1016/j.procir.2020.01.063.

Agrartechnik Bornim. (2005). *Quality Maintenance and Quality Assurance of Organic Fruit and Vegetables in the Postharvest Chain* (No. 48). Potsdam-Bornim.

Ahvenainen, R. (1996). New approaches in improving the shelf life of minimally processed fruit and vegetables. Review. *Trend in Food Science and Technology*, *7*, 179–187.

Almenar, E., Samsudin, H., Auras, R., & Harte, J. (2010). Consumer acceptance of fresh blueberries in bio-based packages. *Journal of the Science of Food and Agriculture*, *90*(7), 1121–1128. https://doi.org/10.1002/jsfa.3922.

Appendini, P., & Hotchkiss, J. H. (2002). Review of antimicrobial food packaging. *Innovative Food Science & Emerging Technologies*, *3*, 113–126. https://doi.org/10.1016/S1466-8564(02)00012-7.

Asgher, M., Urooj, Y., Qamar, S. A., & Khalid, N. (2020). Improved exopolysaccharide production from Bacillus licheniformis MS3: Optimization and structural/functional characterization. *International Journal of Biological Macromolecules*, *151*, 984–992. https://doi.org/10.1016/j.ijbiomac.2019.11.094.

Bajpai, P. (2012). *Biotechnology for Pulp and Paper Processing*. *SpringerLink Bücher*. Springer US. https://doi.org/10.1007/978-1-4614-1409-4.

Barlow, C. Y., & Morgan, D. C. (2013). Polymer film packaging for food: An environmental assessment. *Resources, Conservation and Recycling*, *78*, 74–80. https://doi.org/10.1016/j.resconrec.2013.07.003.

Barry-Ryan, C., & O'Beirne, D. (1998). Quality and Shelf-life of Fresh Cut Carrot Slices as Affected by Slicing Method. *Journal of Food Science*, *63*(5), 851–856. https://doi.org/10.1111/j.1365-2621.1998.tb17913.x.

Bekhit, A. E.-D. A., Holman, B. W., Giteru, S. G., & Hopkins, D. L. (2021). Total volatile basic nitrogen (TVB-N) and its role in meat spoilage: A review. *Trends in Food Science & Technology*, *109*, 280–302. https://doi.org/10.1016/j.tifs.2021.01.006.

Bertin, N. (2018). Fruit quality. In E. Heuvelink (Ed.), *Crop production science in horticulture: Vol. 27. Tomatoes* (2^{nd} ed., pp. 137–179). CABI CAB International.

Bhat, R., & Jõudu, I. (2019). Emerging issues and challenges in agri-food supply chain. In *Sustainable Food Supply Chains* (pp. 23–37). Elsevier. https://doi.org/10.1016/B978-0-12-813411-5.00002-8.

Biscarat, J., Charmette, C., Sanchez, J., & Pochat-Bohatier, C. (2015). Development of a new family of food packaging bioplastics from cross-linked gelatin based films. *The Canadian Journal of Chemical Engineering*, *93*(2), 176–182. https://doi.org/10.1002/cjce.22077.

Buchner, N. S. (1999). *Verpackung Von Lebensmitteln: Lebensmitteltechnologische, Verpackungstechnische und Mikrobiologische Grundlagen*. Springer Berlin / Heidelberg. https://ebookcentral.proquest.com/lib/kxp/detail.action?docID=6491843.

Buendía−Moreno, L., Sánchez−Martínez, M. J., Antolinos, V., Ros−Chumillas, M., Navarro−Segura, L., Soto−Jover, S., Martínez−Hernández, G. B., & López−Gómez, A. (2020). Active cardboard box with a coating including essential oils entrapped within cyclodextrins and/or halloysite nanotubes. A case study for fresh tomato storage. *Food Control*, *107*, 106763. https://doi.org/10.1016/j.foodcont.2019.106763.

Caldeira, C., Laurentiis, V. de, Corrado, S., van Holsteijn, F., & Sala, S. (2019). Quantification of food waste per product group along the food supply chain in the European Union: A mass flow analysis. *Resources, Conservation, and Recycling*, *149*, 479–488. https://doi.org/10.1016/j.resconrec.2019.06.011.

Cazón, P., Velazquez, G., Ramírez, J. A., & Vázquez, M. (2017). Polysaccharide-based films and coatings for food packaging: A review. *Food Hydrocolloids*, *68*, 136–148. https://doi.org/10.1016/j.foodhyd.2016.09.009.

Chakraverty, A., & Singh, R. P. (2016). *Postharvest Technology and Food Process Engineering*. CRC Press. https://doi.org/10.1201/b15587.

Coelho, P. M., Corona, B., Klooster, R. ten, & Worrell, E. (2020). Sustainability of reusable packaging–Current situation and trends. *Resources, Conservation & Recycling: X*, *6*, Article 100037. https://doi.org/10.1016/j.rcrx.2020.100037.

Coles, R. (2003). Introduction. In R. Coles, D. McDowell, & M. J. Kirwan (Eds.), *Food Packaging Technology* (pp. 1–29). Blackwell Publishing.

Coma, V. (2008). Bioactive packaging technologies for extended shelf life of meat-based products. *Meat Science*, *78*(1-2), 90–103. https://doi.org/10.1016/j.meatsci.2007.07.035.

Comi, G. (2016). Chapter 8 - Spoilage of Meat and Fish. In A. Bevilacqua, M. R. Corbo, & M. Sinigaglia (Eds.), *Woodhead Publishing series in food science, technology and nutrition. The microbiological quality of food: Foodborne spoilers* (pp. 179–210). Woodhead Publishing is an imprint of Elsevier. https://doi.org/10.1016/B978-0-08-100502-6.00011-X.

Cooksey, K. (2005). Effectiveness of antimicrobial food packaging materials. *Food Additives and Contaminants*, *22*(10), 980–987. https://doi.org/10.1080/02652030500246164.

Cooreman-Algoed, M., Boone, L., Taelman, S. E., van Hemelryck, S., Brunson, A., & Dewulf, J. (2022). Impact of consumer behaviour on the environmental sustainability profile of food production and consumption chains – a case study on chicken meat. *Resources, Conservation and Recycling*, *178*, 106089. https://doi.org/10.1016/j.resconrec.2021.106089.

Corrado, S., Ardente, F., Sala, S., & Saouter, E. (2017). Modelling of food loss within life cycle assessment: From current practice towards a systematisation. *Journal of Cleaner Production*, *140*, 847–859. https://doi.org/10.1016/j.jclepro.2016.06.050.

Czerwiński, K., Rydzkowski, T., Wróblewska-Krepsztul, J., & Thakur, V. K. (2021). Towards Impact of Modified Atmosphere Packaging (MAP) on Shelf-Life of Polymer-Film-Packed Food Products: Challenges and Sustainable Developments. *Coatings*, *11*(12), 1504. https://doi.org/10.3390/coatings11121504.

Dannehl, D., Huyskens-Keil, S., & Schmidt, U. (2008). Untersuchungen zur Lagerung von Erdbeeren unter Berücksichtigung verschiedener Verpackungsmaterialien. *Erwerbs-Obstbau*, *50*(2), 49–61. https://doi.org/10.1007/s10341-008-0062-3.

Dave, D., & Ghaly, A. E. (2011). Meat Spoilage Mechanisms and Preservation Techniques: A Critical Review. *American Journal of Agricultural and Biological Sciences*, *6*(4), 486–510. https://doi.org/10.3844/ajabssp.2011.486.510.

Detzel, A., Bodrogi, F., Kauertz, B., Bick, C., Welle, F., Schmid, M., Schmitz, K., & Müller, K. (2018). *Biobasierte Kunststoffe als Verpackung von Lebensmitteln*. Heidelberg, Freising, Berlin. Institut für Energie- und Umweltforschung Heidelberg.

Dilkes-Hoffman, L. S., Lane, J. L., Grant, T., Pratt, S., Lant, P. A., & Laycock, B. (2018). Environmental impact of biodegradable food packaging when considering food waste. *Journal of Cleaner Production*, *180*, 325–334. https://doi.org/10.1016/j.jclepro.2018.01.169.

Dungani, R., Khalil, H. P. S. A., Sumardi, I., Suhaya, Y., Sulistyawati, E., Islam, M. N., Suraya, N. L. M., & Aprilia, N. A. S. (2014). Non-wood Renewable Materials: Properties Improvement and Its Application. In K. R. Hakeem, M. Jawaid, & U. Rashid (Eds.), *Biomass and Bioenergy: Applications* (pp. 1–29). Springer International Publishing. https://doi.org/10.1007/978-3-319-07578-5_1.

Ellen MacArthur Foundation and McKinsey & Company (2016). The New Plastics Economy: Rethinking the future of plastics. World Economic Forum. http://www.ellenmacarthurfoundation.org/publications.

European Commission (2018). Environmental impact assessments of innovative bio-based product: Task 1 of “Study on Support to R&I Policy in the Area of Bio-based Products and Services”.

Fischer, M., & Glomb, M. A. (2015). *Moderne Lebensmittelchemie* (1. Aufl.). Behr.

Gill, C. O. (1983). Meat Spoilage and Evaluation of the Potential Storage Life of Fresh Meat. *Journal of Food Protection*, *46*(5), 444–452. https://doi.org/10.4315/0362-028X-46.5.444.

Guillard, V., Gaucel, S., Fornaciari, C., Angellier-Coussy, H., Buche, P., & Gontard, N. (2018). The Next Generation of Sustainable Food Packaging to Preserve Our Environment in a Circular Economy Context. *Frontiers in Nutrition*, *5*, 121. https://doi.org/10.3389/fnut.2018.00121.

Hakeem, K. R. (2014). *Biomass and Bioenergy: Applications* (1st ed.). Springer International Publishing AG. https://books.google.de/books?id=kgVYBAAAQBAJ.

Han, J. H. (2003). Antimicrobial food packaging. In *Novel Food Packaging Techniques* (pp. 50–70). Elsevier. https://doi.org/10.1533/9781855737020.1.50.

Heller, M. C., Selke, S. E. M., & Keoleian, G. A. (2019). Mapping the Influence of Food Waste in Food Packaging Environmental Performance Assessments. *Journal of Industrial Ecology*, *23*(2), 480–495. https://doi.org/10.1111/jiec.12743.

Herbert, U., & Kreyenschmidt, J. (2015). Comparison of Oxygen- and Nitrogen-Enriched Atmospheres on the Growth of Listeria Monocytogenes Inoculated on Poultry Breast Fillets. *Journal of Food Safety*, *35*(4), 533–543. https://doi.org/10.1111/jfs.12203.

Herbert, U., Rossaint, S., Khanna, M.-A., & Kreyenschmidt, J. (2013). Comparison of argon-based and nitrogen-based modified atmosphere packaging on bacterial growth and product quality of chicken breast fillets. *Poultry Science*, *92*(5), 1348–1356. https://doi.org/10.3382/ps.2012-02590.

Huis in't Veld, J. H. (1996). Microbial and biochemical spoilage of foods: an overview. *International Journal of Food Microbiology*, *33*(1), 1–18. https://doi.org/10.1016/0168-1605(96)01139-7.

Ilg, Y., & Kreyenschmidt, J. (2012). Review: Nutzen und Risiken der Anwendung antimikrobieller Werkstoffe in der Lebensmittelkette. *Journal of Food Safety and Food Quality*, *63*(2), 28–34.

Kader, A., & Barrett, D. (2005). Classification, Composition of Fruits, and Postharvest Maintenance of Quality. In D. Barrett, L. Somogyi, H. Ramaswamy, & D. M. Barrett (Eds.), *Processing fruits: Science and technology* (2nd ed.). CRC Press. https://doi.org/10.1201/9781420040074.pt1.

Kalamaki, M. S., Stoforos, N. G., & Taoukis, P. S. (2012). Pectic Enzymes in Tomatoes. In B. K. Simpson (Ed.), *Food biochemistry and food processing* (2nd ed., pp. 232–246). Wiley-Blackwell. https://doi.org/10.1002/9781118308035.ch12.

Kan, M., & Miller, S. A. (2022). Environmental impacts of plastic packaging of food products. *Resources, Conservation and Recycling*, *180*, 106156. https://doi.org/10.1016/j.resconrec.2022.106156.

Korte, I., Kreyenschmidt, J., Wensing, J., Bröring, S., Frase, J. N., Pude, R., Konow, C., Havelt, T., Rumpf, J., Schmitz, M., & Schulze, M. (2021). Can Sustainable Packaging Help to Reduce Food Waste? A Status Quo Focusing Plant-Derived Polymers and Additives. *Applied Sciences*, *11*(11), 5307. https://doi.org/10.3390/app11115307.

Krämer, J., & Prange, A. (2017). *Lebensmittel-Mikrobiologie* (7., vollständig überarbeitete Auflage). *UTB Lebensmittel- und Ernährungswissenschaften, Biologie: Vol. 1421*. Verlag Eugen Ulmer.

Kreyenschmidt, J. (2019, November 20). *Smart Packaging – Die Entwicklung intelligenter Verpackungen für Frischfleisch und Wurst.* afz & Fleischwirtschaft. Deutscher Fleisch Kongress 2019, Wiesbaden.

Kreyenschmidt, J., Albrecht, A., Braun, C., Herbert, U., Mack, M., Rossaint, S., Ritter, G., Teitscheid, P., & Ilg, Y. (2013). Food Waste in der Fleisch verarbeitenden Kette. *Fleischwirtschaft*, *10*, 57–63.

Kreyenschmidt, J., & Ibald, R. (2012). Modeling Shelf Life Using Microbial Indicators. In M. Nicoli & M. C. Nicoli (Eds.), *Food preservation technology series. Shelf life assessment of food* (Vol. 20122242, pp. 127–168). CRC Press. https://doi.org/10.1201/b11871-7.

Kumar, N., Kaur, P., & Bhatia, S. (2017). Advances in bio-nanocomposite materials for food packaging: a review. *Nutrition & Food Science*, *47*(4), 591–606. https://doi.org/10.1108/NFS-11-2016-0176.

Lavoine, N., Givord, C., Tabary, N., Desloges, I., Martel, B., & Bras, J. (2014). Elaboration of a new antibacterial bio-nano-material for food-packaging by synergistic action of cyclodextrin and microfibrillated cellulose. *Innovative Food Science & Emerging Technologies*, *26*, 330–340. https://doi.org/10.1016/j.ifset.2014.06.006.

Lee, D. S., Yam, K. L., & Piergiovanni, L. (2008). *Food packaging science and technology*. CRC Press.

Marsh, K., & Bugusu, B. (2007). Food packaging—roles, materials, and environmental issues. *Journal of Food Science*, *72*(3), R39-55. https://doi.org/10.1111/j.1750-3841.2007.00301.x.

Matche, R. S. (2005). Packaging Aspects of Fruits and Vegetables. In *Food Packaging Technology Department Central Food Technological Research Institute* (pp. 116–132).

Matthews, C., Moran, F., & Jaiswal, A. K. (2021). A Review on European Union's Strategy for Plastics in a Circular Economy and its Impact on Food Safety. *Journal of Cleaner Production*, *283*, Article 125263.

McMillin, K. W. (2017). Advancements in meat packaging. *Meat Science*, *132*, 153–162. https://doi.org/10.1016/j.meatsci.2017.04.015.

Mena, C., Adenso-Diaz, B., & Yurt, O. (2011). The causes of food waste in the supplier–retailer interface: Evidences from the UK and Spain. *Resources, Conservation and Recycling*, *55*(6), 648–658. https://doi.org/10.1016/j.resconrec.2010.09.006.

Mena, C., Terry, L. A., Williams, A., & Ellram, L. (2014). Causes of waste across multi-tier supply networks: Cases in the UK food sector. *International Journal of Production Economics*, *152*, 144–158. https://doi.org/10.1016/j.ijpe.2014.03.012.

Molina-Besch, K., Wikström, F., & Williams, H. (2019). The environmental impact of packaging in food supply chains—does life cycle assessment of food provide the full picture? *The International Journal of Life Cycle Assessment*, *24*(1), 37–50. https://doi.org/10.1007/s11367-018-1500-6.

Opara, U. L., Al-Ani, M. R., & Al-Rahbi, N. M. (2012). Effect of Fruit Ripening Stage on Physico-Chemical Properties, Nutritional Composition and Antioxidant Components of Tomato (Lycopersicum esculentum) Cultivars. *Food and Bioprocess Technology*, *5*(8), 3236–3243. https://doi.org/10.1007/s11947-011-0693-5.

Opara, U. L., Caleb, O. J., & Belay, Z. A. (2019). 7 - Modified atmosphere packaging for food preservation. In C. M. Galanakis (Ed.), *Food Quality and Shelf Life* (pp. 235–259). Elsevier Science & Technology. https://doi.org/10.1016/B978-0-12-817190-5.00007-0.

Opara, U. L., & Mditshwa, A. (2013). A review on the role of packaging in securing food system: Adding value to food products and reducing losses and waste. *African Journal of Agrucultural Research*, *8*(22), 2621–2630.

Pathare, P. B., & Opara, U. L. (2014). Structural design of corrugated boxes for horticultural produce: A review. *Biosystems Engineering*, *125*, 128–140. https://doi.org/10.1016/j.biosystemseng.2014.06.021.

Patrignani, F., Siroli, L., Gardini, F., & Lanciotti, R. (2016). Contribution of Two Different Packaging Material to Microbial Contamination of Peaches: Implications in Their Microbiological Quality. *Frontiers in Microbiology*, *7*, 938. https://doi.org/10.3389/fmicb.2016.00938.

Pauer, E., Tacker, M., Gabriel, V., & Krauter, V. (2020). Sustainability of flexible multilayer packaging: Environmental impacts and recyclability of packaging for bacon in block. *Cleaner Environmental Systems*, *1*, Article 100001. https://doi.org/10.1016/j.cesys.2020.100001.

Peelman, N., Ragaert, P., Vandemoortele, A., Verguldt, E., Meulenaer, B. de, & Devlieghere, F. (2014). Use of biobased materials for modified atmosphere packaging of short and medium shelf-life food products. *Innovative Food Science & Emerging Technologies*, *26*, 319–329. https://doi.org/10.1016/j.ifset.2014.06.007.

Pellissery, A. J., Vinayamohan, P. G., Amalaradjou, M. A. R., & Venkitanarayanan, K. (2020). Spoilage bacteria and meat quality. In *Meat Quality Analysis* (pp. 307–334). Elsevier. https://doi.org/10.1016/B978-0-12-819233-7.00017-3.

Petrovic, Z., Djordjevic, V., Milicevic, D., Nastasijevic, I., & Parunovic, N. (2015). Meat Production and Consumption: Environmental Consequences. *Procedia Food Science*, *5*, 235–238. https://doi.org/10.1016/j.profoo.2015.09.041.

Pro Carton (2010). Wie wichtig ist nachhaltige Verpackung? Die Einstellung von Konsumenten zu Verpackung und Nachhaltigkeit, *3*.

Qin, Y., & Horvath, A. (2022). What contributes more to life-cycle greenhouse gas emissions of farm produce: Production, transportation, packaging, or food loss? *Resources, Conservation and Recycling*, *176*, 105945. https://doi.org/10.1016/j.resconrec.2021.105945.

Ragaert, K., Delva, L., & van Geem, K. (2017). Mechanical and chemical recycling of solid plastic waste. *Waste Management (New York, N.Y.)*, *69*, 24–58. https://doi.org/10.1016/j.wasman.2017.07.044.

Robertson, G. L. (2012). *Food Packaging: Principles and Practice, Third Edition* (3rd ed.). CRC Press. https://doi.org/10.1201/b21347.

Robertson, G. L. (2016). Packaging and Food and Beverage Shelf Life. In P. Subramaniam (Ed.), *Woodhead Publishing series in food science, technology and nutrition: Vol. 297. The stability and shelf life of food* (pp. 77–106). Elsevier/WP Woodhead Publishing. https://doi.org/10.1016/B978-0-08-100435-7.00003-4.

Rossaint, S., & Kreyenschmidt, J. (2014). Intelligent label – a new way to support food waste reduction. *Waste and Resource Management*, *168*(2), Article 1300035, 63–71. https://doi.org/10.1680/warm.13.00035.

Ščetar, M., Kurek, M., & Galić, K. (2010). Trends in fruit and vegetable packaging - a review. *Hrvatski Časopis Za Prehrambenu Tehnologiju Biotehnologiju I Nutricionizam - Croatian Journal of Food Technology, Biotechnology and Nutrition*, *5*(3/4), 69–86.

Scherhaufer, S., Moates, G., Hartikainen, H., Waldron, K., & Obersteiner, G. (2018). Environmental impacts of food waste in Europe. *Waste Management*, *77*, 98–113. https://doi.org/10.1016/j.wasman.2018.04.038.

Schumann, B., & Schmid, M. (2018). Packaging concepts for fresh and processed meat – Recent progresses. *Innovative Food Science & Emerging Technologies*, *47*, 88–100. https://doi.org/10.1016/j.ifset.2018.02.005.

Seier, M., Archodoulaki, V.-M., Koch, T., Duscher, B., & Gahleitner, M. (2022). Polyethylene terephthalate based multilayer food packaging: Deterioration effects during mechanical recycling. *Food Packaging and Shelf Life*, *33*, 100890. https://doi.org/10.1016/j.fpsl.2022.100890.

Seyring, N., Boehlke, K., Kaeding-Koppers, A., & Erbar, I. (2020). *Recyclingfähige und nachhaltige Verpackungen: Ein Leitfaden für Unternehmen*. München.

Sharma, R., & Ghoshal, G. (2018). Emerging trends in food packaging. *Nutrition & Food Science*, *48*(5), 764–779. https://doi.org/10.1108/NFS-02-2018-0051.

Soro, A. B., Noore, S., Hannon, S., Whyte, P., Bolton, D. J., O'Donnell, C., & Tiwari, B. K. (2021). Current sustainable solutions for extending the shelf life of meat and marine products in the packaging process. *Food Packaging and Shelf Life*, *29*, 100722. https://doi.org/10.1016/j.fpsl.2021.100722.

Taleb, M. A., & Al Farooque, O. (2021). Towards a Circular Economy for Sustainable Development: An Application of Full Cost Accounting to Municipal Waste Recyclables. *Journal of Cleaner Production*, *280*, Article 124047. https://doi.org/10.1016/j.jclepro.2020.124047.

Teck Kim, Y., Min, B., & Won Kim, K. (2014). General Characteristics of Packaging Materials for Food System. In *Innovations in Food Packaging* (pp. 13–35). Elsevier. https://doi.org/10.1016/B978-0-12-394601-0.00002-3.

Thakur, S., Chaudhary, J., Sharma, B., Verma, A., Tamulevicius, S., & Thakur, V. K. (2018). Sustainability of bioplastics: Opportunities and challenges. *Current Opinion in Green and Sustainable Chemistry*, *13*, 68–75. https://doi.org/10.1016/j.cogsc.2018.04.013.

Torrieri, E. (2015). Storage Stability: Shelf Life Testing. In B. Caballero, P. Finglas, & F. Toldrá (Eds.), *Encyclopedia of Food and Health* (3rd ed., pp. 188–192). Elsevier Science. https://doi.org/10.1016/B978-0-12-384947-2.00666-8.

United Nations (2015). Transforming our world: the 2030 Agenda for Sustainable Development. *General Assembly*, *A/Res/70/1*.

van Linden, V., Sila, D. N., Duvetter, T., Baerdemaeker, J. de, & Hendrickx, M. (2008). Effect of mechanical impact-bruising on polygalacturonase and pectinmethylesterase activity and pectic cell wall components in tomato fruit. *Postharvest Biology and Technology*, *47*(1), 98–106. https://doi.org/10.1016/j.postharvbio.2007.06.006.

Verghese, K., Lewis, H., Lockrey, S., & Williams, H. (2015). Packaging's Role in Minimizing Food Loss and Waste Across the Supply Chain. *Packaging Technology and Science*, *28*(7), 603–620. https://doi.org/10.1002/pts.2127.

Verma, D., Gope, P. C., Maheshwari, M. K., & Sharma, R. K. (2012). Bagasse Fiber Composites-A Review. *J Mater Environ Sci*, *3*(6), 1079–1092.

Wikström, F., Verghese, K., Auras, R., Olsson, A., Williams, H., Wever, R., Grönman, K., Kvalvåg Pettersen, M., Møller, H., & Soukka, R. (2019). Packaging Strategies That Save Food: A Research Agenda for 2030. *Journal of Industrial Ecology*, *23*(3), 532–540. https://doi.org/10.1111/jiec.12769.

Williams, H., & Wikström, F. (2011). Environmental impact of packaging and food losses in a life cycle perspective: a comparative analysis of five food items. *Journal of Cleaner Production*, *19*(1), 43–48. https://doi.org/10.1016/j.jclepro.2010.08.008.

Yildirim, S., Röcker, B., Pettersen, M. K., Nilsen-Nygaard, J., Ayhan, Z., Rutkaite, R., Radusin, T., Suminska, P., Marcos, B., & Coma, V. (2018). Active Packaging Applications for Food. *Comprehensive Reviews in Food Science and Food Safety*, *17*(1), 165–199. https://doi.org/10.1111/1541-4337.12322.

Youssef, A. M., & El-Sayed, S. M. (2018). Bionanocomposites materials for food packaging applications: Concepts and future outlook. *Carbohydrate Polymers*, *193*, 19–27. https://doi.org/10.1016/j.carbpol.2018.03.088.

2 Can sustainable packaging help to reduce food waste? A status quo focusing plant-derived polymers and additives

Imke Korte[1], Judith Kreyenschmidt[1,2,*], Joana Wensing[3], Stefanie Bröring[4], Jan Niklas Frase[5], Ralf Pude[5], Christopher Konow[6], Thomas Havelt[7], Jessica Rumpf[7], Michaela Schmitz[5,7,*] and Margit Schulze[7,*]

[1] Institute of Animal Sciences, Rheinische Friedrich Wilhelms-University Bonn, Katzenburgweg 7-9, D-53115 Bonn, Germany

[2] Department of Fresh Produce Logistics, Hochschule Geisenheim University, Von-Lade-Straße 1, D-65366 Geisenheim, Germany

[3] Departement of Social Sciences, Wageningen University, P.O. Box 8130, Bode 55, 6700 EW Wageningen, The Netherlands

[4] Institue for Food and Resource Economics, Rheinische Friedrich Wilhelms-University Bonn, Meckenheimer Allee 174, D-53115 Bonn, Germany

[5] Institute of Crop Science and Resource Conservation, INRES-Renewable Resources, Rheinische Friedrich Wilhelms-University Bonn, Campus Klein-Altendorf 1, D-53359 Rheinbach, Germany

[6] Department of Chemistry, MS 015, Brandeis University, 415 South Street, Waltham, MA 02453, USA

[7] Department of Natural Sciences, Institute of Technology, Resource and Energy-Efficient Engineering (TREE), Bonn-Rhein-Sieg University of Applied Sciences, von-Liebig-Strasse 20, D-53359 Rheinbach, Germany

* Correspondence

Korte, I., Kreyenschmidt, J., Wensing, J., Bröring, S., Frase, J. N., Pude, R., Konow, C., Havelt, T., Rumpf, J., Schmitz, M., & Schulze, M. (2021). Can Sustainable Packaging Help to Reduce Food Waste? A Status Quo Focusing Plant-Derived Polymers and Additives. *Applied Sciences*, *11*(11), 5307. https://doi.org/10.3390/app11115307.

2.1 Abstract

The promotion of sustainable packaging is part of the European Green Deal and plays a key role in the EU's social and political strategy. One option is the use of renewable resources and biomass waste as raw materials for polymer production. Lignocellulose biomass from annual and perennial industrial crops and agricultural residues are a major source of polysaccharides, proteins, and lignin and can also be used to obtain plant-based extracts and essential oils. Therefore, these biomasses are considered as potential substitute for fossil-based resources. Here, the status quo of bio-based polymers is discussed and evaluated in terms of properties related to packaging applications such as gas and water vapor permeability as well as mechanical properties. So far, their practical use is still restricted due to lower performance in fundamental packaging functions that directly influence food quality and safety, the length of shelf life, and thus the amount of food waste. Besides bio-based polymers, this review focuses on plant extracts as active packaging agents. Incorporating extracts of herbs, flowers, trees, and their fruits is inevitable to achieve desired material properties that are capable to prolong the food shelf life. Finally, the adoption potential of packaging based on polymers from renewable resources is discussed from a bioeconomy perspective.

2.2 Introduction

The main functions of food packaging are protection/preservation, containment, communication/marketing, and convenience. Thereby, food safety and quality related properties as well as reducing food waste are targeted (Aggarwal & Langowski, 2020; R. Sharma & Ghoshal, 2018; Youssef & El-Sayed, 2018). With an appropriate packaging solution, a high quality, safe (extended) shelf life along the entire supply chain can be ensured. Thus, possible waste of the food product prior to final consumption can be prevented (Accorsi, 2019; Aggarwal & Langowski, 2020; Coelho et al., 2020; Kreyenschmidt et al., 2013; R. Sharma & Ghoshal, 2018).

There are different packaging strategies to achieve a longer shelf life. One of the main strategies is the application of materials with certain barrier functions like gas and water vapor permeability (WVP) guaranteed by various fossil-based materials (Barlow & Morgan, 2013; Matthews et al., 2021) (for definition of terms such as fossil-based, bio-based, etc., see glossary at the end of chapter 2). Many of these fossil-based foils are complex multi-layer materials that represent a large group of individual fossil-based polymers with different chemical and technical characteristics due to diverse requirements regarding food safety and waste reduction (Ellen MacArthur Foundation and McKinsey & Company, 2016; Faraca & Astrup, 2019; Matthews et al., 2021). Another strategy is the use of active packaging (Ilg & Kreyenschmidt, 2012; Youssef & El-Sayed, 2018). Based on the European Union (EU) Guidance to the Commission Regulation No 450/2009, active materials are defined as: "[…] materials […] that are intended to extend the shelf life or to maintain or improve the condition of packaged food; they are designed to deliberately incorporated components that would release or absorb substances into or from the packaged food or the environment surrounding the food." (European Union, 2009)

Today, around 40 % of all plastics circulating are applied for packaging (PlasticsEurope, 2019), of which approximately 60 % are used for food and beverages while the rest covers non-food applications (European Commission, 2018). In Germany, the packaging consumption of glass reached 35.0 kg/head, for paper 98.5 kg/head and for plastic 39.0 kg/head in 2018 (Umweltbundesamt, 2020). Plastic waste represents one of the most complex material mixtures from a recycling perspective (K. Ragaert et al., 2017). Moreover, there are increasing issues concerning the harm caused to the environment mainly due to the manufacturing phase (i.e., oil refinery and material production), problematic end-of-life strategies, and adverse effects on human health (Accorsi, 2019; European Commission, 2018). Despite the negative environmental and health-related effects, fossil-based plastics are favored because of their lightweight nature and low costs (Ellen MacArthur Foundation and McKinsey & Company, 2016; Matthews et al., 2021). As a result, the use of plastic packaging is growing (van Eygen et al., 2017) caused by the need to reduce food waste due to the steadily growing population and market expansion (Sohail et al., 2018). To deal with the issue of adverse impact of fossil-based plastics on human health and environment, the design and production of plastics and plastic products must take account of the end-of-use strategy such as reuse, repair, and recycling needs (European Commission, 2018). This leads to a paradigm shift from linear to circular economy. The core principles "take, make, dispose" of a linear economy are replaced by "take, make, reuse" in a circular economy. "Reuse" involves circular criteria like repair, refurbish, and recycle, as recently reported by Taleb & Al Farooque (2021).

Within the last decade, the development and promotion of more sustainable materials became key roles on social and political levels in the EU (European Commission, 2018). Pursuing these strategies, the European Commission adopted a Circular Economy Action Plan in 2015 (European Commission, 2015). This action plan lays the foundation for a new plastics economy addressing the environmental issues concerning plastics and forcing the EU to move towards a more sustainable model for economic development (European Commission, 2019). The EU launched and developed the action plan in 2018 with the "European Strategy for Plastics in a Circular Economy", the so-called plastics strategy (European Commission, 2018). The plastics strategy forces the industry to rethink plastics design and their usage, disposal, and recycling within the entire value chain. The main goal is to achieve improvements in sustainability (Matthews et al., 2021).

To reduce environmental impacts, one key requirement is the complete reusability and/or recyclability of all plastic packaging placed on the EU market by 2030 (European Commission, 2018). Moreover, Matthews et al. (2021) stated that innovations in food packaging have to focus on maintaining food safety and shelf life and reducing food waste.

Besides current political requirements, sustainable packaging is also an important aspect for consumers (European Commission, 2018; Pro Carton, 2010). As consumer preferences shifted to high quality and safe products with enhanced shelf life, the development of various new trends in packaging systems has arisen (R. Sharma & Ghoshal, 2018). There are different strategies for the development of sustainable packaging such as reducing packaging materials by decreasing the thickness and/or number of layers, applying bio-based and/or biodegradable materials, reducing the amount of layers, and using easy recyclable materials (Pro Carton, 2010). For example, material properties of simple mono-layer materials can be improved by design of packaging itself such as introducing packaging conditions through a modified atmosphere or by using active packaging solutions (Yildirim et al., 2018). Recently, Pauer et al. (2020) reported that the environmental benefit of weight reduction is greater than the benefit from improved recyclability in terms of meat packaging.

Therefore, bio-based and biodegradable polymers represent a growing field in creating environmentally friendly materials (Fabra et al., 2014). The continued use of agricultural and industrial by-products and waste flows such as corn stover, wheat straw, and whey constitutes from dairy and cheese industries as raw materials would provide a significant ecological advantage and would reduce pressure on land use (Ahorsu et al., 2018; Koller et al., 2013; Narodoslawsky et al., 2015). Plant-based (waste) materials such as wood and lignocellulosic residues from agriculture and forestry are a major source of polysaccharides; therefore, they are considered as sustainable alternatives. They have the potential to be used instead of fossil resources (Daioglou et al., 2015; Chunping Xu et al., 2014). Concerns about greenhouse gas (GHG) emissions and the security of industrial feedstock supplies promote substituting conventional fossil-based feedstock in the production of synthetic materials with biomass (Weiss et al., 2012). Several studies considering cradle-to-grave life cycle analysis (LCA) of various bio- and fossil-based plastics showed that the production and use of plastics produced from renewable resources is generally advantageous in terms of saving fossil resources and reducing GHG emissions (Ahorsu et al., 2018; Broeren et al., 2017; Zhu et al., 2016). Biodegradable or compostable plastics can reduce the amount of waste sent to landfills (Matthews et al., 2021; R. Sharma & Ghoshal, 2018). Natural bio-based polymers such as agar, chitosan, cellulose, and starch represent the group of bio-based polymers in food packaging applications (Rhim et al., 2013).

Although significant effort is currently being made to develop novel, sustainable materials, there are currently no competitive alternatives which offer the same level of protection to fossil-based multi-layer plastic packaging, especially for fresh products like meat (Matthews et al., 2021).

Although research focuses on improving the bio-based film characteristics of packaging materials, their mechanical, thermal, and physical properties are still non-satisfactory, and their use in industrial applications is often restricted (Youssef & El-Sayed, 2018). To counteract these disadvantages by replacing fossil-based plastic packaging, research, and developments include active packaging based on bioactive polymers and composites obtained from renewable resources (Oliveira Filho et al., 2019; R. Sharma & Ghoshal, 2018). There are several developments in this field, but up to now, a widespread use in the market is still missing. Today, a limited number of bio-based plastics with food packaging applications are commercially available mainly based on the following polymers: poly (lactic acid) (PLA), poly(hydroxyalkanoates) (PHA), pol(yethylene furanoate) (PEF), poly(butylene succinate) (PBS), thermoplastic cellulose derivatives, and starch-based films (R. Sharma & Ghoshal, 2018).

This review focuses on most recent developments of packaging materials (polymers and composites) that are produced from renewable resources and considered as promising alternatives for fossil-based plastics. Annual and perennial crops, herbs, flowers, lignocellulosic and agricultural residues are a major source of polysaccharides, proteins, and lignin. In addition, they can also be used to obtain plant extracts and essential oils (EO). Therefore, we consider their application in terms of sustainable packaging that contribute to the reduction of food waste. First, challenges along food supply chains related to packaging characteristics are discussed: advantages and disadvantages related to the characteristics of the most prominent fossil-based plastics packaging are compared. They are evaluated regarding food waste, recyclability, and sustainability issues (chapter 2.3). Second, characteristics of selected packaging plastics (mainly plant-derived polymers) in terms of mechanical, thermal, and physical properties are highlighted. Biodegradability will not be addressed in detail but mainly properties, such as gas permeability with influence on the food quality, safety, and shelf life (chapter 2.4). Third, natural additives with focus on plant extracts in the context of active food packaging are presented (chapter 2.5). Incorporating extracts of herbs, flowers, trees, and their fruits is inevitable to achieve desired material properties that are capable to prolong shelf life, resulting in reduced food waste. Finally, in chapter 2.6, the potential of renewable resources is evaluated from a bioeconomy perspective of a packaging core matrix as well as a source for natural additives for active packaging.

2.3 Challenges in food supply chains related to packaging characteristics

2.3.1 Current packaging characteristics

While bio-based systems are the focus of the review, we begin by discussing general issues of packaging and the reasons behind the current ubiquitousness of fossil-based food packaging. Intensive research and development efforts over a long period of time resulted in packaging solutions which are optimized with regard to various important parameters that guarantee their functionality including material weight per unit of packed volume (Matthews et al., 2021; van Sluisveld & Worrell, 2013), mechanical characteristics (impact and tear strength, stiffness, flexibility), durability, and many others (Accorsi, 2019; Ellen MacArthur Foundation and McKinsey & Company, 2016; Hahladakis & Iacovidou, 2018; Marra et al., 2016; van Eygen et al., 2017; Youssef & El-Sayed, 2018). Moreover, fossil-based plastics are less expensive per weight unit compared to most of the bio-based materials. Therefore, currently, alternative bio-based materials are more expensive because of poor commercial availability and lack of efficient production processes. In the future, the prices for fossil resources will rise due to the limited availability, and on the other hand, costs for bio-based plastics may drop due to improvements in production process efficiency (van den Oever et al., 2017). The current focus of research activities is the reduction of packaging waste especially fossil-based material (Accorsi, 2019). LCA showed that a reduction in environmental impact of the packaging itself can be best achieved by minimization of used materials (thinner layers) that retain mechanical and barrier properties rather than emphasizing end-of-life issues (such as recycling or disposal) (Barlow & Morgan, 2013).

Today, multi-layer materials are widely used for food packaging throughout the food industry. As multi-layers of plastic can be easily adjusted to the various requirements of different food types, this kind of packaging can offer effective solutions to maintain food safety and quality, achieve optimal shelf life, and minimize food waste (Matthews et al., 2021). Although from an environmental, sustainability, and biocompatibility perspective, the use of multi-layered packaging materials has to be reduced, their global use for food applications is growing. This development is based on the mechanical and barrier properties of multi-layer materials (higher resistance to water, gas, and aroma transfer) (Barlow & Morgan, 2013; Matthews et al., 2021; R. Sharma & Ghoshal, 2018). Especially, the barrier against oxygen is a key factor for food quality and safety (Barlow & Morgan, 2013). According to McMillin (2017), appearance, color, lipid stability, nutritive value, and palatability (texture, flavor, aroma) are significant factors that must be considered when choosing a packaging solution. Changing to bio-based packaging materials, these factors have to be considered to ensure the shelf life and quality of the food (Molina-Besch et al., 2019; Wikström et al., 2019).

So far, a broad variety of bio-based materials have been investigated to meet the purpose and achieve the properties of commercial packaging plastics. Although many new materials are used, companies worry about their physical inferiority compared to conventional polymers. Currently, European legislation and regulations are forcing companies to eliminate or reduce conventional plastic in packaging (COM/2018/028 final) (Matthews et al., 2021). Even if there is a desire to change, there is a conflicting pressure that prohibits changes in packaging because of different attitudes along the supply chain. The companies are faced with a challenge of alternatives offering higher costs and lower functionality, existing infrastructure, and inconsistent legislation (Garcia-Arce et al., 2016; X. Ma et al., 2020).

Today, many companies are trying to use materials that are more recyclable instead of using bio-based materials. According to the adopted “European strategy for plastics in a circular economy” (COM/2018/028 final) where sustainability is the underlying motivation, recycling of plastic packaging is a key factor (Matthews et al., 2021). Recycling is viewed as the primary mechanism to reduce the environmental and waste management issues that are related especially to the use of conventional plastic (Dilkes-Hoffman et al., 2018). Therefore, sorting and recycling capacities have to be expanded and modernized. Industries are investing in research and innovation activities to develop new technologies that support and increase the recovery of plastic packaging material (European Commission, 2018; Hahladakis & Iacovidou, 2018; Mulakkal et al., 2021).

As recently reported, in a circular economy, resources, materials, and products remain as long as possible on the market, minimizing waste and resources. This results in major economic benefits, innovation, and growth. However, safe disposal and recycling of materials often remain challenging. Reasons are poor management and enforcement, regulatory disparities, lack of infrastructure, and high cost of waste recycle systems (Taleb & Al Farooque, 2021).

Although many polymers are recyclable, due to additives and related quality issues, recycling rates remain low (Hahladakis & Iacovidou, 2018). Currently, the recycling rate of packaging waste in the EU reaches 67 % in total (Statista, 2020a), 42 % of plastic (packaging waste) (Statista, 2019), and 72 % of paper and cardboard (Statista, 2020b), respectively. Until 2025, at least 65 % by weight of all packaging waste has to be recycled. Regarding specific materials contained in packaging waste, 50 % of plastics and 75 % of paper and cardboard are the target rates for recycling. By 2030 the recycling rate of all packaging waste rises to 70 %, and for plastic and paper and cardboard the targets are 55 % and 85 %, respectively (European Union, 2018).

Moreover, the multi-layer structure makes plastic waste one of the most complex material mixture from a recycling perspective (K. Ragaert et al., 2017). Recycling of these materials is accompanied by either high costs, technical difficulties regarding the separation process of the different polymers, or the inability to recycle mixed polymers (Dilkes-Hoffman et al., 2018; Matthews et al., 2021). Current recycling technologies for processing and handling solid plastic waste streams include gasification as a thermo-chemical conversion process for the recycling of polymeric composites, pyrolysis, fluid catalytic cracking, hydrogen technologies (hydrocracking and IH^2 process), and the catalytic pressure-less depolymerization process (Abdou et al., 2021; Deng et al., 2020; Khui et al., 2021; Mumbach et al., 2020; K. Ragaert et al., 2017; Sun et al., 2015).

The purpose of future redesign and waste management is the reduction of the amount of plastic that is accumulated in the environment and disposed on landfills, especially in developing countries (Ingrao et al., 2017; White & Lockyer, 2020). So, further innovations in both recyclable packaging designs and corresponding cost-effective technologies are needed – independent of the material origin (natural or artificial polymers) (Matthews et al., 2021).

2.3.2 Food waste and the meaning of packaging

Food wastage and loss describe a major concern in the food supply chain that takes all of the involved stakeholders into consideration (Bhat & Jõudu, 2019). There is an increasing concern about the amount of food waste in Europe as wasted food has a significant impact on the use of natural resources and the environment (United Nations, 2015). The United Nations set a target of halving the actual amount of global food waste per capita at retail and consumer levels and reducing food losses along production and food supply chains as part of their sustainable development goals for 2030 (goal 12.3 of the UN General Assembly) (United Nations, 2015).

Differentiating between avoidable (edible) food waste and unavoidable (non-edible) food waste, proper waste management, and recycling strategies is required to reduce unavoidable food waste. Various chemical and biological processes can be used to convert food waste into bio-commodity chemicals and bio-energy (Mak et al., 2020).

Packaging prevents avoidable food waste and has the potential to further decrease it (Wikström et al., 2019). According to a study of Bruckner et al. (2012), the shelf life of poultry under aerobe packaging conditions at 4°C accounts for 98.6 h. At 4°C, the shelf life of poultry packed under modified atmosphere packaging (70 % oxygen (O_2)/30 % carbon dioxide (CO_2)) is prolonged to 228 h (Herbert et al., 2015). The kind of packaging has a high impact on the shelf life of poultry and can more than double it.

Caldeira et al. (2019) focused on food waste generated in the EU for the major food groups: sugar beets, oil crops, potatoes, vegetables, fruit, cereals, meat, fish, dairy, and eggs. The food waste generated at each stage of the food supply chain was quantified. In total around 638 mega tons (Mt) primary foods result in approximately 129 Mt of food waste generated along the food supply chain. Fruits and vegetables were the food groups presenting the highest amount of food waste overall, with similar amounts generated at the primary production and consumption stages

(Caldeira et al., 2019). Products of food categories with a relatively short shelf life, like fresh meat, tend to be the most wasteful products (Mena et al., 2011; Rossaint & Kreyenschmidt, 2014).

The amount of wasted food means that not only the products themselves are lost but also a high amount of primary resources of fuel, land, and water, including resources needed for breeding and fattening of animals, for cultivation of plants, and raw materials for processing and packaging during production as well as along the entire supply chain (Rossaint & Kreyenschmidt, 2014; Scherhaufer et al., 2018). Therefore, for a packaging system, it is important to find a balance between the environmental impact of the package itself, on the one hand, and the impact originating from the potential loss of the packaged product, on the other (Scherhaufer et al., 2018; Williams & Wikström, 2011).

Considering the environmental impact, this is much higher for producing the food itself than the (multi-layer) plastic packaging. Therefore, if food waste occurs, the negative overall environmental impact rises with every step in the supply chain due to more used resources (Heller et al., 2019; Matthews et al., 2021; Scherhaufer et al., 2018). An analysis of the food supply chain and the points where food waste is generated showed that reducing packaging is important, but it must still fulfill its duty of protection as the main criteria for sustainability. Otherwise, the supply chain overall will be less sustainable (Heller et al., 2019; Pauer et al., 2020; Verghese et al., 2015).

Steinbuchel (2003) reported that the production of starch plastic granules requires 25 - 75 % less energy, and GHG emissions are reduced about 20 – 80 % compared to poly(ethylene) (PE). Weiss et al. (2012) reviewed the environmental impacts of bio-based materials in a meta-analysis of LCA data. Therefore, one metric ton (t) of bio-based materials saves 55 ± 34 gigajoules (GJ) of primary energy and 3 ± 1 t carbon dioxide equivalents of GHG emissions relative to conventional materials (Weiss et al., 2012).

Conte et al. (2015) assessed the environmental impact of single-layer and multi-layer conventional packaging. The results show that multi-layer surpass single-layer materials by environmental impact when food waste is included in the system boundaries (Conte et al., 2015).

Pettersen et al. (2020) studied the possibility of packaging chicken fillets in recyclable mono-layer materials (high-density polyethylene (HDPE)) instead of complex multi-layered materials (amorphous polyethylene terephthalate (APET)/(PE)) as a replacement for more sustainable packaging system without decreasing the quality of fresh chicken fillets. The results show that a competitive quality and shelf life can be obtained (Pettersen et al., 2020).

H. Zhang et al. (2015) focused on a case study, based on LCA data, where the ability of active packaging to minimize food losses by using thymol/carvacrol-enabled active packaging for fresh beef was investigated. Different scenarios have been considered in terms of overall environmental performance of the food and packaging system, including the effect of food loss reduction by using active packaging. It was shown that a breakeven point can be achieved considering the evaluated impact categories in the scenario using the best-performance active packaging whereas differences were observed between the impact categories. The breakeven point can be achieved as early as 0.1 % food loss elimination occurs, whereas in the case of cumulative energy demand (fossil), it required more effort to reach the breakeven point. In this case, the active packaging performance needs to reduce food losses at least by 0.6 % (H. Zhang et al., 2015).

2.4 Plastics used for food packaging

Within the last decades, a broad variety of polymers prepared from renewable resources have been studied as potential substitute for conventional packaging plastics (X. Ma et al., 2020). The European Committee of the Regions stated that further research on the relation between packaging and food preservation on a life cycle basis is needed, and that possible alternative approaches to prevent food waste without the use of fossil-based (complex) plastics has to be investigated (van de Nadort, 2018). Increasing the exploitation efficiency of natural resources plays an important part on the way to a circular economy (Worrell et al., 2016). Next to circularity, sustainable packaging should be safe for the environment and humans (Verghese et al., 2012). The idea of biodegradable polymers, particularly obtained from renewable sources, stems from the need to close the natural cycle of matter (S. Z. Popović et al., 2018). Bio-based applications can be a useful replacement considering the biodegradability, biocompatibility, and recyclability (Dilkes-Hoffman et al., 2018; W. Wang et al., 2021; Xia et al., 2019).

Different studies show a limited but growing number of natural polymers used as films and coatings applied for food packaging (Asgher, Urooj, et al., 2020; Biscarat et al., 2015). So far, their practical use depending on the material is restricted due to lower performance in fundamental packaging functions. Challenges such as relatively poor thermal, mechanical, and rheological properties; higher costs; lack of compatibility with the processing and recycling systems currently available; or perceived environmental issues of natural polymers must be overcome. In addition, the barrier properties of natural polymers, especially the moisture barrier properties due to the hydrophilic nature of these polymers, are detrimental to existing packaging materials (Aggarwal & Langowski, 2020; Cazón et al., 2017; N. Kumar et al., 2017; S. Thakur et al., 2018).

The WVP is important for fresh foods where dehydration and absorption of moisture should be avoided (Bahrami et al., 2019; Vejdan et al., 2016). In general, the water vapor permeability is affected by several factors: chemical structure of macromolecules, degree of cross-linking, crystallinity and porosity, comparative humidity, and the addition of a plasticizer (Davachi & Shekarabi, 2018). Oxygen permeability is another fundamental parameter of food packaging material. Low values in oxygen permeability are aimed to prevent deterioration in food quality (Saral Sarojini et al., 2019).

To contribute to the reduction of packaging waste by preservation of fresh foods and to enhance their applications, currently, most natural polymers are mixed or blended with synthetic compounds such as PLA, poly(caprolactone) (PCL), and poly(hydroxybutyrate) (PHB). Furthermore, lightweight polysaccharide-based nanomaterials that could replace traditional plastic packaging are shown to improve antimicrobial activity, thermal, mechanical, and gas barrier properties while retaining the biodegradable and non-toxic characteristics of polysaccharides such as chitosan, carboxymethyl cellulose (CMC), and starch (Asgher, Arshad, et al., 2020; Kadam et al., 2013; N. Kumar et al., 2017; P. Ma et al., 2012; Malathi et al., 2014; Marra et al., 2016; Youssef et al., 2017).

2.4.1 Classification of plastics

Currently, the food packaging industry depends on fossil-based plastics which in turn originate from a finite raw material feedstock (Matthews et al., 2021). The finite resources issue induces a movement towards reducing the usage of virgin plastics towards a plastic production based on alternative raw materials such as renewable resources and biomass waste that have the potential to become plastic alternatives (T. Ahmed et al., 2018; X. Ma et al., 2020).

Next to conventional plastics that are fossil-based and non-biodegradable (e.g., PE, PP, poly(ethylene terephthalate) (PET), “bioplastics” were developed. According to the European Bioplastics association, “a plastic material is defined as a bioplastic if it is either bio-based, biodegradable, or features both properties” (European Bioplastics, 2018). Thus, bioplastics involve a range of materials that show different properties and applications (R. Sharma & Ghoshal, 2018). Figure 2.1 illustrates the categories of the plastics used for food packaging applications.

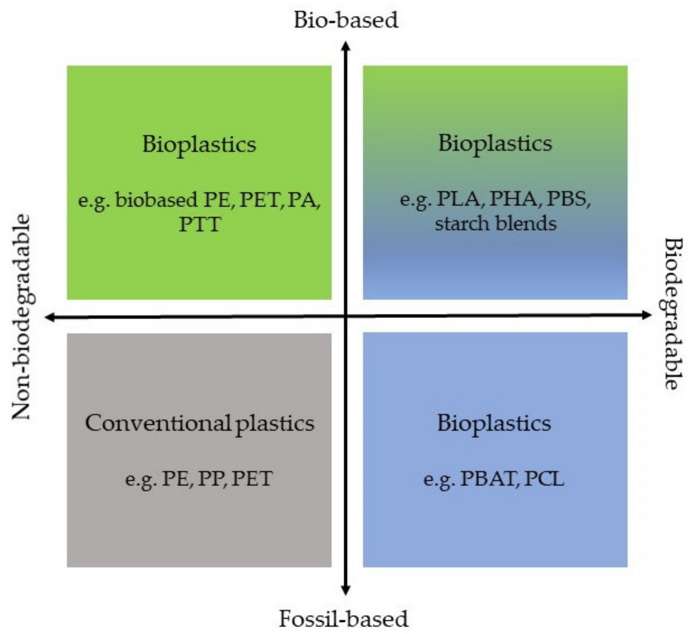


Figure 2.1 Classification of plastics used for packaging applications. Adapted from European Bioplastics (2018). Poly(amide) (PA), poly(butylene adipate terephthalate) (PBAT), poly(butylene succinate) (PBS), poly(caprolactone) (PCL), poly(ethylene) (PE), poly(ethylene terephthalate) (PET), poly(hydroxyalkanoate) (PHA), poly(lactic acid) (PLA), poly(propylene) (PP), poly(trimethylene terephthalate) (PTT).

Thus, biodegradability (and even more compostability) is considered as a useful characteristic providing one option to reduce plastic waste. Biodegradation occurs when a product undergoes a significant change in chemical structure under specific environmental conditions. Biodegradable polymers can, for example, be decomposed to natural substances (CO_2 or methane (CH_4) and water (H_2O)) by microorganisms that are found in the environment like algae, fungi, and bacteria (P. Ma et al., 2012; Matthews et al., 2021; R. Sharma & Ghoshal, 2018; WRAP, 2018). As biodegradation depends on the chemical structure of the material compound rather than on its origin, the basis of biodegradable plastics are not necessarily renewable resources (Lambert & Wagner, 2017; R. Sharma & Ghoshal, 2018).

Biopolymers such as proteins, polysaccharides, and lignin are natural polymers produced by the cells of living organisms (e.g., forestry and agricultural crops, terrestrial and marine animals) (Daioglou et al., 2015; Chunping Xu et al., 2014). These biopolymers can be used for the manufacturing of packaging materials and therefore have a high potential to replace synthetic plastics (Peelman et al., 2013; Saha et al., 2020). Most common biopolymers currently used for food packaging applications are synthesized (Kamdem et al., 2019; N. Kumar et al., 2017; Rhim et al., 2013).

In the following subchapters, the review focuses on different groups of bio-based polymers and their characteristics regarding food packaging applications starting with synthetically manufactured polymers using natural monomers (chapter 2.4.2) followed by polymers isolated from renewable resources (chapter 2.4.3).

2.4.2 Selected biodegradable synthetically manufactured polymers

2.4.2.1 Biomass-derived chemically manufactured polymers

One commercially manufactured example should be discussed here: poly (lactic acid) (PLA) and related products. PLA is a thermoplastic and biodegradable aliphatic polyester (Figure 2.2). As it has its roots in the aliphatic class of polyesters, PLA can be created either by chemical processing of lactic acid monomer or by fermentation of a carbohydrate (Tawakkal et al., 2014).

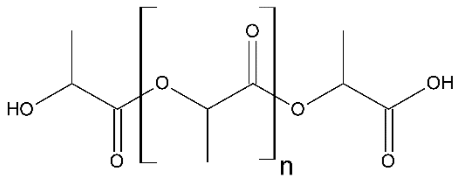


Figure 2.2 PLA structure.

PLA is the first polymer synthesized from bio-based monomers commercialized on a large scale and can be shaped into injection molded objects, films, coatings, and 3D printed materials (Rasal et al., 2010). Next to PHAs, starch, and PCL, PLA is the primary biodegradable polymer used for mono-layer and some multi-layer applications (Byun & Kim, 2014; N. Kumar et al., 2017). So, PLA films are applied as thermoformed trays, cups, bowls, bags, or jars for packaging of fresh salads, ready-to-eat meals, deli products, beverages, potato chips, and yoghurt among other uses (Jabeen et al., 2015; Zinoviadou et al., 2016). Renewability, biodegradability, and biocompatibility are attributes that make PLA one of the best polymeric substitutes for various fossil-based polymers (N. Kumar et al., 2017). However, the PLA synthesis - and in turn the corresponding products - are still rather expensive (van den Oever et al., 2017).

So far, PLA and corresponding copolymers are used to substitute polyolefins as high-density poly(ethylene) (HDPE), low-density poly(ethylene) (LDPE), poly(propylene) (PP), PET, and poly(styrene) (PS) as packaging materials (N. Kumar et al., 2017; Marra et al., 2016) due to comparable mechanical properties like stiffness and tensile strength, gas permeability, and transparency (Table 2.1) (Ahmadzadeh & Khaneghah, 2020; Auras et al., 2004; Benetto et al., 2015).

Table 2.1 Barrier and mechanical properties of selected synthetic polymers and biomass-derived chemically synthesized materials.

Film Composition	Permeability			Mechanical Properties		References
	O_2	CO_2	H_2O Vapor	Tensile Strength	Elongation at Break	
	[$cm^3 \cdot mm/(m^2 \cdot d \cdot atm)$]	[$cm^3 \cdot mm/(m^2 \cdot d \cdot atm)$]	[$g \cdot mm/(m^2 \cdot d \cdot atm)$]	[MPa]	[%]	
Synthetic Polymers						
HDPE	44 – 91 (20°C, 65 % RH)	100 (23°C, 0 % RH)	0.15 (38°C, 90 % RH)	32	150	Crompton (2012); P. Ragaert et al. (2019)
LPDE	98 –183 (23°C, 50 % RH)	NR	0.44 (38°C, 90 % RH)	10	400	Crompton (2012); P. Ragaert et al. (2019)
PE	50 – 200 (23°C, 50 % or 0 % RH)	100 – 1000 (23-25°C, unknown RH)	0.5 – 2 (23°C, 85 % RH)	18	350	Crompton (2012); Lange & Wyser (2003); Massey (2003)
PET	1 – 5 (23°C, 50 % or 0 % RH)	3 – 7 (23°C, 75 % RH)	0.5 – 2 (23°C, 85 % RH)	55	300	Crompton (2012); Lange & Wyser (2003); Massey (2003)
PP	50 – 100 (23°C, 50 % or 0 % RH)	200 – 900 (unknown conditions)	0.2 - 0.4 (23°C, 85 % RH)	26	80	Crompton (2012); Lange & Wyser (2003); Massey (2003)
Biomass-derived chemically synthesized materials						
PLA	3.5 – 15 (23°C, 50 % or 0 % RH)	32.9 – 72 (23°C, 0 % RH)	1.6 - 3.6 (38°C, 85 % RH)	50.45	3	Drieskens et al. (2009); P. Ragaert et al. (2019); Rhim et al. (2009)

NR: Data not reported.

The production of composites by adding nanofillers is a way to extend and improve the properties of PLA (Ahmadzadeh & Khaneghah, 2020; Byun & Kim, 2014; N. Kumar et al., 2017). The addition of many nanofillers (three-dimensional spherical and polyhedral, two-dimensional nanofibers or one-dimensional sheet-like nanoparticles) has been studied and lead to satisfactory achievements in the design of PLA nanocomposites (Raquez et al., 2013).

Panseri et al. (2018) studied the effectiveness of PLA-based packaging solutions compared to a conventional reference package consisting of APET/PET trays wrapped in plastic films of poly(vinyl chloride) (PVC) to store red fresh meat during its refrigerated shelf life. By using PLA packaging in combination with a gas mixture of 66 % O_2, 25 % CO_2, and 9 % N_2, it was possible to maintain an optimum red color together with a reduced content of volatile compounds associated to off-flavors of meat samples (Panseri et al., 2018).

Marra et al. (2016) investigated biocomposite films of PLA with zinc oxide regarding mechanical, barrier, and antimicrobial properties. The results showed that PLA films with 5 wt % of zinc oxide exhibit good mechanical properties related to a high modulus and stress at yielding, decrease of permeability to carbon dioxide and oxygen, and a slight increase of water vapor permeability. Furthermore, the incorporation of 5 % zinc oxide leads to an antimicrobial activity against *E. coli* after 24 h with a reduction value of 99.99 % (Marra et al., 2016). Vanitha & Kavitha (2021) incorporated cellulose natural fibers from palm sprouts in a PLA matrix. The results showed that the mechanical resistance increased, and the water absorption rate decreased significantly with the optimum concentration of palm sprouts fiber in the PLA-film. Interactions between palm sprout and PLA restrict the water infiltration (Vanitha & Kavitha, 2021).

The incorporation of lignin in PLA films via simple blending results in a small but significant increase of the oxygen barrier properties, as well as an improved antiradical efficiency that increases with the severity of the heat treatment of the blends (Domenek et al., 2013). Moreover, the water sorption capacity decreased with an increase of lignin loading from 7 wt % to 15 wt % while tensile strength increased, as shown in a study of Spiridon & Tanase (2018). Gordobil et al. (2014) used commercial alkaline lignin and organosolv lignin from almond shells as PLA filler, which greatly improved the thermal stability and increased the elongation at break. Low percentages up to 1 % unmodified lignin did not affect the maximum strain, while it was decreased with increasing lignin content at percentages greater than 5 % (Gordobil et al., 2014). In addition, kraft and organosolv lignin were examined as nucleating agents, showing that both lignins induce heterogeneous nucleation and increase the crystallization rate in PLA by shortening the crystallization half time and increasing the degree of crystallinity in PLA, while not affecting the processing window of the polymer (Kovalcik et al., 2017). One problem when incorporating lignin is its compatibility with PLA, which can be overcome with the addition of triallyl isocyanurate (TAIC), leading to the formation of PLA-TAIC-lignin cross-linked structures as interface, improving the compatibility in the blend and thus the mechanical, thermal, and hydrolytic degradation properties (A. Kumar et al., 2019). However, using TAIC, the biocompatibility has to be studied before applying it for food packaging. Another possibility is the introduction of lignin nanoparticles (LNP) into PLA using a Pickering emulsion template method where lignin acts as stabilizer. According to the results of this study, lignin could increase the decomposition temperature by approx. 10 %, reduce the light transmission in the UV region, and increase the Young's modulus but also decrease the tensile strength and elongation at break. Moreover, the crystallinity of PLA could be improved with the addition of lignin (X. Li et al., 2019). In another approach, LNP in PLA films could inhibit the growth of bacterial plant pathogens and showed a high antioxidant activity, while migration values remained below the legislative limits, suggesting the exploitation of LNP-PLA films as food packaging material (W. Yang, Fortunati, et al., 2016a).

The combination of LNPs with another lignocellulosic nanofiller, namely, cellulose nanocrystals (CNC), in PLA films can in fact improve UV light blocking capability and strength and modulus values compared to neat PLA or PLA binary systems, confirming a synergic effect of LNP and CNC (W. Yang, Fortunati, et al., 2016b). This was also reported in another study, where Young's modulus, elongation at break, and toughness of neat PLA films were improved by 14 %, 77 %, and 30 %, respectively, by incorporation of high lignin-containing cellulose nanocrystals. In contrast, commercial lignin-coated CNCs showed inferior crystallinity, smaller surface area, and a higher degree of agglomeration, concluding that the presence of LNPs is important for the compatibility between the PLA polymer matrix and CNCs (Wei et al., 2018).

2.4.2.2 Polymers produced by microorganisms

Here, we discuss the most prominent representatives of polymers produced by microorganisms: PHA and PHB and corresponding copolymers or composites. In the group of PHAs, more than 100 known bio-derived polymers exist. The most common ones are PHB and corresponding copolymers such as poly(3-hydroxybutyrate-co-3-hydroxy-valerate) (PHBV) and poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx) (Figure 2.3) (Armentano et al., 2015; P. Ragaert et al., 2019).

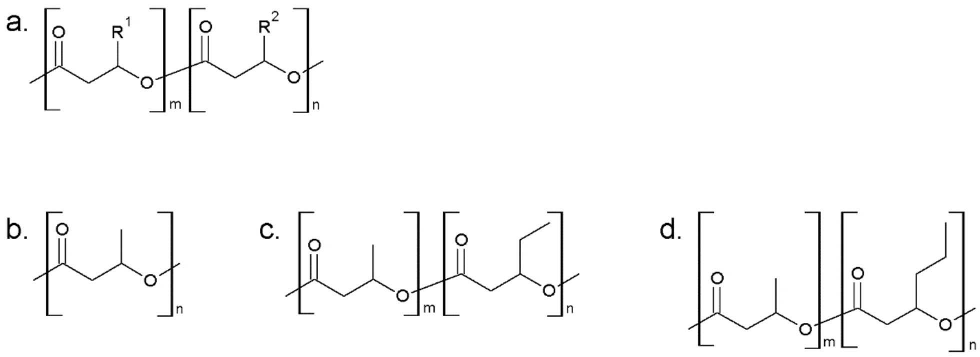


Figure 2.3 Overview of several poly(hydroxyl alkanoate)s (PHA) used for food packaging applications: (a) general structure of PHAs with residues R1/R2 as alkyl chains ranging from 1-13 carbons, (b) poly(hydroxyl butyrate) (PHB), (c) Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), and (d) poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx).

In general, PHAs can be used to coat paper or paperboard to produce water-resistant surfaces, making the coated material completely biodegradable. PHB-coated paperboard has been used for packaging of ready meals, while PHBV-coated paperboard has been used for dry products, dairy products, and beverages (Andersson, 2008). In addition to functionalizing the surface of fiber-based materials, PHAs can also functionalize paper and board’s grease resistance and sealability (P. Ragaert et al., 2019). PHAs involve a range of biodegradable thermoplastic polymers that are produced through fermentation by different microorganisms (Peelman et al., 2013). These polymers are characterized by thermomechanical properties that are similar to synthetic polymers such as PP (Alavi et al., 2014). PHAs can be processed into different products including films, trays, and coatings on other bio-based materials (e.g., paperboard) (P. Ragaert et al., 2019).

Initially, PHAs were used to make everyday articles like shampoo bottles. Moreover, they are used to produce carrier bags, containers and paper coatings, biodegradable bags, and lids. Currently, their use in terms of packaging applications is restricted as they are not transparent (P. Ragaert et al., 2019; Zinoviadou et al., 2016). Their gas and water vapor permeability offer opportunities to be applied as food packaging materials. Copolymerization as well as blending are used to improve physical-mechanical properties of PHAs (P. Ragaert et al., 2019).

Several studies have shown that PHB, PHBV, and PHBHHx films are promising materials for food packaging due to their good barrier properties. The oxygen permeability of PHAs is comparable to PET and PLA. The values are much lower compared to conventional polymers such as PE and PP. The water vapor permeability of PHAs is similar to materials such as PET and PLA but slightly higher than more apolar polymers such as PE and PP. The carbon dioxide permeability of PHAs is higher compared to PET but substantially lower than for common packaging materials such as PP and PE (Table 2.2) (Farmahini-Farahani et al., 2017; Kovalcik et al., 2015; Maes et al., 2018; Siracusa et al., 2012; Siracusa et al., 2017; Vandewijngaarden et al., 2014).

Table 2.2 Barrier and mechanical properties of bio-based polymers produced by microorganisms.

Film Composition	**Permeability**			**Mechanical Properties**		**References**
	O_2	**CO_2**	**H_2O Vapor**	**Tensile Strength**	**Elongation at Break**	
	[$cm^3 \cdot mm/(m^2 \cdot d \cdot atm)$]	**[$cm^3 \cdot mm/(m^2 \cdot d \cdot atm)$]**	**[$g \cdot mm/(m^2 \cdot d \cdot atm)$]**	**[MPa]**	**[%]**	
PHB (P3HB)	2 – 11.4 (23°C, 0 % RH)	3 – 28.9 (23°C, 0 % RH)	1 – 5 (unknown conditions)	35 – 40	3 – 8	Anjum et al. (2016); P. Ragaert et al. (2019)
PHBV (P(3HB-co-3HV))	4.9 – 16.7 (25°C, 0 % RH)	146 (25°C, 0 % RH)	1.5 (38°C, 90 % RH)	38	NR	Anjum et al. (2016); P. Ragaert et al. (2019)
PHBHHx	8.3 (23°C, 0 % RH)	54 (23°C, 0 % RH)	1.42 (23°C, 0 % RH)	20	850	Anjum et al. (2016); Vandewijngaarden et al. (2014)

NR: Data not reported.

Dilkes-Hoffman et al. (2018) summarized that a combination of PHA with thermoplastic starch (TPS), which provides one of the best oxygen barriers of all polymeric materials, seems to have the potential to lower food spoilage rates compared to conventional packaging materials according to good barrier properties.

PHB shows a high crystallinity and a high melting point. Therefore, PHB is often blended with PLA. This results in materials of improved mechanical, thermal, and physical properties compared to neat PLA (Armentano et al., 2015). Arrieta et al. (2014) figured out that blending PLA with 25% (w/w) PHB resulted in improved oxygen and barrier properties, whilst the inherent transparency of PLA was reduced.

Kovalcik et al. (2015) studied the melting and crystallization behavior, thermo-oxidative stability, mechanical and viscoelastic properties, and permeability for oxygen and carbon dioxide of composite materials of microbial PHBHV with methanol fractionated kraft lignin. The results showed that a concentration of already 1 wt % of methanol-extracted kraft lignin can act as an active agent for decreasing the oxygen as well as carbon dioxide permeability of PHBHV films. The gas permeability was decreased for oxygen by 77 % and by 91 % for carbon dioxide, respectively, compared to the native PHBHV film. The low thermo-oxidative stability of pure PHBHV was increased for the lignin-containing films. Based on this results, methanol-extracted kraft lignin is suggested as a suitable active additive in PHBHV films for applications, especially in the field of food packaging (Kovalcik et al., 2015).

Although, high production costs limit the competitiveness in commercial applications, PHAs might have high potential as bio-based and biodegradable plastic packaging materials in the transition towards a circular economy (P. Ragaert et al., 2019).

2.4.3 Selected plant-derived polymers

2.4.3.1 Lignocellulosic biomass and lignin

Lignocellulosic biomass is the major structural component of plants, mainly consisting of cellulose (40 - 60 %), hemicellulose (10 - 40 %), and lignin (15 - 30 %), whereby the latter one is the most complex constituent (Schutyser et al., 2018). Lignin is a randomly cross-linked macromolecule composed of the three monolignols *p*-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol, which form the residues *p*-hydroxyphenyl (H), guaiacyl (G), and syringyl (S), respectively, as shown in Figure 2.4 (Rumpf et al., 2020). It can be obtained from woody biomass (e.g., pine, poplar, birch), annual plants (e.g., wheat straw, miscanthus, switchgrass), or agricultural residues (e.g., sugarcane bagasse) by various extraction processes. The molecular structure of lignins strongly depends on the botanical origin but also on the growing site, season, and isolation process (Rinaldi et al., 2016; Chunbao Xu & Ferdosian, 2017). There are different types of technical lignins that can either be classified as sulfur-containing or sulfur-free. The most common ones are lignosulfonates, kraft lignin, organosolv lignin, and soda lignin (Schutyser et al., 2018). Most industrial lignins originate from the pulp and paper industry with up to 90 million tons of kraft lignin released per year worldwide, though only 2 % of it are used commercially for value-added products (Tribot et al., 2019). One reason for that might be the deficient quality or missing specifications of technical lignins, as they are rather undefined products with a complex composition and impurities from the pulping process (such as remaining sugars or thiol groups). So far, this restricts their industrial exploitation, as resulting products have varying properties, which are inferior to fossil-based products.

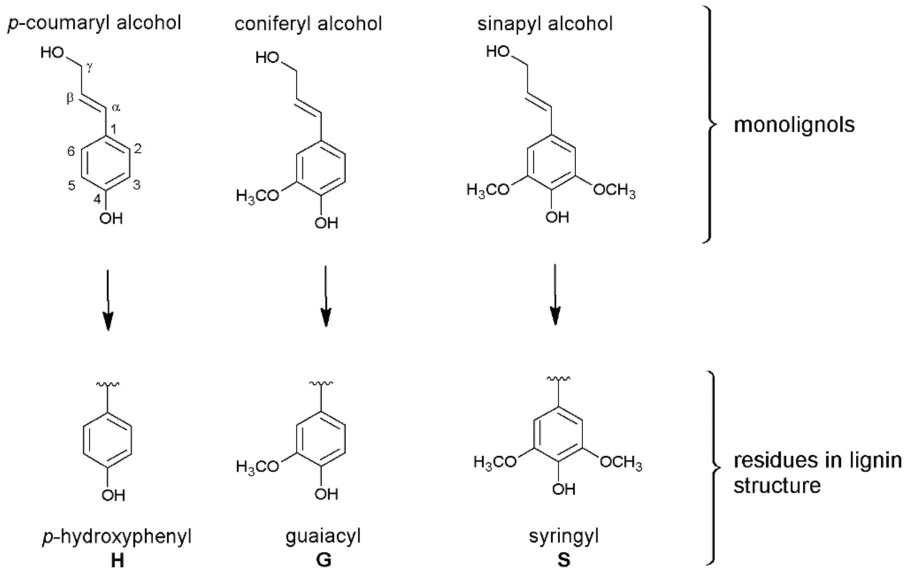


Figure 2.4 Lignin monolignol structures: p-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol forming the specific residues p-hydroxylphenyl (H), guaiacyl (G), and syringyl (S) (Rumpf et al., 2020).

Nevertheless, in consideration of the global energy crisis and the depletion of fossil fuels and petrochemicals, the potential of lignin has been a key topic in biorefinery research (S. S. Hassan et al., 2019; K. H. Kim et al., 2021; Renders et al., 2019; Schutyser et al., 2018). As polyphenols, lignins possess numerous interesting functional properties, such as antioxidant activity, and thus, they are investigated as active packaging materials for the protection of light- or oxygen-sensitive goods (Domenek et al., 2013). Antioxidant polymers are a field of great interest, as the use of macromolecular antioxidants is related to the possibility to produce materials with long-term stability. Due to its cross-linked 3D structure and enzymatic resilience, lignin possesses a higher thermal and biological stability compared to low molecular weight compounds, and thus, could be used in special fields where the exploitation of low-molecular antioxidant substances would be inefficient due to their higher diffusion rates. Moreover, carcinogenic effects have been observed for synthetic antioxidants: butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT), for example, are able to cause cytotoxicity and carcinogenesis as shown in *in vivo* studies (Espinoza-Acosta et al., 2016). Azadfar et al. (2015) have already shown that lignin has the potential to serve as raw material for antioxidants like guaiacol and 4-vinylguaiacol, whereby their antioxidant activity is comparable to that of commercial antioxidants.

Consequently, lignin has gained increasing interest as environmentally benign antioxidant and its ability to improve mechanical, thermal, and barrier properties when incorporated in conventional packaging films. In general, a reduction in WVP of lignin-based films is explained by the hydrophobicity of lignin. It acts as a barrier in the polymeric matrix and increases the path for the diffusion of water vapor, resulting in lower permeation of water molecules through the polymer film (Michelin et al., 2020). Next to the utilization of lignin as additive/blend in different polymer matrices, it could also be used as raw material for the development of polymeric packaging materials, e.g., as polyol substitute in polyurethanes or polyesters or as phenol substitute in resins. Hult et al. (2013) investigated softwood lignin esterified with tall oil fatty acid as coating on paper board. The thermoplastic properties of lignin were enhanced, and the water vapor and oxygen transmission rate decreased while tensile strength was not affected (Hult et al., 2013). Polyurethanes of high transparency and flexibility for construction or packaging applications were prepared by S. E. Klein, Rumpf, et al. (2019), where petroleum-based polyols could be substituted with kraft lignin up to 80 wt %. In addition, demethylated lignins were also used to enhance the reaction selectivity towards polyurethane formation (S. E. Klein, Alzagameem, et al., 2019). In a recently published study by Hao et al. (2019) thermoset coatings with integrated self-healing and removal properties were investigated. They investigated a kraft lignin functionalized with carboxylic acid groups as curing agent with poly(ethylene glycol) diglycidyl ether, which resulted in a cross-linked structure (Hao et al., 2019). Hambardzumyan et al. (2015) designed novel nanocomposite films of lignin and CNC that could be self-supported or used as coatings. This combination of the antibacterial properties of lignin and oxygen barrier properties of CNC films are promising as food packaging material (Hambardzumyan et al., 2015).

Rastogi & Samyn (2015) summarized different possibilities for bio-based paper coatings. Next to polyester and polysaccharides discussed so far, also lipids and proteins could be used (Rastogi & Samyn, 2015). Nevertheless, a full exploitation at industrial scale is not possible, due to different crystallization behavior, brittleness, or melt instabilities that lead to difficulties in processing of these biopolymers. Blending the lipids and proteins with other biopolymers, such as lignin, may provide a route to overcoming this obstacle.

Lignin cannot only be used in paper coatings but also can be used to improve the strength of paperboard. Flory et al. (2013) developed a green binder system with *Salix* lignin that was equal to the wet tear strength of the commercial vinyl acetate binder. Inspired by the reinforcement principle of lignin and cellulose in wood, Jiang et al. (2020) developed a cellulose fiber scaffold with lignin as reinforced matrix via successive infiltration and mechanical hot-pressing treatments. The resulting composite shows a high isotropic tensile strength of 200 MPa, compared to 40 MPa of conventional cellulose paper, and Young's modulus of 10 GPa, which is even higher than many fossil-based plastics. In addition, also the thermostability and UV-blocking performance is enhanced due to the lignin addition (Jiang et al., 2020).

In conclusion, lignins are promising candidates for environmentally benign antioxidants with a great abundance (Alzagameem et al., 2019; Alzagameem, El Khaldi-Hansen, Büchner, et al., 2018; Alzagameem, El Khaldi-Hansen, Kamm, & Schulze, 2018). Regarding the studies summarized in this review, lignin use as an additive seems more favorable than its copolymerization, as copolymerization usually requires at least one functionalization (such as demethylation or carboxylation). Thus, further effort is necessary to get a deeper understanding of the lignin structure and the processing conditions required to maintain and enhance the antioxidant and antimicrobial properties.

2.4.3.2 Protein-based polymers

Research in food packaging has also focused on protein-based films due to their good film-forming properties, low cost, and biodegradable nature (N. Kumar et al., 2017). Materials synthesized from proteins exhibit desirable film-forming and barrier properties, which are often comparable to fossil-based products (Kadam et al., 2013). Proteins from different sources have been investigated for the synthesis of bioplastic films for packaging applications including collagen, gelatin, caseins, soy/whey/quinoa protein, egg white protein, myofibrillar protein, corn zein, wheat gluten, and keratin (Galus & Kadzińska, 2015; N. Kumar et al., 2017; Otoni et al., 2016).

Among all the protein sources, soy proteins got great attention as a potential source for bio-based packaging materials. This development is based on excellent film-forming and oxygen barrier properties of films produced from soy protein isolate. However, these materials cannot meet the requirements of a film with mechanical and water barrier properties guaranteed by conventional plastics (Cho et al., 2010; N. Kumar et al., 2017). Compared to films from other proteins, soy protein-based films are characterized by transparency, flexibility, and cost-effectiveness (Otoni et al., 2016). Furthermore, they show good oxygen barrier properties under low moisture conditions (Denavi et al., 2009). A disadvantage that limits their use beside low mechanical strength is a lack of heat stability compared to LDPE (Acquah et al., 2020; S. Popović et al., 2012; Umaraw & Verma, 2017; Y. Zhang et al., 2018).

Whey proteins are able to form elastic films (Y. Wang et al., 2013), which are transparent, flexible, and exhibit good oil and oxygen barrier properties at low humidity. A disadvantage is a moderate moisture permeability. Nevertheless, whey proteins have been itensively studied as raw material for biodegradable packaging (Ramos et al., 2012).

Collagen and gelatin are proteins originating from animal sources acquired by a controlled hydrolysis reaction. In nature, collagen is the most abundant occurring protein (Fratzl, 2008). Collagen-based bioplastic films are characterized by good mechanical properties (Fadini et al., 2013), and therefore, are suitable for various applications (Oechsle et al., 2017). In contrast to coallgen-based films, gelatin films show poor mechanical and barrier properties according to their hydrophilic nature (Ciannamea et al., 2018). Biscarat et al. (2015) determined functional properties of gelatin-based films. Compared to synthetic polymers, good gas barrier properties were reached by gelatin films cross-linked with ferulic acid. Gelatin films with poly(ethylene glycol) (PEG) 200 showed high gas barrier properties and high permselectivity towards carbon dioxide and oxygen (Biscarat et al., 2015).

Gelatin has been introduced in the manufacturing of packaging films due to its low cost and abundance (Chentir et al., 2019). Furthermore, gelatin is used to produce biodegradable packaging materials due to its good properties, such as low melting and gelling points, good capacity of oxygen barrier, biodegradability, and excellent film formation (Amjadi et al., 2019). The use of gelatin-based composite films incorporating other materials like chitosan, sunflower oil, and corn oil to enhance the barrier and mechanical properties of these films was studied by different authors (Nur Hanani, Roos, & Kerry, 2014). In addition to gelatin, gluten is used to prepare films of high homogeneity, excellent gas barrier properties, and mechanical strength (Mojumdar et al., 2011).

Kanatt (2020) developed a new intelligent-active food packaging film using poly(vinyl alcohol) (PVA) and gelatin incorporated with Amaranthus leaf extract to monitor freshness and increase the shelf life of fish and chicken meat. Incorporation of Amaranthus leaf extract improved its mechanical and water vapor barrier properties next to active functions. The decrease in solubility enables the use for packaging of flesh foods. Samples packed in neat films had a shelf life of 3 days while those in active films spoiled after 12 days. The results of the study suggest the application of Amaranthus leaf extract containing PVA-gelatin films being both active and intelligent ensuring quality and safety of flesh foods (Kanatt, 2020).

For comparative purposes, Table 2.3 shows the discussed barrier as well as mechanical properties of protein-based polymers.

Table 2.3 Barrier and mechanical properties of protein-based polymers.

Film Composition	Permeability			Mechanical Properties		References
	O_2	CO_2	H_2O Vapor	Tensile Strength	Elongation at Break	
		[cm³/(m²·d·atm)]*	[g·mm/(m²·d·atm)]	[MPa]	[%]	
Soy protein isolate film with glycerol (casting method)	NR	NR	1710 (23°C, 50 % RH)	6.97	113.94	Y. Li et al. (2016)
Whey protein isolate film with glycerol	92.448 g/(m²·d) (25°C, 90 % RH)	NR	1680 (25°C, 75 % RH)	3	13	Xin Zhang et al. (2020)
Pumpkin oil cake protein isolate film with glycerol	16.06 cm³/(m²·d·atm) (23°C; unknown RH)	21.15 (23°C; unknown RH)	NR	0.86 - 6.56	22.2 - 196.61	S. Popović et al. (2012)
Fish gelatin film with glycerol	NR	NR	1040 (24°C, 50 % RH)	9.08	44.93	Syahida et al. 2020)
Beef skin gelatin films with corn oil (extrusion)	0.8 – 4.7 x 10^{-4} cm³·mm/(m²·d·atm) (23°C, 50 % RH)	NR	4.05 – 8.61 x 10^{6} (23°C, 50 % RH)	1.43 - 5.37	1.68 - 2.60	Nur Hanani, O' Mahony, et al. (2014)

*In some data, the units were normalized. NR: Data not reported.

2.4.3.3 Polysaccharides

Packaging films based on carbohydrate sources are generally transparent and homogeneous films with effective oxygen barriers at intermediate to low humidity and good mechanical properties. According to their hydrophilic character, they have poor water vapor barrier qualities, and they are relatively sensitive to moisture. Furthermore, the films obtained from several polysaccharides are brittle usually due to interactions between the polymer chains. This leads to limited applications of polysaccharide- and protein-based coatings and films (Han, 2014; Youssef & El-Sayed, 2018; Zinoviadou et al., 2016). To meet the required properties, a pretreatment such as plasticization with small molecular weight-compatible constituents, blending, or chemical modification is needed (Alavi et al., 2014). While considering mechanical properties, the tensile strength of polysaccharide-based films is similar to those of synthetic polymers; differences are observed in elongation at break (Table 2.4) (Cazón et al., 2017).

For several polysaccharides, the film-forming properties and especially their potential for edible packaging has been studied, including starch, cellulose, and its derivatives, alginate, and chitosan (Figure 2.5) (Elsabee & Abdou, 2013; Galus & Kadzińska, 2015; Jiménez et al., 2012; Q. Xu et al., 2016).

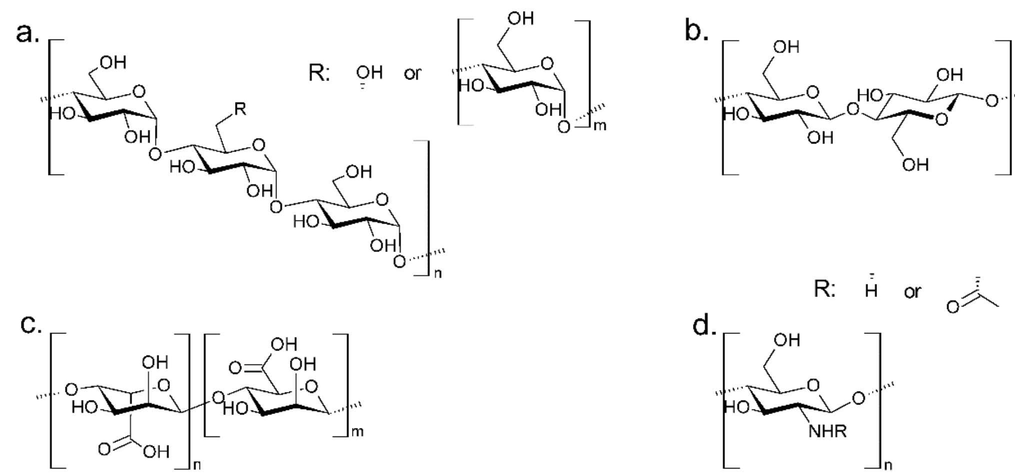


Figure 2.5 Overview of several polysaccharides used for food packaging (edible) films: (a) starch, (b) cellulose, (c) alginate, and (d) chitosan (Witzler et al., 2019).

Table 2.4 Barrier and mechanical properties of polysaccharide-based polymers.

Film Composition	Permeability: O_2 [g·mm/(m²·d·atm)]*	Permeability: H_2O Vapor [g·mm/(m²·d·atm)]*	Mechanical Properties: Tensile Strength [MPa]	Mechanical Properties: Elongation at Break [%]	References
High amylose cornstarch films without plasticizer (amylose:amylopectin ratio 80:20)	NR	1260 (20°C; 52.9 % RH)	34.32	1.41	Muscat et al. (2012)
Low amylose cornstarch films without plasticizer (amylose:amylopectin ratio 25:75)	NR	1430 (20°C; 52.9 % RH)	44.38	2.40	Muscat et al. (2012)
Thermoplastic (cassava) starch (extrusion)	0.182 (Ambient temp., 0 % RH)	36.8 (25°C, 50 % RH)	5.8	78	Dang & Yoksan (2015), Dang & Yoksan (2016)
Methylcellulose mixtures in ethanol	NR	446 – 945 (25°C; 52 % RH)	25 – 33	29 – 14	Nazan Turhan & Şahbaz (2004)
Hydroxypropyl methylcellulose without plasticizer	NR	974000 (23°C, 50 % RH)	61.04	29.51	Hay et al. (2018)
Carboxymethyl cellulose film with glycerol (casting method)	NR	683 (25°C, 52.8 % RH)	6.10	201.73	Jannatyha et al. (2020)

*In some data, the units were normalized. NR: Data not reported.

Table 2.4 Barrier and mechanical properties of polysaccharide-based polymers. *(Continued)*

Film Composition	Permeability		Mechanical Properties		References
	O_2	H_2O Vapor	Tensile Strength	Elongation at Break	
	[g·mm/(m²·d·atm)]*	[g·mm/(m²·d·atm)]*	[MPa]	[%]	
CMC-film with 50 wt% ethanol organosolv lignin with glycerol (casting method)	NR	2570 (20°C, 0 % RH)	20	5.92	Michelin et al. (2020)
Agar/10% lignin composite film with glycerol	NR	13400 (25°C, 50 % RH)	51.8	22.1	Shankar et al. (2015)
(Sodium) alginate film with glycerol (casting method)	NR	13600 (25°C, 50 % RH)	41.1	8.5	Roy & Rhim (2020)
Chitosan film with glycerol	0.188 x 10^{-2} (25°C, <50 % RH)	210 - 3020 (25°C, 0 – 100 % RH)	8.9	38.5	Kurek et al. (2014)

*In some data, the units were normalized. NR: Data not reported.

Starch. Starch is a natural polysaccharide easily available on an industrial scale. Many plant-based polysaccharides and cost-effective starch-based materials have been extensively investigated as an alternative material for fossil-based food packaging applications due to their environmental compatibility and biodegradability (B. Hassan et al., 2018). Although starch-based packaging, which has good film-forming properties and excellent oxygen barrier, is already in wide use, this material still has some disadvantages such as the poor vapor and oxygen moisture barrier and poor mechanical properties compared to conventional non-biodegradable plastics used in food packaging industry (Table 2.1 and Table 2.4) (do Val Siqueira et al., 2021; B. Hassan et al., 2018; Lumdubwong, 2019; Molavi et al., 2015).

The oxygen barrier properties are correlated to a high-ordered hydrogen-bonded network structure. The barrier properties can be improved by increasing the crystallinity or a higher content of amylopectin (Molavi et al., 2015). The poor moisture barrier is caused by a strong hydrophilic behavior (Lumdubwong, 2019). A higher crystalline structure in starch-based films leads to less sensitivity of moisture and to environmental relative humidity. Considering the poor mechanical properties, starch-based films show a relatively high tensile strength while the elongation percentage is low (Molavi et al., 2015). The high tensile strength is attributed to the extensive intra-molecular hydrogen bonds between amylose, amylopectin, and amylose-amylopectin molecules. Amorphous regions in starch-based films formed by amylose cause brittleness and thereby influence the poor mechanical properties (Muscat et al., 2012).

As starch-based films are odorless, colorless, and tasteless, starch is used in either pure or blended form as a biodegradable coating or packaging film. The coatings of edible starch are also applied for other kinds of foods to maintain quality and to extend the shelf life of products (Fakhouri et al., 2015; Lumdubwong, 2019; Ojogbo et al., 2020; R. Thakur et al., 2019). However, as starch blends may contain additives like compatibilizers and plasticizers that can migrate out of the matrix, only some starch blends are suitable for food packaging applications (Zinoviadou et al., 2016).

Starch has already been combined with lignin to improve its poor thermo-mechanical properties while simultaneously decreasing its water vapor permeability significantly (Bhat et al., 2013). Miranda, Ferreira, Magalhães, Bispo, et al. (2015) and Miranda, Ferreira, Magalhães, Santos, et al. (2015) confirmed these findings in their studies, showing that the presence of lignin in combination with CNC increased maximum stress and modulus of elasticity, barrier properties, and the thermal stability of the material. Javed et al. (2018) published a study concerning starch-based coatings for paper packaging materials with lignin, investigating the self-supporting films regarding their mechanical properties and chemical stability in water as well as their barrier properties when used as coating on paper board. When lignin is added, the dissolution of starch from the composites could be significantly decreased. The addition of ammonium zirconium carbonate (AZC) leads to further improvement of the storage modulus, indicating that cross-linking had occurred (Javed et al., 2018).

Cellulose and Derivatives. Cellulose represents the most abundant renewable polymer source available in nature. Biodegradable films made out of this raw material are characterized by renewability, low cost, non-toxicity, biocompatibility, biodegradability, and chemical stability (Sen Wang et al., 2016). For example, cellulose films (known as Cellophane®) are used for wrapping fruits in bio-based trays (Jabeen et al., 2015).

Films made of cellulose exhibit good toughness, tensile strength, high surface gloss, and good transparency (Guzman-Puyol et al., 2019). Whereas hemicellulose-based films are brittle, the flexibility, toughness, and oxygen permeability can be improved by addition of plasticizers (Mendes et al., 2017; Xueqin Zhang et al., 2020). A disadvantage of cellulose films is their poor water vapor barrier. This is caused by the underlying hydrophilic nature of polysaccharides (N. Kumar et al., 2017). Soaking cellulose with alkali to swell the structure followed by different derivatization reactions, CMC, methylcellulose (MC), hydroxypropyl cellulose (HPC), and hydroxypropylmethyl cellulose (HPMC) are available. They are used as raw materials to prepare biodegradable films, which are transparent, water soluble, odorless, tasteless, and flexible and have moderate strength and resistance to lipid compounds (Table 2.4) (Dhall, 2013).

CMC is most often used for biodegradable film production. It is highly soluble (in water) and crystalline and can build solid and flexible films (Bifani et al., 2007; Farshchi et al., 2019). Beneficial characteristics include film-forming properties, good mechanical and gas barrier properties, transparency, ease of processing, and low price (Hasheminya et al., 2018). Next to its good film-forming properties, CMC has been studied as antibacterial food packaging in composites with chitosan (Arnon et al., 2014; Lan et al., 2018) and pectin (Šešlija et al., 2018). Michelin et al. (2020) investigated the incorporation of organosolv lignin from corncob in CMC-based films, which leads to an improved water resistance of approx. 60 % and reduction in the water vapor permeability of 20 %, while also enhancing the thermal stability and antioxidant activity.

As mentioned before, mechanical and barrier properties of cellulose-based films can be improved by the production of nanocomposites. Moura et al. (2008) propesed nanocomposites using chitosan as nanofiller in HPMC to enhance mechanical and film barrier attributes. HPMC films containing different concentrations of chitosan as nanoparticles were analyzed for mechanical properties, water vapor permeability, and oxygen permeability. They realized that chitosan nanoparticles tended to fill up poriferous spaces in the HPMC-matrix. This improves film tensile properties and water vapor permeability, concluding that a HPMC-chitson nanocomposite could be a possible material for food-packaging applications to extend the shelf life of food (Alzagameem, El Khaldi-Hansen, Kamm, & Schulze, 2018; Moura et al., 2008).

Another approach is the preparation of HPMC/chitosan films (Sebti et al., 2007). Alzagameem et al. (2019) and Alzagameem, El Khaldi-Hansen, Kamm, & Schulze (2018) examined the incorporation of different lignins in HPMC and HPMC/chitosan films (Alzagameem et al., 2019; Alzagameem, El Khaldi-Hansen, Kamm, & Schulze, 2018). Results show that lignins are in general more active against Gram-positive bacteria than against Gram-negative bacteria and that films with organosolv lignin possess a higher activity against *Staph. aureus* than films with kraft lignin. It was shown that biomass as well as extraction process influence the properties of the films and that the antioxidant activity of lignins correlates with different parameters such as genotype and phenotype of biomass, pulping and purification of lignin and the resulting heterogeneity (Alzagameem, El Khaldi-Hansen, Büchner, et al., 2018).

Cellulose acetate (CA), cellulose acetate propionate (CAP), and cellulose acetate butyrate (CAB) are thermoplastic cellulosic-derived materials developed through esterification of cellulose. CA gets great attention because of its biodegradable nature, excellent optical clarity, and greater toughness (N. Kumar et al., 2017).

Alginates. Alginate and corresponding derivatives are one of the most promising carbohydrates for packaging applications, especially for foods that are sensitive to gas permeation (Küçük et al., 2020). Alginates are naturally occurring indigestible polysaccharides. They are commonly produced from various genera of brown algae (N. M. Ahmed et al., 2020). A lot of research on alginate has focused on edible coatings or to improve color and flavors (Gammariello et al., 2016; Sangsuwan & Sutthasupa, 2019).

N. M. Ahmed et al. (2020) evaluated the barrier, mechanical, and oil resistance of paper sheets coated with a novel cost-saving coating prepared at different temperatures to be used in packaging purposes. The coating is based on sodium alginate (SA) with new core-shell inorganic particles composed of waste silica fume core covered with cobalt(II) oxide/zinc oxide (CoO.ZnO) oxides. SA is broadly used due to high oil resistance and enhanced greaseproof properties.

Very recently, a binary oxide CoO.ZnO on the surface of silica was shown to decrease the penetration of oil and grease through the paper pores leading to lower oil absorption and enhanced mechanical properties. Tensile strength was decreased whereas stiffness showed slight increase in case of paper sheets coated with SA-CoO.ZnO/SiO_2. Incorporation of SA-SiO_2 and SA-CoO.ZnO/SiO_2 in the fiber matrices improved the tear and burst indices properties. The network created by SA film blended with (CoO.ZnO/SiO_2) pigments on paper sheet substrate drastically changed their mechanical and barrier properties. They are highly dense and organized, causing the creation of smooth distribution of nanoparticles and a strong surface that fills the pores in the paper matrices and increases the air, water vapor, and oil resistance of the samples coated with SA-CoO.ZnO/SiO_2 pigments compared with uncoated paper (N. M. Ahmed et al., 2020).

Chitosan. Chitosan is the second most widely found amino-polysaccharide in nature (Raafat & Sahl, 2009). It is a biopolymer with antibacterial properties that can be found in fungi, insects, crabs, and shrimps and makes up a significant part of the crustacean waste that is discarded every year (Morganti & Stoller, 2017). Chitosan is intensively investigated for biomedical applications (Witzler et al., 2018) and bioactive packaging solutions (Rai et al., 2017). Properties like being microbe resistant, biocompatible, and biodegradable make chitosan attractive for research and use in various applications (Shahid-Ul-Islam & Butola, 2019). In food packaging, potential applications of chitosan blend-based films are for fresh products (vegetable, meat, and fish) and foods with short to medium shelf life (Haghighi et al., 2020). Moreover, chitosan is widely used as a material for nanofibers production due to its ability to form films with antibacterial properties (Lin, Xue, et al., 2018; Sharaf et al., 2019). Moreover, chitosan-based materials exhibit good mechanical properties and a selective permeability to carbon dioxide and oxygen (Kamdem et al., 2019; Priyadarshi & Rhim, 2020). The tensile strength and elongation at break values are comparable to appropriate values of HDPE and LDPE (Table 2.4).

However, the use of chitosan to produce flexible packaging is still limited due to high sensitivity to humidity and moisture and high oxygen permeability. As chitosan films are highly permeable to water vapor, their use in food packaging is restricted (Priyadarshi & Rhim, 2020; Shen & Kamdem, 2015). To overcome these limitations of this otherwise excellent polysaccharide biomaterial, several attempts have been made such as blending the films with other natural or synthetic polymers and the addition of several active and functional substances like fillers, plasticizers, cross-linkers, and natural oils (Priyadarshi & Rhim, 2020).

Several studies are published that focus on improving the barrier and mechanical properties of chitosan-based films. Kamdem et al. (2019) developed a composite flexible film using chitosan as base polymer matrix, xylan to improve mechanical properties, and carvacrol (a monoterpenoid phenol) to control microbial protection. The results show that adding xylan significantly increases the elongation at break of the composite films and exhibits higher tensile strength and Young’s modulus. The incorporation of carvacrol and xylan in the composite films was not effective in terms of antimicrobial activity (Kamdem et al., 2019).

Chitosan is another potential packaging material that is investigated with lignin as additive. The addition of lignin to chitosan improves the tensile strength, storage modulus, glass transition temperature, and degradation temperature, compared to pure chitosan films (Chen et al., 2009). Moreover, lignin confers the scavenging properties to the chitosan films: this antioxidant activity is highly dependent on the film structure, functional properties, and surface activity, which is, in turn, dependent on moisture (Crouvisier-Urion et al., 2016) and the homogenization process of the film-forming suspension (Crouvisier-Urion et al., 2017).

Despite favorable properties of chitosan like its antioxidant and antimicrobial activity as well as a good biocompatibility and biodegradability, there are some drawbacks such as its dissolution in acidic media and poor thermal properties. Thus, the combination of chitosan with other polymers in binary or ternary films is intensively studied for various systems. W. Yang, Owczarek, et al. (2016) investigated films based on PVA, chitosan, and LNPs for food packaging applications and found that lignin improved tensile strength and Young's modulus as well as the thermal stability of the systems.

In conclusion, Table 2.5 summarizes advantages and disadvantages of the polymers discussed in chapter 2.4.

Table 2.5 Advantages and disadvantages of different bio-based polymers discussed in chapter 2.4.

Material	Advantage	Disadvantage	References
PLA-based	Renewable, biodegradable, biocompatible Usable for mono- and multi-layer applications Desirable mechanical properties (stiffness, tensile strength) Good gas permeability Transparen	Expensive (synthesis) Limited to rigid packaging	Ahmadzadeh & Khaneghah (2020); Auras et al. (2004); Benetto et al. (2015); Byun & Kim (2014); N. Kumar et al. (2017); van den Oever et al. (2017)
PHA-based	Water-resistant surfaces by coating Functionalize grease resistance and sealability Good thermomechanical properties Desirable gas permeability and WVP	Not transparent	Alavi et al. (2014); Andersson (2008); P. Ragaert et al. (2019); Zinoviadou et al. (2016)
Lignocellulose biomass and/or lignin-based	Abundance in nature Antioxidant activity with long-term stability Improves mechanical, thermal, and barrier properties Reduction in WVP	Deficient quality (technical lignins) Copolymerization requires functionalization	Alzagameem et al. (2019); Alzagameem, El Khaldi-Hansen, Büchner, et al. (2018); Alzagameem, El Khaldi-Hansen, Kamm, & Schulze (2018); Espinoza-Acosta et al. (2016); Michelin et al. (2020)
Protein-based	Biodegradable Abundance in nature Good film-forming properties Desirable barrier properties Transparent Low cost/cost-effective	Low mechanical strength Lack of heat stability	Acquah et al. (2020); Kadam et al. (2013); N. Kumar et al. (2017); S. Popović et al. (2012); Umaraw & Verma (2017); Y. Zhang et al. (2018)
Polysaccharide-based	Environmentally compatible, biodegradable Abundance in nature Effective oxygen barriers (intermediate to low humidity) Good mechanical properties Transparent Potential for edible packaging	Poor water vapor barrier Sensitive to moisture	Cazón et al. (2017); Han (2014); B. Hassan et al. (2018); Q. Xu et al. (2016); Youssef & El-Sayed (2018); Zinoviadou et al. (2016)

2.5 Natural additives in the context of active food packaging

In the packaging and food industry, active packaging that prolongs shelf life and reduces food losses has already been widely used (Soltani Firouz et al., 2021; Williams & Wikström, 2011). Currently, these packaging types are still mainly based on polymers from fossil-based resources (Oliveira Filho et al., 2019). However, as described previously, several other strategies in the field of bio-based materials are under development.

Currently, active packaging solutions follow different approaches (Mousavi Khaneghah et al., 2018; R. Sharma & Ghoshal, 2018):

- Addition of absorbers and scavengers of gases, off flavors, moisture, taints, UV light;
- Removal of catalyzing undesired food components;
- Addition of emitters/generators of gases and flavors;
- Release of antioxidant and/or antimicrobial compounds;
- Temperature controlled systems (insulting materials; self-heating or cooling).

Thus, basic barrier properties (such as oxygen or moisture) can be improved by adding active ingredients in the packaging system and/or using functionalized polymers. Polymers used in films and coatings that are inherently antimicrobial are chitosan, poly(L-lysine), calcium alginate, acrylic polymers, and sustainable active microbiocidal (SAM) polymers (Appendini & Hotchkiss, 2002; Ilg & Kreyenschmidt, 2012).

A promising technology that is presented and discussed in this chapter is the integration of antimicrobial and antioxidative substances (Han, 2003, 2005). Antimicrobial systems target the control or reduction of microbial growth which often results in the extension of the lag phase or in a reduced growth rate in the exponential phase (Coma, 2008; Lavoine et al., 2014). Different antimicrobial strategies using plant extracts are based on absorption, release, and immobilization systems (Han, 2003).

There are different opportunities to implement antimicrobials in packaging materials. In terms of time-releasing killing, either a volatile or non-volatile antimicrobial agent is temporarily trapped within the backbone material and released from the polymer to the environment. Volatile antimicrobial agents are released through evaporation or diffusion into the headspace of food in most cases without direct contact. Non-volatile antimicrobial agents are released by direct contact through diffusion into the food surface (Appendini & Hotchkiss, 2002; Cooksey, 2005). Another approach is the permanent immobilization of a non-volatile antimicrobial agent to a polymer backbone. The integration of antimicrobial agents in packaging can be realized by direct incorporation of the antimicrobial into the packaging material or by coating the packaging material with antimicrobial agents (Appendini & Hotchkiss, 2002; Cooksey, 2005; Han, 2003).

Natural antimicrobial, antioxidative, and photostabilizing agents used for the preservation of food are bacteriocins and extracts from biomass of animals, plants, and microorganisms. These include enzymes, EOs, and natural extracts from different plant sources (Pereira et al., 2015; Qamar et al., 2020). Phenolic compounds and terpenoids are antioxidative and antimicrobial agents that occur in EOs extracted from different plants. EOs are the most abundant source of bioactive compounds (Ruiz-Navajas et al., 2013). The effect of EOs on the microbial cells depends on different mechanisms such as disrupting the enzyme structures, damaging the phospholipid bilayer of cell membrane, and compromising the genetic makeup of microbes (Asgher, Qamar, et al., 2020). Antioxidants are compounds that react with free radicals, neutralizing them and thereby preventing or reducing their damaging effects. Aromatic plants are a source of natural antioxidants because of the activity of secondary metabolites such as phenylpropanoids and EOs (Burt, 2004). The antioxidant capacity of plant extracts is strongly related to the phenolic content (S. Y. Wang & Stretch, 2001; Zheng & Wang, 2003). Due to their redox property, they can act as reducing agents, hydrogen donors, singlet oxygen quenchers, metal chelating agents, and suppressors of free radicals (Alzagameem, El Khaldi-Hansen, Büchner, et al., 2018; Srivastava & Vankar, 2012). The antioxidant activity is not a property of a single phenolic compound, but it is widely distributed among the phenolic phytochemical constituents. So, anthocyans, flavonoids, phenolic acids, phenolic terpenes, and volatile oils are particularly interesting as antioxidants in food packaging (Alzagameem, El Khaldi-Hansen, Büchner, et al., 2018; Carpena et al., 2021).

The search for appropriate substitutional bio-based core materials is important for tackling environmental issues. Currently, those materials include, but are not limited to chitosan, starch, CMC, PLA, whey proteins, and combinations thereof. As they represent a comparably small amount of the packaging itself when compared to core materials, additives tend to be neglected. However, they are often critical to achieve the desired properties in packaging materials and can be capable of prolonging the shelf life of both packaging materials and packed goods, leading to reduced food loss.

In the following subchapters, the review summarizes applications of plant EOs and plant extracts in the context of active food packaging.

2.5.1 Plant essential oils

Plant-based active stabilizers can be isolated by extracting the appropriate biomasses by steam distillation using various solvents. Generally, EOs are complex mixtures containing over 300 different polar and non-polar volatile organic compounds, usually of low molecular weight (below 300) at quite different concentrations (Dhifi et al., 2016). Often, two or three major components exist at relatively high concentrations (20 - 70 %) while others are present in trace amounts. The major constituents of EOs are terpenoids and phenylpropanoids which provide the characteristic aroma and biological properties. Both families are comprised of phenolic compounds. The sufficiently high vapor pressure of EOs, in general, at atmospheric pressure and room temperature causes them to be found partially in the vapor state (Dhifi et al., 2016; Nieto, 2017; Raut & Karuppayil, 2014).

In comparison to producing plant extracts, a higher amount of plant biomass must be processed to obtain EOs. However, this can prove worthwhile as EOs contain active components of biomasses in particularly high concentrations and are therefore typically highly effective in different applications. This allows manufacturers to obtain great effects with applying only small amounts of oil. Prominent examples of EO applications for active food packaging are listed in Table 2.6. Results obtained so far show that plant EOs are interesting components for active food packaging. However, the corresponding materials have to be specified regarding water resistance; water vapor permeability; mechanical properties; and enhancement of antimicrobial, antioxidant, photostabilizing, and light-absorbing properties. Particular research interest lies on EOs obtained from food sources such as cinnamon, thyme, and rosemary or food production by-products as apricot kernels and banana leaves.

Table 2.6 Spotlight literature for plant essential oils used in the context of active food packaging.

Biomass	Packaging Matrix	Results	References
Apricot kernel EO	Chitosan	Prepared films showed better water resistance and improved antioxidant, antimicrobial, and mechanical properties; fungal growth on Improved antimicrobial properties (against *E. coli* and *Staph. aureus*) of gelatin films enriched with banana leaf EO; improvements on mechanical properties observed packaged bread is inhibited	Priyadarshi et al. (2018)
Banana leaf EO	Gelatin	Improved antimicrobial properties (against *E. coli* and *Staph. aureus*) of gelatin films enriched with banana leaf EO; improvements on mechanical properties observed	Kamari et al. (2018)
Bergamot, lemongrass, rosemary, and clove	PLA	Enhanced mechanical and antimicrobial properties against *E. coli*	Qin et al. (2017)
Cinnamon and ginger EOs	CMC and Chitosan	Decreased water vapor permeability (particularly for cinnamon EO), antifungal activity against *A. niger* with a higher efficacy of cinnamon EO	Noshirvani et al. (2017)
Cinnamon bark EO	Gelatin	Antioxidant and antimicrobial effects observed (against *S. typhimurium* and *L. monocytogenes*), water resistance is increased	H. Kim et al. (2018)
Cinnamon bark EO	PLA and Sea squirt (*Halocynthia roretzi*) shell protein	Enhanced antioxidant activity and antimicrobial effects against *L. monocytogenes*, *Staph. aureus*, *E. coli* and *S. typhimurium*; change of mechanical properties, decrease of water solubility, and water permeability	Beak et al. (2018)
Cinnamon EO	Chitosan and Gum	Better water barrier properties with a decrease in mechanical properties;	T. Xu et al. (2019)
Cinnamon EO	CMC and PVA	Enhancement of antioxidant and photostabilizing properties; highly effective against *Penicillium digitatum;* shelf life of packaged bread was increased	Fasihi et al. (2019)

Abbreviations: Essential Oil – EO; *Escherichia coli – E. coli; Staphylococcus aureus – Staph. aureus; Aspergillus – A.; Salmonella typhimurium – S. typhimurium; Listeria monocytogenes – L. monocytogenes; Salmonella entertidis – S. entertidis; Bacillus cereus – B. cereus.*

Table 2.6 Spotlight literature for plant essential oils used in the context of active food packaging. *(Continued)*

Biomass	Packaging Matrix	Results	References
Cinnamon EO	Gelatin	Water vapor permeability and light-absorbing properties of enriched films increase while water content and elongation at break decrease; antifungal and antimicrobial activity against *E. coli*, *Staph. aureus*, *A. niger*, *Rhizopus oryzae* and *Paecilomyces varioti* observed	J. Wu et al. (2017)
Cinnamon EO	Whey protein	Effect of enriched films against *Staph. aureus*, no effect against *E coli* observed	Aisha & Abdullahi (2017)
Cinnamon leaf oil	Gelatin	Antimicrobial effect against foodborne pathogens reported (*E. coli*, *S. typhimurium*, *Staph. aureus*, *L. monocytogenes*)	S.-Y. Yang et al. (2017)
Clove bud EO	Pectin	Antioxidant and antimicrobial effects observed (against *Staph. aureus*, *E. coli* and *L. monocytogenes*); improved mechanical properties (flexibility, resistance to breakage, water barrier properties, and heat stability)	Nisar et al. (2018)
Clove EO	Starch	Antifungal activity against *Colletotrichum gloeosporioides* and *Colletotrichum musae*, but not against *Saccharomyces bourladii*; enhanced shelf life of packaged bananas	Alves de Figueiredo Sousa et al. (2019)
Eucalyptus and Cinnamon EOs	PLA and PBAT	Antimicrobial activities against *E. coli* and *Staph. aureus* observed for both EOs with cinnamon EO showing a higher antimicrobial effect, increased biofilm inhibition and decreased UV light transmission	S. Sharma, Barkauskaite, Jaiswal, et al. (2020)
Eucalyptus globulus EO	Chitosan	Antibacterial effects against *S. entertidis*, *E. coli*, *B. cereus* and *Staph. aureus* observed (especially in liquid phase); lower antibacterial effect in vapor phase	Azadbakht et al. (2018)

Abbreviations: Essential Oil – EO; *Escherichia coli – E. coli; Staphylococcus aureus – Staph. aureus; Aspergillus – A.; Salmonella typhimurium – S. typhimurium; Listeria monocytogenes – L. monocytogenes; Salmonella entertidis – S. entertidis; Bacillus cereus – B. cereus.*

Table 2.6 Spotlight literature for plant essential oils used in the context of active food packaging. *(Continued)*

Biomass	Packaging Matrix	Results	References
Ginger EO and Eugenol	Gelatin and Chitosan	Antioxidant effect observed for both Eugenol and ginger EO (depending on film formulation); comparable water vapor permeability with increased elasticity	Bonilla et al. (2018)
Lavender EO	Starch, Furcellaran and Gelatin	Enhanced antioxidant and antimicrobial effects against *E. coli* and *Staph. aureus*; change of mechanical properties with addition of Lavender EO (decrease of tensile strength, water absorption, etc.)	Jamróz et al. (2018)
Lemon EO	Starch	Optical and mechanical properties examined; antimicrobial effects against *Staph. aureus* and *E. coli*	Song et al. (2018)
Olive oil, corn oil, sunflower oil	Chitosan	Particularly olive oil enriched films showed better mechanical properties and a high antibacterial activity	Akyuz et al. (2018)
Oregano EO	Whey protein	Higher amounts of EO resulted in higher water vapor permeability and film flexibility; antimicrobial activity against *Penicillium commune*	Oliveira et al. (2017)
Origanum vulgare, O. majorana EOs	Chitosan	Antimicrobial effect against *Staph. aureus* and *B. cereus* observed with both EOs, significantly enhanced effects for *O. vulgare* EO	Sedlaříková et al. (2017)
Pistacia atlantica EO	CMC and Gelatin	Antimicrobial effect against *E. coli*, *Staph. aureus*, *Clostridium sporogenes,* and particularly, *Salmonella enterica*; reduction of e.g., water vapor permeability, film thickness, and tensile strength	Ranjbar et al. (2017)
Rosemary and mint EOs	Chitosan, Pectin, and Starch	Rosemary and mint EOs improved water barrier properties and inhibited *Bacillus subtilis*, *E. coli* and *L. monocytogenes*; both EOs resulted in enhanced antioxidant effects	Akhter et al. (2019)

Abbreviations: Essential Oil – EO; *Escherichia coli – E. coli; Staphylococcus aureus – Staph. aureus; Aspergillus – A.; Salmonella typhimurium – S. typhimurium; Listeria monocytogenes – L. monocytogenes; Salmonella entertidis – S. entertidis; Bacillus cereus – B. cereus.*

Table 2.6 Spotlight literature for plant essential oils used in the context of active food packaging. *(Continued)*

Biomass	Packaging Matrix	Results	References
Rosmarinus officinalis, *Artemisia herba-alba*, *Ocimum basilicum* and *Mentha pulegium* EOs	Alginate	Strong antibacterial activity against *Staph. aureus*, *E. coli*, *Salmonella enterica*, *Enterococcus faecium*, *Klebsiella pneumoniae* and *Enterococcus faecalis*; physical properties analyzed, antioxidant effect observed	Mahcene et al. (2020)
Satureja Khuzestanica EO	Kefiran and CMC	Antimicrobial effects against *Staph. aureus* and *E. coli*, significant antioxidant properties, change in physical properties (e.g., decrease in water vapor permeability)	Hasheminya et al. (2019)
Satureja Khuzistanica Jamzad EO	Whey protein	Antimicrobial effect particularly against *Staph. aureus* with *Pseudomonas aeruginosa* showing the highest resistance of analyzed bacteria; increased elongation at break and water vapor permeability	Kouravand et al. (2018)
Summer savory EO	CMC and Agar	Antimicrobial effects particularly against *Staph. aureus*, *B. cereus* and *L. monocytogenes* with lower effects against *E. coli*; alteration of physical properties (increased water vapor permeability, improved mechanical flexibility)	Abdollahi et al. (2019)
Thyme and Clove EOs	PLA and PBAT	Positive properties observed for both EOs, but particularly for clove EO films, including UV-blocking and highly antimicrobial effects (inhibition of *E. coli*, complete killing of *Staph. aureus*)	S. Sharma, Barkauskaite, Duffy, et al. (2020)
Thyme, rosemary, and oregano EOs	PLA	Significant antioxidant effect on packaged minced fish with moderate alteration of mechanical properties	Zeid et al. (2019)

Abbreviations: Essential Oil – EO; *Escherichia coli – E. coli; Staphylococcus aureus – Staph. aureus; Aspergillus – A.; Salmonella typhimurium – S. typhimurium; Listeria monocytogenes – L. monocytogenes; Salmonella entertidis – S. entertidis; Bacillus cereus – B. cereus.*

2.5.2 Plant extracts of various biomasses

Quality and quantity of plant extracts strongly depend on their biomass origin and type of extraction processes. The most common extracts have been obtained by conventional solvent extraction methods (infusion, decoction, digestion, maceration, and percolation) using solvents such as water, ethanol, methanol, chloroform, or dimethyl-sulfoxide (Aleksic Sabo & Knezevic, 2019; Azwanida, 2015; Karacabey et al., 2013).

Such extracts typically retard bacterial growth and can also introduce antioxidant and photoabsorbing effects. Those properties are reported for various different biomasses including (but not limited to) herbs, flowers, trees, and their fruits (Bati Ay et al., 2018; Biswas et al., 2018; Havelt et al., 2019; Havelt et al., 2020; Havelt & Schmitz, 2018; Karimi et al., 2018; Kimura et al., 2017). In contrast to essential oils, the effects tend to be less extensive; however, less biomass must be processed to obtain such extracts. Furthermore, as high-concentrated essential oils provide lipophilic surroundings for active substances, the application of polar active substances is only possible by (hydrophilic) extraction, thus introducing a whole new group of active compounds. The specific characteristics observed in plant extracts can be utilized by incorporating them in food packaging materials to positively affect the packed food. Prominent examples for this approach are documented in Table 2.7. The studies using plant extracts show similar improvements in packaging characteristics to plant essential oils. Decreased water vapor permeability; enhanced moisture and oil resistance; improved mechanical properties; and enhanced antimicrobial, antioxidant, photoabsorbing, and UV-stability are reported. While it is challenging to directly compare the obtained data with each other due to a variety of tests and extraction methods used, most researchers claim a relevant potential exists for plant-based stabilizers in food packaging applications. Again, plant-based active packaging research is typically focused on biomasses that represents foodstuff.

Table 2.7 Spotlight literature for plant extracts used in the context of active food packaging.

Biomass	Packaging Matrix	Results	References
Coffee beans and de-fatted cocoa beans	Starch	Synergistic antioxidant effect, decreased water vapour permeability, increased shelf life of palm oil	Veiga-Santos et al. (2018)
Grapefruit seed extract	PCL and Chitosan	Films showed better mechanical properties and inhibited bacterial growth of *E. coli* and *P. aeruginosa* for up to 6 days; successful tests with packaged salmon and bread	K. Wang et al. (2019)
Herba Lophatheri extract	Chitosan	Moisture and oil resistance are enhanced, both antioxidant and antimicrobial activities observed (*E. coli*, *Staph. aureus*)	L. Wang et al. (2019)
Kombucha tea extract	Chitosan	Decreased water vapor permeability and improved antioxidant, photoabsorbing, and antimicrobial effects (against *E. coli* and *Staph. aureus*); 3 days extended shelf life for packaged minced beef	Ashrafi et al. (2018)
Grape seed extract	Chitosan	Enhanced antioxidant and antimicrobial activity (total mesophilic aerobic bacteria, coliforms, *E. coli*, *L. monocytogenes*, *Staph. aureus* and *P. aeruginosa*; shelf life extension of refrigerated, vacuum-packed chicken breast fillets)	Sogut & Seydim (2018)
Rosemary extract	Starch	Significant antioxidant effect, increased UV-stability	Piñeros-Hernandez et al. (2017)
Propolis extract and *Zataria multiflora* EO	Chitosan	Antimicrobial effects measured for mesophilic total viable plate counts, lactic acid bacteria, psychotropic bacteria, and *Pseudomonas*; synergistic effects observed; lower microbial load on packaged chicken	Mehdizadeh & Mojaddar Langroodi (2019)
Sumac extract and *Zataria multiflora* EO	Chitosan	Antioxidant effects and prolonged shelf life on packaged meat observed; antimicrobial activity against different bacteria (e.g., *Pseudomonas* spp.)	Mojaddar Langroodi et al. (2018)
Propolis extract and *Zataria multiflora* EO	PLA	Increased shelf life of packaged sausages, antimicrobial effects against common food pathogens (*Staph. aureus*, *E. coli*, *Vibrio parahaemolyticus*, *L. monocytogenes*)	Rezaeigolestani et al. (2017)

Abbreviations: *Escherichia coli – E. coli; Pseudomonas aeruginosa – P. aeruginosa; Staphylococcus aureus – Staph. aureus; Listeria monocytogenes – L. monocytogenes.*

2.5.3 Encapsulated plant essential oils

Advanced methods use plant essential oils after encapsulation by a variety of different techniques, including formation of nanofibers, nanotubes, and nanoparticles (Rehman et al., 2020). This way, the essential oils are more resistant against thermal influences (Qiu et al., 2016; Rashed et al., 2019; Wen et al., 2016). The incorporation of encapsulated essential oil typically also improves the mechanical properties of packaging materials (dos Santos Paglione et al., 2019; Mohsenabadi et al., 2018). Encapsulation furthermore facilitates gradual release of active ingredients, leading to a more durable protection of the packed foodstuff. Encapsulated essential oils are also under investigation in other fields and applications including bio-based insecticides and cleaning agents (Khoobdel et al., 2017; Werdin González et al., 2017). Recent studies utilizing the encapsulation of essential oils in the context of food packaging are presented in Table 2.8. The results confirm that encapsulated plant oils are able to improve water vapor permeability, transparency, and tensile strength as well as antioxidant and antimicrobial effects.

Table 2.8 Spotlight literature for encapsulated plant oils used in the context of active food packaging.

Biomass	Packaging Matrix	Encapsulation Details	Results	References
Chrysanthemum EO	-	Chitosan nanofibers	Antioxidant and antimicrobial effect against *L. monocytogenes* observed e.g., on packaged beef, prolongation of shelf life possible	Lin, Mao, et al. (2019)
Cinnamon EO	PLA	Nanofibers	Better antimicrobial effect against *Staph. aureus* and *E. coli* observed for encapsulated EO; encapsulation process is more suitable formulation method to maintain EO properties; shelf life of packaged pork was prolonged	Wen et al. (2016)
Clove EO	Alginate	Inclusion complex	Successful incorporation of clove EO complexes; resulting in less transparent and flexible films, decreased elasticity, increased water vapor permeability	Maestrello et al. (2017)
Cumin seed oil	-	Nanoemulsion (Whey protein, Guar gum)	Antimicrobial effect of encapsulated oil against *Staph. aureus*, *E. coli*, and *A. flavus*	Farshi et al. (2019)
Cuminum cyminum EO	-	Chitosan nanoparticles	Significant antioxidant effect in packaged white button mushrooms observed, resulting in presumed shelf life prolongation	Karimirad et al. (2018)
Laurel EO and silver nanoparticles	PE	Liposomes in Chitosan	Antioxidant properties observed during 7 days of storage with only about 30% of EO released from liposomes; antimicrobial effect against *Staph. aureus* and *E. coli* results in 6 days prolonged shelf life of packaged pork	Z. Wu et al. (2019)

Abbreviations: Essential Oil – EO; *Listeria monocytogenes* – *L. monocytogenes*; *Staphylococcus aureus* – *Staph. aureus*; *Escherichia coli* – *E. coli*; *Aspergillus flavus* – *A. flavus*; *Pseudomonas aeruginosa* – *P. aeruginosa*.

Table 2.8 Spotlight literature for encapsulated plant oils used in the context of active food packaging. *(Continued)*

Biomass	Packaging Matrix	Encapsulation Details	Results	References
Lavandula angustofolia EO	-	Nanoemulsion (Whey protein)	Encapsulation enhanced thermal stability of EO; antibacterial effect is observed	Rashed et al. (2019)
Menthone, Oregano, Cinnamon, Lavender and Citral EOs		Starch nanoparticles	Enhanced stability of antioxidants against thermal influence after encapsulation; antimicrobial effects against *E. coli* and *Staph. aureus* are prolonged	Qiu et al. (2016)
Moringa oil	Gelatin nanofibers	Chitosan nanoparticles	High antimicrobial activity of encapsulated Moringa oil against *L. monocytogenes* and *Staph. aureus* for 10 days without affecting the sensory properties of packaged cheese	Lin, Gu, & Cui (2019)
Oregano EO	-	PCL nanocapsules	High retention of encapsulated Rosemary EO (determined via carvacrol content) observed, suitability for long-term delivery of carvacrol can be assumed	Fraj et al. (2019)
Oregano EO	Soy protein	Microencapsulation by ionic gelation	Strong antioxidant and antimicrobial properties against *E. coli* and *Staph. aureus*; enhanced effects and mechanical properties with microencapsulated EO in contrast to free EOs	dos Santos Paglione et al. (2019)
Rosemary EO	Starch and CMC	Chitosan nanogel	Films with encapsulated EO show higher water vapor permeability, higher transparency, and tensile strength; immediate (free EO) and gradual (encapsulated EO) antimicrobial effects against *Staph. aureus* were observed	Mohsenabadi et al. (2018)

Abbreviations: Essential Oil – EO; *Listeria monocytogenes* – *L. monocytogenes*; *Staphylococcus aureus* – *Staph. aureus*; *Escherichia coli* – *E. coli*; *Aspergillus flavus* – *A. flavus*; *Pseudomonas aeruginosa* – *P. aeruginosa*.

Table 2.8 Spotlight literature for encapsulated plant oils used in the context of active food packaging. *(Continued)*

Biomass	**Packaging Matrix**	**Encapsulation Details**	**Results**	**References**
Thymbra capitata EO	-	Zein nanoparticles	Both free and encapsulated EO are effective against *E. coli* and *L. monocytogenes*; presumably due to controlled release, encapsulated EO showed lower antimicrobial efficacy compared to free EO	Merino et al. (2019)
Thyme EO	-	Nanofibers (Chitosan, Gelatin)	Both free and encapsulated thyme EO has antioxidant and antimicrobial effects against *Clostridium perfringens*; tests show that such nanofibers could be used to substitute nitrite in meat products	Vafania et al. (2019)
Thyme EO	Gelatin	Nanofibers	Antimicrobial effect against *Campylobacter jejuni* in packaged chicken observed	Lin, Zhu, & Cui (2018)
Thyme EO	Ink (for paper packaging)	Halloysite nanotubes	Strong antibacterial activity against *E. coli*, mesophilic aerobic bacteria, molds, and yeasts for up to 10 days after encapsulation in Halloysite nanotubes	Jang et al. (2017); Lee et al. (2017)
Zataria multiflora EO	PVA	Nanofibers (Chitosan, PVA, Gelatin)	Encapsulated *Zataria multiflora* EO completely inhibited growth of *Staph. aureus*, *P. aeruginosa* and *Candida albicans* for 24 hours; tested material is developed for use as wound dressing	Ardekani et al. (2019)

Abbreviations: Essential Oil – EO; *Listeria monocytogenes* – *L. monocytogenes*; *Staphylococcus aureus* – *Staph. aureus*; *Escherichia coli* – *E. coli*; *Aspergillus flavus* – *A. flavus*; *Pseudomonas aeruginosa* – *P. aeruginosa*.

2.6 Adoption potential of bio-based (active) packaging along the value chain

Active packaging based on biopolymers are identified as the more sustainable alternative compared to conventional packaging. In addition, the integration of natural additives has positive effects on the quality and shelf life of the packaged product (Bos et al., 2010; Schumann & Schmid, 2018; van den Oever et al., 2017). However, in order to be successful in the market, this innovative concept needs to be adopted along the whole agricultural food value chain (Carraresi et al., 2018; Golembiewski et al., 2015).

More specifically, farmers need to collect, process, and deliver raw materials such as annual plants (e.g., miscanthus) or residues from agricultural production (e.g., sugarcane bagasse) (Keegan et al., 2013). The packaging industry must adopt renewable resources as raw materials and might also need to adjust their production processes for the application of bio-based and/or biodegradable polymers as a packaging core matrix and to integrate natural additives into the packaging materials. Food companies need to be willing to pay more for the material to pack their products, and the consumer needs to accept the concept of bio-based active packaging (Theinsathid et al., 2011; Wensing et al., 2020). As the implementation of active packaging based on bio-based polymers entails several changes for farmers, industry, and consumers, the remaining section reviews extant studies exploring the adoption decisions of these value chain actors.

Existing literature looking at the adoption behavior of farmers finds that these value chain actors are generally sceptical towards innovations related to the bioeconomy. Therefore, monetary incentives and assistance with the novel practices and processes might be necessary (Rossi & Hinrichs, 2011). In addition to farmers, food processing companies might also serve as the provider of by-products as raw materials to produce active bio-based packaging. However, there is currently a lack of research regarding the adoption decisions of managers in those companies. Today, a growing number of farmers are interested in adopting practices to valorize by-products (Wensing et al., 2019). Therefore, these farmers need to be targeted by policy initiatives and could then serve as opinion leaders to positively influence the adoption decisions' of their communities (van Eck et al., 2011; Xiong et al., 2016).

Focusing on industry representatives such as packaging producers and food companies, exploratory studies identify several factors driving their adoption decision. First of all, the market prices and the availability of renewable resources for the production of bio-based polymers and natural additives are relevant for the adoption decisions of packaging producers (Theinsathid et al., 2011). Moreover, relevant policy instruments need to be implemented to foster research and development of bio-based polymers with natural additives (e.g., subsidies) or even to ban conventional (multi-layer) plastics (Berg et al., 2018; European Commission, 2018; Theinsathid et al., 2011). This would increase the competitiveness of bio-based and/or biodegradable packaging with conventional plastics (Theinsathid et al., 2011). However, even when policy instruments are in place, the level of consumer demand is the most important factor driving the adoption decision of industry representatives (Berg et al., 2018).

Consumer studies indicate that the final actors in the value chain have both positive and negative associations with bio-based products. They may misunderstand the concept of `bio-based´ (Sijtsema et al., 2016; Sleenhoff et al., 2015; Stern et al., 2018). However, the majority of consumers seem to believe that sustainable packaging is important and useful (Petljak et al., 2019). Moreover, results of two studies provide evidence that bio-based packaging seems to increase the preferences for the packaged product (Herbes et al., 2018; Koutsimanis et al., 2015). In fact, empirical results from a discrete choice experiment indicate that consumers are willing to pay a price premium for bio-based plastic packaging (Wensing et al., 2020). Considering the calculations by van den Oever et al. (2017), this price premium even covers the additional costs for bio-based and/or biodegradable plastics compared to conventional materials. Food companies could therefore switch to bioplastic packaging without expecting any lost profits (Wensing et al., 2020). Moreover, especially those consumers with high levels of environmental awareness and innovativeness seem to prefer bio-based plastics over conventional plastic products (F. Klein et al., 2019; Scherer et al., 2017, 2018). Products packaged with bio-based materials thus need to be presented in retail locations which are preferred by this type of consumers such as organic stores (Wensing et al., 2020). However, as bio- and fossil-based plastic packaging are not easy to be distinguished by consumers, the packaging needs to be labelled accordingly (Rumm, 2016).

After its use as packaging material, end-of-life solutions also need to be considered for active packaging derived from bio-based and/or biodegradable polymers from renewable resources (Wensing et al., 2020). Depending on consumers’ disposal behaviors, bio-based bioplastics can be decomposed given the right conditions (in case of biodegradable and/or compostable compounds) or the material can be used to generate renewable energy. Thus, it is very important that the disposal options are clearly communicated to the consumers (Müller et al., 2014). In fact, consumers are even willing to pay a price premium for biodegradable and recyclable packaging (Klaiman et al., 2016; Yue et al., 2010).

2.7 Conclusion

Besides current political requirements that aim to improve sustainability aspects, the development and promotion of more sustainable materials have gained more importance due to consumer interests. As customers preferences shifted to high quality and safe products with enhanced shelf life, the development of various new trends in packaging systems has arisen. Research focuses on improving the characteristics of bio-based packaging materials, in particular mechanical, thermal, and physical properties. Although bio-based polymers provide significant opportunities in terms of sustainability and biocompatibility, their use in industrial applications is often restricted due to lower performance in fundamental packaging functions. Companies are faced with a challenge of alternatives offering higher costs, limited functionality, existing infrastructure, and inconsistent legislation. Furthermore, a lack of compatibility with conventional processing technologies has to be overcome. Food companies need to be willing to pay more for the material to pack their products and the consumer needs to accept the novel concept of (active) packaging. The *proof-of-concept* is shown by a few commercially available biopolymers with food applications such as PLA, PHAs, PEF, PBS, and thermoplastic cellulose or starch-based films. In the future, the market for sustained active packaging will certainly increase due to enhanced efforts and innovations in material development and processing technologies.

Funding

This research was funded by EFRE/NRW "Biobasierte Produkte" (Grant EFRE 0500035). J.R. gratefully acknowledges a scholarship given by the Graduate Institute of the Bonn-Rhein-Sieg University of Applied Sciences.

List of Abbreviations

APET	Amorphous poly(ethylene terephthalate)
AZC	Ammonium zirconium carbonate
CA	Cellulose acetate
CAB	Cellulose acetate butyrate
CAP	Cellulose acetate propionate
CMC	Carboxymethyl cellulose
CoO.ZnO	Cobalt(II) oxide/zinc oxide
EO	Essential oil
GHG	Greenhouse gas
HDPE	High density poly(ethylene)
HLCNC	High lignin-containing cellulose nanocrystals
HPMC	Hydroxypropylmethyl cellulose
LCA	Life cycle analysis
LDPE	Low density poly(ethelene)
PA	Poly(amide)
PBAT	Poly(butylene adipate terephthalate)
PBS	Poly(butylene succinate)
PCL	Poly(caprolactone)
PE	Poly(ethylene)
PHA	Poly(hydroxyalkanoate)
PHB	Poly(3-hydroxybutyrate)
PEF	Poly(ethylene furanoate)
PET	Poly(ethylene terephthalate)
PHBHHx	Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate)
PHBV	Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)
PLA	Poly(lactic acid)
PP	Poly(propylene)
PS	Poly(styrene)
PTT	Poly(trimethylene terephthalate)
PVA	Poly(vinyl alcohol)
PVC	Poly(vinyl chloride) (PVC)
SA	Sodium alginate
SAM	Sustainable active microbiocidal (SAM)
SiO_2	Silicon dioxide
TAIC	Triallyl isocyanurate
WVP	Water vapor permeability

Glossary

Active packaging	Materials designed to deliberately incorporated components that would release or absorb substances into or from the packaged food or the environment surrounding the food.
Bioactive	Compound that has an effect on a living organism, tissue/cell.
Bio-based	Compound that is composed (in whole or in significant part) of biological products or renewable domestic agricultural or forestry materials (including plant, animal, and marine materials).
Biodegradable	Degradability achieved via microorganisms.
Bioplastics	Plastics that are either bio-based, biodegradable, or features both properties.
Biopolymers	Natural polymers produced by the cells of living organisms (e.g., forestry and agricultural crops, terrestrial and marine animals), examples are polysaccharides, proteins, and lignin.
Compostable	Compounds approved to be degradable by microorganisms at defined conditions (e.g., temperature, humidity, time).
Edible packing	Compounds approved to be metabolized by humans.
End-of-life options	Including re-use, recycling, recovery, disposal, and others (such as littering, ingestion).
European Green Deal	Action plan to boost the efficient use of resources by moving to a clean, circular economy, restore biodiversity, and cut pollution.
Fossil-based	Compounds obtained from crude oil, natural gas, brown or hard coal.
Renewable resource	Resource which will replenish to replace the portion depleted by usage and consumption, either through natural reproduction or other recurring processes in a finite amount of time in a human time scale.

References

Abdollahi, M., Damirchi, S., Shafafi, M., Rezaei, M., & Ariaii, P. (2019). Carboxymethyl cellulose-agar biocomposite film activated with summer savory essential oil as an antimicrobial agent. *International Journal of Biological Macromolecules*, *126*, 561–568. https://doi.org/10.1016/j.ijbiomac.2018.12.115.

Abdou, T. R., Botelho Junior, A. B., Espinosa, D. C. R., & Tenório, J. A. S. (2021). Recycling of polymeric composites from industrial waste by pyrolysis: Deep evaluation for carbon fibers reuse. *Waste Management*, *120*, 1–9. https://doi.org/10.1016/j.wasman.2020.11.010.

Accorsi, R. (2019). A support-design procedure for sustainable food product-packaging systems. In *Sustainable Food Supply Chains* (pp. 61–81). Elsevier. https://doi.org/10.1016/B978-0-12-813411-5.00005-3.

Acquah, C., Zhang, Y [Yujie], Dubé, M. A., & Udenigwe, C. C. (2020). Formation and characterization of protein-based films from yellow pea (Pisum sativum) protein isolate and concentrate for edible applications. *Current Research in Food Science*, *2*, 61–69. https://doi.org/10.1016/j.crfs.2019.11.008.

Aggarwal, A., & Langowski, H.-C. (2020). Packaging Functions and Their Role in Technical Development of Food Packaging Systems: Functional Equivalence in Yoghurt Packaging. *Procedia CIRP*, *90*, 405–410. https://doi.org/10.1016/j.procir.2020.01.063.

Ahmadzadeh, S., & Khaneghah, A. M. (2020). Role of Green Polymers in Food Packaging. In I. Choudhury & S. Hashmi (Eds.), *Encyclopedia of Renewable and Sustainable Materials* (pp. 305–319). Elsevier. https://doi.org/10.1016/B978-0-12-803581-8.10576-4.

Ahmed, N. M., Adel, A. M., & Diab, M. A. (2020). Packaging paper with improved mechanical and oil absorption properties based on novel ingredients. *Packaging Technology and Science*, *33*(8), 303–320. https://doi.org/10.1002/pts.2506.

Ahmed, T., Shahid, M., Azeem, F., Rasul, I., Shah, A. A., Noman, M., Hameed, A., Manzoor, N., Manzoor, I., & Muhammad, S. (2018). Biodegradation of plastics: Current scenario and future prospects for environmental safety. *Environmental Science and Pollution Research*, *25*(8), 7287–7298. https://doi.org/10.1007/s11356-018-1234-9.

Ahorsu, R., Medina, F., & Constantí, M. (2018). Significance and Challenges of Biomass as a Suitable Feedstock for Bioenergy and Biochemical Production: A Review. *Energies*, *11*(12), Article 3366. https://doi.org/10.3390/en11123366.

Aisha, I., & Abdullahi, Y. (2017). Development of Whey Protein Concentrate Edible Membrane with Cinnamon Essential Oil. *Journal of Advances in Biology & Biotechnology*, *11*(2), 1–14. https://doi.org/10.9734/JABB/2017/29136.

Akhter, R., Masoodi, F. A., Wani, T. A., & Rather, S. A. (2019). Functional characterization of biopolymer based composite film: Incorporation of natural essential oils and antimicrobial agents. *International Journal of Biological Macromolecules*, *137*, 1245–1255. https://doi.org/10.1016/j.ijbiomac.2019.06.214.

Akyuz, L., Kaya, M., Ilk, S., Cakmak, Y. S., Salaberria, A. M., Labidi, J., Yılmaz, B. A., & Sargin, I. (2018). Effect of different animal fat and plant oil additives on physicochemical, mechanical, antimicrobial and antioxidant properties of chitosan films. *International Journal of Biological Macromolecules*, *111*, 475–484. https://doi.org/10.1016/j.ijbiomac.2018.01.045.

Alavi, S., Thomas, S., Sandeep, K. P., Kalarikkal, N., Varghese, J., & Yaragalla, S. (2014). *Polymers for Packaging Applications*. Apple Academic Press.

Alves de Figueiredo Sousa, H., Gonçalves de Oliveira Filho, J., Egea, M. B., Da Rosa Silva, E., Macagnan, D., Pires, M., & Peixoto, J. (2019). Active film incorporated with clove essential oil on storage of banana varieties. *Nutrition & Food Science*, *49*(5), 911–924. https://doi.org/10.1108/NFS-09-2018-0262.

Alzagameem, A., El Khaldi-Hansen, B., Büchner, D., Larkins, M., Kamm, B., Witzleben, S., & Schulze, M. (2018). Lignocellulosic Biomass as Source for Lignin-Based Environmentally Benign Antioxidants. *Molecules*, *23*(10). https://doi.org/10.3390/molecules23102664.

Alzagameem, A., El Khaldi-Hansen, B., Kamm, B., & Schulze, M. (2018). Lignocellulosic biomass for energy, biofuels, biomaterials, and chemicals. In S. Vaz JR. (Ed.), *Biomass and Green Chemistry: Building a Renewable Pathway* (pp. 95–132). Springer International Publishing.

Alzagameem, A., Klein, S. E., Bergs, M., Do, X. T., Korte, I., Dohlen, S., Hüwe, C., Kreyenschmidt, J., Kamm, B., Larkins, M., & Schulze, M. (2019). Antimicrobial Activity of Lignin and Lignin-Derived Cellulose and Chitosan Composites Against Selected Pathogenic and Spoilage Microorganisms. *Polymers*, *11*(4). https://doi.org/10.3390/polym11040670.

Amjadi, S., Hamishehkar, H., & Ghorbani, M. (2019). A novel smart PEGylated gelatin nanoparticle for co-delivery of doxorubicin and betanin: A strategy for enhancing the therapeutic efficacy of chemotherapy. *Materials Science & Engineering C*, *97*, 833–841. https://doi.org/10.1016/j.msec.2018.12.104.

Andersson, C. (2008). New ways to enhance the functionality of paperboard by surface treatment - a review. *Packaging Technology and Science*, *21*(6), 339–373. https://doi.org/10.1002/pts.823.

Anjum, A., Zuber, M., Zia, K. M., Noreen, A., Anjum, M. N., & Tabasum, S. (2016). Microbial production of polyhydroxyalkanoates (PHAs) and its copolymers: A review of recent advancements. *International Journal of Biological Macromolecules*, *89*, 161–174. https://doi.org/10.1016/j.ijbiomac.2016.04.069.

Appendini, P., & Hotchkiss, J. H. (2002). Review of antimicrobial food packaging. *Innovative Food Science & Emerging Technologies*, *3*, 113–126. https://doi.org/10.1016/S1466-8564(02)00012-7.

Ardekani, N. T., Khorram, M., Zomorodian, K., Yazdanpanah, S., Veisi, H [Hamed], & Veisi, H [Hojat] (2019). Evaluation of electrospun poly (vinyl alcohol)-based nanofiber mats incorporated with Zataria multiflora essential oil as potential wound dressing. *International Journal of Biological Macromolecules*, *125*, 743–750. https://doi.org/10.1016/j.ijbiomac.2018.12.085.

Armentano, I., Fortunati, E., Burgos, N., Dominici, F., Luzi, F., Fiori, S., Jiménez, A [Alfonso], Yoon, K., Ahn, J., Kang, S., & Kenny, J. M. (2015). Bio-based PLA_PHB plasticized blend films: Processing and structural characterization. *LWT - Food Science and Technology*, *64*(2), 980–988. https://doi.org/10.1016/j.lwt.2015.06.032.

Arnon, H., Zaitsev, Y., Porat, R., & Poverenov, E. (2014). Effects of carboxymethyl cellulose and chitosan bilayer edible coating on postharvest quality of citrus fruit. *Postharvest Biology and Technology*, *87*, 21–26. https://doi.org/10.1016/j.postharvbio.2013.08.007.

Arrieta, M. P., Samper, M. D., López, J., & Jiménez, A [Alfonso] (2014). Combined Effect of Poly(hydroxybutyrate) and Plasticizers on Polylactic acid Properties for Film Intended for Food Packaging. *Journal of Polymers and the Environment*, *22*(4), 460–470. https://doi.org/10.1007/s10924-014-0654-y.

Asgher, M., Arshad, S., Qamar, S. A., & Khalid, N. (2020). Improved biosurfactant production from Aspergillus niger through chemical mutagenesis: characterization and RSM optimization. *SN Applied Sciences*, *2*(5). https://doi.org/10.1007/s42452-020-2783-3.

Asgher, M., Qamar, S. A., Bilal, M., & Iqbal, H. M. N. (2020). Bio-based active food packaging materials: Sustainable alternative to conventional petrochemical-based packaging materials. *Food Research International*, *137*, Article 109625. https://doi.org/10.1016/j.foodres.2020.109625.

Asgher, M., Urooj, Y., Qamar, S. A., & Khalid, N. (2020). Improved exopolysaccharide production from Bacillus licheniformis MS3: Optimization and structural/functional characterization. *International Journal of Biological Macromolecules*, *151*, 984–992. https://doi.org/10.1016/j.ijbiomac.2019.11.094.

Ashrafi, A., Jokar, M., & Mohammadi Nafchi, A. (2018). Preparation and characterization of biocomposite film based on chitosan and kombucha tea as active food packaging. *International Journal of Biological Macromolecules*, *108*, 444–454. https://doi.org/10.1016/j.ijbiomac.2017.12.028.

Auras, R., Harte, B., & Selke, S. (2004). An Overview of Polylactides as Packaging Materials. *Macromolecular Bioscience*, *4*(9), 835–864. https://doi.org/10.1002/mabi.200400043.

Azadbakht, E., Maghsoudlou, Y., Khomiri, M., & Kashiri, M. (2018). Development and structural characterization of chitosan films containing Eucalyptus globulus essential oil: Potential as an antimicrobial carrier for packaging of sliced sausage. *Food Packaging and Shelf Life*, *17*, 65–72. https://doi.org/10.1016/j.fpsl.2018.03.007.

Azadfar, M., Gao, A. H., Bule, M. V., & Chen, S. (2015). Structural characterization of lignin: A potential source of antioxidants guaiacol and 4-vinylguaiacol. *International Journal of Biological Macromolecules*, *75*, 58–66. https://doi.org/10.1016/j.ijbiomac.2014.12.049.

Bahrami, A., Rezaei Mokarram, R., Sowti Khiabani, M., Ghanbarzadeh, B., & Salehi, R. (2019). Physico-mechanical and antimicrobial properties of tragacanth/hydroxypropyl methylcellulose/beeswax edible films reinforced with silver nanoparticles. *International Journal of Biological Macromolecules*, *129*, 1103–1112. https://doi.org/10.1016/j.ijbiomac.2018.09.045.

Barlow, C. Y., & Morgan, D. C. (2013). Polymer film packaging for food: An environmental assessment. *Resources, Conservation and Recycling*, *78*, 74–80. https://doi.org/10.1016/j.resconrec.2013.07.003.

Beak, S., Kim, H., & Song, K. B. (2018). Sea Squirt Shell Protein and Polylactic Acid Laminated Films Containing Cinnamon Bark Essential Oil. *Journal of Food Science*, *83*(7), 1896–1903. https://doi.org/10.1111/1750-3841.14207.

Benetto, E., Jury, C., Igos, E., Carton, J., Hild, P., Vergne, C., & Di Martino, J. (2015). Using atmospheric plasma to design multilayer film from polylactic acid and thermoplastic starch: a screening Life Cycle Assessment. *Journal of Cleaner Production*, *87*, 953–960. https://doi.org/10.1016/j.jclepro.2014.10.056.

Berg, S., Cloutier, L. M., & Broering, S. (2018). Collective stakeholder representations and perceptions of drivers of novel biomass-based value chains. *Journal of Cleaner Production*, *200*, 231–241. https://doi.org/10.1016/j.jclepro.2018.07.304.

Bhat, R., Abdullah, N., Din, R. H., & Tay, G.-S. (2013). Producing novel sago starch based food packaging films by incorporating lignin isolated from oil palm black liquor waste. *Journal of Food Engineering*, *119*(4), 707–713. https://doi.org/10.1016/j.jfoodeng.2013.06.043.

Bhat, R., & Jõudu, I. (2019). Emerging issues and challenges in agri-food supply chain. In *Sustainable Food Supply Chains* (pp. 23–37). Elsevier. https://doi.org/10.1016/B978-0-12-813411-5.00002-8.

Bifani, V., Ramírez, C., Ihl, M., Rubilar, M., García, A., & Zaritzky, N. (2007). Effects of murta (Ugni molinae Turcz) extract on gas and water vapor permeability of carboxymethylcellulose-based edible films. *LWT - Food Science and Technology*, *40*(8), 1473–1481. https://doi.org/10.1016/j.lwt.2006.03.011.

Biscarat, J., Charmette, C., Sanchez, J., & Pochat-Bohatier, C. (2015). Development of a new family of food packaging bioplastics from cross-linked gelatin based films. *The Canadian Journal of Chemical Engineering*, *93*(2), 176–182. https://doi.org/10.1002/cjce.22077.

Bonilla, J., Poloni, T., Lourenço, R. V., & Sobral, P. J. (2018). Antioxidant potential of eugenol and ginger essential oils with gelatin/chitosan films. *Food Bioscience*, *23*, 107–114. https://doi.org/10.1016/j.fbio.2018.03.007.

Bos, H., Meesters, K., Conijn, S., Corré, W., & Patel, M. (2010). *Sustainability aspects of biobased applications*. http://edepot.wur.nl/170079.

Broeren, M. L., Kuling, L., Worrell, E., & Shen, L. (2017). Environmental impact assessment of six starch plastics focusing on wastewater-derived starch and additives. *Resources, Conservation & Recycling*, *127*, 246–255. https://doi.org/10.1016/j.resconrec.2017.09.001.

Bruckner, S., Albrecht, A., Petersen, B., & Kreyenschmidt, J. (2012). Characterization and Comparison of Spoilage Processes in Fresh Pork and Poultry. *Journal of Food Quality*, *35*(5), 372–382. https://doi.org/10.1111/j.1745-4557.2012.00456.x.

Burt, S. (2004). Essential oils: Their antibacterial properties and potential applications in foods—a review. *International Journal of Food Microbiology*, *94*(3), 223–253. https://doi.org/10.1016/j.ijfoodmicro.2004.03.022.

Byun, Y., & Kim, Y. T. (2014). Utilization of Bioplastics for Food Packaging Industry. In *Innovations in Food Packaging* (pp. 369–390). Elsevier. https://doi.org/10.1016/B978-0-12-394601-0.00015-1.

Caldeira, C., Laurentiis, V. de, Corrado, S., van Holsteijn, F., & Sala, S. (2019). Quantification of food waste per product group along the food supply chain in the European Union: A mass flow analysis. *Resources, Conservation & Recycling*, *149*, 479–488. https://doi.org/10.1016/j.resconrec.2019.06.011.

Carpena, M., Nuñez-Estevez, B., Soria-Lopez, A., Garcia-Oliveira, P., & Prieto, M. A. (2021). Essential Oils and Their Application on Active Packaging Systems: A Review. *Resources*, *10*(1), 7. https://doi.org/10.3390/resources10010007.

Carraresi, L., Berg, S., & Broering, S. (2018). Emerging value chains within the bioeconomy: Structural changes in the case of phosphate recovery. *Journal of Cleaner Production*, *183*, 87–101. https://doi.org/10.1016/j.jclepro.2018.02.135.

Cazón, P., Velazquez, G., Ramírez, J. A., & Vázquez, M. (2017). Polysaccharide-based films and coatings for food packaging: A review. *Food Hydrocolloids*, *68*, 136–148. https://doi.org/10.1016/j.foodhyd.2016.09.009.

Chen, L., Tang, C., Ning, N., Wang, C., Fu, Q., & Zhang, Q [Qin] (2009). Preparation and properties of chitosan/lignin composite films. *Chinese Journal of Polymer Science*, *27*(5), 739. https://doi.org/10.1142/s0256767909004448.

Chentir, I., Kchaou, H., Hamdi, M., Jridi, M., Li, S., Doumandji, A., & Nasri, M. (2019). Biofunctional gelatin-based films incorporated with food grade phycocyanin extracted from the Saharian cyanobacterium Arthrospira sp. *Food Hydrocolloids*, *89*, 715–725. https://doi.org/10.1016/j.foodhyd.2018.11.034.

Cho, S. Y., Lee, S. Y., & Rhee, C. (2010). Edible oxygen barrier bilayer film pouches from corn zein and soy protein isolate for olive oil packaging. *LWT - Food Science and Technology*, *43*(8), 1234–1239. https://doi.org/10.1016/j.lwt.2010.03.014.

Ciannamea, E. M., Castillo, L. A., Barbosa, S. E., & Angelis, M. G. de (2018). Barrier properties and mechanical strength of bio-renewable, heat-sealable films based on gelatin, glycerol and soybean oil for sustainable food packaging. *Reactive and Functional Polymers*, *125*, 29–36. https://doi.org/10.1016/j.reactfunctpolym.2018.02.001.

Coelho, P. M., Corona, B., Klooster, R. ten, & Worrell, E. (2020). Sustainability of reusable packaging–Current situation and trends. *Resources, Conservation & Recycling: X*, *6*, Article 100037. https://doi.org/10.1016/j.rcrx.2020.100037.

Coma, V. (2008). Bioactive packaging technologies for extended shelf life of meat-based products. *Meat Science*, *78*(1-2), 90–103. https://doi.org/10.1016/j.meatsci.2007.07.035.

Conte, A., Cappelletti, G. M., Nicoletti, G. M., Russo, C., & Del Nobile, M. A. (2015). Environmental implications of food loss probability in packaging design. *Food Research International*, *78*, 11–17. https://doi.org/10.1016/j.foodres.2015.11.015.

Cooksey, K. (2005). Effectiveness of antimicrobial food packaging materials. *Food Additives and Contaminants*, *22*(10), 980–987. https://doi.org/10.1080/02652030500246164.

Crompton, T. R. (2012). *Physical Testing of Plastics*. Smithers Rapra.

Crouvisier-Urion, K., Bodart, P. R., Winckler, P., Raya, J., Gougeon, R. D., Cayot, P., Domenek, S., Debeaufort, F., & Karbowiak, T. (2016). Bio-based composite films from chitosan and lignin: antioxidant activity related to structure and moisture. *ACS Sustainable Chemistry & Engineering*, *4*(12), 6371–6381. https://doi.org/10.1021/acssuschemeng.6b00956.

Crouvisier-Urion, K., Lagorce-Tachon, A., Lauquin, C., Winckler, P., Tongdeesoontorn, W., Domenek, S., Debeaufort, F., & Karbowiak, T. (2017). Impact of the homogenization process on the structure and antioxidant properties of chitosan-lignin composite films. *Food Chemistry*, *236*, 120–126. https://doi.org/10.1016/j.foodchem.2017.03.094.

Daioglou, V., Wicke, B., Faaij, A. P. C., & van Vuuren, D. P. (2015). Competing uses of biomass for energy and chemicals: implications for long-term global CO 2 mitigation potential. *GCB Bioenergy*, *7*(6), 1321–1334. https://doi.org/10.1111/gcbb.12228.

Dang, K. M., & Yoksan, R. (2015). Development of thermoplastic starch blown film by incorporating plasticized chitosan. *Carbohydrate Polymers*, *115*, 575–581. https://doi.org/10.1016/j.carbpol.2014.09.005.

Dang, K. M., & Yoksan, R. (2016). Morphological characteristics and barrier properties of thermoplastic starch/chitosan blown film. *Carbohydrate Polymers*, *150*, 40–47. https://doi.org/10.1016/j.carbpol.2016.04.113.

Davachi, S. M., & Shekarabi, A. S. (2018). Preparation and characterization of antibacterial, eco-friendly edible nanocomposite films containing Salvia macrosiphon and nanoclay. *International Journal of Biological Macromolecules*, *113*, 66–72. https://doi.org/10.1016/j.ijbiomac.2018.02.106.

Denavi, G., Tapia-Blácido, D. R., Añón, M. C., Sobral, P., Mauri, A. N., & Menegalli, F. C. (2009). Effects of drying conditions on some physical properties of soy protein films. *Journal of Food Engineering*, *90*(3), 341–349. https://doi.org/10.1016/j.jfoodeng.2008.07.001.

Deng, J., Xu, L., Liu, J., Peng, J., Han, Z., Shen, Z [Zhigang], & Guo, S. (2020). Efficient method of recycling carbon fiber from the waste of carbon fiber reinforced polymer composites. *Polymer Degradation and Stability*, *182*, Article 109419. https://doi.org/10.1016/j.polymdegradstab.2020.109419.

Dhall, R. K. (2013). Advances in edible coatings for fresh fruits and vegetables: A review. *Critical Reviews in Food Science and Nutrition*, *53*(5), 435–450. https://doi.org/10.1080/10408398.2010.541568.

Dhifi, W., Bellili, S., Jazi, S., Bahloul, N., & Mnif, W. (2016). Essential Oils' Chemical Characterization and Investigation of Some Biological Activities: A Critical Review. *Medicines*, *3*(4). https://doi.org/10.3390/medicines3040025.

Dilkes-Hoffman, L. S., Lane, J. L., Grant, T., Pratt, S., Lant, P. A., & Laycock, B. (2018). Environmental impact of biodegradable food packaging when considering food waste. *Journal of Cleaner Production*, *180*, 325–334. https://doi.org/10.1016/j.jclepro.2018.01.169.

do Val Siqueira, L., La Arias, C. I. F., Maniglia, B. C., & Tadini, C. C. (2021). Starch-based biodegradable plastics: methods of production, challenges and future perspectives. *Current Opinion in Food Science*, *38*, 122–130. https://doi.org/10.1016/j.cofs.2020.10.020.

Domenek, S., Louaifi, A., Guinault, A., & Baumberger, S. (2013). Potential of Lignins as Antioxidant Additive in Active Biodegradable Packaging Materials. *Journal of Polymers and the Environment*, *21*(3), 692–701. https://doi.org/10.1007/s10924-013-0570-6.

dos Santos Paglione, I., Galindo, M. V., Medeiros, J. A. S. de, Yamashita, F., Alvim, I. D., Ferreira Grosso, C. R., Sakanaka, L. S., & Shirai, M. A. (2019). Comparative study of the properties of soy protein concentrate films containing free and encapsulated oregano essential oil. *Food Packaging and Shelf Life*, *22*, 100419. https://doi.org/10.1016/j.fpsl.2019.100419.

Drieskens, M., Peeters, R., Mullens, J., Franco, D., Lemstra, P. J., & Hristova-Bogaerds, D. G. (2009). Structure versus properties relationship of poly(lactic acid). I. Effect of crystallinity on barrier properties. *Journal of Polymer Science Part B: Polymer Physics*, *47*(22), 2247–2258. https://doi.org/10.1002/polb.21822.

Ellen MacArthur Foundation and McKinsey & Company (2016). The New Plastics Economy: Rethinking the future of plastics. World Economic Forum. http://www.ellenmacarthurfoundation.org/publications.

Elsabee, M. Z., & Abdou, E. S. (2013). Chitosan based edible films and coatings: A review. *Materials Science & Engineering C*, *33*(4), 1819–1841. https://doi.org/10.1016/j.msec.2013.01.010.

Espinoza-Acosta, J. L., Torres-Chávez, P. I., Ramírez-Wong, B., López-Saiz, C. M., & Montaño-Leyva, B. (2016). Antioxidant, Antimicrobial, and Antimutagenic properties of Technical Lignins and Their Applications. *BioResources*, *11*(2), 5452–5481.

European Bioplastics. (2018). *What are bioplastics? Material types, terminology, and labels - an introduction*. https://docs.european-bioplastics.org/publications/fs/EuBP_FS_What_are_bioplastics.pdf.

European Commission (2015). Closing the loop - An EU action plan for the Circular Economy.

European Commission (2018). A European Strategy for Plastics in a Circular Economy.

European Commission (2019). On the implementation of the Circular Economy Action Plan.

European Union (2009). Commission Regulation (EC) No 450/2009 of 29 May 2009 on active and intelligent materials and articles intended to come into contact with food. *Official Journal of the European Union*, *L135/3*.

European Union (2018). Directive (EU) 2018/852 of the European Parliament and of the Council of 30 May 2018 amending Directive 94/62/EC on packaging and packaging waste. *Official Journal of the European Union*, *L 150/141*.

Fabra, M. J., López-Rubio, A., & Lagaron, J. M. (2014). Biopolymers for food packaging applications. In *Smart Polymers and their Applications* (pp. 476–509). Elsevier. https://doi.org/10.1533/9780857097026.2.476.

Fadini, A. L., Rocha, F. S., Alvim, I. D., Sadahira, M. S., Queiroz, M. B., Alves, R., & Silva, L. B. (2013). Mechanical properties and water vapour permeability of hydrolysed collagen–cocoa butter edible films plasticised with sucrose. *Food Hydrocolloids*, *30*(2), 625–631. https://doi.org/10.1016/j.foodhyd.2012.08.011.

Fakhouri, F. M., Martelli, S. M., Caon, T., Velasco, J. I., & Mei, L. H. I. (2015). Edible films and coatings based on starch/gelatin: Film properties and effect of coatings on quality of refrigerated Red Crimson grapes. *Postharvest Biology and Technology*, *109*, 57–64. https://doi.org/10.1016/j.postharvbio.2015.05.015.

Faraca, G., & Astrup, T. (2019). Plastic waste from recycling centres: Characterisation and evaluation of plastic recyclability. *Waste Management*, *95*, 388–398. https://doi.org/10.1016/j.wasman.2019.06.038.

Farmahini-Farahani, M., Khan, A., Lu, P., Bedane, A. H., Eic, M., & Xiao, H. (2017). Surface morphological analysis and water vapor barrier properties of modified Cloisite 30B/poly(3-hydroxybutyrate-co-3-hydroxyvalerate) composites. *Applied Clay Science*, *135*, 27–34. https://doi.org/10.1016/j.clay.2016.08.033.

Farshchi, E., Pirsa, S., Roufegarinejad, L., Alizadeh, M., & Rezazad, M. (2019). Photocatalytic/biodegradable film based on carboxymethyl cellulose, modified by gelatin and TiO2-Ag nanoparticles. *Carbohydrate Polymers*, *216*, 189–196. https://doi.org/10.1016/j.carbpol.2019.03.094.

Farshi, P., Tabibiazar, M., Ghorbani, M., Mohammadifar, M., Amirkhiz, M. B., & Hamishehkar, H. (2019). Whey protein isolate-guar gum stabilized cumin seed oil nanoemulsion. *Food Bioscience*, *28*, 49–56. https://doi.org/10.1016/j.fbio.2019.01.011.

Fasihi, H., Noshirvani, N., Hashemi, M., Fazilati, M., Salavati, H., & Coma, V. (2019). Antioxidant and antimicrobial properties of carbohydrate-based films enriched with cinnamon essential oil by Pickering emulsion method. *Food Packaging and Shelf Life*, *19*, 147–154. https://doi.org/10.1016/j.fpsl.2018.12.007.

Flory, A. R., Vicuna Requesens, D., Devaiah, S. P., Teoh, K. T., Mansfield, S. D., & Hood, E. E. (2013). Development of a green binder system for paper products. *BMC Biotechnology*, *13*, 28. https://doi.org/10.1186/1472-6750-13-28.

Fraj, A., Jaâfar, F., Marti, M., Coderch, L., & Ladhari, N. (2019). A comparative study of oregano (Origanum vulgare L.) essential oil-based polycaprolactone nanocapsules/ microspheres: Preparation, physicochemical characterization, and storage stability. *Industrial Crops and Products*, *140*, 111669. https://doi.org/10.1016/j.indcrop.2019.111669.

Fratzl, P. (2008). Collagen: Structure and Mechanics, an Introduction. In P. Fratzl (Ed.), *Collagen: Structure and mechanics* (pp. 1–13). Springer. https://doi.org/10.1007/978-0-387-73906-9_1.

Galus, S., & Kadzińska, J. (2015). Food applications of emulsion-based edible films and coatings. *Trends in Food Science & Technology*, *45*(2), 273–283. https://doi.org/10.1016/j.tifs.2015.07.011.

Gammariello, D., Incoronato, A. L., Conte, A., & Del Nobile, M. A. (2016). Effect of Sodium Alginate Coating with Ascorbic Acid on Shelf Life of Raw Pork Meat. *Journal of Food Technology Research*, *3*(1), 1–11. https://doi.org/10.18488/journal.58/2016.3.1/58.1.1.11.

Garcia-Arce, J., Trinidad González-Portela Garrido, A., & Prado-Prado, J. C. (2016). Implementing sustainable packaging logistics. An analysis in liquid detergents. *Dirección Y Organización*, *60*, 47–56.

Golembiewski, B., Sick, N., & Broering, S. (2015). The emerging research landscape on bioeconomy: What has been done so far and what is essential from a technology and innovation management perspective? *Innovative Food Science & Emerging Technologies*, *29*(SI), 308–317. https://doi.org/10.1016/j.ifset.2015.03.006.

Gordobil, O., Egüés, I., Llano-Ponte, R., & Labidi, J. (2014). Physicochemical properties of PLA lignin blends. *Polymer Degradation and Stability*, *108*, 330–338. https://doi.org/10.1016/j.polymdegradstab.2014.01.002.

Guzman-Puyol, S., Ceseracciu, L., Tedeschi, G., Marras, S., Scarpellini, A., Benítez, J. J., Athanassiou, A., & Heredia-Guerrero, J. A. (2019). Transparent and Robust All-Cellulose Nanocomposite Packaging Materials Prepared in a Mixture of Trifluoroacetic Acid and Trifluoroacetic Anhydride. *Nanomaterials*, *9*(3). https://doi.org/10.3390/nano9030368.

Haghighi, H., Licciardello, F., Fava, P., Siesler, H. W., & Pulvirenti, A. (2020). Recent advances on chitosan-based films for sustainable food packaging applications. *Food Packaging and Shelf Life*, *26*, 100551. https://doi.org/10.1016/j.fpsl.2020.100551.

Hahladakis, J. N., & Iacovidou, E. (2018). Closing the loop on plastic packaging materials: What is quality and how does it affect their circularity? *Science of the Total Environment*, *630*, 1394–1400. https://doi.org/10.1016/j.scitotenv.2018.02.330.

Hambardzumyan, A., Foulon, L., Bercu, N. B., Pernes, M., Maigret, J. E., Molinari, M., Chabbert, B., & Aguié-Béghin, V. (2015). Organosolv lignin as natural grafting additive to improve the water resistance of films using cellulose nanocrystals. *Chemical Engineering Journal*, *264*, 780–788. https://doi.org/10.1016/j.cej.2014.12.004.

Han, J. H. (2003). Antimicrobial food packaging. In *Novel Food Packaging Techniques* (pp. 50–70). Elsevier. https://doi.org/10.1533/9781855737020.1.50.

Han, J. H. (2005). Antimicrobial packaging systems. In J. H. Han (Ed.), *Food science and technology international series. Innovations in food packaging.* Elsevier.

Han, J. H. (2014). Edible Films and Coatings. In *Innovations in Food Packaging* (pp. 213–255). Elsevier. https://doi.org/10.1016/B978-0-12-394601-0.00009-6.

Hao, C., Liu, T., Zhang, S., Brown, L., Li, R., Xin, J., Zhong, T., Jiang, L., & Zhang, J. (2019). A High-Lignin-Content, Removable, and Glycol-Assisted Repairable Coating Based on Dynamic Covalent Bonds. *ChemSusChem*(12), 1049–1058. https://doi.org/10.1002/cssc.201802615.

Hasheminya, S.-M., Mokarram, R. R., Ghanbarzadeh, B., Hamishekar, H., Kafil, H. S., & Dehghannya, J. (2019). Development and characterization of biocomposite films made from kefiran, carboxymethyl cellulose and Satureja Khuzestanica essential oil. *Food Chemistry*, *289*, 443–452. https://doi.org/10.1016/j.foodchem.2019.03.076.

Hasheminya, S.-M., Rezaei Mokarram, R., Ghanbarzadeh, B., Hamishekar, H., & Kafil, H. S. (2018). Physicochemical, mechanical, optical, microstructural and antimicrobial properties of novel kefiran-carboxymethyl cellulose biocomposite films as influenced by copper oxide nanoparticles (CuONPs). *Food Packaging and Shelf Life*, *17*, 196–204. https://doi.org/10.1016/j.fpsl.2018.07.003.

Hassan, B., Chatha, S. A. S., Hussain, A. I., Zia, K. M., & Akhtar, N. (2018). Recent advances on polysaccharides, lipids and protein based edible films and coatings: A review. *International Journal of Biological Macromolecules*, *109*, 1095–1107. https://doi.org/10.1016/j.ijbiomac.2017.11.097.

Hassan, S. S., Williams, G. A., & Jaiswal, A. K. (2019). Moving Towards the Second Generation of Lignocellulosic Biorefineries in the EU: Drivers, Challenges, and Opportunities. *Renewable Sustainable Energy Reviews*, *101*, 590–599. https://doi.org/10.1016/j.rser.2018.11.041.

Hay, W. T., Fanta, G. F., Peterson, S. C., Thomas, A. J., Utt, K. D., Walsh, K. A., Boddu, V. M., & Selling, G. W. (2018). Improved hydroxypropyl methylcellulose (HPMC) films through incorporation of amylose-sodium palmitate inclusion complexes. *Carbohydrate Polymers*, *188*, 76–84. https://doi.org/10.1016/j.carbpol.2018.01.088.

Heller, M. C., Selke, S. E. M., & Keoleian, G. A. (2019). Mapping the Influence of Food Waste in Food Packaging Environmental Performance Assessments. *Journal of Industrial Ecology*, *23*(2), 480–495. https://doi.org/10.1111/jiec.12743.

Herbert, U., Albrecht, A., & Kreyenschmidt, J. (2015). Definition of predictor variables for MAP poultry filets stored under different temperature conditions. *Poultry Science*, *94*(3), 424–432. https://doi.org/10.3382/ps/peu002.

Herbes, C., Beuthner, C., & Ramme, I. (2018). Consumer attitudes towards biobased packaging - A cross-cultural comparative study. *Journal of Cleaner Production*, *194*, 203–218. https://doi.org/10.1016/j.jclepro.2018.05.106.

Hult, E.-L., Ropponen, J., Poppius-Levlin, K., Ohra-Aho, T., & Tamminen, T. (2013). Enhancing the barrier properties of paper board by a novel lignin coating. *Industrial Crops and Products*, *50*, 694–700. https://doi.org/10.1016/j.indcrop.2013.08.013.

Ilg, Y., & Kreyenschmidt, J. (2012). Review: Nutzen und Risiken der Anwendung antimikrobieller Werkstoffe in der Lebensmittelkette. *Journal of Food Safety and Food Quality*, *63*(2), 28–34.

Ingrao, C., Gigli, M., & Siracusa, V. (2017). An attributional Life Cycle Assessment application experience to highlight environmental hotspots in the production of foamy polylactic acid trays for fresh-food packaging usage. *Journal of Cleaner Production*, *150*, 93–103. https://doi.org/10.1016/j.jclepro.2017.03.007.

Jabeen, N., Majid, I., Nayik, G. A., & Yildiz, F. (2015). Bioplastics and food packaging: A review. *Cogent Food & Agriculture*, *1*(1), Article 1117749. https://doi.org/10.1080/23311932.2015.1117749.

Jamróz, E., Juszczak, L., & Kucharek, M. (2018). Investigation of the physical properties, antioxidant and antimicrobial activity of ternary potato starch-furcellaran-gelatin films incorporated with lavender essential oil. *International Journal of Biological Macromolecules*, *114*, 1094–1101. https://doi.org/10.1016/j.ijbiomac.2018.04.014.

Jang, S.-H., Jang, S.-R., Lee, G.-M., Ryu, J.-H., Park, S.-I., & Park, N.-H. (2017). Halloysite Nanocapsules Containing Thyme Essential Oil: Preparation, Characterization, and Application in Packaging Materials. *Journal of Food Science*, *82*(9), 2113–2120. https://doi.org/10.1111/1750-3841.13835.

Jannatyha, N., Shojaee-Aliabadi, S., Moslehishad, M., & Moradi, E. (2020). Comparing mechanical, barrier and antimicrobial properties of nanocellulose/CMC and nanochitosan/CMC composite films. *International Journal of Biological Macromolecules*, *164*, 2323–2328. https://doi.org/10.1016/j.ijbiomac.2020.07.249.

Javed, A., Ullsten, H., Rättö, P., & Järnström, L. (2018). Lignin-containing coatings for packaging materials. *Nordic Pulp & Paper Research Journal*, *33*(3), 548–556. https://doi.org/10.1515/npprj-2018-3042.

Jiang, B., Chen, C [Chaoji], Liang, Z., He, S., Kuang, Y., Song, J [Jianwei], Mi, R., Chen, G., Jiao, M., & Hu, L. (2020). Lignin as a Wood-Inspired Binder Enabled Strong, Water Stable, and Biodegradable Paper for Plastic Replacement. *Advanced Functional Materials*, *30*(4), 1906307. https://doi.org/10.1002/adfm.201906307.

Jiménez, A [Alberto], Fabra, M. J., Talens, P., & Chiralt, A. (2012). Edible and Biodegradable Starch Films: A Review. *Food and Bioprocess Technology*, *5*(6), 2058–2076. https://doi.org/10.1007/s11947-012-0835-4.

Kadam, D. M., Thunga, M., Wang, S [Sheng], Kessler, M. R., Grewell, D., Lamsal, B., & Yu, C. (2013). Preparation and characterization of whey protein isolate films reinforced with porous silica coated titania nanoparticles. *Journal of Food Engineering*, *117*(1), 133–140. https://doi.org/10.1016/j.jfoodeng.2013.01.046.

Kamari, A., Halim, A. L. A., Yusoff, S. N. M., & Ishak, S. (2018). Gelatin film incorporated with banana leaf essential oil for food preservation. *(Journal of Physics: Conference Series*, *1097*, Article 012047. https://doi.org/10.1088/1742-6596/1097/1/012047.

Kamdem, D. P., Shen, Z [Zhu], Nabinejad, O., & Shu, Z. (2019). Development of biodegradable composite chitosan-based films incorporated with xylan and carvacrol for food packaging application. *Food Packaging and Shelf Life*, *21*, Article 100344. https://doi.org/10.1016/j.fpsl.2019.100344.

Kanatt, S. R. (2020). Development of active/intelligent food packaging film containing Amaranthus leaf extract for shelf life extension of chicken/fish during chilled storage. *Food Packaging and Shelf Life*, *24*, 100506. https://doi.org/10.1016/j.fpsl.2020.100506.

Karimirad, R., Behnamian, M., & Dezhsetan, S. (2018). Development and characterization of nano biopolymer containing cumin oil as a new approach to enhance antioxidant properties of button mushroom. *International Journal of Biological Macromolecules*, *113*, 662–668. https://doi.org/10.1016/j.ijbiomac.2018.02.043.

Keegan, D., Kretschmer, B., Elbersen, B., & Panoutsou, C. (2013). Cascading use: a systematic approach to biomass beyond the energy sector. *Biofuels, Bioproducts and Biorefining*, *7*(2), 193–206. https://doi.org/10.1002/bbb.1351.

Khoobdel, M., Ahsaei, S. M., & Farzaneh, M. (2017). Insecticidal activity of polycaprolactone nanocapsules loaded with Rosmarinus officinalis essential oil in Tribolium castaneum (Herbst). *Entomological Research*, *47*(3), 175–184. https://doi.org/10.1111/1748-5967.12212.

Khui, P. L. N., Rahman, M. R., Jayamani, E., & Bin Bakri, M. K. (2021). Recycling of sustainable polymers and composites. In *Advances in Sustainable Polymer Composites* (pp. 267–282). Elsevier. https://doi.org/10.1016/B978-0-12-820338-5.00012-6.

Kim, H., Beak, S.-E., & Song, K. B. (2018). Development of a hagfish skin gelatin film containing cinnamon bark essential oil. *LWT - Food Science and Technology*, *96*, 583–588. https://doi.org/10.1016/j.lwt.2018.06.016.

Kim, K. H., Jeong, K., Zhuang, J., Jeong, H. J., Kim, C. S., Koo, B., & Yoo, C. G. (2021). Tandem conversion of lignin to catechols via demethylation and catalytic hydrogenolysis. *Industrial Crops and Products*, *159*, Article 113095. https://doi.org/10.1016/j.indcrop.2020.113095.

Klaiman, K., Ortega, D. L., & Garnache, C. (2016). Consumer preferences and demand for packaging material and recyclability. *Resources, Conservation and Recycling*, *115*, 1–8. https://doi.org/10.1016/j.resconrec.2016.08.021.

Klein, F., Emberger-Klein, A., Menrad, K., Moehring, W., & Blesin, J.-M. (2019). Influencing factors for the purchase intention of consumers choosing bioplastic products in Germany. *Sustainable Production and Consumption*, *19*, 33–43. https://doi.org/10.1016/j.spc.2019.01.004.

Klein, S. E., Alzagameem, A., Rumpf, J., Korte, I., Kreyenschmidt, J., & Schulze, M. (2019). Antimicrobial Activity of Lignin-Derived Polyurethane Coatings Prepared from Unmodified and Demethylated Lignins. *Coatings*, *9*(8), 494. https://doi.org/10.3390/coatings9080494.

Klein, S. E., Rumpf, J., Alzagameem, A., Rehahn, M., & Schulze, M. (2019). Antioxidant activity of unmodified kraft and organosolv lignins to be used as sustainable components for polyurethane coatings. *Journal of Coatings Technology and Research*, *55*(5), 97. https://doi.org/10.1007/s11998-019-00201-w.

Koller, M., Sandholzer, D., Salerno, A., Braunegg, G., & Narodoslawsky, M. (2013). Biopolymer from industrial residues: Life cycle assessment of poly(hydroxyalkanoates) from whey. *Resources, Conservation and Recycling*, *73*, 64–71. https://doi.org/10.1016/j.resconrec.2013.01.017.

Kouravand, F., Jooyandeh, H., Barzegar, H., & Hojjati, M. (2018). Characterization of cross-linked whey protein isolate-based films containing Satureja Khuzistanica Jamzad essential oil. *Journal of Food Processing and Preservation*, *42*(3), Article 13557. https://doi.org/10.1111/jfpp.13557.

Koutsimanis, G., Harte, J., & Almenar, E. (2015). Freshness maintenance of cherries ready for consumption using convenient, microperforated, bio-based packaging. *Journal of the Science of Food and Agriculture*, *95*(5), 972–982. https://doi.org/10.1002/jsfa.6771.

Kovalcik, A., Machovsky, M., Kozakova, Z., & Koller, M. (2015). Designing packaging materials with viscoelastic and gas barrier properties by optimized processing of poly(3-hydroxybutyrate-co-3-hydroxyvalerate) with lignin. *Reactive and Functional Polymers*, *94*, 25–34. https://doi.org/10.1016/j.reactfunctpolym.2015.07.001.

Kovalcik, A., Pérez-Camargo, R. A., Fürst, C., Kucharczyk, P., & Müller, A. J. (2017). Nucleating efficiency and thermal stability of industrial non-purified lignins and ultrafine talc in poly(lactic acid) (PLA). *Polymer Degradation and Stability*, *142*, 244–254. https://doi.org/10.1016/j.polymdegradstab.2017.07.009.

Kreyenschmidt, J., Albrecht, A., Braun, C., Herbert, U., Mack, M., Rossaint, S., Ritter, G., Teitscheid, P., & Ilg, Y. (2013). Food Waste in der Fleisch verarbeitenden Kette. *Fleischwirtschaft*, *10*, 57–63.

Küçük, G. S., Çelik, Ö. F., Mazi, B. G., & Türe, H. (2020). Evaluation of alginate and zein films as a carrier of natamycin to increase the shelf life of kashar cheese. *Packaging Technology and Science*, *33*(1), 39–48. https://doi.org/10.1002/pts.2483.

Kumar, A., Tumu, V. R., Ray Chowdhury, S., & S V S, R. R. (2019). A green physical approach to compatibilize a bio-based poly (lactic acid)/lignin blend for better mechanical, thermal and degradation properties. *International Journal of Biological Macromolecules*, *121*, 588–600. https://doi.org/10.1016/j.ijbiomac.2018.10.057.

Kumar, N., Kaur, P., & Bhatia, S. (2017). Advances in bio-nanocomposite materials for food packaging: a review. *Nutrition & Food Science*, *47*(4), 591–606. https://doi.org/10.1108/NFS-11-2016-0176.

Kurek, M., Galus, S., & Debeaufort, F. (2014). Surface, mechanical and barrier properties of bio-based composite films based on chitosan and whey protein. *Food Packaging and Shelf Life*, *1*(1), 56–67. https://doi.org/10.1016/j.fpsl.2014.01.001.

Lambert, S., & Wagner, M. (2017). Environmental performance of bio-based and biodegradable plastics: The road ahead. *Chemical Society Reviews*, *46*(22), 6855–6871. https://doi.org/10.1039/c7cs00149e.

Lan, W., He, L., & Liu, Y. (2018). Preparation and Properties of Sodium Carboxymethyl Cellulose/Sodium Alginate/Chitosan Composite Film. *Coatings*, *8*(8), 291. https://doi.org/10.3390/coatings8080291.

Lange, J., & Wyser, Y. (2003). Recent Innovations in Barrier Technologies for Plastic Packaging - a Review. *Packaging Technology and Science*, *16*(4), 149–158. https://doi.org/10.1002/pts.621.

Lavoine, N., Givord, C., Tabary, N., Desloges, I., Martel, B., & Bras, J. (2014). Elaboration of a new antibacterial bio-nano-material for food-packaging by synergistic action of cyclodextrin and microfibrillated cellulose. *Innovative Food Science & Emerging Technologies*, *26*, 330–340. https://doi.org/10.1016/j.ifset.2014.06.006.

Lee, M. H., Seo, H.-S., & Park, H. J. (2017). Thyme Oil Encapsulated in Halloysite Nanotubes for Antimicrobial Packaging System. *Journal of Food Science*, *82*(4), 922–932. https://doi.org/10.1111/1750-3841.13675.

Li, X., Hegyesi, N., Zhang, Y [Yunchong], Mao, Z., Feng, X [Xueling], Wang, B., Pukánszky, B., & Sui, X. (2019). Poly(lactic acid)/lignin blends prepared with the Pickering emulsion template method. *European Polymer Journal*, *110*, 378–384. https://doi.org/10.1016/j.eurpolymj.2018.12.001.

Li, Y [Ying], Chen, H., Dong, Y., Li, K., Li, L [Li], & Li, J [Jianzhang] (2016). Carbon nanoparticles/soy protein isolate bio-films with excellent mechanical and water barrier properties. *Industrial Crops and Products*, *82*, 133–140. https://doi.org/10.1016/j.indcrop.2015.11.072.

Lin, L., Gu, Y., & Cui, H. (2019). Moringa oil/chitosan nanoparticles embedded gelatin nanofibers for food packaging against Listeria monocytogenes and Staphylococcus aureus on cheese. *Food Packaging and Shelf Life*, *19*, 86–93. https://doi.org/10.1016/j.fpsl.2018.12.005.

Lin, L., Mao, X., Sun, Y., Rajivgandhi, G., & Cui, H. (2019). Antibacterial properties of nanofibers containing chrysanthemum essential oil and their application as beef packaging. *International Journal of Food Microbiology*, *292*, 21–30. https://doi.org/10.1016/j.ijfoodmicro.2018.12.007.

Lin, L., Xue, L., Duraiarasan, S., & Haiying, C. (2018). Preparation of ε-polylysine/chitosan nanofibers for food packaging against Salmonella on chicken. *Food Packaging and Shelf Life*, *17*, 134–141. https://doi.org/10.1016/j.fpsl.2018.06.013.

Lin, L., Zhu, Y [Yulin], & Cui, H. (2018). Electrospun thyme essential oil/gelatin nanofibers for active packaging against Campylobacter jejuni in chicken. *LWT - Food Science and Technology*, *97*, 711–718. https://doi.org/10.1016/j.lwt.2018.08.015.

Lumdubwong, N. (2019). Applications of Starch-Based Films in Food Packaging. In *Reference Module in Food Science.* Elsevier. https://doi.org/10.1016/B978-0-08-100596-5.22481-5.

Ma, P., Hristova-Bogaerds, D. G., Goossens, J., Spoelstra, A. B., Zhang, Y [Yong], & Lemstra, P. J. (2012). Toughening of poly(lactic acid) by ethylene-co-vinyl acetate copolymer with different vinyl acetate contents. *European Polymer Journal*, *48*(1), 146–154. https://doi.org/10.1016/j.eurpolymj.2011.10.015.

Ma, X., Park, C., & Moultrie, J. (2020). Factors for eliminating plastic in packaging: The European FMCG experts' view. *Journal of Cleaner Production*, *256*, Article 120492. https://doi.org/10.1016/j.jclepro.2020.120492.

Maes, C., Luyten, W., Herremans, G., Peeters, R., Carleer, R., & Buntinx, M. (2018). Recent Updates on the Barrier Properties of Ethylene Vinyl Alcohol Copolymer (EVOH): A Review. *Polymer Reviews*, *58*(2), 209–246. https://doi.org/10.1080/15583724.2017.1394323.

Maestrello, C., Tonon, L., Madrona, G., Scapim, M., & Bergamasco, R. (2017). Production and Characterization of Biodegradable Films Incorporated with Clove Essential Oil/β-cyclodextrin Microcapsules. *Chemical Engineering Transactions*, *57*, 1274–1283. https://doi.org/10.3303/CET1757233.

Mahcene, Z., Khelil, A., Hasni, S., Akman, P. K., Bozkurt, F., Birech, K., Goudjil, M. B., & Tornuk, F. (2020). Development and characterization of sodium alginate based active edible films incorporated with essential oils of some medicinal plants. *International Journal of Biological Macromolecules*, *145*, 124–132. https://doi.org/10.1016/j.ijbiomac.2019.12.093.

Mak, T. M. W., Xiong, X., Tsang, D. C. W., Yu, I. K. M., & Poon, C. S. (2020). Sustainable food waste management towards circular bioeconomy: Policy review, limitations and opportunities. *Bioresource Technology*, *297*, Article 122497. https://doi.org/10.1016/j.biortech.2019.122497.

Malathi, A. N., Santhosh, K. S., & Nidoni, U. (2014). Recent trends of Biodegradable polymer: Biodegradable films for Food Packaging and application of Nanotechnology in Biodegradable Food Packaging. *Current Trends in Technology and Science*, *3*(2), 73–79.

Marra, A., Silvestre, C., Duraccio, D., & Cimmino, S. (2016). Polylactic acid/zinc oxide biocomposite films for food packaging application. *International Journal of Biological Macromolecules*, *88*, 254–262. https://doi.org/10.1016/j.ijbiomac.2016.03.039.

Massey, L. K. (2003). *Permeability Properties of Plastics and Elastomers: A Guide to Packaging and Barrier Materials* (2nd ed.). *Plastics Design Library*. Elsevier Science.

Matthews, C., Moran, F., & Jaiswal, A. K. (2021). A Review on European Union's Strategy for Plastics in a Circular Economy and its Impact on Food Safety. *Journal of Cleaner Production*, *283*, Article 125263.

McMillin, K. W. (2017). Advancements in meat packaging. *Meat Science*, *132*, 153–162. https://doi.org/10.1016/j.meatsci.2017.04.015.

Mehdizadeh, T., & Mojaddar Langroodi, A. (2019). Chitosan coatings incorporated with propolis extract and Zataria multiflora Boiss oil for active packaging of chicken breast meat. *International Journal of Biological Macromolecules*, *141*, 401–409. https://doi.org/10.1016/j.ijbiomac.2019.08.267.

Mena, C., Adenso-Diaz, B., & Yurt, O. (2011). The causes of food waste in the supplier–retailer interface: Evidences from the UK and Spain. *Resources, Conservation and Recycling*, *55*(6), 648–658. https://doi.org/10.1016/j.resconrec.2010.09.006.

Mendes, F. R., Bastos, M. S., Mendes, L. G., Silva, A. R., Sousa, F. D., Monteiro-Moreira, A. C., Cheng, H. N., Biswas, A., & Moreira, R. A. (2017). Preparation and evaluation of hemicellulose films and their blends. *Food Hydrocolloids*, *70*, 181–190. https://doi.org/10.1016/j.foodhyd.2017.03.037.

Merino, N., Berdejo, D., Bento, R., Salman, H., Lanz, M., Maggi, F., Sánchez-Gómez, S., García-Gonzalo, D., & Pagán, R. (2019). Antimicrobial efficacy of Thymbra capitata (L.) Cav. essential oil loaded in self-assembled zein nanoparticles in combination with heat. *Industrial Crops and Products*, *133*, 98–104. https://doi.org/10.1016/j.indcrop.2019.03.003.

Michelin, M., Marques, A. M., Pastrana, L. M., Teixeira, J. A., & Cerqueira, M. A. (2020). Carboxymethyl cellulose-based films: Effect of organosolv lignin incorporation on physicochemical and antioxidant properties. *Journal of Food Engineering*, *285*, Article 110107. https://doi.org/10.1016/j.jfoodeng.2020.110107.

Miranda, C. S., Ferreira, M. S., Magalhães, M. T., Bispo, A. P. G., Oliveira, J. C., Silva, J. B., & José, N. M. (2015). Starch-based Films Plasticized with Glycerol and Lignin from Piassava Fiber Reinforced with Nanocrystals from Eucalyptus. *Materials Today: Proceedings*, *2*(1), 134–140. https://doi.org/10.1016/j.matpr.2015.04.038.

Miranda, C. S., Ferreira, M. S., Magalhães, M. T., Santos, W. J., Oliveira, J. C., Silva, J. B., & José, N. M. (2015). Mechanical, Thermal and Barrier Properties of Starch-based Films Plasticized with Glycerol and Lignin and Reinforced with Cellulose Nanocrystals. *Materials Today: Proceedings*, *2*(1), 63–69. https://doi.org/10.1016/j.matpr.2015.04.009.

Mohsenabadi, N., Rajaei, A., Tabatabaei, M., & Mohsenifar, A. (2018). Physical and antimicrobial properties of starch-carboxy methyl cellulose film containing rosemary essential oils encapsulated in chitosan nanogel. *International Journal of Biological Macromolecules*, *112*, 148–155. https://doi.org/10.1016/j.ijbiomac.2018.01.034.

Mojaddar Langroodi, A., Tajik, H., Mehdizadeh, T., Moradi, M., Moghaddas Kia, E., & Mahmoudian, A. (2018). Effects of sumac extract dipping and chitosan coating enriched with Zataria multiflora Boiss oil on the shelf-life of meat in modified atmosphere packaging. *LWT - Food Science and Technology*, *98*, 372–380. https://doi.org/10.1016/j.lwt.2018.08.063.

Mojumdar, S. C., Moresoli, C., Simon, L. C., & Legge, R. L. (2011). Edible wheat gluten (WG) protein films. *Journal of Thermal Analysis and Calorimetry*, *104*(3), 929–936. https://doi.org/10.1007/s10973-011-1491-z.

Molavi, H., Behfar, S., Ali Shariati, M., Kaviani, M., & Atarod, S. (2015). A review on biodegradable starch based film. *Journal of Microbiology, Biotechnology and Food Sciences*, *4*(5), 456–461. https://doi.org/10.15414/jmbfs.2015.4.5.456-461.

Molina-Besch, K., Wikström, F., & Williams, H. (2019). The environmental impact of packaging in food supply chains—does life cycle assessment of food provide the full picture? *The International Journal of Life Cycle Assessment*, *24*(1), 37–50. https://doi.org/10.1007/s11367-018-1500-6.

Morganti, P., & Stoller, M. (2017). Chitin And Lignin: Natural Ingredients From Waste Materials To Make Innovative And Healthy Products For Humans And Plant. *Chemical Engineering Transactions*, *60*, 319–324. https://doi.org/10.3303/CET1760054.

Moura, M. R. de, Avena-Bustillos, R. J., McHugh, T. H., Krochta, J. M., & Mattoso, L. H. C. (2008). Properties of novel hydroxypropyl methylcellulose films containing chitosan nanoparticles. *Journal of Food Science*, *73*(7), 31-37. https://doi.org/10.1111/j.1750-3841.2008.00872.x.

Mousavi Khaneghah, A., Hashemi, S. M. B., & Limbo, S. (2018). Antimicrobial agents and packaging systems in antimicrobial active food packaging: An overview of approaches and interactions. *Food and Bioproducts Processing*, *111*, 1–19. https://doi.org/10.1016/j.fbp.2018.05.001.

Mulakkal, M. C., Castillo Castillo, A., Taylor, A. C., Blackman, B. R., Balint, D. S., Pimenta, S., & Charalambides, M. N. (2021). Advancing mechanical recycling of multilayer plastics through finite element modelling and environmental policy. *Resources, Conservation and Recycling*, *166*, Article 105371. https://doi.org/10.1016/j.resconrec.2020.105371.

Müller, G., Hanecker, E., Blasius, K., Seidemann, C., Tempel, L., Sadocco, P., Pozo, B. F., Boulougouris, G., Lozo, B., Jamnicki, S., & Bobu, E. (2014). End-of-life Solutions for Fibre and Bio-based Packaging Materials in Europe. *Packaging Technology and Science*, *27*(1), 1–15. https://doi.org/10.1002/pts.2006.

Mumbach, G. D., Bolzan, A., & Machado, R. A. F. (2020). A closed-loop process design for recycling expanded polystyrene waste by dissolution and polymerization. *Polymer*, *209*, Article 122940. https://doi.org/10.1016/j.polymer.2020.122940.

Muscat, D., Adhikari, B., Adhikari, R., & Chaudhary, D. S. (2012). Comparative study of film forming behaviour of low and high amylose starches using glycerol and xylitol as plasticizers. *Journal of Food Engineering*, *109*(2), 189–201. https://doi.org/10.1016/j.jfoodeng.2011.10.019.

Narodoslawsky, M., Shazad, K., & Kollmann, R: Schnitzer, H. (2015). LCA of PHA Production – Identifying the Ecological Potential of Bio-plastic. *Chemical and Biochemical Engineering Quarterly*, *29*(2), 299–305. https://doi.org/10.15255/CABEQ.2014.2262.

Nazan Turhan, K., & Şahbaz, F. (2004). Water vapor permeability, tensile properties and solubility of methylcellulose-based edible films. *Journal of Food Engineering*, *61*(3), 459–466. https://doi.org/10.1016/S0260-8774(03)00155-9.

Nieto, G. (2017). Biological Activities of Three Essential Oils of the Lamiaceae Family. *Medicines*, *4*(3). https://doi.org/10.3390/medicines4030063.

Nisar, T., Wang, Z.-C., Yang, X., Tian, Y., Iqbal, M., & Guo, Y. (2018). Characterization of citrus pectin films integrated with clove bud essential oil: Physical, thermal, barrier, antioxidant and antibacterial properties. *International Journal of Biological Macromolecules*, *106*, 670–680. https://doi.org/10.1016/j.ijbiomac.2017.08.068.

Noshirvani, N., Ghanbarzadeh, B., Gardrat, C., Rezaei, M. R., Hashemi, M., Le Coz, C., & Coma, V. (2017). Cinnamon and ginger essential oils to improve antifungal, physical and mechanical properties of chitosan-carboxymethyl cellulose films. *Food Hydrocolloids*, *70*, 36–45. https://doi.org/10.1016/j.foodhyd.2017.03.015.

Nur Hanani, Z. A., O'Mahony, J. A., Roos, Y. H., Oliveira, P. M., & Kerry, J. P. (2014). Extrusion of gelatin-based composite films: Effects of processing temperature and pH of film forming solution on mechanical and barrier properties of manufactured films. *Food Packaging and Shelf Life*, *2*(2), 91–101. https://doi.org/10.1016/j.fpsl.2014.09.001.

Nur Hanani, Z. A., Roos, Y. H., & Kerry, J. P. (2014). Use and application of gelatin as potential biodegradable packaging materials for food products. *International Journal of Biological Macromolecules*, *71*, 94–102. https://doi.org/10.1016/j.ijbiomac.2014.04.027.

Oechsle, A. M., Bugbee, T. J., Gibis, M., Kohlus, R., & Weiss, J. (2017). Modification of extruded chicken collagen films by addition of co-gelling protein and sodium chloride. *Journal of Food Engineering*, *207*, 46–55. https://doi.org/10.1016/j.jfoodeng.2017.03.017.

Ojogbo, E., Ogunsona, E. O., & Mekonnen, T. H. (2020). Chemical and physical modifications of starch for renewable polymeric materials. *Materials Today Sustainability*, *7-8*, Article 100028. https://doi.org/10.1016/j.mtsust.2019.100028.

Oliveira, S. P. L. F., Bertan, L. C., Rensis, C. M. V. B. de, Bilck, A. P., & Vianna, P. C. B. (2017). Whey protein-based films incorporated with oregano essential oil. *Polímeros*, *27*(2), 158–164. https://doi.org/10.1590/0104-1428.02016.

Oliveira Filho, J. G. de, Rodrigues, J. M., Valadares, A. C. F., Almeida, A. B. de, Lima, T. M. de, Takeuchi, K. P., Alves, C. C. F., Sousa, H. A. d. F., Da Silva, E. R., Dyszy, F. H., & Egea, M. B. (2019). Active food packaging: Alginate films with cottonseed protein hydrolysates. *Food Hydrocolloids*, *92*, 267–275. https://doi.org/10.1016/j.foodhyd.2019.01.052.

Otoni, C. G., Avena-Bustillos, R. J., Olsen, C. W., Bilbao-Sáinz, C., & McHugh, T. H. (2016). Mechanical and water barrier properties of isolated soy protein composite edible films as affected by carvacrol and cinnamaldehyde micro and nanoemulsions. *Food Hydrocolloids*, *57*, 72–79. https://doi.org/10.1016/j.foodhyd.2016.01.012.

Panseri, S., Martino, P. A., Cagnardi, P., Celano, G., Tedesco, D., Castrica, M., Balzaretti, C., & Chiesa, L. M. (2018). Feasibility of biodegradable based packaging used for red meat storage during shelf-life: A pilot study. *Food Chemistry*, *249*, 22–29. https://doi.org/10.1016/j.foodchem.2017.12.067.

Pauer, E., Tacker, M., Gabriel, V., & Krauter, V. (2020). Sustainability of flexible multilayer packaging: Environmental impacts and recyclability of packaging for bacon in block. *Cleaner Environmental Systems*, *1*, Article 100001. https://doi.org/10.1016/j.cesys.2020.100001.

Peelman, N., Ragaert, P., Meulenaer, B. de, Adons, D., Peeters, R., Cardon, L., van Impe, F., & Devlieghere, F. (2013). Application of bioplastics for food packaging. *Trends in Food Science & Technology*, *32*(2), 128–141. https://doi.org/10.1016/j.tifs.2013.06.003.

Pereira, M. C., Hill, L. E., Zambiazi, R. C., Mertens-Talcott, S., Talcott, S., & Gomes, C. L. (2015). Nanoencapsulation of hydrophobic phytochemicals using poly (dl-lactide-co-glycolide) (PLGA) for antioxidant and antimicrobial delivery applications: Guabiroba fruit (Campomanesia xanthocarpa O. Berg) study. *LWT - Food Science and Technology*, *63*(1), 100–107. https://doi.org/10.1016/j.lwt.2015.03.062.

Petljak, K., Naletina, D., & Bilogrević, K. (2019). Considering ecologically sustainable packaging during decision-making while buying food products. *Ekonomika Poljoprivrede*, *66*(1), 107–126. https://doi.org/10.5937/ekoPolj1901107P.

Pettersen, M. K., Grøvlen, M. S., Evje, N., & Radusin, T. (2020). Recyclable mono materials for packaging of fresh chicken fillets: New design for recycling in circular economy. *Packaging Technology and Science*, *33*(11), 485–498. https://doi.org/10.1002/pts.2527.

Piñeros-Hernandez, D., Medina-Jaramillo, C., López-Córdoba, A., & Goyanes, S. (2017). Edible cassava starch films carrying rosemary antioxidant extracts for potential use as active food packaging. *Food Hydrocolloids*, *63*, 488–495. https://doi.org/10.1016/j.foodhyd.2016.09.034.

PlasticsEurope (2019). Plastics - the Facts 2019: An analysis of European plastics production, demand and waste data.

Popović, S., Peričin, D., Vaštag, Ž., Lazić, V., & Popović, L. (2012). Pumpkin oil cake protein isolate films as potential gas barrier coating. *Journal of Food Engineering*, *110*(3), 374–379. https://doi.org/10.1016/j.jfoodeng.2011.12.035.

Popović, S. Z., Lazić, V. L., Hromiš, N. M., Šuput, D. Z., & Bulut, S. N. (2018). Biopolymer Packaging Materials for Food Shelf-Life Prolongation. In *Biopolymers for Food Design* (pp. 223–277). Elsevier. https://doi.org/10.1016/B978-0-12-811449-0.00008-6.

Priyadarshi, R., & Rhim, J.-W. (2020). Chitosan-based biodegradable functional films for food packaging applications. *Innovative Food Science & Emerging Technologies*, *62*, Article 102346. https://doi.org/10.1016/j.ifset.2020.102346.

Priyadarshi, R., Sauraj, Kumar, B., Deeba, F., Kulshreshtha, A., & Negi, Y. S. (2018). Chitosan films incorporated with Apricot (Prunus armeniaca) kernel essential oil as active food packaging material. *Food Hydrocolloids*, *85*, 158–166. https://doi.org/10.1016/j.foodhyd.2018.07.003.

Pro Carton (2010). Wie wichtig ist nachhaltige Verpackung? Die Einstellung von Konsumenten zu Verpackung und Nachhaltigkeit, *3*.

Qamar, S. A., Asgher, M., & Bilal, M. (2020). Immobilization of Alkaline Protease From Bacillus brevis Using Ca-Alginate Entrapment Strategy for Improved Catalytic Stability, Silver Recovery, and Dehairing Potentialities. *Catalysis Letters*, *150*(12), 3572–3583. https://doi.org/10.1007/s10562-020-03268-y.

Qin, Y., Li, W., Liu, D., Yuan, M., & Li, L [Lin] (2017). Development of active packaging film made from poly (lactic acid) incorporated essential oil. *Progress in Organic Coatings*, *103*, 76–82. https://doi.org/10.1016/j.porgcoat.2016.10.017.

Qiu, C., Chang, R., Yang, J., Ge, S [Shengju], Xiong, L., Zhao, M., Li, M., & Sun, Q. (2016). Preparation and characterization of essential oil-loaded starch nanoparticles formed by short glucan chains. *Food Chemistry*, *221*, 1426–1433. https://doi.org/10.1016/j.foodchem.2016.11.009.

Raafat, D., & Sahl, H.-G. (2009). Chitosan and its antimicrobial potential—a critical literature survey. *Microbial Biotechnology*, *2*(2), 186–201. https://doi.org/10.1111/j.1751-7915.2008.00080.x.

Ragaert, K., Delva, L., & van Geem, K. (2017). Mechanical and chemical recycling of solid plastic waste. *Waste Management (New York, N.Y.)*, *69*, 24–58. https://doi.org/10.1016/j.wasman.2017.07.044.

Ragaert, P., Buntinx, M., Maes, C., Vanheusden, C., Peeters, R., Wang, S [Sisi], D'hooge, D. R., & Cardon, L. Polyhydroxyalkanoates for Food Packaging Applications. In *Reference Module in Food Science.* https://doi.org/10.1016/B978-0-08-100596-5.22502-X (Original work published 2019).

Rai, S., Dutta, P. K., & Mehrotra, G. K. (2017). Lignin Incorporated Antimicrobial Chitosan Film for Food Packaging Application. *Journal of Polymer Materials*, *34*(1), 171–183.

Ramos, Ó. L., Silva, S. I., Soares, J. C., Fernandes, J. C., Poças, M. F., Pintado, M. E., & Malcata, F. X. (2012). Features and performance of edible films, obtained from whey protein isolate formulated with antimicrobial compounds. *Food Research International*, *45*(1), 351–361. https://doi.org/10.1016/j.foodres.2011.09.016.

Ranjbar, M., Azizi, M. H., & Hashtjin, A. M. (2017). Evaluation of Physico-Mechanical and Antimicrobial Properties of GelatinCarboxymethyl Cellulose Film Containing Essential Oil of Bane (Pistacia atlantica). *Nutrition and Food Sciences Research*, *4*(3), 11–17.

Raquez, J.-M., Habibi, Y., Murariu, M., & Dubois, P. (2013). Polylactide (PLA)-based nanocomposites. *Progress in Polymer Science*, *38*(10-11), 1504–1542. https://doi.org/10.1016/j.progpolymsci.2013.05.014.

Rasal, R. M., Janorkar, A. V., & Hirt, D. E. (2010). Poly(lactic acid) modifications. *Progress in Polymer Science*, *35*(3), 338–356. https://doi.org/10.1016/j.progpolymsci.2009.12.003.

Rashed, M. M., Zhang, C., Ghaleb, A. D., Li, J [JingPeng], Nagi, A., Majeed, H., Bakry, A. M., Haider, J., Xu, Z., & Tong, Q. (2019). Techno-functional properties and sustainable application of nanoparticles-based Lavandula angustifolia essential oil fabricated using unsaturated lipid-carrier and biodegradable wall material. *Industrial Crops and Products*, *136*, 66–76. https://doi.org/10.1016/j.indcrop.2019.04.070.

Rastogi, V., & Samyn, P. (2015). Bio-Based Coatings for Paper Applications. *Coatings*, *5*(4), 887–930. https://doi.org/10.3390/coatings5040887.

Raut, J. S., & Karuppayil, S. M. (2014). A status review on the medicinal properties of essential oils. *Industrial Crops and Products*, *62*, 250–264. https://doi.org/10.1016/j.indcrop.2014.05.055.

Rehman, A., Jafari, S. M., Aadil, R. M., Assadpour, E., Randhawa, M. A., & Mahmood, S. (2020). Development of active food packaging via incorporation of biopolymeric nanocarriers containing essential oils. *Trends in Food Science & Technology*, *101*, 106–121. https://doi.org/10.1016/j.tifs.2020.05.001.

Renders, T., van den Bossche, G., Vangeel, T., van Aelst, K., & Sels, B. (2019). Reductive catalytic fractionation: State of the art of the lignin-first biorefinery. *Current Opinion in Biotechnology*, *56*, 193–201. https://doi.org/10.1016/j.copbio.2018.12.005.

Rezaeigolestani, M., Misaghi, A., Khanjari, A., Basti, A. A., Abdulkhani, A., & Fayazfar, S. (2017). Antimicrobial evaluation of novel poly-lactic acid based nanocomposites incorporated with bioactive compounds in-vitro and in refrigerated vacuum-packed cooked sausages. *International Journal of Food Microbiology*, *260*, 1–10. https://doi.org/10.1016/j.ijfoodmicro.2017.08.006.

Rhim, J.-W., Hong, S.-I., & Ha, C.-S. (2009). Tensile, water vapor barrier and antimicrobial properties of PLA/nanoclay composite films. *LWT - Food Science and Technology*, *42*(2), 612–617. https://doi.org/10.1016/j.lwt.2008.02.015.

Rhim, J.-W., Park, H.-M., & Ha, C.-S. (2013). Bio-nanocomposites for food packaging applications. *Progress in Polymer Science*, *38*(10-11), 1629–1652. https://doi.org/10.1016/j.progpolymsci.2013.05.008.

Rinaldi, R., Jastrzebski, R., Clough, M. T., Ralph, J., Kennema, M., Bruijnincx, P. C. A., & Weckhuysen, B. M. (2016). Paving the Way for Lignin Valorisation: Recent Advances in Bioengineering, Biorefining and Catalysis. *Angewandte Chemie International Edition*, *55*(29), 8164–8215. https://doi.org/10.1002/anie.201510351.

Rossaint, S., & Kreyenschmidt, J. (2014). Intelligent label – a new way to support food waste reduction. *Waste and Resource Management*, *168*(2), Article 1300035, 63–71. https://doi.org/10.1680/warm.13.00035.

Rossi, A. M., & Hinrichs, C. C. (2011). Hope and skepticism: Farmer and local community views on the socio-economic benefits of agricultural bioenergy. *Biomass and Bioenergy*, *35*(4), 1418–1428. https://doi.org/10.1016/j.biombioe.2010.08.036.

Roy, S., & Rhim, J.-W. (2020). Effect of CuS reinforcement on the mechanical, water vapor barrier, UV-light barrier, and antibacterial properties of alginate-based composite films. *International Journal of Biological Macromolecules*, *164*, 37–44. https://doi.org/10.1016/j.ijbiomac.2020.07.092.

Ruiz-Navajas, Y., Viuda-Martos, M., Sendra, E., Perez-Alvarez, J. A., & Fernández-López, J. (2013). In vitro antibacterial and antioxidant properties of chitosan edible films incorporated with Thymus moroderi or Thymus piperella essential oils. *Food Control*, *30*(2), 386–392. https://doi.org/10.1016/j.foodcont.2012.07.052.

Rumm, S. (2016). *Verbrauchereinschätzungen zu Biokunststoffen: eine Analyse vor dem Hintergrund des heuristic-systematic model*. Dissertation. https://mediatum.ub.tum.de/1306582.

Rumpf, J., Do, X. T., Burger, R., Monakhova, Y. B., & Schulze, M. (2020). Extraction of High-Purity Lignins via Catalyst-free Organosolv Pulping from Low-Input Crops. *Biomacromolecules*, *21*(5), 1929–1942. https://doi.org/10.1021/acs.biomac.0c00123.

Saha, T., Hoque, M. E., & Mahbub, T. (2020). Biopolymers for Sustainable Packaging in Food, Cosmetics, and Pharmaceuticals. In *Advanced Processing, Properties, and Applications of Starch and Other Bio-Based Polymers* (pp. 197–214). Elsevier. https://doi.org/10.1016/B978-0-12-819661-8.00013-5.

Sangsuwan, J., & Sutthasupa, S. (2019). Effect of chitosan and alginate beads incorporated with lavender, clove essential oils, and vanillin against Botrytis cinerea and their application in fresh table grapes packaging system. *Packaging Technology and Science*, *32*(12), 595–605. https://doi.org/10.1002/pts.2476.

Saral Sarojini, K., Indumathi, M. P., & Rajarajeswari, G. R. (2019). Mahua oil-based polyurethane/chitosan/nano ZnO composite films for biodegradable food packaging applications. *International Journal of Biological Macromolecules*, *124*, 163–174. https://doi.org/10.1016/j.ijbiomac.2018.11.195.

Scherer, C., Emberger-Klein, A., & Menrad, K. (2017). Biogenic product alternatives for children: Consumer preferences for a set of sand toys made of bio-based plastic. *Sustainable Production and Consumption*, *10*, 1–14. https://doi.org/10.1016/j.spc.2016.11.001.

Scherer, C., Emberger-Klein, A., & Menrad, K. (2018). Consumer preferences for outdoor sporting equipment made of biobased plastics: Results of a choice-based-conjoint experiment in Germany. *Journal of Cleaner Production*, *203*, 1085–1094. https://doi.org/10.1016/j.jclepro.2018.08.298.

Scherhaufer, S., Moates, G., Hartikainen, H., Waldron, K., & Obersteiner, G. (2018). Environmental impacts of food waste in Europe. *Waste Management*, *77*, 98–113. https://doi.org/10.1016/j.wasman.2018.04.038.

Schumann, B., & Schmid, M. (2018). Packaging concepts for fresh and processed meat – Recent progresses. *Innovative Food Science & Emerging Technologies*, *47*(2), 88–100. https://doi.org/10.1016/j.ifset.2018.02.005.

Schutyser, W., Renders, T., van den Bosch, S., Koelewijn, S.-F., Beckham, G. T., & Sels, B. F. (2018). Chemicals from lignin: An interplay of lignocellulose fractionation, depolymerisation, and upgrading. *Chemical Society Reviews*, *47*(3), 852–908. https://doi.org/10.1039/c7cs00566k.

Sebti, I., Chollet, E., Degraeve, P., Noel, C., & Peyrol, E. (2007). Water sensitivity, antimicrobial, and physicochemical analyses of edible films based on HPMC and/or chitosan. *Journal of Agricultural and Food Chemistry*, *55*(3), 693–699. https://doi.org/10.1021/jf062013n.

Sedlaříková, J., Doležalová, M., Egner, P., Pavlačková, J., Krejčí, J., Rudolf, O., & Peer, P. (2017). Effect of Oregano and Marjoram Essential Oils on the Physical and Antimicrobial Properties of Chitosan Based Systems. *International Journal of Polymer Science*, *2017*, 1–12. https://doi.org/10.1155/2017/2593863.

Šešlija, S., Nešić, A., Škorić, M. L., Krušić, M. K., Santagata, G., & Malinconico, M. (2018). Pectin/Carboxymethylcellulose Films as a Potential Food Packaging Material. *Macromolecular Symposia*, *378*(1), Article 1600163. https://doi.org/10.1002/masy.201600163.

Shahid-Ul-Islam, & Butola, B. S. (2019). Recent advances in chitosan polysaccharide and its derivatives in antimicrobial modification of textile materials. *International Journal of Biological Macromolecules*, *121*, 905–912. https://doi.org/10.1016/j.ijbiomac.2018.10.102.

Shankar, S., Reddy, J. P., & Rhim, J.-W. (2015). Effect of lignin on water vapor barrier, mechanical, and structural properties of agar/lignin composite films. *International Journal of Biological Macromolecules*, *81*, 267–273. https://doi.org/10.1016/j.ijbiomac.2015.08.015.

Sharaf, O. M., Al-Gamal, M. S., Ibrahim, G. A., Dabiza, N. M., Salem, S. S., El-Ssayad, M. F., & Youssef, A. M. (2019). Evaluation and characterization of some protective culture metabolites in free and nano-chitosan-loaded forms against common contaminants of Egyptian cheese. *Carbohydrate Polymers*, *223*, Article 115094. https://doi.org/10.1016/j.carbpol.2019.115094.

Sharma, R., & Ghoshal, G. (2018). Emerging trends in food packaging. *Nutrition & Food Science*, *48*(5), 764–779. https://doi.org/10.1108/NFS-02-2018-0051.

Sharma, S., Barkauskaite, S., Duffy, B., Jaiswal, A. K., & Jaiswal, S. (2020). Characterization and Antimicrobial Activity of Biodegradable Active Packaging Enriched with Clove and Thyme Essential Oil for Food Packaging Application. *Foods*, *9*(8). https://doi.org/10.3390/foods9081117.

Sharma, S., Barkauskaite, S., Jaiswal, S., Duffy, B., & Jaiswal, A. K. (2020). Development of Essential Oil Incorporated Active Film Based on Biodegradable Blends of Poly (Lactide)/Poly (Butylene Adipate-co-Terephthalate) for Food Packaging Application. *Journal of Packaging Technology and Research*, *4*(3), 235–245. https://doi.org/10.1007/s41783-020-00099-5.

Shen, Z [Zhu], & Kamdem, D. P. (2015). Development and characterization of biodegradable chitosan films containing two essential oils. *International Journal of Biological Macromolecules*, *74*, 289–296. https://doi.org/10.1016/j.ijbiomac.2014.11.046.

Sijtsema, S. J., Onwezen, M. C., Reinders, M. J., Dagevos, H., Partanen, A., & Meeusen, M. (2016). Consumer perception of bio-based products-An exploratory study in 5 European countries. *Wageningen Journal of Life Sciences*, *77*(SI), 61–69. https://doi.org/10.1016/j.njas.2016.03.007.

Siracusa, V., Blanco, I., Romani, S., Tylewicz, U., Rocculi, P., & Rosa, M. D. (2012). Poly(lactic acid)-modified films for food packaging application: Physical, mechanical, and barrier behavior. *Journal of Applied Polymer Science*, *125*(S2), E390-E401. https://doi.org/10.1002/app.36829.

Siracusa, V., Ingrao, C., Karpova, S. G., Olkhov, A. A., & Iordanskii, A. L. (2017). Gas transport and characterization of poly(3 hydroxybutyrate) films. *European Polymer Journal*, *91*, 149–161. https://doi.org/10.1016/j.eurpolymj.2017.03.047.

Sleenhoff, S., Cuppen, E., & Osseweijer, P. (2015). Unravelling emotional viewpoints on a bio-based economy using Q methodology. *Public Understanding of Science*, *24*(7), 858–877. https://doi.org/10.1177/0963662513517071.

Sogut, E., & Seydim, A. C. (2018). The effects of Chitosan and grape seed extract-based edible films on the quality of vacuum packaged chicken breast fillets. *Food Packaging and Shelf Life*, *18*, 13–20. https://doi.org/10.1016/j.fpsl.2018.07.006.

Sohail, M., Sun, D.-W., & Zhu, Z. (2018). Recent Developments in Intelligent Packaging for Enhancing Food Quality and Safety. *Critical Reviews in Food Science and Nutrition*, *58*(15), 2650–2662. https://doi.org/10.1080/10408398.2018.1449731.

Soltani Firouz, M., Mohi-Alden, K., & Omid, M. (2021). A critical review on intelligent and active packaging in the food industry: Research and development. *Food Research International*, *141*, Article 110113. https://doi.org/10.1016/j.foodres.2021.110113.

Song, X., Zuo, G., & Chen, F. (2018). Effect of essential oil and surfactant on the physical and antimicrobial properties of corn and wheat starch films. *International Journal of Biological Macromolecules*, *107*, 1302–1309. https://doi.org/10.1016/j.ijbiomac.2017.09.114.

Spiridon, I., & Tanase, C. E. (2018). Design, characterization and preliminary biological evaluation of new lignin-PLA biocomposites. *International Journal of Biological Macromolecules*, *114*, 855–863. https://doi.org/10.1016/j.ijbiomac.2018.03.140.

Srivastava, J., & Vankar, P. S. (2012). Principal phenolic phytochemicals and antioxidant property in Eucalyptus bark. *Nutrition & Food Science*, *42*(6), 412–421. https://doi.org/10.1108/00346651211277663.

Statista (2019). Recycling rate of plastic packaging waste in the European Union (EU-28) from 2006 to 2017.

Statista (2020a). Recyclingquote verschiedener Abfallarten in der EU im Jahr 2017.

Statista (2020b). Recyclingquoten von Altpapier in Europa in den Jahren 1991 bis 2019.

Steinbuchel, A. (2003). *Biopolymers: General aspects and special applications*. *10*. Wiley-VCH.

Stern, T., Ploll, U., Spies, R., Schwarzbauer, P., Hesser, F., & Ranacher, L. (2018). Understanding Perceptions of the Bioeconomy in Austria-An Explorative Case Study. *Sustainability*, *10*(11). https://doi.org/10.3390/su10114142.

Sun, H., Guo, G., Memon, S. A., Xu, W., Zhang, Q [Qiwu], Zhu, J.-H., & Xing, F. (2015). Recycling of carbon fibers from carbon fiber reinforced polymer using electrochemical method. *Composites Part a: Applied Science and Manufacturing*, *78*, 10–17. https://doi.org/10.1016/j.compositesa.2015.07.015.

Syahida, N., Fitry, I., Zuriyati, A., & Hanani, N. (2020). Effects of palm wax on the physical, mechanical and water barrier properties of fish gelatin films for food packaging application. *Food Packaging and Shelf Life*, *23*, 100437. https://doi.org/10.1016/j.fpsl.2019.100437.

Taleb, M. A., & Al Farooque, O. (2021). Towards a Circular Economy for Sustainable Development: An Application of Full Cost Accounting to Municipal Waste Recyclables. *Journal of Cleaner Production*, *280*, Article 124047. https://doi.org/10.1016/j.jclepro.2020.124047.

Tawakkal, I. S. M. A., Cran, M. J., Miltz, J., & Bigger, S. W. (2014). A review of poly(lactic acid)-based materials for antimicrobial packaging. *Journal of Food Science*, *79*(8), 1477-90. https://doi.org/10.1111/1750-3841.12534.

Thakur, R., Pristijono, P., Scarlett, C. J., Bowyer, M., Singh, S. P., & Vuong, Q. V. (2019). Starch-based films: Major factors affecting their properties. *International Journal of Biological Macromolecules*, *132*, 1079–1089. https://doi.org/10.1016/j.ijbiomac.2019.03.190.

Thakur, S., Chaudhary, J., Sharma, B., Verma, A., Tamulevicius, S., & Thakur, V. K. (2018). Sustainability of bioplastics: Opportunities and challenges. *Current Opinion in Green and Sustainable Chemistry*, *13*, 68–75. https://doi.org/10.1016/j.cogsc.2018.04.013.

Theinsathid, P., Chandrachai, A., Suwannathep, S., & Keeratipibul, S. (2011). Lead Users and Early Adoptors of Bioplastics: A Market-Led Approach to Innovative Food Packaging Films. *Journal of Biobased Materials and Bioenergy*, *5*(1), 17–29. https://doi.org/10.1166/jbmb.2011.1128.

Tribot, A., Amer, G., Abdou Alio, M., Baynast, H. de, Delattre, C., Pons, A., Mathias, J.-D., Callois, J.-M., Vial, C., Michaud, P., & Dussap, C.-G. (2019). Wood-lignin: Supply, extraction processes and use as bio-based material. *European Polymer Journal*, *112*, 228–240. https://doi.org/10.1016/j.eurpolymj.2019.01.007.

Umaraw, P., & Verma, A. K. (2017). Comprehensive review on application of edible film on meat and meat products: An eco-friendly approach. *Critical Reviews in Food Science and Nutrition*, *57*(6), 1270–1279. https://doi.org/10.1080/10408398.2014.986563.

Umweltbundesamt (2020). Aufkommen und Verwertung von Verpackungsabfällen in Deutschland im Jahr 2018: Abschlussbericht, *166*.

United Nations (2015). Transforming our world: the 2030 Agenda for Sustainable Development. *General Assembly*, *A/Res/70/1*.

Vafania, B., Fathi, M., & Soleimanian-Zad, S. (2019). Nanoencapsulation of thyme essential oil in chitosan-gelatin nanofibers by nozzle-less electrospinning and their application to reduce nitrite in sausages. *Food and Bioproducts Processing*, *116*, 240–248. https://doi.org/10.1016/j.fbp.2019.06.001.

van de Nadort, A. (2018). Opinion of the European Committee of the Regions — Communication on a European Strategy for Plastics in a circular economy. *Official Journal of the European Union*, *C 461/30*.

van den Oever, M., Molenveld, K., van der Zee, M., & Bos, H. (2017). Bio-Based and Biodegradable Plastics: Facts and Figures: Facts and Figures. *Wageningen Food & Biobased Research.* Advance online publication. https://doi.org/10.18174/408350.

van Eck, P. S., Jager, W., & Leeflang, P. S. H. (2011). Opinion Leaders' Role in Innovation Diffusion: A Simulation Study. *Journal of Product Innovation Management*, *28*(2), 187–203. https://doi.org/10.1111/j.1540-5885.2011.00791.x.

van Eygen, E., Feketitsch, J., Laner, D., Rechberger, H., & Fellner, J. (2017). Comprehensive analysis and quantification of national plastic flows: The case of Austria. *Resources, Conservation and Recycling*, *117*, 183–194. https://doi.org/10.1016/j.resconrec.2016.10.017.

van Sluisveld, M. A., & Worrell, E. (2013). The paradox of packaging optimization – a characterization of packaging source reduction in the Netherlands. *Resources, Conservation and Recycling*, *73*, 133–142. https://doi.org/10.1016/j.resconrec.2013.01.016.

Vandewijngaarden, J., Murariu, M., Dubois, P., Carleer, R., Yperman, J., Adriaensens, P., Schreurs, S., Lepot, N., Peeters, R., & Buntinx, M. (2014). Gas Permeability Properties of Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate). *Journal of Polymers and the Environment*, *22*(4), 501–507. https://doi.org/10.1007/s10924-014-0688-1.

Vanitha, R., & Kavitha, C. (2021). Development of natural cellulose fiber and its food packaging application. *Materials Today: Proceedings*, *36*(4), 903–906. https://doi.org/10.1016/j.matpr.2020.07.029.

Veiga-Santos, P., Silva, L. T., Souza, C. O. de, Da Silva, J. R., Albuquerque, E. C., & Druzian, J. I. (2018). Coffee-cocoa additives for bio-based antioxidant packaging. *Food Packaging and Shelf Life*, *18*, 37–41. https://doi.org/10.1016/j.fpsl.2018.08.005.

Vejdan, A., Ojagh, S. M., Adeli, A., & Abdollahi, M. (2016). Effect of TiO2 nanoparticles on the physico-mechanical and ultraviolet light barrier properties of fish gelatin/agar bilayer film. *LWT - Food Science and Technology*, *71*, 88–95. https://doi.org/10.1016/j.lwt.2016.03.011.

Verghese, K., Lewis, H., & Fitzpatrick, L. (2012). *Packaging for Sustainability*. Springer.

Verghese, K., Lewis, H., Lockrey, S., & Williams, H. (2015). Packaging's Role in Minimizing Food Loss and Waste Across the Supply Chain. *Packaging Technology and Science*, *28*(7), 603–620. https://doi.org/10.1002/pts.2127.

Wang, K., Lim, P. N., Tong, S. Y., & Thian, E. S. (2019). Development of grapefruit seed extract-loaded poly(ε-caprolactone)/chitosan films for antimicrobial food packaging. *Food Packaging and Shelf Life*, *22*, 100396. https://doi.org/10.1016/j.fpsl.2019.100396.

Wang, L [Liyan], Guo, H., Wang, J., Jiang, G., Du, F., & Liu, X. (2019). Effects of Herba Lophatheri extract on the physicochemical properties and biological activities of the chitosan film. *International Journal of Biological Macromolecules*, *133*, 51–57. https://doi.org/10.1016/j.ijbiomac.2019.04.067.

Wang, S [Sen], Lu, A., & Zhang, L. (2016). Recent advances in regenerated cellulose materials. *Progress in Polymer Science*, *53*, 169–206. https://doi.org/10.1016/j.progpolymsci.2015.07.003.

Wang, S. Y., & Stretch, A. W. (2001). Antioxidant capacity in cranberry is influenced by cultivar and storage temperature. *Journal of Agricultural and Food Chemistry*, *49*(2), 969–974. https://doi.org/10.1021/jf001206m.

Wang, W [Wangxia], Gu, F., Deng, Z., Zhu, Y [Yang], Zhu, J., Guo, T., Song, J [Junlong], & Xiao, H. (2021). Mulitlayer surface construction for enhancing barrier properties of cellulose-based packaging. *Carbohydrate Polymers*, *255*, Article 117431. https://doi.org/10.1016/j.carbpol.2020.117431.

Wang, Y., Xiong, Y. L., Rentfrow, G. K., & Newman, M. C. (2013). Oxidation promotes cross-linking but impairs film-forming properties of whey proteins. *Journal of Food Engineering*, *115*(1), 11–19. https://doi.org/10.1016/j.jfoodeng.2012.09.013.

Wei, L., Agarwal, U. P., Matuana, L., Sabo, R. C., & Stark, N. M. (2018). Performance of high lignin content cellulose nanocrystals in poly(lactic acid). *Polymer*, *135*, 305–313. https://doi.org/10.1016/j.polymer.2017.12.039.

Weiss, M., Haufe, J., Carus, M., Brandão, M., Bringezu, S., Hermann, B., & Patel, M. K. (2012). A Review of the Environmental Impacts of Biobased Materials. *Journal of Industrial Ecology*, *16*(S1), 169-181. https://doi.org/10.1111/j.1530-9290.2012.00468.x.

Wen, P., Zhu, D.-H., Feng, K., Liu, F.-J., Lou, W.-Y., Li, N., Zong, M.-H., & Wu, H. (2016). Fabrication of electrospun polylactic acid nanofilm incorporating cinnamon essential oil/β-cyclodextrin inclusion complex for antimicrobial packaging. *Food Chemistry*, *196*, 996–1004. https://doi.org/10.1016/j.foodchem.2015.10.043.

Wensing, J., Caputo, V., Carraresi, L., & Broering, S. (2020). The effects of green nudges on consumer valuation of bio-based plastic packaging. *Ecological Economics*, *178*, 106783. https://doi.org/10.1016/j.ecolecon.2020.106783.

Wensing, J., Carraresi, L., & Broering, S. (2019). Do pro-environmental values, beliefs and norms drive farmers' interest in novel practices fostering the Bioeconomy? *Journal of Environmental Management*, *232*, 858–867. https://doi.org/10.1016/j.jenvman.2018.11.114.

Werdin González, J. O., Jesser, E. N., Yeguerman, C. A., Ferrero, A. A., & Fernández Band, B. (2017). Polymer nanoparticles containing essential oils: New options for mosquito control. *Environmental Science and Pollution Research*, *24*(20), 17006–17015. https://doi.org/10.1007/s11356-017-9327-4.

White, A., & Lockyer, S. (2020). Removing plastic packaging from fresh produce – what's the impact? *Nutrition Bulletin*, *45*(1), 35–50. https://doi.org/10.1111/nbu.12420.

Wikström, F., Verghese, K., Auras, R., Olsson, A., Williams, H., Wever, R., Grönman, K., Kvalvåg Pettersen, M., Møller, H., & Soukka, R. (2019). Packaging Strategies That Save Food: A Research Agenda for 2030. *Journal of Industrial Ecology*, *23*(3), 532–540. https://doi.org/10.1111/jiec.12769.

Williams, H., & Wikström, F. (2011). Environmental impact of packaging and food losses in a life cycle perspective: a comparative analysis of five food items. *Journal of Cleaner Production*, *19*(1), 43–48. https://doi.org/10.1016/j.jclepro.2010.08.008.

Witzler, M., Alzagameem, A., Bergs, M., El Khaldi-Hansen, B., Klein, S. E., Hielscher, D., Kamm, B., Kreyenschmidt, J., Tobiasch, E., & Schulze, M. (2018). Lignin-Derived Biomaterials for Drug Release and Tissue Engineering. *Molecules*, *23*(8). https://doi.org/10.3390/molecules23081885.

Witzler, M., Büchner, D., Shoushrah, S. H., Babczyk, P., Baranova, J., Witzleben, S., Tobiasch, E., & Schulze, M. (2019). Polysaccharide-Based Systems for Targeted Stem Cell Differentiation and Bone Regeneration. *Biomolecules*, *9*(12), Article 840. https://doi.org/10.3390/biom9120840.

Worrell, E., Allwood, J., & Gutowski, T. (2016). The Role of Material Efficiency in Environmental Stewardship. *Annual Review of Environment and Resources*, *41*(1), 575–598. https://doi.org/10.1146/annurev-environ-110615-085737.

WRAP (2018). Understanding plastic packaging and the language we use to describe it.

Wu, J., Sun, X., Guo, X., Ge, S [Shangying], & Zhang, Q [Qiqing] (2017). Physicochemical properties, antimicrobial activity and oil release of fish gelatin films incorporated with cinnamon essential oil. *Aquaculture and Fisheries*, *2*(4), 185–192. https://doi.org/10.1016/j.aaf.2017.06.004.

Wu, Z., Zhou, W., Pang, C., Deng, W., Xu, C [Changliang], & Wang, X. (2019). Multifunctional chitosan-based coating with liposomes containing laurel essential oils and nanosilver for pork preservation. *Food Chemistry*, *295*, 16–25. https://doi.org/10.1016/j.foodchem.2019.05.114.

Xia, C., Wang, W [Wenbo], Wang, L [Ling], Liu, H., & Xiao, J. (2019). Multilayer zein/gelatin films with tunable water barrier property and prolonged antioxidant activity. *Food Packaging and Shelf Life*, *19*, 76–85. https://doi.org/10.1016/j.fpsl.2018.12.004.

Xiong, H., Payne, D., & Kinsella, S. (2016). Peer effects in the diffusion of innovations: Theory and simulation. *Journal of Behavioral and Experimental Economics*, *63*, 1–13. https://doi.org/10.1016/j.socec.2016.04.017.

Xu, C [Chunbao], & Ferdosian, F. (2017). *Conversion of Lignin into Bio-Based Chemicals and Materials*. *Green Chemistry and Sustainable Technology* [Buch]. Springer Berlin Heidelberg. https://doi.org/10.1007/978-3-662-54959-9.

Xu, C [Chunping], Arancon, R. A. D., Labidi, J., & Luque, R. (2014). Lignin depolymerisation strategies: Towards valuable chemicals and fuels. *Chemical Society Reviews*, *43*(22), 7485–7500. https://doi.org/10.1039/c4cs00235k.

Xu, Q., Chen, C [Chen], Rosswurm, K., Yao, T., & Janaswamy, S. (2016). A facile route to prepare cellulose-based films. *Carbohydrate Polymers*, *149*, 274–281. https://doi.org/10.1016/j.carbpol.2016.04.114.

Xu, T., Gao, C., Feng, X [Xiao], Yang, Y., Shen, X., & Tang, X. (2019). Structure, physical and antioxidant properties of chitosan-gum arabic edible films incorporated with cinnamon essential oil. *International Journal of Biological Macromolecules*, *134*, 230–236. https://doi.org/10.1016/j.ijbiomac.2019.04.189.

Yang, S.-Y., Lee, K.-Y., Beak, S.-E., Kim, H., & Song, K. B. (2017). Antimicrobial activity of gelatin films based on duck feet containing cinnamon leaf oil and their applications in packaging of cherry tomatoes. *Food Science and Biotechnology*, *26*(5), 1429–1435. https://doi.org/10.1007/s10068-017-0175-2.

Yang, W., Fortunati, E., Dominici, F., Giovanale, G., Mazzaglia, A., Balestra, G. M., Kenny, J. M., & Puglia, D. (2016a). Effect of cellulose and lignin on disintegration, antimicrobial and antioxidant properties of PLA active films. *International Journal of Biological Macromolecules*, *89*, 360–368. https://doi.org/10.1016/j.ijbiomac.2016.04.068.

Yang, W., Fortunati, E., Dominici, F., Giovanale, G., Mazzaglia, A., Balestra, G. M., Kenny, J. M., & Puglia, D. (2016b). Synergic effect of cellulose and lignin nanostructures in PLA based systems for food antibacterial packaging. *European Polymer Journal*, *79*, 1–12. https://doi.org/10.1016/j.eurpolymj.2016.04.003.

Yang, W., Owczarek, J. S., Fortunati, E., Kozanecki, M., Mazzaglia, A., Balestra, G. M., Kenny, J. M., Torre, L., & Puglia, D. (2016). Antioxidant and antibacterial lignin nanoparticles in polyvinyl alcohol/chitosan films for active packaging. *Industrial Crops and Products*, *94*, 800–811. https://doi.org/10.1016/j.indcrop.2016.09.061.

Yildirim, S., Röcker, B., Pettersen, M. K., Nilsen-Nygaard, J., Ayhan, Z., Rutkaite, R., Radusin, T., Suminska, P., Marcos, B., & Coma, V. (2018). Active Packaging Applications for Food. *Comprehensive Reviews in Food Science and Food Safety*, *17*(1), 165–199. https://doi.org/10.1111/1541-4337.12322.

Youssef, A. M., & El-Sayed, S. M. (2018). Bionanocomposites materials for food packaging applications: Concepts and future outlook. *Carbohydrate Polymers*, *193*, 19–27. https://doi.org/10.1016/j.carbpol.2018.03.088.

Youssef, A. M., Malhat, F. M., Abdel Hakim, A. A., & Dekany, I. (2017). Synthesis and utilization of poly (methylmethacrylate) nanocomposites based on modified montmorillonite. *Arabian Journal of Chemistry*, *10*(5), 631–642. https://doi.org/10.1016/j.arabjc.2015.02.017.

Yue, C., Hall, C. R., Behe, B. K., Campbell, B. L., Dennis, J. H., & Lopez, R. G. (2010). Are consumers willing to pay more for biodegradable containers than for plastic ones? Evidence from hypothetical conjoint analysis and nonhypothetical experimental auctions. *Journal of Agricultural and Applied Economics*(42), 757–772. http://ageconsearch.umn.edu/record/60968.

Zeid, A., Karabagias, I. K., Nassif, M., & Kontominas, M. G. (2019). Preparation and evaluation of antioxidant packaging films made of polylactic acid containing thyme, rosemary, and oregano essential oils. *Journal of Food Processing and Preservation*, *83*(5), 1299. https://doi.org/10.1111/jfpp.14102.

Zhang, H., Hortal, M., Dobon, A., Bermudez, J. M., & Lara-Lledo, M. (2015). The Effect of Active Packaging on Minimizing Food Losses: Life Cycle Assessment (LCA) of Essential Oil Component-enabled Packaging for Fresh Beef. *Packaging Technology and Science*, *28*(9), 761–774. https://doi.org/10.1002/pts.2135.

Zhang, X [Xin], Zhao, Y., Li, Y [Yuanyuan], Zhu, L., Fang, Z., & Shi, Q. (2020). Physicochemical, mechanical and structural properties of composite edible films based on whey protein isolate/psyllium seed gum. *International Journal of Biological Macromolecules*, *153*, 892–901. https://doi.org/10.1016/j.ijbiomac.2020.03.018.

Zhang, X [Xueqin], Xiao, N., Chen, M., Wei, Y., & Liu, C. (2020). Functional packaging films originating from hemicelluloses laurate by direct transesterification in ionic liquid. *Carbohydrate Polymers*, *229*, 115336. https://doi.org/10.1016/j.carbpol.2019.115336.

Zhang, Y [Yachuan], Liu, Q., & Rempel, C. (2018). Processing and characteristics of canola protein-based biodegradable packaging: A review. *Critical Reviews in Food Science and Nutrition*, *58*(3), 475–485. https://doi.org/10.1080/10408398.2016.1193463.

Zheng, W., & Wang, S. Y. (2003). Oxygen radical absorbing capacity of phenolics in blueberries, cranberries, chokeberries, and lingonberries. *Journal of Agricultural and Food Chemistry*, *51*(2), 502–509. https://doi.org/10.1021/jf020728u.

Zhu, Y [Yunqing], Romain, C., & Williams, C. K. (2016). Sustainable polymers from renewable resources. *Nature*, *540*(7633), 354–362. https://doi.org/10.1038/nature21001.

Zinoviadou, K. G., Gougouli, M., & Biliaderis, C. G. (2016). Innovative Biobased Materials for Packaging Sustainability. In *Innovation Strategies in the Food Industry* (pp. 167–189). Elsevier. https://doi.org/10.1016/B978-0-12-803751-5.00009-X.

3 Influence of different bio-based and conventional packaging trays on the quality loss of fresh cherry tomatoes during distribution and storage

Imke Korte[1,*], Antonia Albrecht[1], Maureen Mittler[1], Claudia Waldhans[1], Judith Kreyenschmidt[1,2]

[1] Rheinische Friedrich Wilhelms-University Bonn, Institute of Animal Sciences, Katzenburgweg 7-9, D-53115 Bonn, Germany

[2] Hochschule Geisenheim University, Department of Fresh Produce Logistics, Von-Lade-Straße 1, D-65366 Geisenheim, Germany

* Correspondence

Korte, I., Albrecht, A., Mittler, M., Waldhans, C., & Kreyenschmidt, J. (2023). Influence of different bio-based and conventional packaging trays on the quality loss of fresh cherry tomatoes during distribution and storage. *Packaging Technology and Science*, *36*(7), 569–583. https://doi.org/10.1002/pts.2728.

3.1 Abstract

The quality loss and shelf life of cherry tomatoes stored in different types of commercially available bio-based packaging trays were analyzed and compared with the reference materials, corrugated cardboard and recycled polyethylene terephthalate (rPET). During storage under conditions reflecting points in a regional German supply chain, typical quality parameters were investigated. Surface bacterial counts of the packaging materials were investigated before and after storage, and material parameters were determined for selected packaging.

The results showed that groundwood pulp and sugar cane trays showed higher preserving characteristics of tomatoes in comparison with the references. During storage, weight loss was highest for tomatoes stored in polylactic acid (PLA). The highest increase in microbial count with 3.5 $\log_{10}$ units was observed after 20 days for tomatoes stored in packaging trays of bamboo. The lowest increase with 0.5 $\log_{10}$ units and 0.6 $\log_{10}$ units was recorded for tomatoes stored in packaging trays of groundwood pulp and grass paper, respectively. The results of the packaging parameters show the highest (411.0 N) and lowest (79.0 N) tensile strength for cellulose and groundwood pulp. The water absorption capacity ranged from < 10 g/m² (bamboo, cellulose) up to 211.2 g/m² (corrugated cardboard).

3.2 Introduction

The rate of quality loss and length of shelf life of fruits and vegetables is influenced by several parameters, such as initial contamination with spoilage organisms or pathogens, the growth rate of these microorganisms, respiration processes as well as speed of enzymatic, chemical, and physical reactions within the product depending on external factors like temperature, humidity, and packaging conditions (Ahvenainen, 1996; D'Aquino et al., 2016; Kreyenschmidt & Ibald, 2012; Patrignani et al., 2016; Soliva-Fortuny & Martín-Belloso, 2003; Tripathi & Dubey, 2004). The main quality losses of tomatoes during storage are related to softening and decay incidence (Buendía−Moreno et al., 2020; Fagundes et al., 2014; Wei et al., 2018). The softening during tomato storage is caused by enzymatic degradation of cell walls and results in product water losses as well as observable sensory changes (Buendía−Moreno et al., 2020; Kalamaki et al., 2012; Opara et al., 2012). Consequently, softening and decay incidence lead to postharvest economic losses (Buendía−Moreno et al., 2020; Fagundes et al., 2014; Opara & Mditshwa, 2013; Wei et al., 2018).

However, postharvest losses are not only relevant from an economic point of view. There is also an increasing concern about the large quantities of food intended for human consumption that are wasted along the entire supply chain (Godfray et al., 2010). Food losses and waste have a significant impact not only on the use of natural resources for the product and packaging production and the environment, as food production and supply systems are counted as some of the biggest contributors to climate change, but also on malnutrition and hunger in developing countries (Caldeira et al., 2019; Hounsou et al., 2022; Korte et al., 2021; Kummu et al., 2012; Qin & Horvath, 2022; United Nations, 2015). The environmental impact of producing the food itself is much higher than the (plastic) packaging. Thus, the overall negative environmental impact of food waste rises with every step in the supply chain, because of resources used and accumulation along the chain (Heller et al., 2019; Matthews et al., 2021; Scherhaufer et al., 2018).

To reduce food loss and waste, selecting a suitable packaging material is essential (Dannehl et al., 2008; Korte et al., 2021; Wikström et al., 2019). In 2019, plastic was the dominant packaging material with 64 % of the packaging volume for fruits and vegetables. Paper, cardboard, and carton accounted for 31 % and other materials such as wood or cotton for 5 % in Germany (Naturschutzbund Deutschland (NABU) e.V., 2020). Aside from the main functions such as protection, communication, marketing, and convenience, an appropriate food packaging solution offers preservation, ensuring high quality, safety, and longer shelf life along the entire supply chain (Accorsi, 2019; Aggarwal & Langowski, 2020; Coelho et al., 2020; Kreyenschmidt et al., 2013; Sharma & Ghoshal, 2018). This is achieved by slowing down microbial growth and reducing physicochemical spoilage reactions such as respiration rates influencing water loss as well as preventing the products from transport damages (Corrado et al., 2017; Marsh & Bugusu, 2007; Opara & Mditshwa, 2013). Depending on packaging structure and material, the ability for adhesion and persistence of microorganisms on surfaces may lead to cross contamination and thereby affecting the shelf life of the product (Patrignani et al., 2016). Thus, an appropriate packaging can prevent possible waste of the food product prior to final consumption (Accorsi, 2019; Aggarwal & Langowski, 2020; Coelho et al., 2020; Kreyenschmidt et al., 2013; Sharma & Ghoshal, 2018).

Since the application of single-use plastic packaging has grown significantly within the last decade, the development and promotion of more sustainable materials became key roles on political levels worldwide (European Commission, 2018; van Eygen et al., 2017). Moreover, there are increasing concerns in terms of the harm caused to the environment mainly during the manufacturing phase (i.e., oil refinery and material production), problematic end-of life strategies, and adverse effects on human health (Accorsi, 2019; Bertling et al., 2018; European Commission, 2018; Umweltbundesamt, 2018). Despite these well-known negative effects, up to now, fossil-based plastics are favored by the food industry because of their stability, lightweight nature, and low costs (Ellen MacArthur Foundation and McKinsey & Company, 2016; Matthews et al., 2021).

New efforts are continuously being made to increase the sustainability of packaging materials such as applying bio-based and/or biodegradable materials as well as using easy recyclable materials as alternatives to existing fossil-based plastics (García-García et al., 2013; Korte et al., 2021; Pro Carton, 2010; Rhim et al., 2007; Sharma & Ghoshal, 2018). The production of bio-based recyclable packaging and compostable biodegradable packaging is seen as an opportunity for the use of renewable resources, including biomass waste, as raw materials to produce (polymer) packaging materials (European Parliament and the Council of the European Union, 2018). Up to now, a limited number of bio-based (polymer) packaging trays are commercially available as food packaging (Korte et al., 2021; Sharma & Ghoshal, 2018). There are also limited studies about the effect of these packaging solutions on the quality loss and shelf life in comparison with commonly used fossil- or wood-based materials.

Thus, this study aims to investigate commercially available bio-based packaging trays and their influence on quality and shelf life of fresh cherry tomatoes and compare these with reference materials. During cold storage followed by ambient storage to reflect conditions typical for a regional German supply chain, typical quality parameters were investigated. To evaluate material properties, material parameters were determined for selected packaging, and surface bacterial counts of the packaging materials were investigated before an after storage. In this respect, polylactic acid (PLA) and materials from different fibers like wood, sugar cane, bamboo, cellulose, and grass were selected as bio-based materials to be used for storage of fresh cherry tomatoes. As reference materials, conventional corrugated cardboard and recycled polyethylene terephthalate (rPET) were used.

3.3 Materials and methods

3.3.1 Study design and sampling

For the storage trials, fresh cherry tomatoes were delivered by a wholesaler (Bauer Funken GmbH, Kempen, Germany) to the laboratory of the University of Bonn. To ensure the standardization of the tests, the initial surface bacterial counts of the tomatoes were analyzed before the filling process. Approximately 20 fruits were randomly packed into each packaging tray (see tray dimensions in Table 3.1) to reach a weight of 250 ± 5 g. The filled packaging trays of all packaging materials were wrapped with low-density polyethylene (LDPE) foil of 25 µm thickness. The foil was heat sealed and perforated on the top 12 times with a thin needle (Ø 0.5 mm) to ensure aerobe conditions. The LDPE foil was used in combination with all tested packaging trays to avoid (cross) contamination of the tomatoes and to ensure standardized test conditions. Typically, in Germany, fresh cherry tomatoes are only packed in trays without foil or at least without additional equilibrium-modified atmosphere packaging.

In each storage trial, 14 trays of tomatoes were prepared for each packaging material. The temperature and time scenario were defined to reflect specific points (central warehouse, commercialization, and household conditions) in a regional German supply chain (Pelka & Kreyenschmidt, 2013). For 3 days, the trays were stored under controlled temperature conditions in high-precision low-temperature incubators (Sanyo MIR 153, Sanyo Electric Co., Ora-Gun, Gumma, Japan) at 12°C and for further 17 days in an open space at room temperature (approx. 18°C). During storage, the temperature conditions and the humidity levels were recorded continuously every 10 min by data loggers (Data Logger Testo SE & Co. KGaA, Lenzkirch, Germany). For temperature and relative humidity data, see Appendix Figure A.1 – A.4.

Investigations were conducted over a storage period of 20 days with a parameter set comprising assessment of decay incidence (n=8) and weight loss (n=8), microbiological investigations (n=3) (total viable count (TVC), yeasts and molds), sensory evaluation (n=8) as well as instrumental color and texture measurements (n=20). Analyses were conducted at consecutive investigation points after 0, 3, 6, (10), 14, 17 and 20 days of storage, except microbial investigations that were only conducted at day 0 and 20. Packaging trays were analyzed for surface bacterial counts before (day 0) and after storage (day 20) of tomatoes. Selected packaging materials were also analyzed for typical material parameters related to stability like tensile strength, elongation at break, and water absorption capacity.

3.3.2 Packaging characteristics and analyses of the packaging trays

Different types of bio-based packaging trays were compared with a reference, conventional packaging materials such as corrugated cardboard and rPET. The packaging trays for this study were purchased from different manufacturers. Since bio-based packaging trays for tomatoes are rare on the market, packaging trays that were manufactured for different markets and a wide product portfolio were integrated in the study. Therefore, the form and volume of the trays differed depending on material and manufacturer. The packaging trays, their size and thickness are described in Table 3.1.

Table 3.1 Reference materials and bio-based packaging materials used for the study.

Material	Size (l x w x h) [cm]	Thickness [µm]*	Picture
Reference materials			
Corrugated cardboard	21.0 x 7.4 x 2.5	1058.1 ± 4.7	
Recycled polyethylene terephthalate (rPET)	13.5 x 9.0 x 4.5	NA	
Bio-based packaging materials			
Groundwood pulp	13.5 x 8.5 x 4.0	1233.8 ± 142.4	
Sugar cane	13.6 x 13.6 x 2.0	543.6 ± 22.0	
Bamboo (PLA coating inside because of its hydrophobic character)	13.5 x 7.5 x 3.8	330.1 ± 3.4	
Cellulose (PLA coating inside because of its hydrophobic character)	13.7 x 8.6 x 5.2	334.4 ± 22.3	
Grass paper	15.0 x 8.5 x 4.0	NA	
Polylactic acid (PLA)	16.0 x 8.5 x 6.0	NA	

* Measured according to DIN EN ISO 534:2011 with an universal thickness measuring device (Frank-PTI GmbH, Birkenau, Germany). NA - Data not analyzed.

To test the stability and thereby evaluate the protective function, the non-plastic packaging materials (corrugated cardboard, groundwood pulp, sugar cane fiber, bamboo fiber, and cellulose fiber) were analyzed, independent from the storage trials, for tensile strength, elongation at break, and water absorption capacity. Additionally, the surface bacterial counts were investigated before the storage trial (day 0) to assess a possible risk of cross contamination. The bacterial load of the packaging materials was also tested after the storage trials (day 20) to evaluate their potential of taking up microorganisms that increase the spoilage potential of the tomatoes. These analyses were performed in triplicate on each packaging material before and after storage.

3.3.3 Microbiological analysis of cherry tomatoes

For the microbiological assessment of the tomatoes, 25 g of the tomato pericarp was transferred to a filtered sterile stomacher bag and filled up with 225 ml physiological saline tryptone diluent (0.85 %; Oxoid BR0053G, Cambridge, UK) adding 0.1 (m/v) % trypton (BHD Prolab VWR, Pennsylvania, USA). The samples were homogenized separately for 60 s in a Stomacher 400 (Kleinfeld Labortechnik, Gehrden, Germany). Serial decimal dilution series of the homogenates (tomatoes) and solutions (packaging) were made using saline tryptone diluent. Two replicates were prepared for each appropriate dilution step. TVC was determined by pour plate technique (1 mL) on plate count agar (Merck, Darmstadt, Germany). The agar plates were incubated at 30°C for 72 h. Yeasts and molds were detected by spread plate technique (0.1 mL) on Yeast extract glucose chloramphenicol agar (YGC) (Merck KGaA, Darmstadt, Germany), and plates were incubated at 25°C for 120 h. Counts of colony forming units (cfu) were calculated and expressed as $\log_{10}$ cfu/g.

3.3.4 Sensory evaluation

A sensory evaluation of the freshness of the tomatoes was carried out by a trained sensory panel (of at least three panellists). For the sensory evaluation, an evaluation scheme that included five product-specific attributes of the loss of freshness of tomatoes was used. Attributes were defined as color (C), decay incidence (DI), firmness (F), odor (O), and general appearance (GA). The attributes were evaluated based on a graded hedonic five-point scale (0-5): 0 being the highest quality and 5 being equal to “unacceptable”. A weighted sensory index (SI) was illustrated by matching different weighting factors to the various attributes. The weighted sensory index was calculated as a weighted average using equation 3.1 (Pelka & Kreyenschmidt, 2013):

$$SI = \frac{1 * C + 2 * DI + 2 * F + 1 * O + 1 * GA}{7} \quad (3.1)$$

where C is the color, DI is the decay incidence, F is the firmness, O is the odor, and GA is the general appearance.

The sensory acceptance, calculated as sensory index (SI), was plotted as a function of time, and fitted to a linear model. The acceptance level of sensory product quality was reached when the SI was 2.0 based on the evaluation of the sensory panel.

3.3.5 Assessment of weight loss and decay incidence

The weight of filled tomato trays was measured using a balance with an accuracy of ± 0.1 g (Mettler-Toledo GmbH, Gießen, Germany). Weight loss percentage of tomatoes was calculated as the difference between initial weight of the trays with tomatoes and the corresponding weight at consecutive investigation points.

The assessment of visual decay incidence focused on counting the number of rotten and infected tomatoes per packaging tray. Tomatoes were considered as infected if a visible lesion like damage, pressure mark, mold growth or any other sensory deficiency was observed. The number of fruits representing the decay incidence was expressed as percentage of rotten fruits related to overall fruit number.

3.3.6 Color and texture measurements

Color measurements were conducted on 20 tomatoes from each packaging material. To get a representative evaluation, tomatoes were randomly picked from three packaging trays of the specific packaging material. The surface color of the tomatoes was measured using a large view spectrophotometer (ColorFlex EZ 4500L, HunterLab, Murnau). The color measurement was conducted at a wavelength between 400 nm and 700 nm with a 45°/0° geometry. The CIE L*a*b* scale was used, measured with D65 illuminant (6500 K daylight). For the measurement, the tomatoes were placed in equatorial region on the glass surface of the measurement device. The CIE L*, a* and b* values were recorded, and the chroma (color saturation) as well as the hue angle were automatically calculated. As reported by López Camelo & Gómez (2004), the color difference (eq. 3.2) was selected and calculated as an appropriate value for evaluating the ripening stage of tomatoes:

$$Color\ differemce = ((L^* - 50)^2 + (a^* - 60)^2 + b^{*2})^{0.5} \quad (3.2)$$

Additionally, the calculated chroma value, reflecting color purity or saturation of a single color, was used as indicator of consumer acceptance in case of completely ripe tomatoes (López Camelo & Gómez, 2004). Values measured on 20 tomatoes for each packaging material were averaged for each package.

The firmness (texture) of the tomatoes was measured using a digital texture analyzer (TAXTPlusC, Stable Micro Systems, Godalming, United Kingdom) with a 100-N load cell. The firmness of the fruit flesh was calculated by the recorded compression force using a penetration test. For the measurement, the tomatoes were placed central under the measuring tool, a rounded, cylindrical stamp with a diameter of 5.5 mm. Test, pre-test, and post-test speeds were 2 mm/s, 1 mm/s and 20 mm/s. The distance used was 15 mm of penetration depth of a whole tomato. The results were plotted as the needed force in N.

3.3.7 Surface bacterial counts of packaging materials

For the determination of the surface bacterial counts of the packaging materials, three samples per packaging material were analyzed, each before and after use. For the microbiological analysis, a sample from the inner bottom of the packaging tray was cut to a size of 2.5 x 2.5 cm. The sample was transferred into a falcon tube which contained 20 mL of soybean-casein digest broth with lecithin polysorbate (Roth, Karlsruhe, Germany) and washed out by intensive mixing. The microbial count of TVC and yeasts and molds of the solution was determined and expressed as $\log_{10}$ cfu/cm².

Three samples were analyzed for TVC and yeasts and molds on each investigation point. The number of colony-forming units is the average of the analyzed samples.

3.3.8 Evaluation of material parameters

Based on DIN EN ISO 1924-2:2008, tensile strength and elongation at break were determined. The analysis was conducted with a universal testing machine (Frank-PTI GmbH, Birkenau, Germany). The material sample was secured in the clamps and the device pulled on the sample at a steady speed. The maximum force (tensile strength in N) required to tear through the sample was recorded. Sample strips with a width of 15 mm were cut from the packaging material for analysis. Because of the given sizes of the packaging, the standardized test length (distance between the clamping clamps) had to be reduced from 180 mm to 70 mm. The testing device automatically calculated the elongation at break (%) of the material from the given parameters. An average value was calculated for each packaging material from 10 measured values.

Water absorption capacity (g/m²) was measured based on DIN EN ISO 535 and Cobb60. Round samples with a diameter of 5.64 cm, corresponding to 25 cm², were cut out of the packaging materials. 25 mL of distilled water were pipetted into a 500-mL Erlenmeyer flask. The packaging sample was weighed (dry matter) and then placed on the opening of the Erlenmeyer flask. A Petri dish was pressed firmly upside down on the sample. The flask was then turned over creating an approximately 1-cm high water column on the sample. From this moment, the test time of 60 s started while the Petri dish was pressed continuously against the Erlenmeyer flask. After 45 s, the Erlenmeyer flask was turned over, the Petri dish removed, and the sample placed on blotting paper with the wet side down. After further 15 s, a second blotter paper was placed on the sample and rolled over the sample four times with the rolling pin. Then the sample was weighed again (wet weight). The water absorption capacity was calculated using equation 3.3:

$$A = (WW - DM) \times \frac{1000}{X} \quad (3.3)$$

where A is the water absorption capacity in g/m², WW is the wet weight in g, DM is dry matter in g, and X is the surface area of the sample in cm².

Calculated values were averaged for each packaging material.

3.3.9 Data analysis

The standard deviations were calculated for all analyzed parameters. The investigated parameters were analyzed for significant differences using the one-way ANOVA for k-samples according to Kruskal-Wallis for independent samples. A rejected null hypothesis was followed by a pairwise comparison to identify significant differences for selected parameters on different packaging materials. The significance level was defined as a p value ≤ 0.05. The given significance values are Bonferroni corrected. For description of data, bar and line charts were used. Data analysis was conducted with SPSS Statistics 27 (IBM Corp. 1989, 2020, New York, NY).

3.4 Results

3.4.1 Microbiological analysis of cherry tomatoes

Figure 3.1 shows the development of TVC on tomatoes at the beginning and the end of the storage trials. Microbial proliferation differed between the tomatoes stored in the different tested packaging trays. At the beginning of the storage trials, TVC ranged between 3.0 ± 0.2 $\log_{10}$ cfu/g and 6.0 ± 0.7 $\log_{10}$ cfu/g. After 20 days of storage, TVC reached values between 3.9 ± 0.2 $\log_{10}$ cfu/g and 8.8 ± 0.3 $\log_{10}$ cfu/g. The highest increase with 3.5 $\log_{10}$ units was observed for tomatoes stored in packaging trays of bamboo. The lowest increase with 0.5 $\log_{10}$ units and 0.6 $\log_{10}$ units was recorded for tomatoes stored in packaging trays of groundwood pulp and grass paper, respectively. TVC of tomatoes stored in the reference packaging trays of corrugated cardboard and rPET increased by 1.5 $\log_{10}$ units and 2.8 $\log_{10}$ units, respectively.

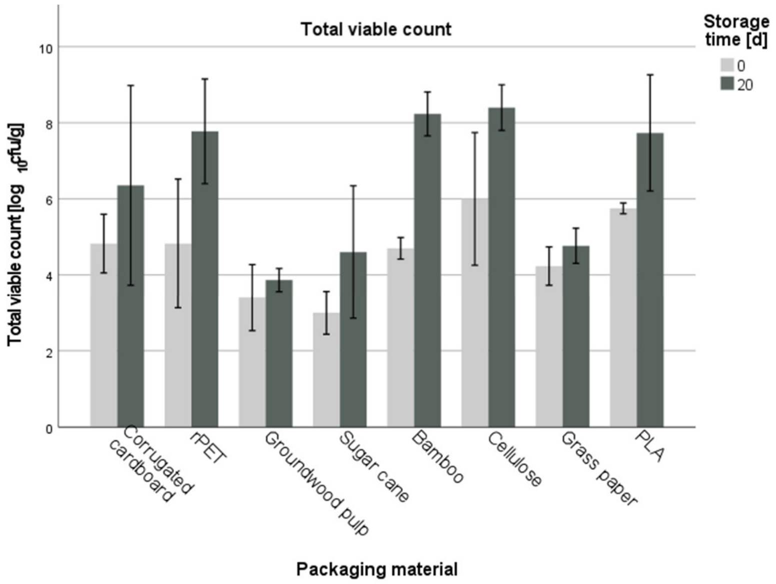


Figure 3.1 Total viable count on fresh cherry tomatoes packaged in reference materials (n=9) and different types of bio-based packaging trays (n=3) during storage. Abbreviations: PLA, polylactic acid; rPET, recycled polyethylene therephthalate.

Figure 3.2 shows the growth of yeasts and molds on tomatoes at the beginning and the end of the storage trials. At the beginning of the storage trials, the load of yeasts and molds varied in a normal, typical range between 2.0 ± 0.0 log_{10} cfu/g and 4.5 ± 0.3 log_{10} cfu/g. After 20 days of storage, yeasts and molds reached values between 4.2 ± 0.1 log_{10} cfu/g and 7.0 ± 0.2 log_{10} cfu/g. The highest increase with 4.3 log_{10} units was observed for tomatoes stored in packaging trays of bamboo. The lowest increase with 1.1 log_{10} units was recorded for tomatoes stored in packaging trays of grass paper. The increase for tomatoes stored in the reference packaging trays was 1.7 log_{10} units (corrugated cardboard) and 2.0 log_{10} units (rPET).

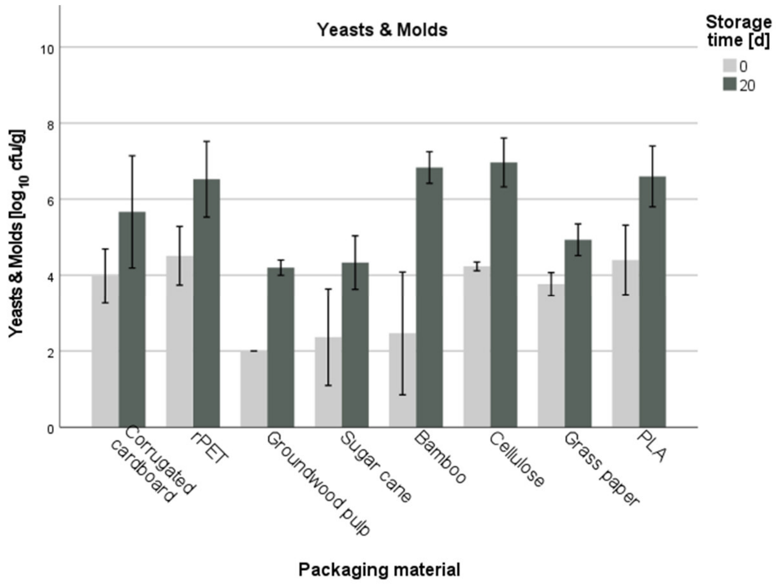


Figure 3.2 Growth of yeasts and molds on fresh cherry tomatoes packaged in reference materials (n=9) and different types of bio-based packaging trays (n=3) during storage. Abbreviations: PLA, polylactic acid; rPET, recycled polyethylene therephthalate.

3.4.2 Sensory evaluation

Different tested packaging trays had a varying strong influence on the sensory quality of tomatoes during storage (data not shown). Nevertheless, important sensory changes were observed especially for the attributes decay incidence, odor, and general appearance from day 0 to day 20. Figure 3.3 illustrates the changes in sensory index as a function of time. Tomatoes packaged in trays of groundwood pulp and sugar cane remained clearly below the acceptance level until the end of storage. These packaging trays preserved an acceptable sensory quality of tomatoes over the entire storage with values for decay incidence, general appearance, and odor below 1. The shortest shelf life with 11 days was observed for tomatoes packed in trays of cellulose because of the high scored attributes decay incidence, general appearance (both 4.0), and odor (3.7), respectively. Using the reference trays, corrugated cardboard and rPET, for storage of tomatoes, the shelf life reached 15 and 19 days, respectively. The shelf life of tomatoes stored in trays of bamboo, grass paper, and PLA reached values between 16 and 19 days. The slope of the linear model, expressing the speed of spoilage, reflects the difference between the shelf life. With values of 0.021 and 0.019 for groundwood pulp and sugar cane, the slope is well below the slope for all other materials, including the references, with values ranging between 0.085 and 0.117. The highest gradient factor of 0.165, confirming the shortest determined shelf life, was shown for cellulose.

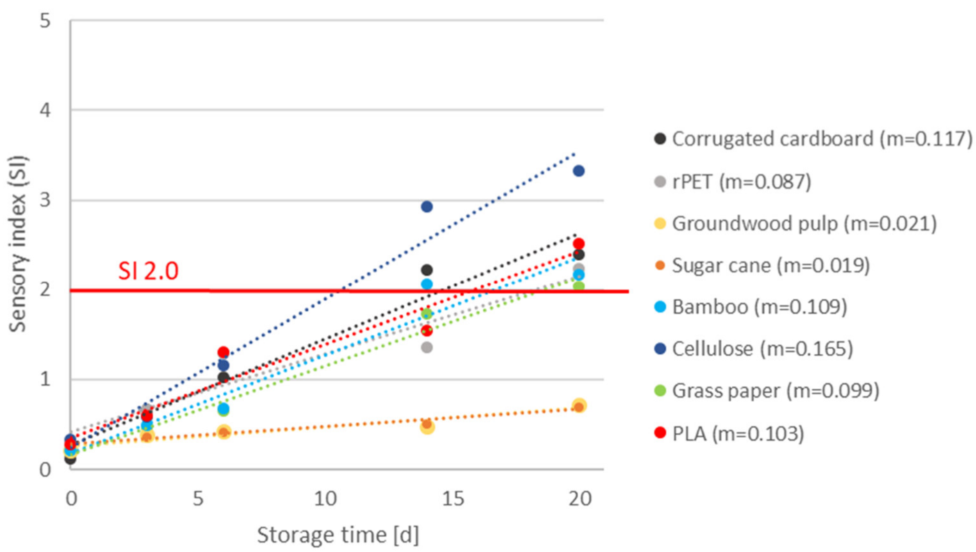


Figure 3.3 Sensory index of fresh cherry tomatoes packaged in reference materials (n=16) and different types of bio-based packaging trays (n=8) during storage. m – slope of linear model expressing speed of spoilage. Abbreviations: PLA, polylactic acid; rPET, recycled polyethylene therephthalate.

3.4.3 Assessment of weight loss and decay incidence

Figure 3.4 indicates changes in weight loss of tomatoes during storage. The weight loss ranges between 1.1 % and 8.5 %. The highest weight loss was observed for tomatoes stored in trays of PLA (8.5 %) and grass paper (8.2 %), followed by trays of bamboo (6.0 %) and cellulose (5.6 %). Tomatoes stored in the reference trays, corrugated cardboard and rPET, showed a weight loss of 4.7 % and 5.8 %, respectively. Interestingly, the weight loss of tomatoes stored in trays of groundwood pulp and sugar cane showed an increase by 2.3 % and 0.7 % during the first 3 days of storage. At the end of storage, tomatoes packaged in trays of groundwood pulp and sugar cane reached the lowest weight loss with values of 1.1 % and 2.4 %, respectively. The weight loss in trays of groundwood pulp and sugar cane is significantly lower compared with that of the reference trays, corrugated cardboard and rPET ($P \leq 0.05$).

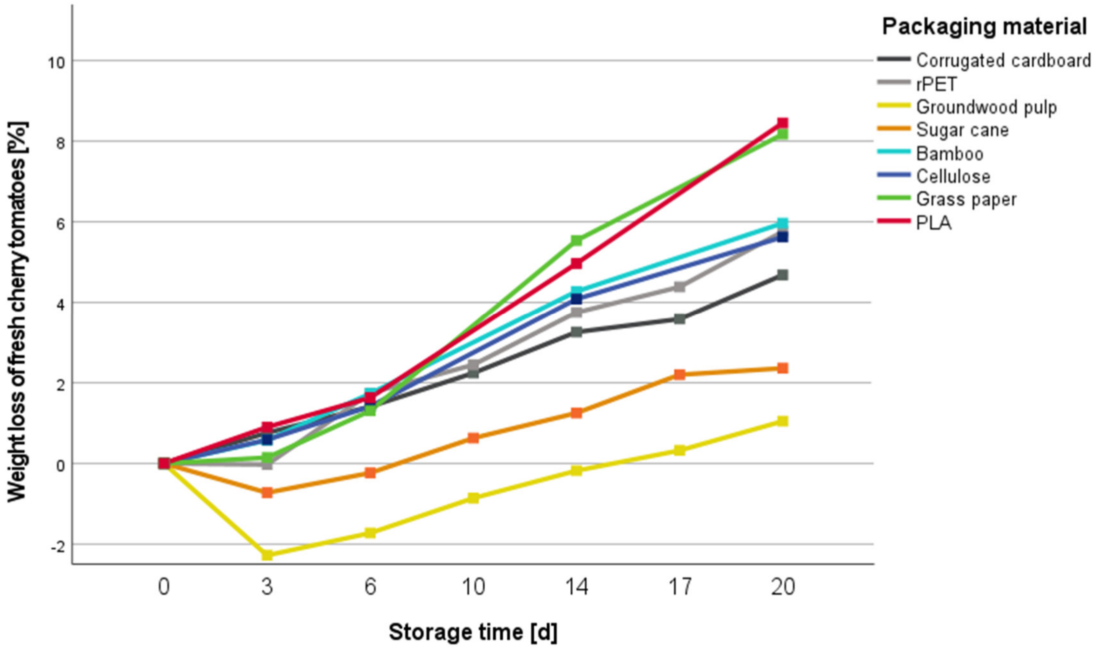


Figure 3.4 Weight loss of fresh cherry tomatoes packaged in reference materials (n=16) and different types of bio-based packaging trays (n=8) during storage. Abbreviations: PLA, polylactic acid; rPET, recycled polyethylene therephthalate.

The decay incidence, measured by rotten and infected tomatoes, increased with storage time in all tested packaging trays (Figure 3.5). The initially determined decay incidence was minimal and varied between 0.5 % for tomatoes stored in the reference tray corrugated cardboard and 5.9 % for tomatoes packed in trays of sugar cane. The trend of decay incidence of tomatoes stored in the tested packaging trays is mostly comparable with the weight loss. At day 3, tomatoes stored in trays of rPET and PLA already showed noticeably high decay incidences with 38.9 % and 56.7 %, while all other packaging trays were still below 15 %. On day 6, the decay incidence of tomatoes stored in trays of corrugated cardboard showed a rapid increase up to 41.4 %. At the end of storage, high decay incidences with 94.9 % and 95.4 % were recorded for tomatoes stored in trays of rPET and cellulose. The highest decay incidence, with 100 %, was shown already at day 17 for tomatoes stored in trays of PLA. This confirms the findings in weight loss that was one of the highest for tomatoes stored in trays of PLA as well. The increase in decay incidence of tomatoes was negligible for trays of sugar cane and groundwood pulp and showed values of 13.7 % and 17.9 %, respectively at the end of storage. These results are in accordance with the results of weight loss that remained also low for these packaging trays. For the packaging trays, groundwood pulp and sugar cane, significant differences ($P \leq 0.05$) in decay incidence were recorded compared with all other tested packaging trays. The packaging trays of groundwood pulp and sugar cane showed lower values for decay incidence than the references, corrugated cardboard and rPET. The recorded decay incidences for the packaging trays of grass paper and bamboo are significantly lower ($P \leq 0.05$) compared with the reference material rPET as well.

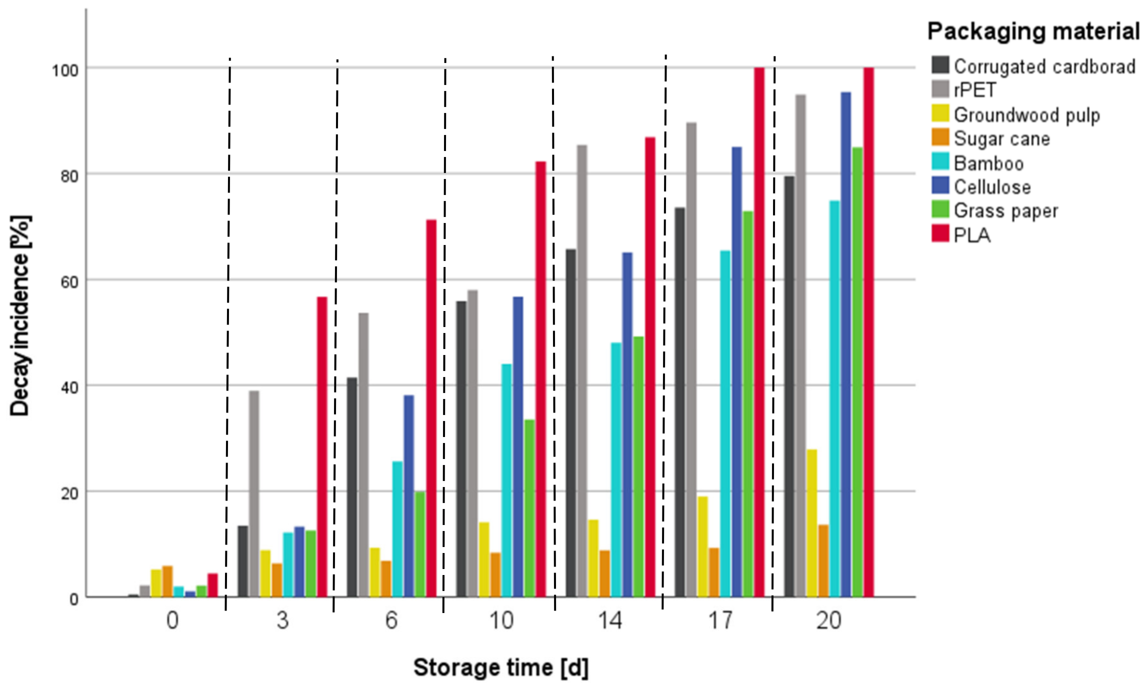


Figure 3.5 Decay incidence of fresh cherry tomatoes packaged in reference materials (n=16) and different types of bio-based packaging trays (n=8) during storage. Abbreviations: PLA, polylactic acid; rPET, recycled polyethylene therephthalate.

3.4.4 Color and texture measurements

The measurements of color difference showed only slight differences over the entire storage period as well as between tomatoes stored in trays of different packaging materials (data not shown). Average color difference values ranged between 51.3 and 44.9. According to the USDA color standard, a color difference of around 50 already classifies tomatoes as red (full ripe) (López Camelo & Gómez, 2004). Additionally, chroma (indicator of consumer acceptance in case of completely ripe tomatoes) showed only slight differences during storage. At the beginning (day 0), chroma varied between 22.1 (rPET) and 24.5 (grass paper) and at the end of storage (day 20), chroma ranged between 23.1 (groundwood pulp) and 26.0 (PLA). The recorded chroma values for bamboo (17.6) and cellulose (14.0) were noticeably lower than these values (data not shown).

In this study, the recorded values for the firmness of the tomatoes varied over the entire storage period in all packaging trays, but no clear trend could be detected (data not shown). The recorded firmness ranged between 32.2 N and 17.1 N. High standard deviations were detected, which indicate natural occurring variations in fruit firmness.

3.4.5 Surface bacterial counts of packaging materials

Figure 3.6 displays the microbial load of TVC on the packaging trays studied before and after storage of tomatoes. TVC of unused packaging materials varied between 0.5 ± 0.0 $\log_{10}$ cfu/cm² (rPET and PLA) and 2.8 ± 0.1 $\log_{10}$ cfu/cm² (corrugated cardboard). After the storage trials, TVC reached values between 2.7 ± 0.5 $\log_{10}$ cfu/cm² and 6.8 ± 0.7 $\log_{10}$ cfu/cm². The highest increase, with 6.2 $\log_{10}$ units, was observed for PLA. The lowest increase, with 0.9 $\log_{10}$ units, was recorded for sugar cane. TVCs of the references corrugated cardboard and rPET increased by 1.3 $\log_{10}$ units and 4.3 $\log_{10}$ units, respectively.

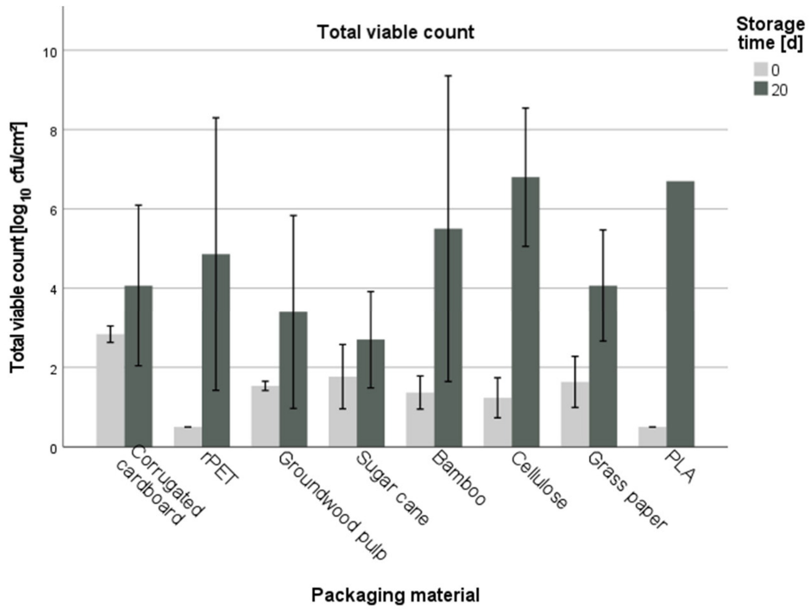


Figure 3.6 Microbial load on reference materials (n=6) and different types of bio-based packaging trays (n=3) before and after storage of fresh cherry tomatoes. Abbreviations: PLA, polylactic acid; rPET, recycled polyethylene therephthalate.

Figure 3.7 displays the microbial load of yeasts and molds on the packaging trays studied before and after storage of tomatoes. The initial load for yeasts and molds of unused packaging trays stayed below the detection limit for all tested packaging materials. After storage, for yeasts and molds values between 2.5 ± 0.8 $\log_{10}$ cfu/cm² and 6.1 ± 0.5 $\log_{10}$ cfu/cm² were reached. The highest increase with 4.6 $\log_{10}$ units was observed for cellulose. In addition, the reference rPET showed a comparatively high increase in yeasts and molds with 4.0 $\log_{10}$ units. The lowest increase with 1.0 $\log_{10}$ units was recorded for sugar cane. As the tomatoes stored in trays of rPET, PLA, and cellulose were completely spoiled after storage, these packaging materials showed a comparable high microbial load with > 6.5 $\log_{10}$ cfu/cm².

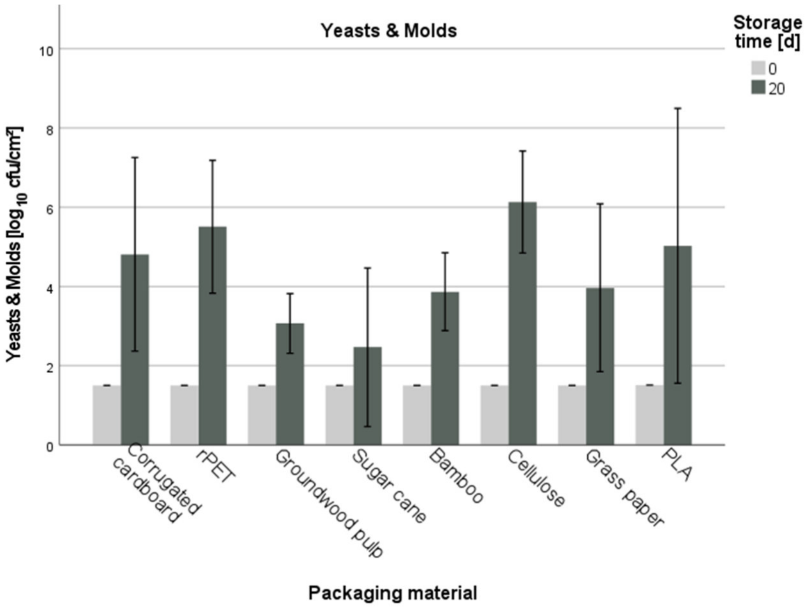


Figure 3.7 Growth of yeasts and molds on reference materials (n=6) and different types of bio-based packaging trays (n=3) before and after storage of fresh cherry tomatoes.

3.4.6 Evaluation of material parameters

Since a mechanically stable packaging is essential for product protection during transport, it is important that packaging materials have appropriate material properties for their specific application. Table 3.2 shows the average values of tensile strength, elongation at break, and water absorption capacity.

The highest tensile strength, with 411.0 N, was shown for cellulose, which is one of the thinnest materials, with a thickness of 334.4 µm. In contrast, the lowest tensile strength (79.0 N) was recorded for groundwood pulp, which was the thickest material (1233.8 µm). The highest measurements for elongation at break were recorded for corrugated cardboard (7.2 %). The values for sugar cane, bamboo, and cellulose varied between 4.2 % and 4.7 %. As shown for the tensile strength, groundwood pulp had the lowest elongation at break with 2.1 %. However, there was no correlation between thickness and tensile strength and elongation at break.

The evaluation of the water absorption capacity of the different packaging materials showed that corrugated cardboard had by far the highest water absorption capacity, with 211.2 g/m². The value for sugar cane was lower by a factor of ten (22.4 g/m²) compared to the reference, corrugated cardboard. The water absorption capacity for bamboo and cellulose was even lower, with values < 10 g/m². A low water absorption capacity for the packaging materials cellulose and bamboo was estimated because of their inner coating with a PLA layer. The low water absorption capacity maintains a high relative humidity, but at the same time, it also favors the growth of yeasts and molds as shown for bamboo and cellulose (chapter 3.4.1).

Table 3.2 Averaged values of the packaging parameters peak force (n=10), elongation at break (n=10), and water absorption capacity (n=5) for different types of bio-based packaging materials and reference material. Lowercase letters donate significant differences (P ≤ 0.05) among the packaging materials for each parameter.

Packaging material	Tensile strength [N]	Elongation at break [%]	Water absorption capacity [g/m²]
Corrugated cardboard	163.3 ± 3.9 *ab*	7.2 ± 0.5 *abc*	211.2 ± 59.3
Groundwood pulp	79.0 ± 5.5 *cde*	2.1 ± 0.4 *ade*	33.6 ± 4.1
Sugar cane	231.0 ± 16.4 *cf*	4.5 ± 0.5 *bd*	22.4 ± 6.0
Bamboo	307.0 ± 11.8 *ad*	4.2 ± 0.2 *c*	5.6 ± 2.0
Cellulose	411.0 ± 17.9 *bef*	4.7 ± 0.3 *e*	8.8 ± 5.9

3.5 Discussion

According to the data of this trial and stated by Alegbeleye et al. (2022), yeasts and molds are more important for spoilage of tomatoes than TVC. García-García et al. (2013) studied the influence of PLA-coated cardboard trays and uncoated cardboard trays (references), each wrapped with thin (20µ) and thick (70µ) LDPE films, on quality changes of tomatoes during 30 days of storage at 20°C. The study showed bacterial counts remaining below 4 log_{10} cfu/g over a storage period of 30 days at 20°C, independent from the packaging material. This means that the packaging materials and storage conditions preserved microbial fruit quality of tomatoes until the end of storage (García-García et al., 2013). Contrary to our results, the authors concluded advantages in terms of extending the shelf life of fresh tomatoes packaged in PLA-lined cardboard trays compared to unlined cardboard trays.

Since the focus of this study was the comparison of the influence of the packaging trays on quality changes of stored tomatoes, a standard LDPE foil was used in combination with all tested packaging trays to avoid (cross) contamination of the tomatoes and ensure standardized test conditions. Therefore, as observed in this study, it is expected that microbial growth among the tested packaging trays does not show any remarkable difference.

Comparing the influence of bio-based packaging trays with the reference materials on the SI, the findings show that groundwood pulp and sugar cane preserved the tomatoes even better than the references, corrugated cardboard and rPET. Bamboo, cellulose, and PLA obtain comparable results with the reference materials. As these visually and organoleptically detected findings by the panel tests are in accordance with the results presented for microbiological results (chapter 3.4.1) and decay incidence (chapter 3.4.3), it is assumed that the packaging materials have no direct influence on sensory changes.

According to Robertson (2013), a weight loss between 3 % and 10 % of the initial product weight leads to significant losses of freshness of fruits and vegetables. In addition, several authors stated that a weight loss of more than 5 % reduces the market value of fruits and vegetables (Almenar et al., 2008; Koide & Shi, 2007). In the presented study, the weight loss stayed below these limits for tomatoes stored in trays of groundwood pulp and sugar cane over the entire storage period.

In the study of García-García et al. (2013), the weight loss was reported to be < 3.5 % for all packaging combinations at the end of storage. In this study, such low weight losses at ambient storage temperature were only observed for tomatoes stored in trays of groundwood pulp and sugar cane. A possible explanation for the results of this study is that the weight loss is significantly influenced by the interaction of the relative humidity inside the packaging trays, the packaging material itself, and its form. It is assumed that the material and form have a direct influence on the exchange and alignment of surrounding conditions, which in turn affects processes of quality changes. In our study, this assumption is also confirmed by the determined water absorption capacity for the packaging materials groundwood pulp and sugar cane, materials with a much lower water absorption capacity compared with the reference corrugated cardboard (chapter 3.4.6). A lower water absorption capacity is normally related to a larger retention of water vapor inside the packaging resulting in lower weight loss.

As widely reported, deterioration, physiological changes and other biochemical activities of tomatoes are primarily related to the storage temperature and consequently also the relative humidity (Al-Dairi et al., 2021; Arah et al., 2015; Majidi et al., 2014; Žnidarčič et al., 2010). Storage of tomato fruits at temperatures of about 20°C, with high relative humidity, is a crucial factor in slowing down many metabolic activities, thereby maintaining fruit quality appearance and avoiding the transpiration process and water loss in tomatoes during storage (Al-Dairi et al., 2021; Arah et al., 2015; Ayomide et al., 2019; Buendía−Moreno et al., 2019; Paull, 1999). Optimal values of relative humidity for tomatoes are within the range of 85 - 95 % (v/v) depending on the mature stage (Arah et al., 2015; Castro et al., 2005; Shewfelt, 1986; Žnidarčič et al., 2010). Especially, because of the water absorption capacity and water transmission rate of packaging materials, the surrounding conditions are directly influenced and thereby leading to undesirable quality changes like high weight losses affecting microbial growth, mainly noticeable through the growth of yeasts and molds.

As decay incidence is strongly related to microbial infection of internal plant tissues, micro-cracks in tomatoes, which originate from the stem scar, are the main factor affecting decay incidence in tomatoes (Buendía−Moreno et al., 2019). A lower decay incidence in some packaging materials may be due to firmness retention of tomatoes. These findings are confirmed by several authors stating that a reduced softening (firmness) of tomatoes is related to a lower decay incidence during storage (Buendía−Moreno et al., 2020; Wei et al., 2018). Preserving plant cell structures with even and smooth packaging trays leads to reduced decay incidence such as pressure marks and micro-cracks and thereby microbial infection of internal tissues is avoided (Buendía−Moreno et al., 2019). The influence of external physical activity or vibration during transportation was supposed to be negligible since packaging trays are boxed in a secondary packaging that adds further layer of protection and absorbs vibration. Even if differences in material properties were shown, the tested packaging trays exhibit comparable mechanical properties (e.g., strength/hardness), and neither protective inlets nor flexible packaging (e.g., paper bag) were used. However, the current study aimed at a basic research on the general suitability of such trays. Further studies have to be conducted under practical conditions to evaluate the influence of other factors such as vibration during transportation.

In general, next to other factors, the percentage of decay incidence elevates with maturity as well as irrespective of maturity stages by storage time (Diaz-Perez et al., 2000; Moneruzzaman et al., 2009). Several authors stated decay incidence levels similar to those found in this study. Moneruzzaman et al. (2009) reported a total rotting percent of full ripen tomatoes of 8.19 % after 3 days of storage. The decay incidence then raised to 74.68 % after 15 days of storage (Moneruzzaman et al., 2009). As non-adequate levels of O_2 and CO_2 as well as water vapor accumulation within the packaging can lead to an increased decay (Akbudak et al., 2012; Linke & Geyer, 2003), differences in decay incidence, as previously demonstrated, are related to the packaging tray themselves and their form having a direct influence on the exchange and alignment of surrounding conditions. In addition to the material properties themselves (e.g., moisture absorption capacity), several factors (e.g., the way the tomatoes are stacked in the packaging, the height of the packaging sides, and holes in the bottom) can influence the air exchange and air circulation, thus having a direct influence on overall quality changes of the stored tomatoes.

The tomatoes analyzed in this study showed almost full ripening at the beginning of the storage trials. According to López Camelo & Gómez (2004), chroma for tomatoes in (visual) red color stage was analyzed to be 29.9. Even if the results of this study are not in this range, the analyzed values are comparable among themselves. Therefore, no conclusion can be drawn about the influence of the tested packaging trays on tomato color change during ripening and even in the ripening state. The low color saturation at the end of storage for tomatoes stored in trays of bamboo and cellulose indicates color changes due to the onset of rotting processes, which were also shown by the assessment of decay incidence and the microbial growth of the tomatoes.

Buendía−Moreno et al. (2019) showed that no differences in tomato color changes occurred during storage between PE and cardboard trays. In the study of García-García et al. (2013), the peak of ripeness was reached after a storage period of two weeks. Especially during ripening, the used packaging material influences color changes. Advantageous storage characteristics of tomatoes are attributed to PLA-coated cardboard trays compared with uncoated trays because of the fact that a PLA coating significantly influences the value of the color parameter a/b by absorbing parts of the ethylene and thereby contributing to a delayed maturation (García-García et al., 2013).

Fruit firmness is an indicator of product freshness (Aurand et al., 2012). The texture of fruits changes immediately after harvest, during ripening and storage because of molecular and biochemical processes causing breakdown and deterioration of membrane and cell structure, cell wall composition and intracellular materials. The softening occurs in relation to turgor loss and enzyme-controlled cell wall loosening (Aurand et al., 2012; Bertin & Génard, 2018; Buendía−Moreno et al., 2019; Fagundes et al., 2015; Page et al., 2010; Rao et al., 2011).

Several authors stated that firmness of stored tomato progressively decreased during storage at 22°C resulting from moisture content loss and activation of enzymes that can degrade the cell wall of tomatoes (Al-Dairi et al., 2021; Tigist et al., 2013). Domínguez et al. (2016) attributed a slowed down process of ripening to the (modified) atmosphere and the high humidity inside packages decreasing the activity of enzymes involved in the cell wall degradation. Buendía−Moreno et al. (2019) reported no difference in product firmness between tomatoes packed in tray materials such as PE and cardboard after 24 days at 8°C. In a comparable study, García-García et al. (2013) demonstrated that the firmness of cherry tomatoes packaged in PLA-coated trays was better preserved compared with uncoated ones because of the positive effect of low concentrations of ethylene and water vapor on the firmness and other quality parameters of tomatoes during storage.

It was expected that an increase in decay incidence results in a decrease of firmness because of enzymatic-induced changes in cell wall structure. Therefore, well-preserved plant cell structures are required to avoid microbial infection of internal tissues reducing decay. However, as proven through color measurement, the investigated tomatoes in this study were already in a full ripened stage. This, in turn, makes it difficult to draw a concluding remark about the influence of the packaging on firmness at this final stage of ripening.

The microbial characteristics of the packaging materials were strongly related to the spoilage rate of the packaged tomatoes. The increase in yeasts and molds until the end of storage depends on the spoilage of the packaged tomatoes and on the material structure and the possibility for bacteria to grow on it. Therefore, the trend in increase of yeasts and molds on packaging materials from start to end of storage was comparable to the changes in TVC. Differences were found for the packaging materials PLA and bamboo. For PLA, the load of yeasts and molds was comparable with the value for the reference, corrugated cardboard. The values for bamboo even remained below.

Even if the clean and unused packaging materials were slightly contaminated with microorganisms due to their nature and production process, this contamination was lower than the microbial load of the tomatoes. However, there is a potential for cross contamination between packaging material and packaged product and vice versa. High initial microbial loads bear the potential of cross contamination with the packaged tomatoes, spreading bacteria and thereby accelerate spoilage of the packaged product upon storage. Considering this fact, sustainable and bio-based packaging materials are suitable in the same way as the reference materials for the storage of tomatoes. It can be assumed that the packaging materials fulfill the requirement of EU regulation No. 543/2011, which demands that packaging protects the product, and that the inner surfaces of the packaging are clean and designed in a way that considering the product, no changes are generated (European Commission, 2011).

While mechanical material properties provide stability and thus guarantee product protection during transport, the water absorption capacity also influences product quality parameters (e.g., weight loss) due to the influence on environmental conditions. The crucial factor is that natural fibers contain hydrophilic constituents, such as cellulose and hemicellulose, and are therefore prone to high humidity and moisture (Asim et al., 2017; Chen et al., 2018; Nor Arman et al., 2021). High water absorption capacities lead to swelling of natural fibers, and the absorbed water molecules will affect the normal matrix interactions and stimulate bacterial growth directly influencing product quality (Khalid et al., 2008; Nor Arman et al., 2021; Robledo-Ortíz et al., 2020). However, the actual water absorption is determined by several internal and external factors. The internal factors mainly comprise of the fiber's inherent characteristics, such as fiber orientation, porosity, and area of exposed surfaces. Among the external factors, humidity and temperature of soaking medium and surface protection are of importance (Dittrich et al., 2014; Jawaid & Abdul Khalil, 2011; Nor Arman et al., 2021; Ramlee et al., 2019). Hence, different materials naturally absorb moisture differently. The results shown for the water absorption capacity of the bio-based materials are in line with the weight loss described (chapter 3.4.3).

For example, the weight loss of tomatoes packaged in the reference trays of corrugated cardboard (which shows the highest determined water absorption capacity) is higher compared with materials such as groundwood pulp and sugar cane (whose water absorption capacity is in the range of one-tenth that of corrugated cardboard). However, it also shows that, despite coating achieving lower water absorption capacity than cellulose and bamboo, other factors have a great influence as well. As shown for decay incidence (chapter 3.4.3), several factors influence the quality changes of the stored tomatoes. In general, Buendía−Moreno et al. (2019) attributed a higher weight loss of tomatoes stored in uncoated cardboard trays compared to polyethylene trays to the water absorption properties of the cardboard itself.

In general, the recorded packaging parameters showed that, in most cases, bio-based packaging materials are preferable compared with the reference, corrugated cardboard, as they are lighter and thinner, resist a higher tensile strength, and exhibit low values for water absorption capacity.

During storage, these values are favorable because of stability and resistance during transportation.

3.6 Conclusion

Quality loss and shelf life of cherry tomatoes are mostly affected by storage temperature, gas concentrations, and relative humidity inside the packages. In practical conditions, both the material and the size of the trays have an impact on quality changes and shelf life of fresh cherry tomatoes and, thus, on the amount of food waste. Considering the totality of the examined parameters, tomatoes stored in groundwood pulp and sugar cane showed consistently good results with only minor quality changes during storage. These materials, at least, partly contribute to a better-preserved quality of the stored tomatoes compared with the references, corrugated cardboard and rPET. PLA and grass paper, on the other hand, showed strong and premature quality changes on the stored tomatoes, which is why these materials are not recommended for tomato packaging. It remains to be clarified whether packaging solutions made of innovative materials can guarantee the quality and safety of food in practical application and, thus, represent packaging that is equivalent to wood-based packaging.

Funding

This research was funded by the European Union via the European Regional Development Fund EFRE.NRW (Grant EFRE 0500035, "Biobasierte Produkte").

Acknowledgements

The authors wish to thank the company Bauer Funken GmbH who kindly provided the tomatoes for the storage trials, Alexandra Glawe and Rebecca Wegner for their highly appreciated, continuous assistance in the experimental parts and Lucas Correa Dresch for the performed final English revision of the manuscript.

References

Accorsi, R. (2019). A support-design procedure for sustainable food product-packaging systems. In *Sustainable Food Supply Chains* (pp. 61–81). Elsevier. https://doi.org/10.1016/B978-0-12-813411-5.00005-3.

Aggarwal, A., & Langowski, H.-C. (2020). Packaging Functions and Their Role in Technical Development of Food Packaging Systems: Functional Equivalence in Yoghurt Packaging. *Procedia CIRP*, *90*, 405–410. https://doi.org/10.1016/j.procir.2020.01.063.

Ahvenainen, R. (1996). New approaches in improving the shelf life of minimally processed fruit and vegetables. Review. *Trend in Food Science and Technology*, *7*, 179–187.

Akbudak, B., Akbudak, N., Seniz, V., & Eris, A. (2012). Effect of pre-harvest harpin and modified atmosphere packaging on quality of cherry tomato cultivars "Alona" and "Cluster". *British Food Journal*, *114*(2), 180–196. https://doi.org/10.1108/00070701211202377.

Al-Dairi, M., Pathare, P. B., & Al-Yahyai, R. (2021). Chemical and nutritional quality changes of tomato during postharvest transportation and storage. *Journal of the Saudi Society of Agricultural Sciences*, *20*(6), 401–408. https://doi.org/10.1016/j.jssas.2021.05.001

Alegbeleye, O., Odeyemi, O. A., Strateva, M., & Stratev, D. (2022). Microbial spoilage of vegetables, fruits and cereals. *Applied Food Research*, *2*(1), 100122. https://doi.org/10.1016/j.afres.2022.100122.

Almenar, E., Samsudin, H., Auras, R., Harte, B., & Rubino, M. (2008). Postharvest shelf life extension of blueberries using a biodegradable package. *Food Chemistry*, *110*(1), 120–127. https://doi.org/10.1016/j.foodchem.2008.01.066.

Arah, I. K., Kumah, E. K., Anku, E. K., & Amaglo, H. (2015). An Overview of Post-Harvest Losses in Tomato Production in Africa: Causes and Possible Prevention Strategies. *Journal of Biology, Agriculture and Healthcare*, *5*(16), 78–88.

Asim, M., Jawaid, M., Abdan, K., & Ishak, M. R. (2017). Effect of pineapple leaf fibre and kenaf fibre treatment on mechanical performance of phenolic hybrid composites. *Fibers and Polymers*, *18*(5), 940–947. https://doi.org/10.1007/s12221-017-1236-0.

Aurand, R., Faurobert, M., Page, D., Maingonnat, J.-F., Brunel, B., Causse, M., & Bertin, N. (2012). Anatomical and biochemical trait network underlying genetic variations in tomato fruit texture. *Euphytica*, *187*(1), 99–116. https://doi.org/10.1007/s10681-012-0760-7.

Ayomide, O. B., Ajayi, O. O., & Ajayi, A. A. (2019). Advances in the development of a tomato postharvest storage system: Towards eradicating postharvest losses. *Journal of Physics: Conference Series*, *1378*(2), 22064. https://doi.org/10.1088/1742-6596/1378/2/022064.

Bertin, N., & Génard, M. (2018). Tomato quality as influenced by preharvest factors. *Scientia Horticulturae*, *233*, 264–276. https://doi.org/10.1016/j.scienta.2018.01.056.

Bertling, J., Hamann, L., & Bertling, R. (2018). *Kunststoffe in der Umwelt: Mikro- und Makroplastik: Ursachen, Mengen, Umweltschicksale, Wirkungen, Lösungsansätze, Empfehlungen.* https://doi.org/10.24406/UMSICHT-N-497117.

Buendía−Moreno, L., Sánchez−Martínez, M. J., Antolinos, V., Ros−Chumillas, M., Navarro−Segura, L., Soto−Jover, S., Martínez−Hernández, G. B., & López−Gómez, A. (2020). Active cardboard box with a coating including essential oils entrapped within cyclodextrins and/or halloysite nanotubes. A case study for fresh tomato storage. *Food Control*, *107*, 106763. https://doi.org/10.1016/j.foodcont.2019.106763.

Buendía−Moreno, L., Soto−Jover, S., Ros−Chumillas, M., Antolinos, V., Navarro−Segura, L., Sánchez−Martínez, M. J., Martínez−Hernández, G. B., & López−Gómez, A. (2019). Innovative cardboard active packaging with a coating including encapsulated essential oils to extend cherry tomato shelf life. *LWT*, *116*, 108584. https://doi.org/10.1016/j.lwt.2019.108584.

Caldeira, C., Laurentiis, V. de, Corrado, S., van Holsteijn, F., & Sala, S. (2019). Quantification of food waste per product group along the food supply chain in the European Union: A mass flow analysis. *Resources, Conservation, and Recycling*, *149*, 479–488. https://doi.org/10.1016/j.resconrec.2019.06.011.

Castro, L. R. de, Vigneault, C., Charles, M. T., & Cortez, L. A. B. (2005). Effect of cooling delay and cold-chain breakage on "Santa Clara" tomato. *Journal of Food, Agriculture & Environment (JFAE)*.

Chen, R. S., Ahmad, S., & Gan, S. (2018). Rice husk bio-filler reinforced polymer blends of recycled HDPE/PET: Three-dimensional stability under water immersion and mechanical performance. *Polymer Composites*, *39*(8), 2695–2704. https://doi.org/10.1002/pc.24260.

Coelho, P. M., Corona, B., Klooster, R. ten, & Worrell, E. (2020). Sustainability of reusable packaging–Current situation and trends. *Resources, Conservation & Recycling: X*, *6*, Article 100037. https://doi.org/10.1016/j.rcrx.2020.100037.

Corrado, S., Ardente, F., Sala, S., & Saouter, E. (2017). Modelling of food loss within life cycle assessment: From current practice towards a systematisation. *Journal of Cleaner Production*, *140*, 847–859. https://doi.org/10.1016/j.jclepro.2016.06.050.

D'Aquino, S., Mistriotis, A., Briassoulis, D., Di Lorenzo, M. L., Malinconico, M., & Palma, A. (2016). Influence of modified atmosphere packaging on postharvest quality of cherry tomatoes held at 20 °C. *Postharvest Biology and Technology*, *115*, 103–112. https://doi.org/10.1016/j.postharvbio.2015.12.014.

Dannehl, D., Huyskens-Keil, S., & Schmidt, U. (2008). Untersuchungen zur Lagerung von Erdbeeren unter Berücksichtigung verschiedener Verpackungsmaterialien. *Erwerbs-Obstbau*, *50*(2), 49–61. https://doi.org/10.1007/s10341-008-0062-3.

Diaz-Perez, J. C., Bautista, S., & Villanueva, R. (2000). Quality changes in sapote mamey fruit during ripening and storage. *Postharvest Biology and Technology*, *18*(1), 67–73. https://doi.org/10.1016/S0925-5214(99)00062-9.

Dittrich, B., Wartig, K.-A., Mülhaupt, R., & Schartel, B. (2014). Flame-Retardancy Properties of Intumescent Ammonium Poly(Phosphate) and Mineral Filler Magnesium Hydroxide in Combination with Graphene. *Polymers*, *6*(11), 2875–2895. https://doi.org/10.3390/polym6112875.

Domínguez, I., Lafuente, M. T., Hernández-Muñoz, P., & Gavara, R. (2016). Influence of modified atmosphere and ethylene levels on quality attributes of fresh tomatoes (Lycopersicon esculentum Mill.). *Food Chemistry*, *209*, 211–219. https://doi.org/10.1016/j.foodchem.2016.04.049.

Ellen MacArthur Foundation and McKinsey & Company (2016). The New Plastics Economy: Rethinking the future of plastics. World Economic Forum. http://www.ellenmacarthurfoundation.org/publications.

European Commission (2011). Commission Implementing Regulation (EU) No 543/2011 of 7 June 2011 laying down detailed rules for the application of Council Regulation (EC) No 1234/2007 in respect of the fruit and vegetables and processed fruit and vegetables sectors. *Official Journal of the European Union*(L157).

European Commission (2018). A European Strategy for Plastics in a Circular Economy.

European Parliament and the Council of the European Union (2018). Directive (EU) 2018/852 of the European Parliament and of the Council of 30 May 2018 amending Directive 94/62/EC on packaging and packaging waste. *Official Journal of the European Union*(L150), 141–154.

Fagundes, C., Moraes, K., Pérez-Gago, M. B., Palou, L., Maraschin, M., & Monteiro, A. R. (2015). Effect of active modified atmosphere and cold storage on the postharvest quality of cherry tomatoes. *Postharvest Biology and Technology*, *109*, 73–81. https://doi.org/10.1016/j.postharvbio.2015.05.017.

Fagundes, C., Palou, L., Monteiro, A. R., & Pérez-Gago, M. B. (2014). Effect of antifungal hydroxypropyl methylcellulose-beeswax edible coatings on gray mold development and quality attributes of cold-stored cherry tomato fruit. *Postharvest Biology and Technology*, *92*, 1–8. https://doi.org/10.1016/j.postharvbio.2014.01.006.

García-García, I., Taboada-Rodríguez, A., López-Gomez, A., & Marín-Iniesta, F. (2013). Active Packaging of Cardboard to Extend the Shelf Life of Tomatoes. *Food and Bioprocess Technology*, *6*(3), 754–761. https://doi.org/10.1007/s11947-011-0759-4.

Godfray, H. C. J., Beddington, J. R., Crute, I. R., Haddad, L., Lawrence, D., Muir, J. F., Pretty, J., Robinson, S., Thomas, S. M., & Toulmin, C. (2010). Food security: The challenge of feeding 9 billion people. *Science (New York, N.Y.)*, *327*(5967), 812–818. https://doi.org/10.1126/science.1185383.

Heller, M. C., Selke, S. E. M., & Keoleian, G. A. (2019). Mapping the Influence of Food Waste in Food Packaging Environmental Performance Assessments. *Journal of Industrial Ecology*, *23*(2), 480–495. https://doi.org/10.1111/jiec.12743.

Hounsou, M., Dabadé, D. S., Götz, B., Hounhouigan, M. H., Honfo, F. G., Albrecht, A., Dresch, L. C., Kreyenschmidt, J., & Hounhouigan, D. J. (2022). Development and use of food packaging from plant leaves in developing countries. *Journal Für Verbraucherschutz Und Lebensmittelsicherheit.* Advance online publication. https://doi.org/10.1007/s00003-022-01390-0.

Jawaid, M., & Abdul Khalil, H. (2011). Cellulosic/synthetic fibre reinforced polymer hybrid composites: A review. *Carbohydrate Polymers*, *86*(1), 1–18. https://doi.org/10.1016/j.carbpol.2011.04.043.

Kalamaki, M. S., Stoforos, N. G., & Taoukis, P. S. (2012). Pectic Enzymes in Tomatoes. In B. K. Simpson (Ed.), *Food biochemistry and food processing* (2nd ed., pp. 232–246). Wiley-Blackwell. https://doi.org/10.1002/9781118308035.ch12.

Khalid, M., Ratnam, C. T., Chuah, T. G., Ali, S., & Choong, T. S. (2008). Comparative study of polypropylene composites reinforced with oil palm empty fruit bunch fiber and oil palm derived cellulose. *Materials & Design*, *29*(1), 173–178. https://doi.org/10.1016/j.matdes.2006.11.002.

Koide, S., & Shi, J. (2007). Microbial and quality evaluation of green peppers stored in biodegradable film packaging. *Food Control*, *18*(9), 1121–1125. https://doi.org/10.1016/j.foodcont.2006.07.013.

Korte, I., Kreyenschmidt, J., Wensing, J., Bröring, S., Frase, J. N., Pude, R., Konow, C., Havelt, T., Rumpf, J., Schmitz, M., & Schulze, M. (2021). Can Sustainable Packaging Help to Reduce Food Waste? A Status Quo Focusing Plant-Derived Polymers and Additives. *Applied Sciences*, *11*(11), 5307. https://doi.org/10.3390/app11115307.

Kreyenschmidt, J., Albrecht, A., Braun, C., Herbert, U., Mack, M., Rossaint, S., Ritter, G., Teitscheid, P., & Ilg, Y. (2013). Food Waste in der Fleisch verarbeitenden Kette. *Fleischwirtschaft*, *10*, 57–63.

Kreyenschmidt, J., & Ibald, R. (2012). Modeling Shelf Life Using Microbial Indicators. In M. C. Nicoli (Ed.), *Shelf life assessment of food* (pp. 127–163). Taylor and Francis.

Kummu, M., Moel, H. de, Porkka, M., Siebert, S., Varis, O., & Ward, P. J. (2012). Lost food, wasted resources: Global food supply chain losses and their impacts on freshwater, cropland, and fertiliser use. *The Science of the Total Environment*, *438*, 477–489. https://doi.org/10.1016/j.scitotenv.2012.08.092.

Linke, M., & Geyer, M. (2003). Postharvest behaviour of tomatoes in different transport packaging units. *Acta Horticulturae*(599), 115–122. https://doi.org/10.17660/ActaHortic.2003.599.13.

López Camelo, A. F., & Gómez, P. A. (2004). Comparison of color indexes for tomato ripening. *Horticultura Brasileira*, *22*(3), 534–537. https://doi.org/10.1590/S0102-05362004000300006.

Majidi, H., Minaei, S., Almassi, M., & Mostofi, Y. (2014). Tomato quality in controlled atmosphere storage, modified atmosphere packaging and cold storage. *Journal of Food Science and Technology*, *51*(9), 2155–2161. https://doi.org/10.1007/s13197-012-0721-0.

Marsh, K., & Bugusu, B. (2007). Food packaging—roles, materials, and environmental issues. *Journal of Food Science*, *72*(3), R39-55. https://doi.org/10.1111/j.1750-3841.2007.00301.x.

Matthews, C., Moran, F., & Jaiswal, A. K. (2021). A review on European Union’s strategy for plastics in a circular economy and its impact on food safety. *Journal of Cleaner Production*, *283*, 125263. https://doi.org/10.1016/j.jclepro.2020.125263.

Moneruzzaman, K. M., Hossain, A. B. M S., Sani, W., Saifuddin, M., & Alenazi, M. (2009). Effect of harvesting and storage conditions on the post harvest quality of tomato (Lycopersicon esculentum Mill) cv. Roma VF. *Australian Journal of Crop Science*, *3*(2), 113–121.

Naturschutzbund Deutschland (NABU) e.V. (2020). *Vorverpackungen bei Obst und Gemüse: Zahlen und Fakten 2010 bis 2016*. https://www.nabu.de/imperia/md/content/nabude/konsumressourcenmuell/201027-nabu-studie-vorverpackungen.pdf.

Nor Arman, N. S., Chen, R. S., & Ahmad, S. (2021). Review of state-of-the-art studies on the water absorption capacity of agricultural fiber-reinforced polymer composites for sustainable construction. *Construction and Building Materials*, *302*, 124174. https://doi.org/10.1016/j.conbuildmat.2021.124174.

Opara, U. L., Al-Ani, M. R., & Al-Rahbi, N. M. (2012). Effect of Fruit Ripening Stage on Physico-Chemical Properties, Nutritional Composition and Antioxidant Components of Tomato (Lycopersicum esculentum) Cultivars. *Food and Bioprocess Technology*, *5*(8), 3236–3243. https://doi.org/10.1007/s11947-011-0693-5.

Opara, U. L., & Mditshwa, A. (2013). A review on the role of packaging in securing food system: Adding value to food products and reducing losses and waste. *African Journal of Agrucultural Research*, *8*(22), 2621–2630.

Page, D., Gouble, B., Valot, B., Bouchet, J. P., Callot, C., Kretzschmar, A., Causse, M., Renard, C. M. C. G., & Faurobert, M. (2010). Protective proteins are differentially expressed in tomato genotypes differing for their tolerance to low-temperature storage. *Planta*, *232*(2), 483–500. https://doi.org/10.1007/s00425-010-1184-z.

Patrignani, F., Siroli, L., Gardini, F., & Lanciotti, R. (2016). Contribution of Two Different Packaging Material to Microbial Contamination of Peaches: Implications in Their Microbiological Quality. *Frontiers in Microbiology*, *7*, 938. https://doi.org/10.3389/fmicb.2016.00938.

Paull, R. E. (1999). Effect of temperature and relative humidity on fresh commodity quality. *Postharvest Biology and Technology*, *15*(3), 263–277. https://doi.org/10.1016/S0925-5214(98)00090-8.

Pelka, M., & Kreyenschmidt, J. (2013). *Ermittlung des Verderbs von frischem Obst und Gemüse in Abhängigkeit der Verpackungsart: Ergebnisbericht.*

Pro Carton (2010). Wie wichtig ist nachhaltige Verpackung? Die Einstellung von Konsumenten zu Verpackung und Nachhaltigkeit, *3*.

Qin, Y., & Horvath, A. (2022). What contributes more to life-cycle greenhouse gas emissions of farm produce: Production, transportation, packaging, or food loss? *Resources, Conservation and Recycling*, *176*, 105945. https://doi.org/10.1016/j.resconrec.2021.105945.

Ramlee, N. A., Jawaid, M., Zainudin, E. S., & Yamani, S. A. K. (2019). Tensile, physical and morphological properties of oil palm empty fruit bunch/sugarcane bagasse fibre reinforced phenolic hybrid composites. *Journal of Materials Research and Technology*, *8*(4), 3466–3474. https://doi.org/10.1016/j.jmrt.2019.06.016.

Rao, T. R., Gol, N. B., & Shah, K. K. (2011). Effect of postharvest treatments and storage temperatures on the quality and shelf life of sweet pepper (Capsicum annum L.). *Scientia Horticulturae*, *132*, 18–26. https://doi.org/10.1016/j.scienta.2011.09.032.

Rhim, J.-W., Lee, J.-H., & Hong, S.-I. (2007). Increase in water resistance of paperboard by coating with poly(lactide). *Packaging Technology and Science*, *20*(6), 393–402. https://doi.org/10.1002/pts.767.

Robertson, G. L. (2013). *Food packaging: Principles and practice* (3. ed.). CRC Press.

Robledo-Ortíz, J. R., González-López, M. E., Rodrigue, D., Gutiérrez-Ruiz, J. F., Prezas-Lara, F., & Pérez-Fonseca, A. A. (2020). Improving the Compatibility and Mechanical Properties of Natural Fibers/Green Polyethylene Biocomposites Produced by Rotational Molding. *Journal of Polymers and the Environment*, *28*(3), 1040–1049. https://doi.org/10.1007/s10924-020-01667-1.

Scherhaufer, S., Moates, G., Hartikainen, H., Waldron, K., & Obersteiner, G. (2018). Environmental impacts of food waste in Europe. *Waste Management*, *77*, 98–113. https://doi.org/10.1016/j.wasman.2018.04.038.

Sharma, R., & Ghoshal, G. (2018). Emerging trends in food packaging. *Nutrition & Food Science*, *48*(5), 764–779. https://doi.org/10.1108/NFS-02-2018-0051.

Shewfelt, R. L. (1986). Postharvest treatment for extending the shelf life of fruits and vegetables. *Food Technology (USA)*.

Soliva-Fortuny, R. C., & Martín-Belloso, O. (2003). New advances in extending the shelf-life of fresh-cut fruits: a review. *Trends in Food Science & Technology*, *14*(9), 341–353. https://doi.org/10.1016/S0924-2244(03)00054-2.

Tigist, M., Workneh, T. S., & Woldetsadik, K. (2013). Effects of variety on the quality of tomato stored under ambient conditions. *Journal of Food Science and Technology*, *50*(3), 477–486. https://doi.org/10.1007/s13197-011-0378-0.

Tripathi, P., & Dubey, N. (2004). Exploitation of natural products as an alternative strategy to control postharvest fungal rotting of fruit and vegetables. *Postharvest Biology and Technology*, *32*(3), 235–245. https://doi.org/10.1016/j.postharvbio.2003.11.005.

Umweltbundesamt (2018). What Matters: Recycling. *Magazine of the German Environment Agency*(1).

United Nations (2015). Transforming our world: The 2030 Agenda for Sustainable Development. *General Assembly*, *A/Res/70/1.* https://doi.org/10.1002/9781119541851.app1.

van Eygen, E., Feketitsch, J., Laner, D., Rechberger, H., & Fellner, J. (2017). Comprehensive analysis and quantification of national plastic flows: The case of Austria. *Resources, Conservation and Recycling*, *117*, 183–194. https://doi.org/10.1016/j.resconrec.2016.10.017.

Wei, Y., Zhou, D., Wang, Z., Tu, S., Shao, X., Peng, J., Pan, L., & Tu, K. (2018). Hot air treatment reduces postharvest decay and delays softening of cherry tomato by regulating gene expression and activities of cell wall-degrading enzymes. *Journal of the Science of Food and Agriculture*, *98*(6), 2105–2112. https://doi.org/10.1002/jsfa.8692.

Wikström, F., Verghese, K., Auras, R., Olsson, A., Williams, H., Wever, R., Grönman, K., Kvalvåg Pettersen, M., Møller, H., & Soukka, R. (2019). Packaging Strategies That Save Food: A Research Agenda for 2030. *Journal of Industrial Ecology*, *23*(3), 532–540. https://doi.org/10.1111/jiec.12769.

Žnidarčič, D., Ban, D., Oplanić, M., Karić, L., & Požrl, T. (2010). Influence of postharvest temperatures on physicochemical quality of tomatoes (Lycopersicon esculentum Mill.). *Journal of Food, Agriculture & Environment*, *8*(1), 21–25.

4 Quality impact of sustainable MA-packaging options for emulsion-type sausage: A German case study

Imke Korte[1,*], Antonia Albrecht[1], Maureen Mittler[1], Claudia Waldhans[1], Judith Kreyenschmidt[1,2]

[1] Rheinische Friedrich Wilhelms-University Bonn, Institute of Animal Sciences, Katzenburgweg 7-9, D-53115 Bonn, Germany

[2] Hochschule Geisenheim University, Department of Fresh Produce Logistics, Von-Lade-Straße 1, D-65366 Geisenheim, Germany

* Correspondence

Korte, I., Albrecht, A., Mittler, M., Waldhans, C., & Kreyenschmidt, J. (2023). Quality impact of sustainable ma-packaging options for emulsion-type sausage: A German case study. *Future Foods*, *7*, 100218. https://doi.org/10.1016/j.fufo.2023.100218.

4.1 Abstract

The industrial packaging of meat products is an important process factor to preserve high quality and prolong shelf life by reducing microbial growth, lipid oxidation, and enzymatic autolysis. Currently, legislation and regulations are forcing companies to eliminate or reduce the use of (conventional fossil-based plastic) packaging.

This case study in the German meat industry was designed to ascertain the effectiveness of four innovative packaging materials, differing in foil thickness and composition, to preserve the quality of MA-packed emulsion-type sausages during its shelf life compared to a conventional control packaging (multi-layer APET/PE 300µ/62µ). During storage, different product parameters and the atmosphere inside the packages were investigated to analyze the effect on product quality and shelf life.

Except for one plastic-reduced multi-layer APET/PE 250µ/47µ, all materials showed significantly higher values for defect packages and gas permeability compared to the control. The decline in product quality varied between the materials showing no correlation between microbial spoilage and a decrease in product quality for emulsion-type sausages in the tested materials in comparison to the control. Besides the control, the multi-layer APET/PE 230µ/62µ was most effective in keeping the quality of MA-packed emulsion-type sausages and preserved an acceptable product quality during its shelf life.

4.2 Introduction

Plastic, with its multiple functions and durability, is an ubiquitous material in our economy. The development and promotion of more sustainable materials and strategies get increasing attention on social and political levels in the EU, leading to the launch of the “European Strategy for Plastics in a Circular Economy” in 2018 (European Commission, 2018). To reduce environmental impacts, all plastic packaging placed on the EU market must be completely reusable and/or recyclable by 2030 (European Commission, 2018). The multi-layer structure makes plastic one of the most complex material mixtures from a recycling perspective (Ragaert et al., 2017). Hence, from an environmental, sustainable, and biocompatible perspective, especially the use of multi-layered packaging materials must be reduced.

Based on their chemical and technical properties, the large group of multi-layer materials is suitable throughout the food industry due to their adjustability to the diverse requirements of different food groups offering effective packaging solutions to increase safety, quality, shelf life and thus to reduce food waste (Ellen MacArthur Foundation and McKinsey & Company, 2016; Faraca and Astrup, 2019; Kreyenschmidt et al., 2013; Matthews et al., 2021; Soro et al., 2021). Therefore, food packaging is of high importance concerning sustainable and resource protecting food production (European Commission, 2020; United Nations, 2020).

Especially the gas and water vapor permeability of the packaging material is of high relevance for meat and meat products (Barlow & Morgan, 2013; Matthews et al., 2021; Sharma & Ghoshal, 2018). As meat consists of a highly concentrated matrix of proteins, moisture, and fats, a combined effect of microbial and endogenous enzymes, such as proteases and lipases, causes food deterioration and promoting lipid as well as protein oxidation (Bekhit et al., 2021; Comi, 2016). To reduce the reaction rate of these processes, the barrier against oxygen is a key factor (Barlow & Morgan, 2013; Herbert et al., 2013). Ethylene vinyl alcohol (EVOH) copolymers are widely used as high barrier layers in multi-layer packaging materials due to their very low permeability of gases (like O_2 and CO_2) and organic vapors (Gavara et al., 2016; López-Rubio, 2011). Therefore, materials for modified atmosphere packaging (MAP) of meat, consisting of a sealed two-component lid/tray system typically comprising multiple joined material layers, comprise low-density polyethylene (PE-LD), linear low-density polyethylene (PE-LLD), polyamide (PA), polypropylene (PP), EVOH and polyethylene terephthalate (PET) (Barukčić et al., 2020; Kargwal et al., 2020; Seier et al., 2022; Walker et al., 2020).

Essential packaging functionalities, such as mechanical stability and barrier function against gas diffusion, depend on the material itself and the integrity of the packaging. Maintaining the modified atmosphere by hindering gas diffusion through the material and damage from external forces like puncture, tearing is of most importance (Chen et al., 2020; Rossaint et al., 2014; Seier et al., 2022). Small leaks affect the gas barrier properties causing an accelerated oxygen, water vapor, and flavor transfer off the packaging leading to a shortened shelf life of gas- or flavor-sensitive products. Furthermore, small leaks in food packaging can lead to microbial penetration (Chung et al., 2003).

There are different strategies to develop more sustainable packaging solutions which still offer the necessary moisture or oxygen barriers to preserve the product quality and to reduce the environmental impact of the packaging itself: reducing packaging materials by decreasing the thickness, reducing the number of layers, using easily recyclable materials or recycled materials, e.g., rPET, applying paper composite, bio-based and/or biodegradable materials (Pro Carton, 2010; Soro et al., 2021; Teck Kim et al., 2014). Reducing the environmental impact of the packaging material for meat can be best achieved by the minimization of used fossil-based materials such as thinner layers and paper composite materials that retain mechanical and barrier properties rather than emphasizing end-of-life issues like recycling or disposal (Barlow & Morgan, 2013). Furthermore, the material properties of simple mono-layer materials can be improved by the design of the packaging itself or by using active packaging solutions (Schumann & Schmid, 2018; Soro et al., 2021; Yildirim et al., 2018). Currently, there are no competitive alternatives that offer the same level of protection as multi-layer plastic packaging, especially for fresh products like meat (Matthews et al., 2021). Innovations in food packaging still need to fulfill the requirement (Matthews et al., 2021) to reduce food waste by maintaining the shelf life, quality and safety while minimizing packaging waste (Molina-Besch et al., 2019; Wikström et al., 2019; Williams & Wikström, 2011). However, investigations about the effects of new packaging on organoleptic and microbial quality, and the shelf life of meat products are rare.

The case study was aimed to investigate the effect of four possible substitutions (test materials) of conventional multi-layered amorphous polyethylene terephthalate/polyethylene (APET/PE) (control) on the quality loss of emulsion-type sausages in order to reduce packaging and food waste. Multi-layer foils variing in foil thickness, a mono-layer foil, and an innovative paper-polyethylene compound (Paper-PE) were tested.

4.3 Materials and methods

4.3.1 Product and packaging samples

Three types of emulsion-type sausage were investigated: Pure emulsion-type sausage (Mortadella), ham sausage (Bierschinken), and mushroom ham sausage (Champignon-Schinkenwurst). The detailed ingredients and nutritional information, as supplied by the manufacturer, are listed in Table 4.1. All product types were sliced and packaged in packaging materials differing in foil thickness using an oxygen-free modified atmosphere with a gas mixture of 30 % CO_2 and 70 % N_2, a typical gas composition used for meat products in Germany. The given shelf life of the products was 28 days.

Table 4.1 Composition and nutritional information as supplied by the manufacturer for the three investigated types of emulsion-type sausage.

	Emulsion-type sausage (Mortadella)	Ham sausage (Bierschinken)	Mushroom ham sausage (Champignon-Schinkenwurst)
Product code	M	B	C
	Ingredients		
Main	85 % pork meat	88 % pork meat	61 % pork meat; 20 % mushrooms
Minor (same ingredients present in all of the products)	Drinking Water; salt; spices; spices extracts; dextrose; glucose syrup; acidity regulator: sodium citrate; stabilizer: diphosphate; antioxidants: ascorbic acid, sodium ascorbate; preservative: sodium nitrite.		
	Nutritions [%]		
Fat	20	10	24
Carbohydrates	1	1	1
Protein	14.0	16.8	9.7
Salt	2.50	2.30	2.25

The selection of packaging materials, as well as upper and under foil combinations, was performed by the meat processing company in cooperation with the University of Bonn. Therefore, this study was designed and conducted as a case study. In total, the case study covered the investigation of an APET/PE multi-layer ((C) APET/PE 300µ/62µ) as control and four material combinations: two multi-layer foils with reduced foil thickness ((1) APET/PE 250µ/47µ and (2) APET/PE 230µ/62µ), one mono-layer foil ((3) Mono APET 200µ/52µ) and one paper-polyethylene compound ((4) Paper-PE/62µ). The characteristic material properties (thickness and permeabilities), based on the packaging specifications provided by the packaging suppliers, are shown separately for upper and under foil in Table 4.2.

Table 4.2 Overview of packaging materials used for storage trials with material characteristics.

Material code	**Material/layers (outside to inside)**	**Thickness [µm]**	**Permeability**	
			O_2 [$cm^3/m^2/d$]	**Water vapor [$g/m^2/d$]**
	Upper foil			
PET 62 (Control)	PET-print-k/PE-hv-PA-EVOH-PA-hv-PE	62	2.5**	1.2***
PET 47	PET-print-k-PE-hv-PA-EVOH-PA-hv-PE	47	2.5**	1.2***
Mono 52	OPET-AD-PE-EVOH-PEEL on PET	52	<3.0**	<9.4****
	Under foil			
APET 300 (Control)	APET/PE-EVOH-PE PEEL	300	<3.0**	<9.0***
APET 250	APET/PE-EVOH-PE PEEL	250	<3.0**	<9.5***
APET 230	APET/PE-EVOH-PE PEEL	230	<3.0**	<9.5***
Mono 200	APET-APET BARRIER-APET	200	<1.0**	<9.5****
Paper-PE compound	Paper-polyethylene with oxygen barrier (EVOH)	400*	<3.0*****	<8.0****

*Grammage (326 g/cm²)
**23°C, 0 % RH; ***23°C, 85 % RH; ****38°C, 90 % RH; *****23°C, 50 % RH

4.3.2 Study design and sampling

The products were produced and MA-packaged in a German meat processing factory and transported to the laboratory in a cooling truck under temperature-controlled conditions. The products arrived at the laboratory within 48 h after production. Packages were stored under temperature-controlled conditions at 7°C, which is the recommended storage temperature of the manufacturer, for 45 days in high-precision low-temperature incubators (Sanyo MIR 153, Sanyo Electric Co., Ora-Gun, Gumma, Japan). During the experiments, data loggers (Testo, Escort Junior; ESCORT JUNIOR Internal Temperature Data Logger, Escort, Auckland, New Zealand) recorded the storage temperatures every 10 min. A total of 349 pork sausage packages were tested in five storage trials to assess the development of quality parameters. Tests were conducted at appropriate time intervals over a storage period of 45 days. The analyzed parameters included measurements of the modified gas atmosphere inside the packages, microbiological investigations, and the color of the product, which was instrumentally measured. Tests were conducted on days 3, 7, 14, 21, 28, 31, 39, and 45 of storage. The number of samples for the different quality parameters are outlined in Table 4.3.

Table 4.3 Experimental design of the conducted analyses for quality parameters and the relevant total number of analyzed samples.

	Product Code	Total Number of Analyzed Samples [n]		
		Gas Atmosphere	Microbiological Analysis	Color
Multi-layer				
(Control) APET/PE 300µ/62µ	M/B/C	73	73	30
(1) APET/PE 250µ/47µ	M/B	94	59	35
(2) APET/PE 230µ/62µ	M	60	31	30
Mono-layer				
(3) Mono APET 200µ/52µ	M/B	55	55	31
Paper-PE compound				
(4) Paper-PE/62µ	M/B/C	67	67	24

4.3.3 Gas barrier properties of the packaging materials

Concentrations of oxygen (O_2) and carbon dioxide (CO_2) inside the trays were monitored over the storage period with a handheld gas analyzer with an accuracy of ± 0.1 % O_2 and ± 2 % CO_2 (Oxybaby V O_2/CO_2, WITT-Gasetechnik GmbH & Co KG, Witten, Germany). The headspace in the packages was sampled with a syringe needle, and per measurement 10 mL of headspace gas was withdrawn through a self-adhesive sealing pad on the package. The gas volume was absorbed in 15 s, and the O_2 concentration was detected by an electrochemical sensor. The gas analyzer uses infrared (IR)-absorption to detect the CO_2 concentration. Gas concentrations are given as volume percentages of the total packaging atmosphere. Every single package was measured three times at the same puncture spot, and the mean value was calculated. According to the producer, packages with an O_2 concentration above 0.5 % are considered as damaged or inappropriate. The value of 0.5 % is the technical limit which is attributed to the accuracy of the machines. A variation below this limit is within the scope.

4.3.4 Microbiological analysis

For microbiological analysis, mixed product samples of 25 g were cut from three meat sausage slices aseptically using a sterile scalpel. Samples were transferred to a filtered sterile stomacher bag and filled up with 225 mL saline tryptone diluent (0.85 % NaCl-saline-tablets (Oxoid BR0053G, Cambridge, UK) with 0.1 (m/v) % tryptone (VWR, Leuven, Belgium)). The samples were homogenized separately for 60 s in a Stomacher 400 (Kleinfeld Labortechnik, Gehrden, Germany). Decimal serial dilutions of the homogenates were made using saline tryptone diluent. Appropriate dilutions of the homogenates were investigated for the natural flora (total viable count (TVC), yeasts and molds, lactic acid bacteria (LAB), and Enterobacteriaceae). TVC was determined by pour plate technique (1 mL) on plate count agar (Merck, Darmstadt, Germany). The agar plates were incubated at 30°C for 72 h. Yeasts and molds were detected by spread plate technique (0.1 mL) on Yeast extract glucose chloramphenicol agar (Merck, Darmstadt, Germany), and plates were incubated at 25°C for 120 h. LAB were determined by pour plating (1 mL) on de Man, Rogosa, Sharpe agar (Merck, Darmstadt, Germany), then agar plates were incubated aerobically at 37°C for 72 h. Enterobacteriaceae were identified by overlay treatment (1 mL) on violet red bile dextrose agar (Merck, Darmstadt, Germany) by incubating the agar plates at 37°C for 24 h. Each sample was analyzed and enumerated in duplicate. Counts of colony-forming units (cfu) were expressed as $\log_{10}$ cfu/g for each medium and sample.

4.3.5 Color measurements

The color of the pure emulsion-type sausage was measured using a large view spectrophotometer (ColorFlex EZ 4500L, HunterLab, Murnau, Germany). The color measurement was conducted at a wavelength between 400 nm and 700 nm and with a 45°/0° geometry. The CIE 1976 L*a*b* scale was used (international standard, internal company software); measured with D65 illuminant (6500 K daylight). For the measurement, the emulsion-type sausage slices were placed on the glass surface of the measurement device. The color of three slices per package was measured and a mean value was calculated for each package (=sampling point).

4.3.6 Data analysis and statistics

Since the criteria for normal distribution and homoscedasticity were not met by most of the parameters, non-parametric testing was selected for all statistical analyses. Differences between the packaging materials were analyzed with the Kruskal-Wallis-Test. The significance level was defined as $P \leq 0.05$. The given significance values are Bonferroni corrected. Box plots were used for describing the data, and the median was applied to compare the results due to its characteristic of not being biased by extreme values or outliers. Data analysis was conducted with SPSS Statistics 27 (IBM Corp. 1989, 2020, New York, USA). The microbial growth data were fitted using the software ORIGIN 8.0G (OriginLab Corporation, Northampton, MA, USA).

The total viable count was used as a parameter for the determination of microbial shelf life, here exemplified by one selected product of emulsion-type sausage (Mortadella). When TVC reached a level of 6.7 $\log_{10}$ cfu/g, the product was considered spoiled according to the microbiological standard and warning values for food (German Society for Hygiene and Microbiology (DGHM) e.V.).

4.4 Results

The measured gas atmosphere inside the packaging was analyzed for significant differences between the different types of emulsion-type sausage to exclude any influence of different product compositions. It was shown that the product itself does not have an influence (data not shown). Thus, all analyses only differentiate between the packaging material (independent of the type of product).

4.4.1 Gas barrier properties of the packaging materials

The measurements of the O_2 concentration inside the packages, monitored over the entire storage period, showed a high variation between the different packaging materials (Figure 4.1). No significant difference in the O_2 concentration was detected between the control and the multi-layer APET/PE 250µ/47µ. Whereas significant differences were shown between the control and the multi-layer APET/PE 230µ/62µ ($P < 0.001$), the mono-layer Mono APET 200µ/52µ ($P = 0.001$) as well as the paper compound paper-PE/62µ ($P = 0.001$). Among the multi-layer APET/PE 230µ/62µ, the mono-layer Mono APET 200µ/52µ, and the paper compound paper-PE/62µ, no significant differences were observed.

When comparing O_2 levels within the different packaging solutions, only the control and multi-layer APET/PE 250µ/47µ can be considered intact packages according to the critical limit of 0.5 % O_2. The median, of the O_2 concentrations, for the control and multi-layer APET/PE 250µ/47µ are below this value.

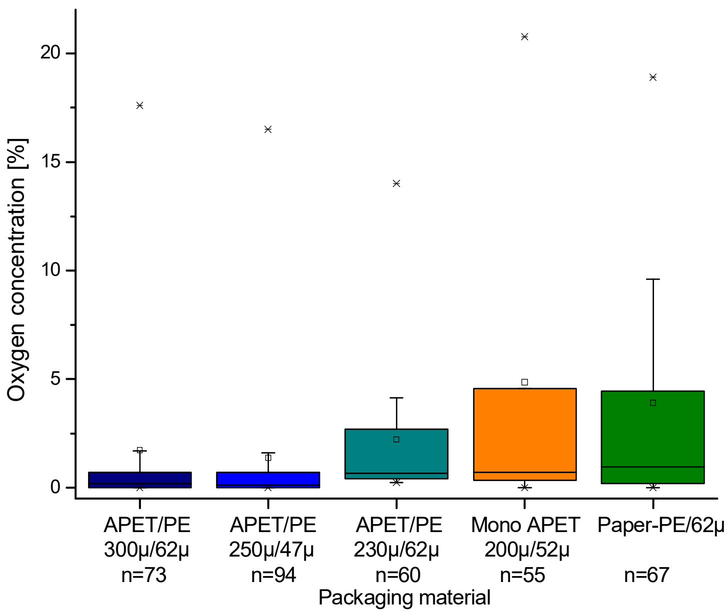


Figure 4.1 O_2 concentrations inside all analyzed packages for different packaging materials over the entire storage.

Considering only the O_2 concentrations of the intact packages ($O_2 \leq 0.5$ %) (Figure 4.2), the median of the control and the paper compound paper-PE/62µ is 0.1 % O_2. The value for the multi-layer APET/PE 250µ/47µ is slightly lower with 0.0 % O_2 compared to the control. The O_2 concentrations of the other packaging materials (multi-layer APET/PE 230µ/62µ and mono-layer Mono APET 200µ/52µ), compared to the control, showed with a median of 0.3 % O_2 higher values. Even the measured O_2 concentrations for the mono-layer Mono APET 200µ/52µ and the paper compound paper-PE/62µ varied more than those of the other investigated packaging materials, including the control.

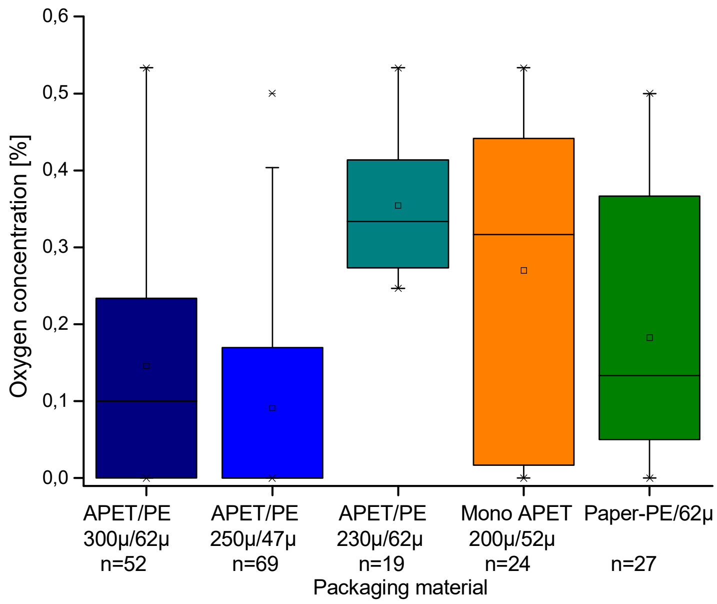


Figure 4.2 O_2 concentrations inside the intact packages ($O_2 \leq 0.5$ %) for different packaging materials over the entire storage.

Taking the critical limit of 0.5 % O_2 into account, the analysis of intact and vice versa defect packages showed differences between the five packaging materials (Table 4.3). For all packaging materials, the percentage of defect packages was higher compared to the control (28.8 %) except for the multi-layer APET/PE 250µ/47µ (26.6 % defective packages) (Table 4.4). The percentage of defect packages increased to 56.4 % when using the mono-layer Mono APET 200µ/52µ without any EVOH layer. The same trend is shown when the thickness of the under foil (multi-layer APET/PE 300µ) becomes too thin and decreases to 230µ (68.3 % defect packages) followed by the paper-PE compound used as under foil (59.7 %).

Table 4.4 Percentage of intact ($O_2 \leq 0.5$ %) and defect ($O_2 > 0.5$ %) packages of different packaging materials.

Packaging material						
	Under foil	APET/PE 300µ	APET/PE 250µ	APET/PE 230µ	Mono APET 200µ	Paper-PE compound
	Upper foil	PET/PE 62µ	PET/PE 47µ	PET/PE 62µ	Mono APET 52µ	PET/PE 62µ
Material code		**Control**	**1**	**2**	**3**	**4**
Intact packages ($O_2 \leq 0.5$ %) [%]		71.2	73.4	31.7	43.6	40.3
Defect packages ($O_2 > 0.5$ %) [%]		28.8	26.6	68.3	56.4	59.7

4.4.2 Microbiological analysis

The microbiological analyses pointed out that emulsion-type sausages showed a high quality upon arrival at the laboratory, with TVC ranging between 1.0 ±0.0 $\log_{10}$ cfu/g and 2.3 ± 0.8 $\log_{10}$ cfu/g. The microbial count of emulsion-type sausages, including all investigation points and samples (intact and defect packages), in different packaging materials, is shown in Figure 4.3. Considering the entire storage, TVC showed the lowest median for the control and multi-layer APET/PE 230µ/62µ with 1.1 $\log_{10}$ cfu/g and 1.0 $\log_{10}$ cfu/g, respectively. A decrease in foil thickness of under and upper foil, or a change in the material itself, led to an increase in the median of TVC during the storage period. No growth of yeasts and molds, and LAB was determined for the control and multi-layer APET/PE 230µ/62µ and the paper compound paper-PE/62µ. The multi-layer APET/PE 250µ/47µ was the only material where the growth of LAB occurred. The mono-layer Mono APET 200µ/52µ led to an increase in the growth of yeasts and molds compared to the other investigated materials. The mean bacterial load of Enterobacteriaceae was under the detection limit for all tested packaging materials (data not shown).

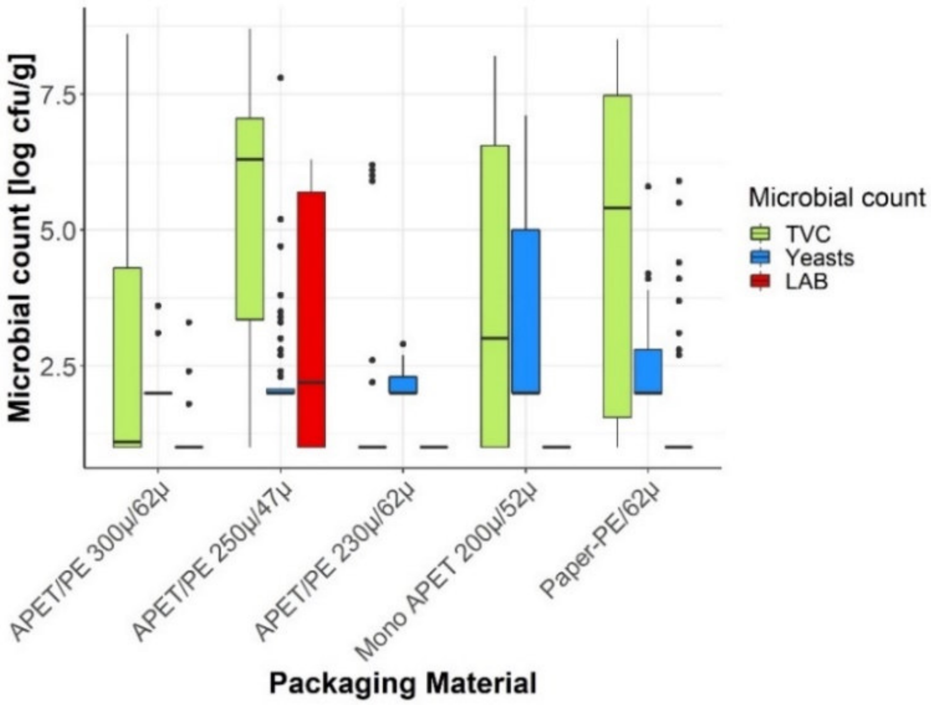


Figure 4.3 Microbial count of total viable count (TVC), yeasts and molds, and lactic acid bacteria (LAB) including all investigation points and samples (intact and defect packages) of emulsion-type sausages packaged in different innovative packaging materials.

4.4.3 Microbial shelf life

According to the German Society for Hygiene and Microbiology (DGHM), a product is considered spoiled when TVC reaches 6.7 log_{10} cfu/g. As the growth curve of TVC shows, exemplified by the emulsion-type sausage Mortadella (Figure 4.4), the time to reach this critical TVC level varied between products packaged in different innovative materials. A TVC of 6.7 log_{10} cfu/g was reached after 26 days for the multi-layer APET/PE 250µ/47µ as well as the mono-layer Mono APET 200µ/52µ and after ten days for the paper compound paper-PE/62µ. A TVC of 6.7 log_{10} cfu/g was not reached for products in control and multi-layer APET/PE 230µ/62µ packages over the entire storage period. Similar results were observed for the investigated ham sausage (Bierschinken) and mushroom ham sausage.

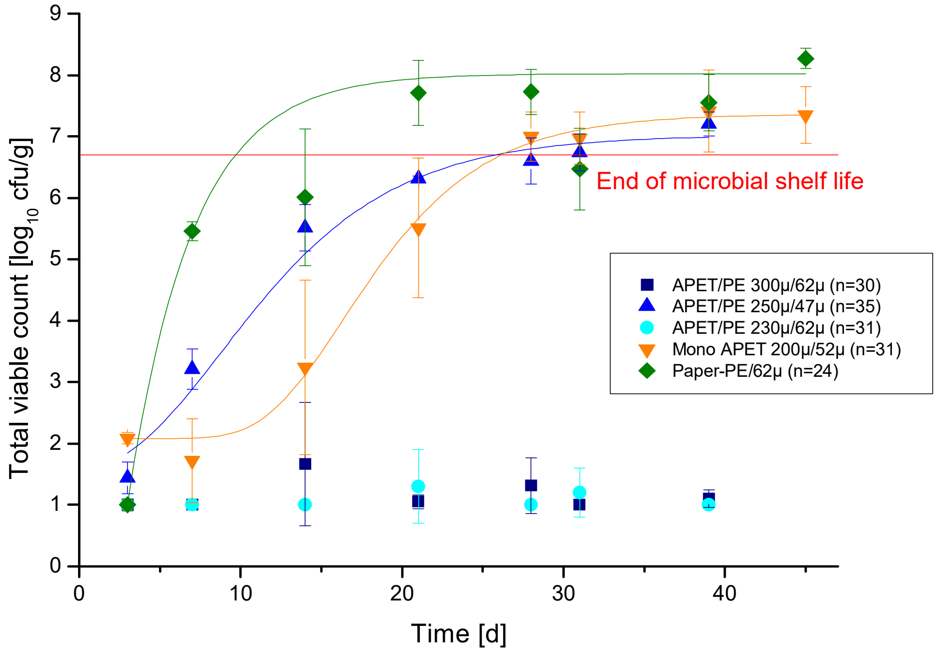


Figure 4.4 Total viable count (TVC) for different innovative packaging materials exemplified by the emulsion-type sausage Mortadella over a storage period of 45 days at 7°C.

4.4.4 Color measurements

No changes and trends were observed for L*, a*, b* values during storage time and between packaging materials. The L* values fluctuated between 69.2 and 70.7 for all packaging materials (data not shown). Only slight differences were observed for a* values during storage time. Over time, a* values differ less than 0.4. Amongst the tested packaging materials, initial a* values ranged from 6.0 to 7.0 (data not shown). No remarkable changes in yellowness (b* values) were measured in this study. B* values ranged from 10.7 to 12.5 (data not shown).

4.5 Discussion

According to the results of this study, both the mono-layer Mono APET 200µ/52µ without any EVOH layer and a decrease in foil thickness of the multi-layer APET/PE 300µ (under foil) as well as the paper compound paper-PE/62µ lead to changes in the gas atmosphere resulting in a high percentage of defect packages. The specified permeabilities for O_2 and water vapor (Table 4.2) refute the assumption that a change in material or its thickness is associated with an increase in permeability. Considering the different investigated packaging materials, it is assumed that the gas atmosphere is affected by the barrier properties compared to the multi-layer foils, especially for the paper compound paper-PE/62µ. As embedding of EVOH is a key factor to guarantee high-barrier properties, it is conceivable that the barrier properties in the paper-PE compound differ greatly (under the same environmental conditions) compared to the multi-layer foils due to the lack of protective layers. Depending on surrounding conditions, especially relative humidity, a moisture uptake by the EVOH layer favors an increased O_2 permeability (Alipour et al., 2015;

Blanchard et al., 2017; Cabedo et al., 2006; Mokwena et al., 2011). Additionally, increased O_2 contents result from permeabilities under test conditions that deviate from the specified values. It must be considered that the specified values refer to different test conditions (temperature and humidity), which makes it difficult to compare these values directly. Furthermore, according to the DIN standard for O_2 permeability (DIN 53380) and water vapor permeability (DIN 53122), the conditions under which permeabilities are tested are not the conditions to which materials in cold chains are exposed typically.

The high percentage of defect packages for the multi-layer APET/PE 230µ/62µ, the mono-layer Mono APET 200µ/52µ, and the paper compound paper-PE/62µ may be related to the sealing quality in this study. Several authors reported that micro-leakage in the sealing caused by improper sealing conditions reduces the gas barrier properties of a packaging material (Chung et al., 2003; Rossaint et al., 2014; Zardetto et al., 2022).

Hauptmann et al. (2021) studied the sealing behavior of paper-based film laminates, among others. Their findings show that the seal strength of paper-based thin film materials depends on the moisture content of the material itself. The heat sealing of paper materials is improved by increased sealing time and sealing pressure as this leads to a sufficient heat transfer. Sealing pressure and moisture content of paper-based materials are the most important variables to be adjusted; as sealing time is typically minimized for the highest productivity, paper-based materials need control of climate conditions during storage, production, and transportation, or conditioning before being applied on the packaging machines (Hauptmann et al., 2021). Due to the specified barrier properties, and the high percentage of defect packages in this study, its results point out that it is difficult to find the optimal sealing settings due to many external factors. Although an attempt was made to adjust the optimal sealing settings, the defect packages of the described level still occurred. Nevertheless, in addition to the barrier properties, this is a factor that causes high defects. Any kind of defect needs to be avoided as this causes additional rejects and increases food waste due to undesirable and early quality changes.

Regarding the microbial results of this study, it must be emphasized that a certain variation for TVC as well as differences in the microflora were shown for different packaging materials contrary to expectations. Neither Enterobacteriaceae nor LAB nor yeasts and molds were clearly dominating germs between different packaging materials. The higher variations of TVC analyzed for the mono-layer Mono APET 200µ/52µ and the paper compound paper-PE/62µ can be explained by the more heterogenous microflora due to the higher percentage of packages with increased O_2 content compared to the control multi-layer. The growth of LAB increasingly occurs on emulsion-type sausage packaged in the multi-layer APET/PE 250µ/47µ whereas the growth of yeasts occurs more frequently with the mono-layer Mono APET 200µ/52µ and the paper compound paper-PE/62µ. This growth behavior can be explained by the relatively large number of packages with increased O_2 content caused by defect packages and also changed barrier properties (differences between specified and achieved application-related). In turn, the growth of yeasts suppresses the growth of LAB. Nielsen et al. (2008) figured out that yeasts do not constitute a spoilage problem in processed, vacuum-packed meat products. But several isolates have the ability and potential to spoil meat products also under a low O_2/high CO_2 atmosphere (0.5 % O_2, 20 % CO_2, 79.5 % N_2) (Nielsen et al., 2008). Especially in MA- and vacuum-packaging, air leakages lead to a loss of functionality and affect the growth of spoilage mold, causing a marked deterioration of the product and leading to rejection of the product by the consumer before the end of shelf life (Koruk & Sanliturk, 2019; Zardetto et al., 2022). Eilamo et al. (1995) reported on minced meat steaks that the size of the leak affects the growth of yeasts and molds. These findings in combination with the present study show that sufficient barrier properties and mechanical resistance are key factors to food quality and that it is very complicated to replace an extremely effective EVOH barrier with just a mono-material. However, Pettersen et al. (2020) showed that: a competitive quality and shelf life can be obtained by packaging chicken fillets in recyclable mono-layer materials (high-density polyethylene (HDPE)) instead of complex multi-layered materials (APET/PE) to achieve more sustainable packaging systems. This points out that the use of more sustainable packaging materials depends on the product itself and its requirements, on the packaging characteristics as well as on the packaging process.

Typically, a specific spoilage organism is needed and selected to determine the most accurate microbial shelf life of fresh meat products (Nychas et al., 2008). As the pattern of microbial growth is not consistent for the different packaging materials, in this study, the suitability of microbial growth as a parameter to evaluate the shelf life, in general, of MA-packed emulsion-type sausage is limited. However, a trend is visible, and due to the analyzed heterogeneous microflora, the TVC shows a promising approach. Based on the modeled growth curve of TVC for the emulsion-type sausage Mortadella, the control and, therefore, currently used multi-layer along with the multi-layer APET/PE 230µ/62µ show the best preservation characteristics by suppressing microbial growth and keeping product quality during storage indicating intact MA-conditions. This indicates that sufficient barrier properties of both under and upper foil are crucial to keep the quality of emulsion-type sausage. The multi-layer APET/PE 250µ/47µ and the mono-layer Mono APET 200µ/52µ represent promising alternatives reaching the value of 6.7 $\log_{10}$ cfu/g after 26 days of storage. For all other investigated packaging materials, reducing packaging material would have the opposite effect on food waste and makes the whole product less sustainable. However, since the shelf life given by the manufacturer is 28 days, investigations are required to determine if a shelf life shortened by even two days will increase food waste depending on logistic aspects, sales, and consumption rates.

Besides microbial spoilage, biochemical changes lead to quality loss. Lipid and protein oxidation as well as oxidation and degradation of pigments by microorganisms, cause a decrease in color saturation and intensity during storage (Jin et al., 2021). For the consumer, the appearance of meat and meat products is crucial when evaluating its suitability for consumption and quality. Slight color changes and deviations quickly lead to rejections causing substantial losses of a valuable and safe nutrient source and thereby generating food waste (Faustman et al., 2023). According to Khorsandi et al. (2019), L* values for emulsion-type sausage are in a range of 53.1 - 57.4, whereas Ruiz et al. (2009) stated L* values for cured sausage and ham ranging from 50.0 - 64.0. The values measured in this study are slightly higher. The color measurements showed higher variations between the emulsion-type sausages packed in the different packaging materials (different batches) than during the storage period. However, as muscles are diverse in color, determining of a typical color range for muscle food products is challenging (Sheridan et al., 2007). As the O_2 concentration inside the packaging is responsible, and even essential, for color changes like fading of cured, cut, and packaged meat products, the results show that even in defective packages, the O_2 concentration is not sufficiently high for color changes leading to fading of the packaged product. This can be caused by certain preservatives, according to the supplier-given recipe. However, packaging materials as well as the gaseous atmosphere in MAP are seen as critical factors influencing the color of red meat during its shelf life (Panseri et al., 2018).

In terms of the sustainability of packaging materials, several analyses of food supply chains showed that reducing packaging materials is crucial, but it must still fulfill its duty of protection. Otherwise, the supply chain, overall, will be less sustainable (Heller et al., 2019; Pauer et al., 2020; Verghese et al., 2015). For a packaging system, it is always important to find a balance between the environmental impact of the package itself, on the one hand, and the impact originating from the potential loss of the packaged product, on the other (Scherhaufer et al., 2018; Williams & Wikström, 2011). Kan & Miller (2022) analyzed several food LCA studies to quantify the impact of packaging in relation to the impact of the food throughout the life cycle. Plastic packaging accounted for less than 10 % of LCA-emissions for more than 75 % of the food items surveyed. Conte et al. (2015) stated that multi-layer surpass single-layer materials regarding the environmental impact when food waste is included in the system boundaries. Based on the results of several LCA studies, minimization of material, whilst retaining mechanical and barrier properties of flexible packaging, should be clearly prioritized over recyclability improvements (Barlow & Morgan, 2013). When considering the pure material properties, the top priority in the (eco)design of meat packaging is food loss prevention instead of minimization of the packaging impacts itself. Additionally, light-weighting should be a priority above circularity improvements (Pauer et al., 2020).

4.6 Conclusion

Since the use of a mono-layer Mono APET 200µ/52µ without any EVOH layer, an under foil reduced in thickness to 230µ and a paper-PE compound lead to an increased O_2 concentration, the material's applicability, and ability to protect the gaseous atmosphere must be considered when using sustainable materials. The control alongside the multi-layer APET/PE 230µ/62µ show the best preservation characteristics by suppressing microbial growth. This indicates that sufficient barrier properties of both under and upper foil are crucial in keeping the quality of emulsion-type sausage during storage. The multi-layer APET/PE 250µ/47µ and the mono-layer Mono APET 200µ/52µ represent promising alternatives, whereas the paper compound paper-PE/62µ was not effective in keeping the quality of emulsion-type sausage approximately over the practical shelf life. In brief, when implementing new packaging materials for perishable products, it is crucial to ensure sufficient barrier properties along an appropriate adjusted combination of sealing settings. Further studies need to focus on optimal sealing settings to reduce the number of defect packages to a minimum. However, although the products were subject to real transport conditions while they were being transported from the production site to the laboratory across Germany, in a next step transport studies are recommended to evaluate the robustness of these studied packaging materials against rigors during transport and handling in a commercial supply chain setup. Finally, introducing new high-performance materials remains highly case-dependent, but represents a suitable replacement for conventionally used multi-layer packaging, as long as food quality is preserved and thereby food waste is prevented. Especially in certain product areas where short residual terms are enough the use of new packaging materials can be favored even in terms of sustainability.

Funding

This research was funded by the European Union via the European Regional Development Fund EFRE.NRW (Grant EFRE 0500035, "Biobasierte Produkte").

Acknowledgement

The authors wish to thank Lucas Correa Dresch for the performed final English revision of the manuscript.

References

Alipour, N., Gedde, U. W., Hedenqvist, M. S., Yu, S., Roth, S., Brüning, K., Vieyres, A., & Schneider, K. (2015). Structure and properties of polyethylene-based and EVOH-based multilayered films with layer thicknesses of 150nm and greater. *European Polymer Journal, 64*, 36–51. https://doi.org/10.1016/j.eurpolymj.2014.12.011.

Barlow, C. Y., & Morgan, D. C. (2013). Polymer film packaging for food: An environmental assessment. *Resources, Conservation and Recycling*, *78*, 74–80. https://doi.org/10.1016/j.resconrec.2013.07.003.

Barukčić, I., Ščetar, M., Marasović, I., Lisak Jakopović, K., Galić, K., & Božanić, R. (2020). Evaluation of quality parameters and shelf life of fresh cheese packed under modified atmosphere. *Journal of Food Science and Technology*, *57*(7), 2722–2731. https://doi.org/10.1007/s13197-020-04308-6.

Bekhit, A. E.-D. A., Holman, B. W., Giteru, S. G., & Hopkins, D. L. (2021). Total volatile basic nitrogen (TVB-N) and its role in meat spoilage: A review. *Trends in Food Science & Technology*, *109*, 280–302. https://doi.org/10.1016/j.tifs.2021.01.006.

Blanchard, A., Gouanvé, F., & Espuche, E. (2017). Effect of humidity on mechani-cal, thermal and barrier properties of EVOH films. *Journal of Membrane Science*, 540, 1–9. https://doi.org/10.1016/j.memsci.2017.06.031.

Cabedo, L., Lagarón, J. M., Cava, D., Saura, J. J., & Giménez, E. (2006). The effect of ethylene content on the interaction between ethylene-vinyl alcohol copolymers and water—II: Influence of water sorption on the mechanical properties of EVOH copolymers. *Polymer Testing*, *25*(7), 860–867. https://doi.org/10.1016/j.polymertesting.2006.04.012.

Chen, X., Zhao, J., Zhu, L., Luo, X., Mao, Y., Hopkins, D. L., Zhang, Y., & Dong, P. (2020). Effect of modified atmosphere packaging on shelf life and bacterial community of roast duck meat. *Food Research International (Ottawa, Ont.)*, *137*, 109645. https://doi.org/10.1016/j.foodres.2020.109645.

Chung, D., Papadakis, S. E., & Yam, K. L. (2003). Simple models for evaluating effects of small leaks on the gas barrier properties of food packages. *Packaging Technology and Science*, *16*(2), 77–86. https://doi.org/10.1002/pts.616.

Comi, G. (2016). Chapter 8 - Spoilage of Meat and Fish. In A. Bevilacqua, M. R. Corbo, & M. Sinigaglia (Eds.), *Woodhead Publishing series in food science, technology and nutrition. The microbiological quality of food: Foodborne spoilers* (pp. 179–210). Woodhead Publishing is an imprint of Elsevier. https://doi.org/10.1016/B978-0-08-100502-6.00011-X.

Conte, A., Cappelletti, G. M., Nicoletti, G. M., Russo, C., & Del Nobile, M. A. (2015). Environmental implications of food loss probability in packaging design. *Food Research International*, *78*, 11–17. https://doi.org/10.1016/j.foodres.2015.11.015.

Eilamo, M., Ahvenainen, R., Hurme, E., Heiniö, R.-L., & Mattila-Sandholm, T. (1995). The effect of package leakage on the shelf-life of modified atmosphere packed minced meat steaks and its detection. *LWT - Food Science and Technology*, *28*(1), 62–71. https://doi.org/10.1016/S0023-6438(95)80014-X.

Ellen MacArthur Foundation and McKinsey & Company (2016). The New Plastics Economy: Rethinking the future of plastics. Word Economic Forum. http://www.ellenmacarthurfoundation.org/publications.

European Commission (2018). A European Strategy for Plastics in a Circular Economy.

European Commission (2020). A new Circular Economy Action Plan: For a cleaner and more competitive Europe. https://eur-lex.europa.eu/resource.html?uri=cellar:9903b325-6388-11ea-b735-01aa75ed71a1.0017.02/DOC_1&format=PDF.

Faraca, G., & Astrup, T. (2019). Plastic waste from recycling centres: Characterisation and evaluation of plastic recyclability. *Waste Management*, *95*, 388–398. https://doi.org/10.1016/j.wasman.2019.06.038.

Faustman, C., Suman, S. P., & Ramanathan, R. (2023). Chapter 11 - The eating quality of meat: I Color. In F. Toldra (Ed.), *Woodhead Publishing Series in Food Science, Technology and Nutrition Ser. Lawrie's Meat Science* (9th ed., pp. 363–392). Elsevier Science & Technology. https://doi.org/10.1016/B978-0-323-85408-5.00023-6.

Gavara, R., Catalá, R., López Carballo, G., Cerisuelo, J. P., Dominguez, I., Muriel-Galet, V., & Hernandez-Muñoz, P. (2016). Use of EVOH for Food Packaging Applications. In *Reference Module in Food Science.* Elsevier. https://doi.org/10.1016/B978-0-08-100596-5.21125-6.

Hauptmann, M., Bär, W., Schmidtchen, L., Bunk, N., Abegglen, D., Vishtal, A., & Wyser, Y. (2021). The sealing behavior of new mono-polyolefin and paper-based film laminates in the context of bag form-fill-seal machines. *Packaging Technology and Science*, *34*(2), 117–126. https://doi.org/10.1002/pts.2544.

Heller, M. C., Selke, S. E. M., & Keoleian, G. A. (2019). Mapping the Influence of Food Waste in Food Packaging Environmental Performance Assessments. *Journal of Industrial Ecology*, *23*(2), 480–495. https://doi.org/10.1111/jiec.12743.

Herbert, U., Rossaint, S., Khanna, M.-A., & Kreyenschmidt, J. (2013). Comparison of argon-based and nitrogen-based modified atmosphere packaging on bacterial growth and product quality of chicken breast fillets. *Poultry Science*, *92*(5), 1348–1356. https://doi.org/10.3382/ps.2012-02590.

Jin, S.-K., Choi, J.-S., & Kim, G.-D. (2021). Effect of porcine plasma hydrolysate on physicochemical, antioxidant, and antimicrobial properties of emulsion-type pork sausage during cold storage. *Meat Science*, *171*, 108293. https://doi.org/10.1016/j.meatsci.2020.108293.

Kan, M., & Miller, S. A. (2022). Environmental impacts of plastic packaging of food products. *Resources, Conservation and Recycling*, *180*, 106156. https://doi.org/10.1016/j.resconrec.2022.106156.

Kargwal, R., Garg, M. K., Singh, V. K., Garg, R., & Kumar, N. (2020). Principles of modified atmosphere packaging for shelf life extension of fruits and vegetables: An overview of storage conditions. *International Journal of Chemical Studies*, *8*(3), 2245–2252. https://doi.org/10.22271/chemi.2020.v8.i3af.9545.

Khorsandi, A., Eskandari, M. H., Aminlari, M., Shekarforoush, S. S., & Golmakani, M. T. (2019). Shelf-life extension of vacuum packed emulsion-type sausage using combination of natural antimicrobials. *Food Control*, *104*, 139–146. https://doi.org/10.1016/j.foodcont.2019.04.040.

Koruk, H., & Sanliturk, K. Y. (2019). Detection of air leakage into vacuum packages using acoustic measurements and estimation of defect size. *Mechanical Systems and Signal Processing*, *114*, 528–538. https://doi.org/10.1016/j.ymssp.2018.05.023.

Kreyenschmidt, J., Albrecht, A., Braun, C., Herbert, U., Mack, M., Rossaint, S., Ritter, G., Teitscheid, P., & Ilg, Y. (2013). Food Waste in der Fleisch verarbeitenden Kette. *Fleischwirtschaft*, *10*, 57–63.

López-Rubio, A. (2011). Ethylene-vinyl alcohol (EVOH) copolymers. In *Multifunctional and Nanoreinforced Polymers for Food Packaging* (pp. 261–284). Elsevier. https://doi.org/10.1533/9780857092786.1.261.

Matthews, C., Moran, F., & Jaiswal, A. K. (2021). A Review on European Union's Strategy for Plastics in a Circular Economy and its Impact on Food Safety. *Journal of Cleaner Production*, *283*, Article 125263.

Mokwena, K. K., Tang, J., & Laborie, M.-P. (2011). Water absorption and oxygen barrier characteristics of ethylene vinyl alcohol films. *Journal of Food Engineering*, *105*(3), 436–443. https://doi.org/10.1016/j.jfoodeng.2011.02.040.

Molina-Besch, K., Wikström, F., & Williams, H. (2019). The environmental impact of packaging in food supply chains—does life cycle assessment of food provide the full picture? *The International Journal of Life Cycle Assessment*, *24*(1), 37–50. https://doi.org/10.1007/s11367-018-1500-6.

Nielsen, D. S., Jacobsen, T., Jespersen, L., Koch, A. G., & Arneborg, N. (2008). Occurrence and growth of yeasts in processed meat products - Implications for potential spoilage. *Meat Science*, *80*(3), 919–926. https://doi.org/10.1016/j.meatsci.2008.04.011.

Nychas, G.-J. E., Skandamis, P. N., Tassou, C. C., & Koutsoumanis, K. P. (2008). Meat spoilage during distribution. *Meat Science*, *78*(1-2), 77–89. https://doi.org/10.1016/j.meatsci.2007.06.020.

Panseri, S., Martino, P. A., Cagnardi, P., Celano, G., Tedesco, D., Castrica, M., Balzaretti, C., & Chiesa, L. M. (2018). Feasibility of biodegradable based packaging used for red meat storage during shelf-life: A pilot study. *Food Chemistry*, *249*, 22–29. https://doi.org/10.1016/j.foodchem.2017.12.067.

Pauer, E., Tacker, M., Gabriel, V., & Krauter, V. (2020). Sustainability of flexible multilayer packaging: Environmental impacts and recyclability of packaging for bacon in block. *Cleaner Environmental Systems*, *1*, Article 100001. https://doi.org/10.1016/j.cesys.2020.100001.

Pettersen, M. K., Grøvlen, M. S., Evje, N., & Radusin, T. (2020). Recyclable mono materials for packaging of fresh chicken fillets: New design for recycling in circular economy. *Packaging Technology and Science*, *33*(11), 485–498. https://doi.org/10.1002/pts.2527.

Pro Carton (2010). Wie wichtig ist nachhaltige Verpackung? Die Einstellung von Konsumenten zu Verpackung und Nachhaltigkeit. Association of European Cartonboard and Carton Manufacturers. www.procarton.com.

Ragaert, K., Delva, L., & van Geem, K. (2017). Mechanical and chemical recycling of solid plastic waste. *Waste Management (New York, N.Y.)*, *69*, 24–58. https://doi.org/10.1016/j.wasman.2017.07.044.

Rossaint, S., Klausmann, S., Herbert, U., & Kreyenschmidt, J. (2014). Effect of package perforation on the spoilage process of poultry stored under different modified atmospheres. *Food Packaging and Shelf Life*, *1*(1), 68–76. https://doi.org/10.1016/j.fpsl.2014.01.002.

Ruiz, A., Williams, S. K., Djeri, N., Hinton, A., & Rodrick, G. E. (2009). Nisin, rosemary, and ethylenediaminetetraacetic acid affect the growth of Listeria monocytogenes on ready-to-eat turkey ham stored at four degrees Celsius for sixty-three days. *Poultry Science*, *88*(8), 1765–1772. https://doi.org/10.3382/ps.2008-00521.

Scherhaufer, S., Moates, G., Hartikainen, H., Waldron, K., & Obersteiner, G. (2018). Environmental impacts of food waste in Europe. *Waste Management*, *77*, 98–113. https://doi.org/10.1016/j.wasman.2018.04.038.

Schumann, B., & Schmid, M. (2018). Packaging concepts for fresh and processed meat – Recent progresses. *Innovative Food Science & Emerging Technologies*, *47*, 88–100. https://doi.org/10.1016/j.ifset.2018.02.005.

Seier, M., Archodoulaki, V.-M., Koch, T., Duscher, B., & Gahleitner, M. (2022). Polyethylene terephthalate based multilayer food packaging: Deterioration effects during mechanical recycling. *Food Packaging and Shelf Life*, *33*, 100890. https://doi.org/10.1016/j.fpsl.2022.100890.

Sharma, R., & Ghoshal, G. (2018). Emerging trends in food packaging. *Nutrition & Food Science*, *48*(5), 764–779. https://doi.org/10.1108/NFS-02-2018-0051.

Sheridan, C., O'Farrell, M., Lewis, E., Flanagan, C., Kerry, J., & Jackman, N. (2007). A comparison of CIE L*a*b* and spectral methods for the analysis of fading in sliced cured ham. *Journal of Optics a: Pure and Applied Optics*, *9*(6), S32-S39. https://doi.org/10.1088/1464-4258/9/6/S06.

Soro, A. B., Noore, S., Hannon, S., Whyte, P., Bolton, D. J., O'Donnell, C., & Tiwari, B. K. (2021). Current sustainable solutions for extending the shelf life of meat and marine products in the packaging process. *Food Packaging and Shelf Life*, *29*, 100722. https://doi.org/10.1016/j.fpsl.2021.100722.

Teck Kim, Y., Min, B., & Won Kim, K. (2014). General Characteristics of Packaging Materials for Food System. In *Innovations in Food Packaging* (pp. 13–35). Elsevier. https://doi.org/10.1016/B978-0-12-394601-0.00002-3.

United Nations (2020). The Sustainable Development Goals Report 2020. United States of America. https://unstats.un.org/sdgs/report/2020/The-Sustainable-Development-Goals-Report-2020.pdf.

Verghese, K., Lewis, H., Lockrey, S., & Williams, H. (2015). Packaging's Role in Minimizing Food Loss and Waste Across the Supply Chain. *Packaging Technology and Science*, *28*(7), 603–620. https://doi.org/10.1002/pts.2127.

Walker, T. W., Frelka, N., Shen, Z., Chew, A. K., Banick, J., Grey, S., Kim, M. S., Dumesic, J. A., van Lehn, R. C., & Huber, G. W. (2020). Recycling of multilayer plastic packaging materials by solvent-targeted recovery and precipitation. *Science Advances*, *6*(47). https://doi.org/10.1126/sciadv.aba7599.

Wikström, F., Verghese, K., Auras, R., Olsson, A., Williams, H., Wever, R., Grönman, K., Kvalvåg Pettersen, M., Møller, H., & Soukka, R. (2019). Packaging Strategies That Save Food: A Research Agenda for 2030. *Journal of Industrial Ecology*, *23*(3), 532–540. https://doi.org/10.1111/jiec.12769.

Williams, H., & Wikström, F. (2011). Environmental impact of packaging and food losses in a life cycle perspective: a comparative analysis of five food items. *Journal of Cleaner Production*, *19*(1), 43–48. https://doi.org/10.1016/j.jclepro.2010.08.008.

Yildirim, S., Röcker, B., Pettersen, M. K., Nilsen-Nygaard, J., Ayhan, Z., Rutkaite, R., Radusin, T., Suminska, P., Marcos, B., & Coma, V. (2018). Active Packaging Applications for Food. *Comprehensive Reviews in Food Science and Food Safety*, *17*(1), 165–199. https://doi.org/10.1111/1541-4337.12322.

Zardetto, S., Fregonese, M., & Pasini, G. (2022). Effects of modified atmospheric packaging configuration on spoilage mould growth in damaged packages of fresh pasta. *Journal of Food Engineering*, *314*, 110760. https://doi.org/10.1016/j.jfoodeng.2021.110760.

5 Antimicrobial activity of different coatings for packaging materials containing functional extenders against selected microorganisms typical for food

Imke Korte[1,*], Michael Petry[2], Judith Kreyenschmidt[1,3]

[1] Rheinische Friedrich Wilhelms-University Bonn, Institute of Animal Sciences, Katzenburgweg 7-9, D-53115 Bonn, Germany

[2] PETRYmade Oberflächentechnik, Merlerstraße 117, D-53340 Meckenheim, Germany

[3] Hochschule Geisenheim University, Department of Fresh Produce Logistics, Von-Lade-Straße 1, D-65366 Geisenheim, Germany

* Correspondence

Korte, I., Petry, M., & Kreyenschmidt, J. (2023). Antimicrobial activity of different coatings for packaging materials containing functional extenders against selected microorganisms typical for food. *Food Control*, *148*, 109669. https://doi.org/10.1016/j.foodcont.2023.109669.

5.1 Abstract

The antimicrobial activity of different coatings for packaging materials consisting of a mineral binder based on liquid potassium silicate, non-conserved acrylates, and non-conserved styrene-acrylates as binders using functional, active extenders was investigated. The influence of time, temperature, and product factors on the antimicrobial activity against different bacteria were determined (ISO 22196:2007).

Results showed a significant reduction in bacterial counts on all tested surface coatings compared to the reference, a glass plate without any coating, (24 h, 35°C). Bacterial counts of *Staphylococcus aureus*, *Escherichia coli*, and *Pseudomonas fluorescens* were reduced between 3.32 (M8 against *Staphylococcus aureus)* and 7.01 $\log_{10}$ units (M1 against *Pseudomonas fluorescens*). A high initial bacteria concentration (10^8 cfu/ml) leads to a bacterial reduction between 1.21 (M8 against *Staphylococcus aureus)* and 7.57 $\log_{10}$ units (M4 against *Pseudomonas fluorescens*). Incubation at a low temperature (7°C) for prolonged storage (120 h) resulted in a reduced reduction rate for *Staphylococcus aureus* < 2.00 $\log_{10}$ units. For most coatings, antimicrobial activity was shown in the presence of several food components as well as different pH values.

The studied coatings show a high antimicrobial activity under different test conditions typical for perishable food supply chains and bear a high potential as a coating for packaging materials to increase the safety and quality of perishable products (in chilled storage).

5.2 Introduction

Generally, packaging materials can be equipped with active agents by direct incorporation or by coating and impregnating (existing) packaging materials depending on the physicochemical possibilities of the components involved (material, antimicrobial agent, technology) (Appendini & Hotchkiss, 2002; Bulitta et al., 2020; Cooksey, 2005; Han, 2003). Coatings are used to protect surfaces and to improve their barrier properties such as water vapor transmission rate. Organic coatings are well known for their protective function of metallic structures; binders, co-binders, extenders, pigments, additives, and solvents, for example, are typical components of organic coatings. The binder is an essential ingredient of the coating composition providing mechanical strength next to its film-forming properties. Well known and studied protective organic coatings are based on oligomeric and polymeric polyurethane, acrylic, epoxy, and polysiloxane-binders (Krauklis & Echtermeyer, 2018; Perrin et al., 2009; Raeissi et al., 2021; Zubielewicz & Królikowska, 2009). For example, acrylic polymers are widespread as coatings, consolidants, or adhesives in art conservation because of their reversibility, long-term stability, easy applicability, good adhesion to the substrate, and hydrophobicity (Kovács et al., 2021). Potassium silicate, sodium silicate, lithium silicate, and phosphate are often used as inorganic binders, ensuring a well-balance of functionality and durability (Chen et al., 2022).

Active packaging are materials that are intended to extend the shelf life, or to maintain or improve the condition of packaged food. Based on the European Union Guidance, these are designed to deliberately incorporate components that would release or absorb substances into or from the packaged food or the environment surrounding the food (Commission Regulation (EC) No 450/2009). One group in the field of active packaging is antimicrobial systems. Antimicrobial materials aim to control or reduce microbial growth, resulting in the extension of the lag phase, or a reduced growth rate in the exponential phase, or even killing them during food storage (Bulitta et al., 2020; Coma, 2008; Dohlen et al., 2016; Lavoine et al., 2014). Depending on the mode of action, the resulting antimicrobial systems are classified as time-release killing or contact-active materials (Adlhart et al., 2018; Bulitta et al., 2020; Han, 2003). The contact-active killing mechanism means that a non-volatile antimicrobial agent is permanently immobilized and attached to a polymer backbone, it is part of the polymer, or the whole macromolecule is antimicrobial on its own (Appendini & Hotchkiss, 2002; Han, 2003; Siedenbiedel & Tiller, 2012). A possible mode of action could be the interaction between the positively charged polymers and negatively charged microbial cell membranes resulting in the leakage of intracellular constituents as well as the denaturation of structural proteins and enzymes in the membrane core (Bulitta et al., 2020; Dehnad et al., 2014; Dutta et al., 2009; Shahidi et al., 1999). Inherently, antimicrobial polymers are chitosan as a cationic polymer, poly(L-lysine), calcium alginate, acrylic polymers, and sustainable active microbiocidal (SAM) polymers (Appendini & Hotchkiss, 2002; Dohlen et al., 2016; Ilg & Kreyenschmidt, 2012).

In the field of active ingredients, the application of metal oxides as antimicrobials instead of organic agents is getting more and more attention. Depending on environmental conditions, the advantages of inorganic antibacterial agents are improved, in terms of safety and stability, when compared with organic antimicrobial materials (AL-Jawad et al., 2017; Yu et al., 2011). In general, simple metal oxide powders (zinc oxide (ZnO), calcium oxide (CaO), magnesium oxide (MgO), and titanium dioxide (TiO_2)) can exhibit significant antimicrobial activities against several microorganisms (Bulitta et al., 2020; Dyshlyuk et al., 2020; Gupta et al., 2013; Jung et al., 2017; Qu et al., 2019; Suresh et al., 2018; Yamamoto, 2001; Yao et al., 2007). The advantages of these oxides are that they contain elements abundant in nature, are essential for humans, and provide high antimicrobial activity in small amounts in their pure and unbound form (Dyshlyuk et al., 2020). However, the modes of action are partly different. Particularly TiO_2, often added as a white pigment, is well known, and associated with a highly efficient photocatalytic activity (induced by visible light, near-UV, or UV) and the resulting formation of free radicals or the generation of high-energy surfaces, high refractive index, chemical stability, and low cost (Arellano et al., 2011; Beyth et al., 2015; Bulitta et al., 2020; Ranganayaki et al., 2014).

Despite these advantages, there are health concerns related to the use of nanoparticles and TiO_2 has recently been reviewed to pose a health risk. The European Framework Regulation (EC) No. 1935/2004 applies to all food contact materials and specifies that materials and objects must be manufactured in such a way that they do not release any components onto food in large quantities that endangers human health or leads to unacceptable changes in the composition of food or impairs the organoleptic properties of food (Regulation (EC) No 1935/2004). However, TiO_2 is approved for use in plastic food contact materials in accordance with Regulation (EU) No. 10/2011 (Commission Regulation (EU) No 10/2011). Only the use of TiO_2 as nanomaterial is prohibited in this context (Bundesinstitut für Risikobewertung, 2021; Commission Regulation (EU) No 10/2011).

It has been shown that a variety of antimicrobial agents applied in packaging materials are effective against various microorganisms under standardized laboratory conditions. However, it is still challenging to develop adequate antimicrobial materials that are in contact with food (Balasubramanian et al., 2009; Dohlen et al., 2017). As stated by several authors, the antimicrobial action of different agents is often (negatively) influenced by various factors, such as cold temperature conditions typical in meat supply chains or the presence of proteins (Asharani et al., 2009; Dohlen et al., 2016; Ilg & Kreyenschmidt, 2011; Kampmann et al., 2008; Lee et al., 2011; Martínez-Abad et al., 2012; Russell & Hugo, 1994).

Due to the surface-active properties of minerals, the use of coatings either based on a mineral binder or using a mineral mixture as a functional extender bears the potential for application as a coating in antimicrobial packaging to increase food safety and shelf life. Up to now, it is not clear if such coatings are active over the broad spectrum of environmental and product factors existing in the food industry.

Thus, the aim of this study was to investigate the antimicrobial activity of several novel surface coatings differing on the binder. The antimicrobial activity of a coating using a mineral binder based on liquid potassium silicate in combination with a new class of biopolymer from renewable resources, a mixture of specially processed fatty acids and oleic acids, as well as natural resins, was determined. Furthermore, non-conserved acrylates, and non-conserved styrene-acrylates using a functional extender, a mineral mixture based on calcium-aluminum hydroxide ($Ca_3Al_2(OH)_{12}$), were evaluated for their antimicrobial activity and potential use as coating of packaging materials and food contact surfaces. Therefore, the influence of several environmental and product factors typically occurring in meat supply chains on the antimicrobial activity against various pathogenic (*Staphylococcus aureus*, *Escherichia coli*) and spoilage bacteria (*Pseudomonas fluorescens*) is investigated.

5.3 Materials and methods

5.3.1 Coating preparation/characterization

The test samples were prepared by coating glass plates with different binder formulations and classified into three groups depending on the used raw material base:

- Mixture of a mineral binder based on liquid potassium silicate in water (binder) and a new biopolymer based on renewable resources (co-polymer);
- Mixture of non-conserved acrylic polymers (binder) and a new functional extender;
- Mixture of non-conserved styrene-acrylic polymers (binder) and a new functional extender.

The used biopolymer based on renewable resources in the first group is a mixture of fatty acids, oleic acids as well as natural resins, which are processed in a thermal process in such a way that they emulsify and can be dissolved in the mineral binder based on liquid potassium silicate ($K_2Si_2O_5$) (Figure 5.1(a)) and water. The mixture of fatty acid is represented by a formulation of linoleic acid ($C_{17}H_{31}COOH$), linolenic acid ($C_{18}H_{30}O_2$), and paraffin (CnH_2n+2, with n approximately between 18 and 32). Paraffin is slightly liquid, oily, or waxy.

The used functional extender in the second and third groups consists of a mixture of minerals based on $Ca_3Al_2(OH)_{12}$. As a complex structure with different cations and anions with and without the addition of TiO_2 as a white pigment. Within these groups, samples with various amounts of binder and biopolymer or functional extender were prepared. In total, eight different coatings were used for the analyses of antimicrobial activity. The detailed composition of the different surface coatings is given in Table 5.1.

Table 5.1 Components and detailed composition of mineral binder based on liquid potassium silicate, acrylic polymer, and styrene-acrylic polymer coatings with percentage composition of solvent, binder, and biopolymer/functional extender.

Sample Code	Components	Solvent (water) [%]	Composition of functional active agents [%]	
			Binder	**Biopolymer/functional extender**
Mineral binder based on liquid potassium silicate				
M1	Mineral binder and biopolymer (transparent)	55	40	5
M4	Mineral binder and biopolymer with mixture of white standard extender	40	50	10
Acrylic polymer				
M2	Acrylic polymer and functional extender with titanium dioxide	58	17	25
M5	Acrylic polymer and functional extender with titanium dioxide	66	14	20
M6	Acrylic polymer and functional extender without titanium dioxide	75	12	13
M8	Dispersion of functional extender in acrylic polymer (transparent)	79.9	17	3.1
Styrene-acrylic polymer				
M3	Styrene-acrylic polymer and functional extender with titanium dioxide	67.9	17	15.1
Pure functional extender				
M7	Suspension of functional extender in water	50	0	50

The chemical raw material basis for the manufacturing of the acrylic polymer is methyl methacrylate ($C_5H_8O_2$) (Figure 5.1(b)) and 2-ethylhexyl acrylate ($C_{11}H_{30}O_2$) (Figure 5.1(c)), produced by companies such as BASF, Alberding & Boley as well as Imparat, and offered to the market as basic raw materials.

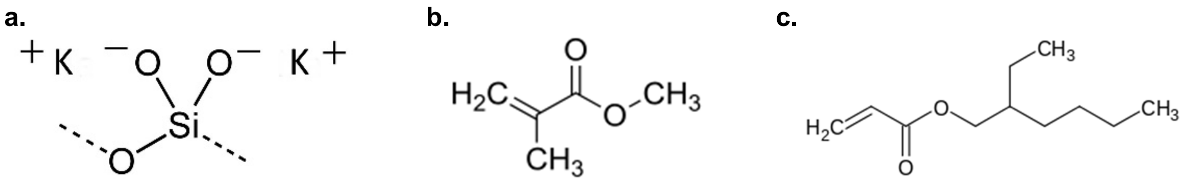


Figure 5.1 Overview of chemical structure of binders used for the coatings: (a) potassium silicate ($K_2Si_2O_5$), (b) methyl methacrylate ($C_5H_8O_2$), and (d) 2-ethylhexyl acrylate ($C_{11}H_{30}O_2$).

Pure acrylic polymer is characterized by the smallest acrylic resin particles that bind the acrylic paint; they are dissolved in water that evaporates completely during the drying process, whereby the binder and the color pigments solidify into an elastic film. The styrene-acrylic polymer is characterized by the smallest acrylic resin particles that bind the acrylic paint as well. They are dissolved in water that evaporates completely during the drying process, whereby the binder and the color pigments solidify into a firm film. Both processes are patent-pending due to the novelty of being able to dispense with classic preservatives and biocides in aqueous binders.

5.3.2 Experimental design

The antimicrobial activity of different surface coatings was tested for the potential to serve as an antimicrobial coating on packaging materials and surfaces against selected gram-positive and gram-negative bacteria under various food supply chain conditions. Square glass plates of 40 mm x 40 mm without any coating were used as references. The antimicrobial activity of the surface coatings was analyzed according to ISO 22196:2007. The standard is based on a comparison of bacteria counts in saline solution on reference and sample materials after defined storage conditions. The standard was modified in order to test conditions typical for food products, as described in detail by Dohlen et al. (2016).

This study was subdivided into four parts. In the first part, the general antimicrobial activity of all eight surface coatings was investigated against *Staphylococcus aureus*, *Escherichia coli*, and *Pseudomonas fluorescens* under standard (24 h, 35°C) and cold temperature conditions (120 h, 7°C) typical for applications in the food industry for perishable products. The second part included the determination of the influence of a high initial bacteria concentration of 10^8 cfu/ml after 24 h at 35°C. In the third part, the influence of pH 5, 7, and 9 on the antimicrobial activity (24 h, 35°C) was analyzed. The fourth part included the determination of the influence of starch, meat extract, and oleic acid as food components for selected coatings (M1, M5, M6, M7, M8). Table 5.2 gives an overview of the different conducted experiments.

Table 5.2 Test conditions (time, temperature, high initial bacteria concentration, pH values, and added food components) to investigate the antimicrobial activity of functional active agents of a mineral binder based on liquid potassium silicate, acrylic polymer, and styrene-acrylic polymer samples against different bacteria.

Test conditions		24 h, 35°C	120 h, 7°C	24 h, 35°C						
		10^5 cfu/ml		10^8 cfu/ml	pH 5	pH 7	pH 9	Starch	Meat extract	Oleic acid
Coating	**Mineral binder based on liquid potassium silicate**									
	M1	+	+	+	+	+	+	+	+	+
	M4	+	+	+	+	+	+	-	-	-
	Acrylic polymer									
	M2	+	+	+	+	+	+	-	-	-
	M5	+	+	+	+	+	+	+	+	+
	M6	+	+	+	+	+	+	+	+	+
	M8	+	+	+	+	+	+	+	+	+
	Styrene-acrylic polymer									
	M3	+	+	+	+	+	+	-	-	-
	Pure functional extender									
	M7	+	+	+	+	+	+	+	+	+

+ Tested against *Staph. aureus*, *E. coli*, and *Ps. fluorescens*; - not tested.

5.3.3 Bacterial cultures

As test organisms, *Staphylococcus aureus* (Strain DSM No. 799), *Escherichia coli* (Strain DSM No. 1576), and *Pseudomonas fluorescens* (Strain DSM No. 50090) were chosen, delivered by the German Collection of Microorganisms and Cell Cultures (DSMZ, Braunschweig, Germany), and were frozen in cryogenic pellets for preservation. Frozen cultures (-18°C) were spread onto plate count agar. The inoculated agar plates were incubated at optimal growth time and temperature for each microorganism according to the instructions of the DSMZ (Table 5.3).

Table 5.3 Overview of the used strains, cultivation and enumeration temperature and growth time.

Bacteria	Strain DSM No.	Cultivation and enumeration temperature [°C]	Cultivation and enumeration growth time [h]
Staphylococcus aureus	799	37	24
Escherichia coli	1576	37	24
Pseudomonas fluorescens	50 090	25	48

5.3.4 Preparation of inoculum

At the beginning of each experimental series, the bacterial solutions for the inoculation of the samples and references were prepared by adjusting the bacterial concentration in the inoculation suspension via photometric measurement. Microorganisms from the incubated agar plates were transferred into a physiological saline solution (0.85 %) (Oxoid, Hampshire, UK) with 0.1 % tryptone (Oxoid, Hampshire, UK). McFarland BBS standard (Roth, Karlsruhe, Germany) was used as the standard for turbidity to adjust the required reproducible cell density of the inoculation suspension. Therefore, using a spectral photometer, the value of the turbidity standard of 0.5 McFarland units was used as it matches the cell density of 1.5 x 10^8 cfu/ml.

The antimicrobial activity under standard and cold temperature conditions (part 1 of experiments) was tested by diluting the inoculum of the pure bacteria in physiological saline solution up to a concentration of 10^5 - 10^6 cfu/ml. To test the influence of a high initial bacteria concentration (part 2 of experiments) on the activity after 24 h at 35°C undiluted bacteria suspension with an adjusted bacteria concentration of 10^8 cfu/ml was used for inoculation. Furthermore, the effect of a range of different pH values (5, 7, and 9) (part 3 of experiments) was investigated. For this test series, Sorenson's buffer was prepared from potassium dihydrogen orthophosphate (VWR, Darmstadt, Germany) and disodium hydrogen phosphate dihydrate (VWR, Darmstadt, Germany). The bacteria concentration was adjusted photometric directly in the solution of Sorenson's buffer. This solution was used as test inocula. To test the influence of food components (part 4 of experiments) on the antimicrobial activity, different food components were added to the inocula. Starch and oleic acid could be used in its pure form and was therefore directly added to the inoculm. The amount depends on the specific miscibility of the food components with distilled water. To test the influence of carbohydrates, 1 g of starch (Merck, Darmstadt, Germany) was added to 10 ml of inocula. The effect of proteins was investigated by meat extract (Merck, Darmstadt, Germany).The meat extract was prepared by dissolving a meat extract granulate (Merck, Darmstadt, German) in sterile distilled water. 1 ml of inocula was added to 9 ml of meat extract. Oleic acid (Sigma-Aldrich, St. Louis, USA) was used to test the influence of lipids. 0.1 g oleic acid was added to 10 ml of inocula. To ensure homogeneous dispersion in the inoculum 0.4 ml of tween ® 80 (polysorbate) (Merck, Darmstadt, Germany) was added to the inocula.

5.3.5 Determination of the antimicrobial activity of surface coatings under standard (24 h, 35°C) and cold temperature conditions (120 h, 7°C) typical for applications in the food industry for perishable products (part 1)

For each experiment, a minimum of three samples for each coating (M1 - M8) and six samples for reference were used. The experiments were conducted in triplicate (n=9). Each sample and reference were inoculated with 0.4 ml of the 10^5 - 10^6 cfu/ml adjusted bacteria suspension. To determine the initial concentration at t_0, the bacterial concentration was determined on three references immediately after inoculation. The other three references and samples were stored in a high-precision incubator (Sanyo model MIR 153, Sanyo Electric Co., Ora-Gun, Gumma, Japan) for 24 h at 35°C. To prevent evaporation and to ensure a standardized contact of the material and the bacteria suspension during storage, the inoculum was covered by a sterile polyethylene film (40 mm x 40 mm). To determine the bacterial counts, each reference and each test sample was washed out with 10 ml of soybean-casein digest broth with lecithin polysorbate. The microbial counts of the solutions were analyzed. Colony-forming units (cfu) were determined by using the drop plate technique and pour plate method (only dilution level 0) with plate count agar (Merck, Darmstadt, Germany). Petri dishes were incubated for 24 h for *Staphylococcus aureus* and *Escherichia coli* at 37°C and 48 h for *Pseudomonas fluorescens* at 25°C. Results were expressed as the number of colony-forming units per milliliter. Detection limits were determined to be 1.41 $\log_{10}$ cfu/ml for the pour plate and 2.72 $\log_{10}$ cfu/ml for the drop plate technique. In total, 384 references and samples were analyzed.

Furthermore, the influence of cold temperature conditions typical in meat supply chains (120 h, 7°C) on the antimicrobial activity was tested. The experiments were conducted with samples M1 - M8 in the same way described above, but references and test samples were inoculated with 0.4 ml of 10^5 - 10^6 cfu/ml adjusted bacteria suspension and stored for 120 h at 7°C. Altogether 333 references and samples were analyzed.

5.3.6 Determination of the antimicrobial activity of a high initial bacteria concentration (part 2)

The influence of a high initial bacteria concentration on the antimicrobial activity after 24 h at 35°C was tested for the samples M1 - M8 in the same way described above, but references and test samples were inoculated with 0.4 ml of 10^8 cfu/ml adjusted bacteria suspension. In this part, in total 357, references and samples were analyzed.

5.3.7 Determination of the influence of pH on the antimicrobial activity of different surface coatings (part 3)

The third part of the study included the determination of the influence of pH 5, 7, and 9. Therefore, the bacterial load was adjusted in the solutions of different pH values to a concentration of 10^5 - 10^6 cfu/ml. The references and samples were inoculated for 24 h at 35°C. This part includes the testing of 876 references and samples in total.

5.3.8 Determination of the influence of food components on the antimicrobial activity of different surface coatings (part 4)

The influence of the food components starch, meat extract, and oleic acid was determined in the fourth part. The bacterial load was adjusted in physiological saline solution to a concentration of 10^5 - 10^6 cfu/ml. The references and treated samples were inoculated with different food components and stored for 24 h at 35°C. For the studies with the food components, only the selected surface coating M1, M5, M6, M7, and M8, which were chosen to cover different binders and showed promising results in former experiments, were used. Here altogether 678 references and samples were analyzed.

5.3.9 Data analysis

5.3.9.1 Antimicrobial activity analysis

The number of viable bacteria counts was calculated according to the following equation (5.1):

$$C_{gew} = \frac{\sum C}{n_1 \times 1 + n_2 \times 0.1} \times d \times \frac{V + l}{l} \times F \quad (5.1)$$

where C_{gew} is the weighted arithmetic mean of the number of viable bacteria, C is the total count of colonies of all evaluated Petri dishes, n_1 is the number of Petri dishes of the lowest evaluated dilution step, n_2 is the number of Petri dishes of the next higher evaluated dilution step, d is the dilution factor that of dilution dispensed into Petri dishes, V is the volume of SCDLP broth for wash-out, l is the volume of inoculation, and F is the factor to bring the analyzed volume to 1 ml.

The value of antimicrobial activity was calculated by subtracting the logarithmic value of the calculated viable counts on the sample from the viable counts on the reference after inoculation and incubation, as shown in the following equation (5.2):

$$log_{10} - reduction = log_{10}(T_{x,Re} - T_{x,Sa}) \quad (5.2)$$

where $T_{x,Re}$ is the bacterial concentration on the reference after x hours of inoculation and incubation, and $T_{x,Sa}$ is the bacterial concentration on the samples x hours after inoculation and incubation. Samples that showed a calculated log_{10}-reduction ≥ 2.0 log_{10} units after inoculation and incubation were considered effective antimicrobials according to ISO 22196:2007.

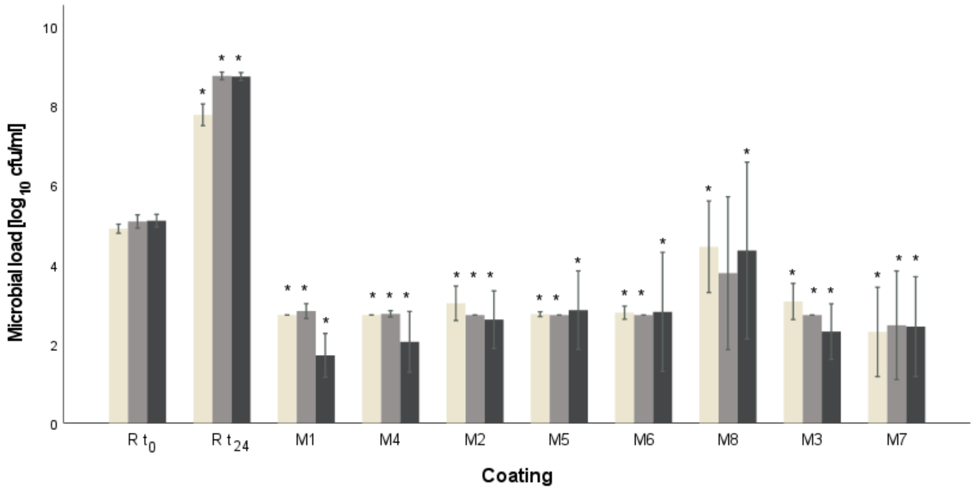
5.3.9.2 Statistical analysis

All data were transformed into log_{10} values before statistical analysis. The standard deviations in bacterial counts on references and samples were calculated. The data were analyzed for statistical outliers. Outliers were excluded from further (statistical) analyses. The differences in bacterial counts on references (t_0 and t_{24}) and coating samples were analyzed for significance using the one-way ANOVA for k-samples according to Kruskal-Wallis for independent samples. A rejected null hypothesis was followed by a pairwise comparison to identify significant differences in bacterial count on different coatings. The significance level was defined as a P value ≤ 0.05, respectively, and the P value of ≤ 0.01 as highly significant. The given significance values are Bonferroni corrected. For the description of data, bar charts were used. Data analysis was conducted with SPSS Statistics 27 (IBM Corp. 1989, 2020, New York, NY).

5.4 Results

5.4.1 Antimicrobial activity of surface coatings under standard (24 h, 35°C) and cold temperature conditions (120 h, 7°C) typical for applications in the food industry for perishable products

The bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on references at t_0 and t_{24} show an increase of 2.87 log_{10} units, 3.67 log_{10} units, and 3.62 log_{10} units, respectively, from an initial bacteria concentration of nearly 5.00 log_{10} cfu/ml after 24 h of storage at 35°C (Figure 5.2). At the same time, a reduction of 3.32 log_{10} units is identified for sample M8 against *Staph. aureus*. The bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on all other samples have highly significantly decreased (P ≤ 0.01) by more than 4.38 log_{10} units compared with the bacterial counts on the references. The standard deviations in bacterial counts are marginal with < ± 1.00 log_{10} cfu/ml for all tested bacteria on all samples except on samples M8 and M7 with standard deviations up to ± 2.13 log_{10} cfu/ml. So, all tested materials can be classified as antimicrobial surfaces under standard conditions.

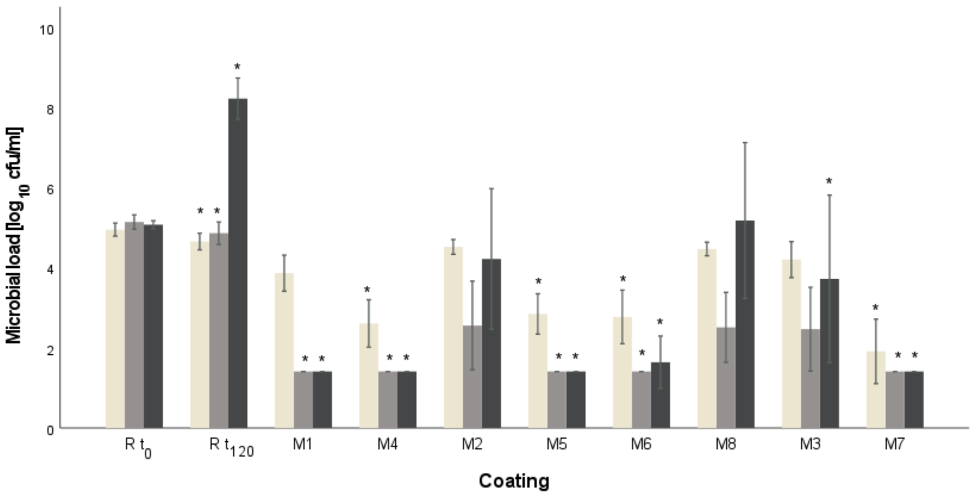


Number (n) of analyzed references (R) and samples M1 - M8 for *Staph. aureus*, *E. coli*, and *Ps. fluorescens* (24 h, 35°C)

	R t_0	R t_{24}	M1	M4	M2	M5	M6	M8	M3	M7
Staph. aureus	21	24	18	18	9	8	8	9	9	8
E. coli	20	18	8	7	9	8	8	10	9	9
Ps. fluorescens	18	25	8	9	9	9	12	12	9	9

Figure 5.2 Arithmetic mean of bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on references and samples M1 - M8 after storage of 24 h at 35°C.
Bacterial counts: (■) *Staph. aureus*, (■) *E. coli*, (■) *Ps. fluorescens*.
*Highly significant differences in bacterial counts on samples and references (P ≤ 0.01).

After 120 h at 7°C (Figure 5.3), conditions usually found in the food industry, only *Ps. fluorescens* shows a significant increase in bacterial count on the reference material of 3.15 $\log_{10}$ units up to 8.23 ± 0.50 $\log_{10}$ cfu/ml. On the reference material, the bacterial counts of *Staph. aureus* and *E. coli* stayed nearly the same. The different test samples (M1 - M8) show different antimicrobial activities at low temperatures. All samples are active against the gram-negative bacteria *E. coli* with reduction rates > 2.31 $\log_{10}$ units. Samples M1, M4, M5, M6, and M7 led to a reduction of bacterial counts of *E. coli* under the detection limit of 1.41 $\log_{10}$ cfu/ml. These samples show a highly significant difference to the reference after 120 h storage at 7°C (P ≤ 0.01). The samples M4 and M7 also show antimicrobial activity against *Staph. aureus* with a reduction in bacteria counts to 2.61 ± 0.56 $\log_{10}$ cfu/ml and 1.91 ± 0.76 $\log_{10}$ cfu/ml, respectively. Furthermore, all samples are active against *Ps. fluorescens* with reduction rates > 3.05 $\log_{10}$ units. The bacterial concentrations on the mineral binder based on liquid potassium silicate (M1, M4) and samples M5 and M7 are highly significantly reduced (P ≤ 0.01) under the detection limit after 120 h compared to the references. As shown under standard conditions, the acrylic polymer M8 exhibits the lowest antimicrobial activity, at low temperature, compared to the other samples.



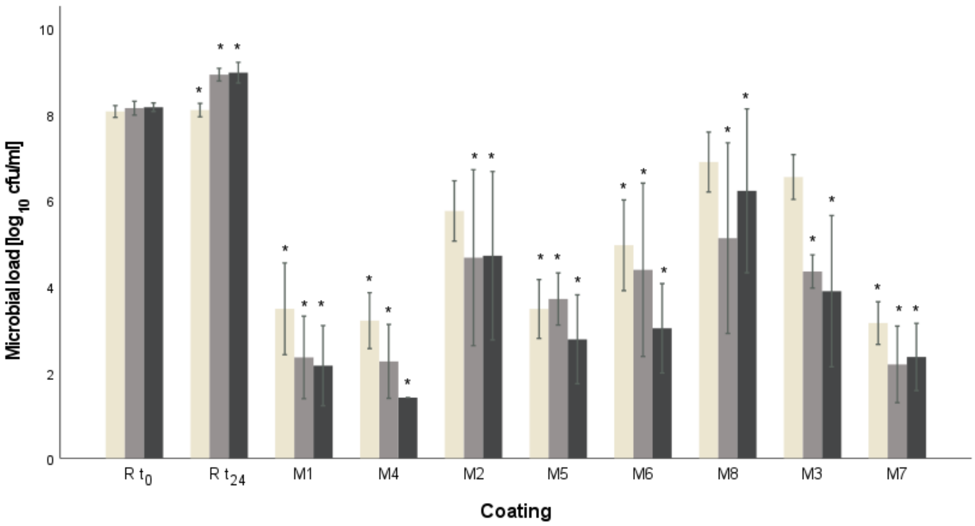
Number (n) of analyzed references (R) and samples M1 - M8 for *Staph. aureus*, *E. coli*, and *Ps. fluorescens* (120 h, 7°C)

	R t_0	R t_{24}	M1	M4	M2	M5	M6	M8	M3	M7
Staph. aureus	17	18	9	9	9	9	9	9	9	9
E. coli	17	16	9	9	9	9	8	9	9	9
Ps. fluorescens	20	18	9	9	12	9	8	9	12	8

Figure 5.3 Arithmetic mean of bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on references and samples M1 - M8 after storage of 120 h at 7°C.
Bacterial counts: (■) *Staph. aureus*, (■) *E. coli*, (■) *Ps. fluorescens*.
*Highly significant differences in bacterial counts on samples and references ($P \leq 0.01$).

5.4.2 Influence of a high initial bacteria concentration on the antimicrobial activity of surface coatings

The bacterial concentrations of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* inoculated with a high initial bacteria concentration of 10^8 cfu/ml on the references and samples after storage of 24 h at 35°C are shown in Figure 5.4. The tested bacteria on the reference materials show only a slight increase in bacterial counts to 8.10 ± 0.15 $\log_{10}$ cfu/ml (*Staph. aureus*), 8.93 ± 0.14 $\log_{10}$ cfu/ml (*E. coli*), and 8.98 ± 0.24 $\log_{10}$ cfu/ml (*Ps. fluorescens*). The bacterial counts of *E. coli* and *Ps. fluorescens* show a highly significant decrease in all samples after 24 h storage in comparison to the references ($P \leq 0.01$). Against *Staph. aureus*, the highest reduction rates, > 4.60 $\log_{10}$ units, could be achieved with the mineral binder based on liquid potassium silicate (M1, M4) and samples M5 and M7, as shown in the trials before. The standard deviations are marginal for all samples ≤ ± 1.00 $\log_{10}$ cfu/ml. The most effective active sample against *E. coli* is the pure functional extender M7, with a reduction rate of 6.75 $\log_{10}$ units. The bacterial count of *Ps. fluorescens* on sample M4 is minimized under the detection limit. As shown before, the lowest reduction rate of 2.76 $\log_{10}$ units is identified for sample M8 against *Ps. fluorescens*.



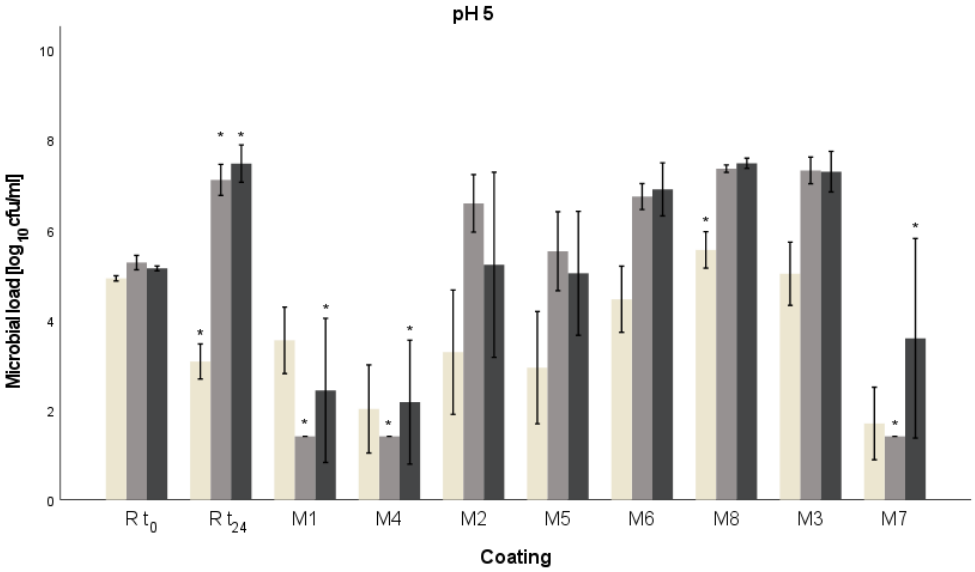
Number (n) of analyzed references (R) and samples M1 - M8 for *Staph. aureus*, *E. coli*, and *Ps. fluorescens* inoculated with a high initial bacteria concentration of 10^8 cfu/ml (24 h, 35°C)

	R t_0	R t_{24}	M1	M4	M2	M5	M6	M8	M3	M7
Staph. aureus	18	17	9	9	9	9	9	8	8	8
E. coli	20	21	9	9	12	8	12	12	7	8
Ps. fluorescens	24	24	9	9	12	9	9	12	9	9

Figure 5.4 Arithmetic mean of bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* inoculated with a high initial bacteria concentration of 10^8 cfu/ml on references and samples M1 - M8 after storage of 24 h at 35°C.
Bacterial counts: (■) *Staph. aureus*, (■) *E. coli*, (■) *Ps. fluorescens*.
*Highly significant differences in bacterial counts on samples and references (P ≤ 0.01).

5.4.3 Influence of different pH on the antimicrobial activity of surface coatings

The influence of pH 5, 7, and 9 on the antimicrobial activity against *Staph. aureus*, *E. coli*, and *Ps. fluorescens* are shown in Figure 5.5, Figure 5.6, and Figure 5.7. At pH 5 (Figure 5.5), the bacterial count of *Staph. aureus* on the reference material decreases by almost 2.00 $\log_{10}$ units to 3.07 ± 0.36 $\log_{10}$ cfu/ml. The bacterial counts on references for *E. coli* and *Ps. fluorescens* show an increase up to concentrations of 7.00 $\log_{10}$ cfu/ml. Due to the reduction of the bacterial counts on the references after 24 h, none of the samples show an antimicrobial activity ($\log_{10}$-reduction > 2.00 $\log_{10}$ units) against the gram-positive bacteria *Staph. aureus* at pH 5. Against *E. coli*, only the mineral binder based on liquid potassium silicate materials (M1, M4) and the pure functional extender (M7) are antimicrobial active with a highly significant difference to the references (P ≤ 0.01) by reduction of bacterial counts under the detection limit as shown for low temperature. As shown for a high initial bacteria count, at pH 5 the lowest bacterial count for *Ps. fluorescens* shows the sample M4 with 2.17 ± 1.32 $\log_{10}$ cfu/ml. The highest bacterial count shows the sample M8 with 7.48 ± 0.11 $\log_{10}$ cfu/ml. Comparing the bacterial counts of *Ps. fluorescens*, the mineral binder based on liquid potassium silicate materials (M1, M4) and the pure functional extender (M7) show a highly significant difference from the references (P ≤ 0.01).

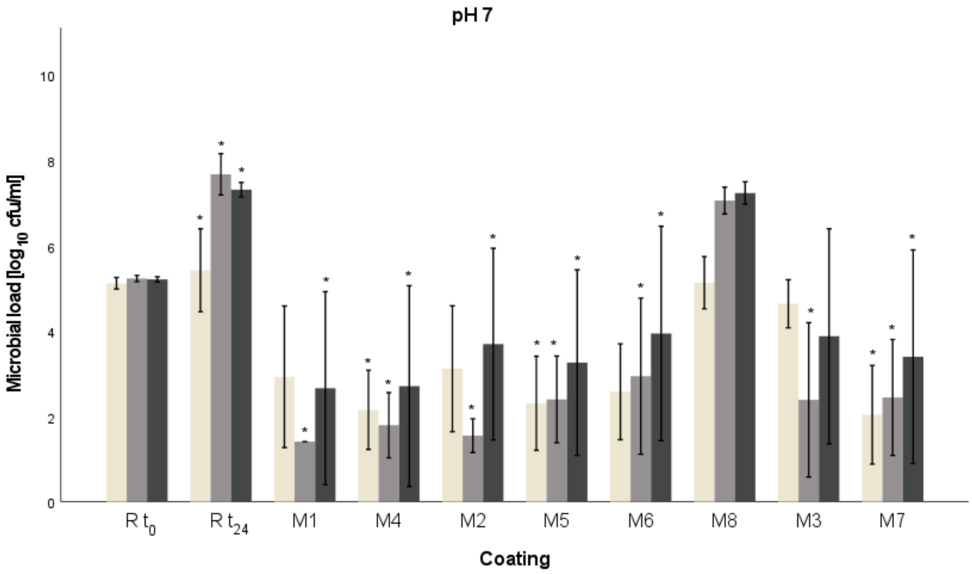


Number (n) of analyzed references (R) and samples M1 - M8 for *Staph. aureus*, *E. coli*, and *Ps. fluorescens* inoculated at pH 5 (24 h, 35°C)

	R t_0	**R t_{24}**	**M1**	**M4**	**M2**	**M5**	**M6**	**M8**	**M3**	**M7**
Staph. aureus	9	6	9	9	9	9	9	9	9	8
E. coli	9	9	8	9	8	9	9	9	9	8
Ps. fluorescens	11	12	12	12	12	12	8	8	9	12

Figure 5.5 Arithmetic mean of bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* inoculated at pH 5 on references and samples M1 - M8 after storage of 24 h at 35°C.
Bacterial counts: (■) *Staph. aureus*, (■) *E. coli*, (■) *Ps. fluorescens*.
*Highly significant differences in bacterial counts on samples and references (P ≤ 0.01).

At pH 7 (Figure 5.6), the bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on references at t_0 and t_{24} show an increase to a final bacteria concentration of 5.43 ± 0.93 $\log_{10}$ cfu/ml, 7.68 ± 0.46 $\log_{10}$ cfu/ml, and 7.31 ± 0.16 $\log_{10}$ cfu/ml, respectively. In contrast to pH 5 for *Staph. aureus*, antimicrobial activity was measured at pH 7 with reduction rates between 0.30 and 3.37 $\log_{10}$ units. No antimicrobial activity was achieved for samples M3 and M8 against *Staph. aureus*. As shown in previous trials, except M8, all tested samples show antimicrobial activity against *E. coli* and *Ps. fluorescens* at pH 7. Except for M3 against *Ps. fluorescens*, the shown antimicrobial activities showed highly significant differences compared to the references (P ≤ 0.01).

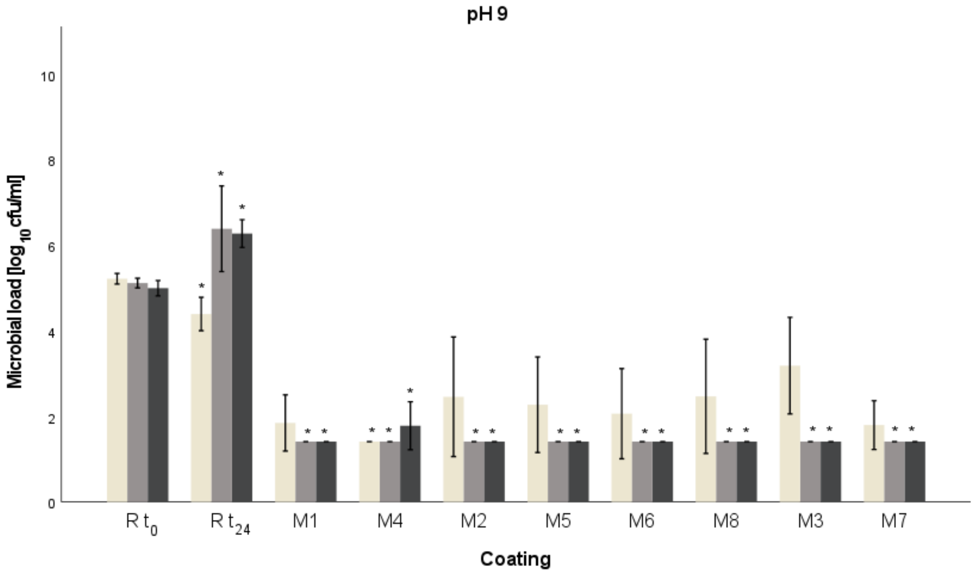


Number (n) of analyzed references (R) and samples M1 - M8 for *Staph. aureus*, *E. coli*, and *Ps. fluorescens* inoculated at pH 7 (24 h, 35°C)

	R t_0	R t_{24}	M1	M4	M2	M5	M6	M8	M3	M7
Staph. aureus	12	12	9	9	9	9	9	9	9	12
E. coli	11	9	9	9	8	9	12	9	12	9
Ps. fluorescens	12	9	12	12	12	12	12	9	12	12

Figure 5.6 Arithmetic mean of bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* inoculated at pH 7 on references and samples M1 - M8 after storage of 24 h at 35°C.
Bacterial counts: (■) *Staph. aureus*, (■) *E. coli*, (■) *Ps. fluorescens*.
*Highly significant differences in bacterial counts on samples and references ($P \leq 0.01$).

At pH 9 (Figure 5.7), the bacterial count of *Staph. aureus* on the references decreases from initial 5.22 ± 0.12 $\log_{10}$ cfu/ml to 4.39 ± 0.37 $\log_{10}$ cfu/ml. *E. coli* and *Ps. fluorescens* show an increase in bacterial counts on the references about 1.30 $\log_{10}$ units to concentrations of 6.30 $\log_{10}$ cfu/ml at pH 9. An antimicrobial activity against *Staph. aureus* is shown on samples M1, M4, M5, M6, and M7, with a highly significant difference in sample M4 compared to the references at pH of 9 ($P \leq 0.01$). The bacterial counts of *E. coli* and *Ps. fluorescens* were reduced under the detection limit on all samples M1 - M8 leading to highly significant differences compared with the bacterial counts on the references at pH 9 ($P \leq 0.01$).



Number (n) of analyzed references (R) and samples M1 - M8 for *Staph. aureus*, *E. coli*, and *Ps. fluorescens* inoculated at pH 9 (24 h, 35°C)

	R t_0	R t_{24}	M1	M4	M2	M5	M6	M8	M3	M7
Staph. aureus	11	9	9	9	9	9	9	12	9	8
E. coli	9	9	8	9	8	8	8	8	9	8
Ps. fluorescens	9	9	9	9	9	9	9	9	9	8

Figure 5.7 Arithmetic mean of bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* inoculated at pH 9 on references and samples M1 - M8 after storage of 24 h at 35°C. Bacterial counts: (■) *Staph. aureus*, (■) *E. coli*, (■) *Ps. fluorescens*. *Highly significant differences in bacterial counts on samples and references (P ≤ 0.01).

5.4.4 Influence of food components starch, meat extract, and oleic acid on the antimicrobial activity of surface coatings

The bacterial concentrations of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on the references and selected samples (M1, M5 - M8) with added food components, starch, meat extract, and oleic acid, which are shown in Table 5.4. The bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on references after inoculation with starch reach concentrations of 9.00 log_{10} cfu/ml. Similar to the results after 24 h storage at 35°C, nearly all tested samples (M1, M5 - M8) show antimicrobial activity against *Staph. aureus*, *E. coli*, and *Ps. fluorescens*. With added starch, the bacterial counts of *Staph. aureus* and *Ps. Fluorescens* on samples M1, M5 – M7 and of *E. coli* on all tested samples show highly significant differences compared to the bacterial counts on references (P ≤ 0.01). Comparable to results at pH 7, the reduction rate is the lowest on sample M8 for all three tested bacteria. The highest standard deviations in bacterial count (± 2.00 log_{10} cfu/ml) are shown on sample M8 for all three tested bacteria. For *E. coli*, the lowest bacterial count shows sample M5, where the bacterial count is reduced to the detection limit of 1.41 log_{10} cfu/ml.

The bacterial count of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on references after inoculation with meat extract increased up to 10.09 ± 0.19 log_{10} cfu/ml. The samples M1, M5 - M7 were antimicrobial active against all tested bacteria with highly significant differences compared to the bacterial counts on references ($P \leq 0.01$), except sample M7 against *Ps. fluorescens*. Similar to the results for the addition of starch, the bacteria count on sample M8 remains high for the tested bacteria, with ≥ 8.00 log_{10} cfu/ml.

By incubation with oleic acid, the bacterial counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on references show an increase in bacterial concentrations, up to 9.24 ± 0.14 log_{10} cfu/ml. The antimicrobial activity of the tested samples in contact with oleic acid is comparable to the results shown for the incubation with meat extract. Sample M8 is the only material in which the bacterial count is decreased only slightly to values around 8.00 log_{10} cfu/ml, as previously demonstrated for the incubation with starch and meat extract.

Table 5.4 Individual bacteria counts of *Staph. aureus*, *E. coli*, and *Ps. fluorescens* on references and samples by added food components starch, meat extract, and oleic acid (24 h, 35°C).

Sample Code	Starch			Meat extract			Oleic acid		
	Staph. aureus	***E. coli***	***Ps. fluorescens***	***Staph. aureus***	***E. coli***	***Ps. fluorescens***	***Staph. aureus***	***E. coli***	***Ps. fluorescens***
Reference									
R t_0	5.04 ± 0.17 (n=12)	5.13 ± 0.12 (n=12)	5.04 ± 0.17 (n=12)	5.10 ± 0.15 (n=12)	5.22 ± 0.09 (n=12)	5.11 ± 0.14 (n=12)	5.08 ± 0.11 (n=12)	4.86 ± 0.09 (n=21)	4.95 ± 0.12 (n=12)
R t_{24}	9.08 ± 0.16* (n=12)	9.07 ± 0.18* (n=12)	9.08 ± 0.16* (n=12)	9.76 ± 0.16* (n=11)	10.05 ± 0.16* (n=12)	10.09 ± 0.19* (n=12)	8.14 ± 0.35* (n=12)	9.16 ± 0.11* (n=19)	9.24 ± 0.14* (n=12)
Mineral binder based on liquid potassium silicate									
M1	2.12 ± 1.00* (n=9)	2.57 ± 1.79* (n=9)	2.12 ± 1.00* (n=9)	3.59 ± 0.68* (n=9)	2.64 ± 1.42* (n=11)	2.64 ± 0.93* (n=9)	3.63 ± 0.53* (n=9)	2.25 ± 0.67* (n=8)	1.90 ± 1.29* (n=8)
Acrylic polymer									
M5	2.55 ± 0.87* (n=9)	1.41 ± 0.00* (n=8)	2.55 ± 0.87* (n=9)	3.28 ± 2.00* (n=12)	1.81 ± 0.70* (n=8)	3.59 ± 0.45* (n=7)	3.25 ± 2.08* (n=9)	4.87 ± 3.20* (n=7)	3.76 ± 2.31* (n=9)
M6	2.93 ± 0.92* (n=8)	2.39 ± 1.84* (n=9)	2.93 ± 0.92* (n=8)	3.23 ± 1.16* (n=11)	1.73 ± 0.55* (n=8)	3.31 ± 1.36* (n=10)	5.00 ± 2.56* (n=12)	2.52 ± 1.00* (n=7)	5.93 ± 2.41 (n=8)
M8	4.27 ± 1.99 (n=12)	3.43 ± 2.00* (n=12)	4.27 ± 1.99 (n=12)	7.98 ± 1.27 (n=10)	9.75 ± 0.07 (n=9)	9.40 ± 0.39 (n=9)	7.89 ± 0.52 (n=10)	8.81 ± 0.26 (n=9)	8.69 ± 0.03 (n=8)
Pure functional extender									
M7	2.95 ± 1.07* (n=9)	2.00 ± 0.91* (n=9)	2.95 ± 1.07* (n=9)	3.34 ± 1.68* (n=12)	2.61 ± 0.87* (n=9)	6.42 ± 2.26 (n=7)	5.62 ± 2.03 (n=11)	4.37 ± 2.45* (n=10)	5.75 ± 2.74* (n=7)

*Highly significant differences in bacterial counts on samples and references ($P \leq 0.01$).

5.5 Discussion

The analyzed coatings characterized either by a mineral binder based on liquid potassium silicate in combination with a new class of biopolymer or non-conserved acrylate polymers or non-conserved styrene-acrylate polymers using a functional extender show high antimicrobial activity against the selected gram-positive and gram-negative bacteria *Staph. aureus*, *E. coli*, and *Ps. fluorescens* under different test conditions. The results show that both samples of the mineral binder based on liquid potassium silicate (M1, M4) show a higher antimicrobial activity under standard conditions than the non-conserved acrylate polymers (M2, M5, M6, M8) and non-conserved styrene-acrylate polymers (M3). However, the functional extender added to acrylic polymers does not show any improved properties compared to the pure functional extender suspended in water, even in combination with TiO_2, and also in comparison to the added biopolymer. This can be explained, as stated by Bulitta et al. (2020), either by the fact that TiO_2-containing surfaces need to be actively activated with UV light, which was not given in this study. Besides, it is conceivable that the formation of free radicals already leads to a degradation of the organic binder resulting in reduced antimicrobial activity.

The specific mode of action of complex chemical reactions, attributed to a mineral mixture based on $Ca_3Al_2(OH)_{12}$, is supposed to explain the antimicrobial activity. In an alkaline milieu, aluminum hydroxide ($Al(OH)_3$) reacts amphoterically with an ionic character; as a result, it has surface-active properties and is generally water-soluble. $Ca(OH)_2$, included as a mineral mixture in the functional extender, is also water-soluble with an ionic character. The amphoteric $Al(OH)_3$ reacts with $Ca(OH)_2$ to form a stable complex called katoite ($Ca_3[Al(OH)_4]_2$), a hydroxide rarely found in nature. The anomaly of water allows this katoite to dissociate and form ions. These ions react with free sodium ions and form temporary sodium hydroxide (NaOH), which in turn is in equilibrium with the formation of disodium oxide (Na_2O) and water (H_2O). In turn, Na_2O reacts with another water molecule, releasing heat (exothermic reaction) to form two molecules of NaOH, occurring in nature only in bound form in numerous minerals. Depending on external conditions, depending on notably water content, temperature, pressure, and light, long-term, highly complex structures are formed through constant equilibrium reactions, leading to changes in mineral-containing compounds, thereby contributing to long-term stability and antimicrobial activity through the dynamic and the response of Na^+ and OH^- ions.

In general, the results of this study validate the formerly stated assumptions explaining the mechanisms of antimicrobial activity. Surface-active properties due to minerals leading to a charged surface, migration of water-soluble minerals, as well as the formation and migration in the dynamics of the sodium ions in an alkaline milieu with water and oxygen, lead to proven antimicrobial activities. Furthermore, the results indicate that the binder influences the antimicrobial activity of the functional extender. These are indicated by the decrease in antimicrobial activity shown for the dispersion of the pure functional extender in the acrylic polymer (M8) compared to the pure functional extender dissolved in water (M7). Here, the results indicate that in combination with the binder, the functional extender is bound in the film, and thus the minerals are not free to undergo chemical reactions leading to a charged surface. In combination with a biopolymer based on renewable resources, the mineral binder based on liquid potassium silicate is a significantly promising approach since it achieves good effectiveness without further additives.

Due to the presence of the mineral ions Ca^{2+}, Al^{3+}, and Na^{+}, the coatings exhibit positively charged surfaces. Therefore, the antimicrobial activity will be attributed to a combined effect of the charged surface of the coatings and a dynamic effect of minerals and Na^{+} and OH^{-} ions. Several authors have shown that the activity of charged antimicrobial polymeric surfaces is influenced by the bacterial strains themselves, due to different charges leading to distinct electrostatic interactions between microorganisms and charged surfaces, leading to a depolarization of the cytoplasmic membrane resulting in cell death (Dohlen et al., 2016; Hewitt et al., 2004; Potter et al., 2005). In general, most bacteria carry a net negative surface charge under most physiological conditions (Jucker et al., 1996). Potter et al. (2005) proved a correlation between electrophoretic mobility and the antimicrobial efficiency of a cationic antimicrobial peptide. The electrophoretic mobility is also more negative for the relevant gram-positive bacteria (*Staph. aureus*) than for the gram-negative bacteria (*E. coli*, *Ps. fluorescens*). Thus, the nearly neutral charge of *Ps. fluorescens* could cause a decreased activity (high resistance) of positively charged surfaces followed by *E. coli* compared to the more negatively charged bacteria. In contrast, Hewitt et al. (2004) assumed, based on their results, the existence of a protective function of the outer membrane of gram-negative bacteria. As stated by Gottenbos et al. (2003), resistance of *Pseudomonas spp.* compared to *E. coli* against cationic surfaces originates from a higher production of exopolysaccharides. Dohlen et al. (2016) demonstrated that the gram-negative bacteria *E. coli* and *Ps. fluorescens* are less sensitive to a poly(TBAMS) polymer compared to other tested microorganisms after 24 h at 7°C due to varying strength of the negative charges that these bacteria exhibit in physiological solutions. Our studies show that no decrease in antimicrobial activity for gram-negative bacteria compared to the tested gram-positive bacteria after 24 h at 35°C and 120 h at 7°C was detected. This leads to the hypothesis that the antimicrobial activity is a combination of the effect of surface charge along with the dynamic of minerals and Na^{+} and OH^{-} ions, leading to cell death.

The temperature is often identified to affect the antimicrobial activity of different materials (Chang et al., 2015; Dohlen et al., 2016; Faúndez et al., 2004; Kampmann et al., 2008). In our studies, the initial bacterial counts of the references for *Staph. aureus* and *E. coli* are higher than after 24 h of incubation, which is explained by the low incubation temperature. Akin, the antimicrobial activity is reduced at low temperatures compared to higher temperatures, which are close to the optimal growth temperatures of the microorganisms. Comparing the results at 35°C and 7°C, these indicate a temperature-dependent activity of the surface coatings for *Staph. aureus* and *E. coli* with reduced activity at cold temperatures. As demonstrated by Dohlen et al. (2016), for surface active materials, poly(TBAMS) showed a lower reduction rate stored at 2°C and 4°C for *Staph. aureus*, *E. coli*, and *B. thermosphacta* in comparison to 35°C. Generally, microorganisms show a higher metabolism and growth at their optimal growth temperature (Jay, 1995). Thus, a lower activity of contact active materials can be explained by changes in bacteria cell structure due to adaption in gene regulation depending on environmental conditions (Di Bonaventura et al., 2008; Liu et al., 2002). A positive correlation is proven between cell surface hydrophobicity and temperature (Di Bonaventura et al., 2008). However, a possible explanation for different antimicrobial activities at low temperatures against *E. coli* and *Ps. fluorescens* can be explained by various fatty acids present in the outer membrane of these microorganisms (Cullen et al., 1971; Gill & Suisted, 1978). Additionally, for migrating systems, the effect is caused by the slower release of active components into the environment at lower temperatures than applied for most tests on the antimicrobial activity at 35°C (Quintavalla & Vicini, 2002). The study results confirm that the antimicrobial activity of the surface coatings is attributed to the effect of a charged surface, especially at low temperatures when a lack of growth for some microorganisms is observed.

The shown antimicrobial activities, leading to highly significantly decreased bacterial counts compared to the references, even under high initial bacteria counts for most of the analyzed coatings, are in accordance with the findings reported by Braun et al. (2017). In the study, the homopolymer poly(TBAMS) showed a highly significant decrease in the initial bacterial counts (Braun et al., 2017). At a high initial bacteria concentration, the effect of the electrophoretic mobilities of the bacteria is more pronounced when compared to a moderate initial concentration. Next to the electrostatic interactions, the availability of active groups is also proportional to the number of bacterial counts (Braun et al., 2017; Kurinčič et al., 2016). In order to guarantee the antimicrobial activity, it must be ensured that killed cells are regularly removed from the surface as the dead biomass, on the one hand, can mask the antimicrobial effect and, on the other hand, it offers an organic substrate for newly introduced microorganisms (Bulitta et al., 2020). The fact that killed cells do not remain on the surface makes a successive killing of bacteria possible with an extension of contact time, leading to comparable results for moderate initial concentrations (Lenoir et al., 2006). The research results led to the hypothesis that the antimicrobial activity is a combination of surface-active properties and a dynamic effect of water-soluble minerals and Na^+ and OH^- ions. As the availability of active groups needs to be proportional to the number of bacterial counts to achieve comparable results between a moderate and high initial concentration, an antimicrobial effect is attributed to the migration of minerals into the cells of microorganisms. Furthermore, the acrylate polymer leads to an inhibition of the antimicrobial activity of the functional extender, comparing the antimicrobial activities of M8 and M7.

In contrast to our findings of the influence of different pH values, studies on the polymers poly(TBAMS) and poly(TBAEMA) showed reduced antimicrobial activities against *E. coli* due to a more neutrally charged surface at alkaline conditions (Buranasompob, 2005). This is attributed to a decreasing zeta potential of compounds of LLDPE and TBAEMA at increasing pH values (Seyfriedsberger et al., 2006). A negative correlation between pH and zeta potential and a positive correlation between zeta potential and antimicrobial activity were substantiated for chitosan (Chang et al., 2015). In our studies, reduced antimicrobial activities at alkaline conditions were not proven. The higher antimicrobial activity under alkaline conditions compared to acidic conditions is explained by the formerly stated assumption of an amphoteric reaction of $Al(OH)_3$ in an alkaline milieu. The reaction of $Al(OH)_3$ with $Ca(OH)_2$ is essential for the formation of the so-called katoite $Ca_3Al_2(OH)_{12}$, which in turn is a prerequisite for constant equilibrium reactions leading to changes in the mineral-containing compounds. It is assumed that reactive, stable, and adaptable compounds are formed that can cause antimicrobial activity in this milieu through Na^+ and OH^- ions. The formed reactive, stable, and adaptable compounds emphasize the potential use of the investigated coatings in various applications. Conceivable applications also include food contact surfaces that are exposed to changing environmental conditions during processing, cleaning, and disinfection.

Based on the results of several other studies (Chen & Cooper, 2002; Dohlen et al., 2016; Lenoir et al., 2006; Roy et al., 2008; Sahalan et al., 2013), it was expected that the level of antimicrobial activity is probably affected by meat components. For different cationic antibiotics and cationic polymers, whose modes of action are related to membrane disruption, it was shown in several studies that divalent cations like Ca^{2+} and Mg^{2+} inhibit antibacterial activity, especially on gram-negative bacteria (Chen & Cooper, 2002; Dohlen et al., 2016; Lenoir et al., 2006; Roy et al., 2008; Sahalan et al., 2013). In nutrient-rich media like meat, bacterial cells are exposed to lower osmotic stress than in media with low ionic strength (Møretrø et al., 2012). In the studies of Noyce et al. (2006) and Cutter (1999), fatty acids seem to provide a protective matrix for microorganisms, leading to reduced antimicrobial activities. Studies of silver-containing materials showed that antimicrobial activity is inhibited in the presence of proteins (Ilg & Kreyenschmidt, 2011; Møretrø et al., 2012). Proteins can interact with antimicrobial agents and occupy the functional amino groups. Furthermore, in the presence of nutrients, the electrical charge of the bacteria is affected (Dohlen et al., 2016). In the present studies, the level of antimicrobial activity of the surface coatings was not affected by meat components; as shown before, this part of the study confirms a combined effect of surface charge along with the dynamic of minerals of Na^+ and OH^- ions on antimicrobial activity.

5.6 Conclusion

The analyzed coatings based on mineral binder based on liquid potassium silicate, non-conserved acrylate polymers, and non-conserved styrene-acrylate polymers exhibit antimicrobial properties in contact with high bacteria concentration, under low-temperature conditions, in acidic and alkaline environments as well as in the presence of meat components against several microorganisms. The findings underline the potential of these coatings as a new antimicrobial material for different applications, such as antimicrobial packaging and antimicrobial food contact surfaces. Especially the mineral binder, based on liquid potassium silicate, in combination with a biopolymer, based on renewable resources, is a promising approach since an effective antimicrobial activity is achieved without further additives. However, additives such as metal ions tend to accumulate in food upon contact. In further experiments, it needs to be studied to what extent components of the organic coatings may migrate in the product and could become a possible health hazard. Overall, the experiments present a first evaluation of the general antimicrobial activity of organic coatings under specific conditions in perishable food supply chains. It is believed that these coatings can be applied to bio-based packaging materials, generating the antimicrobial properties of these materials. In contrast to the direct incorporation of antimicrobial-active substances in packaging materials, the coating of foils with antimicrobial polymers or substances turns out to be a promising approach to produce antimicrobial packaging. Additionally, these coatings attract attention as coatings for organic packaging materials like wood and paper to improve the physical properties next to their antimicrobial activity. In the future, the antimicrobial activity of coated packaging materials needs to be investigated under real conditions. To check the applicability and determine the effect of packaging materials coated with films (containing mineral binders based on liquid potassium silicate, non-conserved acrylate polymers, and non-conserved styrene-acrylate polymers) on quality loss and shelf life of fresh produce. In the long term, these coatings can be implemented to improve hygiene, food safety, and quality, as well as the extended shelf life of perishable produce.

Funding

This research was funded by the European Union via the European Regional Development Fund EFRE.NRW (Grant EFRE 0500035, “Biobasierte Produkte”).

Acknowledgements

The authors wish to thank the company PETRYmade Oberflächentechnik who kindly provided and prepared the sample materials, Claudia Waldhans, Maureen Mittler, Selina Siemens, and Annika Kruse for their highly appreciated, continuous assistance in the experimental parts and Lucas Correa Dresch for the performed final English revision of the manuscript.

References

Adlhart, C., Verran, J., Azevedo, N. F., Olmez, H., Keinänen-Toivola, M. M., Gouveia, I., Melo, L. F., & Crijns, F. (2018). Surface modifications for antimicrobial effects in the healthcare setting: A critical overview. *The Journal of Hospital Infection*, *99*(3), 239–249. https://doi.org/10.1016/j.jhin.2018.01.018.

AL-Jawad, S. M., Taha, A. A., & Salim, M. M. (2017). Synthesis and characterization of pure and Fe doped TiO2 thin films for antimicrobial activity. *Optik*, *142*, 42–53. https://doi.org/10.1016/j.ijleo.2017.05.048.

Appendini, P., & Hotchkiss, J. H. (2002). Review of antimicrobial food packaging. *Innovative Food Science & Emerging Technologies*, *3*, 113–126. https://doi.org/10.1016/S1466-8564(02)00012-7.

Arellano, U., Asomoza, M., & Ramírez, F. (2011). Antimicrobial activity of Fe–TiO2 thin film photocatalysts. *Journal of Photochemistry and Photobiology a: Chemistry*, *222*(1), 159–165. https://doi.org/10.1016/j.jphotochem.2011.05.016.

Asharani, P. V., Hande, M. P., & Valiyaveettil, S. (2009). Anti-proliferative activity of silver nanoparticles. *BMC Cell Biology*, *10*, 65. https://doi.org/10.1186/1471-2121-10-65.

Balasubramanian, A., Rosenberg, L. E., Yam, K., & Chikindas, M. L. (2009). Antimicrobial Packaging: Potential vs. Reality - A Review. *Journal of Applied Packaging Reseach*, *3*(4), 193–221.

Beyth, N., Houri-Haddad, Y., Domb, A., Khan, W., & Hazan, R. (2015). Alternative antimicrobial approach: Nano-antimicrobial materials. *Evidence-Based Complementary and Alternative Medicine: ECAM*, *2015*, 246012. https://doi.org/10.1155/2015/246012.

Braun, C., Dohlen, S., Ilg, Y., Brodkorb, F., Fischer, B., Heindirk, P., Kalbfleisch, K., Richter, T., Robers, O., Kreyenschmidt, M., Lorenz, R., & Kreyenschmidt, J. (2017). Antimicrobial Activity of Intrinsic Antimicrobial Polymers Based on Poly((tertbutyl-amino)-methyl-styrene) Against Selected Pathogenic and Spoilage Microorganisms Relevant in Meat Processing Facilities. *Journal of Antimicrobial Agents*, *03*(01). https://doi.org/10.4172/2472-1212.1000136.

Bulitta, C., Höfer, D., Passoth, N., Schulte, S., & Seifert, M. (2020). *Antimikrobielle Oberflächen zur Infektionsprävention: Werk- und Wirkstoffe, Prüfverfahren sowie rechtliche und regulatorische Rahmenbedingungen.* Verein Deutscher Ingenieure e.V.

Bundesinstitut für Risikobewertung. (2021). *Titandioxid – gibt es gesundheitliche Risiken? Aktualisierte Fragen und Antworten des BfR vom 12. Mai 2021.* Bundesinstitut für Risikobewertung. https://www.bfr.bund.de/de/titandioxid___gibt_es_gesundheitliche_risiken_-240812.html.

Buranasompob, A. (2005). *Kinetics of the inactivation of microorganisms by water insoluble polymers with antimicrobial activity.* https://doi.org/10.14279/depositonce-1049.

Chang, S.-H., Lin, H.-T. V., Wu, G.-J., & Tsai, G. J. (2015). Ph Effects on solubility, zeta potential, and correlation between antibacterial activity and molecular weight of chitosan. *Carbohydrate Polymers*, *134*, 74–81. https://doi.org/10.1016/j.carbpol.2015.07.072.

Chen, C. Z., & Cooper, S. L. (2002). Interactions between dendrimer biocides and bacterial membranes. *Biomaterials*, *23*(16), 3359–3368. https://doi.org/10.1016/S0142-9612(02)00036-4.

Chen, M. C., Koh, P. W., Ponnusamy, V. K., & Lee, S. L. (2022). Titanium dioxide and other nanomaterials based antimicrobial additives in functional paints and coatings: Review. *Progress in Organic Coatings*, *163*, 106660. https://doi.org/10.1016/j.porgcoat.2021.106660.

Coma, V. (2008). Bioactive packaging technologies for extended shelf life of meat-based products. *Meat Science*, *78*(1-2), 90–103. https://doi.org/10.1016/j.meatsci.2007.07.035.

Commission Regulation (EC) No 450/2009 of 29 May 2009 on active and intelligent materials and articles intended to come into contact with food, Official Journal of the European Union (2009)., 52, 3-11.

Commission Regulation (EU) No 10/2011 of 14 January 2011 on plastic materials and articles intended to come into contact with food, Official Journal of the European Union, 12, 1-89.

Cooksey, K. (2005). Effectiveness of antimicrobial food packaging materials. *Food Additives and Contaminants*, *22*(10), 980–987. https://doi.org/10.1080/02652030500246164.

Cullen, J., Phillips, M. C., & Shipley, G. G. (1971). The effects of temperature on the composition and physical properties of the lipids of Pseudomonas fluorescens. *The Biochemical Journal*, *125*(3), 733–742. https://doi.org/10.1042/bj1250733.

Cutter, C. N. (1999). The effectiveness of triclosan-incorporated plastic against bacteria on beef surfaces. *Journal of Food Protection*, *62*(5), 474–479. https://doi.org/10.4315/0362-028x-62.5.474.

Dehnad, D., Mirzaei, H., Emam-Djomeh, Z., Jafari, S.-M., & Dadashi, S. (2014). Thermal and antimicrobial properties of chitosan-nanocellulose films for extending shelf life of ground meat. *Carbohydrate Polymers*, *109*, 148–154. https://doi.org/10.1016/j.carbpol.2014.03.063.

Di Bonaventura, G., Piccolomini, R., Paludi, D., D'Orio, V., Vergara, A., Conter, M., & Ianieri, A. (2008). Influence of temperature on biofilm formation by Listeria monocytogenes on various food-contact surfaces: Relationship with motility and cell surface hydrophobicity. *Journal of Applied Microbiology*, *104*(6), 1552–1561. https://doi.org/10.1111/j.1365-2672.2007.03688.x.

Dohlen, S., Braun, C., Brodkorb, F., Fischer, B., Ilg, Y., Kalbfleisch, K., Lorenz, R., Kreyenschmidt, M., & Kreyenschmidt, J. (2017). Effect of different packaging materials containing poly-2-(tert-butylamino) methylstyrene on the growth of spoilage and pathogenic bacteria on fresh meat. *International Journal of Food Microbiology*, *257*, 91–100. https://doi.org/10.1016/j.ijfoodmicro.2017.06.007.

Dohlen, S., Braun, C., Brodkorb, F., Fischer, B., Ilg, Y., Kalbfleisch, K., Lorenz, R., Robers, O., Kreyenschmidt, M., & Kreyenschmidt, J. (2016). Potential of the polymer poly-2-(tert-butylamino) methylstyrene as antimicrobial packaging material for meat products. *Journal of Applied Microbiology*, *121*(4), 1059–1070. https://doi.org/10.1111/jam.13236.

Dutta, P. K., Tripathi, S., Mehrotra, G. K., & Dutta, J. (2009). Perspectives for chitosan based antimicrobial films in food applications. *Food Chemistry*, *114*(4), 1173–1182. https://doi.org/10.1016/j.foodchem.2008.11.047.

Dyshlyuk, L., Babich, O., Ivanova, S., Vasilchenco, N., Atuchin, V., Korolkov, I., Russakov, D., & Prosekov, A. (2020). Antimicrobial potential of ZnO, TiO2 and SiO2 nanoparticles in protecting building materials from biodegradation. *International Biodeterioration & Biodegradation*, *146*, 104821. https://doi.org/10.1016/j.ibiod.2019.104821.

Faúndez, G., Troncoso, M., Navarrete, P., & Figueroa, G. (2004). Antimicrobial activity of copper surfaces against suspensions of Salmonella enterica and Campylobacter jejuni. *BMC Microbiology*, *4*, 19. https://doi.org/10.1186/1471-2180-4-19.

Gill, C. O., & Suisted, J. R. (1978). The effects of temperature and growth rate on the proportion of unsaturated fatty acids in bacterial lipids. *Journal of General Microbiology*, *104*(1), 31–36. https://doi.org/10.1099/00221287-104-1-31.

Gottenbos, B., van der Mei, H. C., Klatter, F., Grijpma, D. W., Feijen, J., Nieuwenhuis, P., & Busscher, H. J. (2003). Positively charged biomaterials exert antimicrobial effects on gram-negative bacilli in rats. *Biomaterials*, *24*(16), 2707–2710. https://doi.org/10.1016/s0142-9612(03)00083-8.

Gupta, K., Singh, R. P., Pandey, A [Ashutosh], & Pandey, A [Anjana] (2013). Photocatalytic antibacterial performance of TiO2 and Ag-doped TiO2 against S. Aureus. P. Aeruginosa and E. Coli. *Beilstein Journal of Nanotechnology*, *4*, 345–351. https://doi.org/10.3762/bjnano.4.40.

Han, J. H. (2003). Antimicrobial food packaging. In *Novel Food Packaging Techniques* (pp. 50–70). Elsevier. https://doi.org/10.1533/9781855737020.1.50.

Hewitt, C. J., Franke, R., Marx, A., Kossmann, B., & Ottersbach, P. (2004). A study into the anti-microbial properties of an amino functionalised polymer using multi-parameter flow cytometry. *Biotechnology Letters*, *26*(7), 549–557. https://doi.org/10.1023/b:bile.0000021954.82099.a0.

Ilg, Y., & Kreyenschmidt, J. (2011). Effects of food components on the antimicrobial activity of polypropylene surfaces containing silver ions (Ag+). *International Journal of Food Science & Technology*, *46*(7), 1469–1476. https://doi.org/10.1111/j.1365-2621.2011.02641.x.

Ilg, Y., & Kreyenschmidt, J. (2012). Review: Nutzen und Risiken der Anwendung antimikrobieller Werkstoffe in der Lebensmittelkette. *Journal of Food Safety and Food Quality*, *63*(2), 28–34.

Jay, J. M. (1995). Low-Temperature Food Preservation and Characteristics of Psychrotrophic Microorganisms. In J. M. Jay (Ed.), *Food Science Texts Series. Modern Food Microbiology* (pp. 328–346). Springer. https://doi.org/10.1007/978-1-4615-7476-7_15.

Jucker, B. A., Harms, H., & Zehnder, A. J. (1996). Adhesion of the positively charged bacterium Stenotrophomonas (Xanthomonas) maltophilia 70401 to glass and Teflon. *Journal of Bacteriology*, *178*(18), 5472–5479. https://doi.org/10.1128/jb.178.18.5472-5479.1996.

Jung, S.-J., Park, S. Y., Kim, S. E., Kang, I., Park, J., Lee, J [Jungwon], Kim, C.-M., Chung, M.-S., & Ha, S.-D. (2017). Bactericidal Effect of Calcium Oxide (Scallop-Shell Powder) Against Pseudomonas aeruginosa Biofilm on Quail Egg Shell, Stainless Steel, Plastic, and Rubber. *Journal of Food Science*, *82*(7), 1682–1687. https://doi.org/10.1111/1750-3841.13753.

Kampmann, Y., Clerck, E. de, Kohn, S., Patchala, D. K., Langerock, R., & Kreyenschmidt, J. (2008). Study on the antimicrobial effect of silver-containing inner liners in refrigerators. *Journal of Applied Microbiology*, *104*(6), 1808–1814. https://doi.org/10.1111/j.1365-2672.2008.03727.x.

Kovács, R. L., Daróczi, L., Barkóczy, P., Baradács, E., Bakonyi, E., Kovács, S., & Erdélyi, Z. (2021). Water vapor transmission properties of acrylic organic coatings. *Journal of Coatings Technology and Research*, *18*(2), 523–534. https://doi.org/10.1007/s11998-020-00421-5.

Krauklis, A. E., & Echtermeyer, A. T. (2018). Mechanism of Yellowing: Carbonyl Formation during Hygrothermal Aging in a Common Amine Epoxy. *Polymers*, *10*(9). https://doi.org/10.3390/polym10091017.

Kurinčič, M., Jeršek, B., Klančnik, A., Možina, S. S., Fink, R., Dražić, G., Raspor, P., & Bohinc, K. (2016). Effects of natural antimicrobials on bacterial cell hydrophobicity, adhesion, and zeta potential. *Arhiv Za Higijenu Rada I Toksikologiju*, *67*(1), 39–45. https://doi.org/10.1515/aiht-2016-67-2720.

Lavoine, N., Givord, C., Tabary, N., Desloges, I., Martel, B., & Bras, J. (2014). Elaboration of a new antibacterial bio-nano-material for food-packaging by synergistic action of cyclodextrin and microfibrillated cellulose. *Innovative Food Science & Emerging Technologies*, *26*, 330–340. https://doi.org/10.1016/j.ifset.2014.06.006.

Lee, J [Jaesung], Lee, Y.-H., Jones, K., Sharek, E., & Pascall, M. A. (2011). Antimicrobial packaging of raw beef, pork and turkey using silver-zeolite incorporated into the material. *International Journal of Food Science & Technology*, *46*(11), 2382–2386. https://doi.org/10.1111/j.1365-2621.2011.02760.x.

Lenoir, S., Pagnoulle, C., Galleni, M., Compère, P., Jérôme, R., & Detrembleur, C. (2006). Polyolefin matrixes with permanent antibacterial activity: Preparation, antibacterial activity, and action mode of the active species. *Biomacromolecules*, *7*(8), 2291–2296. https://doi.org/10.1021/bm050850c.

Liu, S., Graham, J. E., Bigelow, L., Morse, P. D., & Wilkinson, B. J. (2002). Identification of Listeria monocytogenes genes expressed in response to growth at low temperature. *Applied and Environmental Microbiology*, *68*(4), 1697–1705. https://doi.org/10.1128/AEM.68.4.1697-1705.2002.

Martínez-Abad, A., Lagaron, J. M., & Ocio, M. J. (2012). Development and characterization of silver-based antimicrobial ethylene-vinyl alcohol copolymer (EVOH) films for food-packaging applications. *Journal of Agricultural and Food Chemistry*, *60*(21), 5350–5359. https://doi.org/10.1021/jf300334z.

Møretrø, T., Høiby-Pettersen, G. S., Halvorsen, C. K., & Langsrud, S. (2012). Antibacterial activity of cutting boards containing silver. *Food Control*, *28*(1), 118–121. https://doi.org/10.1016/j.foodcont.2012.05.007.

Noyce, J. O., Michels, H., & Keevil, C. W. (2006). Use of copper cast alloys to control Escherichia coli O157 cross-contamination during food processing. *Applied and Environmental Microbiology*, *72*(6), 4239–4244. https://doi.org/10.1128/AEM.02532-05.

Perrin, F. X., Merlatti, C., Aragon, E., & Margaillan, A. (2009). Degradation study of polymer coating: Improvement in coating weatherability testing and coating failure prediction. *Progress in Organic Coatings*, *64*(4), 466–473. https://doi.org/10.1016/j.porgcoat.2008.08.015.

Potter, R., Truelstrup Hansen, L., & Gill, T. A. (2005). Inhibition of foodborne bacteria by native and modified protamine: Importance of electrostatic interactions. *International Journal of Food Microbiology*, *103*(1), 23–34. https://doi.org/10.1016/j.ijfoodmicro.2004.12.019.

Qu, L., Chen, G., Dong, S., Huo, Y., Yin, Z., Li, S., & Chen, Y. (2019). Improved mechanical and antimicrobial properties of zein/chitosan films by adding highly dispersed nano-TiO2. *Industrial Crops and Products*, *130*, 450–458. https://doi.org/10.1016/j.indcrop.2018.12.093.

Quintavalla, S., & Vicini, L. (2002). Antimicrobial food packaging in meat industry. *Meat Science*, *62*, 373–380.

Raeissi, B., Bashir, M. A., Garrett, J. L., Orlandic, M., Johansen, T. A., & Skramstad, T. (2021). Detection of different chemical binders in coatings using hyperspectral imaging. *Journal of Coatings Technology and Research.* Advance online publication. https://doi.org/10.1007/s11998-021-00544-3.

Ranganayaki, T., Venkatachalam, M., Vasuki, T., & Shankar, S. (2014). Preparation and Characterization of Nanocrystalline TiO2 Thin Films Prepared By Sol-Gel Spin Coating Method. *International Journal of Innovative Research in Science Engineering and Technology*, *3*(10). https://doi.org/10.15680/IJIRSET.2014.03100042.

Regulation (EC) No 1935/2004 of the European Parliament and of the Council of 27 October 2004 on materials and articles intended to come into contact with food and repealing Directives 80/590/EEC and 89/109/EEC, Official Journal of the European Union, 338, 4-17.

Roy, D., Knapp, J. S., Guthrie, J. T., & Perrier, S. (2008). Antibacterial cellulose fiber via RAFT surface graft polymerization. *Biomacromolecules*, *9*(1), 91–99. https://doi.org/10.1021/bm700849j.

Russell, A. D., & Hugo, W. B. (1994). 7 Antimicrobial Activity and Action of Silver. In *Progress in Medicinal Chemistry* (Vol. 31, pp. 351–370). Elsevier. https://doi.org/10.1016/S0079-6468(08)70024-9.

Sahalan, A. Z., Abdul, A. H., Hing, H. L., & Ghani, M. A. (2013). Divalent CATIONS (Mg2+, Ca2+) protect bacterial outer membrane damage by polymyxin B. *Sains Malaysiana*, *42*(3), 301–306.

Seyfriedsberger, G., Rametsteiner, K., & Kern, W. (2006). Polyethylene compounds with antimicrobial surface properties. *European Polymer Journal*, *42*(12), 3383–3389. https://doi.org/10.1016/j.eurpolymj.2006.07.026.

Shahidi, F., Arachchi, J. K. V., & Jeon, Y.-J. (1999). Food applications of chitin and chitosans. *Trends in Food Science & Technology*, *10*, 37–51.

Siedenbiedel, F., & Tiller, J. C. (2012). Antimicrobial Polymers in Solution and on Surfaces: Overview and Functional Principles. *Polymers*, *4*(1), 46–71. https://doi.org/10.3390/polym4010046.

Suresh, J., Pradheesh, G., Alexramani, V., Sundrarajan, M., & Hong, S. I. (2018). Green synthesis and characterization of hexagonal shaped MgO nanoparticles using insulin plant (Costus pictus D. Don) leave extract and its antimicrobial as well as anticancer activity. *Advanced Powder Technology*, *29*(7), 1685–1694. https://doi.org/10.1016/j.apt.2018.04.003.

Yamamoto, O. (2001). Influence of particle size on the antibacterial activity of zinc oxide. *International Journal of Inorganic Materials*, *3*(7), 643–646. https://doi.org/10.1016/S1466-6049(01)00197-0.

Yao, K. S., Wang, D. Y., Ho, W. Y., Yan, J. J., & Tzeng, K. C. (2007). Photocatalytic bactericidal effect of TiO2 thin film on plant pathogens. *Surface and Coatings Technology*, *201*(15), 6886–6888. https://doi.org/10.1016/j.surfcoat.2006.09.068.

Yu, B., Leung, K. M., Guo, Q., Lau, W. M., & Yang, J. (2011). Synthesis of Ag-TiO2 composite nano thin film for antimicrobial application. *Nanotechnology*, *22*, Article 115603. https://doi.org/10.1088/0957-4484/22/11/115603.

Zubielewicz, M., & Królikowska, A. (2009). The influence of ageing of epoxy coatings on adhesion of polyurethane topcoats and protective properties of coating systems. *Progress in Organic Coatings*, *66*(2), 129–136. https://doi.org/10.1016/j.porgcoat.2009.06.014.

6 General conclusion

As customers' preferences shifted to high-quality and safe products with enhanced shelf life, the development of various new trends in packaging systems has risen. An appropriate food packaging solution is a key factor related to food safety and the high-quality properties of fresh produce, as well as reducing food waste prior to final consumption. Food waste describes an increasing concern along the food supply chain, which holds all stakeholders involved accountable. If food waste occurs, the negative overall environmental impact rises with every step in the supply chain due to more used resources. Fossil-based multi-layer packaging offers effective and well-optimized solutions due to mechanical, thermal, and physical properties that are easily adjustable to a broad variety of distinctive food types. Despite these advantages, its use has to be reduced from an environmental, sustainable, and biocompatible perspective. Precisely, there are increasing issues concerning the harm caused to the environment, mainly due to the manufacturing phase, problematic end-of-life strategies, and adverse effects on human health.

Unique strategies like reducing packaging thickness and/or the number of layers, using easily recyclable materials, and applying bio-based and/or biodegradable materials are pursued in the development and implementation of sustainable packaging materials. Although the food industry has shown a broad interest in replacing fossil-based plastics, their use in industrial applications is often restricted due to poor commercial availability, lack of efficient production processes, and lower performance in fundamental packaging functions. Satisfactory packaging characteristics, especially moisture and oxygen barriers, are crucial to preserve product quality and safety, ensuring shelf life and preventing food waste. There are several developments for bio-based materials, but up to now, widespread use in the market is still missing.

Thus, the main objective of this thesis was the assessment of different innovative packaging strategies to maintain product characteristics and quality of different fresh produce, and thus to keep and even improve the resource efficiency of food processing and the packaging material itself. The innovative packaging strategies ranged from bio-based to more recyclable fossil-based packaging materials for use in supply chains of fresh produce. Furthermore, the potential and antimicrobial effectiveness of a novel antimicrobial active compound applied as a coating for active packaging materials was assessed.

The first research question aimed to provide an overview of the status quo on research and practical use of bio-based packaging materials produced from renewable resources and biomass waste. The applicability of bio-based polymers was considered focusing on the relationship between packaging and food preservation. Additionally, the actual use of plant-derived additives in combination with bio-based packaging was screened as active packaging can improve (fundamental) packaging functions with a direct influence on food quality and shelf life.

Based on the literature review it became obvious that, so far, the use of bio-based polymers in industrial applications is often restricted due to lower performance in fundamental packaging functions that directly influence food quality and safety, the length of shelf life, and thus the amount of food waste. The *proof-of-concept* with food applications is shown by a few commercially available biopolymers, such as PLA, PHAs, PEF, PBS, and thermoplastic cellulose or starch-based films. Additives are often critical in achieving the desired properties in packaging materials and can be capable of prolonging the shelf life of both packaging materials and packed goods. Several studies showed that plant essential oils and plant extracts are interesting components for active food packaging due to their antimicrobial, antioxidant, and photo absorbing effects. Additionally, their incorporation typically improves packaging characteristics, especially the mechanical properties of packaging materials. In the future, the market for active packaging will certainly increase due to enhanced efforts and innovations in material development and processing technologies.

For the second research question, an investigation of the effect of different bio-based packaging materials on the quality loss and microbial spoilage of fresh cherry tomatoes was performed. Since the shelf life of cherry tomatoes is very limited mainly due to softening and decay incidence, an appropriate packaging is essential regarding food safety and the high-quality properties of fresh produce as well as reducing food waste. The influence of bio-based packaging materials from renewable resources comprising groundwood pulp, sugar cane, bamboo, cellulose, grass paper, and polylactic acid on quality parameters and microbial spoilage was investigated. Storage trials with fresh cherry tomatoes were performed under dynamic temperature conditions typical in fruit and vegetable supply chains. During storage, the influence on typical quality parameters like decay incidence and weight loss, microbiological spoilage, sensory attributes, as well as the color and texture of cherry tomatoes, was analyzed and assessed. Furthermore, the packaging materials themselves were investigated for surface bacterial counts, followed by general packaging parameters like tensile strength, elongation at break and water absorption capacity testing.

The results show that the above-mentioned bio-based packaging materials themselves have an influence on quality parameters associated with quality changes due to their capacity for moisture absorption and their influence on the exchange and alignment of surrounding conditions. It became evident that groundwood pulp and sugar cane showed higher preserving characteristics of fresh cherry tomatoes compared to the reference materials, whereas in cellulose and PLA packaging, cherry tomatoes reach a level of unacceptable product quality after the shortest storage period. Especially the parameters of weight loss, decay incidence, and microbial contamination of tomatoes packaged in trays made of groundwood pulp and sugar cane point to the advantage of using this kind of bio-based packaging material, even from an economical and sustainable point of view due to increasing prices of fossil-based resources (oil) and packaging waste reduction. The results of the packaging parameters showed the highest tensile strength for cellulose (thinnest material), and the lowest was recorded for groundwood pulp (thickest material). The water absorption capacity showed a wide range, with corrugated cardboard exhibiting, by far, the highest value and bamboo and cellulose the lowest.

For answering the third research question, the ability of different innovative packaging materials to preserve and maintain quality and shelf life of emulsion-type sausages was evaluated compared to a conventional used multi-layer APET/PE multi-layer. During storage, microbiological parameters (total viable count, lactic acid bacteria, Enterobacteriaceae, and yeasts and molds) were analyzed, and instrumental color measurements were investigated. Additionally, the gas composition in the packages was analyzed to identify the gas barrier properties of the packaging materials and the percentage of defective packages.

The study revealed that the reference multi-layer APET/PE 300µ/62µ, and the multi-layer APET/PE 230µ/62µ were the most effective in keeping the quality of ma-packed emulsion-type sausage and preserved an acceptable product quality during shelf life. The growth behavior of several microorganisms varied in the innovative tested packaging materials compared to emulsion-type sausage stored in the reference multi-layer APET/PE 300µ/62µ. The decline of product quality varied between the different materials, with no correlation between microbial spoilage and a decrease in product quality for emulsion-type sausage packaged in the various tested innovative packaging materials. Considering the percentage of intact packages as a critical factor for the machine applicability and the gaseous atmosphere-keeping property, it becomes evident that the use of a mono-layer Mono APET 200µ/52µ without any EVOH layer, a under foil reduced in thickness to 230µ and a paper-PE compound led to an increase in O_2 concentration compared to the reference multi-layer APET/PE 300µ/62µ. It indicates that sufficient barrier properties of both under and upper foil are crucial keeping the quality of emulsion-type sausage during storage. Introducing new packaging materials remains highly case-dependent, but as long as food quality is preserved, these represent a promising alternative for replacing conventionally used multi-layer packaging. Especially in certain product areas where short residual terms are enough, the use can be favored, even in terms of sustainability.

The final research question aimed at the determination of different products, processes, and environmental influence factors, focusing on the antimicrobial activity of several surface coatings without preservatives, biocides, or any similar compound prepared by new binders' treatments, which use functional extenders. Several trial setups were conducted to elucidate the research question. The influence of: selected specific gram-positive and gram-negative organisms, contact time, initial bacteria concentration, temperature, pH value, and the presence of food components on the antimicrobial activity were analyzed by using a test method according to ISO 22196:2007.

The results allowed an evaluation of the potential use of the investigated coatings as packaging coating for active packaging materials.

In general, the results showed a high antimicrobial activity of the coatings based on silicate, non-conserved acrylates, and styrene acrylates under several test conditions. Thus, these coatings bear a high potential for application as packaging material or surface coating in the food industry to increase the safety and quality of perishable products. Overall, no difference in the sensitivity of gram-positive and gram-negative bacteria was observed. All tested surface coatings based on silicate, non-conserved acrylates, and styrene acrylates showed high antimicrobial activities under standard test conditions. In addition, a high initial bacteria concentration for incubation leads to a reduction in bacterial count. Incubation at low temperatures (120 h, 7°C) resulted in a decreased reduction rate for *Staph. aureus* for most of the samples. Furthermore, antimicrobial activity was shown for most of the samples in the presence of the food components starch, meat extract, and oleic acid, as well as when inoculated with adjusted pH values (pH 5, 7, 9). It is expected that these coatings can be applied to different standards, multilayer, and packaging polymers, generating an antimicrobial property of these materials to increase the advantageous characteristics of multilayer foils with high barriers for water and gases, as well as increased mechanical strength.

As a concluding remark, replacing fossil-based packaging materials for fresh produce with innovative packaging systems needs to evaluate and consider a complexity of different factors influencing food safety and high-quality properties of fresh produce, as well as reducing food waste. The implementation of packaging materials that appear to be more sustainable at first glance is recommended if the environmental impact of the entire supply chain has been considered. As the environmental impact of producing the food itself is much higher than that of the packaging, it is crucial for the packaging to maintain product quality and guarantee shelf life to avoid any food waste. According to the results of this thesis, the tested approaches of innovative packaging materials absolutely bear advantages to be used for the packaging of fresh produce. But with every application, the interaction between food, packaging, and environmental factors is crucial for the effects on quality and shelf life. Prior to the implementation of new packaging materials, it is advisable to conduct application-related durability tests to check whether packaging solutions made of innovative materials can guarantee the quality and safety of food. And thus, represent packaging that is equivalent to commercial packaging.

Bio-based materials do not only provide significant opportunities in terms of economics, sustainability, and biocompatibility. A broad interest in the food industry, and the *proof-of-concept*, is shown by a few commercially available biopolymers with food applications such as PLA, PHAs, PEF, PBS, and thermoplastic cellulose or starch-based films. However, their use in industrial applications is often restricted due to poor commercial availability, lack of efficient production, recycling and disposal processes, lower performance in machine applicability, and fundamental packaging functions. Research needs to focus on improving the characteristics of bio-based packaging materials like mechanical, thermal, and physical properties. Approaches to increase packaging functions cover active coatings in combination with bio-based materials as they might have a significant effect on the adsorption of ethylene and water vapor. Organic coatings have the potential to be applied to different standards, packaging polymers, generating an antimicrobial property of these materials to increase the advantageous characteristics of multi-layer foils with high barriers for water and gases as well as increased mechanical strength. Besides their antimicrobial properties, coatings attract attention as coatings for organic packaging materials like wood and paper to improve their physical properties. Due to enhanced efforts and innovations in material development and processing technologies, the market for active packaging will certainly increase in the foreseeable future. Contrasting to the incorporation of antimicrobial active polymers as compounds, the coating of foils with antimicrobial polymers or substances turns out to be a promising approach producing active, antimicrobial packaging. Challenges such as the adhesion of coatings to the substrate need to be overcome. Furthermore, an interesting point for further studies on the packaging of fruits and vegetables is the replacement of LDPE film with a PLA or cellulose-based film to make the packaging even more sustainable, as it was shown that the use of sealed plastic films positively influences the product quality compared to unwrapped stored fruits.

In addition to the points already summarized, end-of-life issues of packaging materials need to be emphasized when assessing their sustainability. Safe disposal and recycling of materials often remain challenging. Except for drop-in solutions, the disposal and recycling of several bio-based materials often remain challenging. One of the main reasons is that investments are required in the overall disposal, including steps of collection, separation, composting, and recycling of upcoming new materials, which, however, are associated with high costs. Due to a still modest market share of such materials, appropriate investments in waste management systems are often not made yet. So, further innovations in both recyclable packaging designs and corresponding cost-effective technologies are needed.

A Appendix

A.1 Appendix for chapter 3

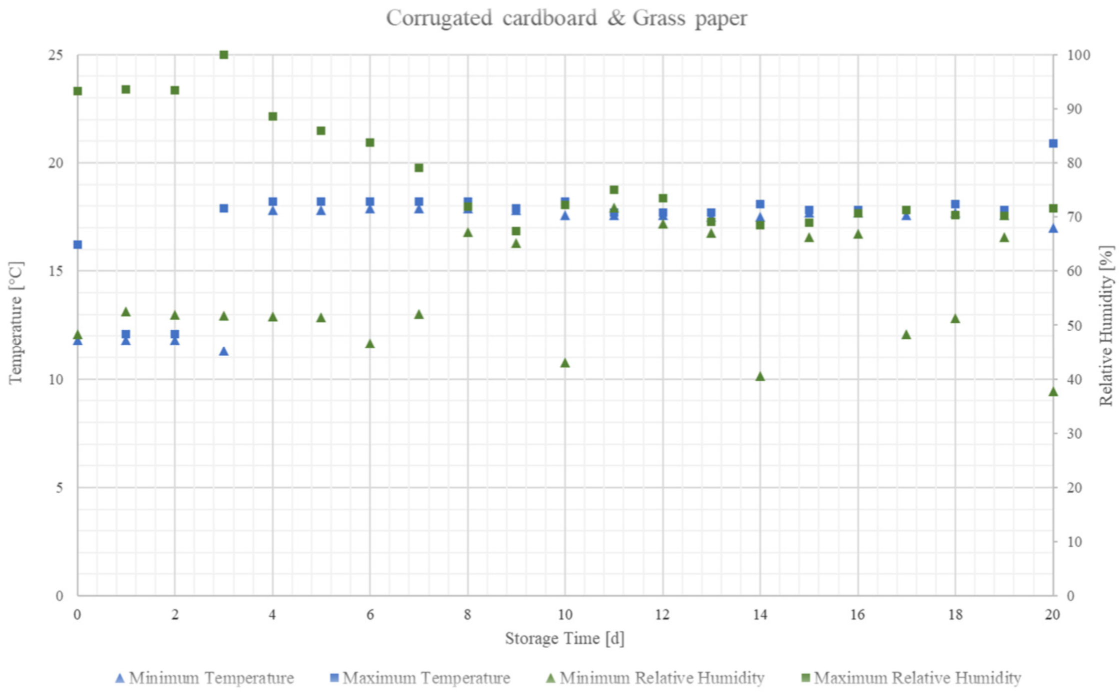


Figure A.1 Recorded temperature and relative humidity, showing minimum and maximum values of each storage day, during the storage trial with the packaging trays of corrugated cardboard and grass paper.

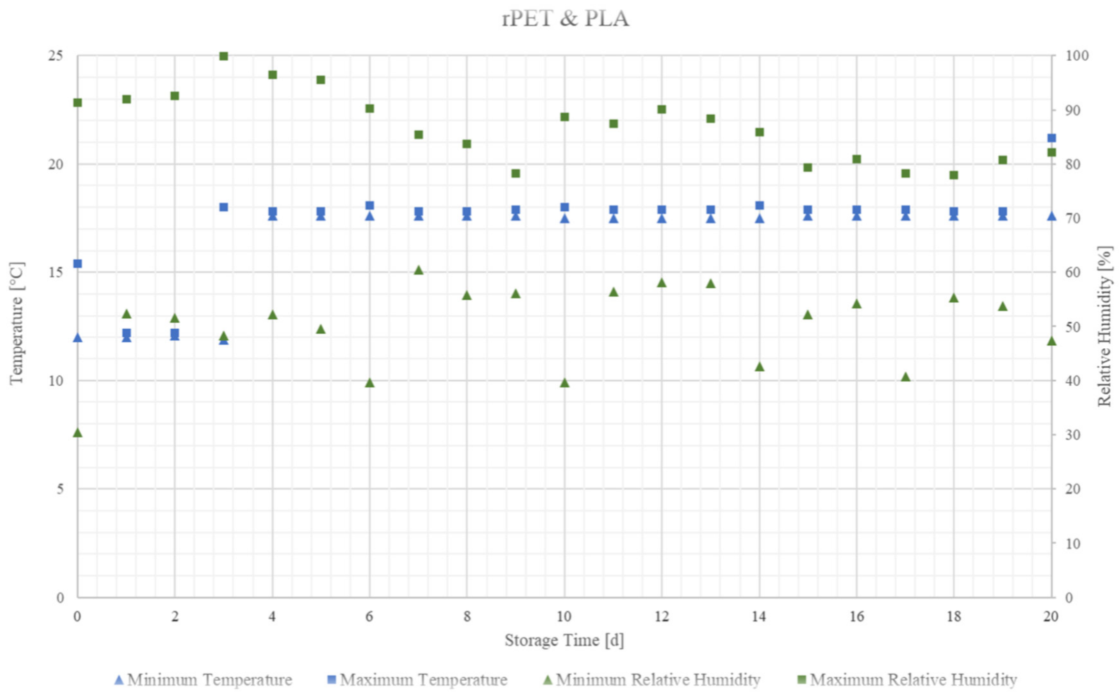


Figure A.2 Recorded temperature and relative humidity, showing minimum and maximum values of each storage day, during the storage trial with the packaging trays of rPET and PLA.

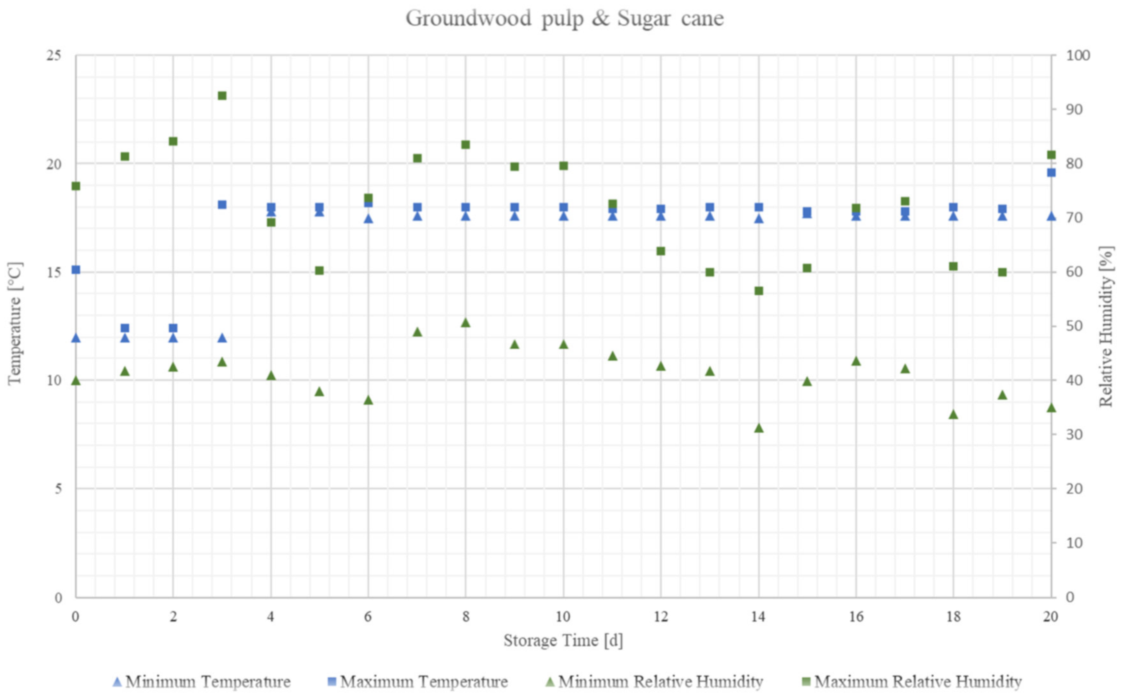


Figure A.3 Recorded temperature and relative humidity, showing minimum and maximum values of each storage day, during the storage trial with the packaging trays of groundwood pulp and sugar cane.

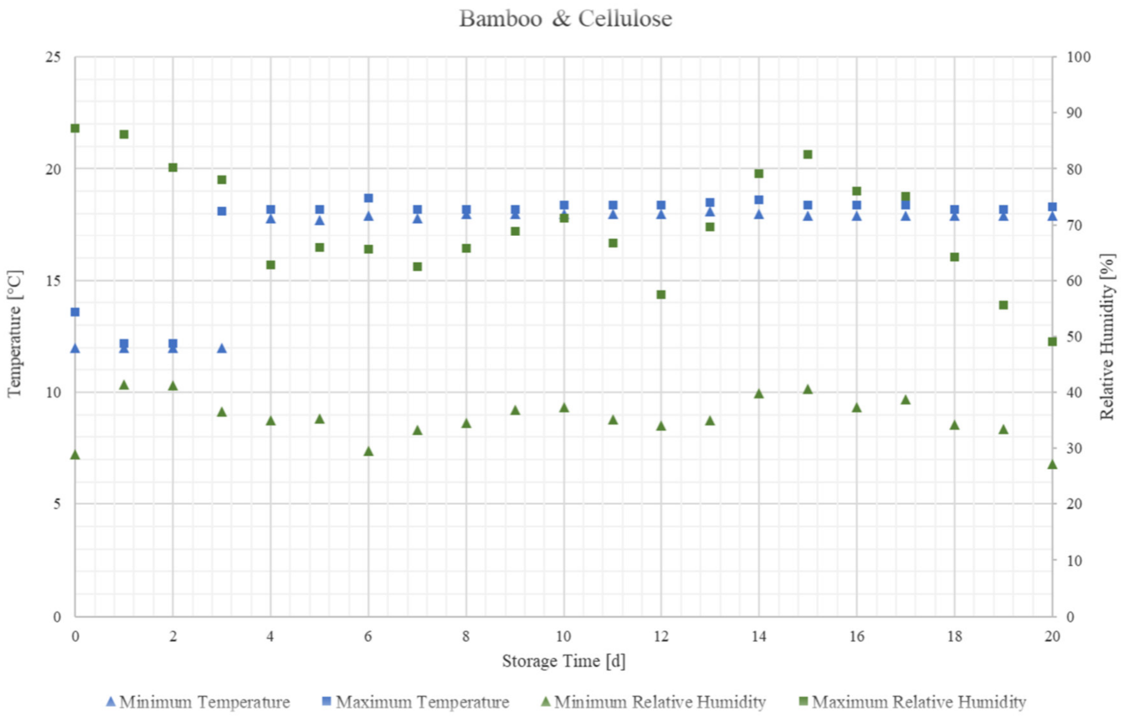


Figure A.4 Recorded temperature and relative humidity, showing minimum and maximum values of each storage day, during the storage trial with the packaging trays of bamboo and cellulose.

List of Figures

List of Tables

List of Publications

2023

Korte, I., Albrecht, A., Mittler, M., Waldhans, C., & Kreyenschmidt, J. (2023). Influence of different bio-based and conventional packaging trays on the quality loss of fresh cherry tomatoes during distribution and storage. *Packaging Technology and Science*, *36*(7), 569–583. https://doi.org/10.1002/pts.2728.

Korte, I., Albrecht, A., Mittler, M., Waldhans, C., & Kreyenschmidt, J. (2023). Quality impact of sustainable ma-packaging options for emulsion-type sausage: A German case study. *Future Foods*, *7*, 100218. https://doi.org/10.1016/j.fufo.2023.100218.

Korte, I., Petry, M., & Kreyenschmidt, J. (2023). Antimicrobial activity of different coatings for packaging materials containing functional extenders against selected microorganisms typical for food. *Food Control*, *148*, 109669. https://doi.org/10.1016/j.foodcont.2023.109669.

2022

Korte, I., Kreyenschmidt, J., Wensing, J., Bröring, S., Frase, J. N., Pude, R., Rumpf, J., Havelt, T., Schmitz, M., & Schulze, M. (2022): Nachhaltige Verpackungen – Quo Vadis. In: Gerhardt, P., Daldrup, J., Eppler, U. (Hg.): Bioökonomie im Lichte der Nachhaltigkeit. Tagungsdokumentation. BfN-Skripten 629. Bundesamt für Naturschutz. Bonn 2022. S. 65-67. doi.org/10.19217/skr629.

2021

Korte, I., Kreyenschmidt, J., Wensing, J., Bröring, S., Frase, J. N., Pude, R., Konow, C., Havelt, T., Rumpf, J., Schmitz, M., & Schulze, M. (2021). Can Sustainable Packaging Help to Reduce Food Waste? A Status Quo Focusing Plant-Derived Polymers and Additives. *Applied Sciences*, *11*(11), 5307. https://doi.org/10.3390/app11115307.

2020

Korte, I., Waldhans, C., Mittler, M., Albrecht, A., & Kreyenschmidt, J. (2020): Comparing the applicability of sustainable packaging materials for meat products and their impact on organoleptic and microbial product quality during stroage. Oral Presentation & Abstract. 34th EFFoST International Conference. 10th – 12th November 2020. Online Event.

Waldhans, C., Albrecht, A., Mittler, M., **Korte, I.**, & Kreyenschmidt, J. (2020): Development of a predictive shelf life model for raw pork sausage to increase sustainability and resource efficiency along the supply chain. Poster Presentation. 34th EFFoST International Conference. 10th - 12th November 2020. Online Event.

Rumpf, J., **Korte, I.**, Kreyenschmidt, J., Dreier, T., & Schulze, M. (2020): Novel cellulose-beesswax-lignin composites as coating material for paper packaging. Conference Paper. European Technical Coatings Congress (ETCC). September 2020. Krakow, Poland.

2019

Klein, S. E., Alzagameem, A., Rumpf, J., **Korte, I.**, Kreyenschmidt, J., & Schulze, M. (2019). Antimicrobial Activity of Lignin-Derived Polyurethane Coatings Prepared from Unmodified and Demethylated Lignins. *Coatings*, *9*(8), 494. https://doi.org/10.3390/coatings9080494.

Havelt, T., Brettschneider, S., Do, X. T., **Korte, I.**, Kreyenschmidt, J., & Schmitz, M. (2019). Sustainable Extraction and Characterisation of Bioactive Compounds from Horse Chestnut Seed Coats for the Development of Bio-Based Additives. *Resources*, *8*(2), 114. https://doi.org/10.3390/resources8020114.

Alzagameem, A., Klein, S. E., Bergs, M., Do, X. T., **Korte, I.**, Dohlen, S., Hüwe, C., Kreyenschmidt, J., Kamm, B., Larkins, M., & Schulze, M. (2019). Antimicrobial Activity of Lignin and Lignin-Derived Cellulose and Chitosan Composites Against Selected Pathogenic and Spoilage Microorganisms. *Polymers*, *11*(4). https://doi.org/10.3390/polym11040670.

Albrecht, A., Hebel, M., Herbert, U., Waldhans, C., **Korte, I.**, Dohlen, S., & Kreyenschmidt, J. (2019): Practical application of predictive models for meat products – challenges and opportunities. Presentation. 11th International Conference on Predictive Modelling in Food. ICPMF, 17.-20.09.2019. Braganca, Portugal.

Hebel, M., Albrecht, A., Breuch, R., Herbert, U., **Korte, I.**, Valler, O., Wörle, V., & Kreyenschmidt, J. (2019): Combining predictive shelf life models with rapid methods in meat supply chains. Poster. 11th International Conference on Predictive Modelling in Food. ICPMF, 17.-20.09.2019. Braganca, Portugal.

Acknowledgement

This doctoral thesis was financially supported by the European Union via the European Regional Development Fund EFRE.NRW (Grant EFRE 0500035, “Biobasierte Produkte”).

Danksagung

An dieser Stelle möchte ich mich bei denen bedanken, die mich während meiner Promotionszeit unterstützt haben und so zum Gelingen dieser Arbeit beigetragen haben.

Zunächst möchte ich Prof. Dr. Judith Kreyenschmidt, meiner Doktormutter, danken, die mir die Möglichkeit gegeben hat, in dem spannenden Themengebiet nachhaltiger Verpackungsmaterialien zu promovieren. Sie hat mich in meinem Vorhaben jederzeit mit wertvollen Ratschlägen und konstruktiver Kritik unterstützt.

Ein besonderer Dank gilt Prof. Dr. Ralf Pude für die Übernahme des Korreferats sowie die gute Zusammenarbeit im Rahmen des Forschungsprojektes „Biobasierte Verpackungen“.

Ich bedanke mich außerdem bei Prof. Dr. Margit Schulze für die Übernahme des fachnahen Mitglieds und ihre Hilfe und Unterstützung während der Zusammenarbeit im Forschungsprojekt „Biobasierte Verpackungen“.

Ein besonderer Dank gilt meinen ehemaligen Kolleginnen und Kollegen der CCM-Arbeitsgruppe, die mir während meiner Promotionszeit mit Rat und Tat zur Seite standen. Besonders danke ich Claudia und Maureen für ihre tatkräftige Unterstützung an langen Labortagen, für konstruktives Feedback in jeglichen Schreibprozessen, besonders im Endspurt meiner Arbeit, und für die engen Freundschaften, die sich während der gemeinsamen Promotionszeit entwickelt haben. Ich bedanke mich bei Michael für eine unvergessliche Zeit auf Station und viele Ratschläge gerade in schwierigen Zeiten. Antonia danke ich für den fachlichen Input, ganz besonders bei der Datenauswertung für alle Manuskripte. Bei Martin bedanke ich mich für seine ruhige und gelassene Art und die dadurch sehr entspannte Arbeitsatmosphäre. Barbara danke ich für den guten fachlichen Austausch bei der Bearbeitung jeglicher Projektaufgaben und Lucas danke ich ganz besonders für die sprachliche Korrektur aller Manuskripte.

Ein großer Dank geht auch an alle meine ehemaligen Studentinnen und Studenten. Darüber hinaus bedanke ich mich bei den ehemaligen Kolleginnen und Kollegen im EFRE-Forschungsprojekt „Biobasierte Produkte“ für die gute und produktive Zusammenarbeit, insbesondere bei bei Jessica Rumpf, Joana Wensing, Jan Niklas Frase und Thomas Havelt.

Nicht zuletzt möchte ich allen Kolleginnen und Kollegen aus der Conditorei Coppenrath & Wiese für ihr offenes Ohr, ihre Zuversicht und ihren Zuspruch in der intensiven Schreibphase dieser Doktorarbeit danken.

Ein ganz besonderer Dank gilt meinen Freundinnen und Freunden, die mich in den letzten Jahren - ganz oder teilweise auf einem Weg mit vielen Höhen und Tiefen - begleitet, motiviert und unterstützt haben und einfach immer für mich da waren.

Meiner Familie, vor allem meinen Eltern und meinem Bruder, danke ich aus tiefstem Herzen für ihre bedingungslose Unterstützung, Motivation in nervenraubenden Zeiten und den unermüdlichen Glauben an mich das Projekt Doktorarbeit erfolgreich zu beenden.

Ohne jeden einzelnen von euch wäre ich nie so weit gekommen. Danke.