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Advances in barrier coatings and film technologies for achieving sustainable packaging of food products – A review

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ABSTRACT

Background: The technology of food packaging is responding to significant market dynamics such as the rapid growth in e-commerce and preservation of fresh food, a sector that accounts for over 40% of plastic waste. Further, mandates for sustainability and recent changes in national governmental policies and regulations that include banning single-use plastic products as observed in sweeping reforms in Europe, Asia, and several US States are forcing industries and consumers to find alternative solutions.

Scope and approach: This review highlights an ongoing shift of barrier coatings from traditional synthetic polymers to sustainable breakthrough materials for paper-based packaging and films. Advantages, challenges and adapting feasibility of these materials are described, highlighting the implications of selecting different materials and processing options. A brief description on progress in methods of coating technologies is also included. Finally, the end fate of the barrier materials is classified depending on the packaging type, coating materials used and sorting facility availability.

Key findings and conclusions: Different types of coatings, such as water-based biopolymers, due to their greater environmental compatibility, are making inroads into more traditional petroleum-based wax and plastic laminate paperboard products for fresh food bakery, frozen food, and take-out containers applications. In addition, nano-biocomposites have been studied at an accelerating pace for developing active and smart packaging. Based on the momentum of recent developments, a strong pace of continuing developments in the field can be expected.

1. Introduction

Food packaging is an essential element to address the key challenge of sustainable food consumption (Coussy et al., 2013; Licciardello, 2017) and has been considered as an added value for waste reduction rather than an additional economic and environmental cost (Matar et al., 2018; Verghese et al., 2015; Wikström & Williams, 2010). Approximately 55% of paper used globally is for packaging, an application area consistently increasing recently from attempts to minimize the use of single-use plastics (Haggith et al., 2018). Paper and paperboard materials exhibit a wide range of thickness, basis weight and fiber quality, which determine the shape stability and mechanical strength. For instance, paperboards are made to provide good mechanical properties such as tensile strength, folding resistance, compression strength, resistance to cracking, bending resistance, etc. These properties can improve the package shape and stability of paper and plastic composites or multilayer materials (Hubbe, 2014). The food and beverage segment accounted for the highest Paper and Paperboard Packaging market share in 2018 (Polaris Market Research, 2018). Coating technology with paper has been proposed as an additional strategy for accomplishing a more rational use of the materials within the food packaging sector. According to the food packaging optimization concept, the use of multifunctional thin layers allows for the replacement of multilayer and heavily structured food packaging materials such as glass, tin, or other metals; the amount of packaging materials is thereby reduced, while meeting the specifications for the functional properties of the final package to pursue the goal of overall shelf life extension of food (Nakaya et al., 2015). Currently, the increasing requirements among consumers for convenience, smaller package sizes, and for minimally processed, fresh, and healthy foods have imposed the necessity of designing highly sophisticated and engineered coatings (Aulin & Ström, 2013; Bobu et al., 2016; Tyagi et al., 2018a).

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Nanotechnology has paved the way for the development of new architectures and unique patterns that eventually have yielded nanostructured and nanocomposite coatings with outstanding performances (Ansari et al., 2018; Sharma et al., 2017). From a sustainability perspective, the range of paper and paperboard products has expanded greatly through the rational use of these coating technologies. To render technological functionalities such as 'gas and moisture permeation resistance, hydrophobicity, antimicrobial protection, scratch resistance, and cohesive strength', the paper-based surfaces of packaging are coated or treated with several conventional and non-conventional coating materials (Karli et al., 2013).

Barrier coatings for food packaging help keep moisture and oils inside of the packaging or keep moisture or oxygen out of the packaging. This is achieved by increasing mean-free-path for the molecules of the respective gases, moisture, or oil by increasing tortuosity (path of resistance in a porous matrix), depending on the type of application. In addition, barrier layers are formulated to have suitably low solubility characteristics relative to one or more permeant of interest. For instance, the coating on a bakery box would have as its primary function the containment of oil and grease inside the box, while the coating on a paper cup keeps fluids inside. The barrier properties of papers are commonly controlled by the application of conventional petroleum-based derivatives such as polyethylene, polyvinyl chloride, polypropylene, polystyrene, waxes and/or fluorine-based derivatives as coatings. While surface hydrophobicity is improved by employing these petro-based polymers, they have become disfavored due to environmental limitations in fossil-oil resources, poor recyclability, and environmental concerns related to the generation of waste due to their very slow biodegradation that can take up to 500 years (Ioakeimidis et al., 2016; Leblanc, 2018; North & Halden, 2013).

Biopolymers including polysaccharides, proteins, and polyhydroxyalkanoates (PHAs) can be used to formulate new pathways for assembly of fully bio-based and functional paper coatings for food packaging. However, difficulties associated in the processing of most biopolymers in their pure form may arise from hydrophilicity, crystallization behavior, brittleness or melt instabilities, sealability, etc., that hinder full exploitation at industrial scale (Rastogi & Samyn, 2015). As an option to enhance the behavior of biopolymers, certain classes of nanomaterials (NPs) such as metals (Ag, Au, Cu), oxides (ZnO), nano-clays (NCs) and nano-emulsions (NEs) have numerous applications to add value to the manufacturing of active food packaging (Ranjan et al., 2014). Nanocomposites, a fusion of traditional food packaging materials with nanoparticles, are gaining interest in the food packaging sector because in addition to their remarkable antimicrobial spectrum of activity, they display great mechanical performance and strong resistance characteristics to external influences (e.g., heat, pressure, pH, salinity, etc.) (Montazer & Harifi, 2017). Nano-sized phases in nanocomposites coatings augment the physical and mechanical properties of the polymer by transferring elastic strain to the nano-reinforced materials, when load is applied (Othman, 2014). Besides improving mechanical properties, nanoparticles also can improve barrier characteristics and tag-on active or smart properties to the packaging system, as discussed in detail in a later section (Duncan, 2014).

In recent years, bioplastics including biodegradable and non-biodegradable bio-based materials have also been widely developed into food packaging applications that have traditionally used conventional plastic processing. Such efforts have been motivated by environmental awareness and the implementation of stringent environmental regulations as a part of replacement of single-use plastics (Jariyasakoolroj & Harnkarnsujarit, 2018; Salem et al., 2020; Souza & Unnikrishnan, 2018). Single-use plastics, which contribute to almost 50% of the waste plastics, are a serious threat to the environment, as only less than 10% of those can be recycled, and the rest are discarded in landfills or incinerated (Geyer et al., 2017), or accumulate as litter. The lack of recycling of the single-use plastics and their non-biodegradable characteristics facilitates these plastics to enter aquatic ecosystems, which have raised wildlife mortalities from ingestion and entanglement (Hale et al., 2020). Moreover, the minute plastic particles can be taken up by different species of the food chain, which is a growing concern for natural habitats and human health (North & Halden, 2013). Thus, the use of biodegradable materials and recycled bioplastics as alternatives to the single-use plastics for food packaging is a preferred solution because bioplastics have included new modalities in packaging such as controlled release (desorption of active ingredients), scavenging (absorption of unwanted materials), barrier (e.g., gas and liquids) technologies, and biodegradability and nontoxicity (Rastogi & Samyn, 2015). Active ingredients derived from organic and inorganic materials can be incorporated into the food packaging in the forms of dispersion, lamination, and coating to improve the functional and barrier properties discussed in detail in a later section (Abdollahi et al., 2013; Rastogi & Samyn, 2015). Other treatments such as cold plasma have also been deployed for rendering disinfection to food packaging (Kuzminova et al., 2013; Pahwa & Kumar, 2018; Pankaj, 2018; Pankaj et al., 2014).

This review is a focused compilation of the state-of-the-art technologies used for barrier coatings to achieve varying modalities such as gas resistance, water resistance, and oil and grease resistance properties for paper-based food packaging. Detailed developments in petro- and bio-based barrier coating materials, coating application methods and coated substrate converting processes are discussed. A shifting focus of food industries on bio-based sustainable packaging materials and their biodegradability, is also described in a later section.

2. Functonal barrier properties and shelf life

2.1. Barrier properties

A barrier coating or film applied on paper-based food packaging should prevent the penetration of specific gases, aromas/odors, moisture vapor, water, oil and grease that could compromise the sensory and hygienic integrity of the packed food product. Gases, especially oxygen, can lead to discoloration, off-flavors and alteration of texture of food due to oxidation or rancidity of unsaturated fats (H. Zhang, Bhunia, et al., 2016). Defining the lower limit of gas permeability of a packaging used to protect food is very subjective and more likely depends on the shelf-life required for the food. For instance, Zhang et al. modeled the shelf life of microwavable packed mashed potato in polymeric pouches for different gas barrier properties (Zhang et al., 2016b; 2016a). Due to the non-polarity of oils, barrier properties against oil should demand similar surface characteristics as for oxygen barrier films, except that the state of the fluid is different (Hirschfelder et al., 1948). A number of factors such as crystallinity, brittleness and surface properties account for the oil and grease barrier properties of coating films (Alkhadra et al., 2017; Kim et al., 2007). One way to increase the oil and grease resistance of ordinary paper is to apply a petro-based polymer such as PE, PP, PVC, waxes, etc., as a laminate film to its surface (Kjellgren et al., 2008). Another strategy is to develop a very dense paper structure in which highly refined cellulosic fibers or applied coating layers of biopolymers such as glassine paper or nanocellulose block the migration (Hubbe & Pruszynski, 2020; Tyagi et al, 2018b, 2019a). For water vapor resistance, hydrophobic petro-based materials such as PP, PVC, and PE are most used in food packaging as films or coatings for paper. For short-term retail applications such a pizza, burgers, cookies, ice cream, etc., grease resistance and water hydrophobicity are the key barrier properties requirements.

2.2. Shelf life

Packed foods that are sensitive to moisture or oxygen, water vapor transmission rate (WVTR) and oxygen transmission rate (OTR) place high demands on defining adequate desired shelf life to maintain food quality. For a maximum allowable H_2O_{max} and $O_{2\ max}$, the shelf life (t_s) can be estimated as following equations (4) and (5) modeled by Yam

et al. (2006):

$$t_s = \frac{H_2O_{max}}{WVTR} \quad (4)$$

$$t_s = \frac{O_{2\ max}}{OTR} \quad (5)$$

where t_s is shelf life in days, and H_2O_{max} and $O_{2\ max}$ are calculated in g/m^2, which can be determined by sensory evaluation (Yam et al., 2006). For calculating t_s, WVTR is generally measured at 38 °C temperature and 90% relative humidity conditions.

There are a few studied mathematical and computational models that can predict the required barrier properties of food packaging for the desired shelf life (Noriega et al., 2014). Most of the food products are packed in packaging materials possessing barrier properties much higher than required.

3. Functional and barrier coating materials

In a state-of-the-art paper-based barrier and active packaging system,

Table 1
Barrier properties of petroleum and bio-based paper coatings.

Coating material	WVP [g. mm/m^2/day]	OP [cm$^{3.}$ mm/m^2/day/atm]	OGR (Kit #)	Applications	Reference
Petro-Based polymers					
High Density Polyethylene (HDPE)	0.1–0.24	26.3–453	–	Bottles for milk and fruit juices, caps for bottled beverage	(Blueridge Films Inc. associate(s), 2020; Hansen & Plackett, 2008; Keller & Kouzes, 2017; van Aardt et al., 2001; Wang et al. 2018)
Low Density Polyethylene (LDPE)	0.39–0.59	98–453	–	Bottles for milk and fruit juices, caps for bottled beverage, packaging films for frozen and dry foods	(Hansen & Plackett, 2008; Lomate et al., 2018; Peroval et al., 2002)
PET	0.28	55	–	Bottles for beverages, salad dressings, cooking oils and peanut butter	(Galdi et al., 2015; Gomes et al., 2019; Peroval et al., 2002; Triantafyllou et al., 2002)
Metalized PET	0.04–0.10	0.16–1.7	6	Yogurt and coffee container	(Geueke et al., 2018a; Polyprint associate (s), 2020) (Pons et al., 2014)
EVOH	0.8–2.4	0.01–0.15	–	Employed with PE/PET/PS for packaging meat, fish, cheese, nut, desserts and alcoholic beverages	(T. Anukiruthika et al., 2020; Blueridge Films Inc. associate(s), 2020; Keller & Kouzes, 2017; Polyprint associate(s), 2020)
PVC	0.94–0.95	3.28–394	–	Films are used for meat and vegetable wrapping, bottles are used for juice and cooking oil	(Keller & Kouzes, 2017; Leadbitter, 2003)
Polyvinylidene Chloride (PVDC)	0.025–0.913	0.00425–0.57	–	Films are used for meat, poultry, confectionary and vegetables packaging	(Keller & Kouzes, 2017; "PVDC Food Packaging Market 2017)
Saran PVDC Films	0.009–0.34	0.00425–0.00625	–	Wrapping films for meat, poultry, seafood cooked food packaging	(Ebnesajjad, 2012; Keller & Kouzes, 2017)
Poly Propylene (PP)	3.9–6.2	35–377	–	Meat, poultry and confectionary packaging	(Martens et al., 2016; McMillin, 2017; Polyprint associate(s), 2020)
Metalized OPP	0.01–0.03	0.019–0.16	–	Snacks, confectionary, nuts and coffee products	(Metalized Oriented Polypropylene (Metalized OPP) Films Market; Polyprint associate(s), 2020)
Nylon-6	0.24–125	0.394–2.50	–	Processed meat packaging	(Blueridge Films Inc. associate(s), 2020; Keller & Kouzes, 2017; Quintavalla & Vicini, 2002)
Metalized Nylon-6	0.1–0.15	0.00078	–	–	(Polyprint associate(s), 2020)
Polystyrene	109–155	4350–6200	–	Egg cartoons	(Blueridge Films Inc. associate(s), 2020; Polyprint associate(s), 2020; Tajeddin et al., 2018)
Polytetrafluoroethylene (PTFE)	0.0045–0.30	222–387	12	Not directly used in food packaging; widely used for coating belts and conveyors in food processing industries for eggs, bacon, sausage, chicken and industrial bakeries	(Dhanumalayan & Joshi, 2018; Keller & Kouzes, 2017)
Bio-Based & Bio-degradable polymers					
Cellulose acetate	4.5–77.85	0.239–1.301		Fresh food baked goods, candy packaging	(Bras et al., 2007; Pawar & Purwar, 2013)
Starches	1.06–2.83	0.01–0.014	–	Wraps for fruits, vegetables, red meat	(Pelissari et al., 2014; Rydz et al., 2018; Talja et al., 2007)
Soy Protein	3.5–4.5	0.94–2.56	–	Films, coating on paper for fruits and vegetable packaging	González and Alvarez Igarzabal (2013)
Pectin	113	2.4	–	Films, coating on paper for cheese, milk powder and beverage	(Mellinas et al., 2020; Vartiainen, Tammelin, et al., 2010)
Chitosan	1.28–21	0.2–9.8	–	Films for fresh fruits and vegetables, bread packaging	(Miranda et al., 2004; Petriccione et al., 2015; Vartiainen, Tuominen, & Naattinen, 2010)
PLA	1.34	0.038–0.042	12	Films for fresh fruit, vegetables, salad and chicken meat packaging	(Hansen & Plackett, 2008; Mohamad et al., 2020; Tee et al., 2016)
PVOH	41.904	0.1–45	–	Packaging for cheese, coffee, nut products eggs and ice-cream	(Musetti et al., 2014; Virtanen et al., 2014)
PHB	0.23	0.42	–	Cheese coatings, water bottles, Mayonnaise containers	(Bucci et al., 2007; Kaci et al., 2014)
CNF	17.0	0.3	–	–	Vartiainen et al. (2013)
Modified CNF	17.0	0.003	5–8	–	(Lavoine et al., 2014; Nair et al., 2014)
CNC-MMT-Soy	20	0.28	6	Fresh food packaging, baked goods packaging	(Rodríguez et al., 2014; Tyagi et al., 2018c)
CNF/CNC-MMT-Soy	17.8	0.03	11	–	Tyagi, Lucia, et al. (2019)

functional materials are coated onto the paper or paperboard to provide a barrier to protect selected packaged goods. Due to the abundance and versatile properties of the petro-polymers in the prevailing plastic-based economy, packaging materials are mainly oriented to the use of synthetic polymers. Functional coatings, providing barriers to the food packaging requirements, may include protection against oxygen, aromas, liquid water and water vapor, oils, and grease, etc. These coating materials, as discussed below, can be classified as synthetic petroleum-based, bio-based, and nano-composite materials. Recent developments in each of the categories are described briefly in the sections that follow.

3.1. Synthetic polymers

Petro-based polymer materials have unique properties such as being malleable. They are more elastic in their responses to physical impacts compared to other types of materials such as metal, glass, and ceramics. Nowadays, quite a wide variety of plastic-coated paper containers can be found in the food and beverage industry. Among all the synthetic polymers, polyolefins (POs) such as polypropylene (PP) and polyethylene (PE) are the highest produced due to their low cost, flexibility, chemical inertness, recyclability, good processability, nontoxicity and biocompatibility (Chaudhry et al., 2012; Guillard et al., 2018). In addition to polyolefins (PP and PE), a wide variety of plastic-coated paper containers can be seen in the food and beverage industry (Zhong et al., 2020). Commonly used plastic (petro-based) coatings for paper-based food packaging include polyethylene terephthalate (PET) (Erlat et al., 1999; Nakaya et al., 2015), polymethylpentene (PMP) (Kirwan, 2008), polyethylene (PE) (López-Carballo et al., 2018), biaxially oriented polypropylene (BOPP), biaxially oriented nylon (BON/OPA), polyesters (Barrientez & Strege, 2003) and polystyrene (Marsh, 2003). All these plastic coatings are either extrusion coated or laminated on paper or paperboard packaging. Optimal barrier performance against multiple permeants is often achieved by co-extrusion of multiple layers of laminate films (Goulas, 2001; Mitsoulis, 2005). A few of the key coating polymers are described below with respect to their recent developments.

3.1.1. Polyolefins

Polyolefins are produced in the highest amount among all the thermoplastics, have high barrier properties against fat, including essential oils and moisture (Table 1) (Feldman, 2016; Heo et al., 2019; Jagannath et al., 2006). Polyolefins have very good sealing properties and can be used as a tie layer on aluminum foil when applying different polymers to foil, such as in the case of paper-based aseptic beverage packaging (Tetra Pak) (Novák et al., 2015). Polyethylene (PE) and polypropylene (PP) are the two most abundant synthetic polymers. Coatings for packaging based on these polymers are discussed briefly in the following segments.

Polyethylene (PE): Both low density (LDPE) and high density (HDPE) versions of PE coating are widely used in food packaging. The LDPE is the most frequently used product for frozen food. LDPE is a good barrier for moisture but is relatively permeable to oxygen and is a poor odor barrier (Allahvaisi, 2012). HDPE has a higher temperature resistant property and can withstand abrasion better and has better gas barrier characteristics than LDPE. HDPE coatings are normally applied on the reverse side of paper packaging that does not come into contact with food (Marichelvam & Nagamathan, 2017).

The poor gas barrier properties of PE laminates/films limit their application in many food packaging applications (Liu et al., 2017). Thus, significant effort has been given to improve the oxygen barrier property of the PE through surface modification or by forming composites incorporating different barrier materials (Heo et al., 2019; Jang & Lee, 2004; Lange & Wyser, 2003; Yeun et al., 2006). PE also fails to restrict the migration of microbes from paper or mineral-coating pigments on paper to the facing food (Suominen & Suihko, 1997). In many recent studies, LDPE and HDPE have been incorporated with other active ingredients such as zinc oxide nanoparticles, fruit seed extracts and chitosan to incorporate antimicrobial properties in food packaging. In addition, multi-walled carbon nanotubes (MWCNTs) and graphite oxides (GO) have been added to impart better mechanical and gas barrier properties (Glaser et al., 2019; Heo et al., 2017; Islam et al., 2016; MUNTEANU et al., 2014; Shankar et al., 2019). PE has also been studied in a multilayer coating system with other polymers such as polycaprolactone (PCL) and active compounds to offer antimicrobial properties for food packaging (Rešček et al., 2016). Though PE is considered a good barrier against oil and water, it has not shown any barrier ability against possible contaminants such as anthracene, benzophenone, dimethyl phthalate, methyl stearate and pentachlorophenol from used packaging paper in contact with the food (Choi et al., 2002; Salafranca & Franz, 2000).

Polypropylene (PP): PP is another extensively used polyolefin for packaging due to its rigidity, toughness, easy processability and low cost (Karian, 2003). Biaxially oriented PP (BOPP) is an excellent candidate for packaging dried foods such as cookies, candies, snack foods, dry fruits, etc., due to its good barrier against moisture, odor and grease (Allahvaisi, 2012). However, its poor oxygen barrier, especially when a thin film is used as laminate or coating over the paper, limits widespread use in the food packaging industry (Khalaj et al., 2016; Sinha Ray & Okamoto, 2003). To overcome this problem, PP/nano-clay (silica, MMT, OMMT) composites have been fabricated, which have shown better oxygen barrier and thermal properties (Azinfar et al., 2014; Fitaroni et al., 2015; Furlan et al., 2011; Gonzalez et al., 2014; Reddy et al., 2010). The melt-compounding method has been used to overcome the dispersion challenge of polar fillers into non-polar PP matrix. Another promising approach to improve barrier properties of the PP is the incorporation of metal-based oxygen scavengers into the PP matrix (Atayev & Oner, 2013; Hannon et al., 2015; Kuorwel et al., 2015; Majeed et al., 2013; Reig et al., 2014). Plasma-enhanced atomic layer deposition (PEALD) has gained a lot of interest recently since it is a controlled continuous growth technique and offers a high degree of reproducibility, homogeneous surface coverage with fewer defects, and has been used to deposit Al_2O_3, SiO_2, SiN_x, and TiO_2 coatings to improve the gas barrier property of the PP films (Andringa et al., 2015; Carcia et al., 2009; Gebhard et al., 2016, 2018; Hoffmann et al., 2015; Meshkova et al., 2018; Ozkaya et al., 2015). In addition, PP/MWCNTs and PP/reduced graphite oxides are also reported to improve the barrier and thermal properties, where incorporation of the reduced graphite oxides showed the best results (Feldman, 2016).

Polyethylene terephthalate (PET): PET is a semi-crystalline thermoplastic that has been extensively used as a packaging material for water and beverage containers (Majdzadeh-Ardakani et al., 2017; Masmoudi et al., 2020; Zekriardehani et al., 2018). PET offers desirable advantages such as chemical, thermal and shatter resistance, flexibility and recyclability, low cost, glass-like clarity, light weight, strong barrier against dilute acids, gases, oils and alcohols (Lubna et al., 2018; Sangroniz et al., 2019). However, its low moisture vapor resistance, compared to PE and PP, limits its use in the food packaging industry (Jin et al., 2015; Majdzadeh-Ardakani et al., 2017; Wu et al., 2012). A number of approaches have been taken to enhance the barrier properties of the PET films and bottles, such as the addition of fillers to create a tortuous path for gas molecules (Frounchi & Dourbash, 2009; Hayrapetyan et al., 2012), incorporation of oxygen scavengers (Galdi et al., 2008; Mahajan et al., 2013; Miranda et al., 2017) and nanoparticles (Meng et al., 2020; Mohd Noh et al., 2019; Wei et al., 2019), copolymerization (Flores et al., 2019; Sangroniz et al., 2019) strain induced crystallization (Zekriardehani et al., 2017) and deposition of inorganic films onto PET using different deposition techniques (Cho et al., 2018; He et al., 2013; Jang et al., 2019; Jin et al., 2015). Recently, a more cost-effective method has been used in which a small amount of low molecular weight diluents (LMWD) is incorporated into PET film to improve the barrier properties (Burgess et al., 2015; Lee et al., 2012).

3.1.2. Ethylene vinyl alcohol (EVOH) and poly vinyl alcohol (PVOH)

EVOH is a copolymer of ethylene and vinyl alcohol having a semi-crystalline structure (Mokwena & Tang, 2012). Its excellent barrier properties against oxygen (O_2) and carbon dioxide (CO_2), chemical resistance and retention of flavor and aroma has allowed it to serve as an alternative to the use of metallic aluminum foil and thus has increased its demand for packaging applications (Cooksey, 2008; Maes et al., 2019, 2018, "Multilayer Flexible Packaging - 2nd Edition," 2020). EVOH exhibits high transparency, oil/solvent resistance, high rigidity and easy processability. The oxygen resistance of EVOH is dependent on the extent of crystalline structure, and EVOH having 70% crystallinity has shown the best oxygen barrier properties (Mokwena & Tang, 2012). However, EVOH is sensitive to moisture due to the presence of hydroxyl groups and is usually used as a copolymer with hydrophobic materials as a good barrier coating system (Kim et al., 2020; Salem et al., 2015; Sun et al., 2019). EVOH films are not biodegradable, but blending with other compounds or copolymerization with other monomers has been found to ease the process of decomposition (Elhamnia et al., 2020).

Poly vinyl alcohol (PVOH) is a synthetic biodegradable polymer with excellent oxygen barrier performance, which is two order magnitude higher than PET due to the stiffness of its chain imparted by hydrogen bonding (Minelli et al., 2010). Like EVOH, PVOH is highly sensitive to moisture and is subject to decreases in its barrier and mechanical properties upon absorption of moisture, which softens and plasticizes the polymer matrix. The water vapor permeability of PVOH increases at high relative humidity and can lead to complete disintegration of the matrix (Schmid et al., 2014). Thus, PVOH is used as a coating in combination with other packaging materials to form a hybrid/composite structure where low gas permeability is required (Cazón et al., 2020; Hamdani et al., 2020). Several advancements have been made to improve the water vapor barrier property of PVOH, such as esterification (Schmid et al., 2014), compositing with nanoparticles (Abdullah et al., 2019), and grafting with non-polar compounds (Schmid, Dallmann, et al., 2012). One of the potential cost effective and commercially feasible methods of improving the water vapor barrier performance of PVOH for paper coating or lamination is metallization with metal oxides or siloxanes.

3.1.3. Polyvinyl chloride (PVC)/Polyvinylidene chloride (PVdC)

PVC/PVdC is clear, moderately tough, amorphous in nature with good flame resistance and has diversified applications, including paper-based food packaging materials (Kumar, 2019). However, the presence of chloride induces a dipole in the PVC chain and hinders the chain mobility due to hydrogen bonding, leading to a brittle structure with low impact strength (Hosney et al., 2018). This limitation can be overcome by using plasticizers, which greatly improves PVC's flexibility, toughness and thermal properties (Shah & Shertukde, 2003). Phthalates have been used as plasticizers due to their low cost and excellent plasticizing effects, which impart flexibility and malleability to the final products. However, the leaching of the plasticizers from the PVC matrix is a major concern for human health and has limited its application in food packaging (Benjamin et al., 2017). The leaching out of phthalates has also been reported for poly (vinylidene chloride) (PVdC), a flexible thermoplastic, which is used for food packaging due to its outstanding oxygen and moisture barrier properties (Wang et al., 2020; You et al., 2020). To overcome this problem, bio-based plasticizers with higher molecular weight have been used; these not only prevent the leaching problems but also improve the mechanical and thermal properties of the PVC and PVdC packaging films (Chavan & Gogate, 2015; Jia et al, 2015a, 2015b).

PVdC is extremely useful for food packaging due to its excellent gas, water vapor and oil barrier properties (Table 1). However, the major disadvantage of polyvinylidene chloride is that it will undergo thermally induced dehydrochlorination at temperatures very near to processing temperatures >150 °C, and hence it is not recommended to be recycled with other polymer or incinerated (Collins et al., 1999; Hsieh & Ho, 1999). Many consumer-packaged goods (CPG) and food companies have marked PVC and PVdC in their material elimination list as part of their sustainability goals.

3.1.4. Polyethylene-vinyl acetate (PEVA)

PEVA is also used as a food packaging polymer due to its clarity, high gloss, flexibility at low temperature, stress-crack resistance and mechanical properties (Amini et al., 2019). One of the major advantages of using PEVA is its good toughness at low temperatures, which makes PEVA processable at room temperature, in contrast to other polymers used in packaging (Eskandarabadi et al., 2019; Wattananawinrat et al., 2014). The properties are largely dependent on the molecular weight and the vinyl acetate content. The water vapor and gas barrier properties of EVA are poor, and thus typically this thermoplastic is copolymerized with other polymers such as PET, cellophane and biaxially-oriented PP films as a part of multilayer film due to good adhesion and heat-sealing properties (Dorey et al., 2020; Huang et al., 2019). The most common used form of PEVA or EVA are laminates or composites with PE or PET; these are used for deep-freeze applications, bag in paper-box applications, milk pouches, cheeses wraps, etc. (Cooper, 2013; Fellows, 2017)

3.1.5. Nylon

Nylon is also used as a packaging material due to its good mechanical properties, odor barrier, and resistance to oil and fat (Bandyopadhyay et al., 2016). But, it has high moisture vapor permeability and low heat-sealing ability, which highly limits its use in the food packaging (Kirwan et al., 2011). This problem can be overcome by using different surface modifications to introduce functional groups, which will impart hydrophobicity on the nylon surface and increase moisture barrier (Jia et al., 2006; Ting et al., 2015). Table 1 compares different barrier properties of the common polymers used for packaging.

3.1.6. Metalized polymers

Most of the single-layer plastic films generally lack the ability to provide an optimum gas and water vapor barrier compared to metal and glass packaging. Metalized aluminum and transparent oxide coated plastic films provide useable O_2 and WVTR performance (Silvestre et al., 2016). Inorganic AlO_x, SiO_x and SiN_x oxides and nitrides deposited by vapor deposition to films is a cost-effective option rather than electron sputtering for improving gas and WVTR barrier properties (Gebhard et al., 2018). PE, PP and PET films are generally used with SiO_x and AlO_x coatings (Pardo-Figuerez et al., 2018). Unfortunately, these films are not heat-sealable; thus, they are used in a multilayer coating sandwich system with a heat damage-susceptible oxide film between plastic film layers (T Anukiruthika et al., 2020). However, if a metalized layer is going to be in contact with food, then a cold seal is required for sealing the food package. These inorganic transparent barrier films are forecasted to grow annually at high rates even with their gas and WVTR limitations compared to aluminum foil (Keane, 2017).

3.1.7. Polyfluoroalkyl substances (PFASs)

Fluorocarbons in the form of polyfluoroalkyl substances (PFASs) have been widely used in nonstick, oil and grease resistant (OGR), and waterproof food packaging until regulated in 2015 (Fig. 1). PFASs possess both hydrophobic and lipophobic characteristics (Li & Rabnawaz, 2018; Schaider, Balan, Blum, Andrews, Strynar, et al., 2017). PFASs have been used in food packaging not only as coatings to prevent the paper material from soaking up fats and water, but also in printing inks and as moisture barriers. The applications particularly target fatty foods intended to be heated in the packaging or stored for an extended period (Schaider et al., 2017; Trier, Xenia; Taxvig, Camilla; Rosenmai, Anna Kjerstine; Pedersen, 2017). Some of the most common applications of PFASs coated paper are sandwich wrappers, french-fry boxes, bakery bags etc. Since the PFASs can migrate into food, and contaminate landfills and compost after disposal, the use of PFAS to treat food packaging can lead to unnecessary long-term exposure to harmful

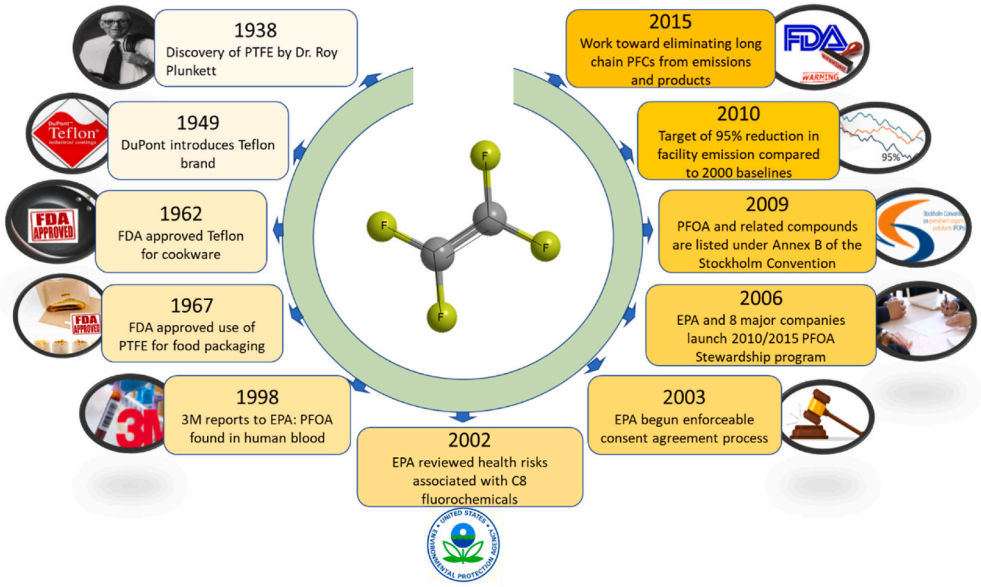


Fig. 1. PFAS from its discovery to the enforced limitation on long chained carbons (>C8).

chemicals. In 2006, EPA ((Environmental Protection Agency) started a program gradual phase-out of long-chain PFAS emissions and products by 2015, and FDA regulated the use of long chain (>C-8) PFASs for direct food contact in the same year (Glenn et al., 2021). Currently, alternatives to PFASs are being used for oil and grease resistance in food packaging discussed in later section '4.4'.

The discussed synthetic polymers, when used as laminates or coatings, provide a low-hanging-fruit solution for paper packaging to be used for direct food contact. This will help in reducing plastic from composting and landfilling sites, but recyclability and biodegradability issue continues, since laminates need to be separated from the paper to be recycled. Recyclability of laminated or plastic-coated paper can be improved by employing a water-soluble layer between the paper and plastic layers that enables their separation.

3.2. Bio-based polymers

Increased environmental concerns over the use of certain plastic packaging and coatings in combination with consumer demands for both higher quality and longer shelf life have led to increased interest in alternative bio-based packaging materials (Zhong et al., 2020). Naturally renewable biopolymers can be used as barrier coatings on paper packaging materials, as they provide high oxygen and oil barriers, are biodegradable and may also retard unwanted moisture transmission in food products (Herniou–Julien et al., 2019). They have the potential to replace current petroleum-based packaging and paperboard coatings (Khwaldia & Arab-tehrany, 2010). Biopolymer-based coating materials obtained from naturally renewable resources including polysaccharides, proteins, and bio-based waxes, etc., offer favorable environmental advantages such as recyclability and reusability compared to conventional petroleum-based synthetic polymers (Mlalila et al., 2018; Wróblewska-Krepsztul et al., 2018). Biopolymer films and coatings may also serve as gas and solute barriers and complement other types of packaging by minimizing food quality deterioration and extending the shelf life of foods (Bilbao-Sáinz et al., 2010; Hubbe et al., 2017; Song & Rojas, 2013; Wu et al., 2009).

The coating of biopolymers to paper provides interesting functionalities while maintaining environment-friendly characteristics such as recyclability and compostability of the material. Renewable biopolymers, such as polysaccharides (Hubbe et al., 2019), poly (lactic acid) (Arrieta et al., 2014a; De Geyter et al., 2010), caseinates (Colak et al., 2015; Pereda et al., 2011), whey protein isolate (Aulin et al., 2010), isolated soy protein (Matikainen, 2017; Wu et al., 2009; S.; Zhang, Yu, et al., 2016), wheat gluten (Das et al., 2018; El-Wakil et al., 2015), corn zein (Moreno et al., 2019; Vahedikia et al., 2019), chitosan (Deng et al., 2011; Honarkar & Barikani, 2009; Sakai et al., 2002; Tang et al., 2016), carrageenan (Azizi & Mohamad, 2018; Savadekar et al., 2012), alginate (Abdollahi et al., 2013; Sirviö et al., 2014), nanocellulose (Hubbe et al., 2017; Puceković et al., 2015; Pulppaper et al., 2014; Salem et al., 2020a; Tyagi et al., 2018c), pectin (Dufresne et al., 1997; Valdés et al., 2015; Vartiainen et al., 2014a) and starch (Brodnjak & Muck, 2017; Vartiainen et al., 2014a) have been investigated as paper-coating materials. The most studied biopolymers as barrier coatings and films for paper-based packaging are discussed in the following sections.

3.2.1. Polylactic acid (PLA)

PLA is a compostable (under industrial conditions) and bioactive thermoplastic aliphatic polyester derived from renewable resources, such as cassava roots and chips, corn starch, and sugarcane. It has been accepted as GRAS (Generally Recognized as Safe) by the Food and Drug Administration (FDA) and is suitable for use in the food and beverage packaging (Oz et al., 2017). The annual production of PLA has been estimated to be 140,000 tons, and it is expected that PLA and its composites have the potential to substitute for petroleum-based products (Siakeng et al., 2019). PLA has a desirable set of qualities such as good transparency and processability, glossy appearance, and high rigidity (Mahmoodi et al., 2019). PLA exhibits better thermal processability compared to other biopolymers and thus, various processing techniques, such as, cast filming, blow filming, fiber spinning, injection molding, etc., can be used to fabricate PLA films (Rasal et al., 2010). PLA-coated paperboards have exhibited improved water barrier properties through a reduction in the water vapor permeability (WVP), water absorptivity (WA) and increase in water contact angle (CA), as achieved with the optimum concentration of 3 w/v % PLA in chloroform (Rhim et al., 2007). However, the obtained PLA films display brittleness and a high extent of crystallization, which limits their applications when plastic deformation at high stress is required (Nagarajan et al., 2016).

The properties of the PLA are highly dependent on the presence of relative amounts of L- and D-lactic acid monomers, which govern the final crystallinity and mechanical properties: pure L-PLA results in high crystallinity, whereas increasing content if D-PLA makes any product

more amorphous with improved film-forming properties. PLA lacks flexibility and elasticity due to its high crystallinity, so a number of methods are employed to increase the flexibility or reduce brittleness of PLA (Awale et al., 2018; Zhao et al., 2017, 2020). In addition, PLA has moderate moisture and oil barrier properties, but poor oxygen and carbon dioxide barrier characteristics (Mattioli et al., 2013). However, these limitations can be overcome by reinforcing PLA with micro- and/or nano-fillers (Nakagaito et al., 2018). For instance, mineral fillers such as clay, montmorillonite, and silica in their nano dimension fortifies the PLA composites in terms of strength and barrier properties (Castro-aguirre et al., 2018). PLA composites with biopolymers have also been studied to improve thermal, mechanical and barrier properties of PLA films and coatings (Bajracharya et al., 2017; Cheng et al., 2015; Li et al., 2017; Siakeng et al., 2019). The oil and grease barrier properties of PLA are not sufficient to protect the oil-bearing food; hence metallization is employed for commercial applications to give the necessary barrier properties (Koppolu et al., 2019). The properties of PLA can be made comparable to those of commercial commodity or engineered polymers by doing adequate modifications to the matrix (Bajracharya et al., 2017; Li et al., 2017). PLA has a great potential in developing antimicrobial packaging because it is an excellent material for successful incorporation with antimicrobial agents such as plant extract, essential oils, enzymes, and metals oxides to develop antimicrobial characteristics (Khosravi et al., 2020; Radusin et al., 2019; Villegas et al., 2019; Yang, Ching, & Chuah, 2019). Several examples can be found of PLA-coated/laminated paper being used commercially for food products such as stand-up bags for dry fruits, pulp molded trays, paper glasses for cold liquids etc.

A conductive polymer composite based on PLA can be used in smart packaging such as antistatic, electromagnetic shielding, and intelligent packaging (Frackowiak et al., 2015; Kruželák et al., 2021; Liu & Zhang, 2011). PLA is stable at room temperature, but it is easy to degrade rapidly in environments that are moderately high temperature, acid-base, or microbial, and finally generate CO_2 and water molecules that are innocuous to the environment (Zaaba & Jaafar, 2020). PLA-coated paper may be considered compostable under specified conditions; however, it is always recommended to confirm compostability before starting.

3.2.2. Polyhydroxy alkenoates (PHAs)

PHA is another non-toxic, crystalline promising biopolymer for packaging. Polyhydroxybutyrate (PHB) is the most common polymer of the polyhdroxyalkanoates (PHAs) family. It was discovered by Maurice Lemoigne in 1923 from the gram positive bacteria *Bacillus megaterium* (Hoseinabadi et al., 2015). It is a naturally produced polyester that can be used as a biodegradable thermoplastic under both the aerobic and anaerobic conditions (Bartczak et al., 2013). PHB has similar properties to PP and is seen as the sustainable replacement candidate for this fossil commodity polymer due to its superior moisture and aroma barrier performance (Bucci et al., 2007; Kamravamanesh et al., 2018; Peelman et al., 2013). Key benefits of PHB over PP are a lower carbon footprint and avoidance of “white pollution”, which manifests itself, e.g. as marine debris and microplastics (Markl et al., 2018). However, high rigidity and difficult processability due to thermal instability has prevented its widespread use. Several attempts have been made to overcome the limitations of using PHA for food packaging not only from the quality perspective but for improving processability.

For instance, blending PHB with PLA helps in improving the flexibility since they have comparable melting point temperatures (Arrieta et al., 2015). Flexibility also can be enhanced by incorporating plasticizer and antioxidant by melt blending or by fabricating composites wthe the incorporation of nanomaterials (Aluthge et al., 2013; Panaitescu et al., 2017, 2018; Seoane et al., 2017). The improved properties of PLA-PHB blends have allowed them to be used as bio-based packaging of fatty products (Levkane et al., 2008). Joyce et al. (2018) studied the photonic drying for PHA coating on a commercial paper. A rapid pulse of photonic energy enables PHA particles to melt and form a film within milliseconds (Joyce et al., 2018).

PHB has been studied successfully for food packaging (Bucci et al., 2007; Guillard et al., 2018; Kamravamanesh et al., 2018), for which it has been found to be more rigid and less flexible than PP (Kamravamanesh et al., 2018). Peelman et al. (2013) reviewed the application of PHB for food packaging, showing it as a competitive bioplastic with petro-based plastics (Peelman et al., 2013). Levkane et al. studied the effect of pasteurization on a meat salad packed in conventional (PE, PP) and bio-based packaging (PLA, PHB) (Levkane et al., 2008). Bucci et al. found that PHB can replace PP for the packaging of fatty products (mayonnaise, margarine, and cream cheese) (Bucci et al., 2007; Levkane et al., 2008).

Starches: Starches have been used and studied as a coating over paper or as a film by extrusion and casting methods (Li et al., 2019; Vartiainen et al., 2014a). The major functional properties rendered by starch coatings are oxygen barrier properties and printability (Pal et al., 2020; Rastogi et al., 2015). The oxygen transmission rate of the starch coatings is strongly dependent on relative humidity conditions (Talja et al., 2008). Increased permeability at higher moisture conditions is most likely due to increased polymer chain mobility, which facilitates the transport process (Forssell et al., 2002). The films and coatings made with starch generally face problems of brittleness and fracture.

Plasticizers, such as glycerol, sorbitol, or xylitol, are typically used for reducing the brittleness of starch (Talja et al., 2007). The concentration of plasticizers has a big impact on strain elongation strength properties of starch coatings (Myllarinena et al., 2002). Crystallization of starch coatings can also be prevented by using binary polyol mixtures as plasticizers. Oxygen barrier properties of starch films can be improved by using sorbitol (Gaudin et al., 2000). This behavior is related to changes in secondary relaxations, which are hindered because of the connections established between starch and sorbitol, leading to decreased diffusion of oxygen molecules. Researchers recently have focused on edible food packaging from starch. Starch has emerged as a potential candidate for this venture where starch from both conventional and non-conventional sources are used to produce starch based edible food packaging (Dai et al., 2019; Dash et al., 2019; Galindez et al., 2019; Pająk et al., 2019).

Studies have been performed to improve functional properties of starch-based films as active and intelligent food packaging by incorporating active materials such as flavonoids and propolis-a resin obtained from beehives (Motelica et al., 2020; Mustafa et al., 2020).

A schematic representation of common bio-based polymers used for packaging is shown in Fig. 2.

3.2.3. Cellulose-based polymers

In recent years, research has been focused on the development of cellulose derivatives for use in packaging applications. Cellulose acetate is the most developed and consumed cellulose-based thermoplastic. High-grade cellulose is treated with a methylene chloride-acetic acid solution to substitute hydroxyl groups with acetyl groups, thereby converting the cellulose to cellulose acetate (Nakanishi et al., 2006). Cellulose acetate has been tagged as “Generally Recognized As Safe” (GRAS) by the FDA, which has prompted the food packaging industry to more aggressively develop innovative applications of cellulose acetate (Shaghaleh et al., 2018). As such, cellulose acetate is commonly used for wrapping baked goods and fresh produce. For making films or using cellulose acetate as coatings, plasticizers are required to impart good gloss, clarity, printability, modified rigidity and dimensional stability (Fordyce & Meyer, 1940).

Cellulose acetate films have lower tear resistance but are tough and resistant to puncture, which is considered good for many applications (Liu, 2006). They possess relatively poor moisture barrier properties and lower thermal resistance compared to conventional thermoplastics (Paunonen, 2013). Also, cellulose acetate films are very rigid, which that makes them brittle. The resulting crinkling sounds while being handled

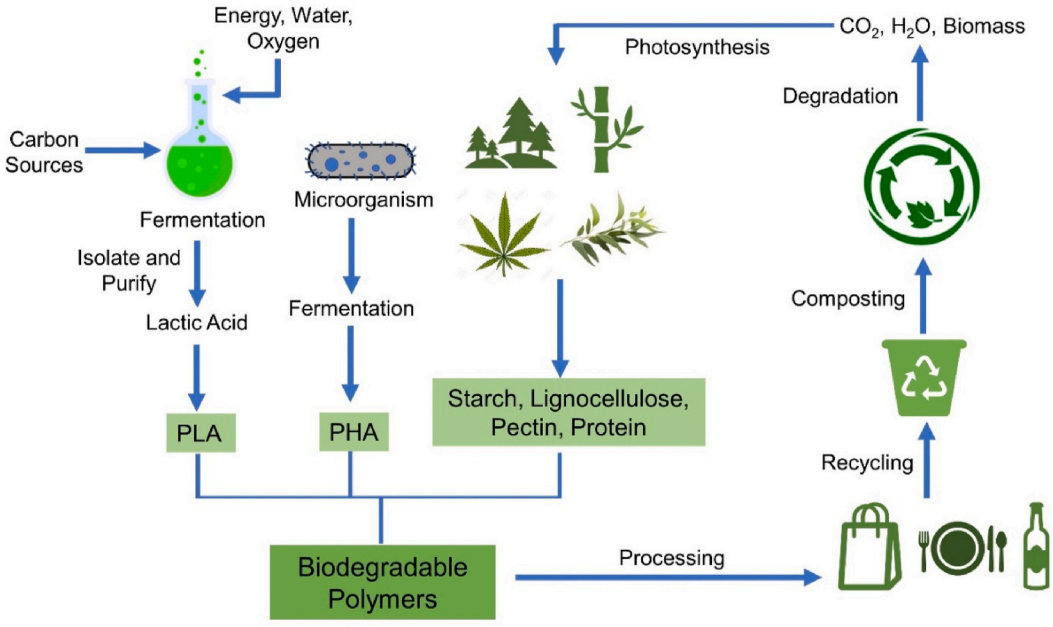


Fig. 2. Schematic illustration of recyclable process of biodegradable polymers.

by consumers means that they are less favored for their application in flexible flow wraps for many commercial food packaging applications. In prolonged applications, cellulose acetate undergoes hydrolysis to produce acetic acid, which is commonly referred to as vinegar syndrome, leaving a smell behind (Puls et al., 2011). These properties have prevented more extensive applications of cellulose acetate films in food packaging.

Several academic and industrial developments have been focused on improving gas and moisture vapor barrier properties of cellulose acetate (Aldana et al., 2014; Arrieta et al., 2014b; Bras et al., 2007; Dellinger & Helou, 2015). Bras et al. (2007) created a fully substituted cellulose acetate using the acyl chloride method and studied its impact on water vapor permeability (WVP) and oxygen permeability (OP) (Bras et al., 2007). With increasing substitution, the WVP was found to decrease, along with increasing OP (Bras et al., 2007). Efforts have been made to incorporate several active components to cellulose acetate films to embed antimicrobial activity in the packaging (Barbiroli et al., 2012). Assis et al. (2020) studied the impact of incorporating the pigments norbixin and lycopene/zeaxanthin for creating active packaging (Assis et al., 2020).

3.2.4. *Micro and nano-fibrillated cellulose*

Different variants of nanocellulose designated as cellulose nanofibril (CNF), cellulose nanocrystals (CNC), microfibrilliated cellulose (MFC), and lignin-containing cellulose nanofibril (L-CNF) have been suggested as suitable for many coating applications such as strength promoters and barrier materials (Agate et al., 2020; Ahola et al., 2008; Hubbe et al., 2017; Taniguchi & Okamura, 1998; Tyagi et al, 2018a, 2019a), as well as for advanced applications, such as in preparing transparent flexible films (Nogi et al., 2009; Okahisa et al., 2009; Yano et al., 2005), magnetic or superabsorbent aerogels (Kettunen et al., 2011; Korhonen et al., 2011) and films with tunable optical properties.(Beck et al., 2011). Using nanocellulose as an additive or applying it as a coating for packaging material is a potentially big area for research (Hubbe et al., 2017). Nanocellulose has been studied intensively for gas and oxygen barrier properties related to their use as coatings for potential applications in packaging (Azeredo et al., 2017; Hubbe et al., 2017; Rastogi et al., 2015). A few studies have focused on barrier performance of nanocellulose against liquids (polar and non-polar both) after employing of physical and chemical modifications (Ansari et al., 2018; Teisala et al., 2014; Zhang et al., 2017). Another area of study for nanocellulose is its antimicrobial activity in conjunction with other materials such as sodium alginate (Tang et al., 2017), allicin (Jebali et al., 2013), polyrhodamine (Tang et al., 2015), nitric oxide and chitosan (Sundaram et al., 2016; Tyagi, Mathew, et al., 2019). Mild chemical and physical modifications have also been studied to render the water and oil barrier properties in nanocellulose films and coatings while maintaining the OTR (Salem et al., 2020,b; Tyagi et al., 2018c).

L-CNF has been found to be more hydrophobic compared to CNF (Osong et al., 2016; Peng et al., 2018; Rojo et al., 2015; Wang, Jia, Liu, & Miao, 2018), and this can be an advantage for many packaging applications. On the other hand, lignin content is considered to interfere in hydrogen bonding between fibrils during coating or film formation (Hubbe, 2014; Przybysz et al., 2016). The mechanical strength of L-CNF films has shown improved results when compared to CNF films (Osong et al., 2016; Peng et al., 2018; Rojo et al., 2015). This has been explained based on a uniform distribution of lignin, which seemingly aids in stress-transfer between fibrils and thus can preserve mechanical properties (Rojo et al., 2015).

3.2.5. *Lignin*

Lignin is one of the three major mass components of lignocellulosic biomass, where the amount of lignin can range from 15 to 25% depending on the plant source (Yang et al., 2019). Lignin is an aromatic-based, cross-linked, amorphous heteropolymer that consists of various phenyl propane units linked by carbon-carbon and ether bonds. Lignin is the only scalable renewable feedstock composed of aromatic units. Only 2% of the 50–60 million tons of lignin produced in pulp- and papermaking processes was utilized for specialty products, whereas 98% was used as low-value fuel (Asgher et al., 2020; Bajracharya et al., 2017; Kropat et al., 2021). In addition to the abundance, its other properties, such as thermal and mechanical stability, antioxidant activity and biodegradability, have made lignin a potential candidate for numerous applications along with its use in food packaging materials. Lignin has been reported to be used as an antioxidant in PP and PLA polymeric matrix to produce active packaging films, where it reduces the amount of free radicals by absorbing and/or scavenging and stabilizes the film (Kai et al., 2016; Morandim-Giannetti et al., 2012; Sanches-Silva et al., 2014). Barrier coating produced using starch and lignin has been found to increase the chemical stability of the packaging film in water, acidic

and alkaline solutions (Espinoza Acosta et al., 2015; Javed et al., 2018). However, the complex structure of lignin is still under study and limits its use in spite of its many advantages. More in-depth study is required to understand the lignin structure and properties so that it can scaled up from laboratory to industry for diversified applications.

3.2.6. Chitosan

Chitosan is a linear polysaccharide similar to cellulose. It is composed of randomly distributed β-(1 → 4)-linked D-glucosamine (deacetylated unit) and N-acetyl-D-glucosamine (acetylated unit). It is obtained by deacetylating its naturally occurring precursor, chitin (Tyagi, Mathew, et al., 2019). Chitosan has been studied for its unique set of biological properties such as biocompatibility, biodegradability, and low to absent toxicity (Baldrick, 2010).

Chitosan has very good film-forming properties, and the reactive amino and hydroxyl groups of chitosan have the potential to form hydrogen bonds, therefore contributing to paper strength development when used as a coating (Wang & Jing, 2016). Several studies have shown the use of chitosan as coating material for improving antimicrobial, barrier, and strength properties of paper-based food packaging (Bordenave et al., 2010; Habibie et al., 2016; Papineau et al., 1991; Song et al., 2018). Divsalar et al. describe the application of chitosan-zinc oxide nanocomposite for cheese packaging (Divsalar et al., 2018). In their study, the proposed chitosan-nanocomposite coating acted as a very effective antimicrobial active packaging.

The mechanism of antimicrobial activity of chitosan has not been well understood yet. However, cationic protonated amine ($-NH_3^+$) groups have been explained to be responsible for hydrolysis of peptidoglycans in a microorganism's wall, which leads to leakage of electrolytes out of the cell and eventually results in death of the bacterial cell (Chen et al., 1998). Chemical and physical modifications have also been employed to improve barrier properties of chitosan (Kopacic et al., 2018a, 2018b; Nicu et al., 2013). Considering the biodegradability, barrier, and strength properties, chitosan appears to be an interesting and promising candidate for environmentally friendly high value-added paper coatings. However, up to now, the economy of the chitosan application in large scale in the papermaking industry has not been considered.

3.2.7. Proteins

In the past, proteins were widely being used in coating and films applications in the paper industry. Compared to polysaccharides and lipids, protein-based polymers possess higher gas barrier properties. The oxygen permeability of soy protein-based films is 260, 500, 540 and 670 times lower than that of low-density methyl cellulose, polyethylene, starch and pectin, respectively (Chen et al., 2019). It is estimated that in the United States, about 25,000 to 50,000 metric tons of soy proteins were being used solely for paper coatings (Myers, 1993). Generally, soy protein films have inadequate mechanical properties and are poor moisture barriers because of the hydrophilic nature of soy protein, which is like starch and nanocellulose coatings. Researchers have attempted to improve the properties of soy protein films, which have major potential applications in the food and packaging industry (JW et al., 1999; Stuchell YM, 1994; Sun, 2011). Soy Protein Isolate (SPI)--coated paper was found to impart gas and oil barrier as well as adequate mechanical properties suitable for extending the shelf life of food products (Nandane & Jain, 2015; Rhim et al., 2006).

Studies have reported that the water resistance of SPI-coated paperboards is higher than that of alginate-coated paperboards (Rhim et al., 2006). The cross-linking technique is an interesting approach to enhance mechanical and water vapor barrier properties of soy protein coatings for food packaging applications (Rhim and J.H., 2004). The most commonly used covalent cross-linking agents are glutaraldehyde, glyceraldehyde, formaldehyde, gossypol, and tannic and lactic acids. However, use of films treated with such cross-linking agents for food packaging is highly questionable due to the possible toxicity of these modifying agents.

Several other proteins such as corn zein (Moreno et al., 2019; Vahedikia et al., 2019), whey protein (Aulin et al., 2010; Bugnicourt et al., 2013), casein (Coltelli et al., 2016), gelatin (Battisti et al., 2017), alfalfa and RuBisCo protein from tobacco (Gutierrez-Pena, 2020) have also been studied for paper coatings. Proteins are applied on paper to improve a wide range of properties such as gas and water vapor barrier (Lange & Wyser, 2003) and sealability (Farris et al., 2010). For instance, whey protein coatings have been shown to be good barriers when applied on paper by increasing oil resistance and reducing water vapor permeability (Gällstedt et al., 2005; Han & Krochta, 1999). Zein-coated paper exhibited oxygen barrier performance that exceeded those of PE; thus zein may be useable on paper boxes as an alternative to paraffin (Trezza et al., 1998). Moreover, RuBisCo proteins, extracted from tobacco and alfalfa leaves, have been found to show excellent performance as co-binders, rheological modifiers, enhanced glueability and coating properties (Gutierrez-Pena, 2020). Thus, alfalfa- and tobacco-based RuBisCo protein can be a potential candidate for coating purpose not only due to its performance, but also for the abundance of the protein and cheap protein sources. When protein-based coatings are combined with biodegradable substrate materials such as paper and PHAs, the materials maintain their ability to be organically recycled. They in fact catalyze the biodegradation of the substrate (Schmid, Dallmann, et al., 2012).

3.3. Other notable polymers

3.3.1. Poly(Dimethylsiloxane) (PDMS)-based polymers

The PDMS or siloxane-based polymers are constituted of alternating silicon (Si) and oxygen (O) atoms, and two organic groups are bound to each Si atom. The most common siloxane is polydimethylsiloxane (PDMS). To manufacture PDMS, the main raw materials are silicon (Si) powder and methyl chloride (obtained from methanol). The PDMS is generally referred to as bio-based if bio-methanol is used in the manufacturing process. Depending on the length of the polymer chains and the degree and nature of crosslinking, silicones are available as fluids, rubbers or resins. PDMS films are transparent, water resistant, microwaveable and provide barriers comparable to ones obtained by metallization (Schneider et al., 2009; Scopece et al., 2009). PDMS films show glass-like behavior, due to which films have low flexibility and low mechanical resistance, and the high cost of production that is mainly related to the use of a vacuum system. To overcome this issue, low-pressure microwave plasma deposition of SiO_x coatings on substrate is an excellent, state-of-the-art and low-cost means for adding the barrier functionalities to paper or other food packaging substrates (Creatore et al., 2002; Schneider et al., 2007). There have been a number of coating technologies such as solvent-based coating, solvent-less coating and emulsion technology for applying siloxane on paper. Among these, emulsion technology, which is based on a platinum-catalyzed reactive system (Lukin et al., 2020), is the preferred coating method for a variety of applications including food packaging.

PDMS coatings are used in two primary paper applications for food packaging where direct food contact is required. One is the general category of food release liners, where the major function of the paper is to exhibit easy and clean release from the surface of processed food. Reusable baking papers, interleaves or single-use papers fall into this category. The second category includes papers designed for greaseproof and grease-resistant food packaging including wet and dry products. Greaseproof and grease-resistant papers are commonly used with fresh food, fast foods, snacks, bagged and boxed items, pet food, microwave cooking products, margarine and butter, and bakery products. Use of PDMS coated paper for food packaging has doubled in past five years, corresponds to 178 thousand tons per year (Alexander Watson Associates, 2010). Continuous growth of PDMS-coated paper for release applications indicates the considerable opportunity of technology transfer to greaseproof applications in the food packaging market.

3.3.2. Waxes

Waxes are in high demand for the packaging of sweets, confectioneries, baked goods and dairy products due to their hydrophobicity, high gloss value and semitransparent nature. Wax is used as treatment, lamination, coating or impregnation of primary food contact paper for the products where grease and water barrier properties are crucial, such as fast-food containment. Waxes can be petroleum-based such as paraffin waxes, montan wax or natural waxes such as beeswax, soywax, candelilla wax, carnauba wax, jojoba wax, rice-bran wax, etc. Waxes are used in food packaging materials because they have good moisture barrier characteristics. As such, they can protect dry foods from environmental moisture or reduce moisture loss of the food stuff (Riederer & Schreiber, 2001). In addition to the moisture and grease resistance, wax coating adds respiratory functionality to the foods which need cold storage and prevent them from sweating which eventually increases their shelf life (Despond et al., 2005). The most common waxes used in paper-based food packaging are petro-based paraffin wax and alkyl ketene dimer (AKD) (Wagner, 2012).

There is concern that waxes and their other components may migrate into food stuff, particularly if they are part of the food contact layer. A study by Varner, Hollifield, & Andrzejewski (1991) showed that it was possible to measure benzophenone in paraffin waxes used in food contact materials, but no migration studies have been done to confirm this point. The natural wax-coated papers can be a potential alternative packaging material the barrier properties required to be in direct contact with food items such as confectionery, dairy, sweets, etc., from drying out and sticking to the packaging material. Garriga et al. (2019) studied the potential of arctic berry wax and beeswax as coating on paper for food packaging and found a good barrier and mechanical properties (Garriga, 2019). Carnauba wax and beeswax have also been studied in several kind composite coating layers of paper to improve the moisture barrier properties of paper (Despond et al., 2005; Garriga, 2019; Tamrin et al., 2019; Zhang, Yu, et al., 2016).

3.4. Multilayer packaging

Multilayer packaging enables the incorporation of unique functionalities of various polymers, giving rise to a package with improved performance in terms of improved barrier properties and mechanical strength (Kaiser et al., 2018). In recent years, the trend of developing multilayer packaging has been emerged not only to improve functionalities but also to reduce the cost. Mostly, with a single monolayer of polymer whether synthetic or natural, it is difficult to meet all the requirements of food packaging, including barrier properties (moisture, gas, light, flavor/odor barrier), strength, sealability, machinability, printability and aesthetic (glossy, transparent) while ensuring cost-effectiveness and adhering to all aspects of food safety (Anukiruthika et al., 2020; Kaiser et al., 2018).

In recent years, a number studies have been performed to develop sustainable multilayer packaging that possesses multiple functional properties in order to meet all the complex functional requirements of food packaging. Conventionally, multilayer packaging has been prepared using techniques such coextrusion, lamination, coating, co-injection with stretch blow molding etc. New developments in multilayer packaging preparation are electrospinning systems (Chalco-Sandoval et al., 2014), UV-curing technology (Ligon et al., 2014; Lin & Goddard, 2018) atomic layer deposition (Hirvikorpi et al., 2011; Vähä-Nissi et al., 2017; Vartiainen et al., 2014b), atmospheric cold plasma (Jothi & Nageswaran, 2019; Tyagi et al., 2018a) etc., (Anukiruthika et al., 2020). Table 2 gives the summary of examples of few recent developments in multilayer sustainable packaging.

Table 2
Recent examples of studied multilayer coatings and films for food packaging.

Packaging material (s)	Preparation method	Function	Reference
CNC-montmorillonite/CNF/paper	Mayer rod	High grease resistance and gas barrier	Tyagi, Lucia, et al. (2019)
Chitosan/PET	Layer by layer assembly	High moisture vapor barrier	Lazar et al. (2019)
Chitosan- (2-carboxyethyl)-β-cyclodextrin)/PLA	Layer by layer assembly	Antimicrobial and antioxidant functional film	Andrade-Del Olmo et al. (2019)
PLA/MFC/Paperboard	Slot-die continuous method	High grease resistance and moisture vapor barrier	Koppolu et al. (2019)
TEMPO-CNF/PE	Extrusion	High oxygen barrier while preserving water vapor barrier property	Vähä-Nissi et al. (2017)
Whey protein isolate/PE	Mayer rod with control	To improve oxygen barrier property of PE	Schmid, Benz, et al. (2012)
(PHA/PVOH/PLA)/Paper	Electrospun and lamination	Barrier to Water vapors, limonene vapors	Cherpinski et al. (2018)
CNC/Chitin/PLA	Spray coating	Barrier to Oxygen while preserving water vapor barrier property pf PLA	Satam et al. (2018)
Zein/(gelatin-Zein composite)/gelatin	Casting	Edible coating to prevent food spoilage and inhibits bacterial growth	Xia et al. (2019)
PLA/(PLA-nanoclay composite)/PLA	Blown film co-extrusion	Improved oxygen barrier property with high mechanical strength	Scarfato et al. (2017)

4. Breakthroughs in recycling and coating technology

4.1. Recycling

Recycling is currently seen as an important measure to manage packaging waste and to encourage a circular economy. For synthetic polymers, thermoplastic recycling can be carried out mechanically as well as chemically (Ragaert et al., 2017). Using mechanically recycled plastic for food packaging may increase the levels of potentially hazardous chemicals in the packaging and, after migration, also in the food (Geueke et al., 2018b). Potency of certain chemicals migrating from food packaging has been associated with chronic disease; therefore, it is of high importance to assess the safety of recycled packaging (Food and Drug Administration, 2006). The presence of non-intentionally added substances (NIAS) such as dyes, additives and their degradation products, chemicals accumulated during recycling etc., in recycled polymers can reach higher levels in recycled food packaging (López de Dicastillo et al., 2020). The presence of NIAS poses a high threat for reuse of recycled polymers in food packaging.

Plastic polymers such as LDPE, HDPE, PP, PET, PS and PVC are all thermoplastics that can be recycled mechanically and can be used as coatings for paper food packaging. In mechanical recycling, thermoplastic polymers products are washed, heated, melted and cleaned using appropriate techniques (Grigore, 2017). There are high chances of NIAS in mechanical recycled polymers, and thus institutions such as FDA, FDHA, etc., have strongly resisted the use of these chemicals in food contact. In response, many research organizations and big companies in the field are doing research to remove NIAS, odor and traces of food contamination from recycled polymers (Hopewell et al., 2009). For example, researchers have considered placing a recycled LDPE layer buried between virgin plastic layers in a multi-layered co-extruded film (Badeka et al., 2003; Chytiri et al., 2005). Composites or compounded

materials are not good candidates for recycling for food packaging for the given reasons. A recent study by Yao et al. employed the supercritical fluid extraction technique to separate waxes from PE and polypropylene plastics for their usage for food packaging (Yao et al., 2019). A recent interest in bio-based polymers such as PLA, also poses interest in their mechanical recycling and their reuse for food packaging (Andrade et al., 2016; Beltran et al., 2016).

The concept of using recycled paper itself faces many threats for its use as food contact packaging. Vápenka et al. studied all the contaminants in paper-based food-packaging. The study found that most of the major contaminants such as dibutyl phthalate, benzophenone and diisobutyl phthalate originate from materials used for modification of functional properties such as water-proof or fat-proof adjustment, coating, lamination with polymer films, etc. (Vápenka et al., 2016). A study by Jamnicki et al., demonstrates reasons for recycled paper not being a good candidate for food contact packaging (Jamnicki et al., 2012). Countries including Switzerland have completely banned the use of recycled paper and paperboard in direct food contact (FDHA, 2016; Geueke et al., 2018c).

4.2. Hybrid coatings

During the past several decades, there has been a shift in coating materials science with the increased use of fiber-reinforced plastics with materials such as carbon, glass fiber, and aramid to make stronger composites for potential applications (Faruk et al., 2012; Taj & Munawar, 2007). However, these synthetic fibers are non-sustainable in terms of their lack of renewability, biodegradability, and recyclability. Also, they have higher cost (carbon and aramid). On the other hand, several sustainable and biodegradable natural fibers such as jute, hemp, flax, banana, and nanocellulose, etc. have gained attention for their use in making polymer composites (Dixit et al., 2017; Islam et al., 2015).

The main advantages of using plant fibers in making bio-composites or semi-synthetic reinforced synthetic polymers coatings is their low weight and density, as well as less abrasiveness to the machines as compared to synthetic fibers (Bledzki & Gassan, 1999; Herrera-Franco & Valadez-González, 2004). Biopolymers or natural fibers have been studied as reinforcing agents in matrices with combinations of petro-based polymers such as PET (Owen et al., 2017), PU (Bledzki & Gassan, 1999), PP (Pandey et al., 2015), HDPE (Ning et al., 2019) and LDPE (Abdelmouleh & Boufi, 2007). All these bio-hybrid polymers have been studied to improve mechanical strength, lower the density, and provide the opportunity to produce a low cost and a sustainable and biodegradable polymer combination for potential applications in food packaging. These bio-hybrid polymers have a few limitations such as hydrophilicity, variable morphology, inferior mechanical properties, etc., which can be addressed with a more focused and application-based chemical and physical modifications (Chakrabarty & Teramoto, 2018).

4.3. Nanocomposite coatings

Nanocomposites for coatings and films are usually made up of a polymer matrix and a selected nanoparticle (Arora & Padua, 2010). The combination is a multiphase material resulting from the amalgamation of the matrix (continuous phase) and a nano-dimensional material (discontinuous phase). Based on the nano-material, the nano-dimensional phase is generally characterized as nano-spheres or nanoparticles, nano-whiskers or nano-rods, nano-tubes and nano-sheets, or nano-platelets (Bratovčić et al., 2015). Polymer nanocomposites, which are mixtures of polymers with inorganic or organic fillers with particular geometries (fibers, flakes, spheres, particulates), have been recently introduced as novel packaging materials (Prateek et al., 2016). The aspect ratio (the ratio of largest to the smallest dimension of filler) of packaging filler material plays a significant role. Fillers having higher aspect ratios typically possess more specific surface area, with associated high reinforcing properties (Dalmas et al., 2007). Various nanomaterials such as silica (Bracho et al., 2012), clay (Schutz et al., 2011), organo-clay (Ham et al., 2013), graphene (Lee et al., 2013), polysaccharide nanocrystals (Lin et al., 2012), carbon nanotubes (Swain et al., 2013), chitosan (Chang et al., 2010), nanocellulose (Sandquist, 2012) and other metal nanoparticles, such as ZnO_2 (Kudilil et al., 2013), colloidal Cu (Cárdenas et al., 2009) or Ti (Li et al., 2011) are under extensive exploration as fillers for composing nanocomposites with enhanced properties.

4.4. Fluorocarbons alternatives

Starting in 2015 some long-chain PFASs have begun being regulated or phased out due to their health risk issues with kidney, cancer, and thyroid related diseases (Barry et al., 2013; Fei et al., 2007; Stein et al., 2009). However, they have been replaced with a wide range of polymers for oil and grease resistance packaging applications. Common replacements include shorter-chain PFASs as well as polyfluorinated polyether-based polymers; those have shorter half-lives and are less bio-accumulative (Schaider et al., 2017). Some petro-based materials such as PE have also been employed for a number of packaging applications. Some other potential replacement polymers include modified starches, poly (vinyl alcohol) (PVOH), PLA composites, carboxymethyl cellulose (CMC), sodium alginate, nanocellulose, etc. (Kjellgren, 2005; Tyagi, Lucia, et al., 2019). These polymers contribute to oil and grease resistance by forming dense, non-porous, and strongly bonded coatings or films. Poly (lactic acid) (PLA), polyhydroxy alkenoates (PHAs), PVOH, and starch-based polymers are some commercially available of the emerging aqueous coatings for oil and grease resistance. They also provide odor and taste protection, temperature or chemical resistance, anti-wicking, product release with good scuff resistance, and an appropriate coefficient of friction (COF) properties, plus gluability, and heat-seal ability have potential to replace PE coatings. However, aqueous coatings do not provide a high barrier against water and water vapor.

Several patented natural grease resistant (NGR) oils are available in the commercial market, launched by big chemical companies (Adamsky, 2009; Egan, 2005). The complete identity to these commercial PFASs-free NGR are not available to academia. However, some of the recent developments on biopolymers including nanocellulose, soy protein, bio-wax, PLA, PHA have been shown to have excellent oil and grease barrier properties that can be a sustainable and biodegradable alternatives for PFASs (Aulin et al., 2010; Kumar et al., 2016a; Tyagi et al, 2018a, 2019a).

4.5. Active packaging

An intentionally designed packaging system that incorporates components that would release (antimicrobial or antioxidant agents) or absorb (oxygen or water vapor) material into or from the packaged food or the food environment is known as active packaging. Inclusion of active compounds such as antimicrobial agents, preservatives, O_2, and water vapor absorbers, ethylene removers, etc., in addition to coating polymers, renders it more effective for boosting the shelf life and quality of a food product (Arora & Padua, 2010). Active food packaging, which has been introduced in conjunction with polymer nanotechnology, aims to improve the principal features of traditional packaging systems. These can include containment (ease of transportation and handling), convenience (being consumer-friendly), protection and preservation (avoids leakage or break-up and protects against microbial contaminants, offering longer shelf life), marketing and communication (real-time information about the quality of enclosed foodstuffs), besides the nutritional constituents and preparatory guidelines (Silvestre et al., 2017).

Several approaches have been reported for the preparation of coatings for active packaging. These techniques include coatings with embedded agents for controlled release, surface immobilization by

covalent bonding or ionic interactions, layer-by-layer (LbL) assembly, and photo-grafting (Fig. 3) (Bastarrachea et al., 2015).

4.6. Aluminum in aseptic packaging

Irrespective of the immense developments in paperboard and barrier coatings, aluminum foils still play an important part in aseptic packaging such as Tetra Pak containers (Bolzon et al., 2015). Aseptic packaging for beverages is commonly made of the functionally layered composite sketched in Fig. 4. The principle function of aluminum in aseptic packaging is to act as a barrier to oxygen, flavor and light (Lamberti & Escher, 2007). An aluminum layer that comprises 5% of total packaging weight also enhances the overall mechanical strength of a Tetrapak container. Szabó et al. (2013) carried out a comprehensive study to compare aluminum and aluminum-free recycled multilayered Tetrapak (Szabó et al., 2013). They found that tensile strength, bending stiffness, and tear strength of aluminum containing packaging was greater than aluminum-free packaging by 44%, 51%, and 11% respectively (Szabó et al., 2013). However, aluminum in Tetrapak containers causes difficulty in recycling processes and yields low quality recycled pulp; the fact that a large part of this packaging is destined to landfills, thus make them less green (Karaboyaci et al., 2017). In order to address these issues, it is necessary to develop greener, sustainable, and recyclable alternative options for replacing aluminum and synthetic coatings in paper-based packaging. To our knowledge, no innovations has been reported in replacing plastic layers with biodegradable polymers in paperboard-based aseptic packaging. Fig. 4 depicts the overall structure of aseptic packaging. Tetrapak recently reported the use of bio-based PE in their aseptic packaging replacing petro-based PE in outer-most layer and cap ("Tetra Pak," 2014). However, overall structure remains non-biodegradable when landfilled or littered in the environment.

4.7. Developments in coating application methods

Several techniques are available for the application of coatings over solid substrates, and these can be divided into three basic categories based on the metering process: self-metered, pre-metered, and doctored (Fig. 5). In the self-metering process, the coating equipment itself controls the final coverage; for example, comma roll, reverse roll (Makhlouf, 2011), and dip coating (Wu et al., 2017) processes. For pre-metering, all fluid fed to the applicator is transferred to the web. The volume of solution supplied to the applicator controls final coverage for the slot die (Kumar et al., 2016a), gravure (Kapur et al., 2011) and curtain coaters (Tripathi, 2005). Doctored processes such as air knife, Mayer rod, and blade & knife coaters use the metering off action at applicator device that removes excess applied coating to control final coverage. Currently there is great interest and promising developments involving in-line applied barrier coating technology.

The most common coating techniques used for paper packaging are described briefly as follows:

4.7.1. Traditional methods

Size presses: The size press, whch is also known as "pond" or *"puddle" size press*, uses two rolls forming a nip in vertical, horizontal or inclined configurations where the coating color or surface size solution is transferred to the paper surface. As the paper web enters this nip, an excess of sizing solution is applied, forming a puddle between the sheet and each roll. A size press is most often employed to apply starch and functional additives at low solids followed by a non-contact drying or steam-heated rolls with or without further coating process. The nip load profile is very important, as an uneven load profile can cause many problems, such as runnability issues, profile issues, uneven wear of the covers, and web breaks during startups (if the nip is closing unevenly) (Hubbe, 2020; Sanchez et al., 2015).

Rod Coater: Due to the simple operation and low cost in comparison to other coaters available, the Metering Rod/Mayer Bar coater is still widely used for paper coatings. In the Mayer rod coating method, a wire-wound bar-rod is used to apply a thin film of the coating suspension over the paper substrate. This method gives users the ability to fine-tune coating thickness quickly and easily without altering the chemistry of their coating material and without time-consuming and expensive changeovers. Coating thickness can be monitored with a selection of a rods having wire wrappings of different diameters, as specified by the number designation of a selected rod. (Afra et al., 2016; Aulin & Ström,

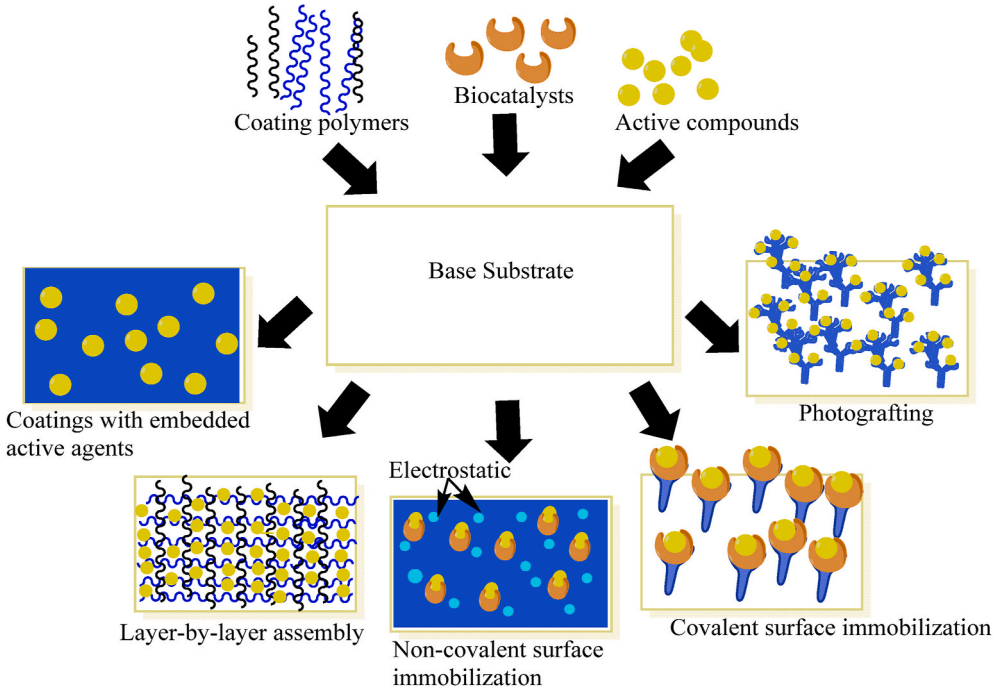
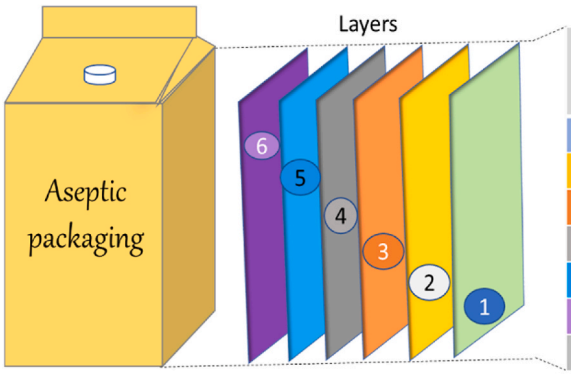


Fig. 3. A graphic description of active coating technologies with embedded agents for controlled release, surface immobilization, layer-by-layer (LbL) assembly, and photografting.



Layers	Function	Conventional material	New develop-ments
1	Protect against outside moisture	PE	Bio-PE
2	For stability and strength	paperboard	Paperboard
3	Adhesion layer	PE	PE
4	Oxygen flavor and light barrier	Aluminum foil	Aluminum
5	Adhesion layer	PE	PE
6	Seals in liquid	PE	PE
Cap		HDPE	Bio-HDPE

Fig. 4. Paperboard-based aseptic packaging layered structure.

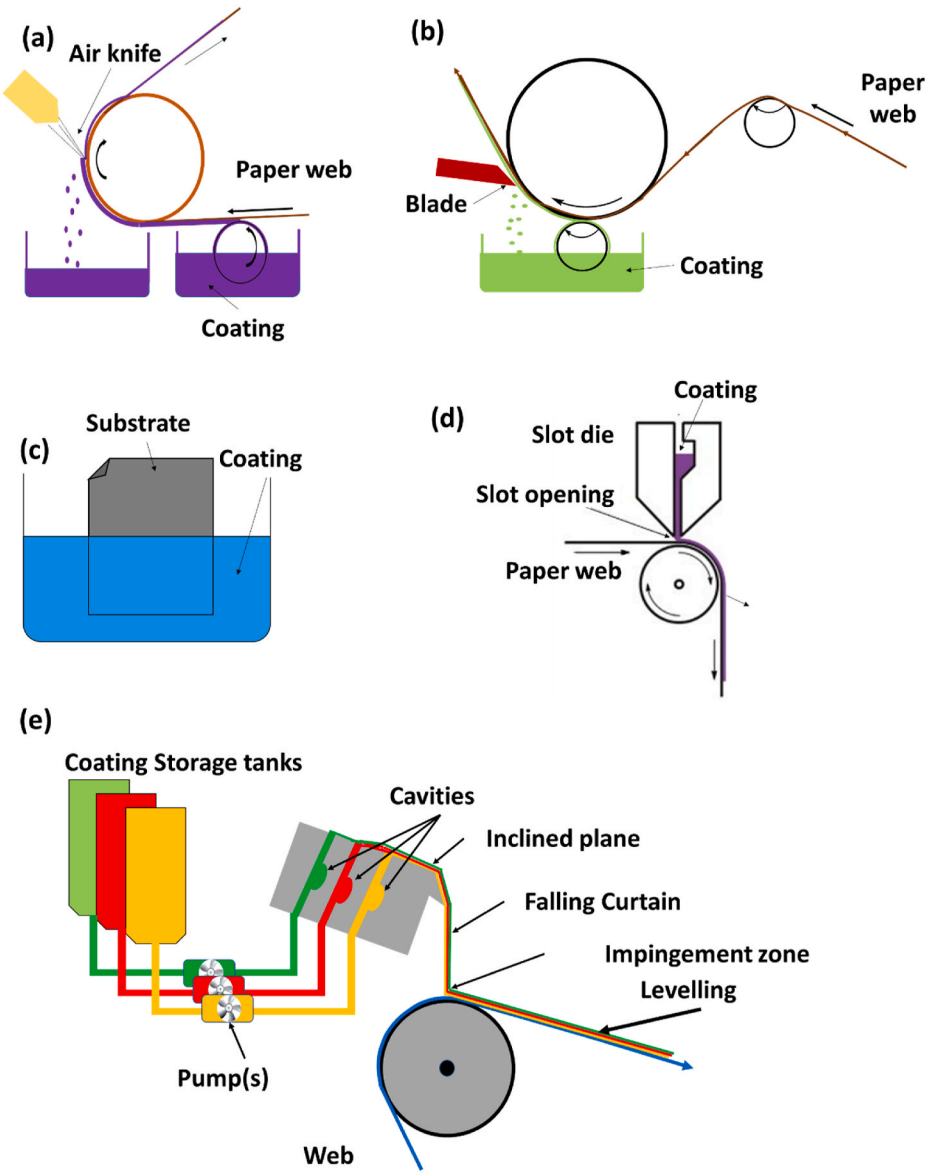


Fig. 5. Illustration of different paper coating methods (a) air knife (b) blade metering (c) dip coating (d) slot die and (e) curtain coating.

2013; Balan et al., 2015; Matikainen, 2017; Mousavi et al., 2017). The Flexrod coater is a further development based on the Metering Rod Coater. When using the Flexrod coater, the rod is loaded, via a pneumatic loading tube, onto a backing roll. By varying the air pressure/rod load pressure, the coating layer weight can be controlled. Profiling screws are used to profile the rod to the backing roll. The predominant use of flexrod coaters is as a pre-coater or back wet coater for boards.

Air-knife Coater: An air-knife coater operates by utilizing the pan-

fed kiss roll applicator, applying an excess of coating to the web (Fig. 5a). The coating layer is then metered and smoothed from the sheet using a jet of air pressure through a specially designed adjustable pressurized *Air Knife*, accurately directed onto the coating layer to shear the fluid. The excess metered coating is collected by an Air Separation Pan. The Separation Pan design changes for high speed applications (Roth et al., 2015; Rastogi et al., 2015).

Blade Coater: The blade coater is the most popular method being used nowadays for top coating of paperboard (Fig. 5b). Blade coaters have the ability to apply high coat weights with high solids and high viscosity at high speeds. Roll coating gives the best possible surface smoothness of any coating method; the coating quality is very uniform. Several variants of blade coaters have been designed and used for paper and board coatings, depending on the requirement and type of application.

For instance, the **Roll-blade coater** constitutes a pan-fed driven applicator roll to apply the coating to the sheet. This process uses a "flooded nip", which means that an excess of coating at the nip is maintained throughout the application.

A **Fountain blade coater** constitutes a fountain to apply an excess coating to the sheet, replacing the applicator roll (Hiorns et al., 1999).

The **Vari dwell fountain blade coater** can vary the dwell time/distance of the coating on the base sheet between the points of application and metering by the blade over a wide range. The independent arrangement of applicator and metering blade assemblies permits the applicator section (jet fountain) to be displaced along the backing periphery. Thus, the distance between the points of application and metering blade can be varied (Aidun & Triantafillopoulos, 1997).

4.7.2. Modern methods

Dip Coater: The dip coater is a process in which the substrate is immersed in a liquid and lifted out of the solution at a preset speed controlled by a continuous motor (Fig. 5c). The dip coating technique is used for making thin films by self-assembly and with the sol-gel technique. Self-assembly can give film thicknesses of exactly one monolayer. Dip coating is generally used in academic research, but it is not very practical for industrial coating for continuously running webs (Puetz & Aegerter, 2004).

Slot die Coater: The slot die is a pre-metered process used for coating a range of thickness uniformly. The slot die head is the main controlling component of slot die coater. It holds the fluid's temperature, distributing a fluid uniformly and defining a coating width. The die is comprised of steel body sections that house the fluid flow chamber (Fig. 5d). A slot coating die can be designed to run an individual or multiple fluid simultaneously and capable for intermittent coating capability and lane coating (Ding & Harris, 2018; Kumar et al., 2016b).

Curtain Coater: A curtain coater uses a curtain of fluid to coat the substrate, located in a gap between two conveyers (Fig. 5e). The coating fluid falls down from a reservoir tank at a certain speed between the two conveyors. The speed of the conveyers and the flow rate of material from tank determine the coating thickness. The process includes no metering step. It results in a non-uniform distribution of coating material but an even looking top surface. Curtain coating is a premetered method, which means that the amount of liquid required is supplying from the tank to the screen, to be deposited on the substrate. The coating thickness can be obtained as low as 12 μm; however, there is no problem in obtaining heavier coating coverage (Husband & Hiorns, 2005; Makhlouf, 2011).

Electrostatic Powder Coatings: Electrostatic coatings use a multi-stage process to apply fluoropolymers to a particular metal object with the help of electricity. The item to be coated is charged using a special machine, which creates a static field on the surface. Then, a dry blend of fluoropolymers is sprayed. These tiny particles are pulled strongly into the static current, forcing them to cling to the target. A thin layer of these particles is applied evenly across the entire surface. Once the surface is completely covered with the powder, the item is sent through an oven, which causes the polymers to melt and bond together. As the material cools, it forms the solid, durable layer that we have come to appreciate as a powder coated surface (Prasad et al., 2016).

Spray Coater: Spray coating is one of the newest developments introduced in paper coating application methods. In the process, coat weight and web running speed are controlled in accordance with the number of spray nozzles fitted. In spray coating, both sides of paper are coated at once, and coating quality can be improved by arranging the nozzles so that the spray fans overlap. It is important to allow the drops time to spread on contact with the paper so that all the surface is covered. An advantage of using spray coating method is a reduction of pressure on the paper web, which allows using low quality paper, cheaper furnish, cheaper pigment and higher efficiency compared with existing technology. One of the biggest disadvantages of using spray method is the need to use a low solids coating solution (Husband & Hiorns, 2005; Koskinen et al., 2000, p. 902).

Lamination with tie layer: Multilayer films/composites require sufficient adhesion between the layers for the structure to perform, especially when the materials are dissimilar. For instance, one of the two layers can be paper, and another one can be synthetic or bio-based non-polar film. To improve adhesion between poorly adhering layers, special adhesive polymers or tie resins have been developed. These resins are typically polyolefin copolymers of polar and nonpolar repeat units and with or without functional reactive groups. Typical non-reactive tie resins include ethylene vinyl acetate (EVA) and ethylene methyl acrylate (EMA) ethylene acrylic acid (EAA) and ethylene methacrylic acd (EMAA). Anhydride-modified polyolefins (AMP) are the most commonly used tie-layer resins for paper, since they are capable of chemically reacting with materials such as paper; EVOH and some metals to provide excellent adhesion (Botros et al., 2007). It has been shown that co-extrusion coupled with primers can dramatically enhance the performance of multilayer structures (Allen et al., 2011). Acid-modified resins are generally considered non-reactive but can provide good adhesion in some applications due to hydrogen bonding and polarity. In the case of metalized films or aluminum foils, tie resins with acid functionalities are often the best choice.

5. Package life cycle and end-fate

After serving its purpose, the paper packaging can have three destinations: recycling, landfilling or composting (Fig. 6).

5.1. Recovery and recycling

The most sustainable destination of used packaging would be its recovery and recycling to the same or other paper grades. Paper packaging such as cardboards, cartons for juice boxes soup, broth and wine cartons, boxboards for cereals, and cake comes under the recyclable category of paper food packaging (Marsh & Bugusu, 2007). Other food packaging such as pizza boxes, frozen food packs, paper egg cartons may

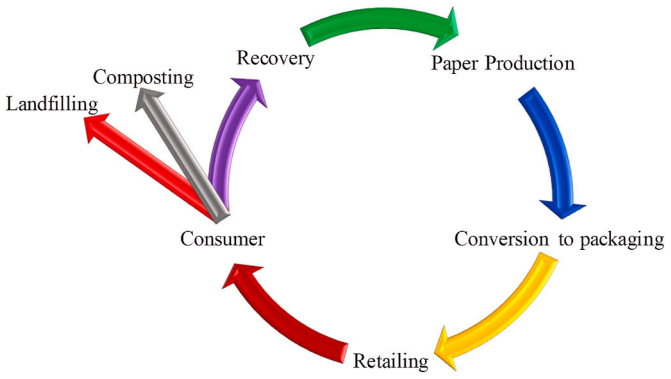


Fig. 6. Life cycle of paper-based food packaging.

also be recycled if not contaminated with the food or greases. As we have become more cognizant towards our environment, several recycling programs have been implemented in developed countries. In fact, today, 96% of the U.S. population has access to curbside or drop-off paper recycling programs according to the American Forest & Paper Association (AF&PA). Paper-based packaging accounts for 71.3% of the nearly 27 million tons of packaging materials recovered for recycling (U.S. EPA). Paperboard and corrugated board are the most easily recyclable packaging substrate in the marketplace. For example, in 2011, 75.4% of all paper-based packaging was recovered for recycling, compared to only 13.5% for plastic (Mourad & Luvison, 2014).

Plastic-coated paper packaging material not only contains paper and plastic but also many functional polymers and viscous substances, inks and metals. Thus, waste pollution to the environment is relatively large. During the paper recycling process, inks and stickies that include mineral oils are separated using steps such as deinking (froth flotation), washing and screening. Indeed, separating laminated or coated plastics from paper is a big challenge. Therefore, we must find a reasonable and feasible way to recycle the waste-plastic-coated paper plastic composite materials effectively. Only in this way can the plastic-coated paper composite packaging technology be sustainable in the long term. A few studies have been done to improve recyclability of plastic-coated paper by adding a water-soluble layer between the paper and plastic layers that enables their separation. Recently, Al-Gharrawi et al. (2021) developed a PE/CNF/coated paperboard, where CNF layer served as middle layer between PE and paperboard to improve the recyclability of paper. In the multilayer system, it was shown that the middle CNF layer weakens fast in presence of water, resulting into as easier separation of PE and paper for better recyclability (Al-Gharrawi et al., 2021).

5.2. Composting

Paper packaging that is not suitable for recycling, such as plastic-coated papers, waxed cardboards and papers, frozen food container, sugar or four bags without plastic liners, paper plates and cups are destined to composting or landfilling. EPA considers composting a form of recycling wherein the controlled aerobic or biological degradation of organic materials such as food and packaging carried out using suitable microorganisms (Marsh & Bugusu, 2007).

5.3. Landfilling

Landfilling is the most extensive method of municipal solid waste disposal. The recycling rate of paper board packaging, especially corrugated boxes, is very high, approximately 85% in developed countries such as in the USA and Canada (Haggith et al., 2018). However, still many old boxes end up in landfills. It is always better to compost an item of paper packaging than to landfill it, as the soiled paper and packaging have long been known to degrade along with food scraps. This is because one provides a source of carbon (the paper) and the other nitrogen (organics) (United States Environmental Protection Agency, 2010).

6. Future trends

6.1. E-commerce

The value of paper-based packaging demand is increasing due to a vast growth of e-commerce business. The sales channel in 2017 for just e-commerce was $28 billion, and it will more than double by 2023 (Smithers Pira report). This is causing a surge in demand for developing new designs for returnability, as well as lighter weight but stronger packaging that minimize the size of postal shipments. Another surge for demand for stronger packaging is that e-commerce is looking for options to expand the shipping by using drone delivery systems. Indeed, this will require tougher and innovative tracking smart packaging (Allen & Walsh, 2017).

6.2. Smart packaging

While talking about food packaging, there is a demand for functional and barrier coatings that will provide added benefits to the product, retailer and consumer. This demand for responsive and active packaging – including antimicrobial products and environmentally sensitive functional coatings – has now gained immense interest for industrial applications after their research field trials (Dutta et al., 2012; Lavoine et al., 2015). By reacting to the stimuli present in the food or the environment, responsive packaging systems are designed to maintain the real-time food quality. Smart packaging involves packaging systems designed with the ability to sense the changes within the food for communicating it to the consumer/user. As a smart packaging approach, the capability to perform intelligent functions such as sensing, detecting, tracking, and communicating about the quality of the packed food, is important to aid in decision-making on the shelf life safety and preserving quality of the food (Kalpana et al., 2019). The technology for smart packaging can be categorized into three types: indicators, sensors, and data carriers. The main function of indicators and sensors is communicating relating to packed food quality (Sun et al., 2020), while data carriers manage the supply chain logistics such as bar codes, radio frequency identification (RFID) tags for information tracking and tracing applications, etc. Examples of commercial indicator types include freshness indicator, time–temperature indicator, integrity indicator and gas indicators (Drago et al., 2020; Janjarasskul & Suppakul, 2018) (Schaefer & Cheung, 2018). The most common sensors in smart packaging senses changes in temperature, humidity, pH, and light exposure.

In recent years, edible sensors have been studied. These are made up of only natural and biodegradable materials without negative or dangerous effects on human health even in the long term. Some examples of materials used for producing edible sensors are red cabbage extract (contains anthocyanin-a pH-change indicator) infused pectin (Dudnyk et al., 2018), radish anthocyanin extract infused gelatin (Mehauden et al., 2007), and genipin (a natural iridoid-sensor for oxygen and biogenic detection) (Mallov et al., 2020). The advantages of smart packaging application in terms of safety and logistics, as well as marketing, indicate that intelligent packaging and active packaging could become a significant part of the industry and may even dominate it in a few years.

6.3. Bio-based coating materials

There is a growing trend in food packaging to use renewable resources to improve the sustainability and their environmental credentials. Many consumer packaged goods (CPG) companies have announced the launch of a new version of paper-based or other sustainable bio-based polymers packaging by 2025–2030, and chemical companies are assisting them to achieve their sustainability objectives (Feber et al., 2020; Reichert et al., 2020). Ellen MacArthur, a UK-based charity (Non-Government Organization) aims to develop and promote the idea of a circular economy by which plastic never becomes part of the waste and also promotes use of renewable and bio-based materials for packaging (“Ellen MacArthur Foundation,” 2009). Also, innovations in the field of bio-based polymers such as micro-fibrillated cellulose (MFC), PHA, PLA and cellulose-based polymers are opening a new potentials to build more sustainable, biodegradable and compostable packaging (Hubbe et al., 2017; Tyagi, Lucia, et al., 2019). Nonetheless, it is still needed to further study whether these bio-based films made from micro and nano fibrillated cellulose, starch, alginate, chitosan etc., can truly enhance the adaptability of new biological substrate for food packaging. For instance, low flexibility, high water absorption and water vapor permeability are some of the performance limiting factors associated with these biopolymers for their application in many packed food packaging and needs to be studied further. Also, higher cost compared to petro-based polymers put these bio-based polymers as less favorable from a commercial perspective. The innovations in bio-based materials

development offer the prospect of a biodegradable or compostable barrier food packaging capable of replacing petro-based polymers and aluminum foil (Allen & Walsh, 2017; Peter, 2017).

6.4. Tailoring of barriers for specific food products

In future studies, to go beyond the scope of the present review article, it is important to recognize that certain food products have specific requirements for protection against different permeating molecules. Consider, for example, food components such as lipids that are especially susceptible to spoilage in the presence of oxygen (Gómez-Estaca, López-de-Dicastillo, Hernández-Muñoz, Catalá, & Gavara, 2014). Foods with such components, in addition to benefiting from the barrier technologies addressed in this article, can also benefit from the use of oxygen-absorbing substances (Cichella, 2015; Dey & Neogi, 2019). Scavengers and antioxidants can work in combination with the barrier technologies. An important question that needs to be considered in future work is whether protection against just one permeant, such as oxygen, can provide sufficient protection of a particular food product of interest, even when other permeants, such as water vapor or an organic compound of interest, may be able to pass into or out from the contained food. Future primary research articles and review articles will be needed to shed more light in these areas, continually aiming toward safe, reliable, eco-friendly, and cost-effective storage and shipment of food products.

7. Conclusions

The expansion and success of the paper-based packaging market is critically dependent on the functional and barrier properties of paper and paperboard coating materials. Coatings that are water-based biopolymers are projected to supplant traditional petroleum-based waxes and plastic laminate paperboard products in food safety and security applications. In addition, nanocomposites have a bright future as emerging materials for introducing active and smart functionalities to food packaging. In the future it is expected that E-commerce will have a great impact on development of coatings for smart food packaging with more functional and barrier properties. Although the economically viable use of advanced biomaterial films in packaging has not been fully established yet on an industrial scale, such films have a great potential for food packaging market with upcoming developments in nanobiomaterials. Although plastic-based applications are growing twice as fast as compared to those using paper for food packaging, recyclability is increasingly viewed as important by consumers. This preference indicates a primary advantage of coated paper and board and encourages new developments in related fields.

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Declaration of competing interest

The authors declare no conflict of interest.

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