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REVIEW PAPER

Biopolymer-based coatings containing active ingredients for cellulosic packaging: A review

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Abstract Cellulosic material is considered to be excellent for food packaging due to its low cost, easy degradability, and high recyclability rates. However, its application is still limited due to its porous structure and high hydrophilicity, which provide high permeabilities to grease, gas and water vapor. One solution to fix these issues and to bring new functionalities that can also extend food shelf life is to coat the paper substrate with biopolymers containing nanoparticles and/or components with antioxidant and/or antimicrobial activities. In this regard, the aim of this study was to review some aspects of the current knowledge critically and didactically on applications of biopolymer-based coatings incorporated with active components and/or nanoparticles on paper/paperboard materials usually applied as primary cellulosic active packaging. Studies have shown that this combination can positively improve the physical, mechanical, and barrier properties of cellulosic packaging, along with the enhancement of antioxidant and antimicrobial activities. A better understanding of these aspects enables the use of other active compounds to develop more functionality to the packaging itself as well as to apply it to food packaging in general.

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Introduction

The development of more innovative, safe, and sustainable food packaging has become a major concern in the food industry for all links in the food supply chain. The reason is that the basic function of packaging is not only to protect and preserve the food, but also to reduce the overall environmental impact of the product and packaging system throughout its entire life cycle (Bahl et al. 2021; Haghighi et al. 2020; Rhim et al. 2013; Robertson 2013).

In this context, and considering consumer concerns, food industries have turned their attention to the use of primary packaging made of cellulose (paper and carton), mostly because they are from renewable sources, relatively easy to recycle and biodegradable (Divsalar et al. 2018). The biodegradability of cellulosic material in soil and seawater is mainly due to the hydrophilicity of cellulose which accelerate the hydrolysis of glycosidic bonds allowing action of numerous enzymes/microorganisms to digest amorphous cellulose chains (Tang et al. 2023).

Normally, non-renewable synthetic polymers are applied as cellulosic packaging coatings and act as barriers to gases, humidity, and grease. Apart from that, those materials can provide good mechanical resistance even in high moisture environments, which is an important characteristic for food preservation. However, due to the environmental impacts that fossil-based materials can bring along, there is an accelerated increase in laboratory scale studies on the use of biopolymer-based coatings for cellulosic packaging (Khwaldia et al. 2010; Guazzotti et al. 2015), remembering that according to IUPAC (2019), biopolymers are "macromolecules (including proteins, nucleic acids and polysaccharides) formed by living organisms."

Several biopolymers including polysaccharides (starch, chitosan, alginate, etc.) and proteins (gelatine, wheat gluten, whey protein, etc.) have been used as coatings for paper and paperboard (Battisti et al. 2017; El-Wakil et al. 2015; Hefft 2018; Koivula et al. 2016; Reis et al. 2011; Santos et al. 2023; Wu et al. 2018; Yoo et al. 2012). Yet, problems related to low mechanical and barrier properties may still persist (Andreuccetti et al. 2017; Soltanzadeh et al. 2022). As an option to enhance the behavior of biopolymers, nanoparticles such as zinc oxide (Wu et al. 2018), montmorillonite clay (Wang and Jing 2017), and cellulose nanocrystals (El-Wakil et al. 2015) were added to the coating-forming solution (CFS).

Another option is the incorporation of fat-soluble components with antioxidant and/or antimicrobial activities in CFSs allowing the development of active packaging, also with improved functional and barrier properties (Doğan et al. 2022; Silva et al. 2022). Active packaging contains specially designed structures incorporated with components intended to release or absorb substances into or from the packaged food or from the environment surrounding the food (Dainelli et al. 2008). These components can be added into sachets placed inside the packaging or can be added to the material used to build the packaging itself (Restuccia et al. 2010). For the latter, the active components can perform by scavenging oxygen, carbon dioxide, moisture, and ethylene, among other chemical species, or releasing compounds such as ethanol, antimicrobials, antioxidants, among others (Restuccia et al. 2010; Yildirim et al. 2018).

In this context, the aim of this paper was to review some aspects of the current knowledge critically and didactically on applications of biopolymer-based coatings incorporated with active components and/or nanoparticles on paper/paperboard material usually applied as primary cellulosic active packaging. A discussion regarding the characterization methodologies used in each reviewed article is out of the objective of this manuscript. Readers interested in the methods used for paper/paperboard packaging characterization are invited to consult classical books such as Holik (2006), Kirwan (2013), and Popil (2017), among others.

Greater understanding of how the coatings have been developed and applied will stimulate improvements of these materials to promote the extended shelf life of food products in an environmentally friend manner.

Concepts of food packaging

The primary food packaging must protect the product by delaying deterioration processes, such as discoloration, lipid oxidation, loss of aromas and nutritional value, as well as microbiological activity, among others. Therefore, while the primary packaging is closely related to these aspects of food preservation, the secondary and the tertiary packaging are more related to the mechanical protection of the product against shocks and vibrations.

Plastic materials are the most common material used as primary food packaging due to their low cost, properties, versatility, durability, and high strength-to-weight ratio (Geyer et al. 2017; Marsh and Bugusu 2007). However, the use of different polymers in multilayer structures and as paper coatings can compromise recycling. This is mostly due to the chemical incompatibility of each present component, which results in different thermal transition temperatures and thermal stabilities. Thereby, the homogeneity and therefore, the quality of the recycled material can be remarkably inadequate or even unpractical for latter uses such as packaging. Without new technologies that enable the return of these materials to the production system by addressing this issue, their increasing use can worsen the environmental impacts related to them perceived only on the last decades (Yao et al. 2022).

Single-use plastics are the plastic-made packaging designed to be disposed right after the packed product is consumed or used, which goes against the principles of Circular Economy (Stahel 2016, Ellen MacArthur Foundation 2022). Few examples are straws, supermarket bags, shopping bags, bottles, coffee cups, and Styrofoam containers. One of the issues with this type of packaging is related to its great volume generated daily, which implies in a high amount of virgin material use, along with the problem of waste management, since they are not always easily recycled, especially if contaminated with the product for food packaging. The latter can contribute to post-consumer plastic being destined to landfills, which is not a long-term solution, as well as to leakages to environmental ecosystems. Since recycling is strongly dependent on government legislation, collection and management infrastructure, alongside consumers' habits, new packaging systems considering the maintenance of the materials into the consumption system for longer, like refillable and reusable packaging, must be put into practice at scale (Merrington 2017).

Those issues highlight that sustainability questions related to single-use plastic packaging are of great concern (United Nations Environment Programme 2018). Still in this context, Europe banned in 2021 certain single-use plastics (Directive (EU) 2019/904) and other countries have been stablishing new restricting rules for plastic use in packaging. In the face of the different scenarios of plastic bags, the packaging market has been focusing on substituting them by cellulosic alternatives.

Cellulosic packaging

Cellulose is the main fibrous raw material used to produce paper, paperboard, corrugated board, and similarly manufactured products (Robertson 2013). It is considered the most abundant biopolymer in nature and attracts the interest of the global packaging market due to its low cost, good mechanical properties, availability, and light weight.

There is a sign of growth in cellulosic packaging consumption worldwide. For example, paper corresponded to 50% of all materials used for packaging purposes in Europe in 2020, which represented 10% more than plastics in the same year (Coelho et al. 2020). Yet, cellulosic packaging can have several limitations due to its porous structure and hydrophilic character related to the cellulose fibers. Moreover, high sensitivity to water (Li and Rabnawaz 2018), grease, and high permeability to gases and water vapor make food packaged in pure cellulosic materials poorly protected from external degrading factors (Nechita and Roman 2020).

To overcome those limitations, the packaging market has been working on laminated material or applying coatings over cellulosic packaging so the needed properties for food storage can be reached. Laminations with aluminum and synthetic polymers and coatings based on waxes and synthetic polymers are already well spread as commercial solutions (Khwaldia et al. 2010; Tyagi et al. 2021). However, those materials can bring other issues, which are discussed on the next topic.

Cellulosic packaging with conventional barrier and bio-based alternatives

Currently, several materials have been applied on paper mainly to deliver barrier (against moisture, grease, water, oxygen etc.), and to improve its mechanical resistance. The way to do so is by using adhesives to bond together laminated structures or by extrusion coating of molten plastics onto the cellulosic substrate. Among the synthetic polymers commercially available for the former process, polyolefins such as polyethylene (PE) and polypropylene (as PP or as BOPP when biaxially oriented) are the most present, along with polyethylene terephthalate (PET), and aluminum foil.

Both low-density and high-density polyethylene (LDPE and HDPE, respectively) are used in laminates with cellulosic substrates, such as in cardboard disposable containers and in many other applications. Because of its branched chains, LDPE is versatile enough to be processed by extrusion, blow molding, or injection molding techniques, with the former being the most used process to coat paper followed by a cold pressure nip step. This cold pressure nip procedure is responsible for intermixing the paper fibers with the polymer matrix, which can make it difficult the recovery of the fibers during paper repulping

(Marichelvam and Nagamathan 2017; Al-Gharrawi et al. 2021).

Disposable paper cups, for instance, are extrusion coated with a thin layer of LDPE which prevents the incorporation of the liquid product into the cup and promotes the necessary mechanical strength for manipulation (Akhdhar et al. 2022). It is indeed most frequently material used as coating for cellulosic food packaging because of its characteristics such as good mechanical behavior, barrier against water vapor, low sealing temperature, and good chemical resistance. The combination of these features can be responsible for prolonging the shelf life of most moisture sensitive foods (Rojas et al. 2019), especially for those that are not very sensitive to oxygen, since PEs do not offer great oxygen barrier (Liu et al. 2017).

Polypropylene (PP) is another polyolefin extensively used in multilayer structures with paper due to its low cost and low water vapor permeability, the latter being linked to its higher crystallinity compared to LDPE, along with its relatively ease of processing (Lahtinen et al. 2009; Wagner et al. 2016). The biaxial orientation of the film promotes the alignment of the crystalline regions, improving its barrier properties even for thin films (Lahtinen et al. 2009; Maddah 2016). However, its limited oxygen barrier does not allow it to be used as paper coating for food that are very sensitive to oxygen (Kim et al. 2018; Ray and Okamoto 2003).

Polyethylene terephthalate (PET) is a semicrystalline thermoplastic, like the others previously mentioned, used worldwide to produce bottles and as outer layer in multilayer flexible packaging mostly due to its high rigidity and excellent optical properties. However, PET does not offer moisture barrier to films, which can be outlined by combining it with other materials (Jin et al. 2015; Majdzadeh-Ardakani et al. 2017; Poulikakos et al. 2017; Wu et al. 2012; Zou et al. 2007).

Aluminum foil can also be applied in laminated paper and paperboard packaging. The purpose of the foil is to serve as a more robust barrier to light, oxygen, moisture, odor and aroma (Rodushkin and Magnusson 2005). As long as the aluminum foil is not compromised with cracks or pinholes, it is considered a complete barrier, which can help prevent food losses and waste. As drawback, the presence of aluminum layers can add issues regarding recyclability, like the need to replace filters used in recycling machines more often, and unwanted activation of metal detectors when packaging made out of recycled content carries them (Fiselier and Grob 2011). In this case, polymers are preferable, but hardly represent complete barriers to cellulosic packaging, and they can awaken the interest of consumers for more environmentally friendly solutions.

Alternatively, some authors have been trying to increase the recovery of the plastic and the paper contents of coated paper packaging. Al-Gharrawi et al. (2021) reported that the application of PE over a cellulose nanofiber (CNF) layer previously applied onto paper improved the oxygen and the oil barrier properties of the cellulosic packaging. In addition, the CNF limited the penetration of the PE coating into the paper porous structure, facilitating paper recovery during the recycling tests. This can be explained by the limited compatibility and poor adhesion between the CNF and the PE layers (Li et al. 2010, 2014).

The use of conventional fossil-based materials as coatings or laminates for paper packaging raises the concern regarding circularity, since excessively complex structures are rarely recycled at" current mechanical recycling plants. For laminates, the strength of the adhesion between the film and the paper can even reduce the amount of fiber that is recovered at the end of the processes (Confederation of Paper Industries 2024). The challenge of dealing with complex structures, which is also one of the main drawbacks of multilayer plastic packaging, has been forcing the brand owners to move their packaging towards monomaterial, simpler and recyclable structures (Ellen MacArthur Foundation 2022).

In order to address environmental concerns related to the current high dependence on petroleum-based materials, the scientific community's attention has been driven to the development of paper packaging made completely, or at least high content of, renewable resources. Some examples of highly investigated biopolymer-based coating materials are starch, sodium alginate, chitosan, cellulose derivatives, wheat gluten and whey protein, which are all discussed as follows.

Starch is a polysaccharide composed of repeated D-glucose units arranged in two different ways. The amylose domain is characterized by a linear α-1,4 bonded structure, whereas the amylopectin arrangement is formed by an α-1,4 backbone containing α-1,6 branches along it. The balance between both components, which is dependent on the plant source, directly influences the properties of the coatings made from starch. The excellent oxygen barrier of starch-coated paper packaging, when dry, can be attributed to the hydrogen bonding between chains, which diminishes the mean free path for molecule diffusion. This strong intermolecular interaction is also responsible for its brittleness, since it is linked to higher crystallinity (Fabra et al. 2021; Thakur et al. 2019; Versino et al. 2016). The latter characteristics along with the moisture sensitivity of starch can be overcome by structure modifications and combinations with other materials. Chi et al. (2020) have derivatized starch by adding a positive group to it in one batch, and a negative group to it in a second batch. Following that, the author mixed both solutions and dip coated the paperboard substrates. The blend containing high molecular weight cationic and anionic starch increased the tensile strength of the paperboard by 18% and the Young's modulus by 21%. Meanwhile, its barrier properties against water vapor were increased by 40%, and regarding grease permeability, determined by the Repellency of Paper and Board to Grease, Oil, and Waxes test, also called as KIT test, was raised from 0 to 12.

Other chemical modifications of the OH groups in starch have been used to provide the necessary water resistance to this polysaccharide. Le and Nguyen (2020) have esterified starch with triglycerides from soybean oil to make it more hydrophobic. The water contact angle (WCA) reached for the paper coated with the modified starch was 121º, indicating that the material became highly hydrophobic. This result was even greater than the ones found by Winkler et al. (2013), mostly because the former used triglycerides composed mainly by C_{18} backbones, while the latter used a C_{12}-fatty acid. Besides, surface morphology was also attributed by the authors to corroborate with the hydrophobic properties of the modified material.

The incorporation of minerals such as bentonite (Breen et al. 2019), calcium carbonate (Thitsartarn and Jinkarn 2020) and montmorillonite (Wang and Jing 2017) increases the tortuosity of the path faced by migrating substances, which enhances the barrier properties of the coatings. Water–vapor transmission rates (WVTR) can reach values as low as under 10 g. m^2.day^{-1} with these materials.

Sodium alginate has been also applied as barrier coating onto cellulosic packaging for food contact (Kopacic et al. 2018). It is composed of α-1,4-L-guluronic acid blocks (GG), β-1,4-D-mannuronic acid blocks (MM), and heteropolymeric sequences of M and G (Rinaudo 2014). The acidic side groups, in the presence of cations are responsible for ionic crosslinking between adjacent chains, which are related to the good gas barrier properties of alginate films and coatings when dry (Singh et al. 2020).

Sodium alginate blended with carboxymethyl cellulose was applied by means of a coating machine to produce greaseproof papers. A KIT number of 9 was obtained with a coating weight as low as 1.9 g/m^2, which resulted in a good oil resistance and met the minimum requirement of a KIT value of 5 from the KIT test for the material to be considered as greaseproof (Sheng et al. 2019). The KIT number in this work has shown to be greater than the KIT number of 3 found in another work for the coating of paper with only sodium alginate (grammage of 3.06 g/m^2) (Wang et al. 2022). However, Kopacic et al. (2018) obtained a KIT value of 12 for alginate coated over a primary fiber paper (coating grammage of 6 g/m^2). Regarding the water–vapor transmission rate, that of the alginate-coated paper of the latter study was reduced by 35%, at 23 ºC and 50% of relative humidity, compared to the uncoated paper substrate. However, the obtained value can still be too high (around 400 g_{water}/m^2.day) for the packing of water-sensitive products. Indeed, uncoated paperboard and paperboard coated with mixtures of alginate/calcium chloride, and alginate/organically modified montmorillonite were found to behave the same towards water vapor permeability (WVP) (Rhim et al. 2006).

Limited research has been found regarding the improvement of alginate water sensitivity for paper coating, which indicates it might suit more appropriately for oil and grease barrier purposes.

Another well-researched polysaccharide for paper coating applications is the chitosan. It is obtained via a deacetylation reaction of chitin, which is mainly extracted from exoskeleton of crustaceans, also from fungi and insects, among other sources. Chitosan is thereby composed of N-acetyl-2-amino-2-deoxy-D-glucopyranose (portion of chitin that has not been deacetylated) and 2-amino-2-deoxy-D-glucopyranose (deacetylated portion of chitin), with the repeating units being linked by β-(1→4)-glycosidic bonds. Chitosan is a linear polysaccharide containing -NH_2 and -OH functional groups, which confer a high polarity to its backbone. Besides, the former group is responsible for its antimicrobial properties when protonated, since with a positive charge it interacts easily with the phosphoryl groups in the cell membrane of microorganisms (Chatterjee et al. 2022). Chitosan is not soluble in water but it can be solubilized by addition of an acid into dispersion (Sobral et al. 2022).

Paperboard coated with a protonated chitosan solution was tested on the work of Naitzel et al. (2023). The WVP of the one-layer-coated paperboard was close to 47% lower than the one for the uncoated paperboard (control) even with no significant differences of thickness and grammage at 95% of confidence between them. The same coating formulation was able to provide a KIT number raise from 3 (control) to 8 (one-layer coating), which indicates the great potential for chitosan to act as barrier toward oil and grease. Regarding mechanical properties, there was also no significant differences of tensile index and elongation at break between control and one-layer-coated paperboard, possibly due to absorption of the coating-forming solution in the paperboard pores instead of a continuous coating formation, as pointed out by the authors. On another work, a similar formulation was applied onto paper as 1, 2, 3 and 5 layers. The WCA was raised from 66.7 ± 2,4 (uncoated) to 94.7° ± 2.8° (one-layer coating) at 0 s. The WCA for the multilayer-coated substrates were the same as for the one-layer coating. This suggests that the coating enhanced the hydrophobicity of the material, independently of the number of coating layers (Tanpichai et al. 2022).

Bhardwaj et al. (2022) coated paper substrates with a mixture of chitosan and beeswax (2.0 to 2.5 g/m^2 of grammage). In addition to not being able to significantly improve the mechanical strength of the coated papers, the chitosan-and-beeswax mixtures contributed with only 20% to the reduction of WVP. In a similar approach, Wang and Jing (2017) used montmorillonite/chitosan nanocomposite coating applied to package paper by the roll coating machine. The formulation was responsible for only an 11% reduction on WVP for a coating grammage of 3,53 g/m^2. Both studies highlight how challenging it is to overcome the hydrophilic effects of this biopolymer.

Chemical modifications of chitosan have been proven to address its high sensitivity to water. Zhu et al. (2023) crosslinked chitosan and tannin extract-based epoxy followed by its application over paper substrates. The obtained KIT value for the coated paper varied from 8.6 to 12 depending on the amount of tannin added to the coating. Regarding oxygen and water vapor permeabilities, they varied from 307.1 to 136.2 cm^3/(m^2.day.0.1 MPa) and from 673.5 to 298.6 g_{water}/(m^2.day), respectively, both variables related only to the tested coated paper containing varying proportions of tannin. The uncoated paper presented an oxygen permeability of 597.1 cm^3/(m^2.day.0.1 MPa) and a WVTR of 1252.9 g_{water}/(m^2.day). Both tensile strength and elongation at break were raised from 6.46 to 7.95 kN/m, and from 1.7% to 2.8%, both comparisons between the uncoated and the substrate coated with the formulation that performed the best regarding the two variables. The mechanical properties of the coated samples were maintained after aging under 340 nm UV light for 24 h, whereas the uncoated paper presented reductions of 32% of tensile strength and 19% of elongation at break.

Tan et al. (2023) derived chitosan with cardanol glycidyl ether at different ratios and found that compared to the uncoated substrate, the formulation that performed the best diminished the WVTR by 57% (90% of relative humidity (RH)) and by 73% (50% of RH), and the oxygen permeability by 34%. The Kit test value was also raised from 0 to 11, and the mechanical properties were also improved by the increase of both tensile strength (125%) and elongation at break (130%).

Cellulose has also been well explored as paper coating in many ways. For such application, cellulose can be found in the literature as nanocrystal (CNC), nano and microfibril (CNF and CMF, respectively), in combination with other materials, as well as in chemically modified structures. Nano-and-micro-scale cellulose is composed of D-glucopyranose units linked together by β-(1,4)-glycosidic bonds, being produced from plant cells and from microbial synthesis. The -OH groups on its backbone are responsible for strong intra and intermolecular hydrogen bonds, which result in great interactions between chains (great mechanical properties), but also in its susceptibility towards water permeation (Spagnuolo et al. 2022).

On the work of He et al. (2021), the authors combined carboxymethyl cellulose with CNC-immobilized silver nanoparticles (CNC@AgNP). As a result of the coating, WVP was reduced by 45.4% with 7% of CNC, probably because of the increase on tortuosity caused by the CNC@AgNP dispersion. Regarding tensile strength and elongation at break, the improvement of both parameters compared to the uncoated paper was attributed to CMC and not to CNC@AgNP, since there was no significant difference between them independently of the concentration of the latter.

TEMPO-oxidated CNF (TOCNF) was combined with beeswax as a Pickering emulsion for paper coating, reaching WCA values of 71.3° when 0.7% of TOCNF was present as stabilizer, and 96.1° when 0.9% of it is in the suspension. Since the WCA of uncoated paper was not able to be determined due to the great interactions between paper and water, these results revealed a good enhancement on surface hydrophobicity of the coated substrates. Moreover, the KIT test values were also improved from 1 (uncoated paper) to 5 (coated paper) (Bayés et al. 2023).

On another work, the authors applied combinations of CNF, alkyl ketene dimer and alkenyl succinic anhydride for water vapor barrier, and CNF, sodium alginate, poly(vinyl alcohol) and illite (a clay mineral) for grease resistance. Even though the maximum decrease on WVTR was 35%, which is still in the same order of magnitude of the uncoated paper, the grease resistance was substantially enhanced, from 0 (uncoated) to 11 (coatings containing high contents of illite and sodium alginate) (Mazega, et al. 2022).

Yi et al. (2023) tested a multilayer paper coating containing chitosan, modified CNF (grafted methyl methacrylate on CNF) (MCNF) and zein. Chitosan and MCNF were expected to promote oil resistance to the packaging, MCNF and zein were expected to deliver water resistance, while zein was thought to give heat-sealing property to the coated paper. The coating structure was able to act as a great barrier towards oxygen and water vapor, with values from 1935 ± 152 $cm^3/(m^2.day)$ (uncoated) to 55 ± 10 $cm^3/(m^2.day)$ for the former and from 1107 ± 106 $g_{water}/(m^2.day)$ (uncoated) to 43 ± 5 $g_{water}/(m^2.day)$ for the latter. Regarding grease barrier, with a zein layer ranging from 1 g/m^2 to 5 g/m^2, the KIT number varied from 9 to 12, which shows that the zein layer considerably contributed to oil repellence.

Other biobased materials available for cellulosic packaging coating are proteins, such as wheat gluten and whey protein, among others. A new approach was presented by Rovera et al. (2020) to improve the barrier properties of paperboard using wheat gluten and silica for this purpose. The coating improved the water vapor barrier of the paperboard by 74%, but the experimental results highlighted some limitations like high brittleness due to the inherent rigidity of the inorganic phase thus limiting market applications. This problem could be avoided by addition of a plasticizer, as glycerol, in the coating-forming solution (Sartori et al. 2018).

Whey protein coatings have good oil barrier properties and reduced water vapor permeability (Yoo et al. 2012). The combination of rice bran wax and whey protein isolate has been used to coat the outer surface of paper cups for popcorn packaging by immersion. The results showed that the coating increased thickness and yellowness, while reducing Young's modulus, and WVTR. Also, the analyses of the surface morphology indicated that the coating covered surface pores (Zavareh et al. 2021). Jeong and Yoo (2020) reported that paperboard coated with whey protein concentrate, beeswax, and sucrose presented good water barrier properties due to the hydrophobic characteristics of beeswax and good oil barrier properties because of the polar moieties of whey protein concentrate.

Active biopolymer-based coatings for cellulosic packaging

The formulation of active biopolymer-based coatings comprises the biopolymer itself, the solvent, plasticizers, and active compounds (Dammak et al. 2021). In addition, nanoparticles can be added into the formulations in order to improve the physical and the functional properties of the coatings contributing to extend the shelf life of the food (Alexandre et al. 2016; Aziz and Karboune 2018; Jamróz et al. 2018).

Regarding the active compounds used in formulations of coating with the purpose of improving the antioxidant and antimicrobial activities of cellulosic packaging, the following stand out: citric, sorbate, acetic, benzoate, and propionate acids; nisin and essential oils; among others (Battisti et al. 2017; Divsalar et al. 2018; Quintavalla and Vicini 2002; Syahida et al. 2021). In the case of essential oils, they can be added in the form of an emulsion (Jamróz et al. 2018), but such compounds can be lost in the development of the coating and in the conditioning period. Due to that, other forms of dispersions of these lipophilic compounds into biopolymer matrix have been studied, such as the use of nanoemulsions (Dammak et al. 2017) or Pickering emulsions (Dammak et al. 2019).

In terms of nanoparticles that can be incorporated into coatings, zinc oxide has received much attention because it has antimicrobial potential, in addition to improving physical properties (Sharma et al. 2020a, b; Wu et al. 2018). Other nanoparticles, such as laponite, montmorillonite and nanocellulose, can also be used to reinforce the biopolymeric matrix (Alexandre et al. 2016; Valencia et al. 2015; Tessaro et al. 2021).

Some biopolymers have proven to possess natural antimicrobial activity, such as chitosan (Yan et al. 2021). However, the incorporation of some fat-soluble components into the coatings can be responsible for the improvement of their antibacterial and antioxidant properties (Ranjbaryan et al. 2019). Mustapha et al. (2019) developed a coating based on cassava starch that incorporated turmeric oil through simple stirring. These authors concluded that the paper showed high mechanical and functional properties and can be used to extend the shelf life of food. However, the application of starch in packaging can be compromised due to its hydrophilicity and non-resistance to microorganisms as discussed before (Ni et al. 2018), especially if glycerol was used as plasticizer (Bergo et al. 2010).

Based on this motivation, several researchers have attempted to improve these characteristics. An example of this is the use of metallic nanoparticles that can significantly improve the hydrophobic surface and the antimicrobial activity of the material (Perkas et al. 2016). Filter papers were coated with composite dispersions made of corn starch and zinc oxide nanoparticles, in which the material presented excellent hydrophobicity and antimicrobial activity against *Escherichia coli* (Ni et al. 2018). The antimicrobial activity of zinc oxide nanoparticles can be explained by the particle coverage formed on the bacterial surface due to the electrostatic forces present (Devrim and Bozkir 2017; Zhang et al. 2008).

In another study, papers coated with silver nanoparticles, chitosan and starch composites demonstrated increased antimicrobial activity against Gram-negative (*E. coli*) and Gram-positive (*S. aureus*) bacteria, which also improved its barrier, mechanical, and oil resistance properties. The increase of these properties occurred due to the combination of the materials and the formation of stronger hydrogen bonds between the amine group of the chitosan and the hydroxyl group of the starch (Xu et al. 2005).

Chen et al. (2022) applied a composite made of micro-fibrillated cellulose and nano-scale silica (n-SiO2) as a potential antimicrobial mixture to the development of new active coatings for paperboard. The coating was sprayed on both surfaces of the paperboard base and the material demonstrated a higher tensile strength and lower permeabilities against air (1.3108 to 0.00317 $\mu m.Pa^{-1}.s^{-1}$), oxygen ($>100{,}000$ to 10 $cm^3.m^{-2}.day^{-1}.atm^{-1}$), and water vapor (1,997 to 378 $g.m^{-2}.day^{-1}$) for a coating grammage of 70 g/m^2.

Shankar and Rhim (2018) coated a paper package with a blend of alginate, carboxymethylcellulose, and carrageenan gum, containing grapefruit seed extract. The coating forming solution was applied by casting it onto the surface of the food wrapping paper. These authors reported that the coating significantly increased the water and grease resistance and mechanical properties of the paper, and it showed strong antimicrobial activity against *L. monocytogenes* and *E. coli*. In addition, Wu et al. (2018) prepared an antibacterial coating for filter paper based on sodium alginate and zinc oxide. The tensile strength and the Young's modulus were increased in 40 and 31%, respectively, when compared to the uncoated paper. Besides, the coating showed good antibacterial activity against *E. coli* and *S. aureus*.

Some authors have tested the performance of active coatings for cellulosic packaging on shelf-life extension of food products (Table 1) and described in detail next. The studies have shown that cellulosic packaging can be applied to the preservation of products susceptible to lipid oxidation or microbial deterioration, such as meat, cheese, milk, almonds, fish, chocolate, and others (Ceylan et al. 2021; Botelho et al. 2014; Ghaedi and Hosseini 2021; Carta et al. 2022).

Battisti et al. (2017) applied a gelatin-based coating incorporated with citric acid (1 and 2% relative to the solution mass) to paper using a low-pressure compressed air pistol. Citric acid was shown to disrupt enzymatic activity, proteins and DNA structures within some microorganisms' cells and their extracellular membrane by compromising the adequate functioning of their electron transport system (Park et al. 2011). The coating improved the water vapor barrier

Table 1 Some studies of the application of cellulosic packaging with biopolymer-based coatings containing active compounds

Biopolymers	Nanoparticles/active compounds	Substrates	Incorporation method	Application of coated paper	Improved properties	References
Alginate/ Carboxymethylcellulose/ Carrageenan	Grapefruit seed extract	Wrapping food paper	The paper was coated with a wire coated bar	Minced fish paste	The coated paper showed strong antimicrobial activity against pathogenic bacteria (*Listeria monocytogenes* and *Escherichia coli*) and high mechanical and water vapor barrier properties	Shankar and Rhim (2018)
Alginate	Thyme essential oil	TSL testliner	An automatic film applicator was used using a smooth roller for coating paper	No application	The active packaging showed antioxidant and antibacterial activity against Gram-negative (*E. coli*, *S. typhimurium*) and Gram-positive (*L. monocytogenes*) bacteria	Aguado et al. (2024)
Chitosan	Not used	Paperboard	Not mentioned	Bilberries and redcurrants	The active packaging showed antimicrobial activity against *Botrytis cinerea*	Hefft (2018)
Chitosan	Rosemary oil/zein	Paper with a weight of 80 g/cm^2	An automatic film applicator was used and then the paper was dried at 40 °C for 10 min	No application	The papers presented high thermal stability and the combination of rosemary oil with zein reduced the water vapor permeability	Vrabič Brodnjak and Tihole (2020)

Table 1 (continued)

Biopolymers	Nanoparticles/active compounds	Substrates	Incorporation method	Application of coated paper	Improved properties	References
Chitosan	Cinnamon essential oil	Wax paper sheets	The paper was subjected to immersion in a chitosan solution with emulsified oil and then dried at 25 °C for 24 h	Fish patty	Papers coated with chitosan incorporated with cinnamon essential oil showed greater antimicrobial, antioxidant and chromaticity activities, significantly improving the shelf life of fish burgers during storage	Valizadeh et al. (2020)
Chitosan	Lemongrass essential oil	Paperboard	An automatic film applicator was used at a speed of 10 mm.s^{-1} to coat the paper. Subsequently, the material was dried at 120 °C for 90 s	Talharim pasta	The active coating showed antimicrobial activity against weevil infestation in cereal-based food-packed products	Silva et al. (2022)
Chitosan/zinc oxide	Nisin	Paper cellulosic	The paper was dip coated for 1 min and dried at room temperature for 24 h	White cheese	The coating had an inhibitory effect on *Listeria monocytogenes*	Divsalar et al. (2018)
Gelatin	Citric acid	Raw paper long fiber	The paper was coated using spraying at low pressure compressed air pistol	Packaged beef	The beef packaged in the active papers showed greater stability to lipid oxidation and greater reduction of the microbial population in 4 days of storage	Battisti et al. (2017)
Gelatin/palm wax	Lemongrass essential oil	Kraft paper	Immersion and drying under different conditions: 24 °C for 48 h 50 °C for 30 min 24 °C for 24 h	Ground beef	Coated kraft paper reduced lipid oxidation and microbial spoilage of ground beef	Syahida et al. (2021)

Table 1 (continued)

Biopolymers	Nanoparticles/active compounds	Substrates	Incorporation method	Application of coated paper	Improved properties	References
Starch	Silver nanoparticle	No. 42 Whatman filter paper	A coating bar was applied to coat the paper. Subsequently, the material was subjected to drying at 90 °C for 10 min	No application	The coated paper showed high resistance to oil and antimicrobial activity, making it viable for application in packaging	Jung et al. (2018)
Corn starch	Zinc oxide nanoparticle	Common filter paper	The filter paper was coated using a coating machine at the dosage of 4 g/m^2 followed by 5 min of drying in a quick-drying machine	No application	The coated paper showed high antimicrobial activity against Escherichia coli in addition to high hydrophobicity	Ni et al. (2018)

properties of the coated paper sheet and improved the antimicrobial activity of the packaging. The latter was proved by the total bacterial count (TBC) assay in which a 3-log-cycles reduction in the order of magnitude of microbial population (expressed as CFU/g – Colony Forming Unit per gram) was seen at the fourth day of packed beef stored at 4 °C, compared to uncoated paper. This study also found that the coatings containing 1% and 2% of citric acid was responsible for the inhibition of lipid oxidation by 27% and 71%, respectively.

Divsalar et al. (2018) reported a nanocomposite formulation based on chitosan, zinc oxide and nisin for cheese packing. Nisin is thought to promote pore formation on the cell membrane and inhibit its biosynthesis (Li et al. 2018). The proposed structure acted as a very effective antimicrobial active packaging against *L. monocytogenes* due to the broad-spectrum inhibitory effect on Gram-positive bacteria of nisin (Brötz and Sahl 2000). For the cheese stored at 4 °C for 14 days inoculated with the studied microorganism, the bacterial population inhibition was found to be of 2.7 $\log_{10}$ CFU/g for the coating containing 500 ppm of nisin, and of 5 $\log_{10}$ CFU/g for the coating containing 1000 ppm of the bioactive compound.

Hefft (2018) tested the antimicrobial activity of pure chitosan coatings against *Botrytis cinerea* on bilberries and redcurrants stored in coated paperboard trays. The coating was responsible for shelf-life extensions of 2 and 3–4 days for the berries, respectively, which also contributes to the reduction of food waste. More recently, Silva et al. (2022) also tested an active chitosan-based coating but containing lemongrass essential oil for paperboard packaging. The active coating reduced the air and the water–vapor transmission rates, and water absorptiveness, also proving to be efficient against adult *Sitophilus zeamais* in wheat grain and pasta package. By increasing the number of coating layers from 1 to 5, the anti-insect efficiency was raised by 59%, after 360 h of exposure.

Syahida et al. (2021) reported that Kraft paper coated with gelatin, palm wax and lemongrass essential oil (GPL/K) through the impregnation method delayed lipid oxidation and microbial spoiling activity of ground beef stored at 4 °C for 7 days maintaining the quality and extending the shelf life of the product at the chilled storage. The total mesophilic bacterial growth of the beef wrapped on this coated paper (GPL/K) was significantly slower than the unwrapped beef (control), and on the beef wrapped on uncoated paper and on paper coated only with gelatin and palm wax.

Active component and/or biopolymer migration

In the scientific literature, studies related to the migration of bio-based coatings for paper packaging are still scarce, not completely standardized, and mainly focused only on overall migration (OM). The availability of migration studies of biopolymer-based films and coatings is much greater in the scientific databases. Nevertheless, this form of packaging is beyond the scope of the present literature review. Also, the papers found and discussed below did not always mention if the amount of the species that migrated or even their chemical compositions endanger human health, unacceptably change the composition of the packed food, or deteriorate its organoleptic characteristics. All those attributes, along with many others, should be addressed according to the Comission Regulation (EC) Nº 450/2009 in the European Union. In addition, active ingredients that should not migrate from the packaging, and that do not possess function over the food must pass through a safety assessment carried out by the European Food Safety Authority.

Kopacic et al. (2018) tested the OM of two different paper grades coated with alginate and chitosan, according to EU-Regulation No. 10/2011. The food simulant used was Tenax® (poly(2,6-diphenyl-p-phenylene oxide)) in order to mimic dry foods. The storage condition of 80 °C for two days was set for long-term storage simulation. The researchers found that the OM of the alginate-coated paper was reduced to 16.3%, and for the chitosan-based coating the reduction was to 29.5%, both compared to the uncoated recycled paper. The virgin paper did not present considerable overall migration.

Javed et al. (2021) investigated the lignin migration from lignin-containing starch coatings to three simulants: (a) 3% w/v acetic acid solution (pH = 2.4); (b) deionized water (neutral pH); and (c) alkaline buffer (pH = 10). The migration test was performed putting 4 mg of the dry coating (4.9 ± 0.9 cm^2 of overall area of specimens) in contact with 15 mL of each simulant at room temperature for 1 h. The addition of ammonium zirconium carbonate (AZC) as a crosslinker between starch chains reduced the amount of lignin migration to the simulants. The level of lignin migration was around 10 times lower for the simulant (b) compared to the other two tested (simulants (a) and (c)), in which the level of migration was similar between them). The lower migration of lignin in simulant (b) was attributed to its low solubility at acidic pH solutions.

Tanpichai et al. (2022) conducted overall migration assays in a paper coated with up to five layers of chitosan, according to the Directive 97/48/EC, in four simulants: (a) 3% w/w acetic acid solution (for aqueous foods with pH lower than 4.5); (b) water (for aqueous foods with pH higher than 4.5); (c) 15% v/v ethanol in water (for alcoholic food); and (d) isooctane (for fatty foods). All migration tests were performed in four contact conditions: (i) contact at cold temperatures and for short and long-term storage (5 °C/10 days for simulants (a), (b), and (c), and 5 °C/12 h for simulant (d)); (ii) contact at ambient temperature and for short and long-term storage (40 °C/10 days for simulants (a), (b), and (c), and 20 °C/2 days for simulant (d)); and two conditions of contact at high temperature and for short duration ((iii) (70 °C/2 h for simulants (a), (b), and (c), and 40 °C/30 min for simulant (d)); and (iv) (100 °C/30 min for simulants (a), (b), and (c), and 60 °C/30 min for simulant (d))). These authors found that regarding simulant (a), there was an intensification of OM as the number of chitosan layer tended to be raised, because of the relatively high solubility of chitosan in acidic solutions. The contact condition (ii) was the one in which all specimens presented OM below 10 mg/dm^2, independently of the number of chitosan layers, indicating that the contact with acidic foods was more appropriate at ambient temperature. For simulant (b), the number of coating layers was significant only for condition (ii), since the five-layered coating presented the lowest OM for this contact condition, as well as the only OM below the limit of 10 mg/dm^2. Still for simulant (b), the contact condition (iii) presented the lowest OM, independently of the number of coating layers. Regarding simulant (c), the OM of the coated samples were below the limit of 10 mg/dm^2 in all contact conditions, independent of the number of coating layers, except for the five-layered coated paper at condition (iv). For simulant (d), at ambient temperature (condition (i)), the coated paper presented OMs lower than the limit with up to three layers of chitosan. The same was observed for conditions (iii) and (iv), but independent of the number of coating layers in this case.

Regarding active coatings for paper packaging, Hassan et al. (2022) developed a formulation based on cellulose nanofibers, pectin and pomegranate extract. The overall migration test was performed only on the face of the specimens coated with the formulation, and according to EU-Regulation No. 10/2011. These authors used three simulants representing water, fatty, and acidic conditions, which were 10% v/v ethanol in water, 50% v/v ethanol in water, and 3% acetic acid solution, respectively. The contact time was 10 days at 40 °C. The amount of coating that migrated to the simulants were 2.3 mg/dm^2 for the fatty one, 2.95 mg/dm^2 for the acidic one, and 3.46 mg/dm^2 for the aqueous condition. Therefore, the coating migration was lower the limit of 10 mg/dm^2 as stated in the mentioned regulation, for all three simulants tested.

Winotapun et al. (2022) studied the overall migration of coatings based on blends of lignin nanoparticles and chitosan, in different proportions, into four simulants: (a) 10% v/v ethanol in water; (b) 3% v/v acetic acid solution; (c) 20% v/v ethanol in water; and (d) n-hexane, according to a standard method described by the Ministry of Food and Drug Safety of South Korea. The coated face of the paper was exposed to the simulants for 30 min at 40 °C. For simulant (a), the coatings containing more than 20% wt of lignin presented an overall migration lower than 10 mg/dm^2. For simulant (b), the opposite was observed, mainly because chitosan is soluble in acidic solutions (Sobral et al. 2022) and because a detachment of the coating was observed in the test for high lignin-content formulations due to the poor adhesion between it and the paper substrate.

Regarding simulant (c), the overall migration of the coating formulations was under the limit stated in the reference in which the test was based. At last, for simulant (d) the only coating formulation that presented overall migration under the limit of 10 mg/dm^2 was the one with a lignin nanoparticle content of 33% wt. Based on the overall migration tests performed, the formulation containing 67% wt of chitosan and 33% wt of lignin nanoparticles, which performed the best regarding migration, was the most appropriate for foods with the characteristics of simulants (a), (c), and (d). However, test conditions, such as temperature and exposure time must be better explored in order to promote a better understanding of the possible real scenarios of the packaging use.

A combination of silver nanoparticles and iron (II, III) oxide (Fe_3O_4) was applied along with carboxymethyl cellulose, poly(vinyl alcohol), glycerol, and citric acid as paper coating. The specific migration of silver and iron ions was performed with: (a) 3% w/v acetic acid; (b) deionized water; and (c) 10% w/v ethanol as simulants, for 168 h (temperature not informed). The amount of Ag released was between 0.076 and 1.30 $mg_{Ag}/kg_{food\ simulant}$, which was below the migration limit of 60 $mg_{Ag}/kg_{food\ simulant}$ as stated in the EU Regulation 10/2011. However, toxicity of migrating Ag ions in food packaging materials is still an issue. The cumulative release of iron ions varied from 2.10 to 3.09 $mg_{Fe}/kg_{food\ simulant}$, lower than the most restrict limit (9 $mg_{Fe}/kg_{food\ simulant}$) stated in the Nordic Nutrition Recommendations (Srichiangsa et al. 2022).

Overall, all studies describe above treated with migration of any component of coating, being interpretated as non-intentional migration. Nevertheless, for active packaging system based on material containing the active component, an intentional migration can be expected favoring the conservation of packed foods (Dammak et al. 2017; Procopio et al. 2023). In this sense, Yang et al. (2023) developed a coating formulation based on an organic metallic structure containing zinc, 2,5-dihydroxyterephthalic acid, and carvacrol, the latter for antibacterial activity, to coat filter paper. The release of carvacrol was measured after the contact with aqueous simulants of (a) 10% v/v, and (b) 95% v/v of ethanol. The contact condition was around 170 h at room temperature. A fast release was noted on the first 24 h of contact for both simulants. However, the total release was more expressive for simulant (a) due to its grater polarity compared to simulant (b), which was thought to be responsible for a grater disrupting of the hydrogen bonds between the organometallic structure and the carvacrol. With respect to the overall migration, the tests were performed with contact at 20 °C for 10 days, with the same simulants. The OM did not exceed the 10 gm/dm^2 limit for both ethanol solutions, with the lowest value seen with simulant (b).

Methods of application of coatings to cellulosic packaging

In general, there are two main routes for the application of coatings to cellulosic packaging: dry and wet methods (Fig. 1). The dry processes are based on the thermoplastic properties of the materials. Two common ways to do so are extrusion and thermocompression methods, which include the heating of the material above its glass transition temperature under low moisture conditions (Cuq et al. 1998). In extrusion, the components of the coating (additives, such as plasticizer, pigments, and active compounds) are added to the dry biopolymer and then a layer of the molten mixture is applied to a sheet of paper in continuous motion (Hanlon et al. 1971; Sharma et al. 2020a, b; Tara et al. 2004). However, the process possesses some limitations such as the need for a thermoplastic behavior of the material, the need for a large amount of coating forming mixture to achieve the desired properties, and the melt instability of the material (Rastogi and Samyn; 2015).

The wet methods are based on the drying of a coating-forming solution, which involves solubilization, application onto the cellulosic material, and drying steps (Fig. 1). In general terms, the coating-forming solution is produced by dissolving the biopolymer in a suitable solvent. Soon after, the solution can be heated or its pH adjusted to improve the coating-forming properties, and then plasticizer is added. In addition, nanoparticles and/or bioactive compounds can also be incorporated. Then, the solution can be dispersed or poured onto the substrate by automatic applicators and dried on a flat surface or on a dryer (Debeaufort et al. 1998; Guilbert 1986; Marangoni et al. 2021; Krochta and Mulder-Johnston 1997; Mustapha et al. 2019; Silva et al. 2022). Depending on the chemical composition of the coating, a layer-by-layer (LBL) method can generate better results. In the LBL application, the alternate deposition of components containing opposite charges is performed, followed by a self-assembly of the materials guided by their electrostatic interactions (Abbadessa et al. 2023). The application itself can be executed by several meanings such as by spraying the material onto the paper surface or by using rods, blades, or bars for this purpose.

Rod coating, consisting of a wire-wound bar-rod is one of the most used methods for thin coating application on paper surface by the paper industry. By

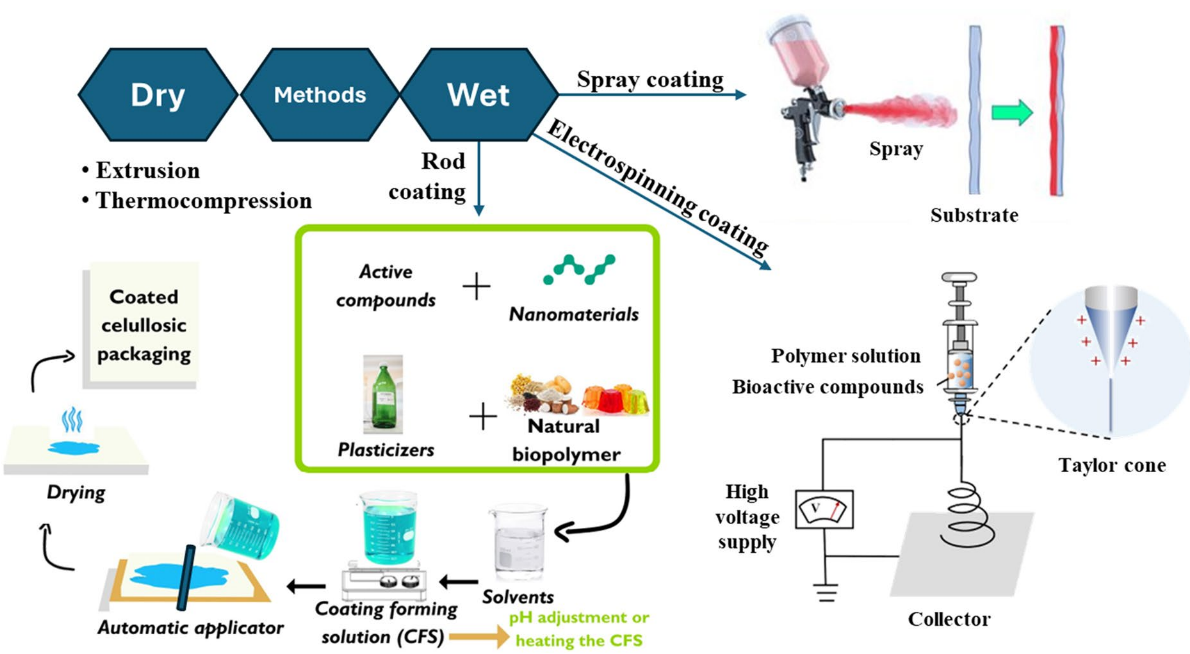

Fig. 1 Schematic representation of the main methods of applying coatings to cellulosic packaging. Adapted from Khan and Liu (2022), Min et al. (2022), and Kunam et al. (2022)

varying the diameter of the wire wrappings, different coating thicknesses can be reached. It has as advantages its simplicity for operation, and low cost. The air-knife coating is another well-stablished method in which an excess of coating is poured on the paper substrate and then a jet of pressured air is applied to spread and remove the excessive amount of coating in a controlled way. For the blade coating, the spread and removal of the excessive amount of coating is done by a blade, which allows the recovery of the non-attached coating, like the air-knife method (Tyagi et al. 2021).

Spray coating is another method of application used to cover cellulosic substrates with biopolymers. It consists of atomizing the liquid in the solution droplet, during spraying, followed by the coalescence of the coating over the substrate (Fig. 1). The spray coating deposition presents as advantages simplicity, rapidity, continuity of the process, ability to coat irregular surfaces, along with the possibility of applying solutions with higher solid contents, which reduces the amount of solvent to be evaporated. However, the controlling of the homogeneity of the coating thickness is considerably challenging (Nadeem et al. 2022; Cherian et al. 2022). Some of the most used techniques to generate the spray are air assisted (best for low-viscosity solutions, complex substrate surface, and small-scale production) or airless systems (best for viscous solutions, simple substrate surface, and large-scale production) (Nadeem et al. 2022); electrostatic spray (the breakdown of the droplets up to the nanoscale is controlled externally by a disturbance of the electrostatic field) (Gui et al. 2023); and hot flame spray (the combustion of a gas fuel transports heat to the coating solution, which also generates the flow towards the substrate to be coated) (Tejero-Martin et al. 2019).

Electrospinning is another method of application of coatings onto cellulosic surfaces. In this technique, micro/nanofibers of the biopolymer are formed when high voltage is applied between the needle tip where the solution flows from and the electrode under the paper substrate. As a result, a charged jet of the polymer solution is created and when the jet travels in the air, the solvent evaporates, leaving behind the charged fibers (Fig. 1). Instrumental factors such as flow rate and applied voltage, along with characteristics of the coating-forming solution (viscosity, concentration of components, electrical conductivity, surface tension etc.) and of the environment (temperature and relative humidity) are crucial for good depositions (Min et al. 2022).

Biodegradability of cellulosic packaging with biopolymer-based coatings containing active compounds

Cellulosic packaging coated with biopolymers containing active compounds are environmentally friendly materials because they can biodegrade into CO_2, H_2O, and biomass easily in less than 6 months after their use through the action of degrading enzymes and microorganisms that occur after disposal under appropriate conditions (Wu et al. 2021). In addition, these materials do not release toxic substances into the environment at the end of their decomposition (Horvat and Krzan 2012). Nevertheless, some concerns due to the presence of active compounds into coating could be expected due to an eventual biocidal effect on biodegradation media. Still, biodegradation was observed with active films incorporated with plant hydroethanolic extracts that are rich in phenolic compounds, which possess antioxidant and antimicrobial activities, by Bonilla and Sobral (2020), and Bonilla et al. (2020, 2021) in studies evaluating compostability or respirometry. No biocidal effect on biodegradation media was observed in these studies.

According to Pathak (2017), the biodegradation process occurs in different stages, which are: biodeterioration, biofragmentation, assimilation, and mineralization (Fig. 2). In biodeterioration, microbial biofilms are formed on the polymer surface. They are responsible for assisting the conversion of polymers into smaller fractions, resulting in altered physical–chemical characteristics of biopolymers (Awasthi et al. 2022; Shaikh et al. 2021). On the biofragmentation step the smaller fractions of the biopolymer are broken down into simpler forms (oligomeric, dimeric, and monomeric) by enzymatic cleavage (Choe et al. 2021; Kjeldsen et al. 2018;). The paper substrate is consumed along with the biobased coating by the degrading microbes as energy source (Villalba et al. 2004). In the next step, the simpler forms of the biopolymers are actively assimilated and consumed by the microorganisms to form cellular biomass and to extract energy from, together

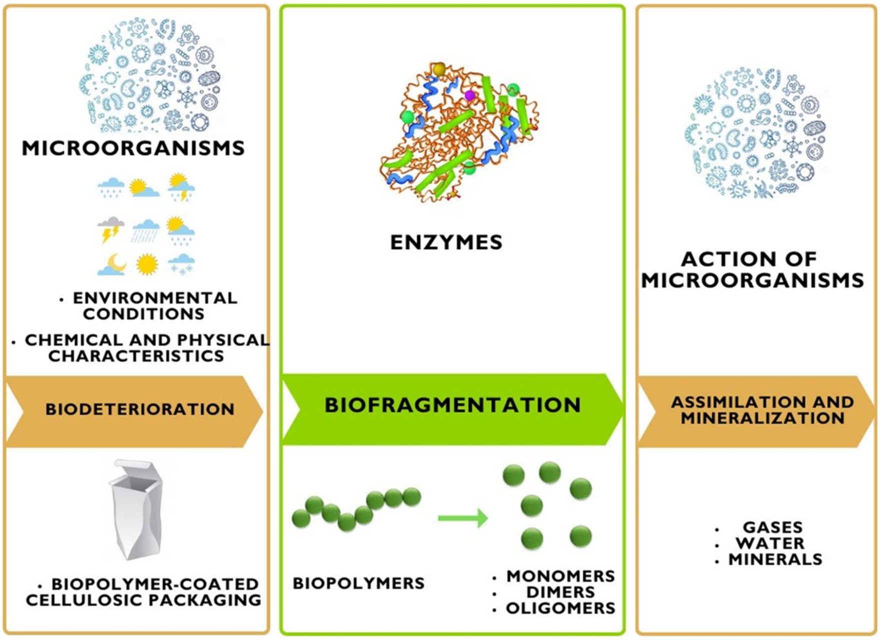

Fig. 2 Schematic representation of biodegradation of cellulosic packaging with biopolymer-based coatings containing active compounds. Adapted from Awasthi et al. (2022)

with other metabolites. Finally, these metabolites (biodegradable materials) are converted into gases (CH_4 and CO_2), water (H_2O), and minerals (inorganic salts) (Pathak 2017).

The biodegradability of coated cellulosic packaging can be affected by several environmental factors such as humidity, temperature, presence or absence of oxygen, and microbiote. In addition, characteristics of the cellulosic packaging (coated paper and paperboard), such as porosity, antibacterial activity, morphology, among other, are determinant for the processes (Fan and Lee 1983; Pommier et al. 2010). More studies on these factors affecting biodegradability of biopolymer coated paper must be encouraged.

Final remarks and perspectives

The success of cellulosic packaging depends on its functional, physical and barrier properties and the coating materials used. Coatings based on biopolymers can be used as replacements for traditional fossil-based coatings, with a serious limitation, ca., high sensitivity to moisture (water as liquid or vapor). Furthermore, the addition of active compounds and/or nanoparticles in the coating-forming solutions can provide enhancements of coating performances towards mechanical, barrier, and antimicrobial properties. Nevertheless, paper coating and the resulting performance of the packaging depend on the application technique used. Thus, further researches on new active compounds, alternative nanoparticles and more industrial-driven application techniques must be privileged. There is a necessity to establish the use of these materials on an industrial scale but they seem to have great potential for many applications in the food market. However, some of the revised papers are based on methodologies and starting materials that could be challenging to scale up.

The next research projects will have to keep that in mind to make the results of their biopolymer-based coatings competitive in the market. A more intensive use of industrial byproducts could be a way to address this issue, which goes along with sustainability principles such as reduction of waste generation, and industrial symbiosis. The migration of components and the security to the human being of the coatings should also be fully studied, along with the real effects of end-of-life disposal of bio-based active materials and of packaged foods.

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Data availability No datasets were generated or analysed during the current study.

Declarations

Ethical approval Not applicable.

Competing interests The authors declare no competing interests.

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