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Electrospinning of sustainable polymers from biomass for active food packaging

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Recent advances in active food packaging have been driven by the integration of electrospun materials, exploiting their inherent advantages. Electrospun materials can be easily functionalized with antioxidant, antibacterial, antifungal, and sensory additives, as well as ethylene scavengers and CO_2 emitters making them ideal for active food packaging. However, it's worth noting that certain electrospun materials utilized in this context are derived from petroleum-based synthetic polymers, which may raise environmental concerns post-usage. In this regard, the use of sustainable polymers for electrospun food packaging materials can address problems like waste generation and the environmental impact of traditional synthetic, petroleum-based polymers. Central to this transition is the utilization of biomass-derived polymers sourced from renewable sources like plants, algae, microorganisms, and wastes. Sustainable polymers, such as poly(lactic acid) (PLA), starch, cellulose and derivatives, polyhydroxyalkanoates (PHA), chitosan, gelatin, and zein have emerged as key sustainable players in active food packaging. This review provides a comprehensive overview of electrospun materials of sustainable polymers derived from biomass for the development of active food packaging films. The review begins with a brief description of the fundamentals and process for active food packaging and electrospinning, followed by a detailed examination of the applications of electrospun materials for active food packaging, categorized by polymer type and bioactivity. Finally, the review concludes with current challenges and provides insights into future perspectives in this area.

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Sustainability spotlight

This review reports the use of sustainable polymers sourced from biomass for the development of active food packaging materials, in line with the Sustainable Development Goals (SDGs). By contributing to SDG-2 (zero hunger) through improving food security, SDG-12 (responsible consumption and production) *via* the efficient utilization of natural resources, SDG-14 (life below water) and SDG-15 (life on land) by mitigating plastic pollution from lands and water resources, the integration of sustainable polymers derived from biomass marks a significant step towards environmentally conscious active food packaging. The use of electrospun food packaging materials made from sustainable polymers such as polysaccharides (*e.g.*, cellulose derivatives and starch-based), protein-based (*e.g.*, zein and gelatin), poly(lactic acid) (PLA), and others reduces reliance on petroleum-based polymers by mitigating the environmental impact of plastics. This sustainable approach not only reduces plastic waste generation but also represents a commitment to renewable resources, laying the foundation for a greener future in food packaging innovation.

1. Introduction

Nowadays, food packaging systems have undergone a remarkable transformation, going beyond traditional barrier films and introducing (bio)active films with functions such as antioxidant, antibacterial, and antifungal properties as well as the ability to monitor changes in packaged foods.[1–3] This transformation is further enhanced by the use of biomass-derived sustainable polymers, not only to meet these functional requirements but also to address environmental aspects, thereby reducing the dependence on petroleum-based polymers.[4–6] This sustainable approach aims to combat food spoilage, protect packaged foods from oxygen, bacteria, and fungi, and reduce the ecological impact of packaging films. Thus, it can not only overcome the challenge of preservation but also contribute to a more environmentally conscious and responsible packaging solution. In this context, the use of the electrospinning technique has seen an upsurge in the production of cutting-edge food packaging materials, exploiting the beneficial properties of the resulting structures for functionalization and structural customization.[7,8]

The electrospinning process enables the integration of a diverse range of molecules or nanostructured materials with tailored functionalities, providing a versatile platform for active food packaging.[9,10] In particular, nanofibers can be equipped with bioactive molecules, transforming them into active food packaging

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materials that can protect packaged foods from radicals, microorganisms, and fungi. Furthermore, these electrospun materials can be engineered as intelligent systems to monitor changes in food samples to ensure and preserve their freshness.[11,12]

Active food packaging goes beyond mere protection and incorporates various active ingredients, including oxygen scavengers,[13] antimicrobials,[14] temperature indicators,[15] ethylene scavengers[16] and CO_2 emitters,[17] improving shelf-life, safety, and overall quality of the packaged foods. This proactive approach not only reduces food waste but also meets consumer preferences for fresher and longer-lasting products. While traditional plastic films (*e.g.*, polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), *etc.*) have dominated food packaging in the past and present,[18–20] electrospun materials are emerging as a viable option for developing active food packaging materials.[8] The increasing environmental concerns surrounding petroleum-based polymers in food packaging have directed research toward sustainable biomass alternatives. Sustainable polymers from biomass are mostly biocompatible and biodegradable, reducing the environmental impact of waste.[21] In this context, cellulose and derivatives, poly(lactic acid) (PLA), polyhydroxyalkanoates (PHAs), proteins (*e.g.*, zein and gelatin), chitosan, and starch have emerged as important options for the development of electrospun food packaging materials, reflecting the commitment to environmentally conscious and effective packaging solutions. These polymers have found extensive applications in the production of a wide range of electrospun food packaging materials through the integration of functional molecules, nanostructures, or groups. In this review, we have summarized advances in electrospun food packaging materials produced from sustainable biomass-derived polymers (Fig. 1). First, we provide a concise overview of active food packaging and electrospinning. We then describe the applications of electrospun materials made from sustainable polymers for various food packaging purposes. Finally, we address the existing challenges and offer insights into future perspectives.

2. Active food packaging: an overview

Every year, the world continues to grapple with the persistent issue of global food waste, wherein a notable amount of food produced is lost due to diverse factors, notably including damage inflicted by microbial and oxidation processes.[22,23] In addressing this challenge and in line with SDG Goal 2 (zero hunger), there has been a notable development in food packaging focused on improving responsiveness to oxidative and microbial degradation of packaged foods. The core of this packaging innovation lies in its ability to strategically improve the interconnected dynamics of chemical, physical, and biological processes occurring within the packaged food and its environment. This complex optimization aims to ensure the quality and freshness of packaged food and ultimately contribute to a reduction in overall food waste. Therefore, apart from the traditional role of food packaging in containment and protection, active food packaging intervenes in the internal environment of the packaged food to ensure the preservation, safety, and quality of the food.[24–28] It uses dynamic mechanisms to protect food from environmental influences and thus preserve the freshness and quality of the packaged food.

Food packaging can also be designed as smart packaging to proactively preserve packaged foods and monitor changes.[29] In this regard, sensory components, such as pH or temperature indicators, are integrated into the packaging to detect any changes to the food during storage.[29–31] Active food packaging can also be employed for the regulation of humidity.[32]

Incorporating additives with free radical scavenging properties into food packaging materials provides them with an antioxidant effect.[33] The polymers can also be modified by introducing radical scavenging groups or molecules to impart antioxidant properties without the need for additional components.[34,35] In this regard, various phytochemicals, especially phenolic compounds, are acknowledged for their radical-scavenging abilities. Essential oils (EOs), quaternary groups,

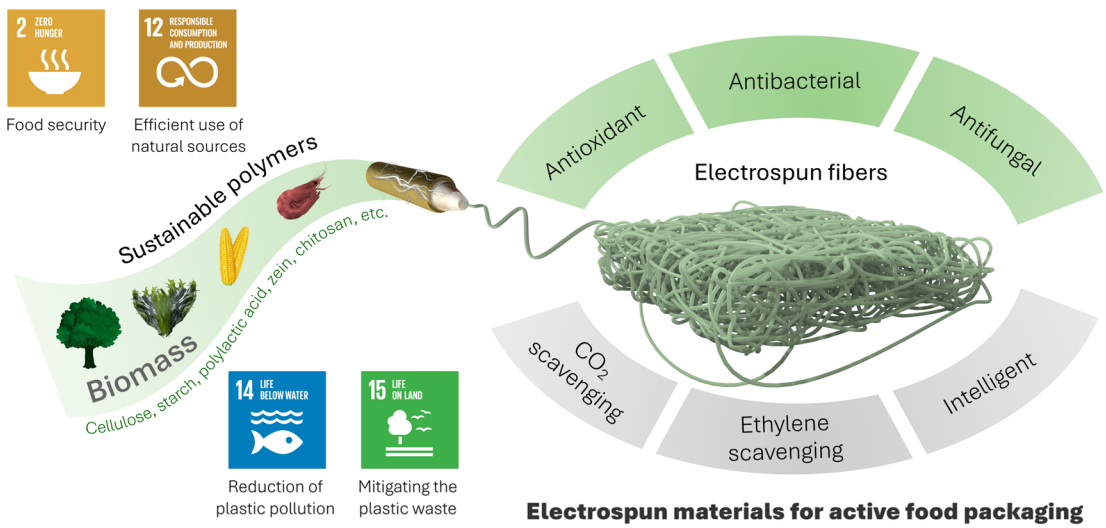

Fig. 1 Schematic representation of the production of electrospun active food packaging materials using sustainable polymers from biomass sources, in line with the Sustainable Development Goals (SDGs).

and metal nanoparticles have been incorporated into electrospun materials for food packaging, conferring antimicrobial properties.[36] Similarly, substances or nanostructures exhibiting antifungal properties have been integrated into food packaging films to enhance their antifungal capabilities.[37] Furthermore, electrospun materials can include ethylene scavengers[38] and CO_2 absorbers[39] to enhance the preservation of packaged foods. Additionally, indicators can be embedded in electrospun food packaging films to monitor variations in temperature, pH, and other factors during food storage.[40]

Key features of active food packaging include:

(1) Improving preservation: the barrier properties of food packaging include the ability of packaging materials to block the passage of gases, moisture, odors, and various external factors that can affect the integrity and safety of the packaged food. A key focus is on preventing the transfer of water vapor, as moisture penetration can lead to spoilage, microbial proliferation, and changes in the texture of food.[32,41]

(2) Oxygen scavenging: the use of oxygen scavengers is crucial to extend the shelf-life and maintain the quality of packaged foods.[13] Oxygen scavengers, either integrated into the packaging material or included as bags in the packaging, interact with oxygen in the packaging environment, reducing its presence. This proactive measure reduces oxygen exposure, preventing oxidative processes that could cause food to spoil, change color and taste, and degrade nutrients.

(3) Antimicrobial activity: antimicrobial components embedded in the packaging material effectively inhibit the proliferation of bacteria, mold, yeast, and other microorganisms responsible for food spoilage and potential health risks to consumers.[42] By creating an inhospitable environment for microbial development, these dynamic packaging solutions help extend the shelf life of perishable foods, reduce reliance on chemical preservatives, and reduce the likelihood of foodborne illness.

(4) Ethylene scavenging: ethylene acts as the promoter for ripening of fruits.[43] However, its presence can accelerate the decay of fruits and vegetables, resulting in a shorter shelf-life. To extend the freshness of foods, ethylene scavengers can be integrated into food packaging materials.[38]

(5) CO_2 emitters: the antimicrobial effects of absorbed CO_2, through carbonic acid formation, acidify the environment, disrupting bacterial cell membranes, cytoplasmic pH, and enzyme activity, thereby inhibiting bacterial growth and enhancing food preservation.[44]

(6) Temperature indicators: temperature-sensitive indicators can be incorporated into packaging materials to show the temperature history of the packaged food.[45]

(7) Intelligent sensing: food packaging materials can be functionalized with sensory elements (*e.g.*, pH sensors) to monitor the condition of the packaged food.[46]

3. Electrospinning: setup and process

Electrospinning represents an electrohydrodynamic technique that employs an electric field to produce micro to nanofibers from a solution containing polymers[47,48] and small molecules.[49] This method relies on the application of an electric field to transform a viscous solution into nanofibers.[50] The fundamental setup comprises (i) a syringe pump, (ii) a high-voltage power source, and (iii) a grounded metal collector.[51] In the electrospinning process, the syringe pump propels the solution, forming a spherical droplet at the tip. Under the influence of a high electric field, this droplet transforms into a cone shape, known as the Taylor cone (Fig. 2). When the electric field surpasses the liquid's surface tension, a charged jet is expelled from the needle, accelerating toward the grounded metal collector.[50,52,53] Throughout the jet's trajectory, solvent evaporation occurs, resulting in a solidified and elongated nanofiber.[54] Fig. 2 illustrates the entire electrospinning setup and process, depicting the formation of the Taylor cone and electrospun nanofibers on the collector. As depicted, the

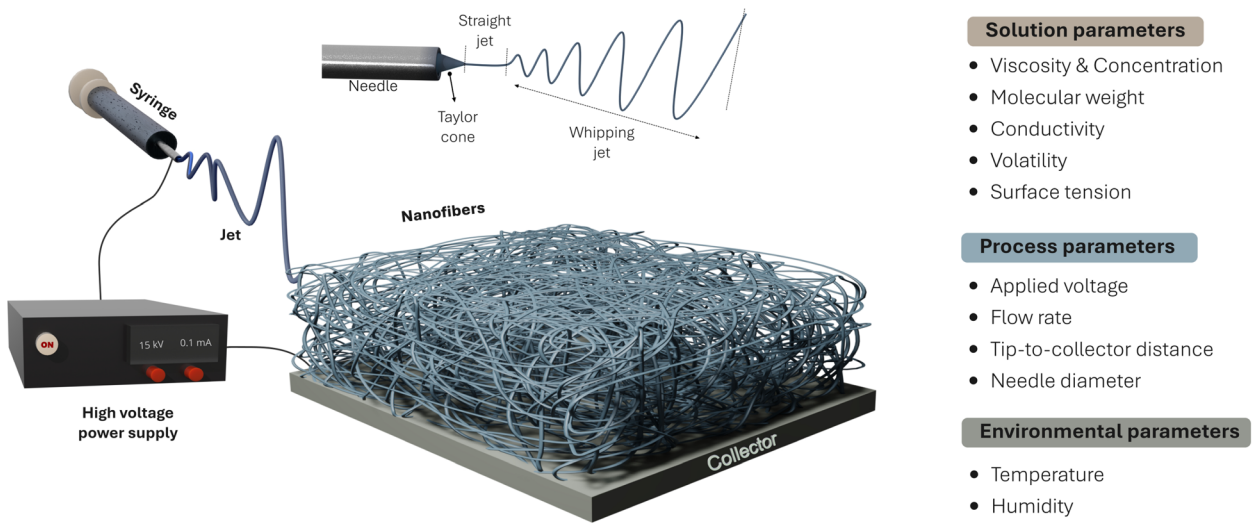

Fig. 2 Electrospinning process from a polymer solution to produce electrospun fiber materials. Important parameters affecting the electrospinning process are highlighted.

electrospinning process is impacted by solution, process, and environmental parameters.

Electrospinning as a versatile process entails the precise deposition of fibers from a polymer solution or melt under the influence of an electric field. The method has garnered considerable interest in recent times due to its promising potential for generating sophisticated and functional materials tailored for active food packaging applications. The electrospinning technique allows for the development of materials using a diverse range of polymers, selected based on their specific barrier properties, mechanical strength, and compatibility with different types of food. These materials can be enriched with active agents, including antioxidants, antimicrobial agents, and oxygen scavengers, enhancing their functionality to extend the shelf life of packaged food by mitigating oxidative processes and preventing microbial growth. The incorporation and release of active agents can be achieved through various methods, such as core–shell structures or emulsion electrospinning, providing sustained release profiles.[55] Additionally, electrospun materials can be applied to traditional food packaging films,[56] imparting them with (bio) activity, including antioxidant, antibacterial, antifungal, and sensory functionalities.

4. Electrospun materials from sustainable polymers derived from biomass for active food packaging

In today's world, there is a noticeable trend toward replacing conventional petroleum-based food packaging materials with sustainable alternatives sourced from biomass.[5,57–59] This transition is motivated by a shared commitment to tackling environmental risks and promoting sustainable initiatives.[59–61] Regarding this matter, electrospun food packaging materials produced from polymers of biomass sources offer several benefits, particularly concerning the environmental footprint of packaging, while also serving as a barrier against light, oxygen, and moisture.[8] Another notable aspect is the easy functionalization of electrospun materials with a variety of antioxidants[62] and antimicrobial agents.[63] This feature contributes significantly to the field of active food packaging as it helps inhibit microbial growth and oxidative reactions that occur in foods. Consequently, the integration of electrospun materials with such properties ensures food safety and helps maintain the high quality of the packaged products. The following section reports the electrospun materials made from sustainable polymers developed for active food packaging. These materials are organized according to both the polymer used and the functionalities of the materials developed.

4.1. Polylactic acid (PLA) based electrospun food packaging materials

Poly(lactic acid) (PLA) stands out as an eco-friendly thermoplastic polymer obtained from renewable resources, offering a sustainable substitute for traditional petroleum-derived plastics due to its biodegradable, biocompatible, and environmentally friendly nature.[64,65] PLA is mostly produced during the fermentation process of sugars obtained from various biomasses, including corn starch, sugarcane, cassava, and other biomass sources (*e.g.*, algae).[66–68] PLA is widely used for the development of a wide range of materials for a broad spectrum of applications, spanning from biomedical to environmental.[69] In this context, electrospun fibers of PLA have been also applied to food packaging.[70] The integration of PLA fibers with functional molecules can endow them with bioactivity to be employed as antioxidant, antibacterial, and antifungal materials, as well as intelligent packaging materials. For example, for the development of pH-sensing intelligent packaging materials, electrospun PLA films were developed by loading anthocyanins from black carrots.[71] The anthocyanin extracted from black carrots loaded PLA fibers exhibited a discernible color transition from reddish pink/dark purple to pinkish gray, suggesting their use as intelligent food packaging materials. Another colorimetric PLA film was developed to monitor cod freshness, utilizing citrated methacrylated urethane (CMU) grafted onto PLA, which was then electrospun into nanofibers.[72] This fibrous film undergoes a color change from white to light orange or pink as cod fish deteriorates, at temperatures of 25 °C and 4 °C, respectively.

PLA fibers were also functionalized with different bioactive molecules and nanostructures, such as plant extracts, to be employed as antioxidant or antimicrobial materials. In this regard, PLA/hydroxypropyl methylcellulose fibers loaded with pomegranate peel extract (PPE)[73] and PLA fibers loaded with ethyl-Nα-dodecanoyl-L-arginate (LAE, ethyl lauroyl arginate)[74] were developed as antimicrobial electrospun films. The latter one tested for the shelf-life of strawberries at 25 °C and the results showed no appearance of infections caused by molds for the tested time compared to unwrapped samples. Likewise, PLA nanofibrous films loaded with a butterfly pea flower extract (BPA) and cinnamaldehyde (CIN) were produced and tested for the packaging of beef pieces (Fig. 3a–d).[75] Nanofibrous electrospun films showed antioxidative and antibacterial properties while they could detect the freshness of packaged food with a fast color responsiveness under acidic–alkaline conditions (Fig. 3b and c), and the film could be used effectively for visual monitoring of beef spoilage (Fig. 3d). In a study, electrospun PLA films were functionalized with Ag_2O-hemp fibers to impart antimicrobial and antifungal properties for fruit preservation against spoilage (Fig. 3e and f).[76] Ag_2O-hemp fibers modified electrospun PLA nanofibrous film showed high mechanical properties in terms of tensile strength and elongation-at-break, and demonstrated effectiveness against *E. coli*, *S. aureus*, *A. niger*, and *Penicillium*, while maintaining cytocompatibility and freshness of red grapes at room temperature (Fig. 3e and f). PLA-based nanofibers loaded with Viola odorata petal anthocyanins, combined with carboxymethyl cellulose/cellulose nanocrystals (CNC) films, were used for monitoring the freshness of various foods like Pacific white shrimps, minced lamb meat, chicken fillets, and rainbow trout fillets (Fig. 3g).[77] These bilayer films, featuring pH-dependent color changes and slow release of encapsulated extract and CNC during refrigerated storage, demonstrate their potential for food freshness monitoring.

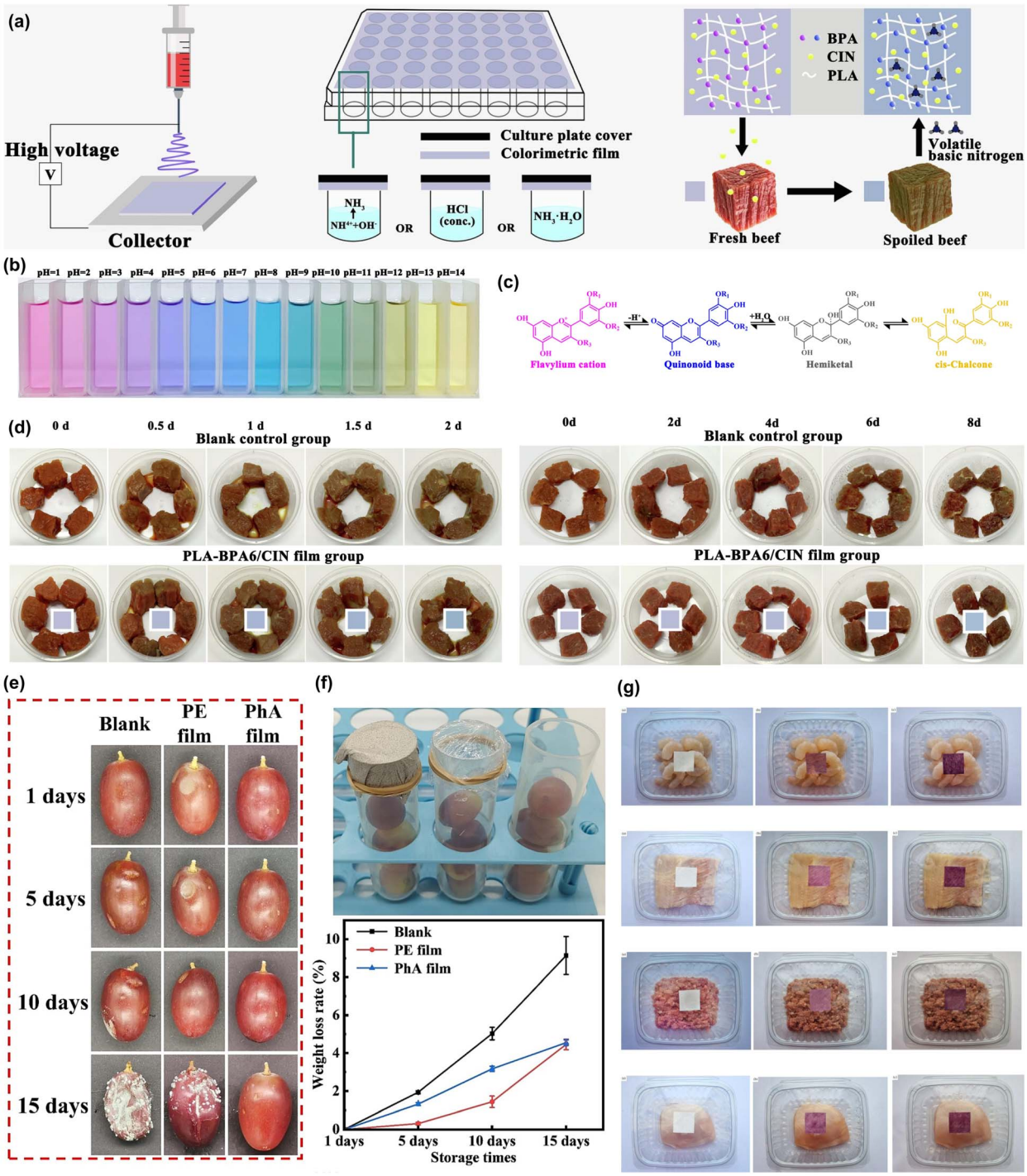

Fig. 3 (a) Evaluation of a hydrophobic PLA-based film for beef meat packaging. (b) Monitoring color variations in BPA solutions relative to pH levels. (c) Structural alterations in BPA corresponding to pH shifts. (d) Assessing long-term performance of films in beef meat wrapping. (e) A longitudinal study of electrospun PLA/AgNPs films on hemp fibers for grape packaging. (f) Experimental setup for grape packaging in centrifuge tubes and monitoring weight loss over time. (g) Freshness assessment of shrimp, rainbow trout fillets, minced lamb meat, and chicken fillets utilizing intelligent double-layer films incorporating carboxymethyl cellulose, CNC, PLA, and viola odorata petal anthocyanins nanofibers. The figures were reproduced from ref. 75–77, Elsevier.

Innovatively, recycled keratin from chicken feather waste was blended with PLA or gelatin and electrospun into antibacterial nanofibers.[78] The resulting nanofibers showed antibacterial activity against *S. aureus* ATCC 6538 and *E. coli* ATCC 25 922 and enhanced thermal properties depending on the keratin loading.

Active ingredients have also been incorporated into PLA nanofibers using excipients such as cyclodextrins (CDs). CD molecules are toroidal cyclic oligosaccharides characterized by a characteristic dual nature: a hydrophobic interior and a hydrophilic exterior.[79] This unique property allows them to

form complexes with molecules that are poorly soluble, thereby rendering them soluble in water.[80] In this context, octyl gallate (OG) as an antibacterial molecule was complexed with β-CD, and the resulting inclusion complex (IC) was incorporated into electrospun PLA nanofibers.[81] The fibers exhibited bactericidal properties, whereby the OG/β-CD IC can act against bacteria by damaging the membrane, penetrating into cells, and subsequently disrupting the activity of the respiratory electron transport chain, leading to the generation of increased intracellular hydroxyl radicals. The PLA-OG/β-CD IC nanofibrous film was tested for the packaging of a Chinese giant salamander fillet at 4 °C, where the freshness of the fillet was significantly extended. PLA/PCL electrospun films with oregano EO/β-CD ICs were also produced for active food packaging applications.[82] The resulting nanofibrous films showed a sustained release profile for the encapsulated oregano EO and showed good biocompatibility and enhanced antibacterial activity, where these electrospun fibers could delay postharvest decay, deterioration, and nutrition loss of blackberries. The Uyar research group also reported CD-ICs of bioactive agents for their higher solubility and enhanced stability. In this regard, α-tocopherol (α-TC)/CD ICs were loaded into PLA nanofibers for potential active food packaging applications.[83] The resulting nanofibers showed antioxidant activity that is high enough to inhibit lipid oxidation. The nanofibers underwent direct testing on beef using the thiobarbituric acid reactive substance (TBARS) method, revealing a lower TBARS content compared to the unpackaged meat. Active packaging notably improved the oxidative stability of the meat samples when stored at 4 °C. In another study, a trilayer food packaging material was developed using an extrusion of PLA and electrospun PLA layer containing ethyl lauroyl arginate (ELA), CNC, and chitosan coating.[84] The trilayer material demonstrated strong bactericidal activity against *L. innocua* and *S. enterica* and degraded over 3 weeks. PLA-based multilayer films were also developed by incorporating ferulic or cinnamic acids using electrospinning.[85] Trilayered films of PLA/starch/PLA with surface-loaded ferulic or cinnamic acid were produced. To promote the compound release of active agents, the active agents were deposited on multilayered films through electrospinning using PLA as a carrier polymer. The multilayer film coated with electrospun fibers demonstrated effective antimicrobial activity against the inhibition of *E. coli* and *L. innocua.* The films functionalized with cinnamic acid showed greater antimicrobial activity. Antimicrobial bilayer films based on PLA and Pickering emulsion were produced to enhance the oxygen barrier property of PLA films.[86] The addition of thymol in the Pickering emulsion layer endowed them with antimicrobial and antioxidant activity.

In contrast to the aforementioned instances utilizing monoaxial electrospinning, PLA-based electrospun fibrous films were also produced through co-axial electrospinning. In this way, the release of active agents could be controlled using a core–shell fiber structure, where the active agent was embedded in the core while the shell layer was mostly hydrophobic to slow down the release of active agents from the fiber matrix. In this regard, cinnamaldehyde (CMA) and tea polyphenols (TP) were encapsulated as core material, while ZnO NPs/PLA served as the shell layer.[87] The synergistic antibacterial efficacy of CMA/TP and ZnO sol induced significant deformation and folding of the cell membrane in *Shewanella putrefaciens* (*S. putrefaciens*). Consequently, there was higher permeability of the cell membrane, leading to the release of intracellular contents and disruption of bacteriophage protein expression. A core–shell nanofiber film utilizing PLA and curcumin (CUR) as the active ingredient was created for active food packaging, employing octadecane solution as the core for phase change thermoregulation and PLA/CUR as the shell, demonstrating both antibacterial properties against *E. coli* and *S. aureus* and antioxidant activity against DPPH radicals, effectively preserving the freshness of bananas compared to conventional plastic bags.[88] In another study, an antioxidant coaxial bionanocomposite was developed using ELA and CNC as active agents.[89] The core solution was composed of PLA and ELA while the shell layer was of PLA/CNC. The core–shell structure could slow down the release of ELA from the fibers. The nanofibers showed antibacterial activity against both Gram-positive and Gram-negative bacteria.

Instead of core–shell structured fibers, bioactive agent release was regulated through moisture-triggered mechanisms, exemplified by carvacrol release from electrospun PLA nanofibers in fresh food packaging.[90] Blending PEG with PLA before electrospinning allowed for varying encapsulation efficiency and loading capacity of carvacrol, leading to sustained release after an initial burst. Application on strawberries illustrated the efficacy of PLA/PEG/carvacrol in maintaining freshness and reducing microbial counts, highlighting the potential of moisture-triggered carvacrol release from PLA nanofibers for active food packaging. A food packaging system utilizing pectin-coated PLA nanofiber films was created to regulate the release of thymol, employing pectin to safeguard thymol from premature release and pectinase to facilitate controlled release.[91] Initially, PLA nanofibers were modified with polyethyleneimine and then coated with pectin to achieve controlled release of thymol, exhibiting potent bactericidal properties against food-related microorganisms, albeit with slightly slower release kinetics compared to thymol-loaded PLA/PEI fibers.

PLA nanofibers were also functionalized with metal–organic frameworks (MOFs) to benefit their strong ability to release highly volatile compounds. In this regard, electrospun PLA/PCL fibers loaded with thymol/MIF-68 (AI) were developed as active nanofibrous film packaging materials.[92] The incorporation of thymol/MIF-68 (AI) improved the water vapor barrier performance and UV resistance of the nanofibers, while it caused the weakening of the mechanical properties of the fibers (*i.e.*, tensile and elongation). The MIF-68 (AI) particles improved the residence time of thymol due to the sustained release of thymol. Another PLA-based nanocomposite electrospun films were developed using an MCM-1 mesoporous molecular sieve loaded with phloridzin, and the resulting films were used for the preservation of strawberries.[93] In this regard, a blend of PLA, phloridzin, and MCM-41 powder was prepared and electrospun into fibers. The electrospun films showed antibacterial activity against *E. coli* growth. Phloridzin was used as an antioxidant

agent, and the phloridzin-loaded electrospun film was used to wrap strawberries, which could delay lipid oxidation in strawberry packaging, promoting their freshness over 3 weeks. Another nanocomposite PLA electrospun films were produced by incorporating TiO_2 nanoparticles and graphene oxide (GO) through an ultrasonic-assisted electrostatic spinning technique.[94] The incorporation of TiO_2 NPs and GO could significantly boost the tensile strength and stretchability of the nanofibrous films while improving their water barrier properties. The UV-exposed fibrous films showed higher inhibition of both *E. coli* and *S. aureus*. The food packaging tests using green peppers through hardness, soluble solids, and chlorophyll content tests revealed that the nanocomposite films could delay the rate of spoilage of green peppers, extending their preservation period.

4.2. Polyhydroxyalkanoates (PHA) based electrospun food packaging materials

Polyhydroxyalkanoates (PHA) are biodegradable polymers produced from microorganisms as intracellular carbon and energy storage compounds.[95–97] PHA could also be produced from plant sugars, vegetable oils, and agricultural byproducts through the fermentation of microorganisms using renewable carbon sources. Their physical and mechanical properties could be tailored by tuning the microbial strains and cultivation conditions. PHA has therefore been employed in various food industries including the development of food packaging materials. Most applications of PHA such as poly(3-hydroxybutyrate) (PHB) for food packaging mainly focus on their use in boosting water resistance by the deposition of PHA fibers on nanocellulose-based films.[98] Likewise, electrospun PHB or electrospun PHB/bacterial cellulose nanowhiskers (BCNW) fibers were also used for improving the barrier properties of thermoplastic corn starch-based films.[99] PHB-BCNW solutions in 2,2,2-trifluoroethanol were prepared and then electrospun onto nanobiocomposites TPCS/BCNW films. The methodology showed good adhesion between the layers and led to higher barrier performance. Such materials with improved barrier properties can be used as food packaging materials. Unlike the above examples, electrospun PHA films were also produced for active food packaging. In one example, a multilayer approach-based electrospun PHA film with/without cellulose nanocrystal (CNC) coatings was reported for active food packaging.[100] The design involved creating an antimicrobial hot-tack layer using poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (PHBV) derived from cheese whey, which was electrospun onto a blown film made of a commercial food contact PHA-based resin. Oregano EO (OEO) and ZnO-NPs were incorporated into the nanofibers of PHVP. The assembly of the multilayer structure took place in a pilot roll-to-roll laminating system, with the blown PHA-based film serving as the outer layers. The electrospun antimicrobial hot-tack PHBV layer and the barrier CNC coating were positioned as interlayers. The resulting multilayers exhibited contact transparency, strong interlayer adhesion, enhanced resistance to water and limonene vapors, and intermediate mechanical properties. Furthermore, these films demonstrated significant antimicrobial and antioxidant activities in both open and closed systems, lasting for up to 15 days. A multilayered electrospun coating with oxygen-scavenging properties was created on conventional cellulose paper through a bilayer electrospinning process.[101] In pursuit of this objective, electrospun fibers of PHB and polycaprolactone (PCL) with palladium nanoparticles (PdNPs) were produced. To diminish porosity and enhance barrier properties and interlayer adhesion, the resulting films were exposed to an annealing process. Following annealing, a significant reduction in the oxygen scavenging capacity of the films was observed compared to the non-heated, highly porous electrospun fibers. Another multilayer concept was developed through the electrospinning of PHB, PCL, or PLA onto both sides of a corn starch film.[102] The water vapor permeability of the membranes experienced a notable decrease upon electrospun coating, irrespective of the polymer used. Similarly, increasing the electrospun coating led to a reduction in the oxygen permeability of the membrane. Another multilayer concept was developed using PHBV electrospun films with antimicrobial properties.[103] Eugenol as a bioactive ingredient was loaded into PHBV fibers, which resulted in the development of antimicrobial fibers. The eugenol-loaded PHBV fibers were used as an interlayer between a structural layer produced through a cast-extruded PHB sheet and a commercial PHBV film as the food contact layer. The resulting multilayer showed hydrophobicity, strong adhesion, mechanical resistance, and higher barrier properties against water vapor and limonene vapors. The antimicrobial efficacy of the multilayer structure was assessed over 15 days in both open and closed environments, revealing notable decreases in the populations of the two food-borne bacterial strains. Another antimicrobial PHB fibrous film was developed through the use of poly(5,5-dimethyl-3-(3′-triethoxysilylpropyl)hydantoin) (PSPH).[104] Following the chlorine bleach step, a bactericidal fibrous membrane could be produced, which showed antimicrobial activity against *S. aureus* and *E. coli* O157:H7. A similar concept was used for the preparation of antimicrobial polyhydroxybutyrate/poly(butyleneadipate-*co*-terephthalate) (PHB/PBAT) nanofibrous membranes.[105] The fibrous membranes were grafted with 1-allyl hydantoin and perfluorooctyl acrylate and then chlorinated with chlorine bleach. The chlorinated membranes showed antibacterial activity against *E. coli* O157:H7 (ATCC 43895) and *S. aureus* (ATCC 6538) with 6.08 and 5.78 log reduction. In another study, PHB of microalgal origin was blended with phenolic compounds and then electrospun into fibers.[106] Phenolic compounds of Spirulina were used for antibacterial activity, and the resulting fibers were tested against the growth of *S. aureus* ATCC 25923. The nanofibers showed good thermal, mechanical, and antibacterial properties, demonstrating their potential for food packaging applications.

Nanocomposite PHB-based fibrous membranes were also developed for food packaging applications. In this regard, PHB/PCL fibrous membranes modified by silica composite hydrol for superhydrophobicity were produced.[107] First, the fibrous membrane was produced through the electrospinning of the PHB and PCL blend, and then the membrane was modified with

silica hydrosol through a dip-pad process. The nanocomposite electrospun films were chlorinated, and the resulting material showed superhydrophobicity with a water contact angle (WCA) of ~150°. The membranes also showed effective antibacterial activity against *E. coli* O157:H7 and *S. aureus* with 99.95% and 99.91% bacterial reduction within 60 min of contact time.

4.3. Starch-based electrospun food packaging materials

Starch, a natural polymer comprised of glucose units, is derived from renewable sources like corn and potato.[108] It serves as a sustainable polymer for creating active food packaging materials, incorporating bioactive compounds and sensing elements. In this regard, various antibacterial/antioxidant/antifungal food packaging materials based on starch have been developed. In one example, a phytochemical with antibacterial activity, cinnamaldehyde EO (CEO), was incorporated into electrospun octenyl succinylated starch-pullulan nanofiber mats.[109] CEO-loaded electrospun nanofibers showed antibacterial activity against *S. aureus* and *E. coli*, and antifungal activity against a saprotrophic and pathogenic fungus, *Aspergillus flavus*. Such materials with incorporated antibacterial agents hold promise in food packaging applications. Likewise, thyme EO (TEO) loaded antioxidant starch electrospun fibers were developed for active food packaging applications.[110] TEO was dissolved in formic acid and blended with starch to produce fibers, where high encapsulation efficiency (>99%) could be reached. The nanofibers showed significant antioxidant activity, which increased with higher TEO loading. Ginger EO (GEO) from avocado seeds was also encapsulated into starch nanofibers.[111] The incorporation of the GEO increased the fiber diameter, and increasing the GEO content led to a drastic rise in antibacterial activity against *E. coli*. Another antimicrobial starch electrospun film was created by incorporating tea polyphenols for active food packaging, with poly(vinyl alcohol) (PVA) added to enhance electrospinnability.[112] This nanofibrous film exhibited improved mechanical strength and water vapor barrier properties while gradually releasing encapsulated tea polyphenols through Fickian diffusion. Demonstrating stronger antimicrobial effects against *S. aureus* compared to *E. coli*, these fibrous films effectively prolonged the shelf-life of strawberries by maintaining freshness for over six days, likely through mechanisms involving cell membrane disruption, DNA fragment degradation, and the generation of intracellular reactive oxygen species (ROS). Polyfunctional starch nanofiber films loaded with tea polyphenols were developed for food packaging, aiming to improve mechanical strength, antioxidant capacity, and hydrophobicity.[113] Fabricated through one-step temperature-assisted electrospinning and cross-linked with glutaraldehyde vapor, the resulting films exhibited altered fiber morphology and a correlation between tea polyphenol concentration and antioxidant properties, albeit with reduced mechanical strength at high polyphenol concentrations.

Antibacterial starch/PVA nanofibers for food packaging have been developed using silver sodium zirconium phosphate (Ag-ZrP) (Fig. 4).[114] Bead-free electrospun fibers with enhanced mechanical strength were produced. The fibers were cross-linked with glutaraldehyde vapor, which also boosted the hydrophobicity of the fibers. The fibers showed antibacterial activity against *E. coli* and *S. aureus*. The antibacterial fibers could prolong the freshness of strawberries over days while a clear sign of rotting was observed for the uncovered strawberries and strawberries covered with Ag-ZrP-free fibers (Fig. 4d). Another silver NP-based starch electrospun fibers were produced for potential food packaging applications.[115] Nonuniform spherical AgNPs were synthesized through the use of crude pulp extract of *Limonia acidissima* and then blended with tapioca starch and guar gum for electrospinning. AgNP loaded electrospun fibers showed antibacterial activity against *E. coli* and *S. aureus*, as well as cytocompatibility over NIH3T3 fibroblast cells, which showed bipolar rod-like shapes in the cells due to a contractile structure. Similar to antibacterial starch-based electrospun films, numerous starch-based electrospun fibers with antioxidant properties have been reported. In one study, β-carotene functional corn starch multilayered electrospun structures were produced for food packaging applications.[116] β-carotene as an antioxidant derived from plants was incorporated into the exterior PCL layers, while the core layer is composed of cornstarch film. Even though the incorporation of β-carotene did not improve the barrier properties of the films, it gave antioxidant characteristics.

In addition to their use as antioxidant and antimicrobial films, starch-based electrospun intelligent food packaging films were also reported. Anthocyanin as a pH sensor was incorporated into the electrospun starch mats for real-time monitoring of food freshness.[117] Bead-free fiber morphology could be obtained at different anthocyanin loadings. The fibers were used to monitor the freshness of pork and shrimp during storage and showed a color change due to loss of freshness. Enzyme and humidity-responsive antimicrobial fibers based on starch were developed for active food packaging.[118] These fibers, comprising CNCs, starch, and zein, were blended with a combination of nature-derived antimicrobials and CD ICs. They demonstrated the ability to release active ingredients either freely in response to microbial enzymes or in the form of CD-ICs when exposed to high humidity levels. This responsive behavior led to significant reductions in bacterial and fungal populations, with greater efficacy observed at higher relative humidity levels. Starch-based nanofibrous films with self-cleaning action were also reported by tuning the hydrophobicity of the films through the surface decoration with hierarchical flower-like micro/nanostructures.[119] To address the extremely low hydrophobicity of starch nanofibrous film, the authors employed a simple and cost-effective solution immersion method to develop a fiber coating using stearic acid (STA). This approach draws inspiration from the superhydrophobic properties observed in biological organisms like lotus leaves. Such films through bioinspired self-assembled coating with self-cleaning ability can be used for the development of self-cleaning packaging materials.

4.4. Chitosan-based electrospun food packaging materials

Chitosan, a biopolymer derived from chitin in crustacean shells, exhibits potent antimicrobial properties, making it ideal

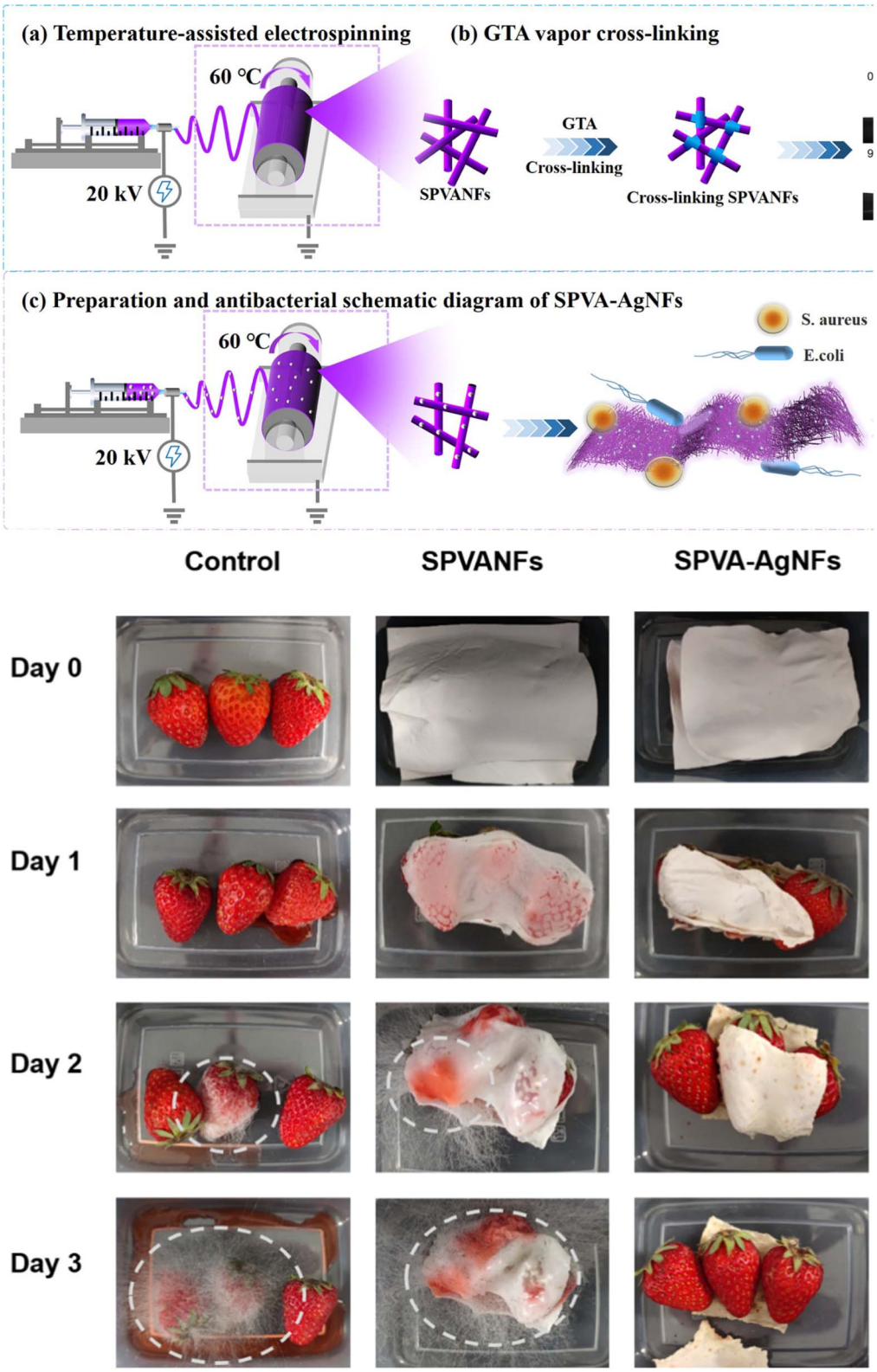

Fig. 4 (a) Schematic diagram illustrating the production of SPVA NFs using temperature-assisted electrospinning; (b) cross-linking of SPVA NFs with GTA vapors depicted; (c) schematic representation of the temperature-assisted electrostatic spinning process for creating SPVA-AgNFs and demonstrating its antibacterial properties. (d) Images of strawberries covered with various protective films were observed under room light and maintained at room temperature for 3 days. The figure was reproduced from ref. 114, Elsevier.

for enhancing the shelf life of perishable foods.[120,121] By incorporating chitosan into electrospun nanofibers, researchers have developed packaging materials with a superior barrier against microbial growth, effectively extending the freshness of packaged foods and reducing food waste. Moreover, utilizing chitosan sourced from biomass waste promotes sustainability by repurposing residues from the seafood industry and reducing dependence on traditional petroleum-based plastics. In this context, various chitosan-based electrospun fibers were reported for active food packaging applications.[122] In one study, chitosan/PCL electrospun fibers incorporated with Chinese yam polysaccharide were developed for active food packaging, demonstrating superior antimicrobial efficacy against *E. coli* compared to *S. aureus*.[123] Antibacterial testing illustrated that these electrospun membranes effectively prevented water-loss rot in cherry tomatoes, maintaining a weight loss rate of 17.6 ± 0.14%. Thymol, an active ingredient, was encapsulated within chitosan fibers using coaxial electrospinning with PEO/chitosan, facilitated by genipin cross-linking (Fig. 5a).[124] These core–shell nanofibers displayed sustained release of thymol, antioxidant properties against DPPH and ABTS radicals, biocompatibility, and antibacterial activity against *E. coli* and *S. aureus*, making them promising for active food packaging applications. Cross-linked chitosan fibers containing bioactive extracts have been explored for active food packaging, as illustrated in Fig. 5b.[125] A blend of chitosan and gelatin mixed with

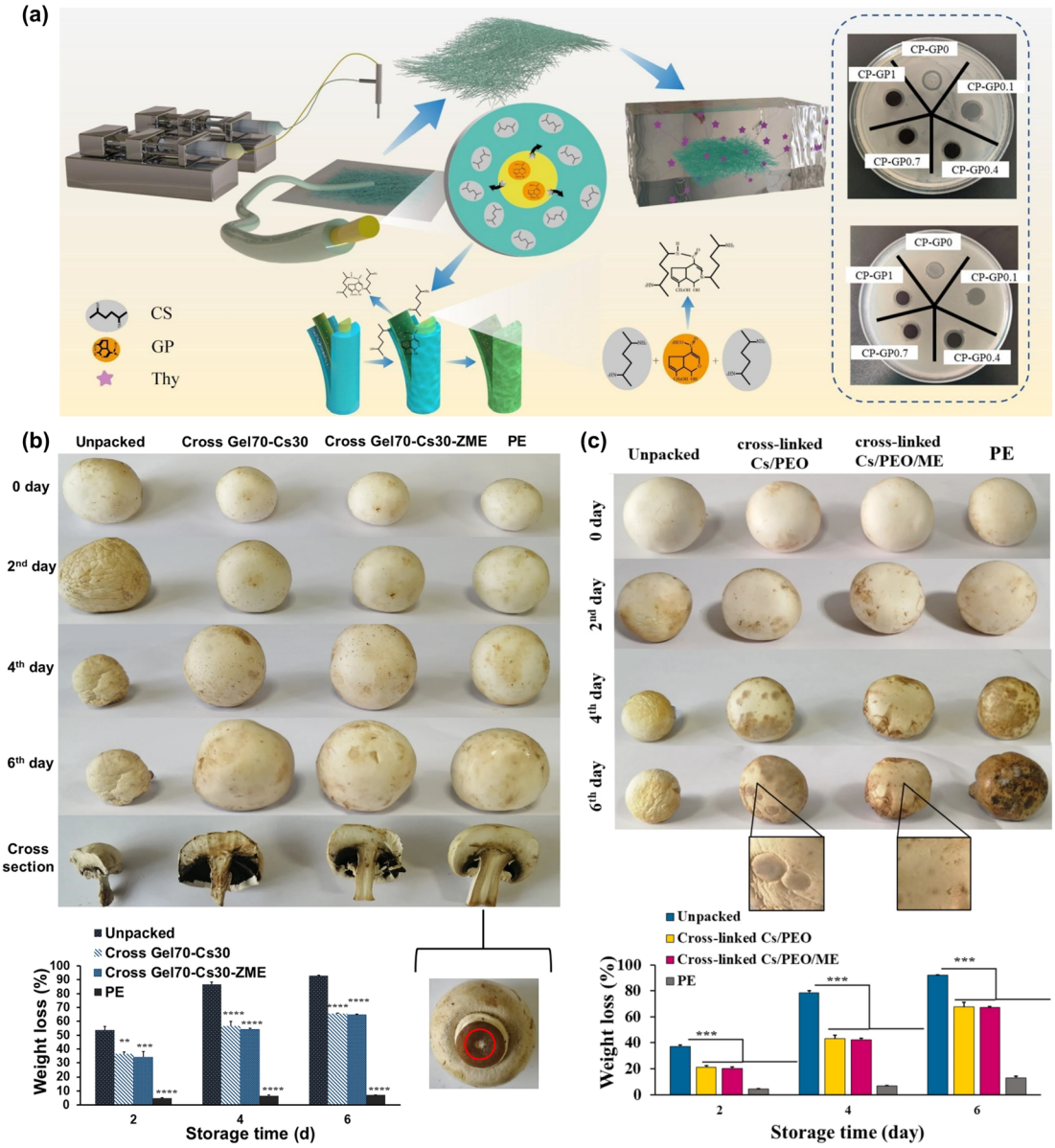

Fig. 5 (a) Schematic depiction of the production of thymol-loaded core–shell chitosan fibers and their use as antibacterial packaging materials. (b) The use of the chitosan/gelatin fibers loaded with *Zataria multiflora* extracts (ZEM) for mushroom packaging over time and the respective changes in weight loss. (c) Appearance changes of mushrooms wrapped by chitosan fibers loaded with MEO during storage and weight loss of mushrooms packed with different samples during storage. The figures were reproduced from ref. 124–126, Elsevier and Springer.

Zataria multiflora extract was electrospun into fibers for mushroom packaging, then cross-linked with citric acid and heat treatment to produce bead-free fibers with an average diameter of 188 nm. These nanofibers exhibited reduced weight loss and water vapor permeability post-cross-linking, along with antibacterial properties, effectively prolonging mushroom preservation compared to conventional polyethylene films, as depicted in Fig. 5b. In another investigation, chitosan fibers were modified with melissa officinalis extract (MOE) for active food packaging, as depicted in Fig. 5c.[126] The resulting nanofibers, formed from a blend of chitosan, PEO, and MOE, exhibited improved mechanical strength through photocrosslinking with benzophenone, while also demonstrating antioxidant and antifungal properties against *C. albicans*, effectively preserving the freshness of wrapped mushrooms for up to 6 days.

In another study, chitosan fibers were used to wrap dry-aged beef, yielding superior results compared to wet-aging over a 3 weeks period, with reduced microbial counts, including yeasts and molds, and enhanced visual appeal.[127] While wet-aged beef showed minimal weight and trimming losses, it also exhibited a significant proliferation of lactic acid bacteria. Likewise, Arkoun *et al.* studied chitosan-based nanofibers' antibacterial action against meat spoilage and pathogenic bacteria.[128] They prepared chitosan fibers *via* electrospinning of chitosan/PEO blends and evaluated their efficacy against *E. coli*, *Salmonella enterica serovar Typhimurium*, *S. aureus*, and *L. innocua*, finding that the antibacterial action depended on amino group protonation and was bactericidal rather than bacteriostatic. The susceptibility of bacteria varied by strain rather than Gram classification, with non-virulent strains showing higher susceptibility, achieving a reduction rate of 99.9%. Ultimately, the nanofibers extended the shelf life of fresh meat by one week.

To boost the antimicrobial activity of chitosan, quaternized chitosan-based fibers were also used for active food packaging applications.[129] In this regard, the mixture of chitosan, quaternized chitosan, vanillin, and PEO was electrospun into fibers. Vanillin could be encapsulated through imine bonds. The use of quaternized chitosan endowed the fibers with high antioxidant, antibacterial, and antifungal activity against relevant strains, such as *E. coli*, *S. aureus*, and *C. albicans*. The fibers could be used for the preservation of raspberries as model fruits, which could extend their shelf-life to 7 days under atmospheric conditions. In another study, a mixture of quaternized chitosan, organic rectorite (OREC), and PVA was electrospun into fibers, which led to antibacterial films for potential applications in active food packaging.[130] The antibacterial activity of the electrospun mats was boosted with increasing amounts of OREC.

Nanocomposite chitosan-based fibers have been developed for active food packaging, such as Fe_3O_4-chitosan/PVA nanofibrous films, exhibiting bead-free morphology and enhanced antibacterial and mechanical properties.[131] Incorporating Fe_3O_4 nanoparticles at varying concentrations improved bacterial adhesion and inactivation efficacy, with the highest effectiveness observed against *E. coli* (90%) and *S. aureus* (66.30%). These films also showed substantial enhancements in tensile strength (up by 46–192%) and elongation at break (increased by 92–141%), along with high biocompatibility, suggesting their potential as active food packaging materials.

CD molecules were utilized for loading bioactive agents into chitosan fibers through inclusion complexation, then blended with chitosan for electrospinning. In one study, 1,8-cineole/CD ICs were incorporated into chitosan/PVA solutions, deteriorating fiber morphology but enabling sustained release of 1,8-cineole, leading to excellent antioxidant and antibacterial activity and extending strawberry shelf-life to 6 days at 25 °C.[132] To address chitosan fibers' hydrophilicity, hydrophobic chitosan derivatives were synthesized to develop water-resistant fibers and improve electrospinnability. In another study, pullulan-carboxymethyl chitosan (CMCS)/PEO core–shell nanofibers loaded with nanogels, specifically CMCS-nisin nanogels (CNNGs), were fabricated and confirmed by TEM analysis.[133] The bead-free nanofibers exhibited smooth textures and demonstrated high antimicrobial activity against *E. coli* and *S. aureus*, effectively extending the shelf-life of bass fish when packed with nisin-loaded core–shell nanofibers from 9 days to 15 days, indicating enhanced stability and bioactivity of the CNNGs. Another intelligent chitosan-based electrospun film was developed through loading shikonin and the use of quaternized chitosan (Fig. 6).[134] Bead-free fibers were produced at different loadings of shikonin, which showed hydrophobicity, barrier, and desired mechanical properties. The nanofibers showed high antibacterial and antioxidant activity, and pH-responsive color change in a reversible manner. The electrospun films were used the monitor shrimp freshness by changing color (Fig. 6e).

An antimicrobial food packaging based on prodigiosin loaded double layered bacterial cellulose and chitosan composites were produced and their properties were explored for food packaging applications.[135] In this regard, two different routes were followed: (i) PVA/chitosan nanofibers were deposited on bacterial cellulose film or (ii) bacterial cellulose/bacterial pigment prodigiosin (PG) blend film was used for the deposition of electrospun film of PVA/chitosan to develop double layered composites. SEM analysis revealed the formation of ribbon-like fibers while the cross-section images revealed the sticking of electrospun fibers on the BC film. WCA analysis revealed the hydrophilic natures of the composites while water vapor transmission rate (WVTR) values BC/PVA-CH-PG and BC-PG/PVA-CH were measured as 1113.71 ± 335.88 g m^{-2} per day and 888.98 ± 125.12 g m^{-2} per day, respectively. The composites demonstrated antimicrobial activity against *S. aureus* and *P. aeruginosa*, suggesting their potential as active food packaging materials.

CUR was also loaded into chitosan fibers to benefit its pH responsiveness. In this regard, G. Sumnu and colleagues reported the natural halochromic CUR-loaded chitosan/PEO nanofibers as an intelligent packaging material.[136] Chitosan/PEO blend was mixed with CUR and electrospun into fibers. The physicochemical properties of the film were notably influenced by the ratio of chitosan to PEO. The nanofilm exhibited substantial color variations correlated with chicken spoilage, as evidenced by changes in pH and total volatile basic nitrogen

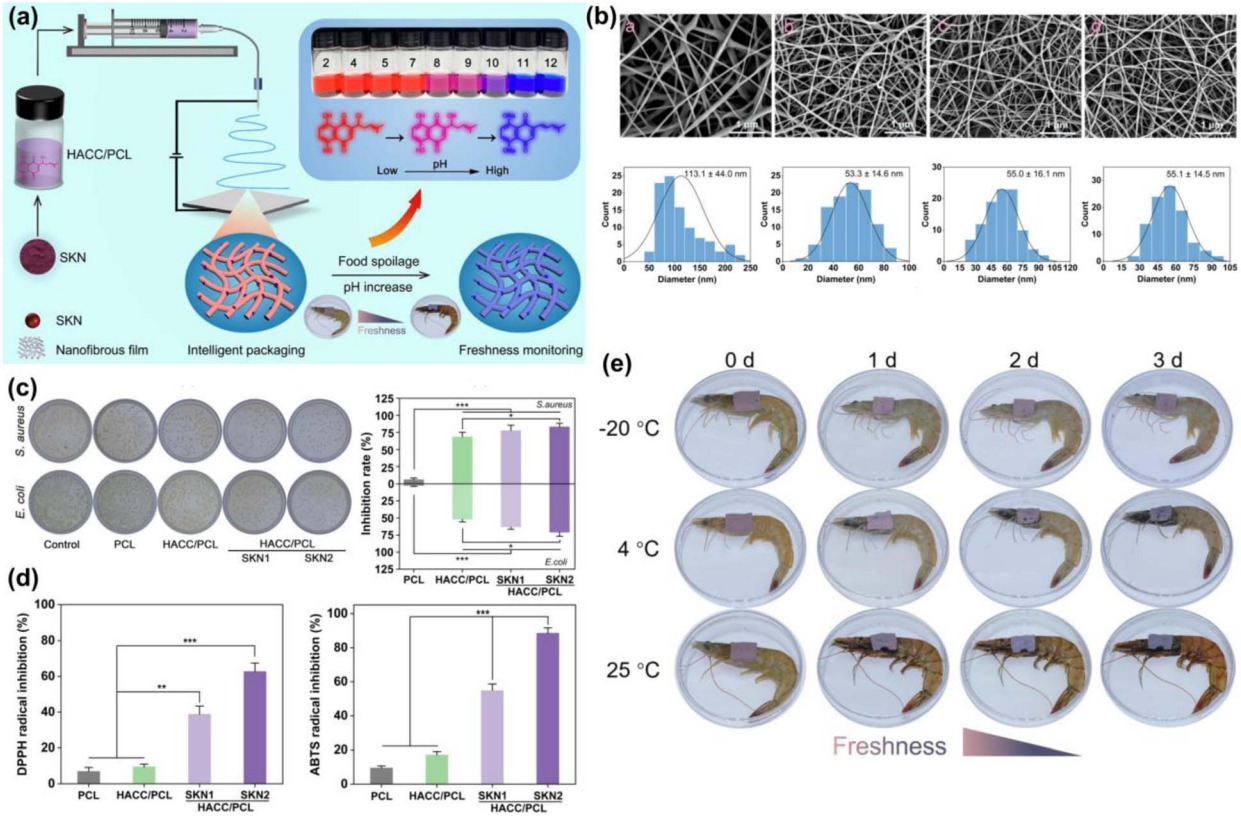

Fig. 6 (a) The development of dual-functional shikonin-loaded quaternized chitosan/PCL electrospun film with pH sensitivity. (b) SEM photos of the respective nanofibers. (c) Antimicrobial activity of the nanofibers against *E. coli* and *S. aureus*. (d) The antioxidant activity of the nanofibers against DPPH and ABTS radicals. (e) The food packaging applications of the films to monitor shrimp's freshness. The figures were reproduced from ref. 134, Elsevier.

(TVB-N) levels. Chitosan-based hybrid electrospun nanofiber mats loaded with eugenol were produced and tested for antimicrobial activity.[137] The solution of chitosan, cellulose acetate (CA), and gelatin was mixed with eugenol of various concentrations and electrospun into fibers. The antimicrobial activity of the nanofibers was confirmed against *Salmonella typhimurium* and *S. aureus.* In addition to the above examples, various chitosan fibers were functionalized with *Zataria multiflora* (ZEO) and cinnamon (CEO) EOs,[138] hordein-quercetin,[139] pomegranate peel extract,[140] oregano EO (OEO),[141] chrysanthemum oil,[142] phenolic compounds,[143] lauric arginate,[144] and clove oil[145] for potential food packaging applications. Nanocomposite chitosan electrospun films were also produced using montmorillonite[146] and silver nanoparticles (AgNPs)[147] for active food packaging applications.

4.5. Cellulose-based electrospun food packaging materials

Cellulose as a natural polymer found in the cell walls of plants, has many intrinsic benefits such as biodegradability and customizable structure.[148] Cellulose and its derivatives have been widely used for the fabrication of food packaging materials.[149] Numerous studies have utilized cellulose and its derivatives in electrospinning processes to fabricate food packaging films, aiming to enhance barrier properties or boost the mechanical strength of the fibers. This has often been achieved through the incorporation of nanoparticles.[150] Likewise, many electrospun cellulose-based active food packaging materials have been developed. In one example, a composite electrospun nanofiber film, comprising ethyl cellulose and soy protein isolate (SPI) blended with bitter orange peel extract, was developed for food packaging.[151] Variation in the ethyl cellulose/SPI ratio and bitter orange peel extract content influenced fiber morphology, antioxidant activity, and antibacterial efficacy against *E. coli* and *S. aureus*, with a WVTR measured at 657 g m^{-2} per day. Gliadin-ethyl cellulose fibers loaded with cumin seed oil and reinforced with adipic acid were developed for food packaging.[152] Acting as a hydrogen bond cross-linker, adipic acid increased polymer viscosity before electrospinning, resulting in nanofibers with antioxidant properties, strong antibacterial and antifungal activity against various pathogens, and demonstrated biocompatibility with human cells, suggesting their promising potential for food packaging applications. Another ethyl cellulose nanofiber film was produced in the presence of PCL and gelatin and loaded with *Zataria multiflora* EO (ZEO) and ZnO NPs to provide an ideal food packaging substrate.[153] The composite nanofibers were nontoxic and showed the highest antioxidant activity (34.61 ± 1.98%) and antifungal properties against *Penicillium notatum* and *Aspergillus niger*. Such fibers are suitable for active packaging materials for foods

prone to fungal spoilage. Likewise, ethyl cellulose electrospun food packaging material was developed using pullulan and cinnamaldehyde.[154] Bead-free uniform nanofibers were produced with hydrophobicity. The antimicrobial activity of the nanofibers was boosted with cinnamaldehyde content added.

Unlike the above example, active agents were not directly released from the fiber matrix, but from the decorated nanogels. Ethyl cellulose/casein nanofibers were modified with ginger essential oil (GEO)-loaded nanogels made of gelatin and carrageenan gum, enabling controlled release of active agents.[155] Synthesized *via* inverse miniemulsion, the gelatin/carrageenan gum aldehyde nanogels measured 95 nm in diameter, increasing to 110 nm after GEO loading, before being blended with the electrospinning solution and spun into nanofibers. These functional nanofibers demonstrated slow release of GEO, antioxidant activity, and antibacterial efficacy against *E. coli* and *S. aureus*, with enhanced performance with higher nanogel content, suggesting their potential application in food packaging. CA nanofibers were functionalized with chitosan nanoparticles loaded with *Ziziphora clinopodioides* essential oil (ZEO) for packaging beef meat samples.[156] Synthesized concurrently with CA electrospinning, these nanostructures showed enhanced tensile strength and low vapor barrier, outperforming ZEO-loaded CA fibers alone. They exhibited potent antioxidant activity against *E. coli* and *S. aureus*, with a release profile featuring rapid initial release followed by gradual release, effectively inhibiting bacterial growth in refrigerated beef samples. A trilayer bionic nanofibrous membrane, consisting of ethylene-vinyl alcohol copolymer, gelatin, and CA, was electrospun for jerk beef packaging, featuring hierarchical pore networks and asymmetric wettability for moisture control.[157] With a high transport index and moisture management capacity, the membrane extended jerk beef shelf-life by 100% under specified storage conditions. Coaxial electrospinning was utilized to produce CA nanofibers with core–shell structures for active food packaging applications.[158] These nanofiber films, composed of CA and gelatin with eugenol loaded into the core, demonstrated slow release of eugenol over multiple days, confirmed by transmission electron microscopy analysis. Showing substantial antimicrobial efficacy against *E. coli* and *S. aureus*, these findings indicate the potential of these nanofibers as effective food packaging materials.

CA was utilized to create colorimetric nanofibrous films incorporating *Perilla frutescens* anthocyanins and chamomile EO (Fig. 7).[159] These films displayed sensitive and reversible color changes over a pH range of 2–12, sustained release of bioactive agents, antioxidant activity, and significant antimicrobial efficacy against *E. coli* and *S. aureus*, particularly effective against the latter. Another colorimetric freshness monitoring system was developed using electrospun ethyl cellulose/gelatin fibers containing purple sweet potato anthocyanin (PSPA) as a pH indicator.[160] These fibers exhibited enhanced wettability and pH responsiveness compared to cast films, with pork wrapped in the nanofilm showing an extended shelf life of up to 6 days, characterized by color shifts from light pink to light brown and then to brownish-green as freshness declined.

Cellulose nanofibers provided support for chitosan/tannic acid bilayers, enhancing antibacterial protection.[161] The resulting composite mats, fabricated *via* LbL assembly with electrostatic interactions, exhibited increased antimicrobial activity against *S. aureus* and *E. coli*, suggesting their potential application in food packaging due to the biocompatibility of the

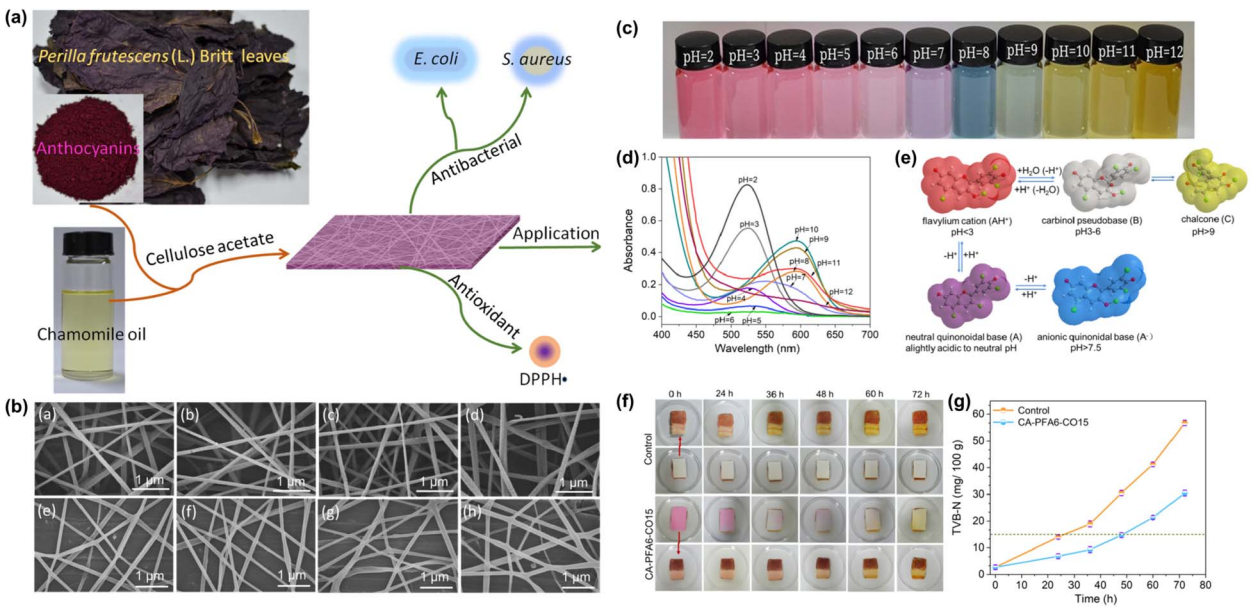

Fig. 7 (a) Schematic illustration of the production of antioxidant and antibacterial CA fibers loaded with chamomile oil and anthocyanins extracted from *Perilla frutescens* (L.) *Britt*. (b) SEM photos of the CA fibers (CA (a), CA-CO5, 10, 15 (b–d), CA-PFA2, 4, 6 (e–g) and CA-PFA6-CO15 (h)). (c) Color changes and absorption spectra (d) of PFA solutions at pH 2–12, corresponding structural transformation (e). (f) CA-PFA6-CO15 for pork freshness monitoring and (g) corresponding levels of TVB-N. The figures were reproduced from ref. 159, Elsevier.

precursors and antimicrobial efficacy of the materials. Another antifungal packaging was developed through the electrospinning of the blend of CA and poly(vinyl chloride) loaded with AgNPs.[162] The incorporation of AgNPs reduced the fiber diameter and decreased the air permeability rates. The hybrid nanofibers could inhibit the growth of yeast and mold because of the AgNPs incorporated. An interesting concept on upcycling blue jeans into fibrous cellulose fibers was reported.[163] Carboxylated carbon nanotubes and GO were integrated into cellulose fiber *via* a wet electrospinning technique. The resulting structures were used for the immobilization of lysozyme enzyme, which could maintain the bioactivity. The fibers were cytocompatible and demonstrated significant antimicrobial activity after immobilization of lysozyme. Another LbL assembly on CA nanofibers was developed using bilayers of chitosan (CS) and epigallocatechin gallate (EGCG) or with bilayers of CS-rectorite (REC) composite (CS-REC) and EGCG.[164] Fibrous structure could be maintained after LbL assembly and the fiber diameter was boosted with the addition of REC. The tensile properties of the fibers did not differentiate significantly with LbL deposition. REC increased the encapsulation efficiency and loading capacity of nanofibers and slowed down the release profile of EGCG. The presence of chitosan and EGCG caused antimicrobial activity against *S. aureus*. A triaxial fiber membrane with CA sheath and PCL as an intermediate a layer and core layer as nisin were developed for long-term antimicrobial activity for potential use in food packaging.[165] The antimicrobial tests were done against *S. aureus* and the antimicrobial activity could be maintained for up to a week. Triaxial electrospun fibers demonstrated excellent antimicrobial activity for up to 5 days and thereafter, they could provide antimicrobial activity for 2 more days. This sustained antimicrobial activity could be attributed to the sustained release of nisin from the fiber core over a long period.

4.6. Zein and gelatin-based electrospun food packaging materials

Utilizing protein-based electrospun materials in food packaging offers a promising path toward sustainable packaging solutions. A number of biomass-derived proteins, including zein and gelatin, have specific properties (*e.g.*, biocompatibility, biodegradability, barrier properties, *etc*) that are well-suited for packaging purposes. These proteins can be isolated from agricultural residues and food wastes to develop active food packaging materials. In one study, gelatin/zein-based nanofibers were produced by incorporating ZnO and gallic acid to develop nanofibers with antioxidant, antibacterial, and antifungal properties for food packaging applications (Fig. 8a and b).[166] The resulting nanofibers were hydrophobic and exhibited antioxidant activity five and nine times higher than that of the

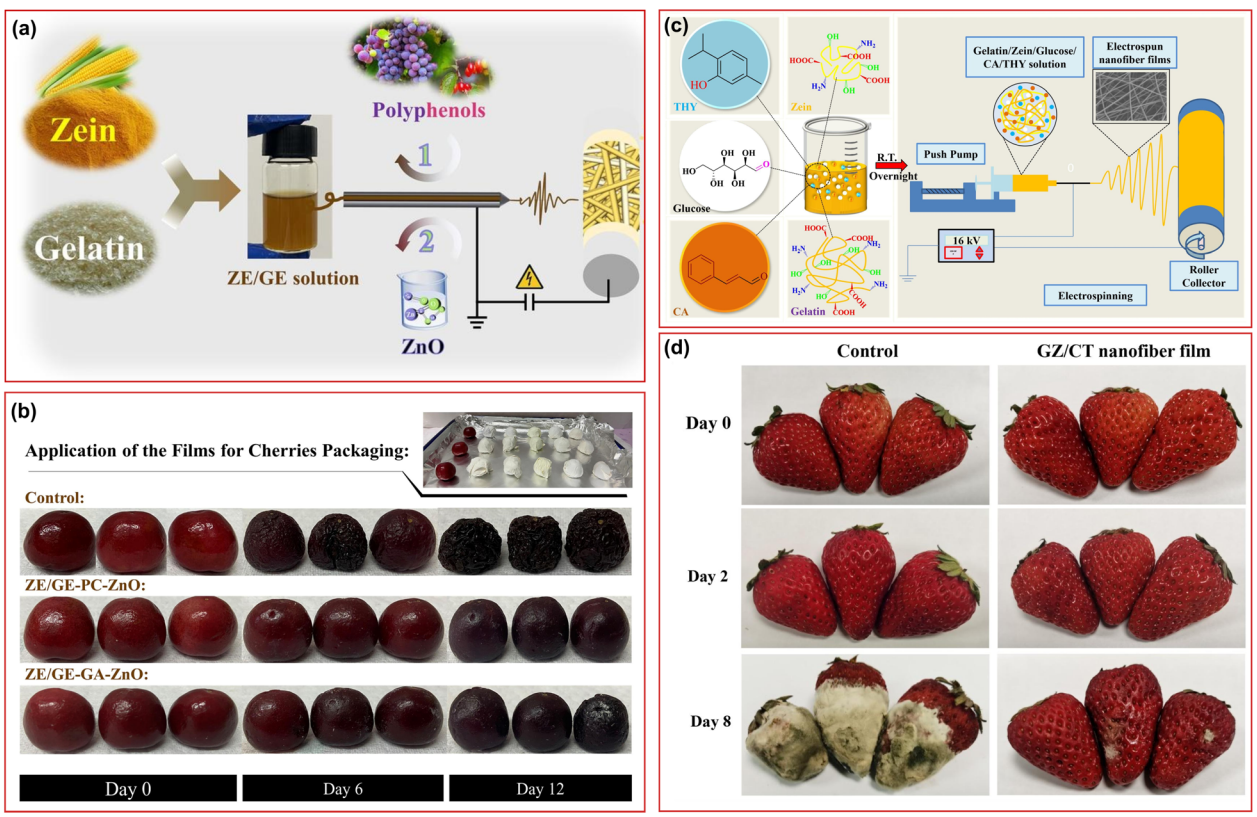

Fig. 8 (a) Development of multifunctional electrospun film using zein and gelatin, functionalized with ZnO and polyphenols. (b) Food packaging performance of the fibers using cherries up to 12 days at 25 °C. (c) Schematic illustration of the production of gelatin/zein nanofibers loaded with cinnamaldehyde and thymol and (d) the use of the electrospun film for strawberry preservation. Figures were reproduced from ref. 166 and 168, Elsevier.

zein/gelatin nanofiber film depending on the composition. The food packaging performance of the fibers was explored over cherry samples (Fig. 8b). On the 11th day of the test, the weight loss and firmness were reduced by more than 20 and 60% to the unwrapped samples. The peak of ethylene release was decreased by nearly half, demonstrating the potential of such fibers for active food packaging applications. In another study, linalool-loaded gelatin/zein fibers were cast on the cast film of carboxymethyl chitosan/Oxalis triangularis ssp. Papilionacea (OTA) extract for the development of a colorimetric film.[167] The resulting film showed good barrier and water resistance while the outer layer of CO-OTA showed colorimetric sensitivity towards pH stimuli with reversible color changes. The inner part of the membrane showed high antibacterial activity against *E. coli* and *S. aureus*, while the encapsulated linalool was released from the fibers in a controlled manner and followed by Fickian diffusion, causing antioxidant activity. The membranes were used for milk freshness monitoring and could double the shelf-life at 25 °C. A gelatin/zein composite nanofiber film loaded with thymol and cinnamaldehyde extended the shelf-life of strawberries by increasing water resistance and reducing the water vapor transmission rate (Fig. 8c and d).[168] It effectively shielded against UV light and inhibited the growth of *E. coli*, *S. aureus*, and *Listeria monocytogenes*. Real-time tests showed that packaged strawberries remained fresh for up to 7 days at room temperature. Another gelatin/zein composite electrospun film loaded with ε-polylysine and gallic acid served as an active food packaging film for tuna.[169] This film exhibited reduced hydrophobicity and antioxidant properties due to the additives, while also displaying antibacterial activity against *Shewanella putrefaciens*. Similarly, electrospun gelatin/chitosan nanofibers loaded with betel leaf ethanolic extract were applied as coatings on PLA film for food packaging, showing antioxidant and antibacterial effects, with improved mechanical properties and reduced water vapor permeability.[170]

Another antimicrobial gelatin electrospun film was developed by incorporating allyl isocyanate (AIC) and supported by pressure-sensitive adhesive (PSA) made of hydroxyethyl cellulose backbone grafted by acrylic acid and methylbutyl acrylate [HEC-*g*-poly(AA-MBA)].[171] The resulting structures with 2% AIC content showed high antimicrobial activity against *E. coli* O157:H7 and *S. aureus* ATCC 25923. Cheese samples covered with these electrospun fibers could maintain their freshness over 5 weeks of storage at 4 °C, demonstrating their effectiveness as antimicrobial food packaging. Antimicrobial fibers were also developed using the combination of zein and PLA, with increased carvacrol content enhancing antioxidant activity and leading to sustained release and these fibers, employed in packaging bread, demonstrated better protection against microbial growth with higher carvacrol levels.[172] Uyar group also developed CD-IC/carvacrol-loaded fibers with gelatin/pullulan as carrier polymers in which CD-ICs of carvacrol minimize the carvacrol loss during electrospinning and enhance its thermal stability.[173] CD molecules were also utilized to form ICs with thymol, enhancing preservation and stability during and after the electrospinning of zein, and zein-thymol/γ-CD ICs nanofibrous mats effectively reduced bacterial growth in beef stored at 4 °C for up to 5 days.[174]

Electrospun nanofibers can be considered as hydrogels after imbibing a significant amount of water while preserving their fibrous structures. Fish gelatin fiber films were functionalized with different loadings of LAE to create food packaging films.[175] These films, produced through electrospinning, could absorb surface water from fish fillets, forming a hydrogel coating. LAE-loaded films exhibited effective antibacterial activity against *S. aureus* and *E. coli*, ultimately extending the refrigerated shelf life of large yellow croaker fillets by about three days. Likewise, electrospun hydrogels of gelatin/chitosan blend loaded with 2-phenylacetic acid were developed for food packaging applications.[176] The resulting hydrogels showed antibacterial activity against foodborne pathogens; *S. aureus* and *E. coli* and could preserve the freshness of chilled chicken for 4 days without spoilage formation. In another study, edible chitooligosaccharide was integrated into a skin gelatin nanofiber-based hydrogel to create antibacterial and antioxidant properties.[177] Cross-linking of the nanofibers occurred *via* the Maillard reaction, facilitated by the addition of glucose, resulting in a highly porous structure upon hydration with a notable swelling ratio of 954%. The incorporation of chitooligosaccharide enhanced both the antioxidant and antibacterial capabilities of the hydrogel, extending the shelf-life of crucian carp by 2 to 4 days, and demonstrating the potential of gelatin-based nanofiber hydrogels for food packaging.

While gelatin-based fibers have been extensively studied for food packaging, their water solubility remains a significant limitation. To address this issue, efforts have focused on developing cross-linked gelatin nanofibers. In one study, water-resistant fish gelatin nanofibers with antioxidant activity were created using chlorogenic acid (CGA) as the active ingredient and citric acid or fructose as cross-linking agents.[178] These fibers exhibited rapid CGA release when exposed to food simulants, with citric acid cross-linked fibers demonstrating superior scavenging activity in DPPH assays.

Gelatin-based nanofibers were utilized for monitoring food freshness through various innovative approaches. Functionalized nanofibers with AuNPs, SnO_2, and black elderberry extract enabled rapid color changes upon exposure to volatiles, facilitating the detection of freshness in packaged Hake fish fillets.[179] Similarly, multifunctional food packaging materials incorporating gelatin/xanthan gum mats containing chitin nanofibers and blackberry anthocyanins were developed for freshness monitoring and extending the shelf-life of Pacific white shrimps, with pH-sensitive color changes indicating microbial growth.[180] Another intelligent packaging system, combining gelatin with anthocyanins from red radish, showed color alterations correlating with pH changes, enabling real-time monitoring of meat spoilage through volatile compound release.[181]

Zein nanofibers loaded with ferulic acid, quercetin, gallic acid, or procyanidin were created for active food packaging, enhancing both antioxidant capacity and hydrophobicity.[182] When applied in cherry packaging, the resulting electrospun film significantly improved water loss, hardness, and gas release compared to unwrapped cherries. Antioxidant zein

nanofibers were created using a core–shell approach, with ferulic acid loaded in the core with PEO and the shell layer composed of zein.[183] This core-loading strategy decelerated the release rate of ferulic acid compared to fibers where it was loaded into the sheath, resulting in enhanced antioxidant activity and reduced weight loss of apple slices compared to blank fibers. Cross-linked zein nanofibers loaded with star anise EO/β-CD ICs were prepared for active food packaging by initially forming the inclusion complexes through mixing, then blending with zein solution for electrospinning.[184] These nanofibers displayed enhanced thermal stability, mechanical strength, and water resistance, and exhibited exceptional antioxidant and antimicrobial properties against *E. coli* and *S. aureus*. Antimicrobial zein-based electrospun films were also produced by incorporating κ-carrageenan, ZnO NPs, and rosemary EO.[185] The resulting electrospun films exhibited satisfactory thermal, mechanical, and surface hydrophobicity while showing both antioxidant and antibacterial activity.

An antiviral multilayer film was developed using cinnamaldehyde-loaded zein fibers and PHB film.[186] Cinnamaldehyde-loaded zein fibers were deposited onto the inner sides of PHB films, which were produced through compression molding. The composite films showed antiviral activity against norovirus surrogates, murine norovirus (MNV), feline calicivirus (FCV), and hepatitis. They observed higher activity for norovirus surrogates compared to the virus HAV in antimicrobial biodegradable multilayer systems. Another antimicrobial multilayer film was developed using polyhydroxybutyrate-*co*-valerate film as a support with cinnamaldehyde-loaded zein fibers and an alginate-based outer layer.[187] The multilayer films exhibited antibacterial activity against *Listeria monocytogenes* and slowly released encapsulated cinnamaldehyde in food simulants, making them promising candidates for active food packaging materials. Food packaging applications of protein-based and other biomass-derived electrospun fibers from various sources are compiled in Table 1.

4.7. Other sustainable polymers-based electrospun food packaging materials

Except polymers listed above, there are other biobased sustainable polymers including bio-based polyamides, which are synthesized from castor oil, sugar-derived polyethylene, polyesters from vegetable oils, lignin, and pectin. Some of these polymers have been used for the fabrication of electrospun active food packaging materials. In a study, PLA/rice husk lignin fibers were prepared by electrospinning technique for food packaging application.[199] The lignin at different concentrations was loaded into ultrafine PLA fibers. Notably, PLA fibers containing 2.5% lignin demonstrated antioxidant activity, displaying approximately 70% reduction for both DPPH and ABTS radicals. SPI-based fibers were also produced for antioxidant food packaging applications. In this regard, antioxidant β-carotene was encapsulated in a mixture of SPI and PVA and then electrospun onto a polyhydroxybutyrate-*co*-valerate (PHB92/PHV8) film, which was attached to the film through the annealing process.[217] The *in vitro* release assay of the antioxidant in soybean oil, mimicking fatty foods, showed that the heat treatment (annealing) resulted in a reduced release rate and a more prolonged presence of the bioactive compound. Likewise, antimicrobial SPI fibers were produced through encapsulation of *Cinnamon Zeylanicum* (Cz) and *Zataria Multiflora* EOs.[218] Nanofibers with 20% Cz showed reductions of 72%, 56%, and 42% in *S. aureus*, *B. cereus*, and *S. Typhimurium*, respectively. Like the above example, SPI fibers loaded with ginger EO were produced for the development of antimicrobial fibers.[219] Soy protein amyloid fibrils (SAFs) were also used to develop antibacterial food packaging materials. The incorporation of SAFs boosted the mechanical properties of the fibers and increased the hydrophobicity. The composite fibers showed antibacterial activity against *E. coli* and *S. aureus*. In another study, antimicrobial electrospun films were also prepared using bio-based polyamide 11 (PA 11) and a nanohybrid of halloysite nanotubes (HNTs) filled with 50 wt% lysozyme as a natural antimicrobial molecule.[220] Antimicrobial evaluations using chicken meat stored at 4 °C for 6, 9, and 13 days demonstrated the efficacy of the nanofibers in inhibiting bacterial growth, highlighting their potential suitability for food packaging applications.

Pectin, a complex carbohydrate found in the cell walls of plants, especially in fruits like apples, citrus fruits, and berries, is widely utilized in the food industry as a gelling agent, thickener, stabilizer, and in the development of active food packaging materials.[221] In one study, pectin/PEO blends were electrospun into fibers in the presence of glycerol.[222] Pectin electrospun fibers were used as an interlayer between two external layers of PHBV films by annealing. The resulting multilayer films showed enhanced barrier performance to water vapor and limonene. In another study, composite fibers of pectin with chitosan and PVA were produced as antibacterial electrospun films.[223] The disk diffusion test showed that the fibers exhibited significant antibacterial activity against *S. aureus* but not against *E. coli*. Another protein-based electrospun food packaging film through an LbL technique was developed using sarcoplasmic protein for the controlled release of CUR.[224] The multilayer structure comprised CA as the base and top layers, with CUR-loaded fibers of direct freeze-dried sarcoplasmic protein (DFSP) or chitosan flocculated sarcoplasmic protein (CFSP) as the middle layer. SEM analysis confirmed the formation of the multilayered nanofiber films, exhibiting slow release of CUR from both DFSP and CFSP layers, suggesting promising applications in food packaging.

5. Performance comparison of biomass-derived electrospun food packaging materials

Each biomass-derived polymer presents unique advantages and challenges when used in electrospinning for food packaging applications. The polymer's intrinsic properties, such as hydrophobicity, bioactivity, electrospinnability, mechanical strength, and barrier characteristics, greatly influence their suitability and performance in this field.

Table 1 Electrospun films made from sustainable polymers for active food packaging applications

Polymer(s)	Active agent(s)	Fiber properties	Activity	Release profile	Description	Ref.
Ethylcellulose, gelatin	ZnO NPs	Bead-free uniform fibers (558–804 nm)	Antimicrobial activity against *E. coli* and *S. aureus*	N.D.	Uniform hydrophobic nanofibers with different ZnO NP loadings were produced. The antimicrobial activity of the nanofibers was enhanced by UV irradiation	188
PLA	Wormwood oil (WO)	Bead-free fibers with a mean diameter of 260 nm	Antimicrobial activity against *E. coli* and *S. aureus*	N.D.	The antimicrobial activity of nanofibers was strongly influenced by the added WO content	189
PLA	AgNPs & *Thymus daenensis* EO	Beaded-fiber morphology	Antimicrobial activity against *E. coli* and *S. aureus*	Sustained and long-term release of AgNPs and EO	An electrospun PLA nanofiber film loaded with AgNPs and EO showed antimicrobial activity due to the sustained release of bioactive agents	190
PLA	Ferulic acid	The morphology of the fibers showed large differences depending on the solvent	Antimicrobial activity against *L. innocua*	N.D.	The encapsulation efficiency of ferulic acid into electrospun PLA fibers was in the range of 84–96%. The antimicrobial PLA fibers were easily produced through the incorporation of ferulic acid	191
PLA	Carvacrol (CRV), nisin (Nis)	Nonuniform fibers	Antimicrobial activity against *L. monocytogenes*, *S. Enteritidis*, *E. coli*, and *S. aureus*	Sustained release of CRV and Nis from the fibers	The incorporation of CRV and Nis improved the elastic modulus of the fibers. The electrospun fibers loaded with 20% CRV showed antimicrobial activity against different bacteria	192
PLA, guar gum (GG)	Thyme EO (TEO)	Nonuniform fibers with mean diameters of 395 nm (with 10% TEO) and 347 nm (with 30% TEO)	Antioxidant activity (over DPPH radical assay) and antibacterial activity against *E. coli* and *S. aureus*	Sustained release profiles: the lesser content is released as the TWO content increases	The composite fibers of PLA and GG showed both antioxidant and antimicrobial activities. The resulting nanofibers were biocompatible	193
PLA	Cinnamaldehyde (CMA), tea polyphenol (TP)	Coaxial electrospinning, core shell-fibers with uniform structure	Antibacterial activity	N.D.	The presence of CMA showed synergistic antibacterial activity. CMA destroyed the	194

Table 1 (*Contd.*)

Polymer(s)	Active agent(s)	Fiber properties	Activity	Release profile	Description	Ref.
					phospholipid layer of the cell membrane, while TP destroyed the extracellular proteins, resulting in cell membrane perforation and cell death	
Zein, PLA, and hydroxypropyl methylcellulose (HPMC)	Zenian (*Carum copticum*) EO (ZO)	Nonuniform fibers with mean diameters between 718 and 335 nm	Antioxidant activity against DPPH radicals and antimicrobial activity against *E. coli* and *S. aureus*	The cumulative release decreased with higher ZO content	Biocompatible composite nanofibers with both antioxidant and antimicrobial activity	195
PHBV	Fe doped ZnO NPs	Beaded fiber morphology	Antimicrobial activity against *E. coli* and *S. aureus*	N.D.	PLA/PHBV/ZnO:Fex electrospun films demonstrate remarkable antibacterial efficacy against *Pseudomonas aeruginosa* (ATCC-27853) by producing a higher quantity of perhydroxyl (˙OOH) radicals	196
PLA, PVA	LAE	Core–shell fibers with ribbon-flat morphology	The highest concentrations of LAE released from the fibers to both simulants corresponded to LAE's MIC values against *L. innocua*	After the initial burst release, sustained release of LAE from the fibers. The release kinetics fit well with Fick's law	LAE-loaded core–shell fibers were prepared for sustained release of LAE from the fibers. The resulting structures showed antimicrobial activity and demonstrated their potential for active food packaging	197
PLA, chitosan nanoparticles	Cinnamon EO (CEO)	The incorporation of nanoparticles worsened the fiber morphology and resulted in beaded fibers	Antibacterial activity	The sustained release of the CEO was observed	The PLA/CS-CEO fibers showed increased, sustained inactivation rates of *E. coli* and *S. aureus* over time, which is attributed to the continuous release of CEO.	198
PLA	Lignin (from rice husk)	Higher lignin load led to more uniform fibers	The nanofibers showed antioxidant activity through DPPH and ABTS assays	N.D.	Lignin-loaded PLA nanofibers were prepared. The resulting nanofibers exhibited antioxidant activity and demonstrated their potential for	199

Table 1 (*Contd.*)

Polymer(s)	Active agent(s)	Fiber properties	Activity	Release profile	Description	Ref.
					food packaging applications	
Gelatin, chitosan	3-Phenyllactic acid	The fiber morphology was deteriorated by the incorporation of 3-phenyllactic acid	Antibacterial activity against *S. enterica Enteritidis* and *S. aureus*	N.D.	Depending on the loading of 3-phenylacetic acid, the nanofibers exhibited tunable water vapor permeability and antibacterial activity, demonstrating their potential for active food packaging applications	200
Chitosan, carrageenan	*Malva sylvestris* extract (MSE)	Rounded/ribbon-like fibers	Intelligent (pH-based sensing)	N.D.	Fiber mats change color depending on the pH value. When assessing the freshness of silver carp fillets, indicators of total bacterial count, the number of psychrotrophic bacteria, pH (8.10), and total volatile basic nitrogen (40.18 mg N/100 g) were measured, which indicate a high bacterial count and showed increased nitrogen levels	201
Gelatin, zein, PVA	—	Beaded fiber morphology for zein/gelatin and gelatin fibers, while bead-free fiber morphology for PVA/gelatin fibers	Antioxidant activity	N.D.	The composite nanofiber mat exhibited favorable thermal and mechanical characteristics, coupled with outstanding antioxidant performance. The sweet potatoes and potatoes demonstrated an extended shelf life of 50 days, while the kimchi maintained its quality for 30 days	202
Gelatin	Cinnamaldehyde EO (CEO), limonene EO (LEO), and eugenol EO (EEO)	Uniform nanofiber structures with different EOs in the presence of β-CD	Antioxidant and antibacterial activity	Sustained release of EOs from gelatin mats	EOs were dissolved with β-CD and the resulting ICs were loaded into electrospun fibers. The resulting nanofibers exhibited both antioxidant and	203

Table 1 (*Contd.*)

Polymer(s)	Active agent(s)	Fiber properties	Activity	Release profile	Description	Ref.
					antibacterial activity, demonstrating their potential for active food packaging	
Gelatin	Eugenol	Beaded-fiber morphology	Antibacterial activity due to loaded eugenol	N.D.	Eugenol-loaded gelatin fibers can prolong the shelf life of beef samples while preserving their textural properties (such as hardness, gumminess, and chewiness) and sensory qualities throughout the storage period	204
Gelatin/*Plantago psyllium* L. seed gum (PPSG)	Cuminum cyminum EO (CCEO)	Bead-free fiber morphology	Antibacterial activity against *S. aureus*	Sustained release of CCEO from the nanofibers	The first nanoemulsion of CCEO was prepared with the emulsifier TWEEN 20 and then added to the polymer solution for electrospinning. Uniform fibers with antibacterial activity, as well as sustained release profiles, have the potential for active applications in food packaging	205
Zein	Phycocyanin & Spirulina (AEES) extract	Bead-free fiber morphology for low loading of AEES and bead-free fiber morphology for fibers loaded with phycocyanin	Higher antibacterial activity against *S. aureus* than *E. coli*	N.D.	Zein electrospun fibers loaded with AEES or phycocyanin showed antioxidant and antibacterial activity. Both fibers could decrease the peroxide value (PV) and 2-thiobarbituric acid (TBA) values of walnut kernels during 24 weeks of storage, demonstrating their effectiveness against lipid oxidation	206
Zein	Sakacin	Bead-free fibers were produced at	The fibers showed antibacterial activity	N.D.	Electrospun zein/sakacin nanofibers showed activity	207

Table 1 (*Contd.*)

Polymer(s)	Active agent(s)	Fiber properties	Activity	Release profile	Description	Ref.
		different loading of sakacin	against *Listeria innocua*		against *L. innocua* and also extended the refrigerator shelf life of quail breasts. The antimicrobial activity of the fibers could be increased by the zein content	
Zein	Phenolic-enriched extracts (PEE)	Bead-free fibers	The fibers showed antioxidant activity because of the loaded PEE.	Sustained release of PPE from the fibers	Antioxidant bead-free zein nanofibers were produced through electrospinning of zein/PEE blends. The cross-linking of zein fibers could improve structural integrity on water contact	208
Zein	Citronellol-rich *Origanum vulgare* EO	Bead-free fibers at various EO loadings	The fibers showed both antioxidant and antibacterial activity	N.D.	The encapsulation efficiency and loading capacity of the EO were in the range of 71.56–85.8% and 8.88–40.93%, respectively. Due to the antibacterial and antioxidant activity of the EO, the fibers exhibited both activities	209
Chitosan, PVA	Catechin	The fiber morphology deteriorated with catechin content added	The nanofibers showed antioxidant activity	Sustained release of catechin	Preservation studies indicated that the electrospun film effectively prolonged the shelf life of strawberries, with the film containing 0.8% catechin concentration demonstrating the most favorable preservation outcomes	210
Chitosan, flaxseed mucilage	*Ziziphora clinopodioides* EO (ZEO) and sesame oil (SO)	Bead-free fibers	The fibers showed both antioxidant and antibacterial activity	Sustained release of SO and ZEO	The nanofibers exhibited a continuous release of ZEO and SO over 96 hours, exhibiting notable antioxidant and antimicrobial properties, thereby indicating their promise for use in active food	211

Table 1 (*Contd.*)

Polymer(s)	Active agent(s)	Fiber properties	Activity	Release profile	Description	Ref.
					packaging applications	
Chitosan, PEO	Anthocyanin	Highly branched fiber morphology	The anthocyanin-loaded fibers showed both antioxidant and antibacterial activity	N.D.	The use of anthocyanin/CS/PEO nanofibers as beef packaging material resulted in a color transition from white or light yellow to yellow-green as the beef decayed over storage time, facilitating visual assessment of beef quality	212
Gelatin, chitosan	CUR	Bead-free fiber morphology at different loadings of CUR	The CUR-loaded fibers showed both antioxidant and antibacterial activity. The fibers also showed color change depending on ammonia content	N.D.	The addition of CUR significantly enhanced the antioxidant and antimicrobial properties of nanofibers. Additionally, the nanofibers containing 0.2% CUR demonstrated higher colorimetric sensitivity to ammonia, with detection occurring within 3 minutes	213
Zein	Jaboticaba peel extract (JPE)		The nanofibers showed antibacterial activity	N.D.	Bilayer films of casted chitosan and electrospun zein/JPE fibers were developed. The bilayer film improved the barrier property while endowing the bioactivity	214
PLA	AgNPs, vitamin E	Bead-free fibers	Antibacterial and antioxidant activity	N.D.	The tests conducted on fresh apple and apple juice revealed that the PLA/Ag/vitamin E nanofiber membrane effectively decreased the activity of polyphenol oxidase	215
Alginate	*Lactobacillus paracasei* KS-199	Beaded fiber structures resulting from bacterial incorporation	—	N.D.	Encapsulating the bacteria at the nanoscale significantly increased its survival in simulated gastric juice, raising the	216

Table 1 (*Contd.*)

Polymer(s)	Active agent(s)	Fiber properties	Activity	Release profile	Description	Ref.
					viability rate from 64.1 to 70.8 log cfu mL^{-1}. Additionally, this nanoencapsulation improved its viability in kefir, with the survival rate increasing from 6.65 to 7.38 log cfu mL^{-1}	

Hydrophobicity plays a crucial role in determining the direct applicability of electrospun films. Polymers like PLA and PHAs are inherently hydrophobic, which enables their electrospun films to be used directly in food packaging. Their water-repellent nature prevents moisture absorption, maintaining the integrity of the packaging and protecting the food from external humidity. Conversely, hydrophilic biomass polymers such as cellulose, chitosan, and various proteins require additional modifications to render them suitable for food packaging. These modifications include cross-linking with appropriate agents or grafting hydrophobic moieties onto their structure. Cross-linking enhances the polymer's structural stability and reduces its water solubility, while hydrophobic modifications improve water resistance. Without these alterations, hydrophilic polymers may dissolve or degrade in high-moisture environments, compromising their packaging function.

However, some hydrophilic polymers offer unique benefits. Chitosan, for instance, possesses inherent antimicrobial properties, making it an excellent candidate for active food packaging.[225] Its ability to inhibit microbial growth can extend food shelf life and maintain food safety without the need for additional antimicrobial agents. This property sets chitosan apart from other biomass-derived polymers in terms of functionality. In this regard, zein or gelatin as biomass-derived polymer is not intrinsically antimicrobial and its applications as antimicrobial films require additional antimicrobial agents/polymers, such as chitosan.[226]

The process of electrospinning itself varies in efficiency depending on the polymer. Some polymers, like chitosan, pose challenges during electrospinning due to their high viscosity and strong hydrogen bonding, resulting in low throughput.[227] To overcome this, chitosan is often blended with more electrospinnable polymers (*e.g.*, PEO, pullulan, PVA) to enhance jet formation and increase productivity.[228] The choice of the co-polymer depends on factors such as compatibility, desired final properties, and the intended application.

The mechanical properties of electrospun films are crucial for packaging applications, as they must endure the stresses of handling, transportation, and storage. These properties vary significantly based on factors such as fiber diameter, distribution, the presence of beads, fiber orientation, and the molecular weight of the polymer used.[229] Additives can also cause notable variations in mechanical properties. PLA is particularly noteworthy, offering high mechanical strength and stretchability compared to many other biomass-derived polymers. The fibrous structure of electrospun PLA can even exceed the mechanical performance of its bulk form in terms of stretchability, making it an attractive option for robust food packaging. Randomly aligned PLA fibers have a tensile strength of 3.9 MPa and a modulus of 43.8 MPa, with an elongation at break of 87.6%.[230] Electrospun CA fibers, on the other hand, exhibit a higher tensile strength (12.1 MPa) and modulus (1170 MPa) but lower stretchability compared to PLA fibers.[231] Conversely, electrospun chitosan fibers have much lower values, with a tensile strength of approximately 0.5 MPa and a tensile modulus of about 10 MPa.[232] Gelatin fibers show better mechanical properties than chitosan, with a tensile strength of 1.6 MPa and an elongation at break of 17%.[233] Fiber alignment also influences mechanical properties. For instance, aligned PLA fibers show higher tensile strength (4.5 MPa) and modulus (62.5 MPa) compared to randomly deposited PLA fibers, although they have a lower elongation at break (27.4%).[230]

Like the mechanical properties, barrier properties, especially against water vapor and gases, are paramount in food packaging to control moisture content and atmospheric composition inside the package. Hydrophobic polymers like PLA and PHA generally exhibit relatively low oxygen, carbon dioxide, and water vapor permeability.[234,235] Whereas, CA has high water vapor permeability compared to PLA and PHA.[235] The porous nature of electrospun films, with gaps between fiber strands, can compromise their gas barrier properties. To address this, researchers have developed bilayer or multilayer electrospun films, along with cast films. These composite structures significantly enhance barrier properties, effectively slowing down gas transmission. In this regard, the incorporation of electrospun zein interlayer decreased the water vapor and oxygen permeability of polyhydroxybutyrate-*co*-valerate films.[236] Increasing the deposition time of the zein fibers decreased both permeability due to increased layer thickness.

In summary, the selection of a biomass-derived polymer for electrospun food packaging involves careful consideration of

various factors. Hydrophobicity influences direct usability, while hydrophilic polymers may require modifications. Some polymers, like chitosan, offer additional functionalities such as antimicrobial activity.[237] Electrospinnability varies among polymers, with some needing co-polymer blending to improve processability.[238] Mechanical strength is crucial for package durability, with PLA excelling in this area. Lastly, barrier properties are enhanced through multilayer designs to overcome the inherent porosity of electrospun films. By understanding these aspects, researchers can tailor electrospun films of biomass-derived polymers to create effective, sustainable food packaging solutions.

6. Environmental impact and economic viability of electrospun food packaging materials

6.1. Environmental impact assessment

While the use of biomass-derived polymers inherently suggests a more sustainable approach compared to petroleum-based plastics, a thorough environmental impact assessment is crucial to validate this assumption. This section compares the environmental footprint of electrospun biomass-derived polymers with traditional packaging materials and explores potential challenges in scaling up production.

The carbon footprint of packaging materials is a key indicator of their environmental impact. Studies have shown that biomass-derived polymers generally have a lower carbon footprint than petroleum-based plastics.[239] For instance, a life cycle assessment (LCA) by Gironi *et al.* compared PET (poly(ethylene terephthalate)), plastic derived from fossil resources, and PLA, bioplastic derived from sugar cane, and found that global warming $kgCO_{2eq}$ per 1000 bottles for PLA (*i.e.*, 17.202) were much lower than for the PET-based bottles (*i.e.*, 38.186).[240] S. Ramakrishna and colleagues reported that the life cycle of PLA involves an energy-intensive process for converting bio-sources to lactic acid and subsequently to PLA, resulting in the release of a substantial amount of CO_2 into the atmosphere.[241] According to the available data, more than 50% (2.8 kg CO_2 per kg PLA) of the released CO_2 in the PLA life cycle belongs to its conversion. Thus, more studies should done on optimizing the conversion process of PLA to make PLA a low carbon-material. It's important to note that the carbon footprint can vary depending on the specific biomass source and processing methods. Factors such as land use change, fertilizer use, and transportation distances can impact the overall emissions.[242] Therefore, locally sourced and sustainably managed biomass feedstocks are crucial for maximizing carbon benefits. Energy consumption is another critical factor in environmental sustainability.[243] G. T. Beckham and his team reported on the energy consumption and greenhouse gas emissions linked to plastic use, revealing that major commodity polymers, each with a global consumption of at least 1 million metric tons annually, contribute to approximately 3.2 quadrillion Btus of energy use and 104 million metric tons of CO_2 equivalent emissions each year in the United States alone.[244] In another study, carbon emissions of traditional plastic products were compared with biodegradable bioplastic products (BPPs), and they found that carbon emissions of 1000 traditional plastic products (plastic bags, lunch boxes, cups, *etc.*) were 52.09–150.36 carbon emissions equivalent of per kilogram (kg CO_{2eq}), with the stage of plastic production contributing 50.71–50.77%. In comparison, 1000 similar BPPs topped out at 21.06–56.86 kg CO_{2eq}, approximately 13.53–62.19% lower than traditional plastic products.[245]

One of the most significant advantages of biomass-derived polymers is their end-of-life disposal options. Many of these polymers, such as PLA,[66] polyhydroxyalkanoates (PHAs),[97] and cellulose derivatives,[246] are biodegradable or compostable under specific conditions. This characteristic reduces the burden on landfills and mitigates issues like microplastic pollution in oceans. In contrast, traditional plastics can persist in the environment for hundreds of years.[247] Even with recycling efforts, only 9% of all plastic waste ever produced has been recycled.[248] However, it's crucial to note that not all biomass-derived polymers are biodegradable, and even those that may require industrial composting facilities for proper degradation. Therefore, investment in composting infrastructure and clear labeling for consumers are essential. While the environmental benefits of electrospun biomass-derived polymers are evident, scaling up production to meet commercial demand presents challenges that could impact their sustainability.

Some biomass processing methods require significant water, which could strain local water resources.[249] Developing water-efficient processes and utilizing wastewater treatment and recycling systems are crucial. While many solvents used in electrospinning are less harmful than those in traditional plastic processing, some are still toxic. Ongoing research into green solvents and solvent recovery systems is essential for minimizing environmental impact.[250] To fully understand and improve the environmental impact of electrospun biomass-derived polymers, ongoing LCAs are crucial. These assessments should cover all stages from feedstock production to end-of-life disposal, considering regional variations in energy mix, transportation, and waste management infrastructure. Furthermore, the principles of green chemistry and circular economy should guide process optimization. This includes designing for recyclability, using renewable energy in manufacturing, and developing closed-loop systems where materials and solvents are recycled.

6.2. Economic viability assessment

While the environmental benefits and technical feasibility of electrospun biomass-derived polymers for active food packaging are obvious, their widespread adoption depends on market acceptance and economic feasibility. This section briefly examines key factors that influence the practical implementation of these materials on a larger scale.

The cost-effectiveness of electrospun biomass-derived polymers is crucial for their market adoption. At present, these materials are more expensive to produce than traditional plastics because of the need for specialized equipment for production and the higher costs of certain biomass-derived polymers.

However, as the technology progresses and economies of scale are realized, these costs are expected to decline. For example, the application of PHAs is constrained by their high production costs, especially due to the expense of substrates.[251] Therefore, a wide range of carbon-rich by-products and agro-wastes from industry (*e.g.*, sugar/starch-based,[252,253] whey,[254] oil/glycerol,[255,256] algal[257]) have been used for PHA production to reduce the cost. Moreover, as governments worldwide introduce more strict regulations on single-use plastics and impose taxes or bans on non-biodegradable materials, the economic landscape is shifting. These policy changes are likely to make conventional plastics more expensive, thereby narrowing the cost gap with biomass-derived polymers. Additionally, increased consumer awareness about environmental issues may drive demand for eco-friendly packaging, creating a larger market that could further reduce production costs. The ability of these polymers, as demonstrated through electrospinning, to extend food shelf life and reduce waste presents a strong economic incentive for food producers and retailers. Adopting this technology could result in substantial long-term savings. In this regard, recent advancements in electrospinning techniques, such as needleless electrospinning[258] and multi-jet electrospinning,[259] as well as using industrial scale electrospinners,[260,261] have demonstrated the potential to enhance production rates and lower costs.

As consumers become more environmentally conscious, the demand for sustainable packaging solutions is on the rise, in line with the United Nations's SDGs. This trend is prompting food companies to explore eco-friendly alternatives. Nonetheless, consumer acceptance relies on the packaging's ability to perform well. For electrospun biomass-derived polymers to be widely accepted, they must match or exceed conventional packaging in terms of food preservation, convenience, and visual appeal. Regulatory approval is vital for the adoption of new food packaging materials. In the United States, the Food and Drug Administration (FDA) ensures that food contact materials are safe for their intended use.[262,263] Similarly, in Europe, the European Food Safety Authority (EFSA) imposes equivalent requirements.[264,265] While obtaining these approvals can be lengthy and costly, they are necessary for entering the market.

Shifting to electrospun biomass-derived polymers demands considerable investment in research, development, and infrastructure. Nonetheless, the rising interest in sustainable technologies has opened up more funding opportunities. For instance, the European Union's Horizon program has dedicated substantial resources to research in bio-based products.[266] Furthermore, collaborations between academia and industry can speed up the development and scaling of these technologies.

7. Current challenges and future perspectives

Despite the numerous benefits they offer, electrospun fibers derived from sustainable polymers face some primary obstacles in their application for food packaging. These include ensuring biocompatibility and compliance with food safety regulations, optimizing barrier properties, and adhesion issues to food surfaces, and addressing scalability and cost-effectiveness issues. Similarly, ensuring the structural stability of these materials is paramount, particularly considering that some sustainable polymers (*e.g.*, chitosan, gelatin, starch, *etc.*) are hydrophilic, posing challenges in maintaining the integrity and performance of the packaging over time, especially in environments with fluctuating moisture levels. In this regard, hydrophilic fibers could dissolve onto the packaged food. Efficient cross-linking techniques are crucial for improving the structural stability of materials. Nonetheless, this procedure introduces complications that are unfavorable for widespread industrial manufacturing. Addressing this challenge could involve modifying these hydrophilic polymers with hydrophobic groups, offering a rational solution to the issue. This strategic alteration can potentially enhance the structural stability of the packaging material, mitigating concerns associated with moisture-induced degradation. Likewise, a significant concern in using electrospun fibers for food packaging lies in the presence of residual fibers on food samples. This can occur due to the soft nanostructured nature of electrospun mats. Unlike traditional film-based packaging, these fibers may inadvertently adhere to food, leading to contamination. In this regard, bioactive electrospun fibers can be employed as an interlayer between the food contact film as previously reported for eugenol-loaded PHBV fibers between cast-extruded PHB sheet and a commercial PHBV film.[103]

Electrospinning of some biomass-derived polymers presents challenges due to their inherent characteristics like high viscosity, high surface tension, or low solubility. Fine-tuning electrospinning parameters to yield fibers with consistent morphology and desired attributes is crucial yet technically complex. Likewise, future research into novel biomass-derived polymers and blends can result in materials tailored for food packaging needs. This entails investigating nanocomposites, bio-based additives, and functional coatings to enhance electrospun material performance. Further refining the electrospinning process, including innovating new techniques and equipment (*e.g.*, electroblowing), can enhance the efficiency, scalability, and reproducibility of producing biomass-derived electrospun materials for food packaging.

While electrospun materials for food packaging hold promise, a key hurdle is achieving mass production at an economically competitive cost. To rival traditional packaging materials, the expense of sustainable biomass-derived polymers must be addressed. Mechanical strength and barrier properties are vital for effective food preservation. When electrospun films alone don't suffice in barrier properties, combining them with conventional films can enhance (bio)activity while improving barrier properties. Adopting circular principles, such as recycling and biodegradability, can further boost the sustainability of electrospun materials sourced from biomass. Pioneering recycling methods or designing materials easily compostable post-use can reduce environmental impact and foster a more sustainable packaging ecosystem.

Overall, the emerging field of electrospun food packaging using sustainable biomass-derived polymers presents a compelling opportunity for environmentally friendly solutions in the active packaging industry. The development of electrospun packaging using sustainable polymers is an example of the dynamic interaction between science, industry, and environmental protection. It highlights the importance of a multifaceted approach to mainstreaming environmentally conscious materials into packaging practices. Hence, addressing these challenges necessitates a multidisciplinary approach, engaging chemists, food engineers, and materials scientists.

8. Conclusion

In the field of active food packaging, electrospun materials offer a variety of benefits, ranging from their antioxidant, antibacterial, and antifungal properties to their innovative smart packaging capabilities, all crucial factors in extending the shelf life of various food products. Incorporating functional molecules or groups into electrospun fibers emerges as a crucial strategy to enhance their effectiveness and impart robust antioxidant, antibacterial, and antifungal properties. Furthermore, the emergence of electrospun nanofibers loaded with sensory elements presents an intriguing challenge for smart food packaging. Through this strategic connection, these materials not only protect but also adapt to environmental fluctuations, providing real-time insights into the freshness and quality of packaged food. To this end, a variety of sustainable polymers have been utilized to produce a comprehensive range of electrospun materials loaded with antioxidant, antibacterial, and antifungal agents, and sensory components to detect changes in foods. Examples such as PLA, PHA, starch, cellulose, chitosan, zein, and gelatin highlight the concerted effort to align packaging methods with environmentally conscious principles. While many of these electrospun materials exhibit bioactive properties and sustainability, certain hydrophilic variants dissolve upon contact with water. Despite efforts to stabilize fibers through cross-linking pathways, this process can increase complexity and potentially result in adverse health effects from the cross-linking agents used. Therefore, there is an urgent need for further research focusing on hydrophobic electrospun fibers from sustainable polymers. In conclusion, electrospun nanofibrous materials are being extensively explored in scientific studies in the field of active food packaging, and due to their tailorable structures and biofunctionalities, these innovative nanofibrous materials are poised to emerge as alternatives to traditional plastic film counterparts and possess superior properties. To this end, and in line with SDG 2030, ongoing and future research efforts would likely continue on the use of sustainable polymers from biomass, taking advantage of their sustainability, environmental friendliness, and inherent structural advantages.

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

Conflicts of interest

There are no conflicts to declare.

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