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REVIEW

Electrospinning of Natural Biopolymers for Innovative Food Applications: A Review

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Abstract

As a result of the growing environmental awareness and increasing needs of functional materials in food industry, electrospinning natural biopolymers has aroused great attention across the aspects of the research and industry. Electrospinning processes are very versatile, simple and scalable, providing an enrichment of products in different areas with improved functionalities. The prepared micro- and/or nanofibers have versatile properties with high porosity, large surface area, and tunable lengthscale, and they are low in cost and easy to be functionalized without the requirement of high-purity raw materials, which showed high potential for industrialization. Natural biopolymers such as proteins and polysaccharides are high quality starting materials used for electrospinning as they not only have abundant sources, but also are biocompatible and degradable that can meet the criteria of safety to use in food areas. However, there are still issues encountered when electrospinning natural biopolymers, such as the difficulty in spinning globular structure mostly appeared in plant proteins, and the poor water vapor barrier properties of some polysaccharide electrospinned fibers. The aim of this review is to offer a comprehensive view of basic knowledge of these commonly utilized natural biopolymers and their fiber fabrication methods, the functional characteristics of micro/nanofibers engineered. Moreover, the emerging applications in the food-related areas including food active packaging, encapsulation, enzyme immobilization, and food biosensing were underlined in order to foster the development of electrospinning fibers in food industry.

Keywords Electrospinning · Nanofibers · Nature polymers · Encapsulation · Food-grade materials

Introduction

Electrospinning is a fiber production technique that uses a very high-voltage source to structure low purity raw materials into nano/micron fibers (Drobota et al., 2020). Several hundred polymers have been electrospun into nanofibers (Bhardwaj & Kundu, 2010). It has many advantages being applicable to a wide variety of materials, ease of material combination, and functionalization, relatively low start-up cost, and capable of large-scale fiber production (Shanesazzadeh et al., 2018). This method can exclude the purification process of recycled raw material prior to use, and does not require any harsh chemical or physical conditions, such as high temperature or acidity (Gaona-Sánchez et al., 2021). The fibers prepared by electrospinning are more porous owing to high specific surface area and air permeability, which are ideal for a variety of applications, including dye adsorption (Chen et al., 2020), wound healing (Rho et al., 2006), tissue engineering (Pham et al., 2006), drug delivery (Hu et al., 2014), biosensing (Sapountzi et al., 2017), and food packaging (Schmatz et al., 2019). Electrospinning technology, emerged as novel, efficient, and alternate technique in fiber production technology, has become a hot spot in the field of material sciences and food technology.

In recent years, electrospinning has gained vast interests in various food-related areas. Active food packaging with incorporated antioxidants, anti-ripening and antimicrobials have been developed using electrospinning to ensure safety and improve the shelf life of fresh and processed food products. Moreover, electrospun fibers have been successfully

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employed in the encapsulation and delivery of bioactive substances and probiotics, and immobilization of enzyme and biosensor development.

A variety of materials were utilized for producing electrospinning nanofiber. For food uses, they must be generally recognized as safe (GRAS); therefore physical, chemical, safety, and biocompatibility traits of the starting ingredients need to be considered carefully. Natural polymers are more superior than the synthetic ones because of their compatibility, low cost, tunable morphological and physical properties, and high availability as byproducts of food and agricultural processing industries (Gough et al., 2020). Food-grade biopolymers like proteins respond differently once structured into electrospun nanofibers, due to their original, chemical and structural diversity. Biopolymer fibers can be adjusted from several nanometers to several microns in diameter with its unit mass of high surface area, high porosity, tunable wetting behavior, and mechanical properties (Mendes et al., 2017). They possess both hydrophilic and hydrophobic domains, and careful attention should be paid on such characteristics to attain electrospun fibers with desirable properties. Moreover, for food packaging purpose, the preparation method, and incorporation of packaging active substances during electrospinning are also crucial to produce functional fibers.

The continuous devotion of scientists to electrospinning fibers has led to rapidly evolving knowledge on their applications in multiple fields. Coelho et al. highlighted the key parameters that influence these techniques in their recent state of art review article on electrospinning and electrospraying (Castro Coelho et al., 2021). Patiño et al. reviewed the more recent approaches based on electrospinning and related techniques for developing composite biopolymeric fiber with tailored antimicrobial properties (Patiño Vidal et al., 2021). Gough et al. summarized basic knowledge on protein and polysaccharide and their fiber fabrication methods from various ionic liquids, as well as the effect of post-treatments on these fiber materials and their applications in biomedical, pharmaceutical, environmental, and sustainable and green chemistry research (Gough et al., 2020). To capture the current progress from a different angle, this paper aims to review the characteristics of electrospun fibers prepared from natural biopolymers and their applications in food-related fields.

Overview of Electrospinning

Fundamentals

Electrospinning is a simple and non-mechanical method for generating ultrafine fibers from a wide range of materials, including polymers, composites, etc. The voltage applied to the polymer solution can lead to the charge accumulation on the solution surface, generating coulombic repulsion among the polymer droplets. Thus, the surface tension can be overcame, which can enable the ejecting of solutions into jet stream and the generating of super thin fibers. Eventually, fibrous products can be collected with a receiving board. The physical aspects of the fibers are conditioned by many factors including the polymer concentration, molecular weight, viscosity of the solution, the spinning voltage, the distance from the tip to the collector, and flow-rate (Bhardwaj & Kundu, 2010; Li & Xia, 2004).

The basic setup of an electrospinning instrument consists of three main parts: a spinneret section, a high-voltage power supply, and a metal collector. The spinneret section is composed of a feed storage (which holds the blending solution), a spinneret (a syringe containing a conductive needle), and an injection pump to ensure that the paint solution is injected at the desired flow rate. The high-voltage power supply can be either direct current or alternating current. It is connected to the metal needle so that the droplets at the tip of the needle are charged to generate coulombic repulsive force (Fig. 1) (Li et al., 2021).

Factors Affecting Electrospinning

In general, the increase in viscosity of the spinning solution is conducive to the increase in diameter of electrospun fibers. To ensure a polymer (natural or synthetic) solution capable of forming an electrospinning fiber, two conditions are required. Firstly, the viscosity of the polymer solution should not be less than 0.1 Pa, and preferably not exceed 2.0 Pa. Secondly, the polymer concentration should be at least 2 to 2.5 times of the entanglement concentration (Ce), which means that the polymer solution with intermolecular entanglement is more easier to be electrospun to obtain nanofibers. In consideration of the two empirical rules, natural and synthetic polymers are often mixed for electrospinning to make electrospinable and morphologically controllable solutions in practical trials. Taking globular proteins as examples, they are characterized with native folding structures and colloid-like behaviors in many cases. To electrospin them, synthetic polymers with sizes larger than proteins are usually incorporated (Zhang et al., 2018b).

Surfactants are often added during electrospinning, which serve to reduce the diameter of nanofibers and make them more uniform. The polymer solution forms a "Taylor cone" and the surface tension of the jet is voltage dependent, which affects the spinnability of the spinning solution. Another determinant is the polymer flow rate, i.e., the volume of solution from the syringe needle tip during the process that affects velocity of the jet. In general, the

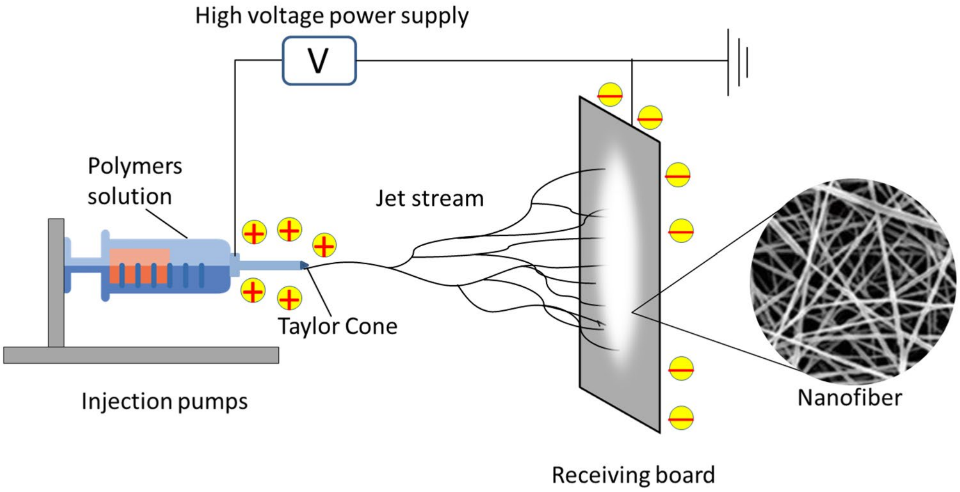


Fig. 1 Schematic diagram of electrospinning

porosity of fibers is positively correlated the flow rate. Environmental factors including humidity and temperature affect the morphological structure and diameter of fibers. Figure 2 listed the main parameters affecting electrospinning and resultant products' properties (Castro Coelho et al., 2021).

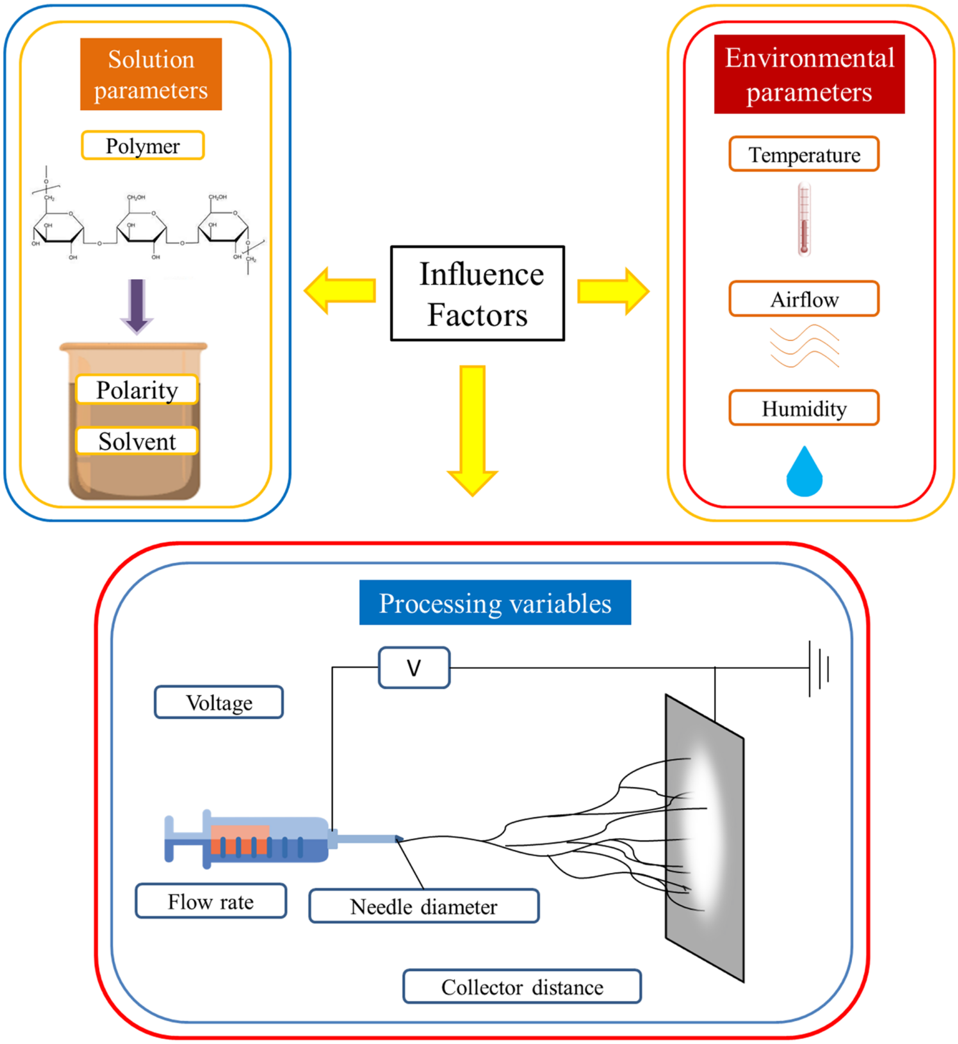


Fig. 2 Factors affecting electrospinning

Natural Biopolymers for Electrospinning

Proteins

Proteins are one of the most commonly used materials for preparing nanofibers by electrospinning technology because of their inherent properties like biodegradability, biocompatibility, and functionalities for the encapsulation and release of active compounds (Silva et al., 2018). When proteins are dissolved, the unfolding of their peptide chains and the interactions between protein molecules are altered, and then electrospinning gives rise to nanofibers (Higashiyama et al., 2021). The common proteins used to prepare fibers in electrospinning are zein, gelatin, soy protein isolate (SPI), whey protein, silk fibroin, pea protein isolate, collagen and keratin, etc. However, the production of protein-based nanofibers by electrospinning is quite challenging because of their spherical structure commonly encountered in most food proteins, the low viscosity of their aqueous solutions, and the lack of intermolecular entanglements. Therefore, proteins are often blended with other bio-based components to obtain mutually compatible hybrid polymer systems, which can then be robustly electrospun to obtain fibers (Drosou et al., 2018). These composite nanofibers often have unique functional properties (Table 1).

Zein

Zein, also known as prolamin, is a protein abundantly present in corn gluten meal, which can be obtained from corn after wet milling (Miyoshi et al., 2005). Zein is rich in sulfur-containing amino acids and hydrophobic amino acid, it can form disulfide bond and promote hydrophobic interactions, making it an attractive candidate to produce fibers via clectrospinning technique (Raeisi et al., 2021). In addition, zein is resistant to heat, oxidation and moisture which makes it favorable for the development of food packaging films produced by electrospinning and the encapsulation of bioactives substances (Prietto et al., 2018). When suitable conditions are set, the nanofibers obtained by electrospinning appear white instead of yellow and have good thermal properties and stability (Torres-Giner et al., 2008). However, zein is hydrophobic and poorly extensible, limiting its application in foods and food packaging (Federici et al., 2020). Compared to solvent cast films, electrospun fibers have more advantages (special molecular structure and high solvent removal efficiency). The fibers obtained by electrospinning of acidified zein solution have a higher glass transition temperature than the unacidified ones, in contrast to the low viscosity of the alkaline zein (Torres-Giner et al., 2008).

Altan et al. used tributyl citrate (TBC) plasticization to make zein nanofibers encapsulated with carvacrol. The nanofibers showed homogeneous round continuous fibers. It was shown that the thermal stability of the electrospun nanofibers was significantly enhanced by the loading of carvacrol. The addition of TBC increased the tensile strength of the fibers (Torres-Giner et al., 2008). Enrico Federici et al. investigated the modification of zein fibers by mixing plasticizers or co-proteins with zein using an aqueous solution of acetic acid or ethanol (with or without sodium hydroxide) as solvent. The results show that the addition of plasticizers and co-proteins has little effect on the properties of spinning solution. The addition of coproteins increases the bead formation in nanofibers, while plasticizer decreases the glass transition temperature of the fibers. Some plasticizers (lactic acid and oleic acid) reduce Young's moduli, while casein has the opposite effect (Federici et al., 2020).

Collagen

Collagen belongs to fibrous protein family and is abundant in animals, and it is the most important structural component that constitutes the extracellular matrix (e.g., connective tissue) (Drobota et al., 2020). Due to its biocompatibility, degradability and excellent biological properties, collagen is commonly used in the biomedical field. Different types of collagen, including type I, type II and III, have been electrospun into nanofibers, the networks of formed nanofibers had high surface area-to-volume ratios, tunable diameters and porosity, and offered desirable bioactivity for modulating cell function and tissue formation. Collagen electrospun fibers promote cell growth, and the structural, material and biological characteristics suggest that this material is a relatively ideal engineered scaffold (Matthews et al., 2002). However, low mechanical property of collagen is a disadvantage of its integration in biomaterial. To enhance the mechanical strength of collagen nanofibers, they are often cross-linked or blended with synthetic polymers for electrospinning (Law et al., 2017).

Andrea Fiorani et al. successfully developed electrospun fibers from type I collagen and showed that the triple helix of collagen was disrupted irrespective of solvent used in the formulation. The collagen scaffold was cross-linked by using 1-ethyl-3-(3-dimethylaminopropyl) carbodiimide and 1,4-butanediol diglycidyl ether and both cross-linking agents effectively stabilize the fibril morphology, with the latter having a superior stabilizing effect (Fiorani et al., 2014). Dems et al. showed that type I collagen could be electrospun into a self-supporting membrane without crosslinkers. And hydration of the dried electrospun collagen fibers was prevented and stabilized using pre-churning vapor. The electrospun collagen retained its original conformation, bioactivity, and ability to self-assemble into fibers (Dems et al., 2020). Zhang et al. investigated the growth and differentiation of dental pulp stem cells on nucleated ribbon collagen bundles

Table 1 Reported natural biopolymers for electrospinning and resultant fibers' characteristics

Natural biopolymer used	Electrospinning conditions	Fibers' characteristics			Functional materials engineered	References
		Lengthscale	Physical properties			
Proteins						
Zein	■Voltage: 18 kV ■Collection distance (distance between the spinneret tip and the collector): 13.5 cm ■Flow rate: 1 mL/h ■Diameter of needle: 0.8 mm ■Carvacrol	■Homogeneous round continuous fibers ■Fiber diameter between 103 ± 35 and 217 ± 47 nm	■Tensile strength of 1.70 ± 0.26 MPa ■Elongation of 5.09 ± 0.69% ■T_g remains unchanged ■The addition of TBC improves tensile strength		■Potential antioxidant packaging fiber materials with controlled morphological properties	(Altan & Çayır, 2020)
Collagen	■Voltage: 15 kV ■Flow rate: 5 μl/min ■Collection distance: 15 cm	■Fibers are smooth on both nanoscale and micron scale surfaces	■The modulus of the nanofibers is higher than that of the microfibers		■Scaffold to promote stem cell differentiation	(Zhang et al., 2018c)
Gelatin	■Voltage: 15 kV ■Flow rate: 1 mL/h ■Collection distance: 10 cm ■Nano-hydroxyapatite	■Smooth surface ■Average diameter: 516 nm	■Young's modulus: 68.6 MPa ■Tensile strength: 1.75 MPa		■Nanofibrous films of bioactive delivery or controlled release in food	(Deng et al., 2018)
Soy protein isolate	■Voltage: 18 kV ■Flow rate: 0.15 mL/h ■Collection distance: 17 cm ■Emulsion electrospinning	■Particle size < 60 μm ■Beaded fibers	■Reduce of water solubility ■Enhance of thermal stability		■Membranes of β-carotene encapsulation ■The modification of bioactive release properties	(Pinheiro Bruni et al., 2020)
Whey protein	■Voltage: 7.5 ~ 25 kV ■Flow rate: 1 ~ 3 mL/h ■Collection distance: 10 cm ■Polyethylene oxide (PEO)	■With increasing polymer concentration, droplets → beaded fibers → spindle-like rod → uniform fibers ■ "Flattened" or "ribbon like" structure ■Average diameter: 290 nm			■Encapsulating matrix	(López-Rubio & Lagaron, 2012)
Silk fibroin	■Voltage: 18 kV ■Flow rate: 0.8 mL/h ■Collection distance: 18 cm ■relative humidity 43%, 18 ℃ ■polyvinyl alcohol (PVA)	■Average diameter < 320 ■Uniform and smooth fibers		■High air permeability ■High adhesion of fibers to adjacent fibers ■Increase of rigid structure and tensile strength	■Silkworm-based micro/nanofibers	(Zhou et al., 2019)

Table 1 (continued)

Natural biopolymer used	Electrospinning conditions	Fibers' characteristics		Functional materials engineered	References
		Lengthscale	Physical properties		
Pea protein isolate	■Voltage: 15 kV ■Flow rate: 1 mL/h ■Collection distance: 15 cm ■PVA	■Average diameter: 485 ± 85 nm ■Fibers appear as nanocapsules after encapsulation of cinnamaldehyde		■Antibacterial mat	(Maftoonazad et al., 2019)
Keratin	■Voltage: 20 kV ■Flow rate: 0.6 mL/h ■Collection distance: 20 cm ■PVA、PEO	■Smooth, bead-free porous fibers ■Average diameter: 13.67 ± 2.95 nm	■T_s up to 7.54 MPa ■EAB up to 27.47 ■Significantly improved tensile properties	■Antibacterial Nanofiber	(He et al., 2020)
Carbohydrates					
Pullulan	■Voltage: 18 kV ■Flow rate: 0.4 mL/h ■Collection distance: 10 cm	■Average diameter: 294 ± 102 nm	■As the PPI content increases, Tm moves toward higher temperatures ■Increased hydrophobicity	■Potential encapsulation matrices of bioactives for functional food	(Jia et al., 2020)
Cellulose and its derivatives	■Voltage: 5–18 kV ■Collection distance: 6 ~ 13 in	■Smooth surfaces and few defects ■Average diameter: 100 nm ~ 1 µm ■Fiber morphology is influenced by the collector		■Fibers with high water retention	(Liu & Hsieh, 2002)
Chitosan	■Voltage: 18 kV ■Flow rate: 0.1 mL/h ■Collection distance: 15 cm	■Average diameter < 500 nm ■Smooth surfaces and few defects	■T_g and T_m increased ■The addition of essential oils reduces the mechanical strength	■Fiber-based antimicrobial nanomaterials in food packaging	(Hasanpour Ardekani-Zadeh & Hosseini, 2019)
Konjac glucomannan	■Flow rate: 0.5 mL/h	■Average diameter: 7.8 ± 0.2 µm ■Uniform, smooth fibers	■WVP: $5.8 \times 10^{-6} \pm 1.44$ g/(m h kPa) ■WCA: 101.0° ■Elongation at break (EB: 223.59 ± 98.14%)	■Polysaccharide micro-nano antibacterial materials	(Lin et al., 2020)

Table 1 (continued)

Natural biopolymer used	Electrospinning conditions	Fibers’ characteristics		Functional materials engineered	References
		Lengthscale	Physical properties		
Alginates	■Voltage: 15–23 kV ■Flow rate: 0.3–1 mL/h ■Collection distance: 15 cm ■Temperature (21–25 °C) and 40–50%RH	■A uniform layer spreads all over with some micron-sized fibers	■Higher ultimate tensile stress (MPa) (from 4.3 ± 2 to 15.1 ± 2 at 10% CU) ■Higher ultimate tensile stress (MPa) (from 4.3 ± 2 to 15.1 ± 2 at 10% CU) ■0.044 ± 0.003 before and after TFA	■Immobilization of hydrophilic proteins in hydrophobic polymers	(Qi et al., 2006)
Hybrid biopolymers					
Protein-polysaccharide		■Low viscosity beaded fiber, high viscosity homogeneous fiber	■Enhanced thermal stability	■Food packaging materials as a potential substitute for petroleum based plastics	(Koshy et al., 2015)
Polysaccharide-polysaccharide	■Voltage: 15–23 kV ■30–33% relative humidity and 23 °C	■Smaller fibers ranging from 87 to 57 nm in diameter ■Smooth and ultra-fine nanofiber	■Elevated thermal stability	■Water-based edible biopolymer nanofibers	(Xiao & Lim, 2018)

of electrospun poly (4-vinylpyridine) fibers by electrospinning of collagen. It was shown that deposition of collagen on the surface ensures the adhesion of dental pulp stem cells (DPSC). The fibers are smooth on both nanoscale and micron scale surfaces. However, it is important to note that in the absence of other cytokines, the supramolecular structure of ECM collagen directs DPSC differentiation (Zhang et al., 2018b, 2018c).

Gelatin

Gelatin is the product of partial hydrolysis of collagen, a large hydrophilic colloid, and is the most abundant protein in ECM. Gelatin, through the destruction of some triple-helix structures of collagen chain, is generally recognized as safe for polymer because of its excellent biodegradability, biocompatibility, and non-immunogenicity (Liu et al., 2018b). Gelatin is rarely used alone owing to its high brittleness, and thus needs to be modified by crosslinking, grafting and blending (Mohammadzadehmoghadam & Dong, 2019). Electrospinning of gelatin can be achieved in relatively mild solvents, such as mixture of acetic acid–water and ethanol-formic acid water. The main limitation of gelatin nanofibers is the poor water-resistance. Cross-linking is necessary to prevent the rapid dis-solution of gelatin nanofibers and to retain the three-dimensional structure (Deng et al., 2019). In addition, polymer blending, as an attractive method to obtain better properties than individual components, has been widely used to incorporate rigid or hydrophobic polymers to prepare high-quality composite gelatin films (Liu et al., 2018b).

Deng et al. reported the surface wettability, mechanical strength and water barrier property of the electrospun gelatin. The average diameter of homogeneous and smooth nanofibers is around 500 nm (Deng et al., 2020). The prepared gelatin, and gelatin/zein film by electrospinning showed well dispersed zein particles in the gelatin network with better nanofiber stability in solvent such as water and ethanol. The zein/gelatin nanofibers prepared after cross-linking Merad reaction have good adjustable wettability, mechanical properties, and biocompatibility. Well-dispersed zein particles in the gelatin network improved dissolution of gelatin in solvent by maintaining the nanofiber structure after being immersed in water or ethanol (Deng et al., 2018).

Soy Protein Isolate

As a renewable resource with good biocompatibility and processability, soy protein isolate (SPI) has great application potential in food industry, agriculture, bioscience and biotechnology (Koshy et al., 2015). The polyelectrolyte structure in SPI hinders the formation of nanofibers by electrospinning. To solve this problem, other polymers (e.g., polyethylene oxide) can be added to the solution to facilitate the formation of fibers (Silva et al., 2018).

Pinheiro Bruni et al. used emulsion electrospinning to encapsulate the antioxidant β-carotene in a mixture of SPI and polyvinyl alcohol (PVA). In this study, the encapsulation of β-carotene reached 65.0% ± 2.6%. Fat food simulation experiments were conducted to demonstrate that the returned electrospinning coating prepared by emulsion electrospinning is an ideal food packaging material for antioxidant hydrophobicity (Pinheiro Bruni et al., 2020).

Whey Protein

As an important by-product of the dairy processing industry, whey proteins are advantageous in gelling and emulsification and they have been widely used in the food industry. López Rubio et al. studied the feasibility of the electrospinning technique for the development of protein-based encapsulation structures avoiding organic solvents and/or high temperatures using whey protein isolate (WPI) (López-Rubio & Lagaron, 2012). WPI allows the preparation of membranes with properties such as degradability, transparency, and odorlessness, which can be used to extend the shelf life of food products. Whey proteins are considered as a possible carrier to deliver bioactive materials and pharmaceutical compounds (Zhong et al., 2018). The preparation of nanofibers by electrospinning WPI is of research interest due to their unique advantages.

Rye flour, whey protein concentrate and other polymers were combined to prepare electrospun nanofibers that encapsulated grape seed extract. The fiber diameter obtained by microwave heating solution of 6% rye flour was larger than that of conventional heating solution. The incorporation of grape seed extract produced strong interactions in the polymer matrix and increased the thermal stability of the films. Although grape seed extract was less stable at high pH, the antioxidant activity of the grape seed extract samples containing both 4% and 6% rye flour exceeded 40% (Zhong et al., 2018). López-Rubio et al. obtained micron, submicron and nanocapsules by electrospinning of aqueous whey protein solutions at different pH conditions, illustrating the potential of electrospinning technology in the development of whey protein-based encapsulation materials. The prepared capsules were able to stabilize the functional additives (López-Rubio & Lagaron, 2012).

Silk Fibroin

Silk fibroin (SF) is a kind of natural protein derived from silkworm cocoon. Since it is a mechanical robust polymeric biomaterial with high stability and strong mechanical properties, SF has a wide range of applications in tissue engineering and regenerative medicine (Sahi et al., 2020). SF has strong intermolecular hydrogen bonds, especially between secondary structures, thus SF can easily structure into fiber by electrospinning. Although SF is difficult to consume, it has potential for a wide range of applications in food packaging. SF has been made into films by solvent casting and electrospinning to obtain colorless, transparent and water-soluble films (Ma et al., 2017).

Instead of extracting proteins from silkworm silk, as is often the case, Zhou et al. extracted proteins from live silkworms and prepared protein solutions. The protein solution and the polyvinyl alcohol solution were mixed in different weight ratios to produce silkworm protein fibers with unique properties like high mechanical strength and good wetting property (Zhou et al., 2019). Drug carriers are used to deliver drugs to a designated location to achieve a lower systemic dose and thus reduce side effects. Gang Tao et al. synthesized AgNPs-silk/PVA films using electrospinning, and in this experiment, AgNPs were homogeneously synthesized on the surface or embedded inside the silk/PVA films. It was shown that the prepared AgNPs-silk/PVA films exhibited excellent mechanical properties and stability, and broad spectrum bacteriostasis to gram-negative bacteria and gram-positive bacteria (Tao et al., 2017).

Pea Protein Isolate

As a by-product produced in pea starch processing, pea protein isolate (PPI) has excellent amino acid composition along with a good content of lysine, owing to its desired functional traits like dispersion forming, stability, water solubility, and gel formation as well as high value-added protein resources (Jia et al., 2020). The globular nature of PPI and probably the potential lack of molecular entanglement make it challenging to produce nanofibers. The addition of a spinnable polymer and other proteins such as egg or soy proteins were studied to overcome these issues. PPI nanofibers can be envisaged as nanocapsule matrix to improve nutritional value of food products or act as a texture modifier (Maftoonazad et al., 2019). PPI has been electrostatically spun into nanomaterials for drug delivery applications, tissue scaffolds and antimicrobial mats (Akhmetova & Heinz, 2021; Pereira et al., 2021).

Oguz et al. prepared a new nanofiber by using pea flour and hydroxypropyl methyl cellulose (HPMC) for electrospinning. At the same time, the effects of pH, pea powder, and HPMC concentration on apparent viscosity and electrical conductivity of spinning solution and on characteristics of nanofiber are investigated. It has been shown that pH is an important factor in obtaining beaded nanofibers and alkaline conditions were favorable for the nanofibers formation. At solutions below a certain concentration, insufficient entanglement of the chains causes instability of the jet, which shrinks driven by surface tension to obtain string bead-like fibers. Besides, pH is also an important factor in obtaining beaded nanofibers. For PPI, the increase of pH from neutral to basic has a positive effect on the morphological structure of nanofibers. The increase of pH and pea flour concentration considerably increases the apparent viscosity of the solution, leading to an increase in diameter. Experimentally, they obtained the suggested solution parameters, containing 0.5% or 0.25% HPMC concentration in a 1% pea flour solution at pH 10, to obtain homogeneous, pea flour-based nanofibers with minimal diameter (Fig. 3) (Oguz et al., 2018). Maftoonazad et al. mixed PPI into polyvinyl alcohol (PVA) and obtained uniform and smooth nanofibers by electrospinning. The antibacterial mat can been further made by adding cinnamaldehyde into these electrospun fibers (Maftoonazad et al., 2019).

Keratin

Keratin is extremely abundant in nature and has aroused great interests in tissue engineering because of its biocompatibility, biodegradability, and biofunctionality. However, it is seldom used in the field of food. Keratin is typically obtained from mammalian nails, horns, and hair (feathers, wool, etc.) and has been extensively studied in tissue scaffold materials, which were shown to promote the growth and proliferation of cells (Su et al., 2021). Since keratin contains higher contents of cysteine, the content of disulfide bonds formed is particularly high; therefore, keratin-based fibers usually has higher mechanical strength.

In recent years, being a byproduct of the poultry industry and animal husbandry keratin is more preferable in electrospinning due to its sustainability and accessibility. The films prepared from wool keratin and citric acid have good transparency and exhibit antibacterial properties in carrot storage experiments, indicating the potential of wool keratin to be used in food-active packaging (Ramirez et al., 2017). Since feather keratin biodegradation produces a highly interconnected porous network, He et al. (Ramirez et al., 2017) prepared antimicrobial feather keratin-based nanofibers by incorporating silver nanoparticles (AgNPs) (He et al., 2020).

Carbohydrates

Carbohydrates are of promising polymers in producing films, which are often edible and biodegradable, with great potential in food application. Among the most used are chitosan, starch, alginate, cellulose, and pectin because they are cheap, biodegradable, and can form gels. In addition, they have excellent mechanical properties and are effective barriers to low polar compounds. At the same time, they have strong resistance to gases such as oxygen and carbon

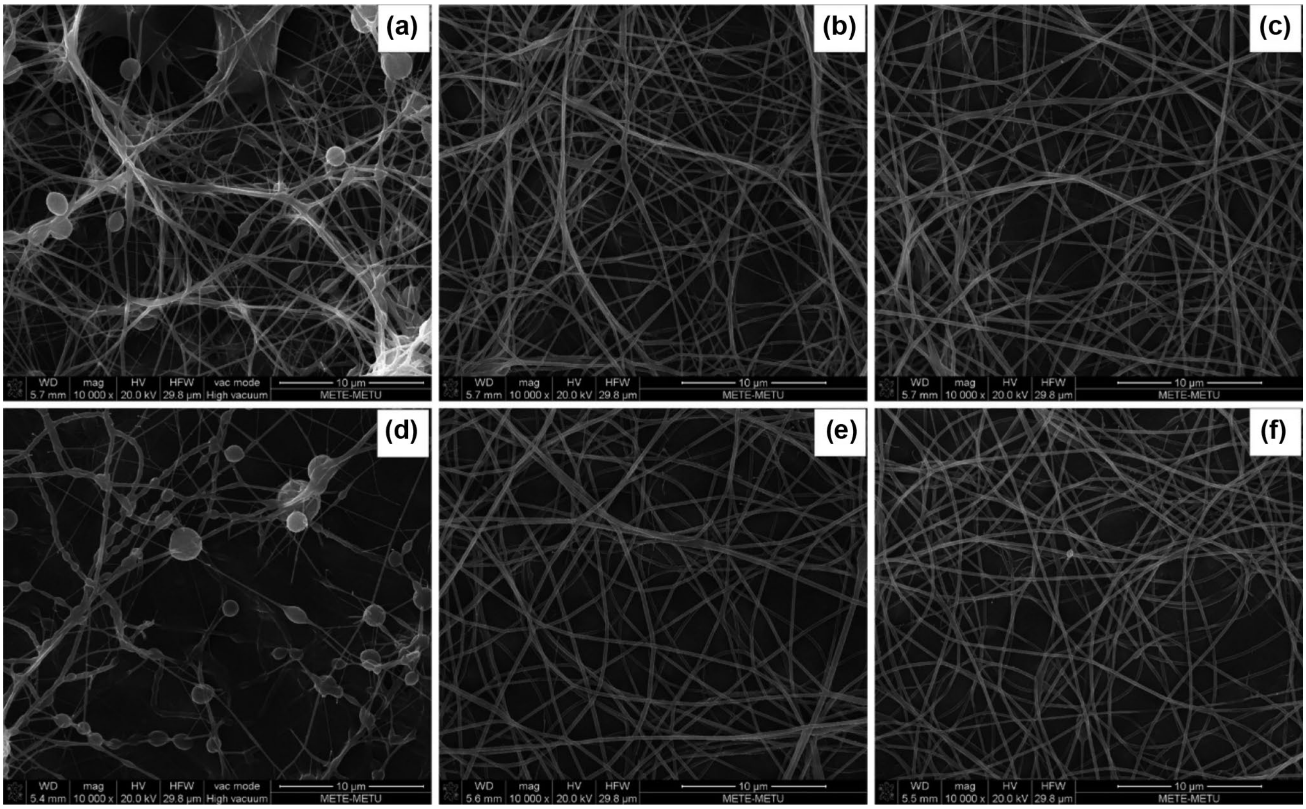


Fig. 3 SEM images (10,000×) of the nanofibers, obtained from solution with 1% pea flour at **a** pH 7, **b** pH 10, **c** pH 12; solution with 2% pea flour at **d** pH 7, **e** pH 10, **f** pH 12 (Oguz et al., 2018). The permission to reproduce it has been obtained

dioxide. However, water vapor issue is often not well prevented, which needs to be further tackled. (Gaona-Sánchez et al., 2021).

Pullulan

Pullulan (PUL) is a microbial and linear polysaccharide, consisting mostly of repeating a-(1 → 6)-linked maltotriose units. With its non-toxic, odorless, tasteless, and edible properties, pullulan has been applied in various fields such as tissue engineering, drug delivery, and food industry (Yang et al., 2020). In addition, as an edible polysaccharide with excellent film-forming properties, pullulan has been used in various ways to prepare food-grade films. The film can block the penetration of oil and oxygen (Drosou et al., 2018). Owing to its unique structural linkage, PUL demonstrated special physical properties, such as the ability to attach to other substances and form very fine fibers by electrospinning. However, the poor water-resistance and thermostability of PUL nanofiber film have significantly limited the applications in food packaging and encapsulation of thermosensitive bioactive substances. In research and applications, other biopolymers are often blended with pullulan to address these issues.

PUL can alter the solution properties for electrospinning through hydrogen bonding with proteins and hence increase the spinnability of globular proteins that are not easily electrospun. Several studies have been focused on PUL as a biopolymer to facilitate the electrospinning process of proteins, resulting in the preparation of nanofibrous membranes (Jia et al., 2020). Aceituno-Medina et al. electrospun microfibers by mixing protein isolate (API)-PUL complexes in formic acid. It was shown that the addition of surfactants can modulate the electrospinning solution properties and thus affect the electrospun fibers (Aceituno-Medina et al., 2013).

Cellulose and its Derivatives

Cellulose is widely and abundantly found in nature, and its strong intra- and intermolecular hydrogen bonds and van der Waals forces make it insoluble in organic solvents. Due to its mechanical properties, it can be used as a support material. The amorphous region of cellulose can be dissolved away by controlled acid hydrolysis, while its water-insoluble highly crystalline region can be converted into a stable suspension by mechanical shearing (Deng et al., 2018). Cellulose forms highly ordered structures in solution, and electrospun fibers are formed by dissolving to destroy crystalline regions and reduce intermolecular interactions in the dissolved state (Xiao & Lim, 2018). Cellulose derivatives, including cellulose acetate, hydroxypropyl cellulose, and hydroxypropyl methyl cellulose, which can be dissolved in volatile solvents for electrospinning, have been extensively studied. In the food sector, cellulose and its derivatives are often mixed with other substances for spinning, and spun films loaded with active substances are often used as carriers for active packaging.

Cellulose (CA) acetate is a cellulose acetate produced from cellulose through an acetylation process. It is a commonly used semi-permeable membrane material for dialysis, ultrafiltration and reverse osmosis operations CA membranes are thermally stable with high fluxes and high flow rates. Liu et al. mixed dimethylacetamide with acetone or acetic acid in a certain volume ratio and found that the resulting solvent system allowed continuous electrospinning of cellulose acetate in the concentration range of 12.5 – 20%, with a positive correlation between fiber size and CA concentration. The obtained cellulose films had similar hydrophilic properties as the commercial cellulose fiber mechanism, but the water absorption was almost ten times higher (Liu & Hsieh, 2002). Li et al. successfully prepared cellulose acetate/poly(ethylene oxide) bioactive fiber membranes loaded with rutin using electrospinning. Due to the high compatibility between the polymers, the encapsulation rate of rutin can be more than 90% and the fibrous membrane displayed antibacterial and antioxidant activity (Li & Yang, 2020).

Hydroxypropyl methyl cellulose (HPMC) is a kind of asviscolizing agent employed for a wide variety of pharmaceutical and food preparations with thickening ability, and its methoxy content has a large impact on physical properties. One study encapsulated gallic acid in hydroxypropyl methylcellulose (HPMC)/polyethylene oxide (PEO) by electrospinning, and the nanofibers showed homogeneous morphology and strong antioxidant properties. It was shown that gallic acid was successfully embedded into the nanofibers. When the gallic acid-loaded nanofibers packed walnuts, it reduced the oxidation of walnuts during storage, thus the nanofibers have the potential for antioxidant active packaging (Aydogdu et al., 2019).

Cellulose nanocrystals (CNCs) are a kind of nanoscale cellulose extracted from natural fibers, which not only has the characteristics of having nanoparticles, but also has co-biodegradability and excellent mechanical properties. Zhang et al. fabricated a novel low-pressure drop poly(vinyl alcohol) (PVA)/cellulose nanocrystals (CNCs) fiber filter by electrospinning. Among them, CNCs have both mechanical enhancement and fiber diameter reduction (increasing the surface charge density of the spinning solution), and the small diameter of the fibers improves the removal efficiency of particulate matter. Considering that PVA and CNCs are non-toxic and biodegradable, this fiber material is promising for air purification and other applications (Zhang et al., 2019).

Chitosan

Chitosan (CS) is composed of 2-amino-deoxy-β-D-glucan, a linear polysaccharide linked by α (1 → 4) glycosidic bond, and has antioxidant and antibacterial properties. Although CS is poorly soluble in water, it is readily soluble in dilute acid solutions (Souza et al., 2021). CS is another widely used biopolymer in electrospinning because of its low toxicity, good compatibility, and degradation, and it can play the role of stabilizer and reducing agent (Li et al., 2020b). Oregano essential oil (OEO), which is sensitive to heat and oxygen, was embedded in an electrospun CS/poly-ε-caprolactone (PCL) antimicrobial fiber mat. The results of the study showed that the CS/OEO/PCL mat had good antimicrobial efficiency with controlled release of essential oil (Hasanpour Ardekani-Zadeh & Hosseini, 2019).

Konjac Glucomannan

Glucomannan and glucose monomers are linked by β-1,4-glycosidic bonds to obtain konjac glucomannan (KGM). KGM is readily soluble in water and is an ideal material for the preparation of nanofibers. There are various health effects associated with the consumption of KGM and related products. KGM has good water binding and gelation capacity as well as film forming property. There has been increasing interest in using KGM for the formulation of functional foods (Zhu, 2018). However, the KGM nanofiber rapidly breaks down upon contacting with water, limiting its use in food packaging. Therefore, modification of the KGM molecule by introducing other components is alternative way to maximize the use of this polysaccharide. Wang and his colleagues developed a facial method to fabricate nanofiber films from biodegradable KGM and corn gluten by electrospinning. Corn gluten was dispersed into KGM solution by electrospinning method to form stable and uniform nanofiber film (Wang et al., 2019). Food packaging films consisting of KGM, PCL, and AgNPs showed excellent antibacterial activity against *Staphylococcus aureus* and *Escherichia coli* due to the good swelling of KGM in the films (Lin et al., 2020).

Alginates

Alginate is a commonly used natural bio-based material. Algal based materials tend to be elastic and durable, and the performance of gels is closely related to the ratio of lactose acid to glutamine (Souza et al., 2021). Fabrication of sodium alginate (SA) nanofibers from its aqueous solutions by electrospinning is still a challenge, because of its rigid chain conformation and lack of chain entanglements. Research shows that Ca^{2+} cations enhanced the intermolecular interactions of SA solutions and improved the electrospinnability of SA solutions (Fang et al., 2011). Gutierrez-Gonzalez et al. used trifluoroacetic acid cross-linking and electrospinning to fabricate a sodium alginate and PEO composite material coated with curcumin. The ultimate tensile stress of the material was elevated and mechanical properties were obtained (Gutierrez-Gonzalez et al., 2020).

Hybrid Biopolymers

As mentioned above, some natural polymers (e.g., globulin) are unable to form nanofibers by electrospinning. The electrospinning properties of natural polymers are closely related to their molecular structure. A new approach is to spin polymers with different electrospinning properties together. Therefore, researchers have combined different biopolymers for electrospinning, including protein–protein, polysaccharide-polysaccharide, and protein-polysaccharide.

There have been many studies on electrospinning of hybrid biopolymers, the most popular of which is protein – polysaccharide electrospinning. SPI solutions have high viscosity and low conductivity, while gelatin solutions have low viscosity and high conductivity. For the above reasons, Francine et al. produced microfibers by electrospinning using a polymer blend of soy protein isolate (SPI), PEO and zein wrapped with ginger essential oil at a 1:1:1 (v/v/v) ratio. It was found that the resulting microfibers exhibited a uniform shape which is not affected by the concentration of the essential oil (Silva et al., 2018). Raeisi et al. encapsulated essential oils in natural polymeric nanofibers in order to prevent their oxidation The pure SPI solution was not spinnable due to the spherical structure of the molecules, and the addition of gelatin up to 30% resulted in uniform, smooth, bead-free nanofibers observed by SEM. Evaluation of the performance of electrospun fibers with 30% gelatin neutralized the charge of SPI and significantly increased the diameter of nanofibers by 314.08 nm. Subsequently, constituents of *Zataria multiflora* and *Cinnamon zeylanicum* essential oils were embedded into the electrospun SPI/Gelatin nanofiber network. The nanofibers obtained by electrospinning exhibited broad-spectrum antibacterial activity (Raeisi et al., 2021). Wen Jia et al. fabricated and characterized the nanofiber films of food-grade PPI and carbohydrate pullulan by electrospinning (Jia et al., 2020). The solubility and rigidity of zein are addressed through adjusting operation parameter and incorporation of plasticizer/biopolymer into feed solution. In one study, zein was made into nanofibers by needle-free electrospinning using a rotating spinneret. With increasing of zein concentration and decreasing applied voltage, the diameters of the fibers raised (Karim et al., 2020). Drosou et al. developed a new electrospun composite amylopection-whey protein isolate fiber. It was shown that the addition of branched starch to the WPI solution improved their electrospinning properties and the thermal stability of the fibers was enhanced compared to pure WPI. It has potential effects such as encapsulation of bioactive substances (Drosou et al., 2018).

Binary polysaccharides have also been employed to produce electrospun nanofibers. Xiao et al. prepared PUL-alginate and PUL-alginate-$CaCl_2$ co-blended fibers by surface electrospinning, and investigated the effects of different concentrations of prulan addition and branched chain starch on the fibers. The prepared smooth and ultrafine nanofibers have the potential to be used as active food packaging materials (Xiao & Lim, 2018).

Food Application

Food Active Packaging

According to World Food Organization (WFO), tens of billions of economic losses occur globally every year due to food deterioration. How to reduce the addition of preservatives and extend the shelf life of food without affecting the quality of food is a new challenge facing the food industry. To ensure food safety, a critical element is the advancement and development of food packaging technology. Most of the food contamination and physical harm occur during transportation (Patiño Vidal et al., 2021). Thus, food packaging plays an integral role in extending the shelf-life of a product without compromising its quality. Unlike traditional packaging that only serves as a seal and barrier, active packaging materials often have multiple features. The addition and release of active agents is a key aspect of active packaging. However, active agents have defects such as thermal instability and loss of activity after storage, and these problems have to be solved in order to make active packaging materials with applications. Therefore, the preparation of packaging structures that can protect active ingredients are of research value. Different from the common technologies for preparing packaging films such as solvent casting, extrusion and thermoforming, electrospinning can prepare continuous polymer fibers with different morphology and structure. The fibers are deposited on the collector and form a fiber film. Electrospinning technology applied to active food packaging has been widely studied and the prepared nanofibers are often functionalized by loading with diverse functional compounds including antimicrobial agents, antioxidant agents, oxygen scavengers, carbon dioxide emitters, and ethylene scavengers. To develop the technique of electrospinning of active food packaging, electrospun nanofibers have been covalently or non-covalently functionalized for loading diverse bioactive compounds including antimicrobial agents, antioxidant agents, oxygen scavengers, carbon dioxide emitters, and ethylene scavengers (Zhang et al., 2020). Qi et al. try to prepare composite fibers from W/O and O/W emulsions, SEM observation reveals the bead-like structures in the fibers, demonstrating that microencapsulation has been effectively carried out (Qi et al., 2006). Among the published articles, there are more studies on the application of natural polymers in the food sector, including active packaging (Table 2).

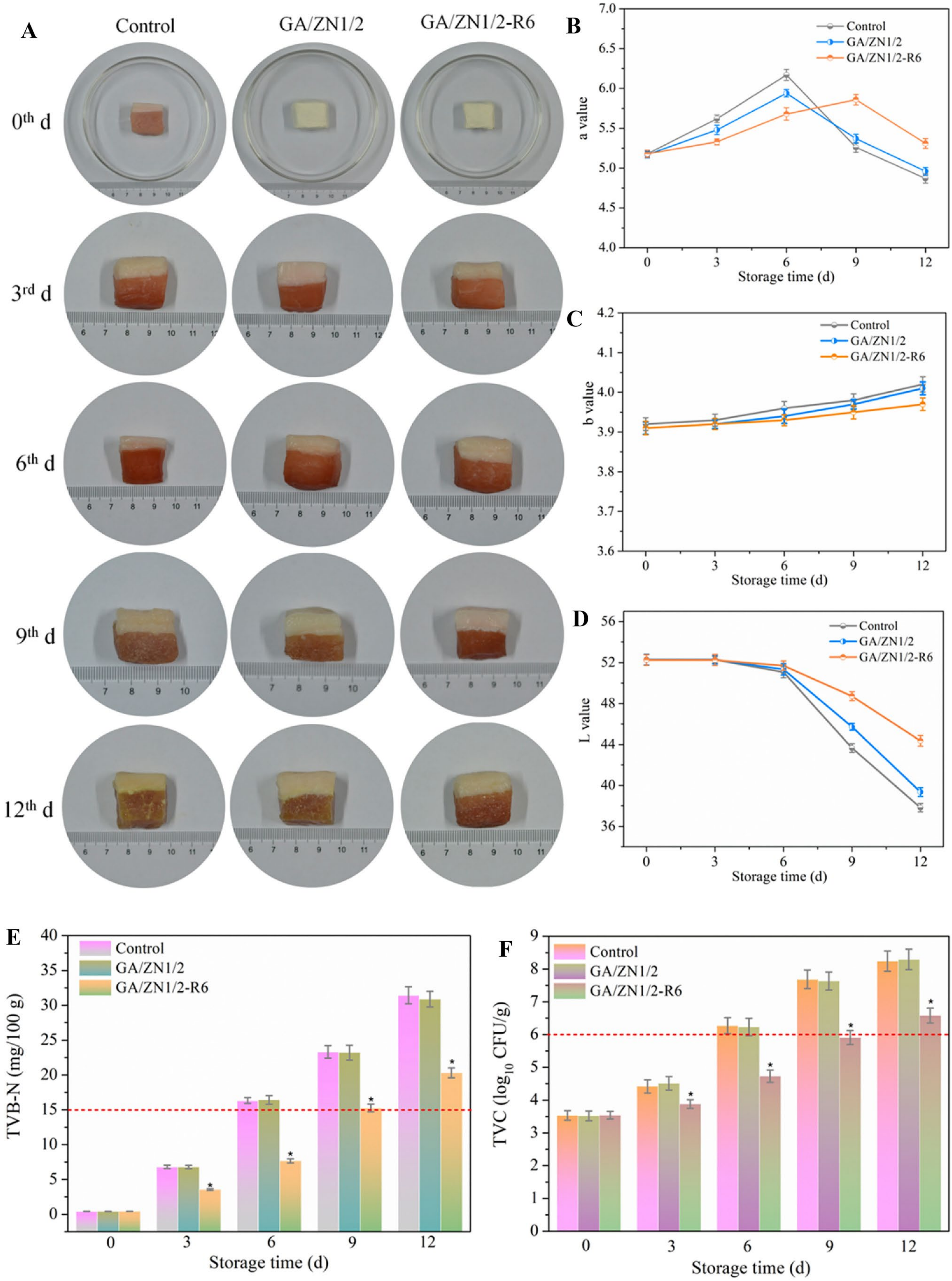
Antibacterial and Antioxidant Materials

Some natural compounds, such as natural phenolic compounds have antioxidant and antimicrobial capabilities. They also have problems such as instability, which limits the application in food packaging (Zhong et al., 2018). For example, eugenol is a natural antibacterial agent with broad-spectrum antibacterial activity and low biological toxicity. Adding it to food packaging can improve the antioxidant, antibacterial activity and hydrophobicity of the packaging. However, eugenol is unstable and has a distinct smell, it is difficult to apply directly. The use of electrospinning technology to embed eugenol in nanofibers can reduce the degradation while retaining its antibacterial and antioxidant effects. Grape seed extract, tannic acid, apple polyphenols, lycopene resveratrol, and other substances have been successfully encapsulated in natural polymer nanofibers (Hajikhani et al., 2020; López de Dicastillo et al., 2019; Monfared et al., 2019). These composite nanofibers packaging showed remarkable inhibitory action against common food-borne microorganisms-*Escherichia coli* and *Staphylococcus aureus*.

Many scholars are looking for new types of nanofiber active packaging materials, and explored their properties and applications. Aslaner et al. used grape seed extract (a mixture of polyphenols such as catechin, epicatechin, and gallic acid) as an antioxidant wrapped in rye flour and whey protein-based nanofibers. Nanofibers have been studied as reliable materials for the fabrication of bio-based films with bioactivity and high stability. However, nanofibers encapsulated with active substances tend to have poor mechanical properties. Therefore, the incorporation of other polymers (natural or synthetic polymers) with proteins is a way of practical to prepare packaging materials with biological activity (such as antioxidant and antibacterial properties) (Zhong et al., 2018). In the study of Zhan et al., tannic acid (TA)/Zein membranes were prepared by the electrospinning technique and then AgNPs were incorporated into electrospun membranes via in situ reductions. TA/Zein and AgNPs/TA/Zein electrospun membranes showed excellent antibacterial and antioxidant activity. Moreover, it was found that the AgNPs/TA/Zein electrospun nanofibers with different TA concentrations had certain catalytic reduction (Zhan et al., 2020). Li et al. encapsulated resveratrol (R) in electrospun

Table 2 Application of electrospinning of biomolecules in published literature

Applications	Fiber details	References
Food active packaging		
Antibacterial and antioxidant materials	■Grape seed extract was wrapped in rye flour and whey protein-based nanofibers	(Zhong et al., 2018)
	■AgNPs were incorporated into TA/zein fibers	(Zhan et al., 2020)
	■Encapsulated resveratrol (R) in electrospun gelatin/zein (GA/ZN) fibers to develop antimicrobial mats that can extend the shelf life of pork	(Li et al., 2020a)
Ethylene absorbent material	■TiO_2 was encapsulated in zein	(Böhmer-Maas et al., 2020)
Food encapsulation		
Bioactive ingredients encapsulation	■CA/PEO-loaded rutin fibrous membrane. The release rate was influenced by the concentration and the pH and temperature of the PBS buffer	(Li & Yang, 2020)
	■β-carotene was encapsulated in zein	(Fernandez et al., 2009)
Probiotic encapsulation	■The viability of probiotics can be improved through encapsulation within coaxial electrospun PLA nanofibers	(Yu et al., 2020)
Enzyme/protein immobilization		
Enzyme immobilization	■α-amylase was immobilized in ultrafine PVA fibers	(Porto et al., 2019)
	■Lipase was encapsulated in poly (glycidyl methacrylate-co-methylacrylate)/feather polypeptide nanofiber	(Liu et al., 2018a)
Biosensing in food testing		
Surface enhanced Raman	■The zein-based sensor platform was formed from zein nanofibers using electrospinning	(Turasan et al., 2019)


A
Control
GA/ZN1/2
GA/ZN1/2-R6
0th d
3rd d
6th d
9th d
12th d
B
a value
Storage time (d)
Control
GA/ZN1/2
GA/ZN1/2-R6
C
b value
Storage time (d)
D
L value
Storage time (d)
E
TVB-N (mg/100 g)
Storage time (d)
F
TVC ($\log_{10}$ CFU/g)
Storage time (d)
*

◂**Fig. 4** Appearance changes (**A**) of pork during storage at 4 °C, corresponding variations in color parameters (**B**–**D**), and TVB-N (**E**) and TVC (**F**) levels. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.) (Li et al., 2020a)

gelatin/zein (GA/ZN) fibers to develop antimicrobial mats that can extend the shelf life of pork. The morphology and thermal stability of the fibers prepared by electrospinning were related to the GA/ZA ratio. The presence of resveratrol ensured the antimicrobial activity and antioxidant activity of the mat, but the fibers became inhomogeneous when the content of R reached 9%. Storage experiments observed and examined changes in the appearance of the pork (Fig. 4), and the results showed that the fiber mats extended the shelf life of the pork by 3 days at 4 °C (Li et al., 2020a).

(Control: the naked one; GA/ZN1/2: GA and ZN (1/2, w/w) electrospinning nanofibers; GA/ZN1/2-R6: resveratrol(R, 6%, w/w, based on the weight of GA/ZN) was added to solution of GA/ZN1/2 to electrospinning). The permission to reproduce it has been obtained.

Ethylene Absorbent Material

During the transportation and storage of fruits and vegetables, ethylene promotes the ripening and aging of fresh vegetables and fruits. Therefore, controlling the content of ethylene is important to extend shelf life of fruits and vegetables. TiO_2 can be used as a catalyst for photocatalytic degradation of ethylene. However, it should be noted that ethylene must be in contact with the surface of TiO_2 in order to function. Therefore, encapsulating TiO_2 in nanofibers with high porosity is an innovation active packaging of fruits and vegetables. Böhmer-Maas et al. worked on zein-TiO_2 nanofibers produced through electrospinning and evaluated fibers as ethylene absorbers to improve the storage of cherry tomatoes (Böhmer-Maas et al., 2020). They found that the nanofibers demonstrated photocatalytic activity against ethylene during the storage because of their high surface area, which optimizes the ethylene photocatalysis, reducing the concentration of ethylene (Gaona-Sánchez et al., 2021).

Food Encapsulations

Bioactive Ingredients Encapsulations

In the context of rapid development of nanotechnology, encapsulation of active substances and their controlled release is an important research direction. On the one hand, encapsulation of active substances into the matrix can protect them from losing their biological activity under the influence of external environment or in vivo environment (e.g., gastric acid, action of internal enzymes). Besides, encapsulation would aid in the controlled release of the active substance (Rostamabadi et al., 2020).

Fibers prepared by electrostatic spinning at the micron or nanoscale have structural similarities to the extracellular matrix and their physicochemical properties were worth of investigation. This type of structure has the potential to achieve a slow release of encapsulated active small molecules rather than an explosive release. Based on this, electrostatic spinning has a promising future in the food industry and pharmaceutical industry (Zhang et al., 2018a). The CA/PEO-loaded rutin fibrous membrane prepared by Li et al. is mentioned above. As a bioactive substance, it is extremely important to study the release rate of rutin in fibrous membranes. The experimental results showed that the release rate was influenced by the concentration and the pH and temperature of the PBS buffer. After kinetic modeling, the release of rutin differs under different conditions, for example, at 37 °C in the buffer, rutin diffuses mainly through swollen membranes and water-filled pores, while at 20 °C, pH 5.6, the release of rutin is due to the swelling and relaxation of the membrane (Li & Yang, 2020). β-carotene is widely used in the food industry as a colorant and antioxidant. Fernandez et al. encapsulated it into zein by electrospinning. β-carotene was stably and widely distributed in microfibers, and its photostability increased significantly under UV–visible light irradiation (Fernandez et al., 2009).

Probiotic Encapsulation

In addition to natural active substances, electrospinning technology is also used to encapsulate active probiotics. Probiotics are a group of microorganisms that have beneficial effects on human health. They are mainly located in the digestive system and can perform a number of functions, such as regulating gastrointestinal motility and maintaining the balance of intestinal microflora. In addition to this, they are able to modulate the immune system, influence intestinal inflammation and modulate potential risk factors for cancer. Probiotics have important functional properties for basic nutritional needs and have beneficial effects on the recovery of some diseases (George Kerry et al., 2018). However, most probiotics have difficulty in surviving the harsh environments as the activity of probiotics is very sensitive to many environmental factors including temperature, oxygen, and mechanical forces. In humans digestive tract, the presence of gastric acid and bile salts can significantly reduce the number of viable beneficial microorganisms (Yu et al., 2020). In order to provide an adequate dose of live probiotics, a suitable delivery system is required. The delivery system needs to be edible, and can offer effective local delivery and ensure the long-term stability of the probiotic under normal conditions (Škrlec et al., 2019). In recent years, electrospinning has become a new delivery technology for probiotics.

Protein and polysaccharide are commonly used in probiotic encapsulation. Whey protein concentrate, zein, and other proteins have been successfully used in the encapsulation of probiotics. However, electrospinning or electrospray of proteins for the encapsulation of probiotics is challenging. The polyelectrolyte nature of protein needed to be considered as it reduces the stability of the electrospinning processing. In turn, the different electrospinning parameters require the adjustment to influence the viability of the probiotics. Desirable protein chain entanglements for electrohydrodynamic processing can be reached by using organic solvents (e.g., hexafluoro-2-propanol or trifluoroethanol), by changing the pH or temperature, or by adding denaturing agents capable of modifying the protein structure/aggregation and intra/inter-molecular disulfide bonds. One classical approach is to mix proteins with polysaccharides as carriers (which may require the addition of a synthetic polymer such as PEO), along with a surfactant to reduce surface tension, and then subject the blended solution to coaxial electrospinning. The core–shell structures can be thus prepared for the encapsulation of probiotics by electrohydrodynamic (Mendes & Chronakis, 2021). A new encapsulation platform for delivering bioactive substances, namely, polylactic acid (PLA) nanofibers were prepared by coaxial electrospinning. It was verified that the viability of probiotics was effectively improved when encapsulated in these nanofibers (Yu et al., 2020).

Enzyme Immobilization

Enzymes are biocatalysts produced by biological cells and mainly composed of proteins. Compared with general catalysts, the enzyme has the characteristics of high efficiency, specificity, and eco-friendly processing. Enzymes are widely used in the food industry, mainly including starch processing, dairy processing, wine brewing, meat processing, baking, food preservation, etc. However, the activity of enzyme is affected by environmental factors (pH, temperature, etc.). Thus, improving the stability of enzymes is very important. Electrospinning nanofibers act as promising carriers improving the stability, efficiency, and storage of the enzymes and, at the same time, avoiding their degradation and off-flavors. The surface of fibers can be functionalized in order to control the release of enzymes (Castro Coelho et al., 2021). The enzyme can be immobilized on the surface or inside of the fiber. The arrangement and type of carriers have a significant effect on immobilized enzymes, and one-dimensional fiber material is one of the ideal carriers for enzyme immobilization (Rather et al., 2022).

Porto et al. immobilized α-amylase in ultrafine PVA fibers by electrospinning and evaluated its stability at different temperatures and pH using various starch substrates such as corn starch and germinated and ungerminated wheat starches. Immobilized enzymes were more active and had higher storage stability compared to free enzymes. For instance, the immobilization of α-amylase provides stability over a wide range of temperatures and pH (Figs. 5 and 6) (Porto et al., 2019). A novel poly (glycidyl methacrylate-co-methylacrylate)/feather polypeptide (P(GMA-coMA)/

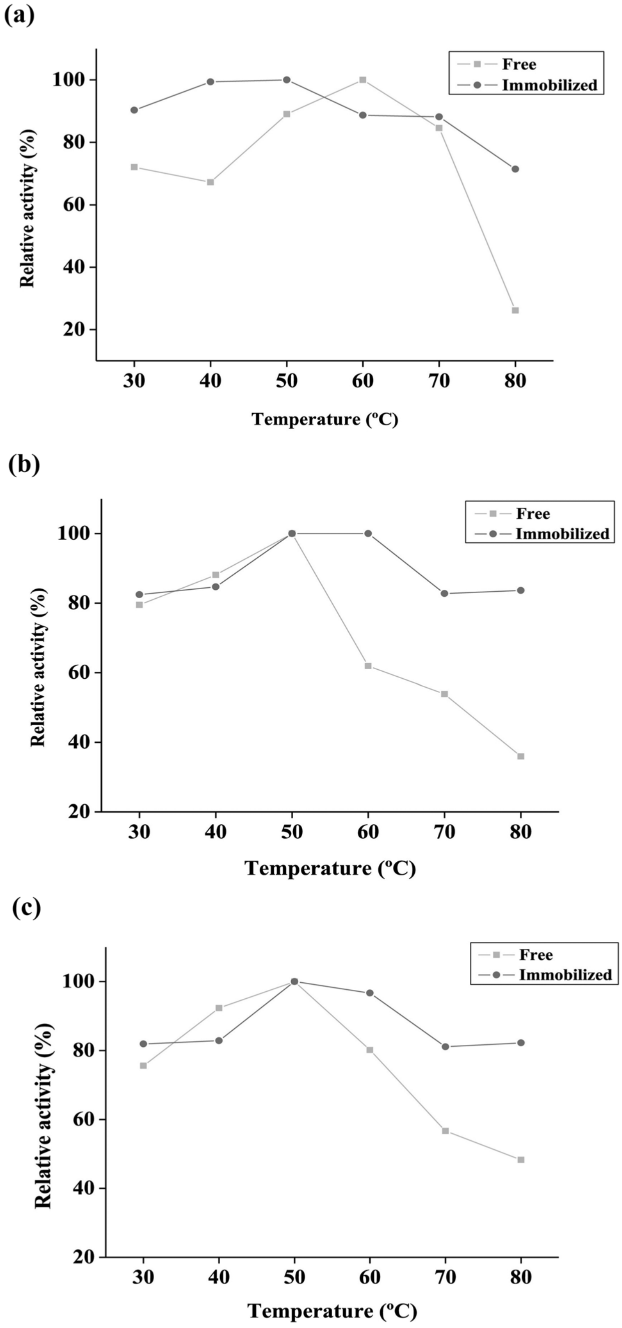


Fig. 5 Relative enzymatic activity of free and immobilized α-amylase at different pH values, different temperatures and with different starch substrates: **a** corn starch, **b** germinated wheat starch, and **c** ungerminated wheat starch (Porto et al., 2019). The permission to reproduce it has been obtained

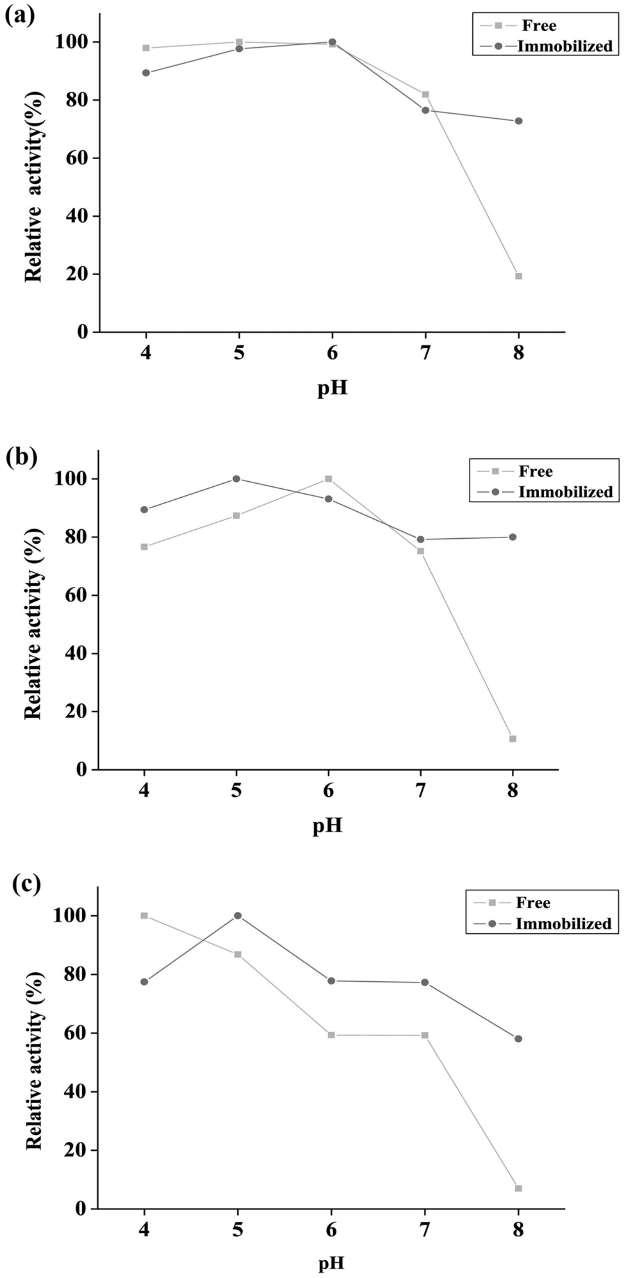


Fig. 6 Relative enzymatic activity of free and immobilized α-amylase at different pH values and with different starch substrates: **a** corn starch, **b** germinated wheat starch, and **c** ungerminated wheat starch (Porto et al., 2019). The permission to reproduce it has been obtained

FP) nanofibers containing reactive epoxy groups and biocompatible feather polypeptide (FP) was fabricated by electrospinning which was the first time used for the covalent immobilization of lipase. This study revealed that the P(GMA-co-MA)/FP-Lipase possesses a wide pH tolerance, high thermal stability, good reuse, and organic solvent stability (Liu et al., 2018a).

Biosensing in Food Testing

Biosensors are popular among medicine, food safety, environmental monitoring and other fields because of their fast detection speed, high specificity, practical, and diagnostic performance. Biosensor materials are readily available and have the potential to be manufactured on a large scale (Turasan et al., 2019). Biosensors are sensitive to living substances and can convert their concentration into other signals (such as electrical signals) for detection. The applications of biosensors in food analysis include the determination and analysis of food ingredients, food additives, harmful poisons, and food freshness.

In recent years, smart (or intelligent) packaging that can measure the freshness of food has been one of the popular application fields for electrospinning of biological macromolecules. Nanofibers structures have a high surface to volume ratio rendering unique characteristics such as small pore size, high porosity, and also a high absorbance capacity. Therefore, they could be considered as appropriate choices for intelligent packaging (Aghaei et al., 2020). Prietto et al. prepared ultrafine zein fiber containing 5% (w/v) anthocyanin, with an average fiber diameter of 510 nm. And it showed that the pH-sensitive membrane exhibited color changes from pink to green, when exposed to acidic and alkaline buffers, respectively (Prietto et al., 2018). To assess the quality of rainbow trout fillets, Aghaei et al. designed a protein-based halochromic nanos-sensor. Zein nanofibers containing alizarin as the indicator dye were electrospun. The prepared sensor was used to detect the color change of the fish during storage at 4 °C. Over time, the sensor showed a color change from yellow to magenta, a color change observable to the human eye. The high sensitivity of the sensor and the apparent color change make it effective for monitoring the quality of fish stored at 4 °C. These results suggested that electrospinning nanofibers has great potential in developing smart materials for intelligent food packaging (Aghaei et al., 2020).

Surface enhanced Raman (SERS) has evolved and enriched over the past decades, demonstrating extremely high sensitivity in sensing in food analysis (Langer et al., 2020). SERS is generally used to study the conformation, adsorption interface surface state and structural analysis of biological macromolecules at interfaces. It is proven to be effective in analyzing the adsorption and orientation of molecules in the interface, etc. It is ideal to use environmentally friendly natural materials instead of plastics and synthetic plastics to manufacture SERS biosensors. A sensor platform based on zein nanofibers has been prepared using electrospinning technology. Due to the high surface area, high porosity and high surface roughness of this nanofiber, the strength of SERS is greatly increased (Turasan et al., 2019).

Commercial Products

Over the past decades, although most of trials stay in the laboratory stage, efforts have been made to convert electrospinning nanofibers into practical applications, and some commercial products have been realized. There are already some companies that have successfully marketed a series of electrospinning products. FILTREX and PARDAM NANOFIBERS have launched a product called RIFTELEN® N15, which is a filtration media. As an electrostatically spun nanofiber filter membrane, it can be used to filter cooking oil, wine, beer, lemonade, and juice. Donaldson, a company based in Minneapolis, USA, has produced a commercial air filter using electrospun nanofiber membranes. In the filter, electrospun nanofibers are covered with a substrate that captures submicron dust particles on the surface of the filter. This product enables dust removal in the food industry and makes the air in the industrial working environment cleaner. Medprin Biotech GmbH produces a nanofiber membrane made of polylactic acid and gelatin that is bioabsorbable and has a wide range of applicability in the food and biopharmaceutical industries. NanoSpun Technologies, dedicated to the development and production of active biological tissues for cosmetics, health, etc., has produced a nanofibrous biomaterials, which can be used in tissue regeneration devices. MANN + HUMMEL produced a nanofiber coated air filter, i.e., Micrograde NF fiber, which consists of a cellulose carrier material coated with a very thin layer of ultra-fine polymer fibers (Fadil et al., 2021). Very fine nanofiber membrane with polysaccharide macromolecule as the backbone can filter smaller impurity particles and has better filtration for gaseous substances, which can be used as a large non-toxic filter material for food industry.

Conclusion and Future Outlook

Fiber materials created from electrospinning of natural biopolymers hold great promise in food-related areas. The fibers prepared by electrospinning have large specific surface area, tunable length scale, versatile physio-chemical properties and are ease of functionalization, which can be further engineered to form multi-scale/type materials (including membranes, films, coatings, antibacterial mat, hybrid fiber composites) with great application potential. Natural biopolymers such as protein (zein, collagen, SPI, gelatin, WPI, SF, PPI, and keratin) and polysaccharides (PUL, cellulose and its derivatives, chitosan, KGM and alginates) come from a wide range of sources and have the advantages of biocompatibility, degradability, and low cost. They are high-quality natural materials and are widely used in food-related fields. Despite the advantages of protein such as biocompatibility and degradability, protein films have relatively poor water vapor barrier capabilities. Because the structure of plant proteins is mostly globular, the viscosity of the aqueous solution is too low to spin. Therefore, the mixture of protein and other polymers is a favorable practice for spinning nanofibers with excellent properties. Polysaccharides have good mechanical properties and effective barrier to low polarity compounds, but with low permeability to gases (mainly oxygen and carbon dioxide) and poor barrier to water vapor. In recent years, protein – polysaccharide hybrid spinning becomes a good method to obtain food-grade nanofibers with excellent properties. Polysaccharides tend to have a linear structure and their solutions are more viscous and easier to be spun than proteins. And under certain conditions, some polysaccharides electrospinning has the possibility of forming very fine fibers within 100 nm. Under the influence of external conditions and solution concentration, the proteins subjected to electrospinning may form beaded fibers or homogeneous fibers. Natural polymers have the ability to encapsulate small molecules of active substances or essential oils by electrospinning, which in turn affects the morphology and mechanical properties of nanofibers.

With the present research and future perspectives in hand, natural biopolymer based biocomposite and bionanocomposite fibers are sure to capture the food markets. Nanofibers can encapsulate and release active substances, active materials with certain functions can be prepared. Packaging materials for prolonging the storage period of food can be developed by encapsulating antibacterial and antioxidant active substances into electrospun fibers. In the preservation of fruits and vegetables, packaging materials that can absorb ethanol are of great significance to prolong the shelf life. As an encapsulation material, nanofibers can encapsulate probiotics and bioactive components, which plays a protective role and help them function. An important topic is the immobilization of enzyme/protein to avoid the reduction of enzyme activity due to environmental impact. Electrospinning of natural biopolymers has also been masterly employed to prepare food biosensing materials. The new SERS made of electrospun materials can be used for surface research and the study of the interface orientation of biomolecules in food analysis. Intelligent packaging that can in situ monitor the freshness of stored food is also one of such smart applications.

Indications are that nanofibers obtained by electrospinning of natural biopolymers have a favorable future as food-grade materials. The development of active packaging materials with unique functional properties is challenging and further research relating to their application in the substitution of petroleum-based materials is imperative.

Electrospinning technique should be further explored to prepare biological macromolecule materials to encapsulate food active substances and release it in a controlled manner. Massive chances still exist to create a new kind of blends of edible polymers (hybrid biopolymers) with new characteristics, which could be a promising area in both food and non-food packaging. It is projected that the applications of electrospinning natural biopolymers to produce functional fiber-based materials in this significant and so far still rising research area will be robustly increasing.

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Declarations

Conflict of Interests The authors declare that they have no conflict of interest.

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