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Materials, Properties, and Future Trends of Electrospinning Nanofiber in Smart Food Packaging Development: a Review

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Abstract: The development of smart food packaging is a rapidly evolving field aimed at enhancing food safety, quality, and shelf life. Among various techniques, nanofiber electrospinning is a versatile and efficient method for synthesizing nanofibers with unique properties suitable for this application. This review aims to provide valuable insights and foster further research in the innovative use of nanofiber electrospinning for smart food packaging solutions. This review paper provides a comprehensive overview of the current advancements in using electrospinning for nanofiber synthesis in smart food packaging. It delves into the advantages of electrospinning, including its ability to produce fibers with high surface area-to-volume ratios and tunable properties. The paper discusses synthetic and natural materials used in the electrospinning process and the incorporation of active compounds to impart functional characteristics to the packaging. A distinctive feature of this review is the in-depth analysis of recent improvements in nanofiber technology to enhance its mechanical properties. This area has received limited attention in previous reviews. Finally, it highlights the future trends and potential developments of electrospinning nanofibers in smart food packaging.

Keywords: Food packaging, nanofiber, electrospinning, polymer, active compound.

1. Introduction

Concerning the increasing global population growth, plastics become a crucial problem and are one of the main issues for international organizations. Most of these materials are derived from non-degradable petroleum and contain dangerous chemicals. They can pollute the environment and contribute to health issues [1]. For instance, plastic waste can be transformed into microplastic (MPs) materials that are dangerous to human health as they can be ingested. Microplastics are found in many foodstuffs and beverages, especially seafood such as bivalves, crustaceans, and fish [2]. A study conducted in 2020 estimated that individuals consume approximately 2,977 microplastics annually through food. Those who eat food

packaged in plastic 4 to 7 times a week could ingest around 12 to 203 microplastics each week from the packaging. Various factors contribute to the degradation of plastic materials, with aging, temperature, and external mechanical forces being the primary causes of microplastic release into food, the human body, and the environment [2,3].

Together with the development of technology and sciences, the scope of biomedical engineering also pays attention to these issues. In the context of food packaging applications within biomedical engineering, biomaterials (BME) play a distinctive and valuable role as they are included in the scope of BME (Figure 1). Biomaterials in this domain are centered around creating packaging solutions that preserve the freshness and quality of food and address environmental and health considerations. One critical application of biomaterials in food packaging involves the development of smart and responsive packaging systems. Biomedical engineers leverage biomaterials to create sensors and indicators embedded in packaging that can monitor and relay information about the condition of the packaged food. For example, bioactive films or coatings made from biomaterials can change color in response to temperature variations or pilage-indicating gases, providing consumers and producers with real-time information about the freshness of the contents [4].

The use of biodegradable and active-health promotion biomaterials is another key aspect. With increasing concerns about plastic pollution, biomedical engineers are exploring biomaterials derived from renewable sources, such as starch, cellulose, or even proteins, to create environmentally friendly and sustainable packaging options. These materials can be engineered to have the necessary barrier properties to protect the food while being biodegradable, contributing to reduced environmental impact. Furthermore, antimicrobial packaging, a crucial consideration for food safety, involves the incorporation of biomaterials with inherent antimicrobial properties. Biomedical engineers design packaging materials that can actively inhibit the growth of microorganisms, extending the shelf life of perishable products. This approach not only ensures the safety of the food but also reduces the need for chemical preservatives [4].

As a result, developing safer and more eco-friendly packaging alternatives to plastic is transforming the market. Biodegradable and renewable materials offer a promising option for enhancing public health and protecting the environment [1]. There has been growing interest in creating smart food packaging films made from biopolymers and natural bioactive compounds free from harmful chemicals [5–7]. These smart films and coatings are increasingly used in food packaging to preserve food quality, providing convenient products while ensuring freshness.

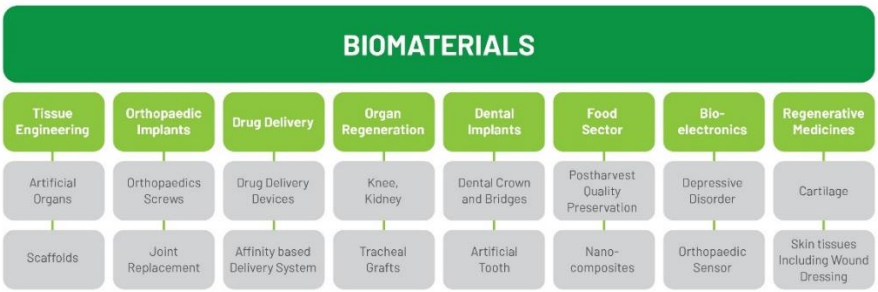

Figure 1. Biomaterials scope applications.

2. Smart Food Packaging Concept

Food packaging systems are a crucial part of human life to preserve food products and maintain their shelf-life. Traditional food packaging has four basic functions: protection and

preservation, containment, communication and marketing, and convenience [7,8]. In recent decades, the food industry has widely used plastic-based food packaging systems because of their unique characteristics. However, due to the massive use of these materials and the increasing human global population, serious environmental problems exist worldwide. Moreover, recent research has revealed negative reports of human health concerns because of plastic-based food packaging [9]. Therefore, to cope with these problems, developing new eco-friendly food packaging systems, together with an innovation concept, has recently become a new focus. Harnessing biocompatibility, biodegradable, and renewable materials, such as food packaging materials, provides a great alternative to protect the environment and public health. Regarding these, bioplastics have begun to gain attention. According to the European Bioplastics Organization, they are defined as plastic materials that are either biobased (partly or entirely), biodegradable, or feature both properties.

With the increase of attention paid to bioplastics, the concept of smart food packaging has become an interesting idea. Smart food packaging is a term that combines active and intelligent concepts in food packaging materials (Figure 2). Active packaging is defined as the modification of food packaging to obtain advanced characteristics such as extending shelf-life, improving its safety, or enhancing food quality. These features can be achieved through the inherent properties of the polymers or by adding specific compounds. Active packaging is designed to either release or absorb substances to or from the packaged food and its surrounding environment. The goal is to directly and positively affectonsumer health by producing healthier packaged foods [9].

On the other hand, intelligent packaging functions as a sensor, detecting changes in the environment or the condition of the food and processing that information to signal potential issues. Like active packaging, intelligent packaging is created by incorporating specific compounds into the system. However, unlike active packaging, it does not directly extend the food's shelf life or release its components into the food. Smart packaging offers a comprehensive solution: it monitors changes in the product or environment as intelligent packaging and responds to these changes to enhance or maintain the food's health quality as active packaging [9].

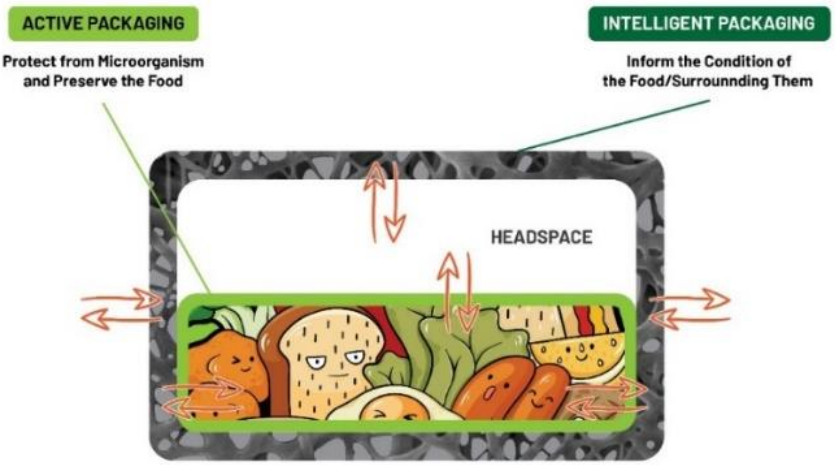

Figure 2. Smart food packaging illustration.

3. Nanofiber Electrospinning

Nanotechnology plays a crucial role in advancing food packaging and creating innovative solutions to meet industry needs [10]. Compared to traditional materials,

nanomaterials offer several benefits, including improved physical, chemical, optical, mechanical, and catalytic properties, making nanotechnology highly promising for food packaging development. Additionally, nanomaterials can serve as carriers for active agents, enhancing packaging performance. For example, integrating antioxidants into packaging materials can help preserve the physical properties of food, such as flavor and color. Polymers at the nanoscale can encapsulate active agents and control their release. However, many active agents are highly volatile and cannot be easily incorporated into polymers using conventional methods. Electrospinning technology can be employed to produce nanofibers for food packaging applications to address this issue [11,12].

Several techniques are available for fabricating nanofibers, including drawing, template synthesis, phase separation, self-assembly, and electrospinning. Among these, electrospinning is the most widely used method for producing polymeric nanofibers due to its repeatability, ease of scaling up, and ability to create long, continuous fibers. This technique typically produces nanofibers ranging from 10 to 1000 nm in diameter, with high elasticity, strength, and a large surface area relative to volume, depending on the polymer. Additionally, the nanofibers have high pore volumes due to their large surface area, which can be used to encapsulate bioactive molecules or other substances, making polymer nanofibers a versatile class of biomaterials [13]. Electrospinning works by applying a high voltage to a polymer solution, causing the nanofibers to form through electrostatic repulsion and the stretching of the solution (Figure 3). The polymer solution is continuously fed through a capillary subjected to a high voltage (10–30 kV). When the electric field exerts enough force to overcome surface tension, a "Taylor cone" forms at the tip of the capillary, and the nanofibers are deposited onto a collector, which may be stationary or rotating. As the solvent evaporates, only nanofibers remain on the collector. Dynamic interactions between polymer chains, such as interlacing, hydrophobic interactions, and hydrogen bonding, play a crucial role in reducing fiber diameter and ensuring continuous fiber formation [13].

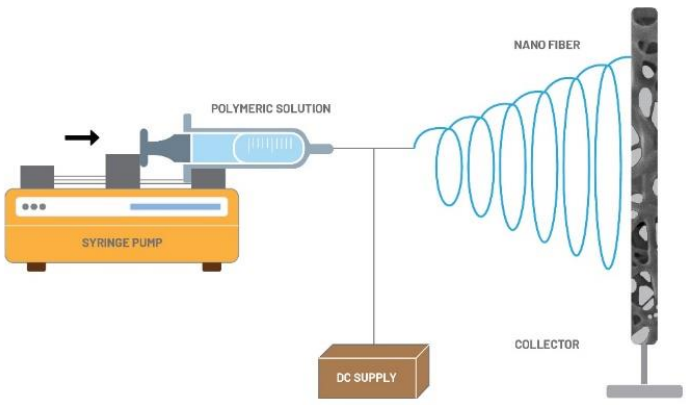

Figure 3. Nanofiber electrospinning illustration.

The electrospinning process is fine-tuned by adjusting various parameters, which can be classified into solution, process, and environmental factors. Solution parameters include polymer concentration, volatile solvent, molecular weight, viscosity, surface tension, and surface charge conductivity/density. Process parameters involve applied voltage, flow rate, types of collectors, and the distance between the capillary and collectors. Environmental parameters encompass humidity, temperature, and airflow. These factors are interconnected and collectively influence nanofiber formation, affecting the morphology and diameter of the resulting nanofibers. Achieving the desired nanofiber characteristics depends on properly

managing these parameters [13]. Materials used in electrospinning include natural ingredients, synthetic materials, and their blends [14]. Natural polymers such as polysaccharides, proteins, and lipids offer benefits like low toxicity, excellent biocompatibility, renewability, and controlled degradability compared to synthetic polymers. In addition to the unique properties of nanomaterials, electrospun biopolymers have advantages in terms of safety and biodegradability. As a result, electrospun nanofibers hold significant potential for applications in the food industry [15].

4. Nanofiber Materials

4.1. Nanofiber matrix.

Polymers commonly fabricate the composition of the nanofiber matrixes. There are two main polymers types: natural and synthetic (Table 1). A synthetic polymer is more favorable than a natural polymer in enhancing the spinnability properties of the polymer. Furthermore, a synthetic polymer commonly has a better mechanical strength characteristic. However, natural polymers have gained more attention in the context of biodegradability and biocompatibility. It is also supported by a diverse binding site of the natural polymer for bioactive compounds [16,17].

4.1.1. Natural polymer.

4.1.1.1. Gelatin.

Gelatin is a partially hydrolyzed version of collagen, containing shorter amino acid chains. While gelatin is technically a collagen type, it is more affordable, easier to obtain, poses a lower immunological risk, and offers enhanced hydrophilicity and better cell adhesion [18]. Nanofiber's research based on gelatin polymer is one of the most studied polymers in various applications. Regarding its use in nanofiber development, gelatin can be used as a single polymer matrix or blended with other polymers. Lin *et al*., Alp-Erbay *et al*., Lin *et al*., Alehosseini *et al*., Zhou *et al*., Yilmaz *et al*., and Cetinkaya *et al*. developed nanofiber using gelatin as a single matrix [19–25]. On the other side, Eghbalian *et al*., Li *et al*., Liu *et al*., Nath *et al*., Shi *et al*., Ghalehjooghi *et al*., Maroufi *et al*., Heydarian and Shavisi, Ullah *et al*., Zhao *et al*., Pandey *et al*., Ertan *et al*., Wu *et al*. using combined polymer as their nanofiber matrixes instead of single gelatin [26–38].

Lin *et al*. [19] developed nanofiber for meat preservation to avoid *Campylobacter jejuni* contamination using thyme essential oil/β-cyclodextrin ε-polylysine nanoparticles (TCPNs) as bioactive and gelatin as their matrix. They used gelatin instead of synthetic polymer because of the biosecurity concern associated with using synthetic polymer. The result showed that the morphology of the gelatin nanofiber showed a uniform structure. However, there was a slight increase in the fiber diameter after adding the bioactive into the nanofiber solution. Lin *et al*. [19] reported an increase in fiber size under the SEM observation after adding the bioactive, which was also supported by the AFM assessment. Some small bumps were observed on the nanofiber surface, which can be seen in the 3-dimensional image. The bumps showed the bioactive incorporation in the fibers.

Furthermore, Pandey *et al*. [36] blended PVA and chitosan as the matrix of the nanofiber with the addition of silver nanoparticles (AgNPs) as bioactive. These blending polymers can combine the different characteristics of each polymer to obtain the desired

effects. The result of the morphology characterization of the fiber showed the uniform-cylindrical morphology. Meanwhile, Ullah *et al*. [34] combined both natural polymer and synthetic polymer to make the desired nanofiber from gelatin, zein, and PVA. Natural polymer is considered for its safety and sustainability, while synthetic polymer is good for considering its easy electrospinnability. According to the result, combining PVA and gelatin can produce uniform and beadles fiber. On the other side, adding the gelatin into the zein nanofiber led to the decrease of the beads in the zein nanofiber.

4.1.1.2. Zein.

Zein protein, a natural polymer, is deemed safe by the US Food and Drug Administration (FDA). It is utilized in various fields, including tissue engineering, the food industry, and medical applications. Its hydrophobic nature makes it suitable for controlling the release of hydrophobic drugs. While many protein polymers struggle to form nanofibers due to their complex structure, zein can easily do so. Zein scaffolds exhibit high porosity, resistance to microbial attack, biocompatibility, and antioxidant properties. However, the stability of zein fibers in wet conditions and their mechanical strength are significant limitations [39].

Like gelatin polymer, zein is also used as a single polymer matrix or blended with other polymers. Aghaei *et al*. [40] developed a food packaging nanofiber using zein protein and alizarin as an organic dye to detect the freshness of the trout fillets. The result showed that the low concentration of zein protein leads to low viscosity and produces more beads in the nanofiber morphology. Furthermore, adding alizarin dye did not change the zein fiber structure significantly. On the other side, Mohammadi *et al*. [41] developed a food packaging system using zein and PLA-based nanofiber. The addition of PLA in the blended polymer with zein increases the strength of the fiber and makes it easier to handle. Moreover, they also added HPMC to modulate more in the control-release of Zenian (*Carum copticum*) essential oil (ZO) as its bioactive. The result showed that regarding the increase of zein concentration in the nanofiber solution, the diameter of the fiber revealed a smaller size. This can be done because increasing the zein content may increase the conductivity of the nanofiber solution, which leads to greater elongation forces for the biopolymer jet and reduces fiber size.

4.1.1.3. Cellulose derivatives.

Cellulose consists of a linear arrangement of repeating $(C_6H_{10}O_5)n$ units, forming large macromolecules. It is typically found in the form of microfibrils in the cell walls of wood and plants and algae tissues, membranes, tunicate epidermal cells, and bacterial byproducts. Cellulose derivatives can be easily synthesized through simple reactions involving the hydroxyl groups and functional substituents [42]. These materials possess a semi-crystalline structure with both highly ordered and loosely disordered regions, and the crystalline-to-amorphous phase ratio varies depending on the synthesis method. The hierarchical organization of cellulose, from the nano- to microscale, contributes to its excellent mechanical properties, particularly the high specific strength of cellulose-based fibers. Combined with their biocompatibility, renewability, functionality, and environmental friendliness, cellulose and its derivatives aredeal for certain biomedical applications [43]. However, cellulose in its original form has poor solubility in common solvents, making it difficult to produce as nanofibers. Researchers often use cellulose derivatives to address this challenge, which are easier to dissolve.

Compared to other cellulose derivatives, ethyl cellulose (EC) is one of the most used in developing nanofiber using electrospinning for food packaging applications. Ethyl cellulose is produced by reacting cellulose with ethyl chloride or ethylene oxide. In this process, some of the cellulose molecules' hydroxyl groups (-OH) are replaced by ethyl groups (-CH_2CH_3). EC is solved in organic solvents such as chloroform, toluene, and ethanol. Yang *et al*., Rashidi *et al*., Beikzadeh *et al*., and Nath *et al*. used EC as the main matrix of their nanofiber [44–47]. They blended it with other polymers, commonly with an organic polymer, except for Beikzadeh *et al*., which blended with PCL as a synthetic polymer [46].

Nath *et al*. [47] developed nanofiber with EC as its matrix for bioactive trapping. This research extracted anthocyanin from red cabbage and encapsulated it into the gelatin nanoparticle. According to the result, adding anthocyanin-gelatin nanoparticles to the EC nanofiber showed no significant change in the structure compared to the pure EC nanofiber. However, the bioactive material can be seen trapped in the fiber as a round-shaped particle, which makes the fiber slightly bead-formed. On the other side, Beikzadeh *et al*. [46] developed nanofiber using blended polymers among EC, gelatin, and PCL that encapsulated ZnO and Zataria multiflora essential oil (ZEO) as the active compound. The combination of these polymers produced a uniform nanofiber structure. Then, after the encapsulating process, the size of the fiber increases depending on the concentration of the active compound.

Meanwhile, other cellulose derivatives that are also used in the development of food packaging nanofiber include hydroxypropyl methylcellulose (HPMC), cellulose acetate (CA), and carboxymethyl cellulose (CMC). HPMC and CMC are generally soluble in water, but CA is more soluble in organic solvents. Aydogdu *et al*. [48] and Hashmi *et al*. [49] combine the cellulose derivatives with synthetic polymers, while Shi *et al*. [30] use natural polymers to develop composite material nanofiber. Furthermore, Mohammadi *et al*. [41] combined both natural and synthetic polymers instead of using one of them to develop food packaging nanofiber. The almost similar trend from the previous research was the fiber diameter increased together with the increase of the viscosity of the solution.

4.1.1.4. Chitosan.

Chitosan is a polysaccharide widely studied as a biomaterial for creating scaffolds used in tissue regeneration. Chitosan nanofibers are frequently utilized because their structure and chemistry closely resemble the natural extracellular matrix (ECM), making them biocompatible and biodegradable. Additionally, chitosan has been noted for its antimicrobial, antiulcer, and antitumor properties, as documented in the literature [50–52]. It is produced through the deacetylation of chitin. One of its key advantages is its ability to adopt various conformations and bind to a wide range of functional groups, allowing for versatility in specific applications [53].

In the context of natural polymer-based nanofiber food packaging development, chitosan is a natural polymer mostly used after gelatin and zein proteins. Similar to other natural polymers, chitosan is rarely used as a single polymer or nanofiber matrix. This is because natural polymers commonly possess drawbacks, such as poor mechanical strength and spinnability, compared to synthetic polymers. Hence, blended polymer or developed composite material is preferred for use in the chitosan-based nanofiber. Ardekani-Zadeh and Hosseini, Zou *et al*., Ahmed *et al*., Zou *et al*., and Zang *et al*. developed chitosan-based nanofiber blended with synthetic polymers [54–58]. On the other side, Roshanak *et al*., Liu *et al*., Duan *et al*., Shavisi and Shahbazi, and Shavisi *et al*. blended chitosan with natural polymers [59–63].

Furthermore, Duan *et al*. [64] and Boonmahitthisud *et al*. [65] used chitin nanofiber instead of chitosan. Chitosan is derived from chitin through deacetylation, which removes some of the acetyl groups from the N-acetylglucosamine units. Different from chitosan, chitin is harder soluble in various solvents because of its rigid structure. Hence, chitin is more rarely used than chitosan to form nanofiber.

Zou *et al*. [55] developed nanofiber food packaging using chitosan and PCL as its matrix. Moreover, they encapsulated chlorogenic acid (CGA) as the active compound into halloysite nanotubes (HNTs) before incorporating it into the nanofiber to obtain more sustained release. According to the result, chitosan did not produce fiber after the SEM assessment, and it can form a uniform nanofiber after adding PCL—furthermore, the addition of active compounds beyond 6 wt% results in the merged fibers. In the next years, Zou *et al*. [57] researched to improve chitosan usage in nanofiber applications. They used hydroxypropyltrimethyl ammonium chloride chitosan (HACC) instead of chitosan. Chitosan has limited antibacterial activity in the condition of pH above 6.5 due to its poor solubility in the alkaline condition. Hence, they used HACC, which has higher antibacterial activity than chitosan in its original form.

4.1.1.5. Pullulan.

Pullulan, a natural polymer, is originally from the yeast-like fungus *Aureobasidium pullulans*. It is non-toxic, non-immunogenic, non-carcinogenic, and non-mutagenic [66]. Its structure is characterized by a distinctive arrangement of glycosidic bonds, including two -(1→4) and one - (1→6) bonds within maltotriose repeating units (G3). The unique physical characteristics of pullulan stem from the nine hydroxyl groups present on the glucopyranose rings of its G3 unit [67]. In recent research about the development of smart food packaging, pullulan was used as a single polymer or blended with other natural polymers. Duan *et al*. [64] and Qin *et al*. [68] fabricated nanofiber for food packaging with pullulan as a single matrix. Furthermore, Özgen and Özbaş. [69], Yang *et al*. [44], Jia *et al*. [70], and Ertan *et al*. [37] developed nanofiber using pullulan for food packaging blending with other natural polymers.

Qin *et al*. [68] synthesized nanofiber for active packaging using pullulan as its matrix and *Zanthoxylum bungeanum* essential oil/β-cyclodextrin inclusion complexes as its active compound. Pullulan-only nanofiber showed uniform morphology in the 20 wt% concentration. After the addition of the active compound, there was a change in the structure. The result showed that adding *Zanthoxylum bungeanum* essential oil/β-cyclodextrin with a similar total polymer concentration can reduce the fiber size. However, the research stated that the reduction of the fiber diameter is related to the reduction of the solution viscosity, which gave more beads fiber morphology. Furthermore, according to the thermal analysis, adding the active compound led to increased thermal stability. Moreover, an increase in the polymer concentration gave more mechanical strength regarding the total pullulan concentration.

Yang *et al*. [44] developed food packaging nanofiber using blended polymer pullulan and ethyl cellulose (EC), while cinnamaldehyde (CA) is used as the active compound. As a result of the research, the addition of CA decreased the diameter of the nanofiber. This result was similar to [68]the research of Qin *et al*. [68], which found that adding an essential oil compound decreased the fiber diameter. According to the research, it decreased because of the reduction of viscosity and increased conductivity of the solution. In contrast, the thermal stability was not affected by adding CA into the nanofiber. Furthermore, blending pullulan with

EC was a good approach to make the fiber more hydrophobic, as the result showed. Besides, adding CA also improved the water hydrophobicity of the nanofiber.

4.1.1.6. Others.

Other natural polymers are also used to fabricate nanofiber for food packaging applications. In this review, a natural polymer was used to synthesize nanofiber for food packaging, but it was not as prevalent as previous polymers, which were summarized in this section. The natural polymer included starch, carrageenan, soybean protein, casein, xanthan gum, and alginate. The details of the research are displayed in Table 1. Almost all previous natural polymers had been synthesized into nanofiber with a single polymer for food packaging. Conversely, based on a recent research journal, in the natural polymers summarized in this section, only starch has been used mostly as a single matrix of nanofiber for food packaging.

Starch was employed as a single matrix nanofiber for food packaging applications by Cai *et al*. [71], Zhu *et al*. [72], and Yang *et al*. [73], while Lv *et al*. [74] blended it with PVA. All of them incorporated starch nanofiber with organic-based active compounds except Zhu *et al*. [72], which did not combine with any active compound. Instead of developing active nanofiber packaging with active compounds. Zhu *et al*. [72] focused on modifying starch to be more water resistant. However, the modification of the fiber matrix was discussed separately in this review paper. Meanwhile, Yang *et al*. [73] fabricated starch-based nanofiber for active packaging with tannic acid (TA) and Fe^{3+} as its active compound. The result of the morphology of the nanofiber showed a uniform structure. The addition of TA to the solution increases the diameter of the fiber, and as the solution is more viscous, the fiber tends to increase. However, adding Fe^{3+} increases the conductivity of the solution and decreases the fiber diameter.

Alongside starch, carrageenan is a widely used natural polymer, although it is not as prevalent as previous natural polymers. In recent publications, carrageenan-based nanofiber for active food packaging was commonly blended with other synthetic polymers. Forghani *et al*. [75] and Goudarzi *et al*. [76] fabricated carrageenan nanofiber blended with PVA, while Panwar *et al*. [77] blended with PEO. On the other side, Shavisi *et al*. [63] synthesized carrageenan nanofiber blended with chitosan. Similar to the starch-based nanofiber, carrageenan-based nanofiber in recent publications mostly used organic active compounds except for Panwar *et al*. [77], which used titanium oxide as its active compound. The rest research used soybean protein (Bruni *et al*. [78], Rashidi *et al*. [45], Raeisi *et al*. [79]), casein (Eghbalian *et al*. [26], and Maroufi *et al*. [80]), xanthan gum (Heydarian and Shavisi [33]), and alginate (Ghalehjooghi *et al*. [31]).

Besides other research that tried to entrap an active compound, both organic and inorganic compounds, the nanofiber in the context of smart food packaging development, Ghalehjooghi *et al*. [31] tried to encapsulate probiotic microorganisms into a nanofiber. In this research, sodium gelatin (SA) was used with gelatin as the nanofiber matrix. Probiotics were used, including Lactobacillus acidophilus, *Limosilactobacillus reuteri*, *Lacticaseibacillus casei*, and *Lacticaseibacillus rhamnosus*. They encapsulated them to delay the growth of *Vibrio parahaemolyticus*, *Salmonella Typhimurium*, *Staphylococcus aureus*, and *Listeria monocytogenes*. The morphology result from SA and gelatin nanofiber showed a uniform structure. Furthermore, adding these probiotics changes the fiber structure to become oval in certain parts. Moreover, the entrapment also leads to a decrease in tensile strength while the flexibility increases.

4.1.2. Synthetic polymer.

4.1.2.1. Polyvinyl alcohol.

Poly(vinyl alcohol) (PVA) is a synthetic polymer consisting of repeating units of $CH_2CH(OH)$. While this polymer is derived from vinyl monomers, it's not directly synthesized from vinyl alcohol due to its unstable nature, which triggers a tautomerization process resulting in acetaldehyde [81]. It exhibits strong mechanical properties, including high tensile strength and flexibility, alongside effective barriers against oxygen and aroma. Furthermore, it lacks odor, is safe for use, and excels in forming films, emulsifying substances, and adhering surfaces. Additionally, it demonstrates resilience against grease, oil, and various solvents. However, being water-soluble, its performance relies on moisture content [81].

PVA is the most used synthetic polymer based on recent publications developing food packaging using nanofiber. According to recent journals, PVA was commonly developed as a single nanofiber matrix without adding other polymers. This may be due to PVA nanofiber's excellent mechanical properties and good spinnability. Feng *et al*., Shao *et al*., Maftoonazad and Ramaswamy, Lan *et al*., Nazari *et al*., He *et al*., Kowsalya *et al*., Goksen *et al*., Dogan *et al*., Ansarifar *et al*., Zhang *et al*., Quintero-Borregales *et al*., Han *et al*., and Turgay Cetinkaya developed nanofiber for active packaging using PVA as a single matrix [82–95]. On the other side, Hashmi *et al*., Xiao *et al*., Forghani *et al*., Goudarzi *et al*., Ullah *et al*., Thakur and Satapathy, and Lv *et al*. developed it using PVA blended polymer [34,49,74–76,96,97].

Cetinkaya [95] synthesized nanofiber for active food packaging using PVA and black carrot extract (BC) with SnO_2 incorporated as the active compound. The usage of BC and SnO_2 was employed as pH and gas sensing to monitor the freshness of the food. The addition of the active compounds increased the fiber diameter and gave a crystal-like shape to the fiber surface, showing the successful incorporation of SnO_2. Furthermore, the addition of SnO_2 increased the roughness of the nanofiber surface. It can be beneficial properties for PVA-based nanofiber as the rough surface can increase the water-resistant effect. On the other side, Hashmi *et al*. [49] fabricated nanofiber using PVA, PVP, and CMC to protect food. According to the result, the addition of CMC into the PVA solution enhanced the viscosity dramatically. Even adding CMC 2 wt% into PVA can not form the fiber. However, adding PVP to the PVA/CMC solution helped reduce the solution's viscosity. It gave a uniform and beads-free nanofiber. Furthermore, adding both CMC and PVP into PVA nanofiber showed no significant change in air permeability, but it showed a significant change in hydrophobicity. Adding CMC to the PVA/PVP can induce a crosslinking mechanism among the chains and enhance the water-resistant effect.

4.1.2.2. Polylactic acid.

Poly(lactic acid) or PLA, a synthetic biopolymer extensively employed in the biomedical sector, can be sourced from natural materials like sugar cane, rice, and corn starch. Its manufacturing involves condensation polymerization or ring-opening polymerization of lactic acid [98]. Lately, there has been an increasing focus on creating nanofibrous structures using the electrospinning method, particularly with PLA as the base material. These PLA nanofibers possess notable traits resembling the extracellular matrix (ECM), along with a significant specific surface area, high porosity featuring small pore size, and suitable mechanical properties [98]. Similar to the PVA nanofiber for food packaging, PLA-based has

also been widely used in recent publications, and most of them used PLA as a single matrix for developing nanofiber. Wen *et al*., Aytac *et al*., Altan *et al*., Liu *et al*., Zhang *et al*., Li *et al*., Zhu *et al*., Xie *et al*., and Liu *et al*. developed nanofiber using PLA as a single matrix [99–107]. On the other side, Râpă *et al*., Li *et al*., Dehghani *et al*., Liu *et al*., Bodbodak *et al*., Mohammadi *et al*., Min *et al*., and Shi *et al*. developed nanofiber with PLA and blended with other polymers [28,41,108–113].

Xie *et al*. [106] synthesized PLA-based nanofiber with phloridzin adsorbed onto an MCM-41 mesoporous silica sieve as the active compound to form active food packaging. The morphology result showed that pure PLA nanofiber revealed a uniform structure without any beads. However, after adding phloridzin and MCM-41, the structure worsened, and the film became visible as nodule-like structures. Furthermore, the thermal stability analysis revealed that PLA-base nanofiber loaded MCM-41/phlorizin showed better stability, and it was also supported that the pore volume of the nanofiber was lower and denser than MCM-41 itself. Hence, the nanofiber composite was suitably applied as active food packaging. Meanwhile, Min *et al*. [112] fabricated nanofiber for active food packaging using PLA-pectin nanofiber loaded thymol and crosslinked using polyethyleneimine (PEI). The result showed that the addition of PEI led to an increase in the porous size of the fiber compared to the PLA only. However, there was no significant change in the diameter of the fiber compared to both. Furthermore, the TGA results demonstrated that the pectin coating improved the thermal stability of PLA-PEI nanofibers.

4.1.2.3. Polycaprolactone.

Polycaprolactone (PCL) is an FDA-approved polymer widely studied for its use in several biomedical applications, including tissue engineering, drug delivery systems, and implantable biomaterials [114]. PCL is an aliphatic, semicrystalline thermoplastic polyester that remains soft at room temperature. The two main methods for producing PCL, as cited in various sources, are ring-opening polymerization (ROP) of ε-caprolactone (εCL) and polycondensation of 6-hydroxyhexanoic acid [114]. It also has good rheological properties; a low melting temperature (glass transition (Tg) ≈60°C, melting temperature (Tm) ≈60°C) [114]. PCL has been widely blended with other polymers and biological compounds with unique properties. Recent studies showed that PCL was blended with natural polymers, including Ardekani-Zadeh and Hosseini, Zou *et al*., Beikzadeh *et al*., Maroufi *et al*., Zou *et al*., and Zang *et al*. [46,54,55,57,58,80]. On the other side, Alonso-González *et al*., Jovanska *et al*., and Shi *et al*.. Furthermore, Dumitriu *et al*. and Liu *et al*. developed active food packaging nanofiber only using PCL polymer as the nanofiber matrix [113,115–118].

Dumitriu *et al*. [117] developed nanofiber using polycaprolactone as a matrix and incorporated α-tocopherol antioxidant, a major component of vitamin E as its bioactive. The result showed that the fibers from PCL had hydrophobic characteristics, reflected by the high contact angle of 121°. This characteristic was enhanced by the incorporation of α-tocopherol, with small amounts of 1 wt% having the highest effect, which could be due to the increased roughness of the fibers. Furthermore, the addition of α-tocopherol could decrease the viscosity of the solution compared to the PCL-only solution. It resulted in finer and smoother fibers, as revealed in the SEM result analysis. Conversely, Zou *et al*. [56] developed food active packaging nanofiber using a blended polymer between PCL and chitosan as a natural polymer. Chlorogenic acid (CGA) loaded halloysite nanotubes (HNTs) were used as an active

compound. Pure chitosan could not be synthesized in this research, while pure PCL revealed a slightly homogenous fiber.

Furthermore, the addition of chitosan into the PCL solution resulted in a decrease in the nanofiber size. In contrast, incorporating CGA@HNTs into the composite polymer increased the fiber size until they merged with each other in the 6 wt% of CGA@HNTs. Shi *et al*. [113] developed polycaprolactone (PCL) electrospun nanofibers with added oregano essential oil (OEO)–loaded β-cyclodextrin (β-CD) as an active food packaging material. The result showed that blended polymer increased the fiber size by around 1144.5 nm. Furthermore, the addition of OEO@β-CD enhanced the thermal stability of the PLA/PCL fibers in the DSC result. They suggested that the enhanced thermal stability may be because of the formation of hydrogen bonds, which led to the chemical conjugation on the surface.

4.1.2.4. Polyethylene oxide.

Polyethylene oxide (PEO) is a high-molecular-weight, non-ionic polymer that is hydrophilic, linear, and non-cross-linked. It is highly soluble in both water and organic solvents. PEO is produced through the polymerization of ethylene oxide using a metallic catalyst [119]. Previous studies showed that harnessing PEO as a synthetic polymer for developing active food packaging nanofiber was widely used and commonly combined with other polymers. Jovanska *et al*. [116] combined PEO with PCL as a similar synthetic polymer, while other research, including Aydogdu *et al*., Aydogdu *et al*., Yildiz *et al*., Yildiz *et al*., Zeren *et al*., Jiang *et al*., Panwar *et al*., Lin *et al*. combined PEO with natural polymers [48,77,120–125].

Lin *et al*. [125] developed active food packaging nanofiber using *Lycium barbarum* polysaccharides (LBP) and PEO as the matrix. Furthermore, Eugenol (EO) was encapsulated into silk fibroin nanoparticles to incorporate them into the nanofiber (EO/SFNPs). The result showed that the increase in LBP concentration resulted in a less likely fiber structure due to the lower viscosity of the final solution. Furthermore, adding EO/SFNPs decreased the fiber diameter and showed a similar structure to PEO-only nanofiber. The decrease in the fiber size may be caused by the larger the conductivity and the greater the electrostatic repulsion. Next, the thermal stability analysis revealed that the PEO-based nanofiber-loaded EO/SFNPs had lower mass loss than the LBP nanofiber, indicating more thermal stability. On the other side, Jovanska *et al*. [116] combined PEO with PCL to make active food packaging nanofiber-loaded anthocyanin extract (AE) and silver nanoparticles (AgNPs). The morphology result showed that the blended polymer between PCL and PEO had a uniform fiber structure. The addition of AE resulted in a lower fiber size than PCL-PEO nanofiber. However, the contact angle of both showed a similar result at 24°. Furthermore, AgNPs were incorporated into the PCL/PEO-AE nanofiber, resulting in a more hydrophobic contact angle (33°), while the fiber size was not significantly changed.

4.1.2.5. Others.

Other types of synthetic polymers were sometimes also used by researchers to develop active food packaging nanofiber. Polyvinylidene fluoride (PVDF) and poly(ethylene-co-vinyl alcohol) (EVOH) were the least common synthetic polymers that can be used in active food packaging development. Torres-Giner *et al*. [126] fabricated poly(ethylene-co-vinyl alcohol) (EVOH) and then embedded graphene nanoplatelets (GNPs) that had been synthesized from

graphite powder for intelligent food packaging. According to the result, EVOH nanofiber revealed a uniform structure and was completely free of beaded regions, with a mean diameter of approximately 675 nm. Next, the embedded GNPs in the nanofiber showed a decreasing trend in the fiber size. This can be related to an increase in the solution conductivity. However, this result was also accompanied by forming certain bead fibers and increasing GNP contents. Next, Guan *et al*. [127] developed active food packaging nanofiber using PVDF-based polymer. Guan *et al*. [127] fabricated PVDF fibrous film incorporated with β-cyclodextrin (β-CD) and Cinnamon essential oil (CEO), and it was designed for griskin preservation to enhance its shelf time. The result showed that PVDF-only nanofiber was uniform and bead-free. However, the addition of β-CD-CEO changes the fiber size and makes the structure more ribbon-shaped.

Furthermore, all samples showed hydrophobic characteristics as the water contact angle was above 90℃. This was due to the natural hydrophobicity of PVDF polymer. However, the addition of β-CD-CEO decreased the WCA result due to hydrogen bond interactions.

Table 1. Polymer types of nanofiber matrixes.

Nanofiber matrix		**References**
Natural polymer	Gelatin	Lin *et al*. [19], Alp-Erbay *et al*. [20], Lin *et al*. [21], Alehosseini *et al*. [22], Zhou *et al*. [23], Yilmaz *et al*. [24], Cetinkaya *et al*. [25], Eghbalian *et al*. [26], Li *et al*. [27], Liu *et al*. [28], Wang *et al*. [29], Shi *et al*. [30], Ghalehjooghi *et al*. [31], Maroufi *et al*. [32], Heydarian and Shavisi (2023) [33], Ullah *et al*. [34], Zhao *et al*. [35], Pandey *et al*. [36], Ertan *et al*., [37], Wu *et al*. [38]
	Zein	Aghaei *et al*. [40], Mohammadi *et al*. [41]
	Cellulose derivatives	Yang *et al*. [44], Rashidi *et al*. [45], Beikzadeh *et al*. [46], and Nath *et al*. [47], Aydogdu *et al*. [48], and Hashmi *et al*. [49], Shi *et al*. [30], Mohammadi *et al*. [41]
	Chitosan	Ardekani-Zadeh and Hosseini [54], Zou *et al*. [55], Ahmed *et al*. [56], Zou *et al*. [57], and Zang *et al*. [58], Lin *et al*. [21], Zhao *et al*. [60], Duan *et al*. [61], Shavisi and Shahbazi [62], and Shavisi *et al*. [63], Duan *et al*. [64] and Boonmahitthisud *et al*.[65]
	Pullulan	Qin *et al*. [68], Yang *et al*. [44]
	Others	Cai *et al*. [71], Zhu *et al*. [72], Yang *et al*. [73], Lv *et al*. [74], Forghani *et al*. [75], Goudarzi *et al*. [76], Panwar *et al*. [77], Shavisi *et al*. [63], Bruni *et al*. [78], Rashidi *et al*. [45], Raeisi *et al*. [80], Eghbalian *et al*. [26], and Maroufi *et al*. [80], Heydarian and Shavisi [33], and Ghalehjooghi *et al*. [31]
Synthetic polymer	Polyvinyl alcohol	Feng *et al*. [82], Shao *et al*. [83], Maftoonazad and Ramaswamy [84], Lan *et al*. [85], Nazari *et al*. [86], He *et al*. [87], Kowsalya *et al*. [88], Goksen *et al*. [89], Dogan *et al*. [90], Ansarifar *et al*. [91], Zhang *et al*. [92], Quintero-Borregales *et al*. [93], Han *et al*. [94], Turgay Cetinkaya [95], Hashmi *et al*. [49], Xiao *et al*. [96], Forghani *et al*. [75], Goudarzi *et al*. [76], Ullah *et al*. [34], Thakur and Satapathy [97], Lv *et al*. [74]
	Polylactic acid	Wen *et al*. (2016) [99], Aytac *et al*. [100], Altan *et al*. [101], Liu *et al*. [102], Zhang *et al*. [103], Li *et al*. [104], Zhu *et al*. [105], Xie *et al*. [106], Liu *et al*. [107], Râpă *et al*. [108], Parhi *et al*. [109], Dehghani *et al*. [110], Liu *et al*. [28], Bodbodak *et al*. [111], Mohammadi *et al*. [41], Min *et al*. [112], Shi *et al*. [113]
	Polycaprolactone	Ardekani-Zadeh and Hosseini [54], Zou *et al*. [55], Beikzadeh *et al*. [46], Maroufi *et al*. [80], Zou *et al*. [57], Zang *et al*. [58], Alonso-González *et al*. [115], Jovanska *et al*. [116], Shi *et al*. [113], Dumitriu *et al*. [117] and Liu *et al*. [118], Zou *et al*. [55]
	Polyethylene oxide	Jovanska *et al*. [116], Aydogdu *et al*. [48], Aydogdu *et al*. [120], Yildiz *et al*. [121], Yildiz *et al*. [122], Zeren *et al*. [123], Jiang *et al*. [124], Panwar *et al*. [77], Lin *et al*. [125]
Others		Torres-Giner *et al*. [126], Guan *et al*. [127]

4.2. Active compound.

In recent years, the development of active food packaging has increasingly focused on incorporating bioactive compounds into nanofiber matrixes through electrospinning technology (Table 2). This approach leverages the unique properties of nanofibers, such as high surface area-to-volume ratio and tunable porosity, to enhance the functionality of packaging materials. Bioactive compounds are encapsulated within these nanofibers to extend the shelf life and maintain the quality of food products [128]. The encapsulation process protects these

sensitive compounds from degradation and allows for controlled and sustained release, improving their efficacy in real-world applications. Commonly used bioactive compounds in this context include natural extracts like curcumin, essential oils, chitosan, and various plant-derived phenolic compounds, each contributing distinct protective and preservative benefits [129]. Integrating these bioactive substances into nanofibers represents a promising strategy to meet consumer demand for safer, more natural food preservation methods while addressing the global push towards sustainable and environmentally friendly packaging solutions.

4.2.1. Essential oils.

Essential oils (EOs) are intricate blends of volatile compounds plants produce for defensive and communicative functions [130]. EOs are the active compounds mostly used in the food packaging nanofiber. According to the recent literature, *Zataria multiflora* essential oil (ZMEO), cinnamon essential oil (CEO), and thyme essential oil (TEO) were EOs widely used for the active compound in the nanofiber for food packaging development. Moradinezhad *et al*., Beikzadeh *et al*., Ebrahimzadeh *et al*., and Raeisi *et al*. used ZMO as their active compound [46,79,131,132]. Next, Zhang *et al*., Nazari *et al*., Shao *et al*., Feng *et al*., Wen *et al*., and Wen *et al*. used CEO as their active compound [82,83,86,99,133,134]. Then, Lin *et al*., Min *et al*., and Aytac *et al*. used TEO as their active compound [19,135,136]. The use of active compounds in developing nanofiber food packaging is commonly used to extend food self-life or improve the effects of intelligent food packaging. However, incorporating EOs is mostly for antimicrobial and antioxidant effects for food preservation.

Based on the previous research, there were two approaches for using EOs as the active compound in developing food packaging nanofiber. The first approach was to incorporate the EOs into the nanofiber as an active compound, and the second approach was to encapsulate the EOs in another matrix and incorporate them into the nanofiber. Previous research that used the second approach to develop nanofiber for food packaging based on the EOs was mostly using β-cyclodextrin (β-CD) to encapsulate the EOs. Cyclodextrins are primarily utilized to enhance the solubility of substances not soluble in water. Additionally, the hydrophobic cavity of cyclodextrins offers a protective microenvironment for the guest, shielding it from volatilization and detrimental environmental influences [137]. Hence, these characteristics can overcome the limitations of the EOs, such as increasing the aqueous solubility and their chemical stability in the presence of light, oxygen, humidity, and heat.

Maroufi *et al*. [80] developed nanofiber for active food packaging using polycaprolactone (PCL)/casein (Cas) as the matrix's fiber and incorporating green tea essential oils (GTO) as an active food packaging to prevent spoilage and food loss. The morphology result showed that the combination of PCL and Cas was beads-free and had a uniform fiber structure. Furthermore, with the increase in the Cas weight ratio to the PCL, the fiber diameter was increased, making the distribution of the fiber size narrower. This result may come from the increase in the viscosity solution as adding Cas content enhances the viscosity. Because it will make a stronger resistance of the viscoelastic force to the electric field force, leading to the production of thicker nanofibers. A similar result was shown after the addition of GTO into the solution. The fiber diameter was increased, which may be due to the reduction of the conductivity of the solution because of the characteristic of GTO. Moreover, the mechanical properties of the nanofiber increased with the addition of Cas and GTO compounds. The addition improved the flexibility as adding Cas increased the tensile stress while adding GTO increased the elongation at the break parameter.

In a similar year, Shi *et al*. [113] also fabricated nanofiber for active food packaging with polymers that used polylactic acid (PLA) and polycaprolactone (PCL). For the active compound, they used an encapsulation approach: oregano essential oil (OEO) encapsulated by β-cyclodextrin (β-CD). Based on the result, combining PLA and PCL resulted in the beads-free nanofiber, but the fiber diameter increased significantly compared to its single polymer nanofiber. Furthermore, the addition of complex OEO- β-CD also gave a slight increase. However, the diameter decreased, and the weight increased for the complex. The blending of polymer and complex active compounds also led to an increase in the thermal stability of the nanofiber. This result was shown by increased melting temperature and temperature at the maximum weight loss rate. Next, the complex of OEO- β-CD in the PLC/PCL nanofiber showed variable mechanical properties results. The addition of 6wt% of OEO- β-CD showed the highest elastic modulus value, which represented the most stiff material compared to the other formulas.

4.2.2. Plant extract.

The use of isolated bioactive compounds from medicinal plants often called synthetic medicinal plants, is expensive and has been linked to adverse side effects in humans. These side effects are thought to arise from the purification process, which can produce toxic byproducts. Recently, there has been growing scientific interest in using medicinal plants, herbal extracts, and natural bioactive compounds instead of isolated alternatives. This shift aims to create safer, more accessible treatments for various diseases while preserving traditional herbal medicine knowledge. Herbal extracts are formulations made from natural sources, mainly plants, containing bioactive ingredients that offer biological benefits [138]. In developing nanofibers for smart food packaging, plant extract-based compounds are among the most commonly used bioactive ingredients.

Based on recent journals, tea-based extract was the most common plant used for smart food packaging development. Some other plants were used, such as pomegranate and barberry. Shahbzi *et al*. [139] developed carboxymethyl cellulose-gelatin (CMC-GE) nanofibers and used *Mentha longifolia* L. essential oil (MEO) as its bioactive. According to the result, the morphological nanofiber showed a uniform and beads-free structure. Furthermore, The incorporation of MEO into the CMC-GE nanofibers resulted in relatively beaded nanofiber morphology and increased the diameter of nanofibers. This result is due to the spinning solution having higher surface tension and electrical conductivity after adding MEO and changing the morphology from uniform-smooth nanofibers to nanofibers with beaded strands. In the thermal stability result using TGA analysis, the addition of MEO showed the reduction of thermal stability of the nanofiber compared to the MCM-GE nanofiber. It can be caused by the addition of MEO disrupting intermolecular interaction in the polymer matrix, inhibiting cohesive structural integrity and yielding a weaker polymer network.

Next, Liu *et al*. [107] fabricated nanofiber film using polylactide (PLA), butterfly pea flower extract (BPA), and cinnamaldehyde (CIN). The result showed that the microstructure was beads-free, and the diameter of the fiber increased with the higher concentration of BPA. Align with that, the porosity size of the fiber was reduced due to BPA increasing the viscosity of the spinning solution. In contrast, adding cinnamaldehyde (CIN) reduced the fiber diameter after adding BPA. CIN, as a liquid form, can reduce the viscosity of the solution, leading to a thinner fiber diameter.

Furthermore, the contact angle result of all samples showed hydrophobic characteristics at an angle above 90°. However, there was a reduction in the angle after adding BPA and CIN, although it was not significant. On the other side, the mechanical properties of the fiber were improved with the addition of BPA and CIN. This was shown by increased tensile strength and elongation at break after the addition. Improvements may come from additional interaction with the materials.

Ullah *et al*. [140] developed smart food packaging material using a combination of synthetic and natural polymers as they used zein and polycaprolactone. Then, they incorporated *Aster yomena* extract-loaded halloysite nanotubes (*A. yomena*-HNT) as bioactive nanofibrous food packaging. The morphology result showed that the fiber structure was flat ribbon-like morphology. The distinct characteristic was that the diameter of the fiber was highly heterogeneous, which can be due to the low Taylor cone stability at the needle tip during the spinning process. After adding HNT-loaded *A. yomena*, the fiber's diameter increased as the solution's viscosity also increased. Next, the thermal stability analysis revealed that the inclusion of HNT-loaded *A. yomena* increased the onset of thermal degradation, thus suggesting improved thermal stability. This was in line with the DSC result, as adding active compounds into the fiber gradually increased the melting temperature. Conversely, the mechanical strength analysis showed that the fiber with the active compound became stiffer than the fiber with only zein and polycaprolactone. Adding the active compound gave the fiber better tensile strength but worse in the strain parameter.

4.2.3. Inorganic materials.

In the field of nanofiber development, organic materials are generally more commonly used as active compounds due to their flexibility, ease of processing, and compatibility with various applications. However, inorganic active compounds have emerged as crucial supporting elements that significantly enhance the functionality and performance of nanofibers. While not typically serving as the main active compounds, these inorganic materials provide valuable properties such as mechanical strength, thermal stability, and unique optical or electronic characteristics. Additionally, integrating inorganic nanoparticles can improve the overall durability and efficiency of nanofiber-based systems [141]. As research advances, the synergistic combination of organic and inorganic components in nanofibers continues to unlock new potential, leading to innovative applications in fields including smart food packaging development.

Cetinkaya *et al*. [25] developed smart food packaging nanofiber using black elderberry (BE) extract, Au nanoparticles (AuNPs), and SnO_2 for Hake fish (*Merluccius merluccius*) fillets, while gelatin was used as the nanofiber matrix. The morphology result showed that the nanofiber produced was uniform and free of beads for gelatin and gelatin-BE extract. Furthermore, there was an increase in fiber size in the nanofiber containing BE. A similar result was also shown when AuNPs and SnO_2 were incorporated into the nanofiber. This may show that these compounds were encapsulated inside the nanofiber. The DSC thermograms of electrospun nanofibers revealed that adding BE, SnO_2, and Au shifted the melting temperature. The improvement in the regularity and compactness of polymer chains can be attributed to forming hydrogen bonds between gelatin, BE extract, and Au. The creation of cross-linking bonds among these components limits the mobility of the polymer chains, resulting in increased thermal stability. Furthermore, the TGA analysis supported this result, which showed that nanofiber with active compounds had a lower decomposition rate than polymer nanofiber.

On the other side, Torres-Giner *et al*. [126] developed Graphene nanoplatelets (GNPs) embedded in poly(ethylene-co-vinyl alcohol) (EVOH) fibers by electrospinning. According to the morphology result, the pure EVOH nanofiber showed homogenous, free of beads, and wide porosity structures. After the addition of GNPs to the fiber, the morphology showed that the diameter of the fiber decreased with the higher GNP concentration. This can be due to the increased conductivity of the solution. Furthermore, the incorporation of GNPs gave rise to the formation of certain beads. This preliminary observation suggests that GNPs may agglomerate in high concentrations within the EVOH. Furthermore, the thermal stability of the nanofiber showed that the addition of GNPs delayed crystallization as the crystallization temperature value decreased with the existence of GNPs. This suggests the embedded GNPs acted as an anti-nuclear agent, particularly at low contents due to their improved dispersion.

Râpă *et al*. [108] fabricated poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) and PLA, which were combined with Fe-doped ZnO nanoparticles (NPs). The morphology result showed that both polymers only (PHBV and PLA) and polymers with Fe-doped ZnO NPs showed uniform and beads structure. However, the beads increased significantly at the highest Fe concentration at 1%. Furthermore, incorporating ZnO nanoparticles into the PHBV matrix enhances the mechanical properties of the nanofibers. Specifically, the tensile strength and elongation at break increase with adding ZnO, indicating improved flexibility and strength. To support the SEM result, EDX was used to determine the elemental composition of the nanostructures. The analysis revealed the presence of Zn, O, and C as the main constituents of the PLA/PHBV/ZnO nanostructures.

4.2.4. Others.

In the development of functional nanofiber, many active compounds are used, such as protein or colorant agents. Souri *et al*. [142] developed smart food packaging nanofiber using Persian gum (PG) and poly (ethylene oxide) (PEO) as the matrix and ε-polylysine as the active compound (ε-PL). The morphology result showed that the fiber was not significantly different between polymer only and polymer/ε-polylysine. However, nanofiber with incorporated ε-PL showed less ribbon structure than polymer-only nanofiber. Furthermore, the XRD analysis assessed the nanofiber's crystallinity degree. The result showed that the crystallinity peak was higher with increased PEO content. In contrast, PG is an amorphous polymer that reduces the nanofiber's crystallinity peak. Moreover, the addition of ε-PL did not change the XRD result. In a similar way, the inclusion of -PL in the solutions did not lead to major changes in the viscosity of the spinning solutions. The influence of -PL on the viscosity of biopolymers largely depended on the electrical charge of the hydrocolloids.

Moreover, Jiang *et al*. [143] developed polyvinyl alcohol (PVA) and polyacrylate sodium (PAAS) nanofiber, which added nisin nanoemulsions (EN) as active compounds. They fabricated a nanoemulsions system to enhance the stability and solubility of nisin in the nanofiber. The result showed that PVA/PAAS nanofiber showed uniform structure without cracking or bead formations. After the addition of EN, there were small bumps in the structure, which corresponded with the immobilization of the EN. Furthermore, the fiber size also increased compared to the PVA/PAAS nanofiber. Next, the addition of EN significantly influenced the mechanical properties of the nanofibers ($p < 0.05$). The tensile strength (TS) values increased with higher EN content. This increase in EN content led to strong hydrogen bonding between the functional groups in EN and the hydroxyl groups in PVA and PAAS. However, at very high EN levels, aggregation occurred, reducing surface free energy,

weakening certain nanofiber sections, decreasing hydrogen bonding among EN, PVA, and PAAS, and ultimately reducing TS. The elongation at break (EAB) of PVA/PAAS nanofibers also increased with EN content, reaching a maximum of 15% EN before significantly decreasing. Strong hydrogen interactions and enhanced aggregation likely impeded chain movement at the highest EN concentrations, reducing plasticizing effects and nanofiber flexibility. These observations suggest that EN addition improved the nanofibers' mechanical properties, particularly TS and EAB.

He *et al*. [144] fabricated polycaprolactone/gelatin/zein nanofibers containing alizarin and thymol as a protein-based halochromic nanosensor for smart food packaging. Halochromic or pH-sensitive sensors, such as those containing alizarin, are indicators that change color based on the surrounding pH levels. These sensors are non-destructive and easy to manage, offering immediate visual cues about the quality of a product. Based on the result, adding alizarin might alter the solution's viscosity, but it does not impact the morphology or structure of the nanofibers. According to the thermal property result, the degradation of the nanofiber occurred at 160–380 ◦C, and alizarin addition had little effect on the thermal resistance of the nanofiber.

Table 2. Active compounds incorporated in nanofiber.

Active compound		**References**
Essential oils	*Zataria multiflora* essential oil	Moradinezhad *et al*. [131], Beikzadeh *et al*. [46], Ebrahimzadeh *et al*. [132], and Raeisi *et al*. [79]
	Cinnamon essential oil	Zhang *et al*. [133], Nazari *et al*. [86], Shao *et al*. [83], Feng *et al*. [82], Wen *et al*. [99], and Wen *et al*. [134]
	Thyme essential oil	Min *et al*. [135], Aytac *et al*. [136], and Lin *et al*. [19]
	Green tea essential oils	Maroufi *et al*. [80]
	Oregano essential oil	Shi *et al*. [113]
Plant extract	*Mentha longifolia* L. extract	Shahbazi *et al*. [139]
	Butterfly pea flower extract	Liu *et al*. [107]
	Aster yomena extract	Ullah *et al*. [140]
Inorganic material	AuNPs	Cetinkaya *et al*. [25]
	SnO_2	Cetinkaya *et al*. [25]
	Graphene nanoplatelets (GNPs)	Torres-Giner *et al*. [126]
	Fe-doped ZnO nanoparticles	Râpă *et al*. [108]
Others		Souri *et al*. [142], Jiang *et al*. [143], He *et al*. [144]

5. Nanofiber Improvement

Nanofiber fabrication, primarily through electrospinning, has garnered significant attention due to its potential in various applications, including biomedical, environmental, and food packaging sectors. Traditional electrospinning techniques utilize a single jet to create nanofibers from a polymer solution. However, recent advancements in this field have led to the development of several innovative methods to improve nanofibers' quality, functionality, and performance (Table 3). This part explores these advancements, focusing on multilayer film fabrication and post-treatment preparation.

5.1. Multilayer film.

Nanofiber with multilayer films has been developed in recent years. This multilayer is used to optimize each part to conduct its function. Alonso-González *et al*. [115] synthesized glucose oxidase (GOX) enzyme immobilized polyvinyl alcohol (PVOH) nanofiber. Then, the layer was protected with two hydrophobic polycaprolactone (PCL) membranes (multilayer system) for food packaging applications. The enzyme was used as an antioxidant and antimicrobial agent. According to the morphology result, the multilayer system containing the enzyme showed a more uniform fiber diameter size distribution compared to each single polymer. On the other side, the enzyme encapsulation led to a decreased stress-strain value. This suggests that the composite materials were easier to fracture than each single polymer PVOH and PCL.

Conversely, Shi *et al*. [30] fabricated food packaging nanofiber using coaxial electrospinning to create core and shell-structured nanofiber. Cellulose acetate (CA) was used as the shell layer, which has excellent water stability, while gelatin (Gel) was used as the core layer to encapsulate the active compound (eugenol (Eg)). Based on the result, all the nanofiber films have a typical porous and smooth structure—furthermore, the addition of Eg decreases the nanofiber diameter. The results might be attributed to physical interactions and hydrogen bonding between essential oil and gelatin molecules. Based on thermal stability, all samples showed three degradation stages, which revealed a similar trend. However, the nanofiber with Eg content was more vulnerable to high temperatures because Eg has lower thermal degradation. Next, as the concentration of Eg increased, the nanofiber diameter became smaller. The reduction in fiber diameter reduced the amount of air trapped at the interface between the fiber membrane and water, decreasing the water contact angle. CA/Gel-Eg nanofiber also exhibited higher elastic modulus and tensile strength compared to CA/Gel when Eg was incorporated into its core, indicating increased stiffness and decreased flexibility. They suggested that thinner nanofibers in the films led to improved mechanical performance.

Next, Zhang *et al*. [145] fabricated a colorimetric bilayer film for pork freshness detection and preservation using a layer with polyvinyl alcohol - sodium alginate - alizarin as the sensor layer and a layer with polyvinylidene fluoride - vanillin as the antibacterial layer. They developed hydrophilic and hydrophobic layers to enhance the stability of the food packaging sensor ability. Over an extended period, pH-sensitive dyes in the film may leach out, and the polymeric substrate could be compromised by water vapor, leading to color instability of the indicator. Hence, research is being undertaken to address this issue by incorporating a hydrophobic layer. The bilayer film nanofiber was injected by injecting the antibacterial layer and then the sensor layer into the first layer. The morphological result revealed that the antibacterial layer showed a grainy-porous structure, and the diameter was thinner than the sensor layer. In contrast, the sensor layer showed a smooth-cylindrical shape with a thicker diameter.

Furthermore, there were some beads in the sensor layer. The mechanical properties showed that the tensile strength (TS) and Elongation Break (EB) were better for bilayer film than its single film. Moreover, the fabrication bilayer film also improved the nanofiber's water contact angle, water solubility, and swelling index to meet the desired parameters for smart food packaging applications.

5.2. Post-treatment process.

Overall, the post-treatment process is conducted to enhance the nanofiber's physical ability, especially to enhance the hydrophobicity or water resistance effects. A crosslinking approach is used in this process, either by using additional compounds or only by other physical processes [146]. Li *et al*. [147] synthesized hordein (HO) and chitosan (CS) nanofiber with quercetin (QU) as an active compound. After fabricating the nanofiber, heat treatment was applied to it. The temperature was set at 90℃, 120℃, 150℃, and 180 ℃ with treatment for 6h. The times were set as 3h, 6h, 9h, and 12h with treatment at 150℃. Based on the result, the HO-QU-CS nanofiber film exhibited a hydrophilic surface before heat treatment, with water droplets rapidly spreading into the fiber matrices within the first 4 seconds. Following heat treatment at 90°C, the nanofiber film maintained a stable contact angle of 60°. Furthermore, the contact angle result kept increasing with the temperature increase until 150℃. Interestingly, the nanofiber morphology did not alter significantly after heating treatment, although the fiber diameter was reduced slightly by adding fiber branches after heat treatment.

On the other side, Jia *et al*. [70] developed allicin-loaded pea protein isolated/pullulan electrospun nanofiber films with Maillard reaction as the crosslinking approach. This approach harnessed a reducing sugar to build an interaction with the amino group as a crosslinker and was conducted under heat treatment at 120℃ for 3 h to fasten the process. According to the result, the morphology showed that adding glucose made the nanofiber merge and increase its diameter. In addition, glucose levels above 3% diminished fibrous structure, leading to a network of holes and bumps. The melting point (Tm) and degradation temperature of the nanofibers (NFs) with glucose cross-linking were higher than that of the sample without glucose. The findings suggested that the glucose-cross-linked nanofibers exhibited better thermal stability compared to the nanofibers without cross-linking. Furthermore, the water resistance of the nanofiber was increased with the glucose content dependent until 2%, which gave the highest contact angle value of around 104.5°. Furthermore, in up to 2% glucose concentration, the swelling ratio, water solubility, and water vapor transmission rate (WVTR) gradually decreased, indicating enhanced hydrophobicity and improved water vapor barrier properties.

Next, Goksen *et al*. [89] fabricated polyvinyl alcohol (PVOH) containing essential oils (EOs) from *Laurus nobilis* (LEO) and *Rosmarinus officinalis* (REO). Citric acid (CA) was used as a crosslinker by evaporation to the nanofiber at 170℃ for 10 min. After morphological analysis, incorporating EOs reduced the nanofiber's diameter, suggesting the conductivity enhancement of the solution after adding EOs. Furthermore, the annealing treatment increased sharply to the diameter of the fiber without CA. However, nanofiber with CA did not significantly increase the fiber diameter.

Table 3. Nanofiber improvement.

Nanofiber improvement	References
Multilayer film	Alonso-González *et al*. [115], Shi *et al*. [30], Zhang *et al*. [145]
Post-treatment process	Halim *et al*. [146], Li *et al*. [147], Jia *et al*. [70], Goksen *et al*. [89]

After the annealing process, the nanofiber was immersed in water to analyze the stability of the structures. Nanofiber without CA conducted a degraded fiber structure, while nanofiber with CA still can maintain the fiber structure, although some regions were also degraded. Next, the thermal stability using TGA analysis showed that the annealing treatment

improved the stability of the nanofiber. However, the addition of EOs and citric acid decreased the thermal stability compared to the PVOH-only nanofiber.

6. Conclusion and Future Perspective

Integrating electrospinning nanofibers into smart food packaging represents a groundbreaking approach to enhancing food preservation, safety, and overall quality. By leveraging the unique properties of nanofibers, such as their high surface area, porosity, and customizable nature, researchers have been able to design packaging solutions that offer superior performance compared to traditional materials. The careful selection of both natural and synthetic polymers, such as chitosan, cellulose, PVA, and PLA, in combination with bioactive compounds like essential oils, plant extracts, metal-based agents, and phenol-based compounds, has enabled the development of packaging materials with enhanced antimicrobial, antioxidant, and barrier properties. These advancements are particularly crucial for extending the shelf life of food, preventing spoilage, and ensuring that food remains fresh and safe during storage and transport. Furthermore, incorporating multilayer films and post-treatment processes has opened up new possibilities for enhancing the mechanical strength, thermal stability, and environmental resistance of nanofiber-based packaging. As a result, the packaging industry now has access to more flexible, functional, and sustainable materials that meet the growing demand for environmentally friendly and high-performance packaging solutions.

Despite these advances, several challenges still impede the widespread adoption of electrospun nanofibers in food packaging, particularly at an industrial scale. One of the most significant hurdles lies in the scalability of the electrospinning process itself. While lab-scale production has demonstrated promising results, scaling up the technology to meet the demands of mass production remains a technical challenge. Issues such as low production rates, high costs, and difficulties maintaining consistent fiber morphology and functionality must be addressed for the technology to become economically viable for commercial applications. Additionally, regulatory safety concerns regarding the potential migration of nanomaterials into food require further investigation. Ensuring that nanofiber-based packaging is safe for consumer use is essential, and rigorous testing and compliance with food safety standards will be critical to gaining regulatory approval and consumer trust.

Future research must focus on overcoming these technical and regulatory challenges. One promising direction is the development of eco-friendly, biodegradable materials that meet the necessary safety standards and address the growing environmental concerns surrounding plastic waste. Natural polymers such as cellulose, alginate, and starch, combined with non-toxic, sustainable electrospinning processes, offer a pathway toward more environmentally responsible packaging. Additionally, smart packaging functionalities represent an exciting frontier in food packaging innovation. Integrating nanosensors, biosensors, and intelligent indicators into nanofiber matrices could enable real-time monitoring of food conditions, such as temperature, humidity, gas levels, and microbial contamination. These intelligent systems could alert consumers to spoilage or changes in food quality, reducing food waste and improving food safety. Furthermore, multifunctional nanocomposites that combine preservation properties with active sensing capabilities could lead to packaging solutions that are passive barriers and active participants in food preservation.

In conclusion, the future of electrospun nanofibers in smart food packaging is filled with potential for innovation and impact. By addressing the current challenges in scalability,

cost-effectiveness, and regulatory safety and by continuing to explore new materials and functionalities, electrospun nanofibers have the potential to revolutionize the food packaging industry. With continued research and development, these materials could play a key role in creating smarter, safer, and more sustainable packaging solutions that meet the needs of both consumers and the environment.

Author Contributions

Conceptualization, M.M.J.; E.M. and A.K.; methodology, M.M.J.; E.M. and A.K.; writing—original draft preparation, M.M.J.; E.M. and A.K.; writing—review and editing, M.M.J.; E.M. and A.K.; All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest

The authors declare no conflict of interest.

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