Current issues and potential solutions for the electrospinning of major polysaccharides and proteins: A review

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Abstract

Biopolymers, especially polysaccharides and proteins, are the promising green replacement for petroleum based polymers. Due to their innate properties, they are effectively used in biomedical applications, especially tissue engineering, wound healing, and drug delivery. The fibrous morphology of biopolymers is essentially required for the effectiveness in these biomedical applications. Electrospinning (ES) is the most advanced and robust method to fabricate nanofibers (NFs) and provides a complete solution to the conventional methods issues. However, the major issues regarding fabricating polysaccharides and protein nanofibers using ES include poor electrospinnability, lack of desired fundamental properties for a specific application by a single biopolymer, and insolubility among common solvents. The current review provides the main strategies for effective electrospinning of the major biopolymers. The key strategies include blending major biopolymers with suitable biopolymers and optimizing the solvent system. A systematic literature review was done to provide the optimized solvent system of the major biopolymers along with their best possible biopolymeric blend for ES. The review also highlights the fundamental issues with the commercialization of ES based biomedical products and provides future directions to improve the fabrication of biopolymeric nanofibers.

Keywords:

Biopolymers; Electrospinning; Nanofibers; Biological macromolecules

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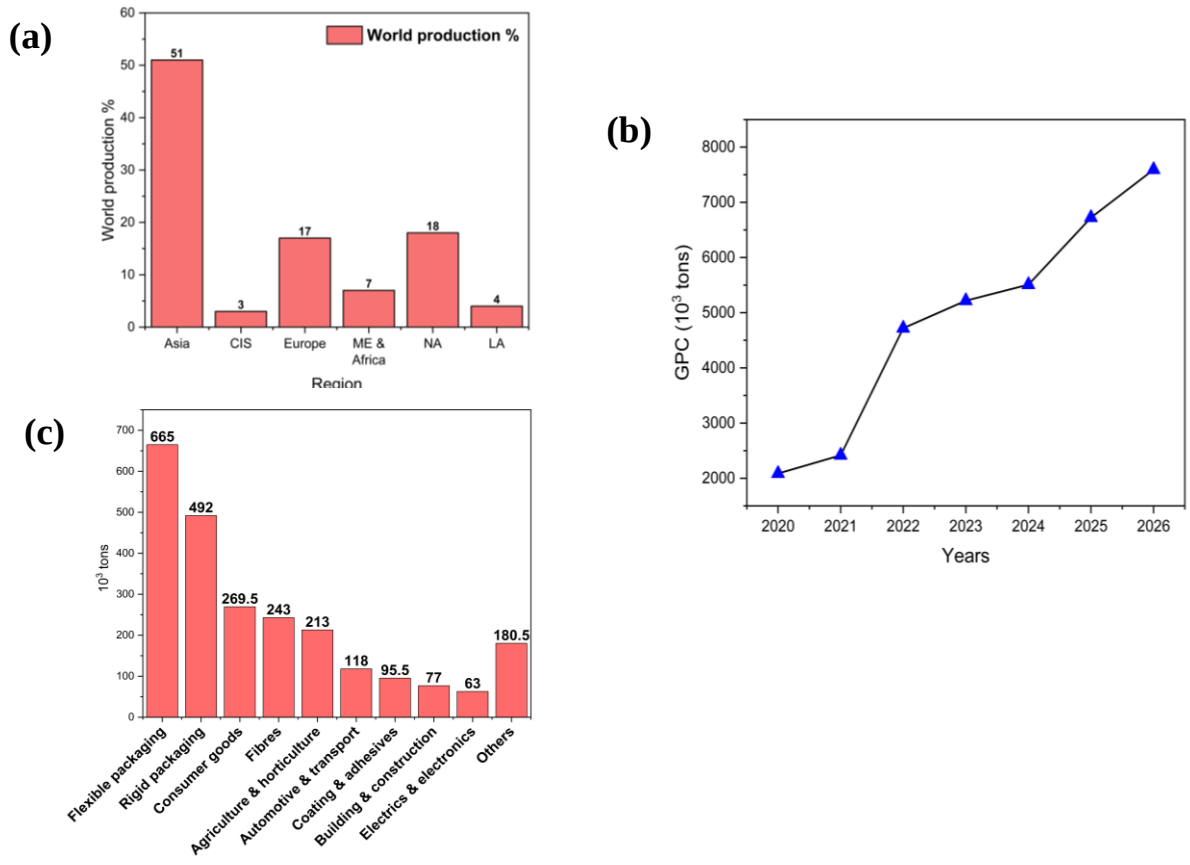
1. Introduction

In the mid-20th century, significant policy changes were made to discourage using petroleum-based polymers. The main reasons for this initiation were the recycling and hazardous issues associated with petroleum based polymers [15]. However, the industry continues to grow as it employs almost 1.6 million people in Europe, and the continent's total annual revenue is roughly 360 billion euros [16]. There has been a progressive rise from 2 million tons of plastic manufacture in 1950 to 380 million tons in 2015; just 18% of this was recycled. An estimated 12.5 million tons of plastic made their way into the ocean in 2010, mainly from nations around the coast [19]. Fig. 1a shows the percentage of world percentage production of plastics based on different regions [20].

Irrespective of the production and growth of the plastic industry, synthetic polymers are becoming a constant threat to the environment [21]. Due to their nondegradable nature, these plastic materials poison the environment when disposed of in landfills or incinerators, whereas tertiary recycling methods include chemical, thermochemical, and pyrolysis [22]. However, reusing all petroleum based plastics is difficult because of their environmental toxicity and recycling difficulties [23]. Above all, these petroleum based polymers are not suitable at all for any biomedical application since they are toxic and non-biodegradable. All these issues caused the researchers to look for alternatives [24]. Researchers increasingly focus on environmentally benign, biodegradable, and readily accessible materials instead of petroleum based polymers. The most promising alternatives to these petroleum based polymers are biopolymers [25]. In the past decade, a sharp increase in industrial and research fields has been observed for biopolymers (Fig. 1c). Biopolymers are the polymers produced by living things and can be degraded biologically. Biopolymers are taking over the market from synthetic polymeric materials. These

compounds, such as biopolymers, bioplastics, vitamins, and organic acids, may be made from natural resources using biotechnology [26].

About 1.1% of plastic output worldwide comes from bioplastics, with the European Union (EU) leading the charge [32]. Biopolymers have seen a steady rise in investment over the last several years. A recent survey shows that 1% of the world's plastics are produced from biopolymers [27]. The report also estimated that biopolymers' global production capacity (GPC) would be increased by 214.15% by 2026 (Determined by the authors based on data https://ect-center.com/blog/biodegradable-polymers) (Fig. 1b).



***CIS: Commonwealth independent states; ME: Middle east; NA: North America; LA: Latin America**

Fig. 1 Production comparison of polymers, (a) Plastic production in major regions, (b) Prediction of biopolymers production, (c) Utilization of biopolymers in different industrial sectors

Biopolymers can be divided into three distinct classes depending on the origin of their raw materials. The three classes include (1) natural biopolymers, (2) synthetic biopolymers, and (3) microorganism-based biopolymers (Fig. 2) [28]. Some of the most diverse polymeric macromolecules with exceptional structural and metabolic properties may be found in microorganisms, including bacteria, fungi, yeasts, molds, smuts, and many more forms of what we consider "primitive life" [29]. These biopolymers comprise various polysaccharides and proteins, including cellulose, chitin, chitosan, dextran, hyaluronic acid, silk, keratin, and many more. For a long time, plant-based polysaccharides and proteins have been valuable and sustainable resources [30]. All living organisms ranging from microorganisms to highly evolved terrestrial, aquatic, or aerial animals, may produce natural structures with great potential in biomedicine [31]. These structures are present in a large variety, from glycosaminoglycans (such as chitin and hyaluronic acid) and proteoglycans to proteins (majorly collagen, elastin, gelatin) to the genetic material deoxyribonucleic acid (DNA) [32].

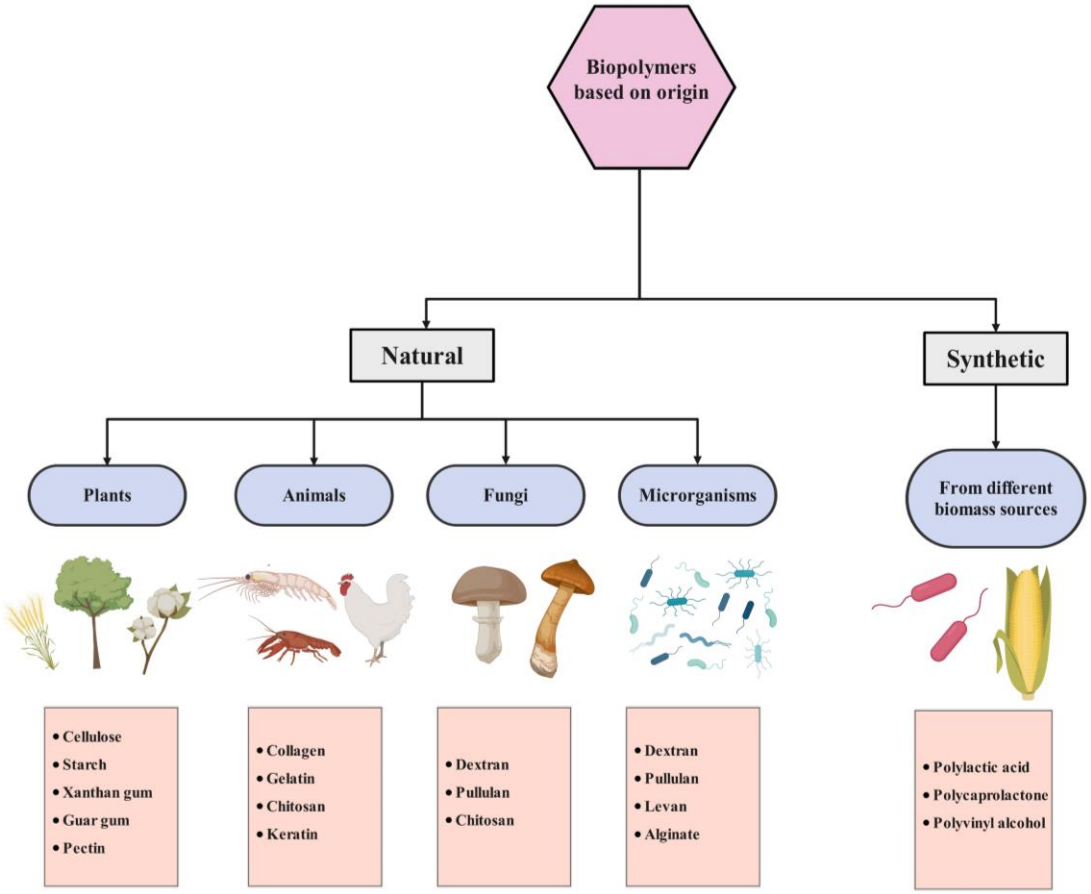


Fig. 2 Types of biopolymers based on the origin

This review focuses on the electrospinning of major biopolymers and highlights the issues and the potential solution of each biopolymer. The review highlights three main strategies to overcome biopolymers ES, including chemical modifications, blending with other biopolymer, and optimizing the solvent system for the major biopolymeric combinations. The review also highlights the ES insights of the two unexplored polysaccharides, levan, and kefiran, which could be useful in future biomedical applications.

1.2 Biopolymers in biomedical applications

Biopolymers occur naturally in the human body and play vital roles, from enclosing cells to cell communication. They regulate the skin's moisture and elasticity to keep it in its native state.

They lubricate body joints and the digestive system and prevent infection by building up mucus linings in the eyes and airways of humans [33]. All the significant events at the cellular level of mammals revolve around the extracellular matrix (ECM), as it contains collagen and elastin. It provides a high surface area based habitat for cells and tissues by providing cell adherence, controlling cell signaling, and promoting cell differentiation [34]. Hence, mimicking the natural ECM is the fundamental requirement of biomedical applications [35].

The most challenging part of fabricating a scaffold for tissue engineering is ECM replication and creating a tissue-specific scaffold that meets the requirements of natural biocompatibility, biodegradability, and high mechanical strength [36]. The other additional requirements of the biopolymeric scaffolds include signal transportation for maintaining cell growth, skin hydration, and pliability maintenance, a barrier for pathogens protection, less friction via lubrication, and so on [37, 38]. However, the conventional techniques lack the fundamental mimicking of the ECM [39].

The essential prerequisite for any drug delivery system is the controlled release of a therapeutic agent. In addition, the drug carrier should prevent the therapeutic substance from being degraded by enzymes before it is released slowly [40]. The drug carrier must be nontoxic, biodegradable, and, most critically, should be released from the body without any adverse effect after effective delivery. However, most biopolymers lack natural interaction, and the system lacks mechanical strength to survive in the physiological environment [41]. Conventional drug delivery systems usually use a crosslinker to overcome this issue, but the crosslinkers are mostly toxic, limiting the options for developing a drug delivery system [42].

Biopolymers also play a significant role in wound healing by replacing traditional dressings. Since traditional wound dressings were made from simple coverings, such as plant fibers or animal fats, and were used to prevent further bleeding [37]. Modern technological advances have

allowed the fabrication of artificial materials for wound dressings with a wide range of applications. Quick hemostasis and antimicrobial efficacy are two must-haves for every contemporary wound dressing. The primary role of wound dressing is to stop bleeding, which must be antibacterial to prevent infections from spreading [43, 44]. However, for treating major wounds, a sustained drug release and high surface area to volume ratio to promote growth and differentiation of the cells is the major requirement. Conventional wound healing dressings mostly lack these properties owing to the methods for fabrication [45].

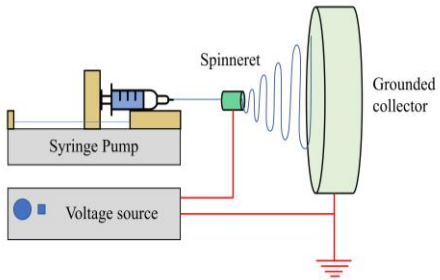
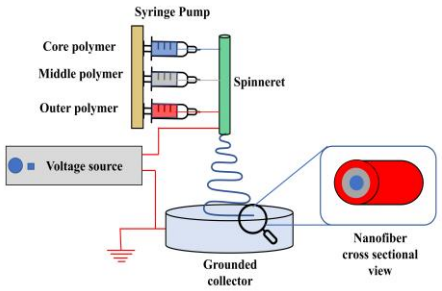
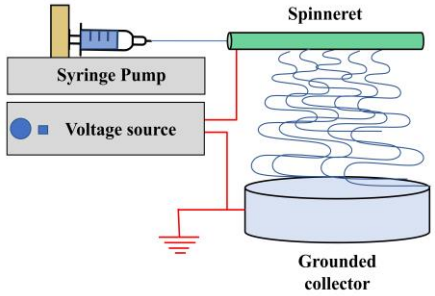
1.3 Electrospinning

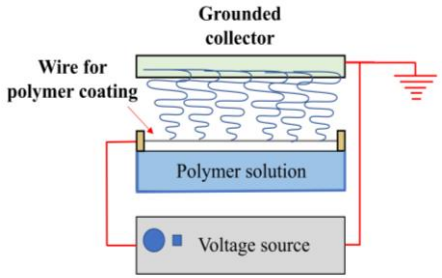
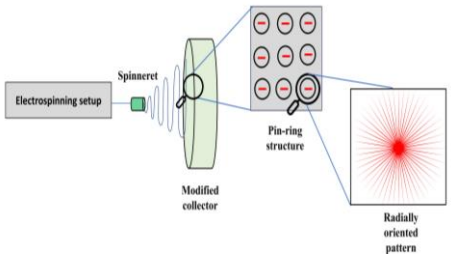
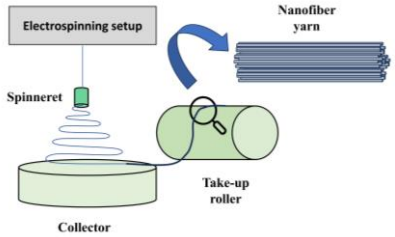
The major issues highlighted in all three biomedical applications can be resolved using an advanced, more flexible method to fabricate fibrous morphology [46]. Since the fibrous morphology provides cellular-control properties, high porosity, better mechanical performance, and high surface to mass and volume ratios compared to alternative morphologies [47]. Scaffold-cell interactions may occur over a more extensive region due to the high surface to area and volume ratio. The manufacturing procedure determines the fiber diameter, which may range from nanometers to millimeters, allowing for the regeneration of multi-layered tissues [48]. The potential biological uses of fibrous scaffolds are broadened by their malleable nature, the simplicity with which their structures can be altered, and the precision with which they may be shaped [49].

There are many techniques for the fabrication of nanofibers. However, the most promising and flexible technique to fabricate nanofiber is electrospinning (ES). ES uses the blend of biopolymers in a solvent system and fabricates nanofibers with the help of voltage. It provides the flexibility of using a unique combination of biopolymers without any natural interactions, which was challenging to achieve with old methods [50]. Incorporating unique combinations of

biopolymers provides the nanofibers with many critical properties crucial for biomedical applications [51]. Many ES techniques are currently being used to fabricate different nanofibrous morphologies for various biomedical applications. Some major ES techniques are listed in Table 1, along with their recent applications, and Fig. 3 shows the various morphologies of biopolymers that can be achieved by using electrospinning and can be utilized for a particular application.

113 **Table. 1 Major strength and drawbacks of various electrospinning techniques**

Electrospinning methods	Scheme of setups	Strengths	Drawbacks	Recent applications	References
Single needle (emulsion/ blend)		High and uniform drug distribution, a high initial burst	The bioactivity of therapeutics is lost due to due voltage and solvent; an initial high dosage burst can cause toxicity	1. Chitosan/ polyhydroxy butyrate based nanofibers for tissue engineering 2. Alginate/ chitosan and polycaprolactone based nanofibers for capsaicin drug delivery for cancer treatment	[52, 53]
Co/tri axial		Hollow fibers formation, two separate solutions for the outer and inner core, saves the therapeutic from the solvents to dissolve biopolymers; the final structure is optimal for sustained drug delivery since the encapsulation of soluble drugs in core	Consideration of variables like interfacial tension and viscoelasticity makes the procedure designing complex	Polycaprolactone/chitosan/poly vinyl alcohol shell with resveratrol loaded core for tissue engineering	[54]
Multineedle		Enhanced rate of production	Non-uniform fiber production, electrostatic interaction of needles which are side by side	No recent potential biomedical application was observed	[55]

Needleless		Superior production rate as compared to needle based techniques, no spinneret clogging issue, viscous natured biopolymers can be used	High voltage, non-uniform diameter distribution of fibers, high cost, and random generation of taylor cones	Gelatin/ polycaprolactone based nanofibers for ibuprofen sustained release	[56]
Radial nanofibers		Fabricated nanofibers have specific patterns providing biological cues for cells growth and differentiation	The process used a ring and pin structure in the collector while the area without the structure still has random nanofibers, so overall, the technique is complex with low throughput	Polylactic acid/gelatin based radially aligned nanofibers for chronic wound healing	[57]
Yarn based nanofibers		The resultant nanofibers show exceptional mechanical strength, surface area and pores mimicking natural extracellular matrix for cell culture	The process is complex as it requires specialized and complex collectors and is not suitable for all biopolymers, limiting the options	Polycaprolactone based nanofiber yarns for tendon repair	[56]

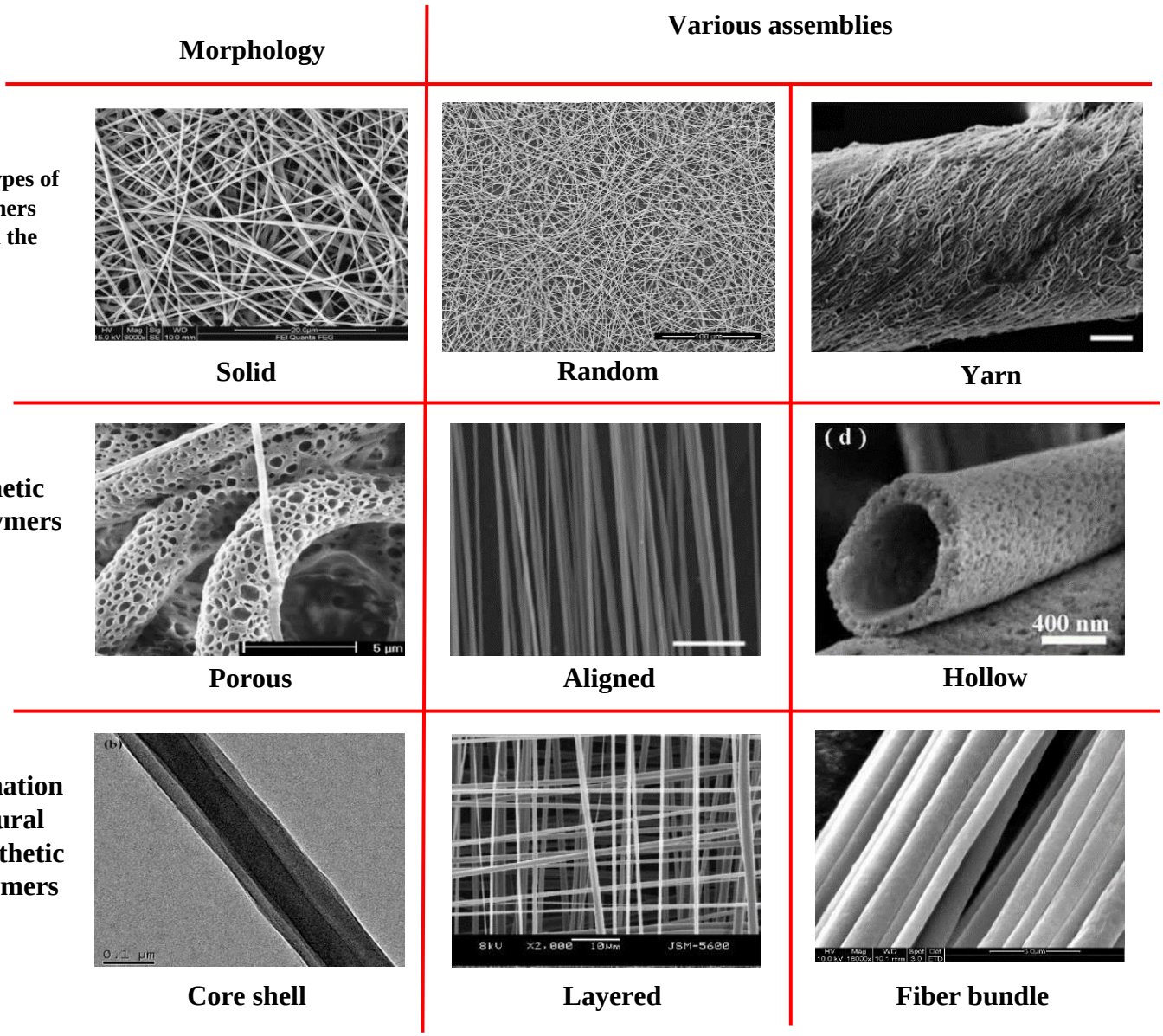


Fig. 3 Types of biopolymers based on the sources.

Fig. 3 Various morphologies and assemblies of natural, synthetic or combination of biopolymers [6]

1.4 Importance of biopolymeric nanofibers

For the application of tissue engineering, in addition to the previously mentioned features, biopolymeric nanofibrous scaffold also enhances cell adherence by including specific patterns, such as RGD (arginine, glycine, and aspartate tripeptide) [58]. All of the listed features encourage the growth and proliferation of tissues. The manufacturing process fabricating specific

fibers varying in size from nanosized to millimetric may be employed to recreate hierarchical tissues at various levels. Fig.4a shows the schematic representation of how the cells interact with various other factors and therapeutic agents in a nanofibers based scaffold. Fig. 4b shows the SEM micrographs of the nanofibrous yarns developed by Cai et al., (2012) [7]. The study employed human embryonic stem cell-derived mesenchymal stem cells to be developed in the scaffold. The cells changed from a round to a spindle shape, and at around 6 weeks mark a 3D bony tissue was observed around and inside the scaffold. The study showed how nanofibrous scaffolds can guide the development of bone formation.

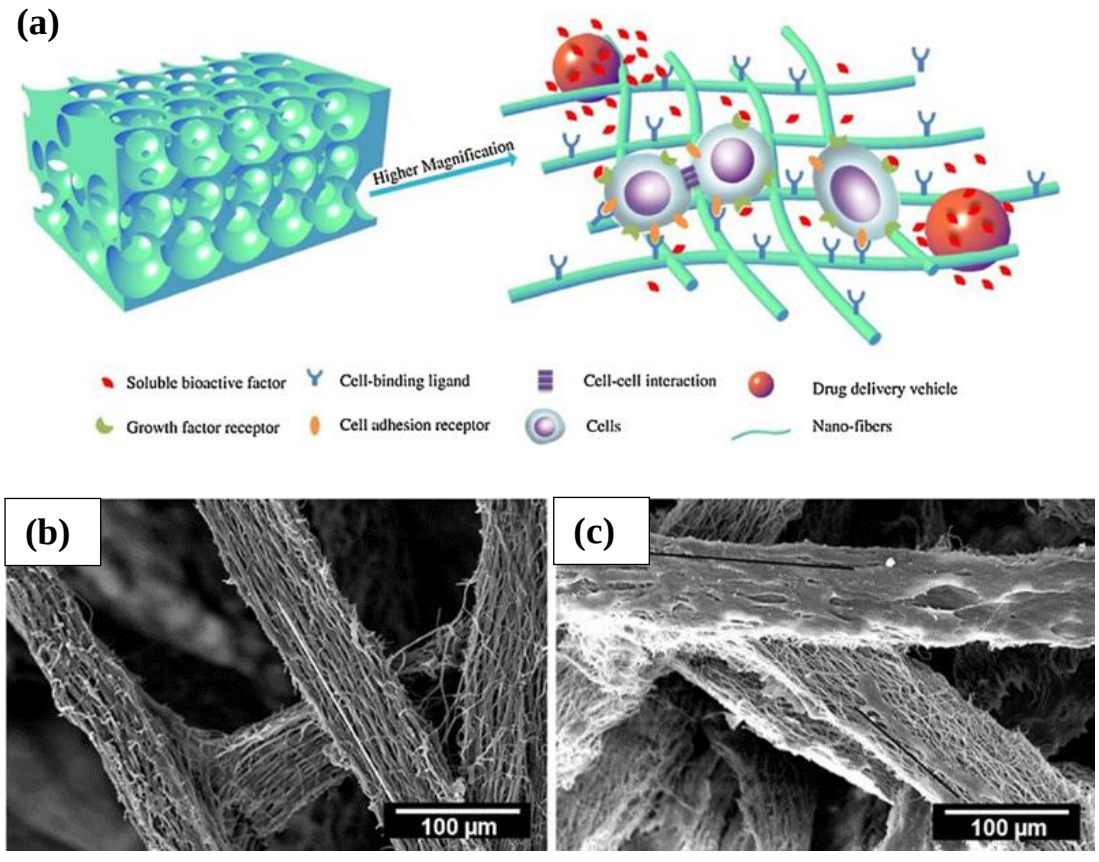


Fig. 4 Nanofibers based scaffolds for tissue engineering, (a) Schematic illustration, (b-c) SEM images at various magnifications [7]

For the application of drug delivery, nanofibers provide a potential solution for sustained drug delivery and overcoming the issue of burst release. The achievement is only possible due to the fibrous morphology, which increases the surface area to volume ratio [59]. The other more effective approach is encapsulating therapeutic to provide protection and sustained release. Coaxial electrospinning produces nanofibers with core-shell morphology and encapsulates drugs and other therapeutics in the core to form smart drug delivery systems [60]. Abdolbaghian et al., (2022) [2] developed PLA/PVA/PVP based core-shell nanofibers for sage drug delivery. The core consisted of the PLA and sage, while the shell comprised PVP and PVA (Fig. 5a & b). They compared drug release of the core, shell, and core-shell structures. Their results showed that the drug release was reduced by 35% in the core-shell structure compared to the dispersed drug loaded structures (Fig. 5c).

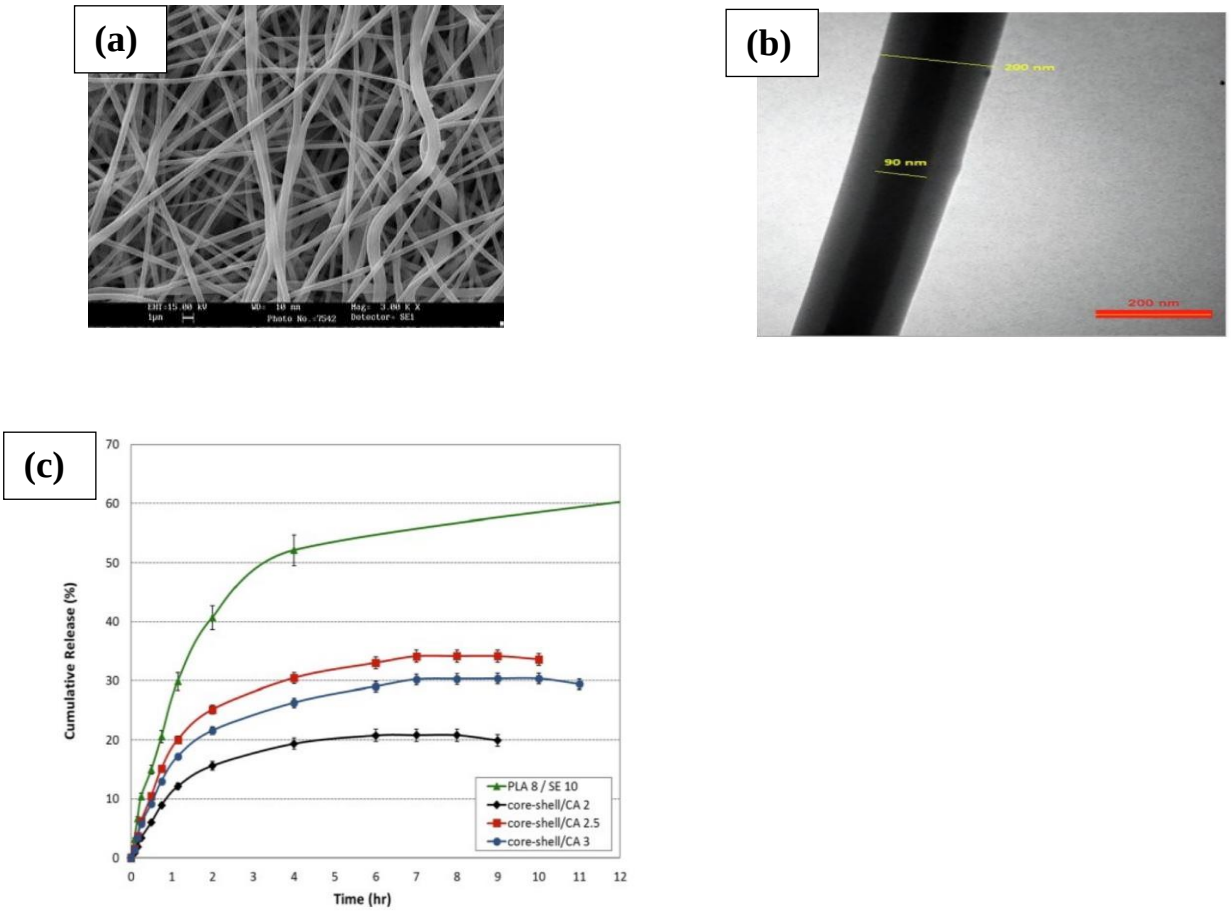


Fig. 5 Core-shell nanofibers based drug delivery system (a) Overall biocomposite SEM image, (b) Single nanofiber TEM image, (c) Drug release comparison between core-shell and simple morphology [2]

Nanofibers can also play a vital role in fabricating smart wound dressing since the ES allows the combination of any biopolymers, including the therapeutic, which helps advance wound healing [45]. The traditional dressing comprises polymers with no antimicrobial activities, but the biopolymeric nanofibers based dressing is innately antimicrobial. The other important concern about chronic wound healing is that the dressing can not be replaced daily due to the danger of wound infection and exhaustion of the therapeutic. This change of dressing results in disturbing the wound healing process. The nanofibrous based smart wound healing system reduces the frequency of dressing changes since there are fewer chances of wound infection, and with sustained drug release, the drug is not finished too early [61]. Due to the wide variety of options for fibrous scaffolds, the ability to adjust fiber orientation in the scaffold, and the flexible

structural modification, the field of biomedical applications has been greatly widened [62]. In a study by Wu et al. (2022) [57], a wound healing patch for diabetic patients was developed. The developed patches utilized the effect of patterns on the differentiation and growth of the cells. They fabricated nanofiber and hydrogel-based bi-layered dressing consisting of the radially aligned nanofibers of gelatin and polylactic acid in a radial manner (Fig. 6). The developed patches showed better cell recruitment and guidance ability for better differentiation of the human dermal fibroblasts (HDFS). Fig. 6a & b shows the graphical representation and fluorescent imaging of the migration of HDFS along the radially aligned nanofibers from day 3 to 7. Fig. 6c shows the cell viability test for the developed patches and confirms the successful differentiation environment mimicking the natural ECM.

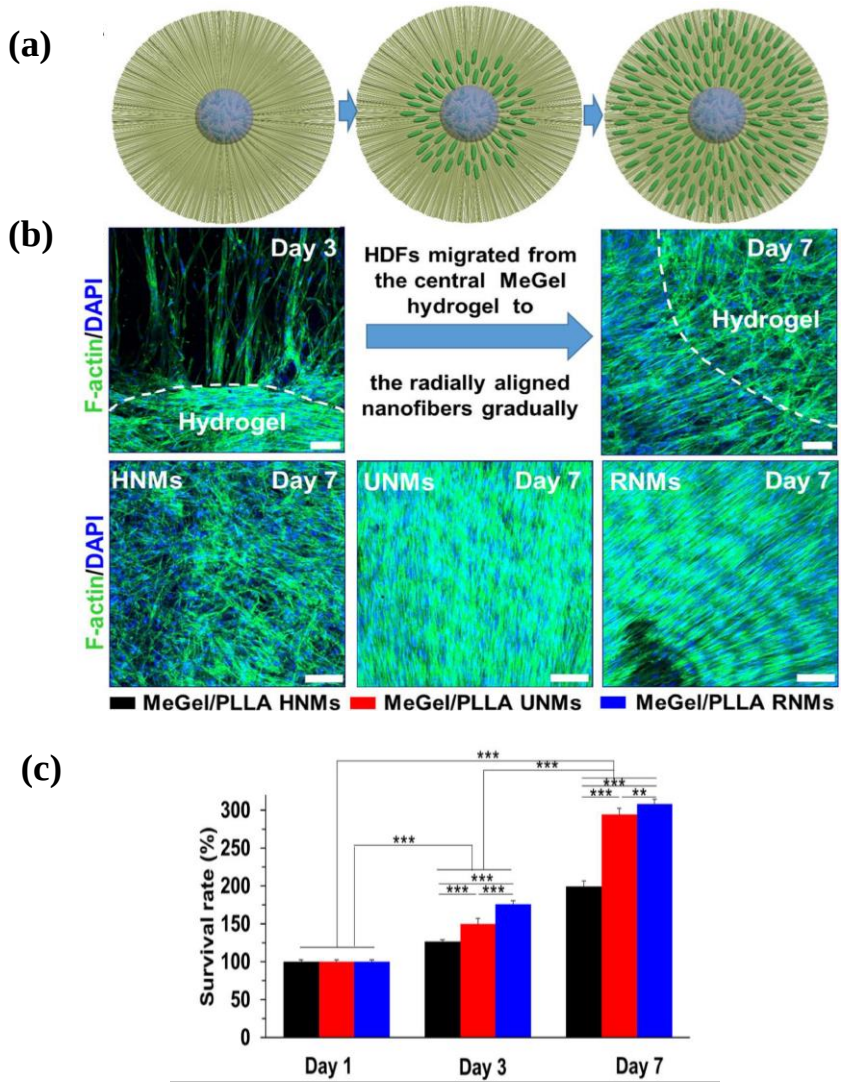


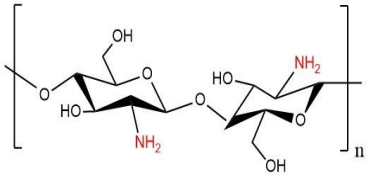
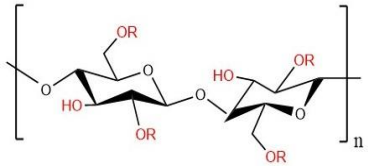
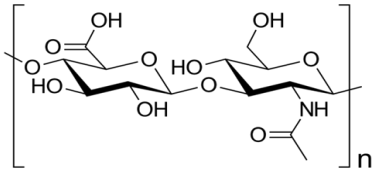
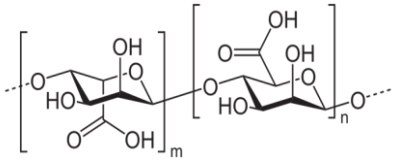
Fig. 6 Radially aligned nanofibers based wound healing patches, (a) Graphical representation, (b) Fluorescent microscope analysis, (c) Cell viability test [4]

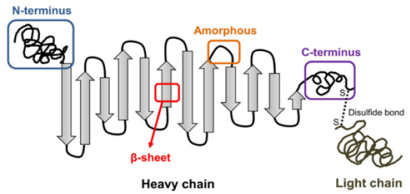
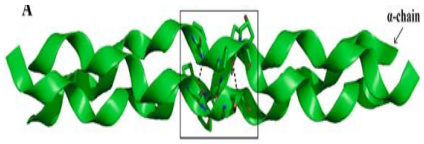
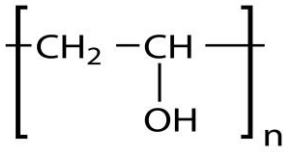
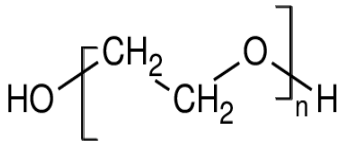
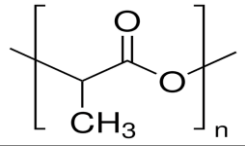
2. Major biopolymers for nanofibers fabrication

The major classes of biopolymers for major biomedical applications include polysaccharides, proteins, and phospholipids due to their abundance, cost-effectiveness, and high level biocompatibility [63]. However, using only a single natural biopolymer has various drawbacks and difficulties in forming a system for a biomedical application. The fundamental issue is that a single biopolymeric system cannot manifest desired properties for the nanofibers to accomplish

the objective of biomedical applications as they lack the whole spectrum of innate properties required by a particular system including biocompatibility, cell adherence, mechanical strength and antimicrobial effect [64]. The other major biopolymer issues are their insolubility and innate low electrospinnability potential. There are two main approaches to overcoming the electrospinnability and properties issue: biopolymers are chemically modified or blended with other biopolymers for ES [65]. The blending provides the resultant nanofibers with additional features for biomedical applications. Some of the major biopolymers used for ES, along with their merits and demerits, are listed in Table. 2

191 **Table 2. Merits and demerits of commonly used biopolymers in electrospinning**

Type	Biopolymers	Chemical structures	Advantages	Disadvantages	References
Natural	Chitosan		Incorporates antimicrobial & antioxidation activity, regulates inflammation, promotes hemostasis	Less solubility and biodegradation rate are uncontrollable	[66-68]
	Cellulose		Improves thermal stability and scaffolding for the therapeutics	Highly immunogenic, less solubility and limited water holding capacity	[69-71]
	Hyaluronic acid		Natural component of the body and actively involves in the clot formation, inflammation, proliferation, and re-epithelialization	Highly viscous even at low concentration	[72-74]
	Alginate		Structure similarity with extracellular matrix, abundant availibity and cost effective	High viscosity and immunogenic response	[75]

	Silk fibroin		Enhanced mechanical properties, rate of biodegradation is controllable, improve water, and oxygen permeability	Extraction process is tedious, and degumming affects mechanical properties	[76]
	Collagen		Less immunogenic, better biocompatibility, good for cell proliferation and adhesion	Sudden breakdown during degradability and extension	[77, 78]
Synthetic	Polyvinyl alcohol		Bio-adhesive, nontoxic and better chemical resistance	Bio-inertness and low mechanical strength	[79, 80]
	Polyethylene oxide		Good viscoelastic behavior in physiological environment, improves the electrospinnability as easy to blend with natural biopolymers	Non-biodegradable within the biological time frame	[81]
	Polylactic acid		Excellent electrospinnability, good mechanical strength	Hydrophobic in nature and can cause inflammation and results in immunogenicity	[82, 83]

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2.1 Influences of the solvent system in nanofibers production

The solubility issue of biopolymers is more complex than the other mentioned issues, as the solvent system should not only makes the homogenous blend but also fabricates nanofibers. Since it is evident in the next examples, sometimes the system will dissolve the biopolymers, but the nanofibers will not be fabricated, and the ES will result in bead formation or spraying. Each combination of biopolymers requires an optimized solvent system to fabricate nanofibers. The properties of the solvent system also play a crucial role in the fabrication of nanofibers and their morphology. The major properties of the resultant solvent system include the dielectric constant, polarity, surface tension, and viscosity [84]. Georgiadou et al., 2014 [3] showed the effect of the different solvent systems on the morphology of the same concentration of polylactic acid. Out of the six different solvent systems, all were dissolving PLA, but only acetone/dimethylacetamide formed the uniform nanofibers. The rest solvent systems either failed to fabricate the nanofibers or formed the beads (Fig. 7)

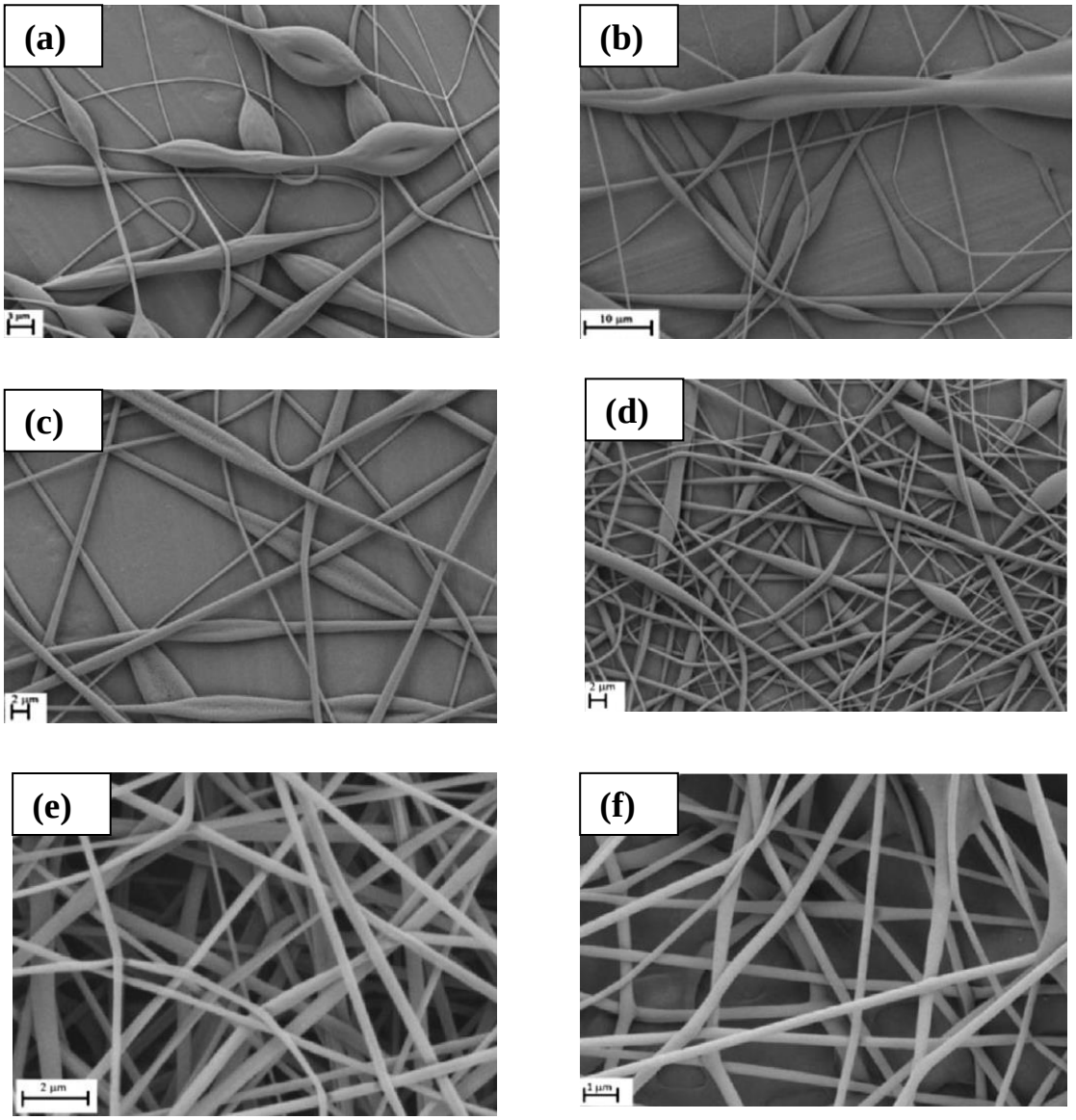

Fig. 7 Effect of different solvent systems on the nanofibers morphology of polylactic acid (10% w/v), (a) acetone/1,4-dioxane, (b) acetone/tetrahydrofuran, (c) acetone/dichloromethane, (d) acetone/chloroform, (e) acetone/dimethylformamide, (f) acetone/dimethylacetamide [3]

Another study by Choktaweesap et al. (2007) [6] demonstrated the effect of various solvent systems (Glacial acetic acid (GAA) (Fig. 8a), GAA/2,2,2-trifluoroethanol (TFE) (Fig. 8b), GAA/dimethyl sulfoxide (DMSO) (Fig. 8c), GAA/ethylene glycol (EG) (Fig. 8d), and AA/formamide (F) (Fig. 8e) on the morphology of gelatin NFs. They conducted the ES process with the same parameters (voltage: 7.5 kV and the distance between needle and collector 7.5 cm)

and the same biopolymer (gelatin) but changed the solvent system to see the direct effect of solvent systems on the final NFs. Different solvent systems resulted in the change in properties of the solution, like surface tension and conductivity, and hence the change in morphology and the diameter of the NFs was observed.

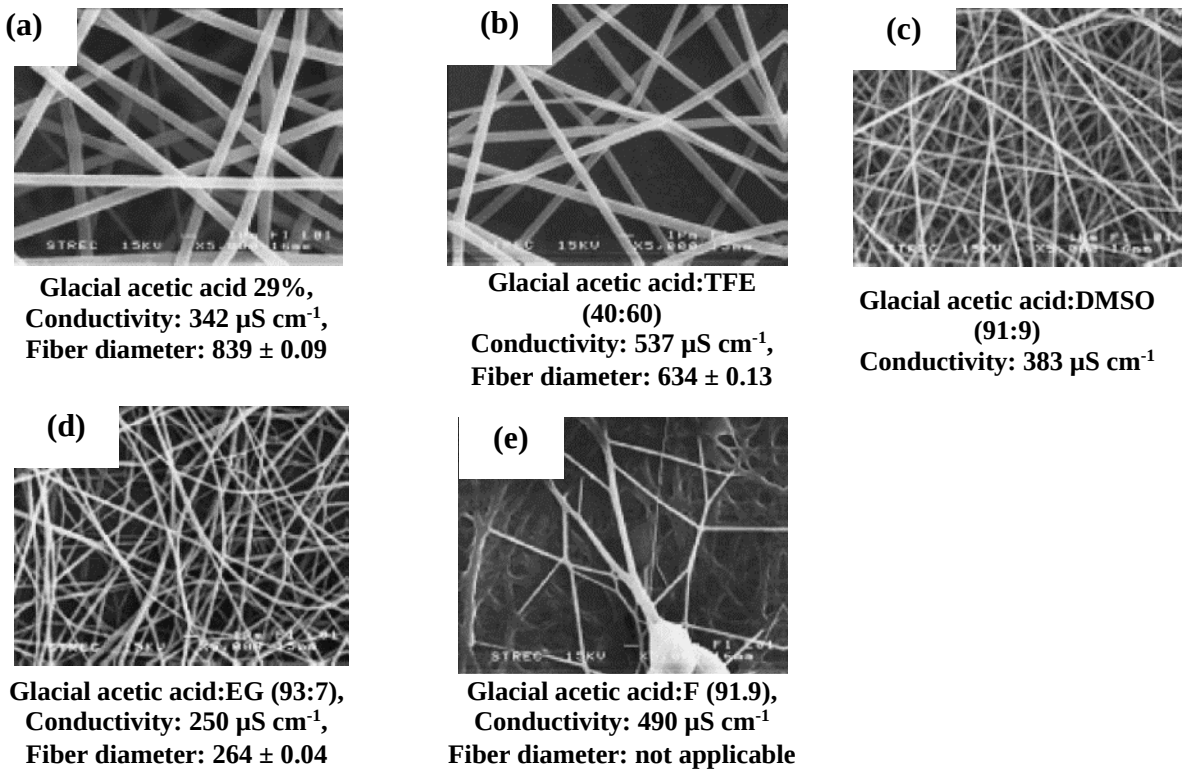

Fig. 8 Effect of different solvent systems on the gelatin nanofibers, (a) Glacial acetic acid, (b) GAA:TFE, (c) GAA:DMSO, (d) GAA:EG, (e) GAA:F [5]

The following section highlights the major issues with the fabrication of nanofibers for major biopolymers. It also provides potential approaches to solve those issues, which mainly include blending the biopolymer or developing an optimized solvent system. It also enlists the possible combinations of various biopolymers and the particular solvent system for a specific biomedical application.

2.2.1 Cellulose and their derivatives

According to the International Union for the Conservation of Nature (IUCN), cellulose is the most abundant biopolymer, which is a homopolymer of glucose units homopolymer which is connected by (1,4)-linkages [85]. Cellulose is biodegradable, bio-friendly, nontoxic, cost-effective, and has high mechanical strength and chemical resistance since insoluble in most polar and nonpolar solvents [86]. Cellulose source, the extraction process, polymerization (DP) degree, and treatment procedure may determine cellulose polymorphism (i.e., I, II, III, and IV). DP mostly depends on the source type, but the overall molecular structure stays constant and significantly impacts cellulose viscosity, mechanical characteristics, and dissolving properties. Thus, these three characteristics determine cellulose electrospinning and the application of its fiber form [87]. These will be the advantages of employing cellulose in the nanofibers. Developing technology for cellulose extraction from waste items such as food, agricultural, and textile waste further increases its sustainability and promotes a green environment [88].

Despite the many advantages of cellulose, the major hindrances are the non-solubility of cellulose. Hence, it is mainly modified into various derivatives that somehow solve the solubility issue and improves the chances of electrospinnability. Most alterations are made to cellulose for its better affinity for polar/nonpolar molecules [89, 90]. The advantages and disadvantages of significant cellulose derivatives for ES and their common solvent systems are listed in the Table. 3. Fig. S1 shows the morphology of the major cellulose derivatives blending with other biopolymers [8, 91, 92]

Table 3. Merits and demerits of common cellulose derivatives

Derivative	Advantages	Solvent system	Disadvantages	Potential applications	References
Cellulose acetate	Better thermal stability, solubility, biocompatibility	Acetone: dimethyl acetamide (2:1 v/v)	Low Maximum operating temperature (40°C), Low pH working range (3-6)	Drug delivery, wound healing, microfiltration	[93-95]
Cellulose acetate butyrate	Better orientation stability, chemical and moisture resistance, innately hydrophobic	Acetone: dimethyl acetamide (1:4 v/v)	Hydrophobic nature limits the application in the biomedical field	Oil absorption	[96]
Carboxymethyl cellulose	Enhanced biodegradability, Good interaction with carboxyl and amine groups in drugs	Distilled water	Rigid structure and high viscosity result in gel formation	Drug delivery, Food preservation	[97-99]
Hydroxy propyl cellulose	Better solubility in common solvents, especially water and methanol, constant fibers diameter	Chloroform and dimethyl formamide	Low mechanical strength	Transdermal drug delivery, sensors	[100, 101]

2.2.2 Lignin

Lignin is a highly preferred option for ES because of its low cost, less pollution, and availability of renewable sources (the second most abundant biopolymer in nature behind cellulose). Lignin is mainly derived from wood pulping byproducts as the secondary cell wall of higher plants contains lignin [102]. It provides mechanical stability, the polysaccharide binding site, and aids in resistance to pathogenic invasion [103]. Lignin is considered a phenolic biopolymer since enzyme based polymerization of three primary phenolic alcohols (monolignols-p-coumaryl, coniferyl, and sinapyl) produces plant lignin. Since lignin molecules are widely crosslinked with one other and different polysaccharides, the "native structure" is still unknown as it cannot be removed from plants in their natural state [103]. The significant properties of lignin include biocompatibility, biodegradability, antioxidant, and more chemical resistance than other

cellulose-based biopolymers. These properties have considerable potential for biomedical applications [104]. However, the major issues regarding the ES of lignin are low solubility, poor mechanical strength, impurities during the extraction process, low interaction with other biopolymer systems, and less viscoelasticity for ES [12]. It is mostly ES with other biopolymers to overcome the nanofibers fabrication of lignin. Some of the major biopolymers blended with lignin and their solvent systems are listed in Table 4. The SEM images of various biopolymers blended with lignin are shown in Fig. S2 [12, 105].

Table 4. List of commonly blended biopolymers with lignin for the electrospinning

Source	Commonly blended biopolymers	Solvent system	Potential applications	References
Natural	Chitosan	Glacial acetic acid (1.5% V/V or 5% v/v)	Food packaging, Ultrafiltration	[106, 107]
	Cellulose	Trifluoroacetic acid: dichloromethane (1:1 w/w)	Packaging, Filtration	[108, 109]
Synthetic	Polylactic acid	1,1,1,3,3,3-hexafluoro-2-propanol	Tissue engineering	[110]
	Polyethylene oxide	Dimethyl formamide	Tissue engineering	[111]
	Polyvinyl alcohol	Distilled water	Antimicrobial, Drug delivery	[112, 113]

2.2.3 Chitosan and Chitin

Chitin and chitosan (CS) have a repeating structure of (1,4)-b-N-acetyl glycosaminoglycan and its deacetylate derivative, respectively [114]. The chemical structure of chitin is almost identical to that of cellulose except for the organic group R, which in chitin is NHAc rather than OH. Crab, shrimp, prawn, and insect exoskeletons, as well as the cell walls of mushrooms, include the

most readily accessible animal derived biopolymer, chitin [115]. Chitin is produced yearly at 1011 tons but is mainly discarded in the fishing industry. The chitin nanofibers had a 10–20 nm diameter and a good aspect ratio [116]. The application of chitin is somewhat limited due to its insoluble character in most solvents [117].

Chitin deacetylation produces the polysaccharide biopolymer chitosan. In CS, the R group is NH_2 instead of OH in cellulose. Because D-glucosamine repeat unit C-2 possesses a -NH_2 functional group, chitosan is a better chelating agent than cellulose [118]. Deacetylated polysaccharides are transformed into polyelectrolytes in an acidic medium (pH < 6.5) and are more likely to dissolve because they lose their charges and precipitate due to amine (-NH_2) group deprotonation [119]. Chitosan properties in a solution are generally determined by molecular weight, deacetylation, polymer charge, ionic strength, and pH [120]. The most well-known chitosan properties include biocompatibility, biodegradability, metal chelation, mucoadhesiveness, and antibacterial [121, 122]. Owing to the mentioned properties, it is a polymer of high importance in biomedical goods, cosmetics, chemicals, and medicines [123].

The primary issue with the CS is that it can only be dissolved in an acidic medium (pH 2-6) and becomes polycationic, forming a viscous solution. So, high voltage is required for the ES of CS due to high viscosity. Due to the application of high voltage, CS NFs generally show bead formation [124-127]. The other issue includes the strong intramolecular hydrogen bonding created in chitosan solutions since the polymer chain motion in response to the electric field is usually suppressed, and for continuous fiber fabrication to occur during ES, it induces minimal accessibility for the spinning of the polymer [128, 129]. These problems can be addressed by employing solvents like trifluoroacetic acid (TFA), or a combination of TFA and dichloromethane (DCM), or by adding additional solvents to the former mixture such as ethanol,

1,4-dioxane, or acetone [130, 131]. Chitosan solution in TFA generates soluble salt residues of -$NH_3^+CF_3COO^-$ because the amino groups of CS form salts with TFA, disrupting the regular rigid interaction between chitosan molecules and hence resulting in the dissolution of the polymer, increasing the accessibility for ES and the charge density of CS solution [132]. Due to the high volatility of the TFA solvent, the resulting NFs solidify rapidly. The fundamental issue with the TFA based CS NFs is the low mechanical strength in the aqueous media which is the main requirement for any biomedical application [133]. However, blending CS with other biopolymers can efficiently address this issue. Some of the major blended biopolymers used with CS to produce NFs and the solvent systems are listed in Table 5. The morphologies of various blended chitosan nanofibers are shown in Fig. S3 [17, 134].

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316 **Table 5. List of various commonly blended biopolymers for the electrospinning of chitosan**

Source	Commonly blended biopolymers	Solvent system	Application	References
Natural	Alginate	Ethanol in water (20% v/v) OR Acetone: dimethyl acetamide (2:1 w/w)	Wound dressing, Skin repair, Drug delivery	[135-137]
	Pullulan	Aqueous acetic acid (50% v/v)	Drug delivery, wound dressing	[138, 139]
	Cellulose	Trifluoro acetic acid: acetic acid (7:3 w/w)	Adsorption, Food packaging	[133, 140]
	Hyaluronic acid	Distilled water	Drug delivery, wound dressing	[11, 67]
Synthetic	Polyethylene oxide	Acetic acid (various percentages)	Tissue engineering, Sensors, wound healing	[141-143]
	Polyvinyl alcohol	Acetic acid (various percentages) OR Distilled water	Metal ion removal, Drug delivery	[144, 145]
	Polylactic acid	Trifluoro acetic acid: dichloromethane (70:30 v/v)	Water filter, Wound dressing, Drug delivery	[146-148]

317 ### 2.2.4 Hyaluronic Acid

318 Natural Hyaluronic Acid (HA) is a linear polysaccharide composed of alternating disaccharide
319 units of D-Glucuronic acid and N-acetyl D-glucosamine. Its molecular weight ranges from 100
320 to 8,000 kDa depending upon the source [149]. The HA molecular weight is essential in several
321 biological activities, as shown in Table 6. As a component of most mammalian connective
322 tissues, HA helps to maintain structural integrity and viscoelasticity. HA's primary distribution in
323 connective tissues and important role in cell adhesion, proliferation, migration, and granulation
324 tissue development are all HA's biological features [150].

325

Table 6. Correlation of hyaluronic acid molecular weight with biomedical applications

Molecular weight (Da)	Applications	References
6×10^6	Skin inflammation treatment	[151]
$>1 \times 10^6$	Joint inflammation treatment	[152, 153]
1.2×10^5	Cancer treatment	[154, 155]
2×10^4	Immunomodulation	[156]
1×10^6	Removal of reactive oxygen species by UV-rays	[157, 158]
1×10^6	Epidermal cells growth regulation	[159]
$<1 \times 10^4$	Skin metabolism regulation and antiaging	[160, 161]
$2–3 \times 10^3$	Promoting collagen synthesis	[162, 163]
$<1 \times 10^4$	Joint health improvement by oral formulation	[164]

The most distinguishing features of HA are its biodegradability, biocompatibility, and negligible induction of immunogenic response, which makes HA an important biopolymer for various biomedical applications. The half-life of HA depends upon the location of the organs. It has a short half-life of 24 hours in skin tissues and is around 70 days long in some eye regions (up to 70 days) [165]. Injectable HA gels and solutions are used as dermal fillers to restore skin volume and reduce wrinkles' appearance. HA is also used in other cosmetics because of its safety and effectiveness. Drug delivery [166], osteoarthritis [167], and cosmetics [168] are only a few of the medical uses of HA. Furthermore, HA is often recognized as the most hydrophilic, naturally occurring polymer. The strong absorption characteristics of HA for wound exudates make it a preferred option for wound dressings [169].

The major issue with HA is the low mechanical strength in physiological conditions. HA is easily depolymerized invitro by acids and oxygen in the presence of thiols and ferrous based

reducing agents. [170]. Additionally, enzymatic or reactive species-mediated HA degradation may also occur in vivo and limit the biomedical applications of HA. Since HA is only soluble in the aqueous based media but even at low concentrations, the electrospinnability is poor due to the high viscosity and surface tension due to its ionic nature. Counter-ions in aqueous solutions also increase HA solutions' viscosity [171]. Due to the high viscosity, HA polymer chains could not interact with one another, contributing to jets' instability and the subsequent creation of discontinuous and heterogeneous NFs. Due to aqueous based solvents, the jet does not evaporate between the collector and the needle, leading to droplets or clogging in the needle. HA's relatively high surface tension requires a high voltage to generate more intense electrostatic forces to fabricate NFs, resulting in beading. Therefore, the high surface tension of HA must be decreased for the fabrication of continuous NFs [172]

To address these mentioned issues, the first approach is to blend HA with other biopolymers for the NFs fabrication [173]. Major biopolymers used for the ES of HA are listed in Table 5. The second approach uses solvents that disrupt the H-bonding in the HA chains [172]. That approach increases the flexibility of the chains, but the downside is the addition of toxic materials that reduce the potential for biomedical applications [174]. The third approach uses a surfactant to reduce surface tension [175]. The fourth approach is the modification of HA, which can be physical or chemical crosslinking. Conventional crosslinking is mainly accomplished by using aldehydes, hydrazides, or sulfides. However, the downside of the chemical crosslinking is the increase in cytotoxicity of the resultant NFs. Recently, 1-ethyl-3-(3-dimethylaminoisopropyl) carbodiimide modification has been used and considered better than the conventional crosslinking as the latter result in a non-cytotoxic product [175]. The physical modification is mostly done using silver nanoparticles to develop self-crosslinking [176]. Some of the major

363 biopolymers used with HA to produce NFs and the solvent systems are listed in Table 7. Some
364 important HA blended nanofibers morphologies are shown in Fig. S4 [11, 177].

365 **Table 7. List of various biopolymers blended with hyaluronic acid for electrospinning**

Source	Commonly blended biopolymers	Solvent system	Application	Reference
Natural	Cellulose	Distilled water	Therapeutic delivery	[178]
	Collagen	Hexafluoro isopropanol	Wound healing, Tissue engineering	[179, 180]
Synthetic	Polyvinyl alcohol	ethanol: water: benzyl alcohol (50:40:10 v/v)	Wound healing, drug delivery	[175, 181, 182]
	Polyethylene oxide	Water and 2,2,2-Trifluoroethanol: ethanol (1:10)	Drug delivery, Wound healing	[183, 184]
	Polycaprolactone	Ethanol: chloroform (7:3) OR formic acid: acetic acid (7:3)	Drug delivery, Wound healing, Tissue engineering	[185, 186]

366

367 ### 2.2.5 Alginate

368 Alginate (AG) is a linear anionic polysaccharide having mannuronic acid and glucuronic acid,
369 referred to as M-blocks and G-blocks, respectively, and (1,4)-glycosidic bonds connect both
370 blocks. AG is mainly obtained from bacteria and algae, with algae being the most common
371 source [187]. AG has a similar role to cellulose in plants by giving strength and flexibility to the
372 algae. Some bacteria (such as Azotobacteria and Pseudomonas) may also make AG [188].
373 Several features of AG vary depending on the source. Due to their high G-block concentration,
374 seaweed AG is often employed for biomedical applications [189]. In contrast, bacterial source-
375 based AG has high M-blocks concentration and often demonstrates enhanced immunogenicity
376 and cytokine production [190].

AG is an appealing material for increasing production and industrial applications since it is readily available, economically viable, and ecologically beneficial. AG is also nontoxic when employed in medical applications and has a low immunogenetic response in people because it has excellent absorptivity, biocompatibility, biodegradability, and nontoxicity [191]. Despite numerous beneficial characteristics, AG is difficult to electrospun, and pure AG electrospinning in organic or aqueous solvents has not been reported. Some of the properties of AG that have been recognized for its poor electrospinnability include its polyelectrolyte nature, stiff and prolonged structure, a tendency to form a gel at low concentration, high electrical conductivity, and high surface tension [191-193]. These problems are mainly addressed using a cosolvent system, blending with other biopolymers, surfactants, and chemical modifications [194]. The co-spinning polymers with better electrospinnability when blended with AG result in enhanced AG electrospinnability. The blending with other biopolymers boost the electrospinnability by producing charge repulsion in polymer chains of AG hence improving the flexibility of AG and generating new hydrogen bonds [191]. Some of the significant blended biopolymers and solvent systems of AG are listed in Table 8, and some major nanofiber morphologies are shown in Fig. S5 [14, 195].

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399 **Table 8. List of various biopolymers blended with alginate for the electrospinning**

Source	Commonly blended biopolymers	Solvent system	Application	Reference
Natural	Gelatin	Distilled water	Cell culture, Wound healing, dye adsorption	[196-198]
	Pullulan	Distilled water	Food packaging	[199]
Synthetic	Polycaprolactone	Aqueous acetic acid	Cell culture, Drug delivery, Wound healing	[200, 201]
	Polyethylene oxide	Distilled water	Wound dressing, Dye adsorption	[202-204]
	Polyvinyl alcohol	Acetone: dimethyl formamide (2:3 v/v)	Tissue engineering, Sensor	[205-207]

400

401 ## 2.2.6 Levan

402 For years, scientists have been intrigued by levan, a fructan homopolysaccharide with
403 amphiphilic properties [208]. The structure consists of residues of D-fructo-furanose are held
404 together by 2,6-glycosidic linkages and is synthesized by a wide range of microorganisms
405 (including Acetobacter, Bacillus, Halomonas, Microbacterium, and Streptococcus) and some
406 plants [209] (Fig. 9).

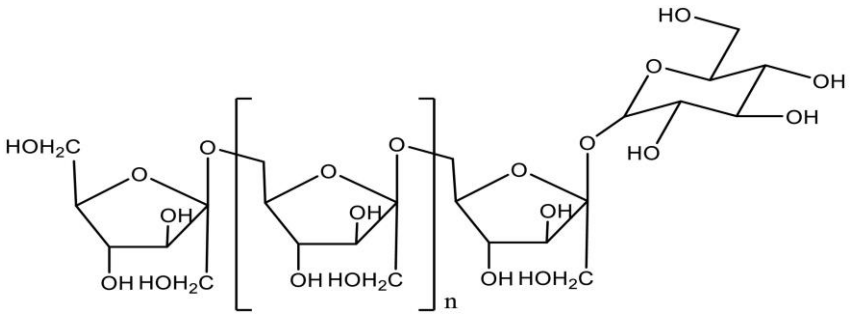

Fig. 9 Chemical structure of levan [1]

407

Any enzymes do not digest Levan once it enters the body since it does not affect the physiological environment of animals, unlike other polysaccharides like cellulose and starch [210]. This property can make this material an ideal candidate for drug delivery applications. In addition, they are intrinsically mucoadhesive and can be used in tissue engineering [1]. The main issue with levan ES is its inconsistent fluid properties at low or high concentrations. Levan displays Newtonian behavior for concentrations of up to 30% by mass, and between 30% and 55%, it shows shear thinning behavior, and for greater than 55% by weight, no plateau at low shear rates is obtained [211]. However, its nanofibers have been reported at 60% weight concentration in water [212]. Therefore, this material requires further studies to acknowledge its ES potential and to find better biopolymer for blending to enhance its natural properties and overcome its drawbacks.

2.2.7 Kefiran

Kefir is a probiotic and jelly-like grains complex of bacteria and yeasts that produces kefiran, a glucogalactan heteropolysaccharide copolymer of D-glucose and D-galactose [213] (Fig. S6). Kefiran has long been recognized as a possible biomaterial. It was electrospun for the first time in 2014 from a water-based solution with no additives. Since then, various efforts have been made to produce multifunctional nanofibrous scaffolds based on kefiran [214, 215].

Kefiran has anti-inflammatory, antibacterial, antifungal, and excellent biocompatibility properties. Probiotic properties have been proven to stimulate the development of healthy microflora while inhibiting the growth of cancer cells and bacteria. The best advantage of kefiran is its solubility in water and other commonly available solvents [216]. However, additional study is needed from the ES perspective, as kefiran is expected to generate a new, fascinating category of biomaterials with enhanced properties. Some recent biopolymers blended with kefiran and the

431 solvent system for ES are listed in Table 9, and some major nanofiber morphologies are shown
432 in Fig. S7 [18].

433 **Table 9. List of various biopolymers blended with kefiran for the electrospinning**

Source	Commonly blended biopolymers	Solvent system	Potential applications	References
Natural	None	Deionized water	Drug delivery	[18]
	Chitosan	Aqueous acetic acid (2% v/v)	Tissue engineering	[217]
Synthetic	Polyvinyl alcohol	Distilled water	Yet to be reported	[218]
	Polyethylene oxide	Distilled water	Food packaging	[215]

434

435 Fig. 10 shows the SEM images of major polysaccharides nanofibers with different biopolymers
436 and their ratios.

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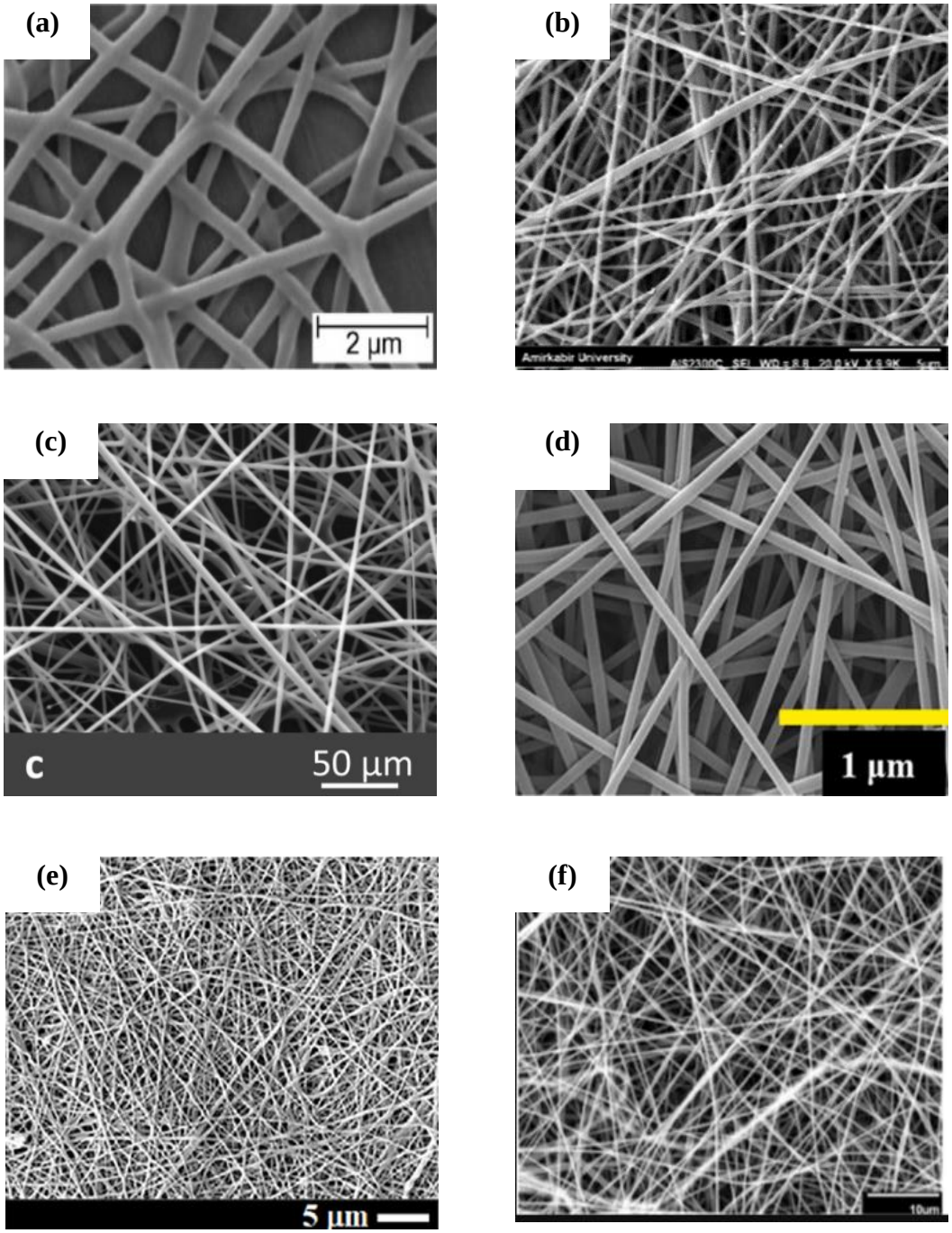

Fig. 10 SEM images of major polysaccharides nanofibers with other biopolymers, (a) Carboxymethyl cellulose: polyvinyl alcohol: (2:10 % w/v) [8] , (b) Hyaluronic acid: polyvinyl alcohol (1:2) [11], (c) Lignin: polyethylene oxide (97:3) [12], (d) Alginate: polyvinyl alcohol (8:2) [14], (e) Chitosan: alginate (1:1) [17], (f) Pure kefiran 4% [18]

442

2.2.8 Collagen and Gelatin

Collagen is the most common protein in mammals and makes up the ECM of numerous mammalian organs and tissues, including skin, bone, blood vessels, tendons, and ligaments [219]. Crosslinked tropocollagen units comprise three polypeptide chains that form a right-handed triple helix stabilized by interstrand hydrogen bonding and intrastrand interaction. Collagen protects the ECM's biological and structural integrity because of its high tensile strength and mechanical resilience [220]. Only one of the 29 collagen variations, collagen I, is used to produce collagen-based biomaterials. Because of its biodegradability, low antigenicity, customizable mechanical properties, ability to interact with a range of cell types, and capacity to form three-dimensional scaffolds, collagen is a material of interest in tissue engineering and therapeutic applications [221]. The major problems associated with collagen ES include loss of physiochemical structure due to high voltage, no swelling in the water; instead, it is dissolved instantly, and low denaturation temperature [222]. These shortcomings have been overcome by blending with other biopolymers (Table 10) and by using modifications in the structure [222]

Table 10. List of various biopolymers blended with gelatin for the electrospinning

Source	Commonly blended biopolymers	Solvent system	Potential applications	Reference
Natural	Chitosan	Distilled water	Tissue engineering, Wound dressing	[223, 224]
	Hyaluronic acid	Aqueous acetic acid	Tissue engineering, Wound healing	[179, 180, 225]
Synthetic	Polycaprolactone		Ulcer treatment, Wound healing, Tissue engineering	[226-228]
	Polylactic acid	Ethanol (80% v/v)	Tissue engineering,	[229, 230]
	Polyvinyl alcohol	Aqueous acetic acid (2% v/v)	Drug delivery, Wound dressing	[231, 232]

When collagen is hydrolyzed, it loses its native structure and transforms into gelatin. Two major events occur of this loss: breaking the intermolecular glycosylation linkage between lysine and hydroxylysine and breaking intramolecular disulfide bridges [233]. Collagen and gelatin are biopolymers with a fundamental structure composed of as many as 20 amino acids in varying quantities, depending on the hydrolysis procedure used to convert collagen to gelatin. However, there is a three amino acid sequence of L-arginine-glycine-aspartic acid (RGD) which is recognized by integrins to facilitate cell attachment.

The major features of gelatin include non-immunogenicity, biocompatibility, biodegradability, adaptability, cheap cost, and no denaturation phenomena. Thus, gelatin is used in many ways in the food and biomedical sectors, especially in tissue engineering. The major plus point is that after being subjected to ES, it has no denaturation phenomena and is preferred over collagen. The major drawback for gelatin NFs is the dissolution in the aqueous conditions. Gelatin is usually crosslinked by several crosslinkers, mostly aldehydes, including glutaraldehyde (GA), carbodiimides, succinimide, and genipin (GEN), for resolving dissolution issues. GA and GEN are considered the most suitable crosslinking agents because of their low toxicity and resistance to degradation in cell culture conditions at 37 °C [234].

Collagen and gelatin are usually methacrylate modified and resulting in ColMA and GelMA, respectively. ColMA has been widely researched for biological uses in the form of hydrogels [235], but its usage in ES has been restricted due to structural denaturation issues. Since ES produces GelMA rather than ColMA, it is a common electrospinnable material because of its high biocompatibility and morphological and chemical resemblance to ECM [236]. Furthermore, it promotes neovascularization and water adsorption, and its biodegradation rate is rapid and

simple to control. It has been widely used in tissue engineering [237]. Fig. S8 shows some essential collagen and gelatin nanofibers with commonly blended biopolymers [10, 238].

2.2.9 Elastin

Elastin is a naturally occurring protein with natural fiber morphology and is majorly found in animal ECM. This insoluble, elastomeric protein is formed by crosslinking tropoelastin, the protein's precursor (Fig. S9) [239] . Elastin has remarkable qualities, including a half-life of more than 70 years, outstanding durability, reversible stretchability, biocompatibility, and resistance to chemical and mechanical impacts. Elastin fibers have Young's modulus between 300 and 600 kPa and can be stretched up to 220% of their original length. They can withstand billions of stretch and recoil cycles without breaking [240].

Elastin found its application in wound healing and tissue engineering due to the mentioned intrinsic properties [241]. Since elastin is both resistant and insoluble, it presents specific difficulties as a starting material for ES. However, the solubility of elastin may be increased by acidic or basic hydrolysis. These preparations have several drawbacks, including compositional variability, potential loss of cellular signaling capabilities, and the need for crosslinking by glutaraldehyde to increase the water stability of the fiber mats. It is frequently mixed with additional polymers such as tropoelastin or elastin-like recombinamers (ELRs) 200 for rendering pure elastin spinnable. Major biopolymers blended with elastin are enlisted in Table 11, and some morphologies are shown in Fig. S10 [13, 242].

Table 11. List of various biopolymers blended with elastin for the electrospinning

Source	Commonly blended biopolymers	Solvent system	Potential application	References
Natural	Alginate	Deionized water and hexafluoro isopropanol	Tissue engineering	[34]
	Collagen	Deionized water	Tissue engineering	[243, 244]
	Hyaluronic acid	Hexafluoro isopropanol	Tissue engineering	[225, 245]
	Chitosan	Trifluoroacetic acid: dichloromethane (7:3)	Tissue engineering, Wound healing	[13, 246, 247]
Synthetic	Polylactic acid	Deionized water and dichloromethane	Tissue engineering, Wound healing	[248, 249]

2.2.10 Silk fibroin

Silk fibroin (SF) is a fibrous protein from silkworm larval cocoons, often Bombyx mori or Antheraea assama. It consists of polypeptide chains ranging in size from 200 to 350 kDa, and each chain consists of a tandem repeat of hydrophobic heavy and hydrophilic light chains held together by disulfide bonds [250]. As a result, a semi-crystalline fishnet structure is formed, with the crystalline regions acting as pressure sponges that disperse that force evenly over the whole fibroin network [247, 248]

Due to its accessibility, cheap cost, cytocompatibility, bioactivity, biodegradability, thermostability, excellent mechanical qualities, and minimal immunogenicity, SF has seen an uptick in usage as a biomaterial for tissue engineering and various biomedical applications in the previous decade [251, 252]. The solution is prepared in formic acid or hexafluoro-2-propanol for the ES of pure SF [253]. Organic solvents like methanol are required to stimulate the production

of a more stable antiparallel-sheet conformation (silk II) in the NFs. There is a risk that organic and caustic solvents can alter the structure and bioactivity of biomolecules, particularly sensitive therapeutics in biomedical applications [254]. As a result, it is recommended to use less severe conditions and refrain from employing organic solvents to reduce their toxicity. Mostly SF is also electrospun with other biopolymers to overcome the ES issues. Some of the most common blended biopolymers are listed in Table 12. The effect of the solvent system on the SF can be seen in Fig. 11; aqueous media results in uniform morphology, while the organic solvent hexafluoro isopropanol results in a wide range of diameter. Overall, SF seems promising for developing various tissue engineering materials, especially for treating burnt or infected wounds.

Table 12. List of various biopolymers blended with silk fibroin for the electrospinning

Source	Commonly blended biopolymers	Solvent system	Potential applications	References
Natural	Collagen	Aqueous acetic acid	Tissue engineering	[255, 256]
	Hyaluronic acid	Deionized water	Tissue engineering, Wound healing	[257]
Synthetic	Polyethylene oxide	Deionized water	Tissue engineering, Wound healing	[258, 259]
	Polyvinyl alcohol	Aqueous acetic acid (90% v/v)	Tissue engineering, Drug delivery, Wound dressing	[260-263]

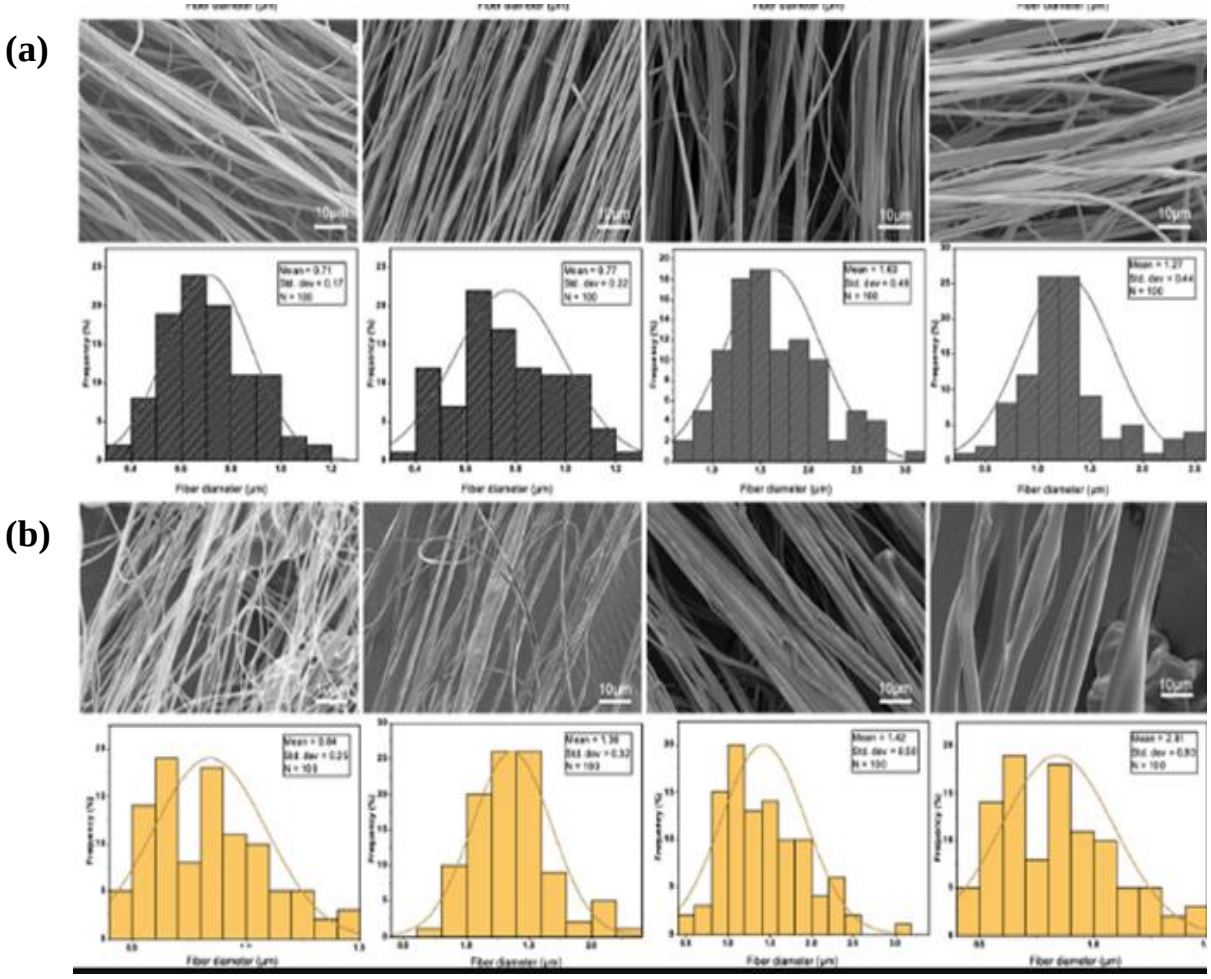

Fig. 11 SEM images of silk fibroin nanofibers with other biopolymers, (a) Polyethyleneimine: silk fibroin (10:90) in aqueous media, (b) Silk fibroin (8% w/v) in organic solvent (hexafluoro isopropanol) [9]

533 Fig. 12 shows the SEM images of major proteins nanofibers with different biopolymers and their

534 ratios.

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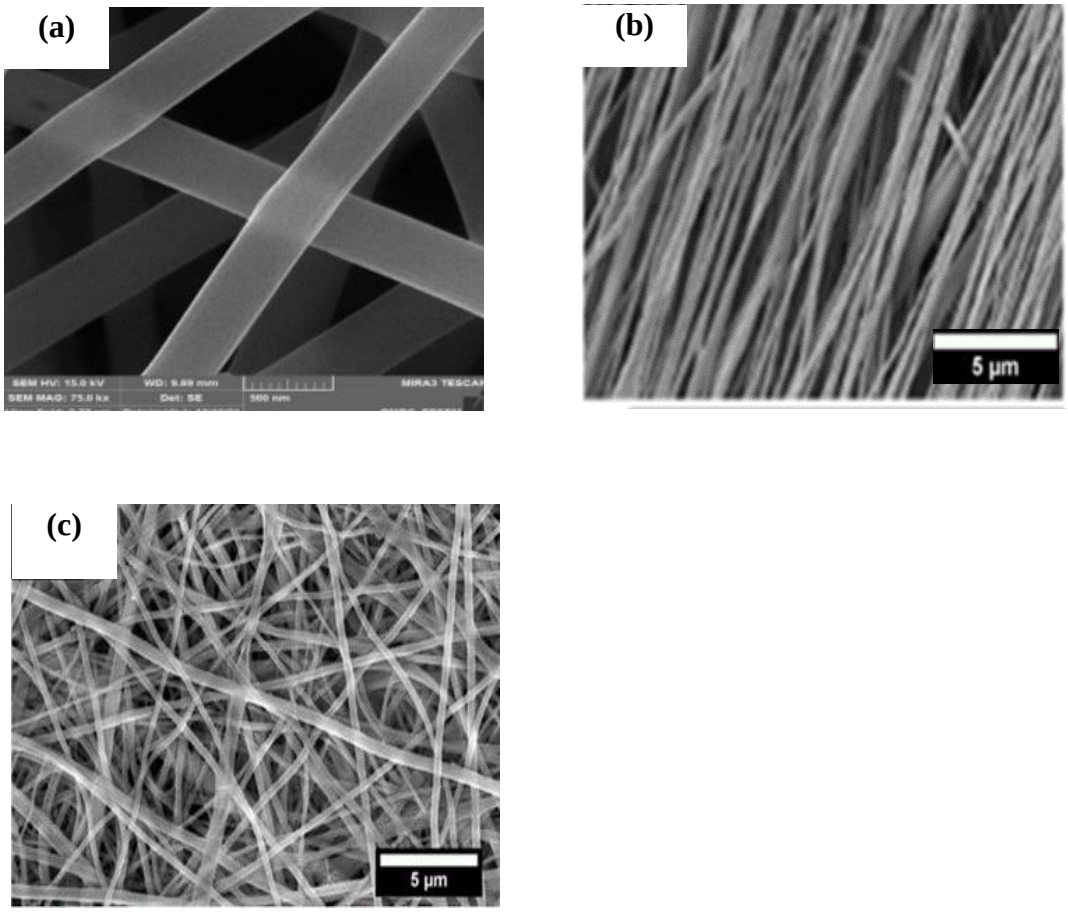


Fig. 12 SEM images of major proteins nanofibers with other biopolymers, (a) Gelatin: Alginate (1:1) [10], (b) Silk fibroin: polyethyleneimine (90:10) in aqueous media [9], (c) Elastinn: chitosan (5.5:4 w/v) [13]

539

540 3. Trends in biopolymers electrospinning

541 We use the ScienceDirect database to study the current trends and progress for biopolymeric
542 nanofibers (Fig. 13). Biopolymers were divided into two groups of polysaccharides and proteins,
543 depending on their biochemical nature. In the case of the polysaccharides group 212% increase,
544 while in the case of proteins, a 152% increase is seen in the nanofibers research from 2018 to
545 2022. For polysaccharides, cellulose is the top-researched biopolymer, while collagen and gelatin
546 have been the top research biopolymers since 2018. However, in our review, we have

highlighted polysaccharides newly introduced in ES. Kefir and levan have been gaining interest for the past few years, but still, much study is required to unveil their potential.

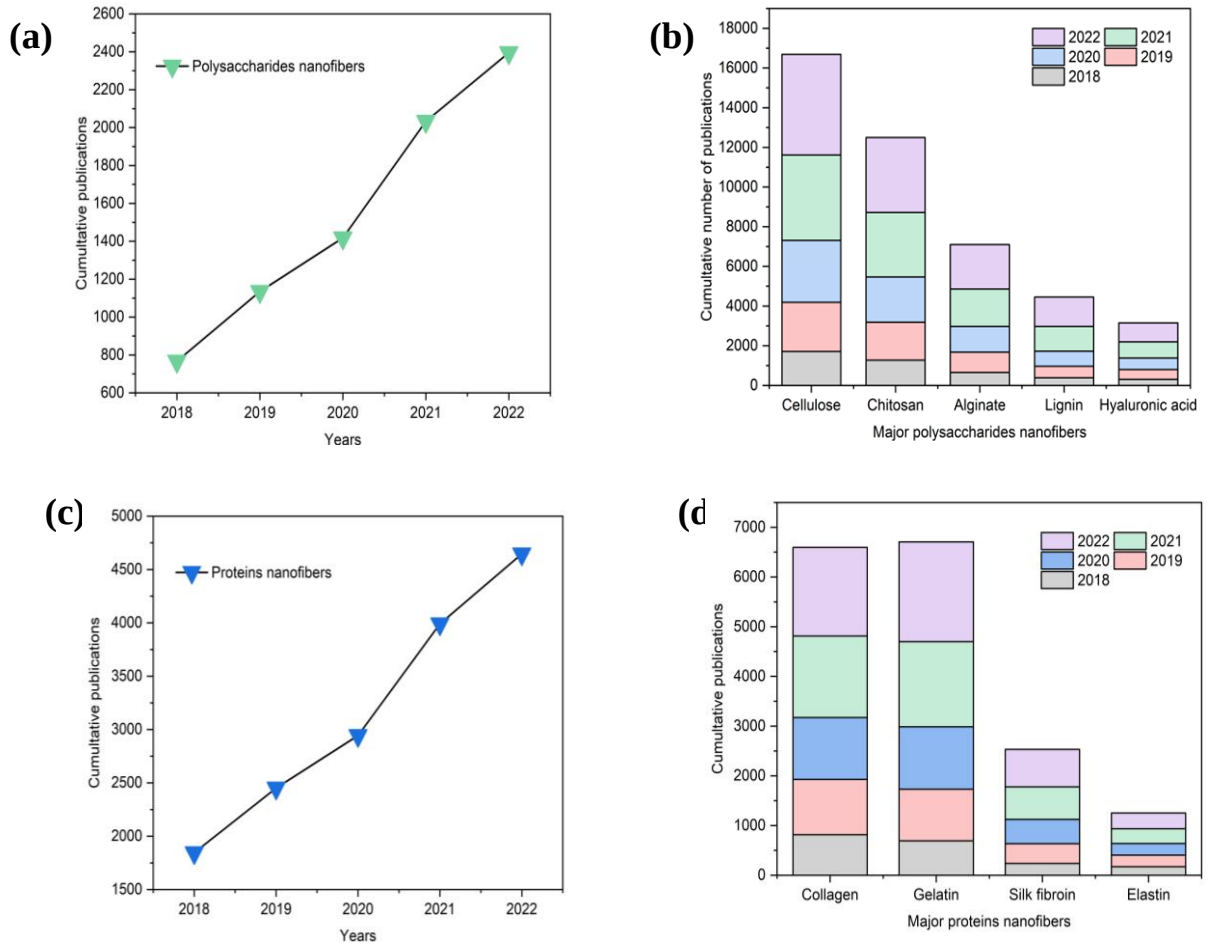


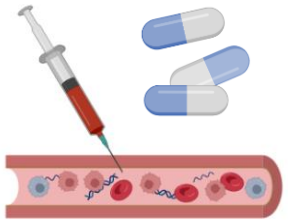
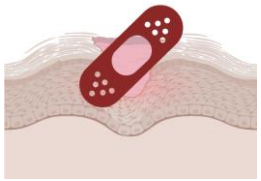
Fig. 13 Cumulative number of publications in ScienceDirect platform in the last five years (2018-2022), (a) Total polysaccharides based nanofibers, (b) Major polysaccharides nanofibers, (c) Total protein based nanofibers, (d) Major proteins nanofibers

4. Commercial products

Much study has been dedicated to the fast expanding field of electrospinning. Upscaling to industrial standards and commercialization are the ultimate goals of every research field. Regarding electrospinning, however, repeatability issues and batch-to-batch variability are the biggest problems across all methods [264]. This is a major hindrance to the widespread

554 commercialization and industrialization of electrospinning, particularly for use in the medical
555 field. Table 13 details several cutting-edge electrospinning-based medicinal supplies.
556

557 **Table 13. Recent applications of electrospinning methods for biopolymers nanofibers based wound healing system**

Product category	Brand name	Manufacturer	Main component	Key features	References
Drug delivery patches 	Rivelin® patch	Bioinicia (Valencia, Spain)	---	The system is specially designed for mucosal surface unidirectional drug delivery.	[265]
Surgical sutures and wound healing dressings 	Zeus Bioweb™	Zeus Industrial Products, Inc. (Orangeburg SC, USA)	Polytetrafluoroethylene	Ultrasmall fibers with the least chemical reactivity	[266]
	ReDura™	MEDPRIN (Guangzhou, China)	Polylactic acid	Material is similar to extracellular matrix (ECM) and promotes rapid repair and regeneration.	[267]
	ReBOSSIS®	Ortho ReBirth (Yokohama-shi Kanagawa pref., Japan)	TCP (β-Tricalcium Phosphate), Bioabsorbable Polymer and SiV (Silicone-containing Calcium Carbonate	Bone filler that promotes the bone formation	[268]
	HealSmart™	PolyRemedy®, Inc. (Concord, MA, USA)	Hyaluronic acid	Antimicrobial Dressings	[269]
	PK Papyrus®	Biotronic (Berlin, Germany)	Polyurethane	Thin and elastic membrane for stent coating	[270]
	Surgiclot®	St. Theresa Medical Inc. (Eagan, USA)	Dextrin and fibrin	Sealent for the bone bleeding	[271]

5. Future prospectives

Despite widespread agreement that electrospinning is a fascinating method with promising applications, issues still need to be addressed. Various hurdles to fabricating these biopolymeric nanofibers include innate poor electrospinnability of biopolymers, lack of individual biopolymers properties to accomplish the applications solely, and optimizing the solvent system for the nanofibers fabrication. This review focusses on all three mentioned issues and addresses the solutions for individual biopolymers. The blending of biopolymers can solve the lack of properties issue. The major results of this study suggested that the functionalization of biopolymeric materials and then the blending with other biopolymers is crucial for developing and improving these materials for use in targeted applications. However, the major concern is the insolubility of biopolymers and the optimization of the solvent system. The solvent system plays a critical role in the fabrication of the nanofibers. This study provides insight into the current solvents systems for the major biopolymers nanofibers fabrication.

However, it was observed that most of the solvents employed for the fabrication are organic solvents, and they are a concern from the environmental and the user's point of view. During the ES procedure, most of the solvents are evaporated; however, some traces are still left in the fabricated nanofibers, resulting in the commercial level pharmaceutical development of biopolymeric nanofibers. Melt electrospinning might seem like a straightforward way to overcome the organic solvent based procedures. However, the process has drawbacks regarding the fibers' complexity, fibers with a large diameter, polymer-related thermal degradation, and incompatibility with several high throughputs. On the other hand, needleless electrospinning is considered the most advance electrospinning method, but in this technique, a large liquid surface that is exposed to the air and evaporates extremely volatile solvents into the environment results

in raised concerns for the user and the developed product. To overcome the solvent related issues, researchers should develop "green" solvents as the alternative to the harmful toxic solvents whose residues will not be harmful or will be affecting the biomedical applications and, above all, will increase the electrospinnability of the blend solution.

The other major issue noted in the research is the lack of generalized solvent-based models for the ES process biopolymers. Even though stimulation models have been studied, no one has yet created a model that can reliably forecast the needle-based or needleless electrospinning parameters. Therefore, most ES experiments depend on parametric analyses and empirical knowledge of the process requirements. The current literature lacks studies on developing standard protocols to make the fabrication more standardized rather than focusing on just changing the process parameters (voltage, needle distance, and flow rate). To overcome these restrictions, scientists should be more forthcoming with their findings on optimizing the solvent systems and how the solvent system will affect different properties of biopolymeric under the same electrospinning parameters and vice versa. In addition, more focus should be put on developing green solvents for biopolymeric nanofibers to replace harmful and toxic solvents.

6. Conclusion

Biopolymers are the materials in focus to replace petroleum based polymers. Various biopolymers have proved to be perfect candidates in different industries, especially in the health sector, due to their natural features like biocompatibility, antimicrobial activity, and biodegradability. The fibrous morphology of biopolymers is essentially required for effective biomedical applications, including tissue engineering, drug delivery and wound healing.

Electrospinning (ES) has emerged as the most productive and modern technique for fabricating biopolymeric nanofibers. ES provides the flexibility of combining various biopolymers without

toxic crosslinkers, which is nearly impossible with other conventional techniques. The resultant nanofibrous morphology provides better surface area, mechanical properties, and compatibility. However, electrospinning of biopolymers is still going through the developmental stages, and there are many issues, including the use of toxic solvent systems, no focus on parameter based studies in the current literature and the lack of reproducibility. The solvent system issue can be resolved by formulating environmental friendly green solvent systems. For the other issues, more parametric studies are required to make the ES procedure universal and user-friendly to reduce variation among the batches. The focus on these recommendations can lead to better utilization of the ES technique and increased commercialization of biopolymeric nanofibrous products.

Declaration of Competing Interest

The authors state that no potential conflicts of interest or personal ties might have influenced the work reported in this research.

Declaration of generative AI in scientific writing

The authors state that no AI software or AI based technologies were used for the writing purposes of this review article.

Authors contribution

All authors contributed towards drafting and critically revising the paper and agree to be accountable for all aspects of the work. The authors confirm that this manuscript has not been previously published and is not currently under consideration by any other journal.

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