International Journal of Biological Macromolecules 171 (2021) 130–149

Contents lists available at ScienceDirect

International Journal of Biological Macromolecules

journal homepage: http://www.elsevier.com/locate/ijbiomac

Review

Electrospun fibers based on botanical, seaweed, microbial, and animal sourced biomacromolecules and their multidimensional applications

K.P. Akshay Kumar [a,1], Ehsan Nazarzadeh Zare [b], Rafael Torres-Mendieta [c], Stanisław Wacławek [c], Pooyan Makvandi [d,*], Miroslav Černík [c,*], Vinod V.T. Padil [c,*], Rajender S. Varma [e,*]

[a] Department of Applied Chemistry, Cochin University of Science and Technology (CUSAT), India
[b] School of Chemistry, Damghan University, Damghan 36716-41167, Iran
[c] Institute for Nanomaterials, Advanced Technologies and Innovation (CxI), Technical University of Liberec (TUL), Studentská 1402/2, Liberec 1 461 17, Czech Republic
[d] Istituto Italiano di Tecnologia, Centre for Micro-BioRobotics, Viale Rinaldo Piaggio 34, 56025 Pontedera, Pisa, Italy
[e] Regional Centre of Advanced Technologies and Materials, Palacký University in Olomouc, Šlechtitelů 27, 783 71 Olomouc, Czech Republic.

ARTICLE INFO

Article history:
Received 1 November 2020
Received in revised form 20 December 2020
Accepted 28 December 2020
Available online 5 January 2021

Keywords:
Electrospinning of bio-macromolecules
Fiber morphology and structures
Multidimensional applications

ABSTRACT

This review summarizes and broadly classifies all of the major sustainable natural carbohydrate biomacromolecular manifestations in nature – from botanical (cellulose, starch, and pectin), seaweed (alginate, carrageenan, and agar), microbial (bacterial cellulose, dextran, and pullulan), and animal (hyaluronan, heparin, chitin, and chitosan) sources – that have been contrived into electrospun fibers. Furthermore, a relative study of these biomaterials for the fabrication of nanofibers by electrospinning and their characteristics viz. solution behavior, blending nature, as well as rheological and fiber attributes are discussed. The potential multidimensional applications of nanofibers (filtration, antimicrobial, biosensor, gas sensor, energy storage, catalytic, and tissue engineering) originating from these polysaccharides and their major impacts on the properties, functionalities, and uses of these electrospun fibers are compared and critically examined.

Contents

* Corresponding authors.
E-mail addresses: pooyan.makvandi@iit.it (P. Makvandi), miroslav.cernik@tul.cz (M. Černík), vinod.padil@tul.cz (V.V.T. Padil), Varma.Rajender@epa.gov (R.S. Varma).
[1] Present address: Future Energy and Innovation Laboratory, Central European Institute of Technology, Brno University of Technology, Purkyňova 123, 61200 Brno, Czech Republic.

https://doi.org/10.1016/j.ijbiomac.2020.12.205
0141-8130/

1. Introduction

Polysaccharides are abundant in the environment and have a wide range of functionalities and structural characteristics, originating from plants, animals and microorganisms (Fig. 1). The potential applications of these bio-macromolecules in various fields such as food, medical, pharmaceutical, energy harvesting, cosmetics, packaging, surface coatings, adhesives, dyes and pigments, paper making, construction, antimicrobial, water treatment, etc. have been well documented [1–9]. Furthermore, natural polymers are used extensively due to their outstanding functionalities, which include thickening, gelling, emulsifying, foaming, creaming, dispersive, and adhesive abilities, and they are also used in the development of films and coatings. A comprehensive list of the various natural carbohydrate polymers acquired from botanical, seaweed, animal, and bacterial sources are presented in this review.

Commercial natural polymers such as polysaccharides and proteins are widely used biological macromolecules that are present in food, medical, and other industries. Natural polysaccharides are used in the food and non-food fields as viscosity enhancers, gel formers, emulsion stabilizers, dispersion producers, and film generators [10–13]. These natural polymers command a market in the food industry [14,15] where starch and its derivatives account for ~70% of usage compared to gelatin, pectin, and carrageenan, whereas polysaccharides such as alginates, celluloses, and others are limited to less than 4%. Hyaluronan, another polysaccharide extracted from both animal and bacterial sources, is used in tissue engineering, ophthalmic surgery, and in the treatment of arthritic joints due to its remarkable viscoelastic properties and biocompatibility [16].

Electrospinning is one of the simplest, most economical, and flexible ways of fabricating polymers at a micro or nano scale [17,18]. Polysaccharides display variable intrinsic characteristics such as molecular weight distribution, degree of deacetylation, purity, charged functional groups, and crystallinity, all of which depend on their source of extraction [19,20]. There are many unique factors to electrospinning systems and their corresponding process parameters. The most significant polymer properties (viz. molecular weight distribution, glass-transition temperature and solubility) and solution characteristics (viz. viscosity, viscoelastic behavior, concentration, surface tension and electrical conductivity) – together with other inherent attributes (viz. substrate behavior, solution feed rate, field strength, geometry of electrode, vapor pressure of the solvent and relative humidity) – all serve to control and stabilize the electrospinning process [18,21].

Many natural materials comprising proteinaceous or saccharide building blocks have been electrospun into nanofibers and have found application in many areas. These include the food [10,22], environment [23], energy [24], biosensor [25], and biomedical (tissue engineering, drug delivery and pharmaceutical) sectors [26]. Several researchers have extensively worked on the green electrospinning of biopolymers and polysaccharides. This article focusses on the characterization of electrospinning and the ensuing application of commercial natural carbohydrate polymers extracted or obtained from botanical, seaweed, animal and bacterial sources. Furthermore, electrospun fibers of commercial natural polymers and their properties and applications in diverse fields such as agriculture, biomedical materials, energy storage, biosensor, water treatment, removal of pollutants, and analytical sciences are comprehensively described in detail.

2. Electrospinning parameters

The electrospinning of nanofibers based on natural polymers depends on solution, processing, and environmental factors. They bestow changes in the morphology (size and shape) of the resulting fibers, such as straight and aligned, bead-free, non-uniform, beads-in-string, spindle, ribbon, rod, porous, wrinkled, grooved, rough, cactus, hollow, and core-shell shaped nanofibers [27–30].

Bio-macromolecules possess a high molecular weight and complicated structures, including their molecular configurations in main chain and side chains. The electrospun fibers and their properties,

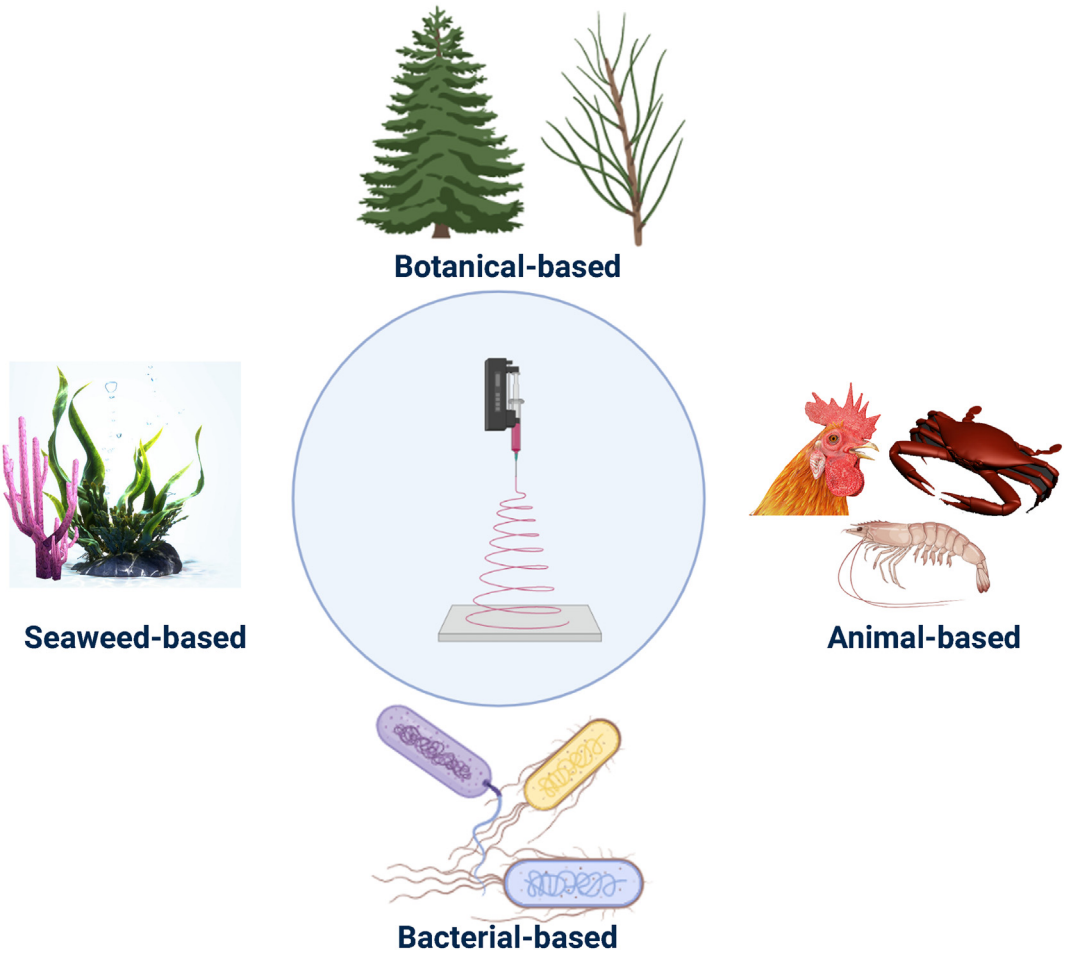

Fig. 1. Sources of bio-macromolecules originating from plants, animals and microorganisms.

characteristics and applications depend on the electrospinning parameters. Table S1 (Supplementary information) summarizes how different electrospinning parameters may influence the morphology, size and other characteristics of nanofibers based on bio-macromolecules.

3. Effect of solvent, structure-properties, and the mechanical attributes of bio-macromolecule electrospun fibers

Electrospinning parameters highly influence the outcome of the final nanofiber membranes providing outstanding properties and functionalities such as high porosity, high thermal stability and water permeability, interrelated pore structure, and a well-ordered composition [31]. The selection of the proper solvent systems for the polymers is the key challenge in electrospinning. The type and molecular weight of the solvent, low temperature and deficient solvent volatility may lead to wet fusion during the deposition of the fibers thus promoting inhomogeneous fiber formation [32]. Pristine cellulose acetate (CA) nanofibers have certain drawbacks such as reduced mechanical attributes and structural defects. To overcome these limitations, additives (nanoscale fibers or nanoparticles, NPs) can be added to the CA nanofibers to enhance the structural and mechanical properties of the composite nanofibers [33]. Other important factors such as the viscoelastic properties, electric conductivity, and specific surface energy of the polymeric materials determine the electrospinnability and the nano-structural formation [34]. To develop enhanced structural, physical, chemical, and mechanical properties of the electrospun fibers, natural and synthetic polymers can be blended to create fibers with highly desirable properties such as improved biodegradation, biological response, flexibility and toughness for their subsequent applications in tissue engineering and other specific fields. For example, poly(lactic-co-glycolic acid) (PLGA), poly(ethylene oxide)(PEO) and polycaprolactam (PCL) have been mixed with carbohydrate polymers such as starch, alginate, hyaluronic acid, chitosan, dextran, and gelatin to fabricate highly specialized nanostructured and mechanically robust electrospun fibers [35].

Electrospun composite fibers of CA and polyacrylonitrile (PAN) with varying compositions have been fabricated via a multi-electrospinning method [36]; an increase in CA content in the composite fibers, the dimeters of the fibers could be increased from 303 ± 43 to 502 ± 119 nm. Electrospinning solvents such as *N,N*-dimethylacetamide (DMAc) (Boiling point = 166 °C) and dimethylformamide (DMF) (Boiling point = 152.8 °C) can also affect the fiber morphology due to differences in their volatility. Furthermore, low volatility and high temperature are preferred in the electrospinning process to enhance the evaporation rate of the solvent. In one study, the ensuing nanofibers exhibited high water contact angle (131°), bulk density (0.252 g/cm^3), surface roughness, and mechanical attributes (elastic modulus (σmax) = 5.0 MPa; elongation at break (εb) = 29.2%; and Young's moduli (E) = 1.3 MPa) after increasing the amount of CA (75% content) in the composite fiber [37,38]. Further, the thermal properties of the composite electrospun fibers are depended on the spinning parameters, especially the higher content of CA in the spinning solution has contributed to the inherent higher thermal stability of the PAN nanofibers.

Several solvents such as acetone, DMAc, DMF, trifluoroethylene (TFE), chloroform, methanol, water or their mixtures in various ratios have been successfully used for electrospinning CA. However, the low boiling point of acetone makes it difficult to use for longer duration of electrospinning on industrial-scale CA nanofibers. In this context,

Naragund et al. [39], developed a solvent system comprising methyl ethyl ketone (MEK) and DMAc in two different ratios (2:1 and 1:1 v/v, respectively) for the electrospinning of CA [39]. This study highlighted the fact that the morphology (cylindrical) of the produced CA fibers depended on the polymer concentration and solvent ratios. Furthermore, considering the reduced evaporation rate of MEK, MEK/DMAc has been found to be a better solvent than acetone/DMAc for the industrial-scale production of CA nanofibers.

To develop biodegradable composites, cellulose nanofibers have been fabricated as a reinforcing agent via coaxial electrospinning. Cellulose nanofibers with a diameter of 560 nm have been incorporated with poly (butylene succinate) (PBS) to make a bio-composite, which showed that the storage modulus of the composites was 67% higher than that of a neat PBS matrix. Similarly, a polylactic acid (PLA) composite reinforced by cellulose was found to have enhanced mechanical attributes (higher storage modulus) compared to the bare PLA [40]. These results show that coaxial electrospinning may provide highly mechanically robust electrospun reinforced fibers based on carbohydrate polymers.

The effect of solvent composition, concentration and rheological properties with respect to the electrospinnability of pullulan (Pu) polymers has been investigated [41]. The effect of the rheological behavior of Pu (concentrations = 0.1–20% w/v) in aqueous dimethyl sulfoxide (DMSO) solutions with varying concentrations (0%, 20%, 40%, 60%, 80%, and 100% v/v) was studied. Pu concentrations lower than 10% (w/v) were observed to display Newtonian behavior, whereas concentrations above 10% showed shear thinning attributes. By applying the power law model ($\eta = K\gamma^{n-1}$) to the data observed, the power law indices (n) were 0.83 and 0.82 for 20% (w/v) pullulan in 60% and 40% DMSO, respectively. This effect was corroborated with starch dispersions in an aqueous/DMSO medium, where the flow rate was reported to display Newtonian behavior [42].

The entanglement concertation of Pu in DMSO/aqueous solution ranged from 3.99% to 4.41% (w/v) in DMSO concentrations of 20% and higher. However, the Pu had a much higher entanglement concentration (5.33% w/v) in the aqueous solution compared to DMSO/aqueous dispersions. This shows that the DMSO had no effect on the molecular interactions of Pu in the solvent and the rheological behavior with respect to the molecular entanglement and shear viscosity was influenced by the electrospinnability of Pu, as reported for starch polymer electrospinning [42]. The pullulan concentration was 1.88 to 2.25 times the entanglement concentration depending on the DMSO concentration and shear viscosity (0.06–2.2 Pa s) for the production of uniform electrospun nanofibers. The uniform fiber diameter and pore size of the Pu electrospun fibers were produced with DMSO concentrations ranging from 0% to 20% and the results were comparable with electrospun Pu fibers produced via green electrospinning [43].

Chitosan (Ch)-Pu nanofibers using different cross-linking methods (thermal cross-linking by a Maillard reaction and cinnamaldehyde cross-linking by a Schiff base) have been described and their mechanical properties (elastic modulus, tensile strength, and elongation at break) compared [44]. With an enhanced concentration of Ch in the blend nanofibers, both the elastic modulus and the tensile strength increased from 344.3 ± 66.4 to 1150.9 ± 355.3 MPa and 11.36 ± 0.70 MPa to 22.5 ± 2.6 MPa. However, the elongation at the break decreased from 4.5 ± 0.8% to 2.8 ± 0.8%. This effect may be explained by the intermolecular entanglement and interaction between the polymeric chains (Ch and Pu) governed by covalent bonding [45]. Overall, the mobility of the polymer chains was reduced, but the rigidity increased [46]. The solvent effects, as well as electrospinning parameters (system, process, and ambient conditions and their structural-functional relationships), mechanical, thermal and applications of various biomaromolecules electrospun fibers are summarized in Table S2 (Supplementary Information).

4. Bio-macromolecules from botanical sources

Plant-based homopolysaccharide glucans such as cellulose (cotton, wood and unmodified), starch (amylose (potato), and amylopectin (potato and waxy maize)) and heteropolysaccharides (pectin, arabinogalactan, arabinoxylan, galactomannan, and glucomannan) are included in this section, which highlights their structural and functional attributes in relation to their electrospinning characteristics (Fig. 2).

4.1. Cellulose

4.1.1. Structure and properties

Cellulose is a renewable product with excellent characteristics such as high physical strength and biocompatibility, including impeccable ecological credentials. It also has great potential in medical, pharmaceutical, environmental and industrial applications. The major structural linkages occurring in cellulose are β-(1–4) D-glucopyranosyl residues with the β-sheet symmetry of this molecule harboring intra- and

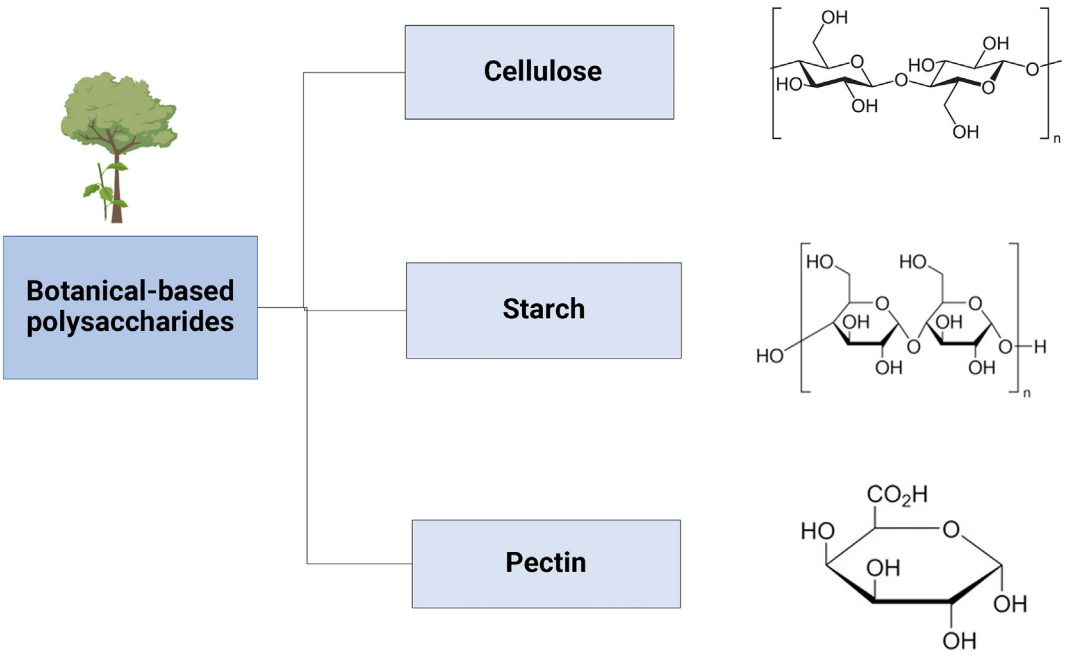

Fig. 2. Botanical sourced bio-macromolecules and their structures.

intermolecular hydrogen bonds. Native cellulose is crystalline and insoluble in aqueous and common organic solvents. Major cellulosic sources are wood pulp, dissolved wood pulp, cotton, cotton linter and other fiber pulps. Its uses are diverse and include the production of paper, casing, sanitary products, rayon, cellophane derivatives, textile fibers, fillers and medical fabrics etc. [47].

There have been remarkable applications of cellulose extracted from both plants and bacteria in the form of cellulose nanofibers and NPs [48], hydrogels, and aerogels [49], with their novelty being due to their nano rather than micro dimensions. However, nanofibers of cellulose have a peculiar advantage over other forms, owing to their excellent physical and chemical, and mechanical properties, coupled with their advanced applications in various domains viz. medical, catalytic, environmental, biosensory [50] and energy [51].

4.1.2. Electrospinning of cellulose

Many natural polymers depend on a 'top-down' approach to produce their corresponding nanofibers. To attain good solubility and homogeneity in various solvents, natural polymers are subjected to acid or alkali treatment in order to cleave the non-crystalline regions in their structures. Furthermore, factors such as high solution viscosity, inter/intramolecular hydrogen bonding, gelling behavior, and distinctive rheological properties all impact negatively on the smooth electrospinning of natural polysaccharides [22,52].

Various strategies employing physical, chemical, biological and oxidative methods are commonly used for the preparation of cellulose nanofibers. Physical methods (viz. ultracentrifugation, high-speed homogenization, ultra-sonication, etc.), acid/alkali digestion, enzymatic degradation, and (2,2,6,6-tetramethylpiperidin-1-yl) oxyl radical (TEMPO) oxidation are the general routes undertaken to generate the corresponding cellulose nanofibers [53].

The selection of a proper solvent system for the electrospinning of cellulose acetate is critical. Acetone, chloroform, *N*,*N*-dimethylformamide (DMF), dichloromethane (DCM), formic acid, methanol (MeOH), pyridine, and multiple phase solvents viz. acetone–dimethylacetamide (DMAc); chloroform–MeOH; DCM–MeOH and acetone/DMF/trifluoroethanol have been utilized for this purpose [54].

Balgis et al. [55] fabricated a dual-sized nanofiber composite via the electrospinning of TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl)-oxidized cellulose nanofibers using PVP (polyvinylpyrrolidone) with an ethanol-water system, serving as both a copolymer and a solvent. Attributes of the precursor (cellulose fibers) such as thixotropic nature and zeta potential determined the produced electrospun composite fibers of PVP-CNF (cellulose nanofibers). By using, the needle voltage to control the size of single and dual nanofibers, enhanced air purification efficiency may be achieved, whereby promoting its performance applicability in aerosol filtration.

Freire et al. [56] introduced ionic liquids for the electrospinning of cellulose nanofibers either using ionic liquid alone (1-ethyl-3-methylimidazolium acetate) or in combination with 1-decyl-3-methylimidazolium chloride, yielding nanofibers with dimensions of (120 ± 55) nm and (470 ± 110) nm, respectively. Extensive studies have also been conducted on ionic liquids in conjunction with a co-solvent (glycerol) to enhance the solubility and electrospinning characteristics of cellulose [57]. A recent review described the use of organic electrolyte solutions as effective greener solvent systems for the dissolution and regeneration of cellulose [58].

The cellulose derivative, namely cellulose acetate has been electrospun into fibers because of its solubility in many electrospinning solvents**,** along with the deacetylation process. The potential applications of cellulose acetate have been recently summarized [59]. The common solvents applied for the electrospinning of cellulose acetate include acetone, acetic acid and their mixture with dimethylacetamide/DMF/trifluoroethylene (TFE) [54]. Researchers have also used a mixture of cellulose acetate and water along with organic solvents (acetone) for electrospinning to avoid clogging and reduce the evaporation rate of the organic solvent. Furthermore, the addition of ammonia or formic acid to maintain pH during electrospinning is vital. Silver incorporated nanoparticles or mixed blended nanofibers of cellulose acetate have been produced via electrospinning with good antibacterial properties [60].

Electrospun fibers of cellulose derivatives have been investigated in the immobilization of biologically active molecules or drug and vitamins – e.g. alkannin [61], amoxicillin [62], curcumin [63], ibuprofen, indomethacin, naproxen, [64] shikonin, sulindac [65], vitamin A and vitamin E [66] – to enhance cell adhesion, proliferation and differentiation properties in biomedical applications, especially in tissue engineering and drug delivery. Rodríguez et al. [67] embedded gentamicin sulfate and halloysite clay into cellulose ether-PVA nanofiber mats to deliver therapeutics to the skin tissue; presence of halloysite enhanced the mechanical properties while gentamicin was used to impart antibacterial activity. The scaffold composite exhibited faster in-vitro and in-vivo wound healing as depicted in Fig. 3. [68]

Taajamaa et al. [69] produced biocomponent fibers by electrospinning hydrophobized cellulose derivatives (trimethylsilyl cellulose (TMSC) and cellulose triacetate (CTA) in chloroform; super-hydrophobic fibers with diverse morphologies were generated depending on the polymer blend ratios. Wang et al. [70] fabricated TiO_2 immobilized CA electrospun fibers by electrospinning and used them for the photocatalytic degradation of methylene blue (methylthioninium chloride) from textile effluents. The higher photocatalytic activity of the fibers was discerned with high TiO_2 content as CA composite ultrafine fibers with 5 wt% TiO_2 were as high as 90% after 4 h of degradation.

Fu et al. [71] devised an effective means of transferring polyaniline nanorods to electrospun carboxymethyl cellulose (CMC)-modified cellulose nanofiber surfaces, wherein a CMC/cellulose/polyaniline hierarchical nanostructure was fabricated by in-situ polymerization of aniline on the electrospun CMC/cellulose nanofibers. The ensuing CMC/cellulose/PANI nanofibers maintained the cylinder-shaped fibrous structures of singular fibers as well as the 3-D morphology of fibrous mats, providing a great electroactive surface area for simplistic electron transfer and mass diffusion.

Amino-cellulose-based nanofibers were synthesized by electrospinning of 6-deoxy-6-trisaminoethyl-amino (TEAE) cellulose and polyvinyl alcohol (PVA) blended solutions in aqueous media [72]. The study emphasized that TEAE-containing nanofibers caused a robust fall of *S. aureus* growth as well as substantial antibacterial effectiveness against Gram-negative *K. pneumoniae* bacteria, which can enter the respiratory tract causing *pneumonia*.

Moreover, cellulose-based nanofibers have been actively utilized in the field of gas sensors and energy storage systems. Electrospinning of sodium-doped cellulose nanocrystals together with a tungsten (W) precursor and further subjected them to calcination to fabricate tungsten oxide (WO_3) nanotubes [65]. Interestingly, the formed $Na_2W_4O_{13}$ on WO_3 showed a selective response to hydrogen disulfide (H_2S), exhibiting an outstanding sensor performance. Ultrafine cellulose fibers with titanium particles were synthesized and used as separators for lithium-ion batteries [66]. Cellulose-based nanofibers have been used for a polymer-ion battery [73] where the specific capacity reached above 200 mAh g^{-1}. Active research is still being pursued for exploiting cellulose-based nanofibers for versatile applications, which may open various research opportunities in the near future.

4.2. Starch

4.2.1. Structure and properties

Starch is produced in higher plants as a storage material and has been extensively used for food and non-food applications. Structurally, starch consists of complex polysaccharides with α-D-glucopyranosyl (α-D-Glcp) units viz. amylose and amylopectin. Glycosyl residues (α-(1 → 4) links in amylose and (1 → 4)-linked α-D-Glcp units, and a significant proportion of branch points α-(1 → 6) linked in amylopectin) are

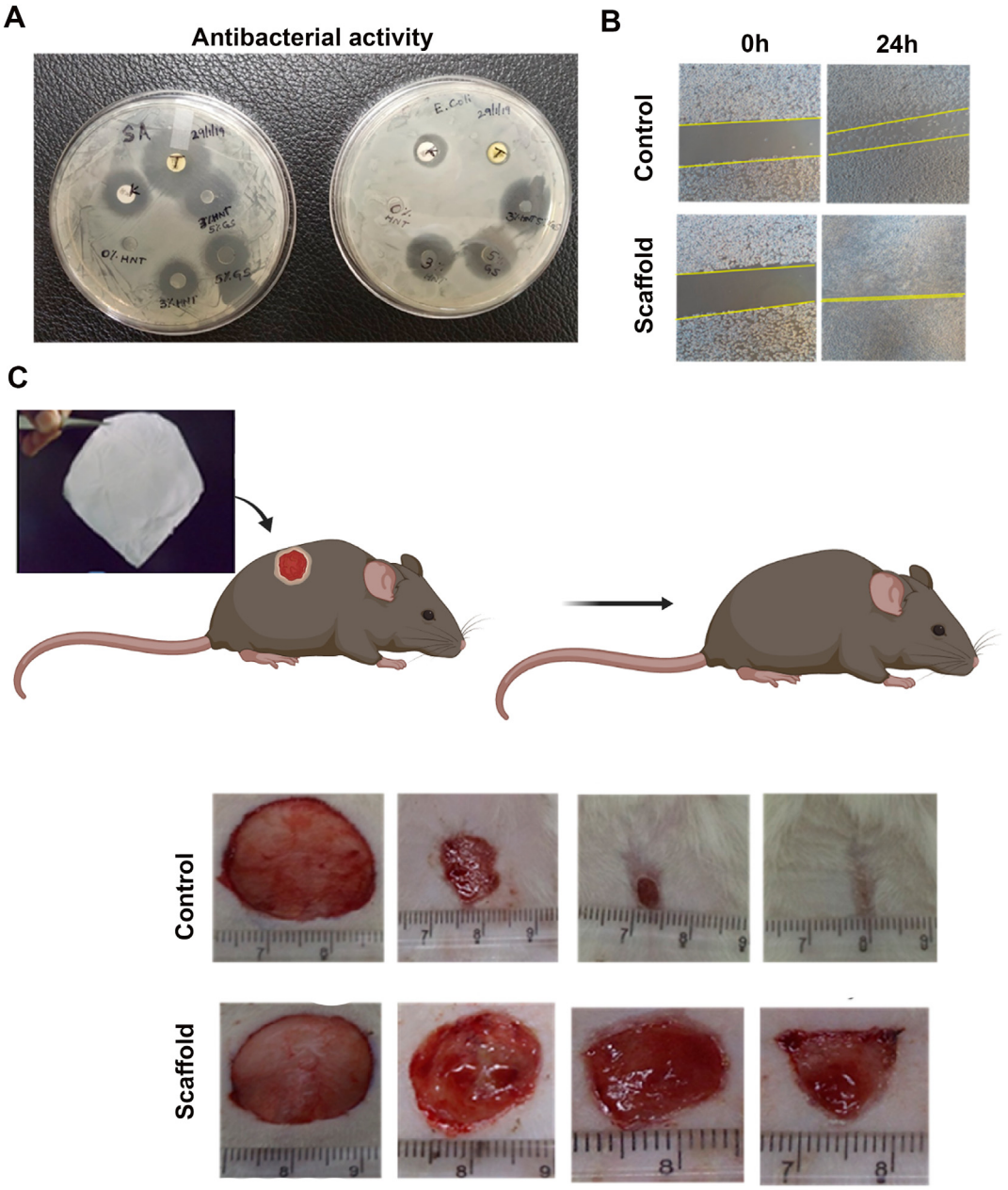

Fig. 3. Antibacterial activity (A), in-vitro cell migration (B), and in-vivo wound healing experiments of the control and the scaffold.
Reprinted with permission from "A. Wali, M. Gorain, S. Inamdar, G. Kundu, M. Badiger, In Vivo Wound Healing Performance of Halloysite Clay and Gentamicin-Incorporated Cellulose Ether-PVA Electrospun Nanofiber Mats, ACS Applied Bio Materials. 2 (2019) 4324–4334" Ref. [68], Copyright (2019) American Chemical Society.

found in wheat, maize, rice, potato, sago, tapioca/cassava, rye, barley and oats. The ratios of amylase to amylopectin vary in the starches, depending on their sources. In addition to food applications, starch and its chemical derivatives bearing carboxyl, acetyl and hydroxypropyl functional groups synthesized via cross-linking, grafting, and esterification have been widely studied in environmental and biomedical sectors [74]. A comprehensive review on the chemical derivatives of starch, especially by etherification and esterification, has been published recently [75].

4.2.2. Electrospinning properties of starch

Electrospinning of starch and its derivatives has been documented in the literature and among patented technologies [76,77]. An example by Kong et al. [78] prepared pure electrospun starch fibers where the main criterion to be satisfied for effective electrospinning is that the polymer concentration must be double the entanglement concentration.

In another study, electrospinning solutions were prepared by dissolving high amylose content starch (15% w/w) in a 95% aqueous DMSO solution [79]. This method was more effective than electrospinning starch with inclusion complexes (e.g. palmitic acid) in ethanol or ethanol/water systems [80]. Kong et al. [81] systematically investigated molecular entanglement, conformation, and shear viscosity, all critical factors for the electrospinning of starch along with a solvent (DMSO). The research established that the starch concentration should be in excess of 1.2–2.7 times that of the entanglement concentration.

High-amylose Hylon VII maize starch together with nanographene oxide (nGO) derived from starch (via carbonization) have been electrospun, using formic acid as the solvent for electrospinning [82]. Additionally, the integration of nGO into starch resulted in enhanced electrospinnability, reduced fiber diameter, improved hydrophilicity and thermal stability. Jeong et al. [83] reported on combining starch-

derived carbon nanofibers (CNFs) with carbon nanotube (CNT) for application in rechargeable batteries. The fabricated electrode exhibited an outstanding performance (510 mAh g^{-1} after 30 cycles), which may be attributed to its large surface area.

4.3. Pectin

Pectin is a natural hetero-polysaccharide from of terrestrial plant tissue, largely composed of poly (α-1,4-D-galacturonic acid) as a main chain, with insertions of α-1,2-L-rhamnopyranose units [84]. The major commercial sources of pectin are citrus peel, apple pomace, and sugar beet pulp. Pectin is utilized in the food industry as a thickening, gelling, and stabilizing agent, in addition to its huge potential in biomedical applications. The viscosity of pectin can be increased by the addition of metal salts [85].

4.3.1. Electrospinning of pectin

Pectin with PEO (polyethylene oxide) as a copolymer together with Triton X-100 as an additive, dispersed in a DMSO/aqueous mixture was successfully electrospun into fibers [86]. Pectin (5–8 wt%) and PEO (5 wt%) both in an aqueous solution were prepared separately and mixed in various weight ratios (pectin/PEO: 60/40, 70/30, 80/20 and 90/10) and subsequently blended with 1 wt% and 5 wt% of Triton X-100 and DMSO, respectively. The dispersions were then mixed for 3 h before electrospinning. Except for the 90/10 ratio of pectin/PEO, all of the other combinations of these two components of the mixtures yielded nanofibers that were both uniform and smooth. These nanofibers were treated with DCM for the elimination of PEO and other surfactants from the electrospun fibers. Electrospinning of charged natural polysaccharides such as chitosan and alginate etc. on their own proved to be difficult, emphasizing the need for support from the co-polymers, additives and suitable solvents. This is due to the fact that such systems having complex molecular chain entanglements in solution and possess diverse rheological properties [87]. In another study [88], pectin nanofibers were cross-linked using Ca^{2+}, glutaraldehyde, and adipic acid dihydrazide (ADH) to evaluate the physical and chemical, mechanical and biomedical characteristics of these generated nanofibers (Fig. 4A and B).

The investigations have revealed that cross-linking ADH enhanced the mechanical strength of the nanofibers appreciably, while simultaneously decreasing the rate of degradation, reducing cytotoxicity, increasing cell adhesion and promoting the proliferation of the pectin nanofibers.

Cui et al. [89] determined folic acid entrapped on electrospun pectin/alginate/PEO nanofibers. Furthermore, NMR and FTIR suggested that the entrapment between the fiber and folic acid was physical in nature. The same authors investigated the in vitro release of folic acid under different pH conditions [90]. The ensuing data confirmed the findings of these studies, which implied that sodium alginate/pectin electrospun fibers were prospective carriers of folic acid. Moreover, Rockwell et al. [91] fashioned electrospun fibers with different degrees of esterification from pectin extracted from various sources (apple pomace, citrus peel, and sugar beet pulp) combined with PEO. Various morphologies, fiber diameters, and tensile characteristics were exhibited by the three types of nanofibers generated by electrospinning. The result showed that the degree of esterification and crystallinity of the pectin fractions influenced the fiber characteristics such as their diameter.

5. Polysaccharides from seaweeds

5.1. Alginate

5.1.1. Structure and properties

The major source of alginates is marine brown algae (*Phaeophyceae*) although it is also produced by some bacteria. Structurally, alginates comprise α-L-guluronic and 1,4 linked-β-D-mannuronic acid residues and these marine polysaccharides possess high viscosities at low concentrations (Fig. 5) [92]. Alginates display very interesting properties, which include water holding capability, stabilizing, and sol gelling abilities, all of which account for their numerous applications in food, medicine, and biotechnology fields [93].

5.1.2. Electrospinning of alginate

Due to repulsive forces between the polyanionic chains in alginate, flexible and uncharged copolymers such as PVA or PEO are added to decrease these forces via hydrogen bonds. This facilitates the chain entanglement, enabling the alginate to be successively electrospun into fibers [6]. Electrospinning of alginates has been conducted in combination with aqueous samples of PVA, PEO, glycerol, acetic acid, citric acid, ethanol and chitosan. Fu et al. [94] fabricated an antibacterial wound dressing membrane consisting of PVA, sodium alginate and moxifloxacin hydrochloride via electrospinning.

Nie et al. [87] illustrated a method for electrospinning sodium alginate in aqueous solution with glycerol as a partner solvent. The addition of glycerol to the sodium alginate solution greatly influenced the electrospinning parameters. Furthermore, the polymeric chain entanglement of alginate in the presence of glycerol resulted in significant improvement and the enhanced flexibility of the polymeric chains. The research highlighted the importance of co-solvents or additives, which are key factors in the regulation of their solution properties and rheological characteristics.

Bonino et al. [95] used high and low molecular weight alginates, combined with PEO and additives to generate uniform fiber morphology compared with blends consisting of alginate. This type of alginate, which only yielded electrospun fibers devoid of PEO/Pluronic F127 materials, is in great demand, especially for in vivo biomedical applications.

Nista et al. [96] illustrated the preparation of coaxial nanofibers from alginate and chitosan with PEO as an additional co-polymer and ethanol as a co-solvent in aqueous media. Core-shell type nanowires were obtained by mixing sodium alginate (2.4%) with PEO (1.6%) at a ratio of 1:1 (w/w), in an aqueous solution of 5% ethanol and chitosan (5%)/PEO (4%), 70:30 (w/w) as shell structure nanofibers. Carbon-based nanostructures derived from alginate are also noteworthy, as Li et al. [97] reported on the novel synthesis of *N*-doped CNFs with high number of mesopores, which exhibited outstanding performance in oxygen reduction reaction, lithium-ion batteries, and supercapacitors.

A green seaweed *Ulva rigida* sourced from Ulvan, an acidic sulfated bio-macromolecule, was successfully electrospun into fibers by Toskas et al. [98]. Various blends of Ulvan/PVA solutions were prepared and the resultant nanofibers had a diameter of ~84 nm by imparting a higher concentration of the polysaccharide utilized by co-blending with PVA; ensuing fibers have been used for biomedical applications such as drug delivery. Further exploration of the research based on Ulvan, biocompatible synthetic polymers such as PEO or polycaprolactone (PCL), and Ulvan nanocomposite fibers were fabricated via electrospinning [99]. The developed interconnected spider-web-like morphology fibers highlighted that the diameter of Ulvan/PCL nanofibers decreased as the Ulvan content was increased in the blend; diameter of the fibers was 300 ± 65 nm with Ulvan/PCL blended in a 3:2 ratio. Green electrospinning of Ulvan with PEO using water as solvent also generated a diameter of 935 ± 65 nm (Ulvan/PEO:1:1) and 505 ± 65 nm (Ulvan/PEO:2:1), respectively. Due to the exceptional properties such as anti-thrombogenic, and biodegradation, the ensued Ulvan/PCL or Ulvan/PEO electrospun fibers can be applied for biomedical applications (drug release, wound healing, and tissue engineering).

5.2. Carrageenan

5.2.1. Structure and properties

Red seaweed is a major source of carrageenan, which can be divided into kappa, iota and lambda carrageenan. The dominant structural units

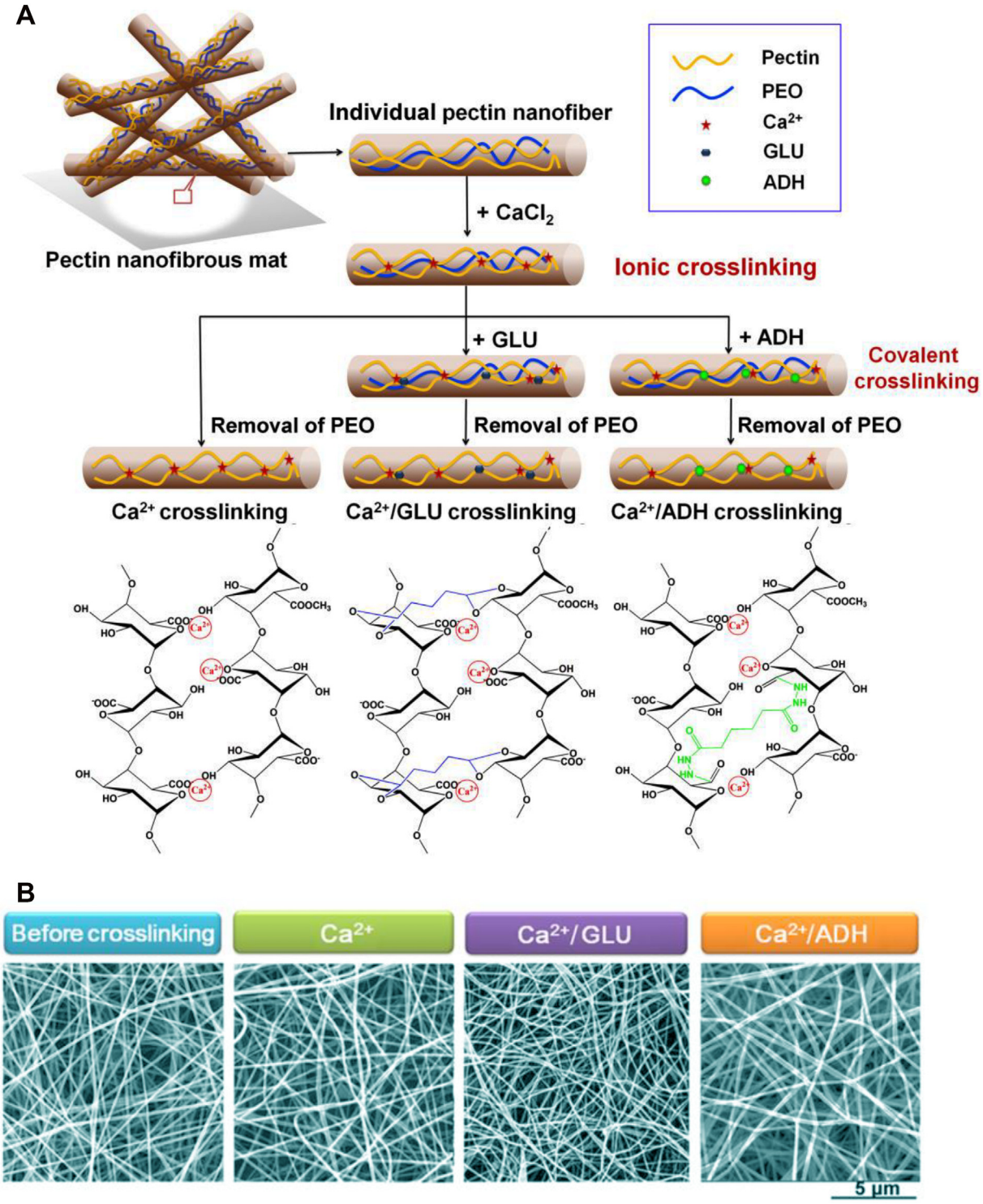

Fig. 4. (A) Schematics and (B) SEM images of individual cross-linked nanofibers with Ca^{2+}, glutaraldehyde (GLU and adipic acid dihydrazide (ADH). Reprinted with permission from Ref [88]. Copyright 2017 Elsevier.

in kappa carrageenan comprise repeating disaccharide units of alternating (1 → 3)-α-D-galactose-4-sulphate and (1 → 4)-β-3,6-anhydro-D-galactose, joined in a linear chain (Fig. 5) [100]. The kappa carrageenan shows non-toxic, ionotropic, thermotropic, chelating and encapsulating behavior, as well as thickening and stabilizing abilities. Therefore, it is not surprising that kappa carrageenan serves as a scaffold in tissue engineering and has the capacity to immobilize enzymes, whole cells, proteins, and biocatalysts [101].

5.2.2. Electrospinning of carrageenan

Goonoo et al. [102] prepared electrospun fibers comprising kappa carrageenan and fucoidan with poly(ester-ether) polydioxanone in varying ratios and in two different solvents (1,1,1,3,3,3-hexafluoroisopropanol (HFIP) and chloroform). Manufactured electrospun mats were configured for cell culture studies and utilized in tissue engineering.

Electrospun fibers composed of a mixture of kappa carrageenan, polyhydroxybutyrate (PHB) or polyhydroxybutyrate valerate (PHBV)

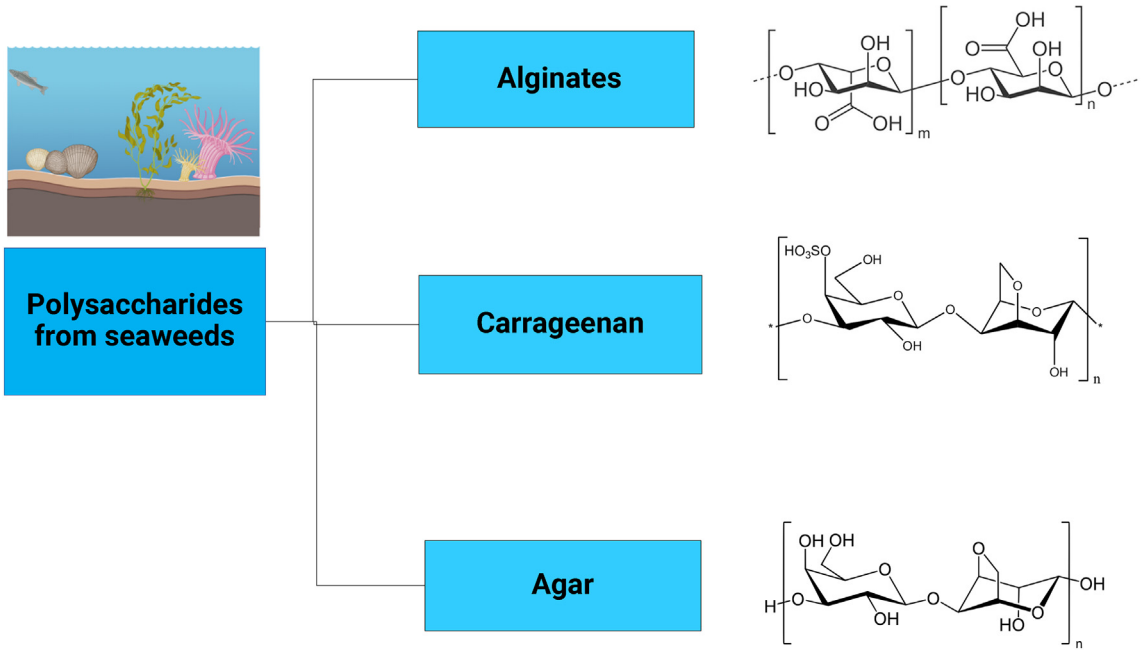

Fig. 5. Seaweed-based macromolecules and their structures.

blended in solvent systems HFIP and chloroform have been fabricated [103]. Both types of fibers (PHB/kappa-carrageenan and PHBV/kappa carrageenan) exhibited excellent bioactivity and were consequently used as scaffolds in bone tissue engineering.

Recently, innovative research has appeared for kappa carrageenan in combination with ZnO nanoparticles and rosemary essential oil [104]. ZnO nanoparticle and rosemary essential oil served as active agents, which can be applied for novel packaging system. The limitations of kappa carrageenan (poor mechanical and barrier properties) were circumvented by making reinforced zein electrospun fiber containing ZnO nanoparticles and rosemary essential oil.

5.3. Agar

5.3.1. Structure and properties

Agar is a polysaccharide extracted from red seaweeds (Gelidium species). It consists of linear chains of β (1 → 3) linked galactose and (1 → 4) linked 3,6-anhydro-α-galactopyranose repeat units, forming a double helix and is widely used as a culture medium for the growth of microorganisms (Fig. 5). It is mainly deployed in food industries as a thickener and clarifying agent [105]. Agarose, one of the major components of agar, has been extensively utilized in important areas such as cell encapsulation, topical, and injectable applications [106].

5.3.2. Electrospinning of agar

Forget et al. [107] designed electrospun fibers of carboxylated agarose in the ionic liquid solvent 1-butyl-3-methylimidazolium chloride. This overcame three major obstacles, namely the poor solubility of agarose in organic solvents, viscosity of agarose in aqueous solutions, and the avoidance of highly toxic solvents. The morphology and fiber diameter distribution of various fiber meshes, may influence various physical and chemical properties. Furthermore, the manufactured fibers may be endowed with antibacterial properties for combating microorganisms (*Staphylococcus aureus* and *Pseudomonas aeruginosa*) (Fig. 6).

Agar/polymer-based hybrid electrospun fibers have been devised with the incorporation of ampicillin via electrospinning [108]. Agar (0.5 to 4.0% w/v) and PAN (polyacrylonitrile, 7% w/v) were prepared separately in *N*,*N*-dimethylformamide (DMF). Uniform nanofibers with diameters (30–230 nm) were produced from different concentrations of the mixed solutions of agar (4%) and PAN (3.2%). The prepared hybrid nanofiber possesses high antibacterial efficiency and controlled drug release.

Sousa et al. [109] reported electrospinning of a mixture of agar and PVA in aqueous solution with varying combinations of PVA/Agar (100/0, 50/50, 40/60, 30/70, 20/80 and 0/100, respectively) by maintaining them at 50 °C inside the chamber during the electrospinning process to avoid agar gelation. The result indicates that PVA, the co-polymer, improved the spinning processability of agar, thereby modifying the solution and rheological characteristics. The same authors reported that agar could be electrospun by using (2-hydroxyethyl) trimethylammonium chloride/urea (1:2 molar ratio) as the solvent by replacing the aqueous media [110]. Rheological and solution properties of the agar were greatly influenced by changing the solvent system from water to (2-hydroxyethyl) trimethylammonium chloride/urea. The solvent used was a deep eutectic solvent (DES), which is environmentally-friendly, economical, and easier to prepare than ionic liquids [111].

6. Bacterial polysaccharides

6.1. Bacterial cellulose

6.1.1. Structure and properties

Owing to its unique nano-scaled three-dimensional network structure, bacterial cellulose (BC) has high water retention, high mechanical strength, and outstanding biocompatibility, features which enables it to serve as a natural scaffold material for the regeneration of a wide variety of tissues. BC fibers have a high aspect ratio with a diameter of 20–100 nm. As a result, BC has a very high surface area per unit mass. This quality, combined with its extremely hydrophilic nature, results in a very high liquid loading capacity. The species of bacteria that produce cellulose is generally called *Acetobacter xylinum*, although bacteria of different names have been mentioned in the literature [112,113].

6.1.2. Electrospun bacterial cellulose

One strategy to improve the bioactivity and physical and chemical properties of scaffolds it to enhance their hydrophilicity. For instance, keratin/bacterial cellulose fibers have been fabricated that were

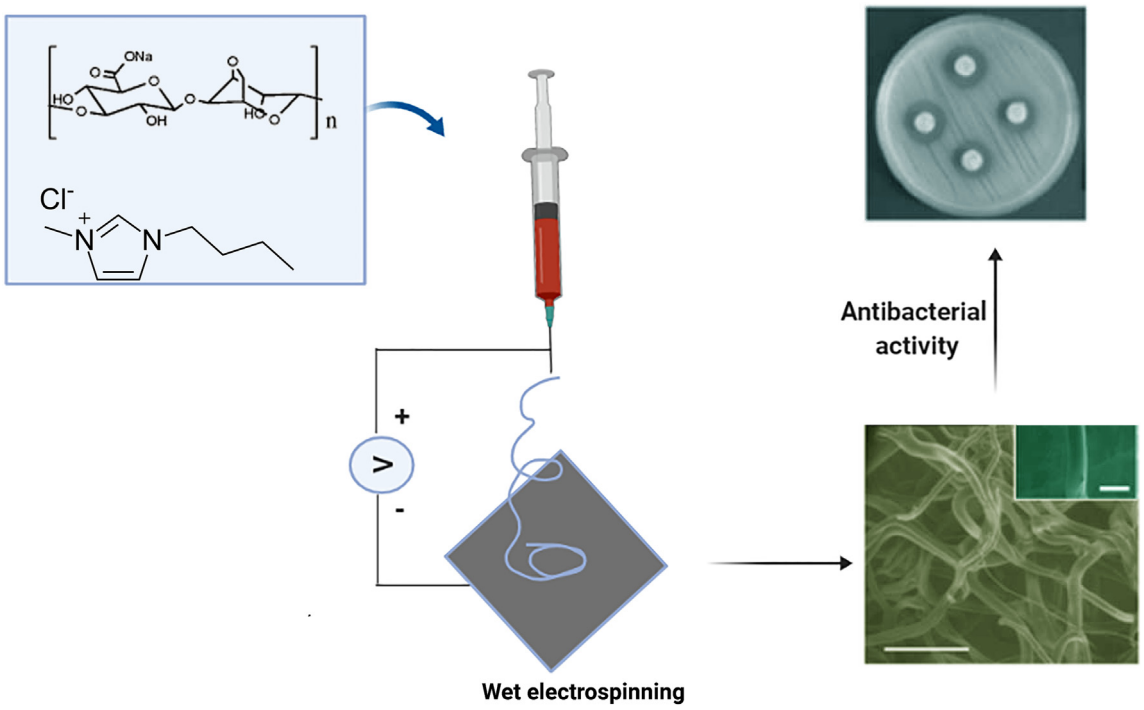

Fig. 6. Schematic illustration of electrospun carboxylated agar and its antibacterial properties against *Staphylococcus aureus*.
Adapted with permission from "A. Forget, N. Arya, R. Randriantsilefisoa, F. Miessmer, M. Buck, V. Ahmadi, D. Jonas, A. Blencowe, V.P. Shastri, Nonwoven carboxylated agarose-based fiber meshes with antimicrobial properties, Biomacromolecules. 17 (2016) 4021–4026". Ref [107]. Copyright (2016) American Chemical Society.

hybridized with hydrogel particles based on polymers conjugated with tragacanth gum. The thermoresponsive hydrogel particles were incorporated into the fibrous network. The scaffold demonstrated higher cell adhesion due to the greater hydrophilicity of the nanofibers (Fig. 7) [114].

6.2. Pullulan (Pu)

6.2.1. Structure and properties

Pullulan (Pu) is an extracellular microbial polysaccharide produced by the fungus *Aureobasidium pullulans*. Structurally, pullulan is an α-

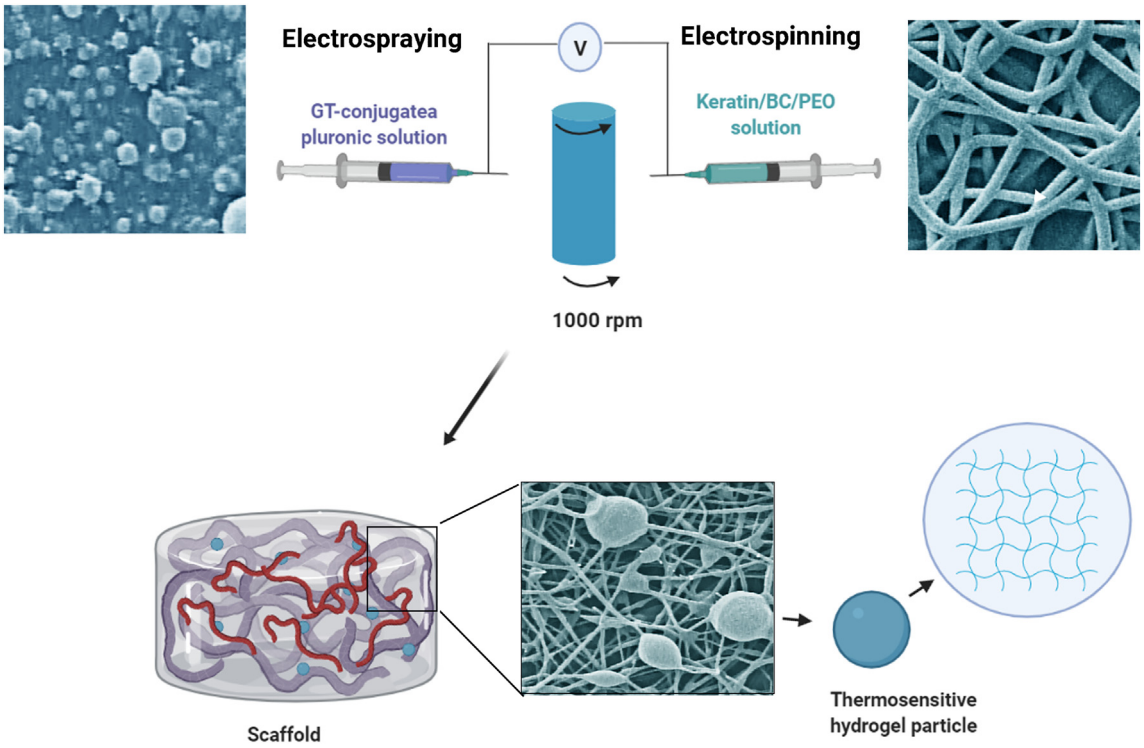

Fig. 7. Schematic illustration of concurrent electrospinning/electrospraying for the fabrication of BC hybrid fibrous/hydrogel matrix.
SEM images reprinted from Ref. [114].

(1 → 6)-linked (1 → 4)-α-D-triglucoside (maltotriose) and depends on its structural flexibility and high solubility in water (Fig. 8) for varied applications [115].

6.2.2. Electrospinning of pullulan

A mixture of pullulan and whey protein isolate (WPI) in varying ratios (viz. pullulan:WPI at 20:80, 30:70, 50:50, 70:30 and 80:20 w/w.) were electrospun into fibers by Drosou et al. [116]. The solvent deployed for electrospinning was distilled water and the effects of various parameters (viscosity, surface tension, electrical conductivity, fiber morphology, flow rate, applied voltage, and tip-to-collector distance) were investigated. The various concentrations of pristine WPI (all with over 40% w/v protein content) cannot be electrospun into fibers, whereas pullulan with 20% (w/v) may easily be electrospun into beadless fibers as it acquires adequate molecular entanglement. The optimum ratio was found to be 50/50 wt/wt of WPI/Pu, highlighting the effectiveness of producing uniform bead-free nanofibers in the range of 231 nm, attributed to improved molecular entanglements through the interactions between both biopolymers. The critical factors for fabricating WPI/pullulan fibers with a uniform morphology and diameter include the blend polymer ratio, flow rate (0.5 mL/h), applied voltage (20 kV), and tip to collector distance (10 cm) [117].

Several pullulan-based multifunctional nanofibers have been well-synthesized by using electrospinning. Jia et al. combined a pea protein isolate together with pullulan [118], which showed better thermal stability and improved hydrophobic properties. Fast dissolving oral films used for drug delivery were also demonstrated by pullulan-based nanofibers [119]. Román et al. employed pullulan nanofibers containing the palindromic peptide LfcinB (21–25)Pal to obtain antimicrobial properties [120]. The study provided a significant milestone in the potential development of antibacterial wound dressings. In another study, pullulan/poly(vinyl alcohol) sponges (with 2–5 μm pores) were fabricated from short electrospun nanofibers with rapid liquid uptake (Fig. 9) [121].

6.3. Dextran

6.3.1. Structure and properties

Dextran is a polysaccharide extracted from bacteria. It has a linear backbone of α-l,6-linked D-glucopyranosyl repeating units, with branches of smaller chains of D-glucose attached to the backbone by α(1 → 2)-, α(1 → 3)- or α(1 → 4)-glycosidic bonds (Fig. 8) [122].

Dextran has high water solubility and its solutions behave as Newtonian fluids. Its solution viscosity is dependent on concentration, temperature and molecular weight, the latter having a characteristic distribution. Due to its biodegradability and biocompatibility, dextran has been used as a food additive and in the pharmaceutical and cosmetic industries. The other applications include biomedical usage as in plasma volume expanders, drug delivery and as protein and enzyme imaging agents. The major functional group in dextran–OH is responsible for conjugation of other molecules. Most importantly, dextran is soluble in both water and organic solvents and may be easily blended with hydrophobic polymers (such as PLGA) to generate composite electrospun fibers [123]. Therefore, it is possible to control the mechanical strength of dextran, its swelling properties in water and its biological activity.

6.3.2. Electrospinning of dextran

Jiang et al. [124] produced electrospun fibers of dextran in various solvent systems (water, DMSO/water and DMSO/DMF) and the process parameters were optimized. An addition of 10% (w/w) of bovine serum albumin (BSA) or lysozyme did not affect the electrospun fibers of dextran. Furthermore, the feasibility of preparing electrospun membranes

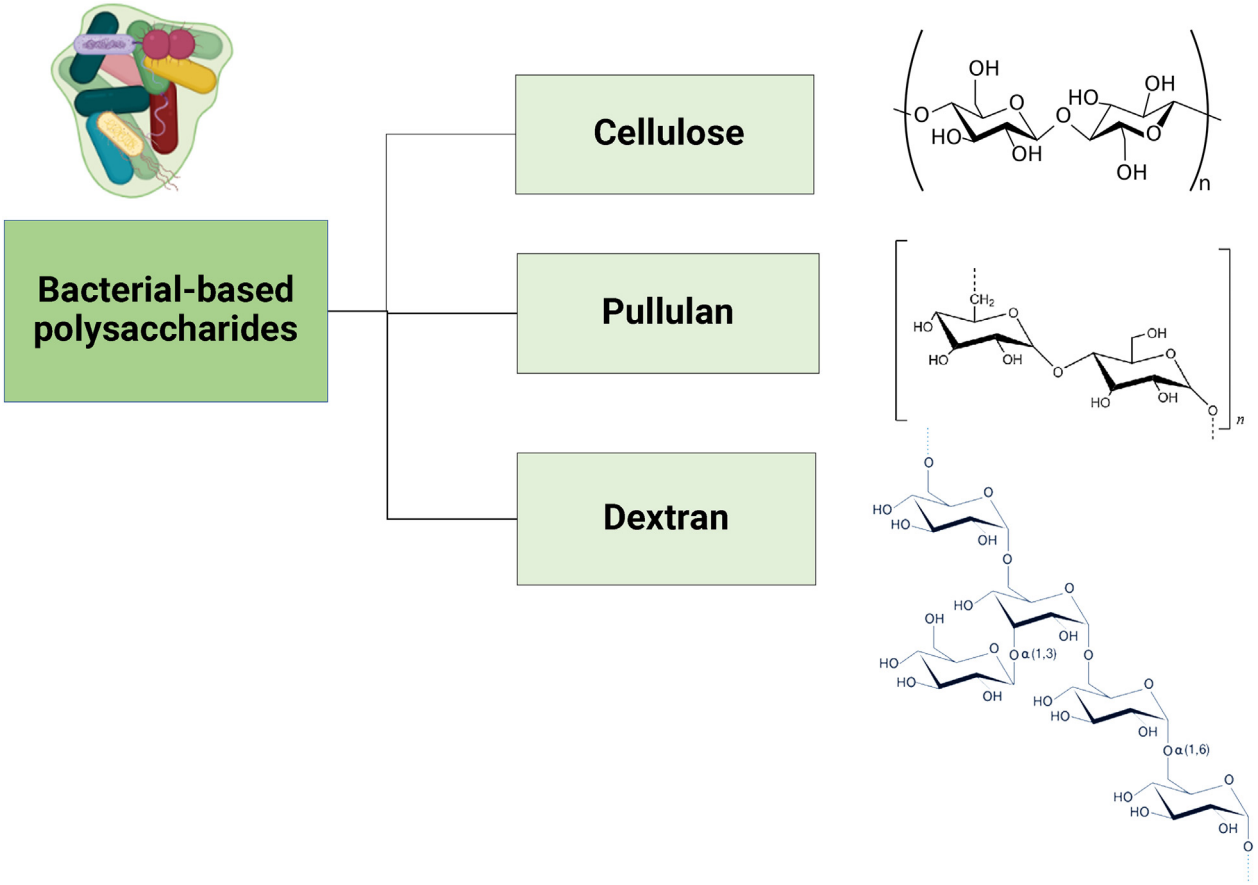

Fig. 8. Bacterial-based macromolecules and their structures.

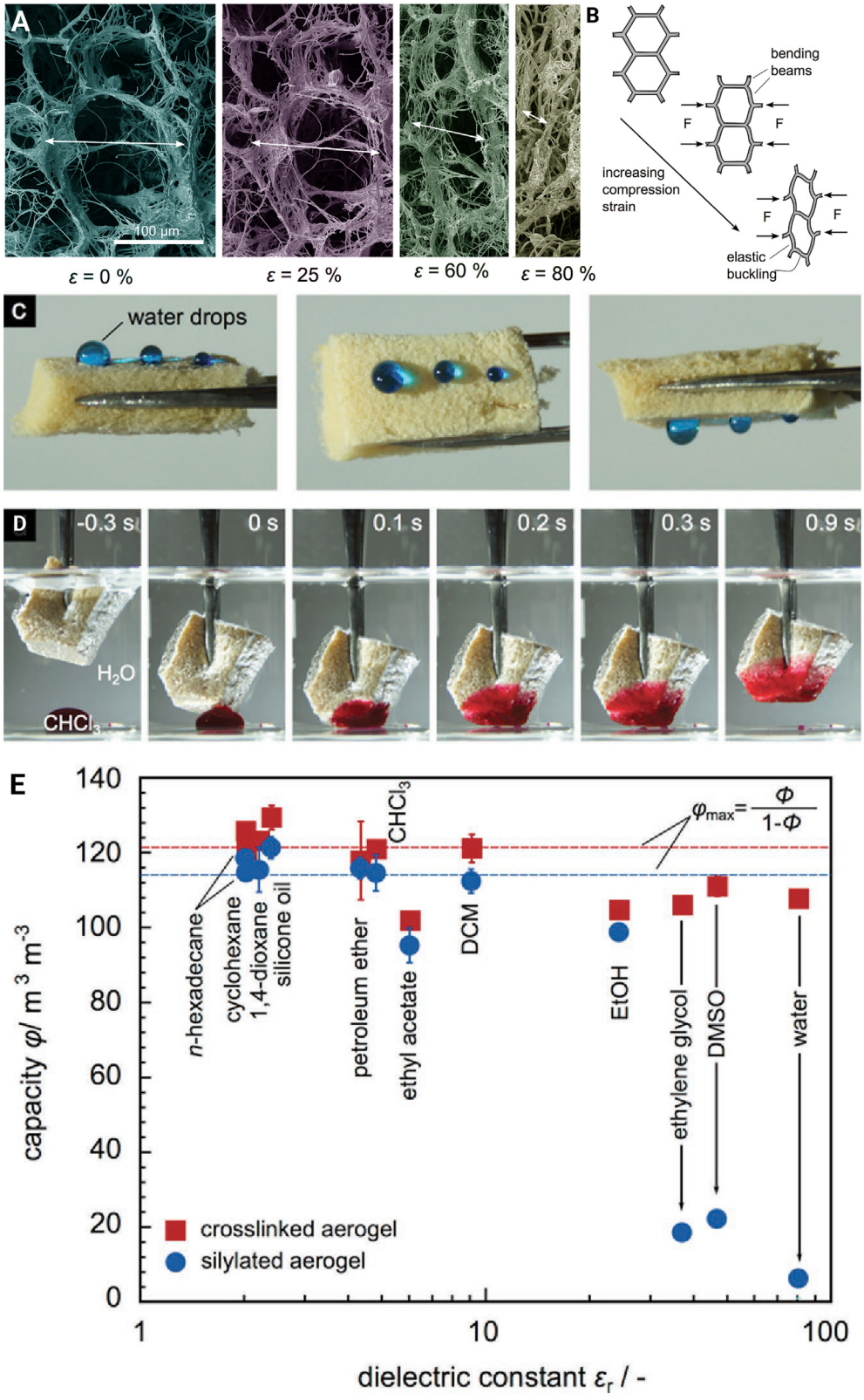

Fig. 9. (A) SEM images of in situ compression test with enhancing compression degree from parallel to the freezing direction. The suggested elastic buckling can be detected by the pore walls of the secondary pores as shown by white arrow; (B) mechanistic illustration to compare the open-pore structures; (C) schematic illustration of water adhesion on the silylated aerogel (rotation from 0° to 180°); (D) rapid and selective uptake of a nonpolar liquid (e.g., $CHCl_3$, dyed with Sudan red IV) from water through the aerogel within 0.3 s. (E) Absorption capacity ($\varphi = V_{liquid} / V_{fiber}$) of amphiphilic cross-linked and silylated pullulan/PVA aerogels). The dashed lines indicate the maximum absorption capacity (φ_{max}) with assuming 100% pore filling with water.
Reprinted with permission from Ref. [121].

via photo cross-linking in the solid state using methacrylated dextran was investigated.

Intriguing dextran-based nanostructures have also been fabricated as shown by Davidson et al. via electrospinning approach to dextran vinyl sulfone polymeric fibers [125] which were found to have modifiable mechanical properties both for single fibers and the bulk matrix. Dextran may also be combined with other polymers, such as polycaprolactone and graphene oxide [126] as exemplified by Nematpour et al. where fabrication of a nanofibrous transdermal patch was accomplished, with tetracycline as the antibacterial agent. These nanofibrous transdermal patches showed remarkably good antibacterial activity against both *E. coli* and *S. aureus*. Poly(vinylpyrrolidinone) (PVP) was also combined with dextran sulfate for application as a drug delivery system [127]. Yang et al. employed electrospinning and photo cross-linking to fabricate gelatin/dextran-maleic anhydride composite fibers [128] where the photo cross-linkable dextran-maleic anhydride was first synthesized, and gelatin was further added. The mechanical properties were greatly enhanced by the introduction of dextran-maleic anhydride, whereby the degradation increased to above 40%. Dextran has been used to further modify a polyurethane fibrous membrane [129], where it exhibited high biocompatibility and enhanced curcumin release. Various studies, ranging from combining dextran with inorganic materials to using dextran for modification, have also been carried out.

7. Polysaccharides from animal sources

7.1. Hyaluronic acid

7.1.1. Structure and properties

Hyaluronan (hyaluronic acid; HYA) is an anionic, non-sulfated glycosaminoglycan is composed of chains of repeating units linking [β-1 → 3] D-glucuronic acid and [β-1 → 4] *N*-acetyl-D-glucosamine (Fig. 10) [130]. It exists in the extracellular matrix of tissues in all higher animal species and is also produced by some bacteria. HYA and its derivatives have potential applications in the fields of medicine (drug delivery and tissue engineering), biotechnology, pharmaceutical technology and cosmetics [131–133].

7.1.2. Electrospinning of hyaluronic acid

Purcell et al. [131] created electrospun fibers of sulfated HYA macromers and these nanofibers were utilized to carry heparin and induce protein interaction, whereby proving their applicability in a variety of fields, and especially in drug delivery and tissue engineering.

Li et al. [134] investigated the electrospinning characteristics of both HYA and blended mixtures of HYA/gelatin in DMF/water-mixed solvent systems with the generation of HYA and HYA/gelatin (with diameters of ~200 nm and 190–500 nm, respectively). In another study [135], gelatin/HYA blended mixtures were electrospun into fibers, which showed that the addition of HYA may improve the electrospinning and processability of aqueous gelatin solutions culminating in the enhancement of viscosity and conductivity of the blended mixtures.

A 'green' procedure based on two biopolymeric system such as hyaluronic acid and chitosan coacervate has been electrospun into nanofibers with an average fiber diameter of 115 ± 30 nm [136]. The effect of salt, solvent (water) and co-solvent (ethanol) on electrospinning parameters such as applied voltage, polymer concentration, viscosity, solvent impacts etc. was studied.

HYA-based nanofibers have been actively employed for biomedical applications. El-Aassar et al. conducted in-vitro and in-vivo studies to examine the wound healing properties of nanofibers comprising polygalacturonic/hyaluronic acid, and silver nanoparticles [137]. HYA-based nanofibers were also further employed for skin tissue engineering [138], which were found to be completely biocompatible. Hyaluronic acid was also immobilized on random or aligned electrospun polylactic acid nanofibers [139], which were capable of capturing cancer cells. Liu et al. synthesized poly(lactide-co-glycolide) grafted hyaluronic acid-derived electrospun nanofibers [140], which showed sustainable anti-infection and immunoregulation properties. In another work by

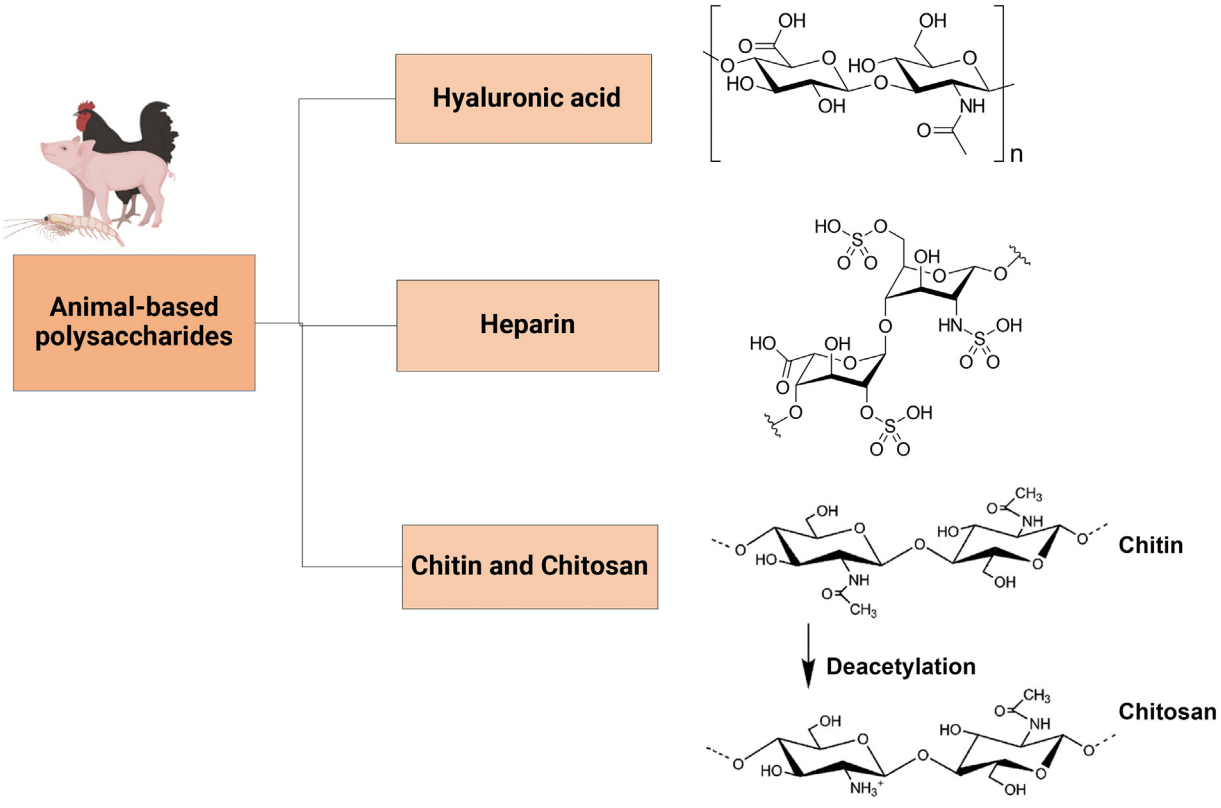

Fig. 10. Biomacromolecules from animal sources and their structures.

Séon-Lutz et al. [141], hyaluronic acid/poly(vinyl alcohol) nanofibers were fabricated by electrospinning, and cyclodextrin was added; ensuing nanofibers exhibited excellent wound dressing properties with a controlled drug release.

7.2. Heparin

7.2.1. Structure and properties

Heparin, a natural glycosaminoglycan, consists of D-glucuronic acid, L-iduronic acid and D-glucosamine units, with a high content of sulfonate and carboxylic functionalities (Fig. 10). Major applications of heparin in the biomedical field rely on its anticoagulant, antiviral, anti-cancer credentials. Heparin also promotes angiogenesis, new blood vessel formation, which is crucial for functional tissue generation [142–145].

7.2.2. Electrospinning of heparin

The major applications of heparin-conjugated electrospun nanofibrous scaffolds are in vascular tissue engineering and delivery systems. Feng et al. [146] studied the development of core-shell type fibers via electrospinning of blended solutions of heparin, Rosuvastatin calcium (an antihyperlipidemic drug) and poly (L-lactide-co-caprolactone) with corresponding solvents. The work highlighted the ability of fabricated electrospun fibers to retain bioactivity to allow sustained release of both heparin and Rosuvastatin calcium and to function efficiently as a scaffold for stent-grafts for the treatment of aneurysms.

Research on heparin is more focused on multicomponent nanofibers as shown by Amand et al. on the electrospun nanofibers made up of hybrid polymers, such as chitosan, polyvinyl alcohol, polyaniline, and polyurethane [147]; they exhibit a kinetic release of drugs, and their cytotoxicity was also assessed. Jin et al. developed small-diameter tissue-engineered vascular grafts composed of heparin, which was further combined with end-capped poly(ε-caprolactone) [148]; they exhibited anticoagulation activity and improved hydrophilicity.

7.3. Chitin and chitosan

7.3.1. Structure and properties

Chitin, an animal polysaccharide, is the second most abundant (after cellulose) natural biological polymer with numerous applications in the biocatalytic, energy storage, filtration, biosensing, and biomedical fields [149]. The structures of chitin are composed of (1, 4)-linked *N*-acetyl-β-D-glucosamine, while its derivative (chitosan) is composed of (1–4)-linked 2-acetamide-2-deoxy-β-D-glucopyranose and 2-amino-2-deoxy-β-D-glucopyranose (Fig. 10) [150].

A major drawback for the electrospinning of chitin and chitosan is the need to conclusively establish an ideal solvent for processing. Furthermore, properties such as the polycationic nature and presence of hydrogen bonds in chitin and chitosan are parameters that may impair their smooth electrospinning [150].

Nevertheless, contemporary research on ionic liquids as viable solvents for both, the chitin and chitosan has been conducted. The objective was to ensure that the physical and chemical properties of the two aforementioned animal-sourced polysaccharides have not been altered, prior to their use in electrospinning.

7.3.2. Electrospinning of chitin

Zavgorodnya et al. [151] devised the electrospinning methodology for three types of chitin [(P-Chitin (Thermally Preprocessed Shrimp Shells), R-Chitin (Unprocessed Shrimp Shells) and PG-Chitin (Practical Grade Chitin)] in the ionic liquid (1-ethyl-3-methylimidazolium acetate). Various factors such as conductivity, polymer concentration, viscosity, and surface tension of the electrospinning solution were studied, which revealed that both, the concentration and viscosity of chitin were key factors influencing the electrospinning process.

Wu et al. [152] contrived an environmentally-friendly filtration membrane composed of poly *N*-isopropylacrylamide-co-*N*-methylolacrylamide (PNIPAm-co-NMA) and chitin nano-whiskers for application in oil-in-water emulsion separation. The electrospun membrane (PNIPAm-co-NMA)/ChNWs) was constructed via electrospinning of PNIPAm-co-NMA and ChNW (chitin nanowhiskers) in a water/THF mixture (at a 1:2 solvent ratio) with a concentration of 15 wt%. Its recyclability and reusability, more than 5 times, is better than other methods for the treatment of oil-in-water emulsions, spanning a broad range of pH values and high salinity atmospheres.

7.3.3. Electrospinning of chitosan

Nanofibers of chitosan blended with the polyelectrolyte poly(acrylic acid) (PAA), have been produced using a ternary solvent system comprising formic acid, glacial acetic acid, and water via electrospinning [153]. The swelling and mechanical properties of the fibers and films were pH dependent. Hence, the promising potential of these materials lies in drug delivery and release.

Koosha et al. [154] fabricated nanofibrous nanocomposites by the electrospinning technique based on chitosan/poly (vinyl alcohol) blended with Na-montmorillonite (MMT) nanoclays. The cytotoxicity of nanocomposite mats was examined by means of an MTT assay using the A-431 cell line. They did not display any detrimental effect on the viability of the cells, with the A-431 cells observed to be properly attached to the surface of the mats. Croisier et al. [155] described chitosan-coated, charged electrospun fibers based on electrospinning a mixture of PCL with a poly(MMA-MA) copolymer of methyl methacrylate and maleic anhydride, making it negatively charged by deprotonation of the carboxylic acid under neutral or basic, aqueous conditions.

Annur et al. [156] formulated electrospun fibers of chitosan/polyethylene oxide (PEO) containing Ag NPs generated by the reduction $AgNO_3$ on the surface of the nanofibers through the application of argon plasma treatment; Ag NPs deposited on the surface of the nanofibers had an average diameter of 1.5 nm and the data showed the enhancement of antibacterial efficiency of the nanofibers after argon plasma treatment, compared to untreated samples. This is ascribed to the strong interaction between Ag NPs and the amino group of chitosan, which in turn brought about uniform exposure of Ag NPs on the nanofiber surfaces.

Chauhan et al. [157] successfully designed biopolymeric nanofibers composed of chitosan, PVA and Fe (III) by electrospinning in an aqueous/acetic acid solution. The ensuing fibers were subsequently utilized for the effective removal of As (III), As (V), Cr (VI) and F^- ions from contaminated water. In addition, they exhibited promising antibacterial potential against *Escherichia coli*, a Gram-negative, optional anaerobic, rod-shaped, coliform bacterium. Chen et al. [158] developed environmentally-friendly fibers via electrospinning, using *N*-maleoyl functionalized chitosan/PVA in an aqueous (water) medium. Cross-linking of the fibers was carried out under UV-irradiation with allyl disulfide as the cross-linker, thereby avoiding harmful reagents. The nanofibers synthesized were infused with suitable antibiotic agents and used in wound healing applications. Makaremi et al. [159] developed electrospun PAN nanofibrous membranes that were functionalized with zinc oxide (ZnO) nanoparticles and coated with a layer of electrospun chitosan. The aim was to improve the mechanical and antibacterial properties, as well as the water filtration performance. The conjugated fiber layers exhibited high mechanical properties (tensile strength and Young's modulus) after the introduction of chitosan fibers. The incorporation of ZnO NPs was found to further enhance the adsorption capacity of Cr (III). Additionally, the antibacterial efficiency and high-water permeability of this sandwiched membrane indicated its suitability for water treatment.

Core-shell nanofibers based on L-ascorbic acid/poly (vinyl alcohol)-chitosan have been controlled via coaxial electrospinning. Acetic acid was used as a shell solution for the chitosan while ethanol/propylene glycol/water served as the core solution for L-ascorbic acid. The release

of ascorbic acid (as a model drug) from the core-shell fibers was evaluated and the results highlighted the suitability of functionalizing such materials for transdermal drug delivery systems [160].

Talebian et al. [161] successfully electrospun a blended solution composed of chitosan-PEO/bioactive glass and functionalized its use as a scaffold in bone tissue engineering. The synthesized composite nanofiber scaffold exhibited better mechanical properties after incorporation with bioactive glass. In view of the good bone-bonding capacity and non-cytotoxicity, it was deemed beneficial for the tissue engineering applications.

Chitosan-based electrospun nanofiber membrane was fabricated as a sorbent for the removal of As(V) from aqueous solutions [162]. The SEM images showed that the chitosan nanofiber membrane was highly porous, which was favorable for the adsorption of As(V) (Fig. 11(A)). It was reported that the solution pH had a key role in the adsorption of As (V) onto chitosan nanofiber membrane, and higher adsorption capacity was observed at lower pH (Fig. 11(B)). The presence of humic acid showed a negative effect on the As(V) adsorption (Fig. 11(C)). Indeed, the presence of humic acid blocked some active adsorption sites of the chitosan nanofiber membrane and had a major impact on the adsorption performance. The humic substances are generally negatively charged in the natural water environment, and it can be electrostatically sorbed by the positively charged adsorbent.

8. Future perspectives: new directions and scope

Successful electrospinning of an abundant number of natural polymers from botanical, animal, bacterial, and seaweed sources is being pursued worldwide. The adopted methodology mainly utilizes natural biomaterials, a selection of solvents (both organic and aqueous) and the maintenance of processing and system parameters appropriate to produce electrospun fibers with uniform dimensions. The major drawbacks of this procedure are the use of imprecise evaluation tools for gauging stability, fiber diameter, aspect ratio and morphology. Also problematic is the fact that owing to the diverse physical, chemical and mechanical properties of the ensuing nanofibers, deciding which particular application best suits them is not a straightforward task.

Many natural biomaterials and their electrospun fibers have been extensively deployed in environmental, energy and biomedical applications (e.g. in tissue engineering scaffolds, drug/gene delivery and wound healing). This is in view of their biocompatibility, non-toxicity, biodegradability and the availability of functional groups of medicinal value in their structures. The important prospects for natural polymer-based electrospun fibers rely on the commercialization of these products in the field of food packaging, tissue engineering and disposable sanitary products. In this context, both academic and commercial organizations and their enhanced networks and interaction among them should strive to promote the newer nanofiber-derived products. To attain the suggested objectives, the highlighted routes to produce biomacromolecules-based electrospun fibers should provide our society with appropriate functional polymeric materials. This will be helpful in the development of sustainable polymeric products to meet economic, environmental, and social needs. However, the substitution of petroleum-based polymers by biopolymers is currently limited by challenges that need to be addressed in the future. These challenges comprise creating simple, low-cost, and less time-consuming methods for all stages of various biopolymer production, search for newer biomaterials or biowaste from nature embracing their isolation, and purification from the bio-sourced materials, synthesis of appropriate forms or chemical modification of natural polymers with desired properties or polymerization of the biomaterials to construct the desirable product. Polysaccharides especially cellulose, chitin, and chitosan are important biomacromolecules because of their high abundance, wide distribution, and low cost of production. Exploiting biomacromolecules, which are copious and can be obtained from biosource material at a lower cost, as the key component of biopolymeric products, can be a smart strategy to alleviate the cost-related matters. Besides, the processes for making the integration of two biopolymeric systems to generate multiple phases of polymer composites fully compatible need to be developed for a variety of biomacromolecules. The above-described challenges and the related

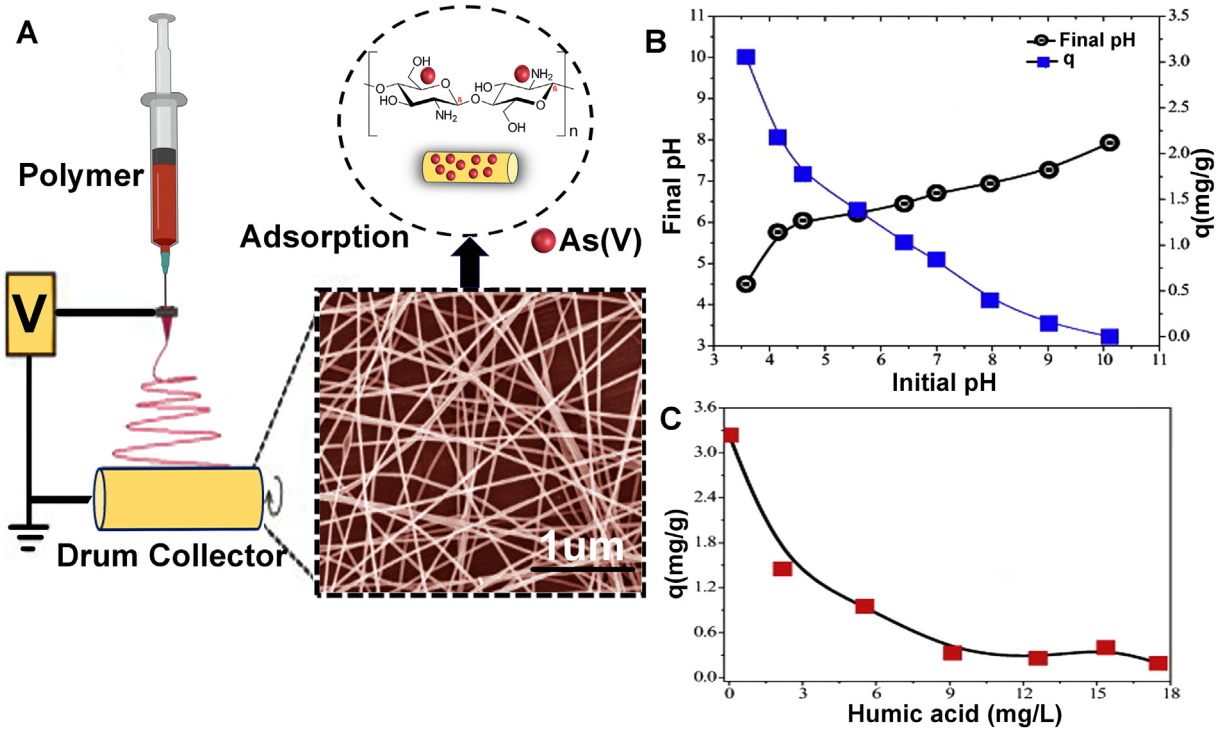

Fig. 11. The schematic illustration of chitosan-based electrospun nanofiber membrane (A), effect of solution pH on As(V) adsorption on the chitosan-based electrospun nanofiber membrane(B), and effect of humic acid on As(V) adsorption on chitosan-based electrospun nanofiber membrane (C).
Reproduced with modification from Ref. [162] with permission from Elsevier.

solutions to these thought-provoking problems require many diverse approaches, based on chemistry, material science, and process engineering. The systematic coordination by researchers and the industry in the planning, selection, and development stages of the production of biopolymer-based electrospun fibers could impart sustainable, competitive and economical advantages so that their use will become more widespread in real-world applications. To create awareness in the society for the deployment of biopolymer-based products and their biodegradability, biocompatibility, abundance, and renewability in nature is also the prime duty of the academician and environmentalists.

NP-functionalized electrospun fibers have proven their worth as highly sensitive conduits for bio-sensing environmental pollutants in water and other applications e.g. as biomarkers for estimating glucose levels in blood plasma. This property may lead to the development of bio-sensing kits for environmental pollutants in drinking water – as well as for the detection of biological fluids in the human body – utilizing eco-friendly and biocompatible electrospun fibers.

Moreover, indications are that natural, biomaterial-based electrospun fibers have a favorable future as replacements of petroleum-based plastics, which have become the scourge of the oceans. The development of bioplastics from biodegradable, biobased electrospun fibers is challenging and further research relating to their application in food packaging, carrier bags and antibacterial pads is imperative if the viability and value of bioplastics are to be firmly established. Natural polymers may then serve as a renewable and highly economical bio-resource for a greener future. Regarding the chemical modification of biomaterials and their electrospinning parameters, the important aspects to be considered are the incorporation of additives or bioactive compounds into the nanofibers and the development of greener solvents for the electrospinning process, especially in applications of nanofibers for the biomedical and food technology sectors, particularly in view of safety concerns.

Electrospun fibers and membranes based on natural polymers/biomaterial functionalized substances will attract much attention in the future in many areas such as filtration/separation, adsorption, catalysis, imaging, energy storage, alternative (greener) corrosion inhibitors, nano-filtration (for removal of nanoparticles, pesticides, pharmaceutical waste, etc.), air filtration, acoustics, a medical diagnosis such as the use of nanofibers coated with antibodies for binding and trapping cancerous cells and the bio-sensing of biological fluids, production of artificial cartilage, delivery of therapeutic drugs, wound healing, tissue engineering, and controlled drug delivery.

Furthermore, the enhancement of the physical and chemical and mechanical properties via surface modification through the adoption of various greener technologies such as plasma, γ-ray irradiation and laser treatment involving chemical grafting, heat treatment and functionalization, will help generate economical, sustainable and bio-organic products with enhanced processability for the future. Improvements in electrospinning techniques and processes such as coaxial electrospinning may also facilitate the preparation of core-shell, multifunctional and layered fibers for advanced applications.

Despite extensive investigations into the electrospinning of natural polymers, the lingering problem has been that without blending them with synthetic polymers or additives, natural polymers alone cannot be electrospun using a 'green' solvent. This is due mainly to critical issues such as high molecular mass, gelling properties, solution behavior and rheological parameters, all of which interfere with the smooth electrospinning of natural polymers. Apart from conventional needle-based electrospinning, other methods such as multiple needle and needle-less electrospinning techniques may favor smooth electrospinning of natural polymers, thereby avoiding blockages incurred by the high viscosity of polymer solutions and their associated gelling properties. Another important avenue for improving the functionality and applicability of 'green' nanofibers based on natural polymers is the fusion of electrospinning with 3D printing technology. Natural polymer-based nanofibers with highly porous carbon nanostructures may facilitate the development of new strategies and help create a framework for the manufacture of environmentally friendly materials for energy harvesting and environmental catalysis applications. They have already found interesting utilization in developing nanofiber-based 3D scaffolds for tissue engineering ventures. The scaling up of production of nanofibers from the laboratory to commercial scale is one of the main challenges. In this regard, multiple needle, needle-less and AC electrospinning systems herald a promising future. The major advantages of developing natural polymer-based fibers that should be emphasized include their minimal effect on the environment in terms of pollution from the solvents, toxic chemicals and flammable gases used in the processing stages, which bodes well for both humans and other living species. The ensuing products and their application will allay safety concerns and consequently be highly sought after in various fields, particularly in the biomedical, food and pharmaceutical sectors.

List of abbreviations

ADH	adipic acid dihydrazide
BC	bacterial cellulose
BSA	bovine serum albumin
CA	cellulose acetate
CTA	cellulose triacetate
CMC	carboxymethyl cellulose
CNF	cellulose nanofibers
Ch	chitosan
ChNWs	chitin nanowhiskers
DMAc	*N*,*N*-dimethylacetamide
DMF	*N*,*N*-dimethylformamide
DMSO	dimethyl sulfoxide
DCM	dichloromethane
FTIR	Fourier transform infrared spectroscopy
HYA	hyaluronic acid
HFIP	1,1,1,3,3,3-hexafluoro-propan-2-ol
MMT	montmorillonite
MEK	methyl ethyl ketone
MMA	methyl methacrylate
MA	methacrylic acid
NPs	nanoparticles
NMR	nuclear magnetic resonance spectroscopy
MeOH	methanol
PLGA	poly(lactic-co-glycolic acid)
PEO	polyethylene oxide
PCL	polycaprolactone
PAN	polyacryloniril
PANI	polyaniline
PVP	polyvinyl pyrrolidone
PVA	poly(vinyl)alcohol
PLA	poly lactic acid
Pu	pullulan
PHB	polyhydroxybutyrate
PHBV	poly(3-hydroxybutyrate-co-3-hydroxyvalerate)
PBS	polybutylene succinate
poly(NIPAAm-co-NMA)	
poly(N-isopropylacrylamide-co-*N*-methylol acrylamide)	
TEMPO	(2,2,6,6-tetramethylpiperidin-1-yl)oxyl
TFE	trifluoroethylene
TEAE	triethylaminoethyl cellulose
THF	tetrahydrofuran
WPI	whey protein isolate

CRediT authorship contribution statement

K.P. Akshaykumar: Writing – original draft, Validation. **Ehsan Nazarzadeh Zare:** Writing – original draft, Validation. **Rafael Torres-Mendieta:** Writing – original draft, Validation. **Stanisław Wacławek:**

Writing – original draft, Validation. **Pooyan Makvandi:** Writing – original draft, Validation. **Miroslav Černík:** Writing – original draft, Validation. **Vinod V.T. Padil:** Writing – original draft, Validation. **Rajender S. Varma:** Writing – original draft, Validation.

Declaration of competing interest

Authors declare no conflict of interest.

Acknowledgements

The authors would also like to acknowledge the assistance provided by the Research Infrastructure NanoEnviCz, supported by the Ministry of Education, Youth and Sports of the Czech Republic in the framework of Project No. LM2018124. This work was also supported by the project "Tree Gum Polymers and their Modified Bioplastics for Food Packaging Application" granted by Bavarian-Czech-Academic-Agency (BTHA) (registration numbers LTAB19007 and BTHA-JC-2019-26) and the Ministry of Education, Youth and Sports in the Czech Republic under the "Inter Excellence – Action Programme" within the framework of project "Bio-based Porous 2D Membranes and 3D Sponges Based on Functionalized Tree Gum Polysaccharides and their Environmental Application" (registration number LTAUSA19091) - TUL internal No.: 18309/136.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijbiomac.2020.12.205.

References

[1] G.A. Valencia, E.N. Zare, P. Makvandi, T.J. Gutiérrez, Self-assembled carbohydrate polymers for food applications: a review, Compr. Rev. Food Sci. Food Saf. 18 (2019) 2009–2024.
[2] A. Venkateshaiah, J.Y. Cheong, S.-H. Shin, K.P. Akshaykumar, T.G. Yun, J. Bae, S. Wacławek, M. Černík, S. Agarwal, A. Greiner, V.V.T. Padil, I.-D. Kim, R.S. Varma, Recycling non-food-grade tree gum wastes into nanoporous carbon for sustainable energy harvesting, Green Chem. 22 (2020) 1198–1208, https://doi.org/10.1039/C9GC04310A.
[3] A. Venkateshaiah, J.Y. Cheong, C. Habel, S. Wacławek, T. Lederer, M. Černík, I.-D. Kim, V.V.T. Padil, S. Agarwal, Tree gum–graphene oxide nanocomposite films as gas barriers, ACS Appl. Nano Mater 3 (2020) 633–640, https://doi.org/10.1021/acsanm.9b02166.
[4] E. Nazarzadeh Zare, P. Makvandi, A. Borzacchiello, F.R. Tay, B. Ashtari, V.V.T. Padil, Antimicrobial gum bio-based nanocomposites and their industrial and biomedical applications, Chem. Commun. 55 (2019) 14871–14885, https://doi.org/10.1039/C9CC08207G.
[5] E. Mele, Electrospinning of natural polymers for advanced wound care: towards responsive and adaptive dressings, J. Mater. Chem. B 4 (2016) 4801–4812, https://doi.org/10.1039/C6TB00804F.
[6] M. Norouzi, S.M. Boroujeni, N. Omidvarkordshouli, M. Soleimani, Advances in skin regeneration: application of electrospun scaffolds, Adv. Healthc. Mater 4 (2015) 1114–1133, https://doi.org/10.1002/adhm.201500001.
[7] R. Sridhar, R. Lakshminarayanan, K. Madhaiyan, V. Amutha Barathi, K.H.C. Lim, S. Ramakrishna, Electrosprayed nanoparticles and electrospun nanofibers based on natural materials: applications in tissue regeneration, drug delivery and pharmaceuticals, Chem. Soc. Rev. 44 (2015) 790–814, https://doi.org/10.1039/C4CS00226A.
[8] A. Farrán, C. Cai, M. Sandoval, Y. Xu, J. Liu, M.J. Hernáiz, R.J. Linhardt, Green solvents in carbohydrate chemistry: from raw materials to fine chemicals, Chem. Rev. 115 (2015) 6811–6853, https://doi.org/10.1021/cr500719h.
[9] E.N. Zare, P. Makvandi, F.R. Tay, Recent progress in the industrial and biomedical applications of tragacanth gum, Carbohydr. Polym. 212 (2019) 450–467, https://doi.org/10.1016/J.CARBPOL.2019.02.076.
[10] V.V.T. Padil, C. Senan, S. Wacławek, M. Černík, S. Agarwal, R.S. Varma, Bioplastic fibers from gum Arabic for greener food wrapping applications, ACS Sustain. Chem. Eng. 7 (2019) 5900–5911, https://doi.org/10.1021/acssuschemeng.8b05896.
[11] V.V.T. Padil, S. Wacławek, M. Černík, R.S. Varma, Tree gum-based renewable materials: sustainable applications in nanotechnology, biomedical and environmental fields, Biotechnol. Adv. 36 (2018) 1984–2016, https://doi.org/10.1016/j.biotechadv.2018.08.008.
[12] K. Liang, P. Han, Q. Chen, X. Su, Y. Feng, Comparative study on enhancing oil recovery under high temperature and high salinity: polysaccharides versus synthetic polymer, ACS Omega 4 (2019) 10620–10628, https://doi.org/10.1021/acsomega.9b00717.
[13] L. Alison, P.A. Rühs, E. Tervoort, A. Teleki, M. Zanini, L. Isa, A.R. Studart, Pickering and network stabilization of biocompatible emulsions using chitosan-modified silica nanoparticles, Langmuir 32 (2016) 13446–13457, https://doi.org/10.1021/acs.langmuir.6b03439.
[14] F. Froiio, A. Mosaddik, M.T. Morshed, D. Paolino, H. Fessi, A. Elaissari, Edible polymers for essential oils encapsulation: application in food preservation, Ind. Eng. Chem. Res. 58 (2019) 20932–20945, https://doi.org/10.1021/acs.iecr.9b02418.
[15] A. Ali, S. Ahmed, Recent advances in edible polymer based hydrogels as a sustainable alternative to conventional polymers, J. Agric. Food Chem. 66 (2018) 6940–6967, https://doi.org/10.1021/acs.jafc.8b01052.
[16] N. Broguiere, E. Cavalli, G.M. Salzmann, L.A. Applegate, M. Zenobi-Wong, Factor XIII cross-linked hyaluronan hydrogels for cartilage tissue engineering, ACS Biomater. Sci. Eng 2 (2016) 2176–2184, https://doi.org/10.1021/acsbiomaterials.6b00378.
[17] E. Ewaldz, B. Brettmann, Molecular interactions in electrospinning: from polymer mixtures to supramolecular assemblies, ACS Appl. Polym. Mater 1 (2019) 298–308, https://doi.org/10.1021/acsapm.8b00073.
[18] J. Xue, T. Wu, Y. Dai, Y. Xia, Electrospinning and electrospun nanofibers: methods, materials, and applications, Chem. Rev. 119 (2019) 5298–5415, https://doi.org/10.1021/acs.chemrev.8b00593.
[19] Z. Wang, S. Li, S. Ge, S. Lin, Review of distribution, extraction methods, and health benefits of bound phenolics in food plants, J. Agric. Food Chem. 68 (2020) 3330–3343, https://doi.org/10.1021/acs.jafc.9b06574.
[20] A. Basu, K.R. Kunduru, E. Abtew, A.J. Domb, Polysaccharide-based conjugates for biomedical applications, Bioconjug. Chem. 26 (2015) 1396–1412, https://doi.org/10.1021/acs.bioconjchem.5b00242.
[21] A. Memic, T. Abudula, H.S. Mohammed, K. Joshi Navare, T. Colombani, S.A. Bencherif, Latest progress in electrospun nanofibers for wound healing applications, ACS Appl. Bio Mater 2 (2019) 952–969, https://doi.org/10.1021/acsabm.8b00637.
[22] P. Wen, Y. Wen, M.H. Zong, R.J. Linhardt, H. Wu, Encapsulation of bioactive compound in electrospun fibers and its potential application, J. Agric. Food Chem. 65 (2017) 9161–9179, https://doi.org/10.1021/acs.jafc.7b02956.
[23] B. Balusamy, O.F. Sarioglu, A. Senthamizhan, T. Uyar, Rational design and development of electrospun nanofibrous biohybrid composites, ACS Appl. Bio Mater 2 (2019) 3128–3143, https://doi.org/10.1021/acsabm.9b00308.
[24] P.S. Kumar, J. Sundaramurthy, S. Sundarrajan, V.J. Babu, G. Singh, S.I. Allakhverdiev, S. Ramakrishna, Hierarchical electrospun nanofibers for energy harvesting, production and environmental remediation, Energy Environ. Sci. 7 (2014) 3192–3222, https://doi.org/10.1039/C4EE00612G.
[25] M. Zhang, X. Zhao, G. Zhang, G. Wei, Z. Su, Electrospinning design of functional nanostructures for biosensor applications, J. Mater. Chem. B 5 (2017) 1699–1711, https://doi.org/10.1039/C6TB03121H.
[26] S. Iravani, R.S. Varma, Plants and plant-based polymers as scaffolds for tissue engineering, Green Chem. 21 (2019) 4839–4867, https://doi.org/10.1039/C9GC02391G.
[27] C. Zhang, H. Zhang, Formation and stability of core–shell nanofibers by electrospinning of gel-like corn oil-in-water emulsions stabilized by gelatin, J. Agric. Food Chem. 66 (2018) 11681–11690, https://doi.org/10.1021/acs.jafc.8b04270.
[28] N. Cai, C. Han, X. Luo, G. Chen, Q. Dai, F. Yu, Fabrication of core/shell nanofibers with desirable mechanical and antibacterial properties by pickering emulsion electrospinning, Macromol. Mater. Eng. 302 (2017), 1600364, https://doi.org/10.1002/mame.201600364.
[29] B. Zaarour, L. Zhu, X. Jin, A review on the secondary surface morphology of electrospun nanofibers: formation mechanisms, characterizations, and applications, ChemistrySelect 5 (2020) 1335–1348, https://doi.org/10.1002/slct.201903981.
[30] C. Zhang, Y. Li, P. Wang, H. Zhang, Electrospinning of nanofibers: potentials and perspectives for active food packaging, Compr. Rev. Food Sci. Food Saf. 19 (2020) 479–502, https://doi.org/10.1111/1541-4337.12536.
[31] Y. Liao, C.-H. Loh, M. Tian, R. Wang, A.G. Fane, Progress in electrospun polymeric nanofibrous membranes for water treatment: fabrication, modification and applications, Prog. Polym. Sci. 77 (2018) 69–94, https://doi.org/10.1016/j.progpolymsci.2017.10.003.
[32] J. Ding, J. Zhang, J. Li, D. Li, C. Xiao, H. Xiao, H. Yang, X. Zhuang, X. Chen, Electrospun polymer biomaterials, Prog. Polym. Sci. 90 (2019) 1–34, https://doi.org/10.1016/j.progpolymsci.2019.01.002.
[33] E.A. Morris, M.C. Weisenberger, M.G. Abdallah, F. Vautard, H. Grappe, S. Ozcan, F.L. Paulauskas, C. Eberle, D. Jackson, S.J. Mecham, A.K. Naskar, High performance carbon fibers from very high molecular weight polyacrylonitrile precursors, Carbon 101 (2016) 245–252, https://doi.org/10.1016/j.carbon.2016.01.104.
[34] G. Yang, X. Li, Y. He, J. Ma, G. Ni, S. Zhou, From nano to micro to macro: electrospun hierarchically structured polymeric fibers for biomedical applications, Prog. Polym. Sci. 81 (2018) 80–113, https://doi.org/10.1016/j.progpolymsci.2017.12.003.
[35] S.-B. Park, E. Lih, K.-S. Park, Y.K. Joung, D.K. Han, Biopolymer-based functional composites for medical applications, Prog. Polym. Sci. 68 (2017) 77–105, https://doi.org/10.1016/j.progpolymsci.2016.12.003.
[36] D. Wang, Y. Yue, Q. Wang, W. Cheng, G. Han, Preparation of cellulose acetate-polyacrylonitrile composite nanofibers by multi-fluid mixing electrospinning method: morphology, wettability, and mechanical properties, Appl. Surf. Sci. 510 (2020), 145462, https://doi.org/10.1016/j.apsusc.2020.145462.
[37] H.P. Karki, L. Kafle, D.P. Ojha, J.H. Song, H.J. Kim, Cellulose/polyacrylonitrile electrospun composite fiber for effective separation of the surfactant-free oil-in-water mixture under a versatile condition, Sep. Purif. Technol. 210 (2019) 913–919, https://doi.org/10.1016/j.seppur.2018.08.053.
[38] Q. Feng, D. Wu, Y. Zhao, A. Wei, Q. Wei, H. Fong, Electrospun AOPAN/RC blend nanofiber membrane for efficient removal of heavy metal ions from water, J. Hazard. Mater. 344 (2018) 819–828, https://doi.org/10.1016/j.jhazmat.2017.11.035.
[39] V.S. Naragund, P.K. Panda, Electrospinning of cellulose acetate nanofiber membrane using methyl ethyl ketone and N,N-dimethylacetamide as solvents, Mater.

Chem. Phys. 240 (2020), 122147, https://doi.org/10.1016/j.matchemphys.2019.122147.
[40] S. Jiang, Y. Chen, G. Duan, C. Mei, A. Greiner, S. Agarwal, Electrospun nanofiber reinforced composites: a review, Polym. Chem. 9 (2018) 2685–2720, https://doi.org/10.1039/C8PY00378E.
[41] L. Kong, G.R. Ziegler, Rheological aspects in fabricating pullulan fibers by electro-wet-spinning, Food Hydrocoll. 38 (2014) 220–226, https://doi.org/10.1016/j.foodhyd.2013.12.016.
[42] L. Kong, G.R. Ziegler, Role of molecular entanglements in starch fiber formation by electrospinning, Biomacromolecules 13 (2012) 2247–2253, https://doi.org/10.1021/bm300396j.
[43] X. Bin Sun, D. Jia, W.M. Kang, B.W. Cheng, Y. Bin Li, Research on electrospinning process of pullulan nanofibers, Appl. Mech. Mater. 268–270 (2012) 198–201, https://doi.org/10.4028/www.scientific.net/AMM.268-270.198.
[44] Z. Qin, X. Jia, Q. Liu, B. Kong, H. Wang, Enhancing physical properties of chitosan/pullulan electrospinning nanofibers via green crosslinking strategies, Carbohydr. Polym. 247 (2020), 116734, https://doi.org/10.1016/j.carbpol.2020.116734.
[45] N. Zhao, Y. Chai, T. Wang, K. Wang, J. Jiang, H. Yang, Preparation and physical/chemical modification of galactomannan film for food packaging, Int. J. Biol. Macromol. 137 (2019) 1060–1067, https://doi.org/10.1016/j.ijbiomac.2019.07.048.
[46] J.M. Frick, A. Ambrosi, L.D. Pollo, I.C. Tessaro, Influence of glutaraldehyde crosslinking and alkaline post-treatment on the properties of chitosan-based films, J. Polym. Environ. 26 (2018) 2748–2757, https://doi.org/10.1007/s10924-017-1166-3.
[47] A. Barhoum, P. Samyn, T. Öhlund, A. Dufresne, Review of recent research on flexible multifunctional nanopapers, Nanoscale 9 (2017) 15181–15205, https://doi.org/10.1039/C7NR04656A.
[48] A. Sato, D. Kabusaki, H. Okumura, T. Nakatani, F. Nakatsubo, H. Yano, Surface modification of cellulose nanofibers with alkenyl succinic anhydride for high-density polyethylene reinforcement, Compos. A: Appl. Sci. Manuf. 83 (2016) 72–79, https://doi.org/10.1016/J.COMPOSITESA.2015.11.009.
[49] L. Heath, W. Thielemans, Cellulose nanowhisker aerogels, Green Chem. 12 (2010) 1448–1453, https://doi.org/10.1039/c0gc00035c.
[50] S. Ge, L. Zhang, Y. Zhang, F. Lan, M. Yan, J. Yu, Nanomaterials-modified cellulose paper as a platform for biosensing applications, Nanoscale 9 (2017) 4366–4382, https://doi.org/10.1039/C6NR08846E.
[51] H. Shaghaleh, X. Xu, S. Wang, Current progress in production of biopolymeric materials based on cellulose, cellulose nanofibers, and cellulose derivatives, RSC Adv. 8 (2018) 825–842, https://doi.org/10.1039/C7RA11157F.
[52] R.J. Moon, A. Martini, J. Nairn, J. Simonsen, J. Youngblood, Cellulose nanomaterials review: structure, properties and nanocomposites, Chem. Soc. Rev. 40 (2011) 3941–3994, https://doi.org/10.1039/c0cs00108b.
[53] K. Ohkawa, Kousaku, Nanofibers of cellulose and its derivatives fabricated using direct electrospinning, Molecules. 20 (2015) 9139–9154, https://doi.org/10.3390/molecules20059139.
[54] H. Lee, M. Nishino, D. Sohn, J.S. Lee, I.S. Kim, Control of the morphology of cellulose acetate nanofibers via electrospinning, Cellulose 25 (2018) 2829–2837, https://doi.org/10.1007/s10570-018-1744-0.
[55] R. Balgis, H. Murata, Y. Goi, T. Ogi, K. Okuyama, L. Bao, Synthesis of dual-size cellulose-polyvinylpyrrolidone nanofiber composites via one-step electrospinning method for high-performance air filter, Langmuir 33 (2017) 6127–6234, https://doi.org/10.1021/acs.langmuir.7b01193.
[56] M.G. Freire, A.R.R. Teles, R.A.S. Ferreira, L.D. Carlos, J.A. Lopes-da-Silva, J.A.P. Coutinho, Electrospun nanosized cellulose fibers using ionic liquids at room temperature, Green Chem. 13 (2011) 3173–3180, https://doi.org/10.1039/c1gc15930e.
[57] M.T. Clough, J.A. Griffith, O. Kuzmina, T. Welton, Enhancing the stability of ionic liquid media for cellulose processing: acetal protection or carbene suppression? Green Chem. 18 (2016) 3758–3766, https://doi.org/10.1039/C6GC00027D.
[58] M.T. Clough, Organic electrolyte solutions as versatile media for the dissolution and regeneration of cellulose, Green Chem. 19 (2017) 4754–4768, https://doi.org/10.1039/C7GC01776F.
[59] R. Konwarh, N. Karak, M. Misra, Electrospun cellulose acetate nanofibers: the present status and gamut of biotechnological applications, Biotechnol. Adv. 31 (2013) 421–437, https://doi.org/10.1016/j.biotechadv.2013.01.002.
[60] K. Kalwar, M. Shen, Electrospun cellulose acetate nanofibers and Au@AgNPs for antimicrobial activity — a mini review, Nanotechnol. Rev. 8 (2019) 246–257, https://doi.org/10.1515/ntrev-2019-0023.
[61] K.N. Kontogiannopoulos, A.N. Assimopoulou, I. Tsivintzelis, C. Panayiotou, V.P. Papageorgiou, Electrospun fiber mats containing shikonin and derivatives with potential biomedical applications, Int. J. Pharm. 409 (2011) 216–228, https://doi.org/10.1016/j.ijpharm.2011.02.004.
[62] M.M. Castillo-Ortega, A.G. Montaño-Figueroa, D.E. Rodríguez-Félix, G.T. Munive, P.J. Herrera-Franco, Amoxicillin embedded in cellulose acetate-poly (vinyl pyrrolidone) fibers prepared by coaxial electrospinning: preparation and characterization, Mater. Lett. 76 (2012) 250–254.
[63] O. Suwantong, P. Opanasopit, U. Ruktanonchai, P. Supaphol, Electrospun cellulose acetate fiber mats containing curcumin and release characteristic of the herbal substance, Polymer 48 (2007) 7546–7557, https://doi.org/10.1016/J.POLYMER.2007.11.019.
[64] X. Wu, C.J. Branford-White, L. Zhu, N.P. Chatterton, D. Yu, Ester prodrug-loaded electrospun cellulose acetate fiber mats as transdermal drug delivery systems, J. Mater. Sci. Mater. Med. 21 (2010) 2403–2411, https://doi.org/10.1007/s10856-010-4100-y.
[65] S. Tungprapa, I. Jangchud, P. Supaphol, Release characteristics of four model drugs from drug-loaded electrospun cellulose acetate fiber mats, Polymer 48 (2007) 5030–5041, https://doi.org/10.1016/J.POLYMER.2007.06.061.
[66] P. Taepaiboon, U. Rungsardthong, P. Supaphol, Vitamin-loaded electrospun cellulose acetate nanofiber mats as transdermal and dermal therapeutic agents of vitamin A acid and vitamin E, Eur. J. Pharm. Biopharm. 67 (2007) 387–397, https://doi.org/10.1016/j.ejpb.2007.03.018.
[67] K. Rodríguez, S. Renneckar, P. Gatenholm, Biomimetic calcium phosphate crystal mineralization on electrospun cellulose-based scaffolds, ACS Appl. Mater. Interfaces 3 (2011) 681–689, https://doi.org/10.1021/am100972r.
[68] A. Wali, M. Gorain, S. Inamdar, G. Kundu, M. Badiger, In vivo wound healing performance of halloysite clay and gentamicin-incorporated cellulose ether-PVA electrospun nanofiber mats, ACS Appl. Bio Mater 2 (2019) 4324–4334.
[69] L. Taajamaa, E. Kontturi, J. Laine, O.J. Rojas, Bicomponent fibre mats with adhesive ultra-hydrophobicity tailored with cellulose derivatives, J. Mater. Chem. 22 (2012) 12072–12082, https://doi.org/10.1039/c2jm30572k.
[70] S.-D. Wang, Q. Ma, H. Liu, K. Wang, L.-Z. Ling, K.-Q. Zhang, Robust electrospinning cellulose acetate@TiO_2 ultrafine fibers for dyeing water treatment by photocatalytic reactions, RSC Adv. 5 (2015) 40521–40530, https://doi.org/10.1039/C5RA03797B.
[71] J. Fu, Z. Pang, J. Yang, F. Huang, Y. Cai, Q. Wei, Fabrication of polyaniline/carboxymethyl cellulose/cellulose nanofibrous mats and their biosensing application, Appl. Surf. Sci. 349 (2015) 35–42, https://doi.org/10.1016/j.apsusc.2015.04.215.
[72] K. Roemhild, C. Wiegand, U.-C. Hipler, T. Heinze, Novel bioactive amino-functionalized cellulose nanofibers, Macromol. Rapid Commun. 34 (2013) 1767–1771, https://doi.org/10.1002/marc.201300588.
[73] W. Kang, X. Ma, H. Zhao, J. Ju, Y. Zhao, J. Yan, B. Cheng, Electrospun cellulose acetate/poly(vinylidene fluoride) nanofibrous membrane for polymer lithium-ion batteries, J. Solid State Electrochem. 20 (2016) 2791–2803, https://doi.org/10.1007/s10008-016-3271-y.
[74] Q. Chen, H. Yu, L. Wang, Z. ul Abdin, Y. Chen, J. Wang, W. Zhou, X. Yang, R.U. Khan, H. Zhang, X. Chen, Recent progress in chemical modification of starch and its applications, RSC Adv. 5 (2015) 67459–67474, https://doi.org/10.1039/C5RA10849G.
[75] A. Gilet, C. Quettier, V. Wiatz, H. Bricout, M. Ferreira, C. Rousseau, E. Monflier, S. Tilloy, Unconventional media and technologies for starch etherification and esterification, Green Chem. 20 (2018) 1152–1168, https://doi.org/10.1039/C7GC03135A.
[76] G. Liu, Z. Gu, Y. Hong, L. Cheng, C. Li, Electrospun starch nanofibers: recent advances, challenges, and strategies for potential pharmaceutical applications, J. Control. Release 252 (2017) 95–107, https://doi.org/10.1016/j.jconrel.2017.03.016.
[77] T. Hemamalini, V.R. Giri Dev, Comprehensive review on electrospinning of starch polymer for biomedical applications, Int. J. Biol. Macromol. 106 (2018) 712–718, https://doi.org/10.1016/J.IJBIOMAC.2017.08.079.
[78] L. Kong, G.R. Ziegler, Quantitative relationship between electrospinning parameters and starch fiber diameter, Carbohydr. Polym. 92 (2013) 1416–1422, https://doi.org/10.1016/J.CARBPOL.2012.09.026.
[79] L. Kong, G.R. Ziegler, Fabrication of pure starch fibers by electrospinning, Food Hydrocoll. 36 (2014) 20–25, https://doi.org/10.1016/J.FOODHYD.2013.08.021.
[80] L. Kong, G.R. Ziegler, Formation of starch-guest inclusion complexes in electrospun starch fibers, Food Hydrocoll. 38 (2014) 211–219, https://doi.org/10.1016/J.FOODHYD.2013.12.018.
[81] L. Kong, G.R. Ziegler, Role of molecular entanglements in starch fiber formation by electrospinning, Biomacromolecules 13 (2012) 2247–2253, https://doi.org/10.1021/bm300396j.
[82] D. Wu, A. Samanta, R.K. Srivastava, M. Hakkarainen, Starch-derived nanographene oxide paves the way for electrospinnable and bioactive starch scaffolds for bone tissue engineering, Biomacromolecules 18 (2017) 1582–1591, https://doi.org/10.1021/acs.biomac.7b00195.
[83] Y. Jeong, K. Lee, K. Kim, S. Kim, Pore-structure-optimized CNT-carbon nanofibers from starch for rechargeable lithium batteries, Materials 9 (2016) 995, https://doi.org/10.3390/ma9120995 (doi:10.3390/ma9120995).
[84] E.G. Maxwell, N.J. Belshaw, K.W. Waldron, V.J. Morris, Pectin — an emerging new bioactive food polysaccharide, Trends Food Sci. Technol. 24 (2012) 64–73, https://doi.org/10.1016/J.TIFS.2011.11.002.
[85] S. Pina, J.M. Oliveira, R.L. Reis, Natural-based nanocomposites for bone tissue engineering and regenerative medicine: a review, Adv. Mater. 27 (2015) 1143–1169, https://doi.org/10.1002/adma.201403354.
[86] S. Cui, B. Yao, X. Sun, J. Hu, Y. Zhou, Y. Liu, Reducing the content of carrier polymer in pectin nanofibers by electrospinning at low loading followed with selective washing, Mater. Sci. Eng. C 59 (2016) 885–893, https://doi.org/10.1016/j.msec.2015.10.086.
[87] H. Nie, A. He, W. Wu, J. Zheng, S. Xu, J. Li, C.C. Han, Effect of poly(ethylene oxide) with different molecular weights on the electrospinnability of sodium alginate, Polymer 50 (2009) 4926–4934, https://doi.org/10.1016/j.polymer.2009.07.043.
[88] S. Cui, B. Yao, M. Gao, X. Sun, D. Gou, J. Hu, Y. Zhou, Y. Liu, Effects of pectin structure and crosslinking method on the properties of crosslinked pectin nanofibers, Carbohydr. Polym. 157 (2017) 766–774, https://doi.org/10.1016/j.carbpol.2016.10.052.
[89] S. Alborzi, L.T. Lim, Y. Kakuda, Encapsulation of folic acid and its stability in sodium alginate-pectin-poly(ethylene oxide) electrospun fibres, J. Microencapsul. 30 (2013) 64–71, https://doi.org/10.3109/02652048.2012.696153.
[90] S. Alborzi, L.T. Lim, Y. Kakuda, Release of folic acid from sodium alginate-pectin-poly(ethylene oxide) electrospun fibers under invitro conditions, LWT Food Sci. Technol. 59 (2014) 383–388, https://doi.org/10.1016/j.lwt.2014.06.008.
[91] P.L. Rockwell, M.A. Kiechel, J.S. Atchison, L.J. Toth, C.L. Schauer, Various-sourced pectin and polyethylene oxide electrospun fibers, Carbohydr. Polym. 107 (2014) 110–118, https://doi.org/10.1016/j.carbpol.2014.02.026.

[92] V. Jakobi, J. Schwarze, J.A. Finlay, K.A. Nolte, S. Spöllmann, H.-W. Becker, A.S. Clare, A. Rosenhahn, Amphiphilic alginates for marine antifouling applications, Biomacromolecules 19 (2018) 402–408, https://doi.org/10.1021/acs.biomac.7b01498.
[93] K. Wang, M.-M. Wang, H.-X. Dou, S.-Y. Xing, B.-L. Zhu, J.-H. Cui, Comparative study on the supramolecular assemblies formed by Calixpyridinium and two alginates with different viscosities, ACS Omega 3 (2018) 10033–10041, https://doi.org/10.1021/acsomega.8b01554.
[94] R. Fu, C. Li, C. Yu, H. Xie, S. Shi, Z. Li, Q. Wang, L. Lu, A novel electrospun membrane based on moxifloxacin hydrochloride/poly(vinyl alcohol)/sodium alginate for antibacterial wound dressings in practical application, Drug Deliv. 23 (2016) 828–839, https://doi.org/10.3109/10717544.2014.918676.
[95] C.A. Bonino, M.D. Krebs, C.D. Saquing, S.I. Jeong, K.L. Shearer, E. Alsberg, S.A. Khan, Electrospinning alginate-based nanofibers: from blends to crosslinked low molecular weight alginate-only systems, Carbohydr. Polym. 85 (2011) 111–119, https://doi.org/10.1016/j.carbpol.2011.02.002.
[96] S.V.G. Nista, J. Bettini, L.H.I. Mei, Coaxial nanofibers of chitosan-alginate-PEO polycomplex obtained by electrospinning, Carbohydr. Polym. 127 (2015) 222–228, https://doi.org/10.1016/j.carbpol.2015.03.063.
[97] D. Li, C. Lv, L. Liu, Y. Xia, X. She, S. Guo, D. Yang, Egg-box structure in cobalt alginate: a new approach to multifunctional hierarchical mesoporous N-doped carbon nanofibers for efficient catalysis and energy storage, ACS Cent. Sci 1 (2015) 261–269, https://doi.org/10.1021/acscentsci.5b00191.
[98] G. Toskas, R.-D. Hund, E. Laourine, C. Cherif, V. Smyrniotopoulos, V. Roussis, Nanofibers based on polysaccharides from the green seaweed Ulva rigida, Carbohydr. Polym. 84 (2011) 1093–1102, https://doi.org/10.1016/j.carbpol.2010.12.075.
[99] S. Kikionis, E. Ioannou, G. Toskas, V. Roussis, Electrospun biocomposite nanofibers of ulvan/PCL and ulvan/PEO, J. Appl. Polym. Sci. 132 (2015)https://doi.org/10.1002/app.42153 n/a-n/a.
[100] S. Muthuswamy, A. Viswanathan, R. Yegappan, V. Selvaprithiviraj, A.K. Vasudevan, R. Biswas, R. Jayakumar, Antistaphylococcal and neutrophil chemotactic injectable κ-carrageenan hydrogel for infectious wound healing, ACS Appl. Bio Mater 2 (2019) 378–387, https://doi.org/10.1021/acsabm.8b00625.
[101] K. Ganesan, L. Ratke, Facile preparation of monolithic κ-carrageenan aerogels, Soft Matter 10 (2014) 3218–3224, https://doi.org/10.1039/c3sm52862f.
[102] N. Goonoo, A. Bhaw-Luximon, U. Jonas, D. Jhurry, H. Schönherr, Enhanced differentiation of human preosteoblasts on electrospun blend fiber mats of polydioxanone and anionic sulfated polysaccharides, ACS Biomater. Sci. Eng 3 (2017) 3447–3458, https://doi.org/10.1021/acsbiomaterials.7b00350.
[103] N. Goonoo, B. Khanbabaee, M. Steuber, A. Bhaw-Luximon, U. Jonas, U. Pietsch, D. Jhurry, H. Schönherr, κ-Carrageenan enhances the biomineralization and osteogenic differentiation of electrospun polyhydroxybutyrate and polyhydroxybutyrate valerate fibers, Biomacromolecules 18 (2017) 1563–1573, https://doi.org/10.1021/acs.biomac.7b00150.
[104] S. Amjadi, H. Almasi, M. Ghorbani, S. Ramazani, Reinforced ZnONPs/ rosemary essential oil-incorporated zein electrospun nanofibers by κ-carrageenan, Carbohydr. Polym. 232 (2020), 115800, https://doi.org/10.1016/j.carbpol.2019.115800.
[105] K. Nishinari, Y. Fang, Relation between structure and rheological/thermal properties of agar. A mini-review on the effect of alkali treatment and the role of agaropectin, Food Struct. 13 (2017) 24–34, https://doi.org/10.1016/j.foostr.2016.10.003.
[106] N. Ninan, A. Forget, V.P. Shastri, N.H. Voelcker, A. Blencowe, Antibacterial and anti-inflammatory pH-responsive tannic acid-carboxylated agarose composite hydrogels for wound healing, ACS Appl. Mater. Interfaces 8 (2016) 28511–28521, https://doi.org/10.1021/acsami.6b10491.
[107] A. Forget, N. Arya, R. Randriantsilefisoa, F. Miessmer, M. Buck, V. Ahmadi, D. Jonas, A. Blencowe, V.P. Shastri, Nonwoven carboxylated agarose-based fiber meshes with antimicrobial properties, Biomacromolecules 17 (2016) 4021–4026, https://doi.org/10.1021/acs.biomac.6b01401.
[108] H. Yang, P.F. Gao, W.B. Wu, X.X. Yang, Q.L. Zeng, C. Li, C.Z. Huang, Antibacterials loaded electrospun composite nanofibers: release profile and sustained antibacterial efficacy, Polym. Chem. 5 (2014) 1965–1975, https://doi.org/10.1039/C3PY01335A.
[109] A.M.M. Sousa, H.K.S. Souza, J. Uknalis, S.C. Liu, M.P. Gonçalves, L. Liu, Electrospinning of agar/PVA aqueous solutions and its relation with rheological properties, Carbohydr. Polym. 115 (2015) 348–355, https://doi.org/10.1016/j.carbpol.2014.08.074.
[110] A.M.M. Sousa, H.K.S. Souza, J. Uknalis, S.C. Liu, M.P. Gonçalves, L.S. Liu, Improving agar electrospinnability with choline-based deep eutectic solvents, Int. J. Biol. Macromol. 80 (2015) 139–148, https://doi.org/10.1016/j.ijbiomac.2015.06.034.
[111] D.H. Kim, K. Shin, H.M. Lees, CO oxidation on positively and negatively charged Ag 13 nanoparticles, J. Phys. Chem. C 115 (2011) 24771–24777.
[112] H.M.C. Azeredo, H. Barud, C.S. Farinas, V.M. Vasconcellos, A.M. Claro, Bacterial cellulose as a raw material for food and food packaging applications, Front. Sustain. Food Syst 3 (2019)https://doi.org/10.3389/fsufs.2019.00007.
[113] G. Chantereau, N. Brown, M.-A. Dourges, C.S.R. Freire, A.J.D. Silvestre, G. Sebe, V. Coma, Silylation of bacterial cellulose to design membranes with intrinsic antibacterial properties, Carbohydr. Polym. 220 (2019) 71–78.
[114] A. Azarniya, E. Tamjid, N. Eslahi, A. Simchi, Modification of bacterial cellulose/keratin nanofibrous mats by a tragacanth gum-conjugated hydrogel for wound healing, Int. J. Biol. Macromol. 134 (2019) 280–289.
[115] S. Farris, L. Introzzi, J.M. Fuentes-Alventosa, N. Santo, R. Rocca, L. Piergiovanni, Self-assembled pullulan-silica oxygen barrier hybrid coatings for food packaging applications, J. Agric. Food Chem. 60 (2012) 782–790, https://doi.org/10.1021/jf204033d.
[116] C. Drosou, M. Krokida, C.G. Biliaderis, Composite pullulan-whey protein nanofibers made by electrospinning: impact of process parameters on fiber morphology and physical properties, Food Hydrocoll. 77 (2018) 726–735, https://doi.org/10.1016/j.foodhyd.2017.11.014.
[117] C.G. Drosou, M.K. Krokida, C.G. Biliaderis, Encapsulation of bioactive compounds through electrospinning/electrospraying and spray drying: a comparative assessment of food-related applications, Dry. Technol. 35 (2017) 139–162, https://doi.org/10.1080/07373937.2016.1162797.
[118] X. Wen Jia, Z. Yu Qin, J. Xin Xu, B. Hua Kong, Q. Liu, H. Wang, Preparation and characterization of pea protein isolate-pullulan blend electrospun nanofiber films, Int. J. Biol. Macromol. 157 (2020) 641–647, https://doi.org/10.1016/j.ijbiomac.2019.11.216.
[119] Z. Qin, X.-W. Jia, Q. Liu, B. Kong, H. Wang, Fast dissolving oral films for drug delivery prepared from chitosan/pullulan electrospinning nanofibers, Int. J. Biol. Macromol. 137 (2019) 224–231, https://doi.org/10.1016/j.ijbiomac.2019.06.224.
[120] J.T. Román, C.A. Fuenmayor, C.M. Zuluaga Dominguez, D. Clavijo-Grimaldo, M. Acosta, J.E. García-Castañeda, R. Fierro-Medina, Z.J. Rivera-Monroy, Pullulan nanofibers containing the antimicrobial palindromic peptide LfcinB (21–25) Pal obtained via electrospinning, RSC Adv. 9 (2019) 20432–20438, https://doi.org/10.1039/C9RA03643A.
[121] F. Deuber, S. Mousavi, L. Federer, C. Adlhart, Amphiphilic nanofiber-based aerogels for selective liquid absorption from electrospun biopolymers, Adv. Mater. Interfaces 4 (2017), 1700065, https://doi.org/10.1002/admi.201700065.
[122] A.J. Manaster, C. Batty, P. Tiet, A. Ooi, E.M. Bachelder, K.M. Ainslie, K.E. Broaders, Oxidation-sensitive dextran-based polymer with improved processability through stable boronic ester groups, ACS Appl. Bio Mater 2 (2019) 3755–3762, https://doi.org/10.1021/acsabm.9b00399.
[123] W.E. Hennink, C.F. van Nostrum, Novel crosslinking methods to design hydrogels, Adv. Drug Deliv. Rev. 64 (2012) 223–236, https://doi.org/10.1016/j.addr.2012.09.009.
[124] H. Jiang, D. Fang, B.S. Hsiao, B. Chu, W. Chen, Optimization and characterization of dextran membranes prepared by electrospinning, Biomacromolecules 5 (2004) 326–333, https://doi.org/10.1021/bm034345w.
[125] C.D. Davidson, D.K.P. Jayco, D.L. Matera, S.J. DePalma, H.L. Hiraki, W.Y. Wang, B.M. Baker, Myofibroblast activation in synthetic fibrous matrices composed of dextran vinyl sulfone, Acta Biomater. 105 (2020) 78–86, https://doi.org/10.1016/j.actbio.2020.01.009.
[126] N. Nematpour, N. Farhadian, K.S. Ebrahimi, E. Arkan, F. Seyedi, S. Khaledian, M. Shahlaei, S. Moradi, Sustained release nanofibrous composite patch for transdermal antibiotic delivery, Colloids Surf. A Physicochem. Eng. Asp. 586 (2020), 124267, https://doi.org/10.1016/j.colsurfa.2019.124267.
[127] A.M. Moydeen, M.S.A. Padusha, B.M. Thamer, A. Ahamed N., A.M. Al-Enizi, H. El-Hamshary, M.H. El-Newehy, Single-nozzle core-shell electrospun nanofibers of PVP/dextran as drug delivery system, Fibers Polym. 20 (2019) 2078–2089, https://doi.org/10.1007/s12221-019-9187-2.
[128] X. Yang, D. Yang, X. Zhu, J. Nie, G. Ma, Electrospun and photocrosslinked gelatin/dextran–maleic anhydride composite fibers for tissue engineering, Eur. Polym. J. 113 (2019) 142–147, https://doi.org/10.1016/j.eurpolymj.2019.01.059.
[129] P. Sagitha, C.R. Reshmi, S.P. Sundaran, A. Binoy, N. Mishra, A. Sujith, In-vitro evaluation on drug release kinetics and antibacterial activity of dextran modified polyurethane fibrous membrane, Int. J. Biol. Macromol. 126 (2019) 717–730, https://doi.org/10.1016/j.ijbiomac.2018.12.155.
[130] H. Kim, M. Shin, S. Han, W. Kwon, S.K. Hahn, Hyaluronic acid derivatives for translational medicines, Biomacromolecules 20 (2019) 2889–2903, https://doi.org/10.1021/acs.biomac.9b00564.
[131] B.P. Purcell, I.L. Kim, V. Chuo, T. Guenin, S.M. Dorsey, J.A. Burdick, Incorporation of sulfated hyaluronic acid macromers into degradable hydrogel scaffolds for sustained molecule delivery, Biomater. Sci. 2 (2014) 693–702, https://doi.org/10.1039/C3BM60227C.
[132] P. Makvandi, G.W. Ali, F. Della Sala, W.I. Abdel-Fattah, A. Borzacchiello, Hyaluronic acid/corn silk extract based injectable nanocomposite: a biomimetic antibacterial scaffold for bone tissue regeneration, Mater. Sci. Eng. C 107 (2020), 110195, https://doi.org/10.1016/j.msec.2019.110195.
[133] P. Makvandi, G.W. Ali, F. Della Sala, W.I. Abdel-Fattah, A. Borzacchiello, Biosynthesis and characterization of antibacterial thermosensitive hydrogels based on corn silk extract, hyaluronic acid and nanosilver for potential wound healing, Carbohydr. Polym. 223 (2019), 115023, https://doi.org/10.1016/j.carbpol.2019.115023.
[134] J. Li, A. He, C.C. Han, D. Fang, B.S. Hsiao, B. Chu, Electrospinning of hyaluronic acid (HA) and HA/gelatin blends, Macromol. Rapid Commun. 27 (2006) 114–120, https://doi.org/10.1002/marc.200500726.
[135] J. Li, A. He, J. Zheng, C.C. Han, Gelatin and gelatin — hyaluronic acid nanofibrous membranes produced by electrospinning of their aqueous solutions, Biomacromolecules 7 (2006) 2243–2247, https://doi.org/10.1021/bm0603342.
[136] J. Sun, S.L. Perry, J.D. Schiffman, Electrospinning nanofibers from chitosan/hyaluronic acid complex coacervates, Biomacromolecules 20 (2019) 4191–4198, https://doi.org/10.1021/acs.biomac.9b01072.
[137] M.R. El-Aassar, O.M. Ibrahim, M.M.G. Fouda, N.G. El-Beheri, M.M. Agwa, Wound healing of nanofiber comprising polygalacturonic/hyaluronic acid embedded silver nanoparticles: in-vitro and in-vivo studies, Carbohydr. Polym. 238 (2020), 116175, https://doi.org/10.1016/j.carbpol.2020.116175.
[138] M. Movahedi, A. Asefnejad, M. Rafienia, M.T. Khorasani, Potential of novel electrospun core-shell structured polyurethane/starch (hyaluronic acid) nanofibers for skin tissue engineering: in vitro and in vivo evaluation, Int. J. Biol. Macromol. 146 (2020) 627–637, https://doi.org/10.1016/j.ijbiomac.2019.11.233.
[139] Y. Zhao, J. Xiong, X. Shi, F. Ko, Capturing cancer cells using hyaluronic acid-immobilized electrospun random or aligned PLA nanofibers, Colloids Surf. A Physicochem. Eng. Asp. 583 (2019), 123978, https://doi.org/10.1016/j.colsurfa.2019.123978.

[140] W. Liu, G. Xi, X. Yang, X. Hao, M. Wang, Y. Feng, H. Chen, C. Shi, Poly(lactide-co-glycolide) grafted hyaluronic acid-based electrospun fibrous hemostatic fragments as a sustainable anti-infection and immunoregulation material, J. Mater. Chem. B 7 (2019) 4997–5010, https://doi.org/10.1039/C9TB00659A.
[141] M. Séon-Lutz, A.-C. Couffin, S. Vignoud, G. Schlatter, A. Hébraud, Electrospinning in water and in situ crosslinking of hyaluronic acid/cyclodextrin nanofibers: towards wound dressing with controlled drug release, Carbohydr. Polym. 207 (2019) 276–287, https://doi.org/10.1016/j.carbpol.2018.11.085.
[142] I. Capila, R.J. Linhardt, Heparin–protein interactions, Angew. Chem. Int. Ed. 41 (2002) 390–412, https://doi.org/10.1002/1521-3773(20020201)41:3<390::AID-ANIE390>3.0.CO;2-B.
[143] S. Mizrahy, D. Peer, Polysaccharides as building blocks for nanotherapeutics, Chem. Soc. Rev. 41 (2012) 2623–2640, https://doi.org/10.1039/C1CS15239D.
[144] X. Liu, L. Yuan, D. Li, Z. Tang, Y. Wang, G. Chen, H. Chen, J.L. Brash, Blood compatible materials: state of the art, J. Mater. Chem. B 2 (2014) 5718–5738, https://doi.org/10.1039/C4TB00881B.
[145] C. Cheng, S. Sun, C. Zhao, Progress in heparin and heparin-like/mimicking polymer-functionalized biomedical membranes, J. Mater. Chem. B 2 (2014) 7649–7672, https://doi.org/10.1039/C4TB01390E.
[146] W. Feng, P. Liu, H. Yin, Z. Gu, Y. Wu, W. Zhu, Y. Liu, H. Zheng, X. Mo, Heparin and rosuvastatin calcium-loaded poly(L-lactide-co-caprolactone) nanofiber-covered stent-grafts for aneurysm treatment, New J. Chem. 41 (2017) 9014–9023, https://doi.org/10.1039/C7NJ01214D.
[147] F. Khalili Amand, A. Esmaeili, Investigating the properties of electrospun nanofibers made of hybride polymer containing anticoagulant drugs, Carbohydr. Polym. 228 (2020), 115397, https://doi.org/10.1016/j.carbpol.2019.115397.
[148] X. Jin, X. Geng, L. Jia, Z. Xu, L. Ye, Y. Gu, A. Zhang, Z. Feng, Preparation of small-diameter tissue-engineered vascular grafts electrospun from heparin end-capped PCL and evaluation in a rabbit carotid artery replacement model, Macromol. Biosci. 19 (2019), 1900114, https://doi.org/10.1002/mabi.201900114.
[149] R. Jayakumar, M. Prabaharan, S.V. Nair, H. Tamura, Novel chitin and chitosan nanofibers in biomedical applications, Biotechnol. Adv. 28 (2010) 142–150, https://doi.org/10.1016/j.biotechadv.2009.11.001.
[150] F. Ding, H. Deng, Y. Du, X. Shi, Q. Wang, Emerging chitin and chitosan nanofibrous materials for biomedical applications, Nanoscale 6 (2014) 9477–9493, https://doi.org/10.1039/C4NR02814G.
[151] O. Zavgorodnya, J.L. Shamshina, J.R. Bonner, R.D. Rogers, Electrospinning biopolymers from ionic liquids requires control of different solution properties than volatile organic solvents, ACS Sustain. Chem. Eng 5 (2017) 5512–5519, https://doi.org/10.1021/acssuschemeng.7b00863.
[152] J.-X.X. Wu, J. Zhang, Y.-L.L. Kang, G. Wu, S.-C.C. Chen, Y.-Z.Z. Wang, Reusable and recyclable superhydrophilic electrospun nanofibrous membranes with in situ co-cross-linked polymer-chitin nanowhisker network for robust oil-in-water emulsion separation, ACS Sustain. Chem. Eng 6 (2018) 1753–1762, https://doi.org/10.1021/acssuschemeng.7b03102.
[153] R.Y. Zhang, E. Zaslavski, G. Vasilyev, M. Boas, E. Zussman, Tunable pH-responsive chitosan-poly(acrylic acid) electrospun fibers, Biomacromolecules 19 (2018) 588–595, https://doi.org/10.1021/acs.biomac.7b01672.
[154] M. Koosha, H. Mirzadeh, M.A. Shokrgozar, M. Farokhi, Nanoclay-reinforced electrospun chitosan/PVA nanocomposite nanofibers for biomedical applications, RSC Adv. 5 (2015) 10479–10487, https://doi.org/10.1039/C4RA13972K.
[155] F. Croisier, P. Sibret, C.C. Dupont-Gillain, M.J. Genet, C. Detrembleur, C. Jérôme, Chitosan-coated electrospun nanofibers with antibacterial activity, J. Mater. Chem. B 3 (2015) 3508–3517, https://doi.org/10.1039/C5TB00158G.
[156] D. Annur, Z.K. Wang, J. Der Liao, C. Kuo, Plasma-synthesized silver nanoparticles on electrospun chitosan nanofiber surfaces for antibacterial applications, Biomacromolecules 16 (2015) 3248–3255, https://doi.org/10.1021/acs.biomac.5b00920.
[157] D. Chauhan, J. Dwivedi, N. Sankararamakrishnan, Facile synthesis of smart biopolymeric nanofibers towards toxic ion removal and disinfection control, RSC Adv. 4 (2014) 54694–54702, https://doi.org/10.1039/C4RA11172A.
[158] C.K. Chen, S.C. Huang, Preparation of reductant-responsive N-maleoyl-functional chitosan/poly(vinyl alcohol) nanofibers for drug delivery, Mol. Pharm. 13 (2016) 4152–4167, https://doi.org/10.1021/acs.molpharmaceut.6b00758.
[159] M. Makaremi, C.X. Lim, P. Pasbakhsh, S.M. Lee, K.L. Goh, H. Chang, E.S. Chan, Electrospun functionalized polyacrylonitrile–chitosan bi-layer membranes for water filtration applications, RSC Adv. 6 (2016) 53882–53893, https://doi.org/10.1039/C6RA05942B.
[160] R. Najafi-Taher, M.A. Derakhshan, R. Faridi-Majidi, A. Amani, Preparation of an ascorbic acid/PVA–chitosan electrospun mat: a core/shell transdermal delivery system, RSC Adv. 5 (2015) 50462–50469, https://doi.org/10.1039/C5RA03813H.
[161] S. Talebian, M. Mehrali, S. Mohan, H. Rao Balaji Raghavendran, M. Mehrali, H.M. Khanlou, T. Kamarul, A.M. Afifi, A.A. Abass, Chitosan (PEO)/bioactive glass hybrid nanofibers for bone tissue engineering, RSC Adv. 4 (2014) 49144–49152, https://doi.org/10.1039/C4RA06761D.
[162] L.-L. Min, Z.-H. Yuan, L.-B. Zhong, Q. Liu, R.-X. Wu, Y.-M. Zheng, Preparation of chitosan based electrospun nanofiber membrane and its adsorptive removal of arsenate from aqueous solution, Chem. Eng. J. 267 (2015) 132–141, https://doi.org/10.1016/j.cej.2014.12.024.