

Development and potential applications of gelatine, honey, and cellulose electrospun nanofibres as a green polymer

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Abstract

Nanofibres have emerged as a brilliant technology to be applied in various areas due to their excellent properties that include having a great flexibility, prominent specific surface area and structural strength. Electrospinning is one of the most effective and favourable methods to fabricate nanofibres mainly because electrospun nanofibres have been demonstrated to possess small pore sizes, large specific surface area, and can be produced with different functions to fill the need of various applications in industries. Due to their remarkable properties, electrospun nanofibres have been proven to be suitable for applications in food packaging, medical, pharmaceutical and even in tissue engineering. Currently, there have been numerous research utilising both electrospun synthetic and natural polymers. Natural or green polymers are considered more favourable due to their biodegradable properties and potential biocompatibility. Therefore, there has been a shift to include more research regarding these green polymers. Green polymers can source from both plant polysaccharides and animal protein. Considering the different characteristics of synthetic polymers, the processing and fabrication methods may differ and must be adjusted accordingly. To well summarise the development of these green polymer nanofibres, we review fabrication methods of gelatine, honey and cellulose-based nanofibre and their potential applications in industries. There are indeed numerous promising areas for the usage of these green polymers which are based on their splendid individual properties especially when combined to form nanofibres via electrospinning. We hope this will promote continuous research and development for the applications in various industries including but not limited to tissue engineering, biomedical, food and pharmaceutical industries.

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1.0 Introduction

Green nanotechnology has transpired as one of the promising areas to be explored as it holds a large range of applications for various fields. One particular field involves the synthesis and utilisation of green polymers. Green polymers, as opposed to synthetic polymers are considered biopolymers which are not only natural but have biodegradable properties and potential biocompatibility. This review will explore three types of green polymers which are cellulose, gelatine, and honey in the form of nanofibres and their potential applications.

An efficient and well-accepted method to synthesise nanofibres is electrospinning. Electrospinning is a simple process which produces

nanofibres from its polymer solution under exposure of high electric field voltage at atmospheric pressure and room temperature (Ibrahim & Klinger, 2020). The usage of electrostatic force produces a membrane or continuous nanofibre mat with sizes in the range of 5–100 nm which is about 10,000 times smaller than fibres fabricated by solution or melt spinning (Kalantari et al., 2019). Better yet, this technique is applicable for both synthetic and natural polymers.

A noteworthy characteristic of nanofibres is the ease of handling and high response rate to external stimuli such as pH, temperature, and chemicals (Bhardwaj & Kundu, 2010). In addition to that, electrospun nanofibres have unique characteristics, such as large surface area to volume ratio, high porosity, and small pore size (Kalantari et al., 2019).

These characteristics enlist the usage of the electrospun nanofibres in numerous applications, including medical, food, pharmaceutical, membranes, drug delivery, wound dressing, and even tissue engineering. In this review, we will discuss the basics for the electrospinning process, the green polymers used in electrospinning, and finally, we will emphasise the various potential applications of this versatile technology.

2.0 Electrospinning technique

2.1 Principle of electrospinning technique

Electrospinning is a very simple technique that can generate fibres with lengths varying from nanometres up to the micrometre range (Bhat et al., 2017). There are three main components of electrospinning; a high voltage power source, the spinneret (or also known as the needle), and a conducting collector (Patil et al., 2017). The utilisation of electro hydrodynamic atomisation method in electrospinning allows it to be used for various scale production of nanofibres in industrial applications (Sabra et al., 2020). When compared to self-assembly and phase separation techniques, electrospinning offers a simpler method in fabricating nanofibres with an interconnected pore structure and fibre diameters in the submicron range (Hasan et al., 2014). During the electrospinning process, the high voltage applied ranging between 10–50 kV produces an electrically charged jet of polymer solution or melt extruding out of the spinneret. Under the high voltage, a cone shaped polymer is formed from the polymer solution droplet and is directed to the counter electrode (Hasan et al., 2014). The general electrospinning configuration is shown in Fig. 1. The electrospinning setup can be designed into two different types of orientations which are the horizontal and vertical configurations, and both these configurations will have an effect towards the resulting nanofibres formed. These arrangements basically refer to the orientation in which the syringe pump and collector are located.

For the horizontal electrospinning technique, the syringe pump is located parallel to the floor and the collector is located perpendicular to the floor, across the syringe tip. Such equipment set up causes the electrical field source being parallel to the floor. In contrast when the resulting electrical field source is perpendicular to the floor; it is named the vertical electrospinning technique. In this technique, the

collector is located on the insulating floor and the syringe pump is positioned above the collector. The collector type typically used is aluminium with a surface area of $15 \times 15 \text{ cm}^2$ (Rodoplu & Mutlu, 2012).

Normally, the syringe pump would be adjusted and positioned to where we would like to start the electrospinning process, which is usually at the centre of the collector plate. For both configurations, gravitational forces which may affect the polymer extruded, is negligible with respect to the larger electric field forces which causes the polymers to spin while electrospinning (Rodoplu & Mutlu, 2012). However, it is important to note that gravity does have an effect towards the shape of the polymer droplet and the Taylor cone. Therefore, other parameters such as the flowrate and critical voltage must be adjusted accordingly (Rodoplu & Mutlu, 2012).

When an electrical force is applied to an ejected polymer solution, its pendant droplet at the tip of the needle is deformed. A charge is generated on the surface of the droplet because of the electrical field. This charge off sets the forces of surface tension and the droplet changes its form from hemispheric to conical. This conical form of the ejected solution is usually referred to as Taylor cone. The conical surface is formed with an angle of 49.3° when the droplet is exposed to an external electrical field (Sill & von Recum, 2008).

When the electrical field voltage exceeds the critical voltage value, the electrostatic forces surpass the surface tension of the polymer solution and cause the ejection of the liquid jet (cone-jet) from the tip of the Taylor cone (Sill & von Recum, 2008). At the tip of the cone from where the jet is started, the maximum

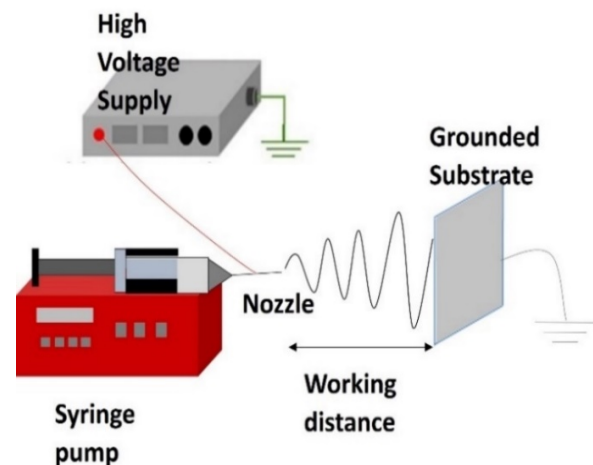


Fig. 1: A horizontal electrospinning configuration where the syringe pump is placed parallel to the floor

charge density is present. Cone-jet speeds up to the grounded collection plate. Besides the electrospinning technique, there are also methods to be used to fabricate hydrophobic surfaces such as the coaxial electrospinning or emulsion electrospinning method (Wang et al., 2019). However, coaxial electrospinning requires the polymers used to have similar dielectric properties. Another modification that can be made is using triaxial electrospinning, in which three polymer solutions are provided to the spinneret (Zhang et al., 2020).

2.2 Effects of various parameters in electrospinning technique

Generally, there are three key conditions in the electrospinning technique that can be tuned to modify the nanostructure of the fibres. These three main parameters are the polymer solution, processing conditions and surrounding environment. Each of these conditions is made up of a few parameters that contributes to the resulting nanofibres produced. Firstly, the factors that can influence the electrospinning polymer solution is the viscosity, concentration, molecular weight, and surface tension of the polymer solution (Patil et al., 2017). Whereby the adjustable parameters for the processing conditions are amount of voltage applied, ejected solution flowrate, and distance between the collector and the needle tip. According to Patil et al. (2017), surrounding temperatures and humidity are also considered contributing parameters for these techniques. All these conditions directly influence the quality of the electrospun nanofibres and finding an optimum setting for these parameters is essential to yield smooth and bead free electrospun nanofibres.

Nowadays, most researchers are conscious and centred on studying the effect of these electrospinning parameters technique towards the chemical and physical properties of final nanofibre structure that has been developed. Most studies characterise the effects of these parameters towards the physical properties of the nanofibre and include theories regarding the manufacturing process. In this review, the corresponding effects of all the parameters towards the development of the nanofibre structure are reported and summarised. Table 1 gathers information from several literatures which use either cellulose, gelatine or honey in their polymer solution and compare the amount of voltage supplied, the flow rate of the ejected polymer solution, the distance of the needle tip to the collector, the ambient conditions and finally the

characteristics of the nanofibre produced.

All these literatures effectively report the parameters used for them to synthesise nanofibres from these green polymers and may act as a basis for future studies to further improve or develop the technique used to produce satisfactory nanofibres. In fact, the nanofibre characteristics presented with each setting allows researchers to plan ahead on the applications of the synthesised nanofibres. The effects of each respective parameter on the electrospinning technique will be further discussed in the sections to follow.

2.3 Effect of the electrical voltage supplied

The application of high voltage is a very important factor in electrospinning and is usually supplied ranging between 7–32 kV. This high voltage connection is usually directed onto the metal needle that functions as a nozzle and the metal plate that functions as a collector. The dynamics of the solution movement from the spinneret are directly affected by the voltage regulation. The droplet shape exiting from the needle tip, dripping raft, rate of surface charge and most significantly the structural morphology are all influenced by these dynamic changes. In addition to that, the charge transport between the needle tip and collector will be generated from the electrical supply. The Taylor cone that forms ultrafine nanofibres at the collector will then be deformed from the dripping of the solution.

The structural morphology that is greatly influenced by the applied voltage is the fibre diameter. Generally, the fibre diameter or size depends on two factors which are the evaporation of the solvent and rate of the charged solution stretching, that is caused by the electrical force. The voltage data influences the fibre diameter by narrowing the fibre dimension with the increase of the applied voltage due to the increasing in stretching of the polymer solution (Law et al., 2017).

Other than that, the upsurge of voltage often results in formation of beaded nanofibres. This is mainly because the formation of Taylor cone will decrease and the corresponding velocity of jet at constant flow rate will increase. Therefore, the voltage applied ultimately affects the morphology of nanofibre, but the changes also depend on the concentration of polymer and the distance between collector and needle tip (Law et al., 2017).

Lin et al. (2008) revealed how the diameter of poly (ether sulfone) nanofibres produced can vary with changes in voltage supplied, and in fact, an optimum voltage to produce the smallest nanofibre diameter can

be obtained. This allows users to plan out the required voltage to acquire a certain sized nanofibre for any application. Thus, there is a need to have the basic information or validate the electrical voltage supplied to produce the nanofibres via electrospinning. From the literatures shown in Table 1, the most common range of voltage used for electrospinning cellulose, gelatine & honey-based nanofibres are in between 15 to 30 kV.

2.4 Effect of ejected flow rate of the electrospinning solution

The flow rate of spinning fluid from the syringe is crucial as it influences the transfer rate of materials and velocity of the jet ejected. The nanofibre dimensions and its diameter formed is affected by the regulation of the flow rate. If the flowrate used is too high and exceeds the critical value, larger diameter fibres with larger pore size and beads will be formed (Qasim et al., 2018). This is because high flow rate would result in the high speed of the jet and it will cause insufficient drying rate of the nanofibre jet during its trip to the collector (Qasim et al., 2018). Hence, to ensure the solution ejected will have enough time for the drying process before it reaches the collector, a lower flow rate is more suitable.

A moderate flow rate can keep the balance between the rate of leaving of the polymeric solution and the fresh polymer solution in the syringe that turn into a new jet at the tip of the nozzle. This effect can produce stable Taylor cone from the tip or sometimes also lead to formation of receded jets (Zhu et al., 2017). Receded jets are unstable formation that emerge directly from the syringe with no other apparent or noticeable droplet. Other than that, the density of the surface charge also correlates with the applied flow rate. The electric current and flow rate are highly correlated to each other which was demonstrated by Theron et al. (2005) in their study which combined experiments and modelling of multiple jets during the electrospinning of polymer solutions. Effects of density of the surface charge and flow rate using different type of polymers were also studied. They found that the increase in electric current and the reduction of density of the surface charge was affected by the increase in the flow rate. Merging of nanofibers could happen during extrusion of the jet with low charge density of the surface.

Considering all the factors that can influence the nanofiber produced, it can be seen that a rather slower flowrate has been preferred for the fabrication of

cellulose, gelatine and honey-based nanofibers via electrospinning which ranges from about 0.5 to 2 mL/h as shown in Table 1.

2.5 Effect of distance between the needle tip to the collector

In the electrospinning technique, one of the key process parameters is the distance between the needle tip and the metal plate collector which is one of the properties that can be tuned to achieve the desired nanofibre structure. This distance also corresponds with the polymer concentration and viscosity of the spinning solution which is similar to other parameter conditions. The deposition time, rate of evaporation, and instability interval or whipping are all affected by the morphology of nanofibre and changes with the distance (Ibrahim & Klingner, 2020). Lin et al., (2008) concluded from their study with poly (ether sulfone) that increasing the tip to collector distance will lead to a decrease in fibre diameter until an optimum value is achieved. After this optimum value, the fibre diameter will increase. Conversely, Ibrahim & Klinger (2020) reported that in studies involving polysulfone fibres, the diameter generally decreases with reduction in distance between the needle tip and the collector. In short, to ensure smooth and uniform nanofibres are obtained, an optimum distance that is suitable with the solution condition needs to be applied (Bhardwaj & Kundu, 2010).

For the green polymers considered in this paper and summarized in Table 1, the common distance between the needle tip to the collector ranges between 10–20 cm which might well be the suggested optimum range for these materials. However, it cannot be denied that the usage of different electrospinning solutions could alter the resulting nanofibre structure morphology. Therefore, it is indeed important to validate each parameter before further experimentation.

2.6 Effect of solution viscosity and polymer concentration

Both polymer solution and concentration are interrelated. This is because the viscosity and concentration of the respective solution depend on the polymer content. During the electrospinning process, the polymer solution is a crucial part in ensuring that the nanofibres can be formed. The viscosity is basically a portrayal of the intermolecular interactions in the polymer solution. The electrospinning technique relies on the ejected charged solution that has uniaxial

Table 1: This table summarises the different parameters that can be tailored or adjusted and the resulting nanofibre characteristics produced for some cellulose, gelatine, and honey-based electrospinning solution

Electrospinning Solution	Voltage (kV)	Flow Rate (mL/h)	Distance of		Ambient Conditions	Nanofibre Characteristics	Ref.
			Tip to Collector (cm)	Tip to Collector (cm)			
<i>Viburnum opulus</i> L/ cellulose acetate; acetic acid/ ethanol	35	3.0	10	-	-	Fibre diameter ranges from 544 ± 117 nm to 292 nm	Çanga & Dudak, 2019
Cellulose acetate; acetaminophen; acetone/ DMF mixture	30 ± 0.2	1.0	8	21 ± 3 °C	21 ± 3 °C	Radius of fibre is shown as 52.9 ± 0.1 nm	Mehrabi et al., 2017
Cellulose acetate/ acetone/ N,N-dimethylacetamide	75–80	2.4–8.6	20	20–22 °C	20–22 °C	Diameter in the range from 0.1–24 µm	Chattopadhyay et al., 2016
Cellulose (neat)	22	0.03–0.10	10	60% relative humidity	60% relative humidity	Interconnected nanofibres	Absar et al., 2015
Cellulose acetate (neat)	25	0.5–1.0	15	35% relative humidity	35% relative humidity	Regular nanofibres	Absar et al., 2015
Cellulose + cisplatin	28	0.05–0.10	10	60% relative humidity	60% relative humidity	Particles attached to fibre surface	Absar et al., 2015
Cellulose + cisplatin	25	0.3	21	35% relative humidity	35% relative humidity	Highly branched fibres	Absar et al., 2015
Gelatine/ polycaprolactone	10	2.0	13	Room temperature	Room temperature	Average fibre diameter of 441.6 ± 118.0 nm	Feng et al., 2019
Ultrafine fish gelatine nanofibre	15	0.3	15	Relative humidity below 60%	Relative humidity below 60%	Increasing fish gelatine concentration from 23.7 to 35.5% (w/v), the average fibre diameter increased from 91.42 to 200.48 nm	Kwak et al., 2017
Gelatine electrospun (GES) nanofibre electrolytes	23	-	7	45–50 °C	45–50 °C	Fibre diameters in the 306.2–428.4 nm range	Kotatha et al., 2019
Gelatine powder in water/acid solvent	25	0.6	10	Room temperature	Room temperature	The average fibre diameter is 95.19 nm	Aliakbarshirazi & Talebian, 2017
Electrospun of polycaprolactone and manuka honey scaffolds	28	4	12.5	Room temperature	Room temperature	The honey scaffolds and control scaffolds had pore sizes ranging from 800 to 7000 µm ²	Minden-Birkenmaier et al., 2015
Honey loaded alginate/ PVA nanofibre	15	0.4	10	Room temperature	Room temperature	Diameter of the nanofibres increase from 379 ± 65 nm to 528 ± 160 nm with the increase of honey concentration	Tang et al., 2019
Manuka honey-cellulose acetate nanofibre	15	0.7	20	Relative humidity of 55%	Relative humidity of 55%	Manuka honey (MH) loaded composite nanofibres diameter increase from 357 to 389 nm and 403 nm as the quantity of the MH in the composite fibres increases from 10 to 20%	Ullah et al., 2020

stretching phenomenon. The viscosity of solution also highly influences the rate of stretching of the ejected solution making it directly proportional to the concentration of the polymeric content. A higher polymeric concentration will result in high viscosity of the spinning solution. The solution that has optimum levels of content, molecular weight and viscosity will result in an optimum surface tension that will improve the stretching rate.

On the other hand, at low solution concentration, the interaction between the high voltage applied and unsuitable surface tension at low solution concentration would cause polymer chains entanglement fracture into fragments during their projection. This fragment breaking phenomenon that causes beads to be formed on the collector plate (Haider et al., 2015; Pillay et al., 2013). By increasing the solution viscosity and the polymeric concentration content, the entanglement of the polymer chain can be improved. The chain entanglement during uniform formation of nanofibre can overcome the surface tension effect. However, if the polymer concentration exceeds the critical concentration of the electrospinning technique, the bead will be formed on the collector. This occurs as the solution becomes difficult to force through a very small syringe needle opening at higher viscosity which cause complications or an unstable control of the flow rate of the solution.

Apart from the formation of beads, the aspect ratio and fibre diameter are also directly proportional to the polymer concentration. There are six types of forces that acts on the charged jet small segment and effects the diameter which are surface tension forces, Coulombic forces, electrostatic forces, viscoelastic forces, gravitational forces and drag forces. However, only surface tension, Coulombic and viscoelastic forces have a strong influence on the ratio of fibre diameter aspect and correlate well with the changes in concentration (Subbiah et al., 2005; Bhardwaj & Kudu, 2010). The effects of viscosity on the nanofibre morphology have also been studied previously and the reported optimum viscosity for a PEO polymeric solution is between 800–4000 p to ensure the uniform nanofibres can be generated (Doshi & Reneker, 1995). Meanwhile, in the same study using a different type of polymer which is polyacrylonitrile (PAN) solution, the optimum viscosity required to generate uniform nanofibres is in the range of 1.7–215 cp (Doshi & Reneker, 1995).

2.7 Effect of humidity

Other than the process and solution conditions, the surrounding conditions also have their own influences on the electrospinning technique. The humidity and temperature are the main parameters that will affect the ambient condition. For example, the humidity affects the electrospinning by interfering with the solidification process of the charged jet to induce the diameter of the nanofibres (Qasim et al., 2018). The solidification process is mainly affected by the nature of the chemical used. This is because, the rate of volatility of the solvent and other chemicals added are different. The evaporation rate is faster when the chemical has a higher rate of volatility when exposed to the ambient atmosphere. As the humidity of surrounding condition will also affect the evaporation rate, there will be some influence of it towards the solidification process.

The effects of humidity specifically on green polymers indeed require further exploration. In a study by Absar et al. (2015), using various cellulose based polymer solutions, the changing relative humidity indeed affected the resulting nanofibre morphology. However, its mechanism that influences the properties of the constructed nanofibre is not well understood.

3.0 Potential applications of green polymer-based nanofibres

There is a wide range of polymers that can be used in electrospinning and are able to form fine nanofibres within the submicron range and used for varied applications. Electrospun nanofibres have been reported as sourcing from various synthetic polymers, natural polymers, or a blend of both including proteins, nucleic acids and even polysaccharides (Bhardwaj & Kundu, 2010). The natural biopolymers are biomolecules produced by living cells and normally exhibit better biocompatibility and low immunogenicity along with efficient interactions with cells by courtesy of bearing a high morphological resemblance to natural extracellular matrix (ECM) as well as high hydrophilicity, compared to synthetic polymers (Heidari et al, 2019). A strong reason for using natural polymers for electrospinning is their inherent capacity for binding cells since they carry specific protein sequences which is the arginylglycylaspartic acid or RGD for short (Pierschbacher & Ruoslahti, 1984). Natural polymers

can source from both plant polysaccharides and animal protein. Examples for plant polysaccharides include starch, cellulose, and chitosan, while animal proteins include silk fibroids, collagen, gelatine, and albumin (Wsoo et al., 2020). For this review, we will focus on three natural polymers which are cellulose, gelatine, and honey. Their development of applications as electrospun nanofibres will be discussed and reviewed.

3.1 Applications of cellulose-based nanofibre

As a natural resource of biodegradable materials, the research and application of cellulose are quite extensive. The separated cellulose, after proper physical and chemical modification, can be made into a variety of fibre films for various purposes such as filtration, biomedical applications, and protective clothing (Lv et al., 2018). They are ideal biodegradable polymer materials and easy to be decomposed by microorganisms.

A trait that needs to be considered regarding cellulose fibres is its hydrophilic nature which leads to a poor interaction between the natural fibres and hydrophobic polymer especially in a composite system (Lv et al., 2018). Nonetheless, this is where electrospinning technique plays an important role as the acetylation of cellulose fibres could improve the interaction with the polymer during the process (Chattopadhyay et al., 2016).

In fact, the hydrophilic features of the cellulose fibres make it a splendid material to be used in absorption of water or other liquids. A study by Yadav et al., (2016) utilised this feature and discovered its potential use for feminine hygiene applications. A challenge with the development of this is that some solvents used for cellulose which are not completely volatile (i.e., ionic liquids) requires an additional coagulation step to stabilise the fibres. Alternatively, there are some cellulose derivatives such as cellulose acetate, hydroxypropyl cellulose and hydroxypropyl methyl cellulose which can be easily dissolved in a variety of volatile solvents rendering them suitable for electrospinning (Yadav et al., 2016).

In addition to that, many cellulose derivatives have been developed to overcome the limited solubility of cellulose in general organic solvents (Wang et al., 2020). Amongst these derivatives, cellulose acetate is biodegradable, insoluble in water, biocompatible non-toxic, has high affinity, good hydrolytic stability, has excellent chemical resistance, and most importantly is easily available as well as has a low cost (Khoshnevisan et al., 2018).

Due to the long list of magnificent qualities, cellulose acetate can be widely applied for many types of potential applications such as antimicrobial membranes, filament-forming matrix, biomedical nanocomposites, affinity membranes, and biomedical separation (Teixeira et al., 2020). In fact, it can also be potentially utilized for wound dressing due to the antimicrobial properties (Teixeira et al., 2020).

Table 2 summarises the various usage of cellulose derivatives together with their nanofibre characteristics. The literature basically highlights the vast potential of using cellulose-based nanofibres as well as how it has progressed to become more favourable to fit various applications ranging from filtration to drug delivery.

3.2 Applications of gelatine-based nanofibre

Gelatine originates from partial hydrolysis of collagen from animal tissues. Since it owns the RGD sequences of collagen which makes it an efficient microenvironment for cell attachment, it has been discovered to be an appropriate candidate for tissue regeneration (Wu et al., 2011). Research involving the electrospinning of gelatine has been performed continuously for a few decades. In fact, due to its low cost, biocompatibility, safety, and biodegradability, gelatine derived from animal tissues have been broadly used in the pharmaceutical, biomedical and food disciplines (Shalumon et al., 2015).

Based on the aforementioned characteristics, gelatine nanofibres is an excellent candidate for wound dressing material. In fact, their lack of antimicrobial properties which limits their use can be resolved by incorporating it with antimicrobial properties (Inal & Mülazımoğlu, 2019). In a study by Inal and Mülazımoğlu (2019) poly([2-(methacryloyloxy) ethyl] trimethyl-ammonium chloride) or PMETAC was used and the nanofibres containing PMETAC showed good anti bactericidal activity particularly against *Staphylococcus aureus*, *Escherichia coli*, methicillin-resistant *Staphylococcus aureus* and *Acinetobacter baumannii* (Inal & Mülazımoğlu, 2019). The findings from that study shows that the obtained nanofibres can be utilised effectively and safely as an antimicrobial wound dressing material.

Gelatine based nanofibre also have a high potential for clinical application as a novel pericardial substitute in cardiac surgery due to its suitability as a barrier to prevent cardiac operative adhesions (Feng et al., 2019).

Table 2: A summary of various applications of cellulose based nanofibres including the characteristics of the nanofibres formed

Polymers	Applications	Nanofibre Characteristics					Ref.
		Size	FTIR	In vitro release assay/ Absorbency	Thermal strength	DSC/ XRD	
<i>Viburnum opulus</i> L (gum arabic)/ cellulose acetate nanofibre	Edible coatings for food industries	From 544 ± 117 nm to 292 nm	Peak height at 1030 cm ⁻¹ demonstrated the presence of GA	n < 0.45 indicating release of fruit extract from fibres	150 and 160 °C	-	Çağa & Dudak, 2019
Cellulose acetate (CA)/ Acetaminophen (A) nanofibre	Delivery of morphine for drug delivery	52.9 ± 0.1 nm	FTIR indicated presence of C-H, OH, C=O, and C-O functional groups	n = 0.24 for conventional nanofibre, n = 0.35 for coaxial nanofibre	-	A highly dispersed in CA nanofibre matrix and present in amorphous mode (R ² = 0.9844)	Mehrabi et al., 2017
Cellulose acetate nanofibre	Aerosol filtration	0.1–1 µm	-	Mean penetration from 0.03–70	-	-	Solidity: 0.1–0.2 and filter thickness: 7–51 µm Chattopadhyay et al., 2016
Cellulose acetate (CA) nanofibre	Feminine sanitary napkin	111.9 ± 88.3 nm	-	Absorption capacity for 180 s was 2333.1%	-	-	29.8 ± 5.5 MPa Yadav et al., 2016
Cellulose acetate-poly-acrylonitrile (CA-PAN) nanofibre	Membrane separation	502 ± 119 nm	Peak at 1090 to 1070 cm ⁻¹ attributing to C-C	-	From 92 to 128 °C	-	E = 1.28 MPa, σ _{max} = 5 MPa, ε _b = 29.2% Wang et al., 2020
Chitosan-bacterial nanocellulose nanofibre	Antibacterial wound dressing	Avg D = 85 nm	Peak at 1735 cm ⁻¹ corresponding to the COO- group	-	-	-	Ardila et al., 2016
Poly-(vinylidene fluoride)/ cellulose acetate/ AgTiO ₂ nanofibre	Electrolyte membrane for lithium-ion battery	250 nm	Bands at 2870 and 2933 cm ⁻¹ are assigned to C-H stretching vibration	-	601 °C	DSC: crystallinity 30.23%, XRD: low intensity peak confirmed to be Ag peak	Bhute & Kondawar, 2019

Table 3: A summary of applications of gelatine-based nanofibres and their characteristics

Polymers	Applications	Nanofibre Characteristics						Ref.
		Size	FTIR	In-vitro degradation	Thermal strength	Tensile strength	DSC/ XRD	
Gelatine/polycaprolactone nanofibre	Barrier to prevent cardiac postoperative adhesion	441.6 ± 118.0 nm, pore size of 5.6 ± 1.8 µm	-	-	-	7.4 ± 1.6 MPa, 41.1 ± 8.9 MPa and 28 ± 7%	-	Feng et al., 2019
Ultrafine fish gelatine nanofibre	Biomedical engineering field	91.42 to 200.48 nm	-	7.5% of their total weight (24 h)	-	44.28 ± 0.10 MPa	-	Kwak et al., 2017
Gelatine electrospun (GES) nanofibre electrolytes	Nonaqueous electrolyte in electric double-layer capacitor	306.2 to 428.4 nm range	1169 cm ⁻¹ by C–N (imidazolium ring), 1049 and 525 cm ⁻¹ of B–F	-	423 °C	147.3 ± 33.7 cN	T _m = 110 °C, T _{degradation} = 277 °C	Kotatha et al., 2019
Gelatine/glycerlaldehyde nanofibre	Cartilage tissue engineering	95.19 nm	1449 cm ⁻¹ form cross-linking bonds in gelatine chains	-	-	-	-	Aliakbar-shirazi & Talebian, 2017
PCL/gelatine/grapheme nanofibre	Nerve tissue engineering and drug delivery systems	135 ± 29 nm	-	-	-	-	-	Heidari et al., 2019
Gelatine nanofibre	Bactericidal wound dressing	201.16 ± 29.95 nm	1635 cm ⁻¹ (C=O), 1539 cm ⁻¹ (N–H), 1454 cm ⁻¹ (C–H), 1238 cm ⁻¹ (C–N)	-	450 to 720 °C	-	-	Inal & Mülazımoğlu, 2019
PCL/gelatine composite nanofibre	Effective guided bone regeneration membranes	200–600 nm	Asymmetric CH ₂ (2943 cm ⁻¹), symmetric CH ₂ (2866 cm ⁻¹)	Degraded for 42 days	50 °C	-	-	Ren et al., 2017
Graphene oxide-gelatine nanofibres	Tissue engineering and wound dressing	200 ± 50 nm	3288 cm ⁻¹ N–H bond or OH group of the free –COOH vibrations	-	110 to 116 °C	56.4 ± 2.03 MPa	-	Jalaja et al., 2016
Chitosan/gelatine	Wound dressing	288.3 ± 74 nm	Gelatine affirmed by shifting of carbonyl stretching and NH ₂ bending peaks of chitosan (1655 & 1570 cm ⁻¹)	Decomposition of gelatine polymeric structure at 390–425 °C	285 to 390 °C	Young's: 410 ± 36 MPa, UTS: 16.86 ± 1.6 MPa, Strain%: 9.07 ± 0.99	-	Bazmandeh et al., 2020

Other than that, gelatine can also be widely used as a scaffold for tissue engineering (Ji et al., 2017). One important thing to note about electrospinning gelatine is that mammal gelatine aqueous solution cannot be electrospun at room temperature due to gelation. Fortunately, the usage of formic acid, hexafluoroisopropanol (HFIP), 2,2,2-trifluoroethanol (TFE), and acetic acid solvent systems were found to be suitable for the electrospinning to resolve this issue (Kwak et al., 2017).

Table 3 provides a summary of several literatures' usage of gelatine based nanofibres and their respective characteristics. Ultimately, literature confirms that

gelatine based nanofibres are indeed very promising to be developed especially for tissue engineering due to its excellent biodegradability, biocompatibility, ability to promote cell adhesion and proliferation, and resistance to immunogenicity.

3.3 Applications of honey-based nanofibre

Honey is an aromatic hydrocarbon which has many organic compounds and has been used as a natural medication since ancient time. When compared to other naturally occurring compounds which demonstrated antimicrobial efficiency such as tea tree oil, mastic gum and cranberry juice, honey is the most

Table 4: Summary of various applications of honey-based nanofibre and their characteristics

Polymers	Applications	Nanofiber Characteristics								Ref.
		Size	FTIR	Water vapor transport rate	Swelling and Weight Loss	Anti-bacterial activity	Anti-oxidant activity	Tensile strength	DSC/ XRD	
Polycaprolactone and manuka honey scaffolds	Dermal repair	Pore sizes ~ 80–7000 μm^2	-	50 and 300 $\text{g}/\text{m}^2/\text{day}$	-	Against both Gram+ and Gram- bacteria	-	-	-	Minden-Birkenmaier et al., 2015
Honey loaded alginate/PVA nanofiber	Potential bioactive wound dressing	528 $\pm$ 160 nm	3317 cm^{-1} (–OH), 2943 cm^{-1} (C–H), and 1092 cm^{-1} (C–O)	-	Weight losses of 84.82 $\pm$ 0.42% after 24 h	Against both <i>E. coli</i> and <i>S. aureus</i>	66 $\pm$ 7% after 9 h	-	-	Tang et al., 2019
Manuka honey-cellulose acetate nanofiber	Wound dressing	389–403 nm	1654 cm^{-1} (keto carbonyl C–O) in the spectrum of the MH loaded CA	2600 to 1950 $\text{g}/\text{m}^2/\text{day}$	-	Against <i>Escherichia coli</i> was better than against <i>Staphylococcus aureus</i>	47.25 $\pm$ 1.15 at 12 h	8.26 MPa	Amorphous micro-structure at 2 θ = 10° and 19°	Ullah et al., 2020
Honey/chitosan nanofiber enriched with <i>allium sativum</i> and <i>cleome droserifolia</i>	Enhanced antimicrobial and wound healing activity	145 $\pm$ 58 nm	-	-	90% swelling & 60–70% weight loss	Against <i>E. coli</i> and <i>MDR Pseudomonas aeruginosa</i>	-	-	-	Sarhan et al., 2016
Honey/poly (1,4-cyclohexane dimethylene isosorbide terephthalate) nanofiber	Wound dressing	482 nm	1270 & 1850 cm^{-1} C–O stretching and C–H bending	-	Release of honey was 72 mg/L in 10 min	-	-	2.7 MPa	-	Khan et al., 2017

efficient antimicrobial compound (Abou et al., 2020). This has allowed honey to be exploited for safe and effective antimicrobial wound dressings.

Honey consists of water (20%), fructose (40%), glucose (30%), sucrose (5%), and other substances (minerals, vitamins, amino acids, and enzymes) (Bagde et al., 2013). There are many types of bee's honey which have their own properties. It has been reported that 'Makshika' honey is considered medicinally the best (Scagnelli, 2016). Meanwhile, manuka honey (*Leptospermum scoparium*) has been promoted as an inexpensive and effective means of treating soft tissue wounds for veterinary patients and in particular exotic animal species (Scagnelli, 2016).

Honey has been incorporated into wound dressing due to its antimicrobial activity and capacity to perform the topical nutrition to the wound, debriding activity, minimise inflammation, and stimulate angiogenesis, granulation, wound contraction, and epithelialisation

(Simões et al., 2018). Electrospun manuka honey scaffolds have enormous potential as wound dressings and precursors to tissue engineered skin (Minden-Birkenmaier et al., 2015). Sarhan et al. (2016) has successfully produced nanofibres which consist of honey, polyvinyl alcohol (PVA) and chitosan enriched with *Allium sativum* and *Cleome droserifolia* extracts for use as wound dressings. The cytotoxicity of obtained wound dressings was compared with Aquacel®Ag, a commercially available wound dressing. For nanofibres with 100% extract, the cell viability was found to be 68% while for Aquacel®Ag, it was found not to exceed 20% even at the lowest concentration of cell viability (Sarhan et al., 2016). This highlights the potential of honey-based nanofibres for the commercial use as wound dressing. Table 4 includes a summary of several other literature utilising honey-based nanofibres and their characteristics. From the literatures, it can be shown that honey-based

nanofibres have a high potential for usage particularly for healing or wound dressing applications. Exploiting its natural antibacterial properties and further incorporating them into the fabrication of nanofibres produce meaningful outcomes.

The nanofiber characteristics detailed in Table 4, when combined with the respective electrospinning parameters detailed in Table 1 can aid researchers who plan to further develop honey-based nanofibers for utilization in commercial applications

4.0 Conclusions

This paper has effectively summarised literatures that involve the fabrication method of cellulose, gelatine and honey based electrospun nanofibres and their potential applications. Electrospinning is a very attractive, complex, and versatile technique to prepare advanced functional nanofibrous scaffolds for a variety of applications. We have summarised the fundamentals of electrospinning process, particularly in regard to the various effects of the electrospinning parameters when producing cellulose, gelatine and honey-based nanofibres. The morphology of electrospun nanofibres is influenced by three main parameters which are the solution, process conditions and environmental parameters. Hence, uniform nanofibres are obtained by optimizing these factors. Therefore, it is critical to gather and compare these various processing conditions to enable more progress moving forward with these green polymers.

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Natural or green electrospun nanofibres have been demonstrated to be potentially applicable in many fields including food, pharmaceutical, drug delivery, medical, electronic, and tissue engineering. Technology advancements through multiple spinnerets in electrospinning have proven to be effective methods for large scale production of nanofibres for industrial applications. Developing the green polymers through electrospinning method to produce nanofibres will make the process eco-friendly and facilitate the industrial production. However, there are still much more that needs to be explored and investigated before these green polymers can be included for various applications in industries. For example, prototypes, validation, and testing must be done before considering large scale industry productions, regardless the product and usage.

Nonetheless, cellulose, gelatine and honey are all very promising green polymers to be exploited. Therefore, further advancement in the performance of electrospun nanofibres based natural polymer will be crucial to move forward to obtain more widespread applications in industry.

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