

## MINI-REVIEW

# Electrospinning for the manufacture of biosensor components: A mini-review

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### Abstract

One of the key modern advancements of the biomaterial's world has been the recent progress with fibre manufacturing techniques which have allowed for novel approaches to biosensor fabrication. Electrospinning is capable of producing ultrafine and highly tailored fibres that can be made from a wide variety of natural and synthetic materials to be exploited for use as medical sensors. In recent years, electrospinning has been more widely researched and great strides have been made in its ability to produce fibres with more desirable properties. In this mini-review, the process of electrospinning is overviewed with an emphasis on its properties in producing biosensor components. It is seen how the advancements of nanotechnology allows for easier incorporation of bioreceptors to a larger range of membranes that are capable of higher sensitivity and broader applications. Furthermore, newer manufacturing techniques that are also capable of producing ultra-thin fibres are elaborated and the future of biosensor production in taking advantage of micro- and nano-scaled components are discussed.

### KEYWORDS

biosensor, electrospinning, nanofiber, nanomaterials, polymer

## 1 | INTRODUCTION

Biosensors have become an indispensable tool in the medical field to detect and quantify biological analytes that can then be easily quantified to the end user (Goode et al., 2015). A great example of a highly utilized biosensor can be found within blood-glucose tests which have surely transformed the way this disease affects the human population and have revolutionized the ease-of-use of such devices (Newman & Turner, 2005; Vigneshvar & Senthilkumaran, 2018; Wang, 2001). As the technology for biological detection advances, there is a necessity for novel methods to improve the sensitivity and accuracy of the readings, as well as to detect for newer and less obviously characterized diseases.

One approach to increasing the sensitivity of sensors is to manufacture the transducers at a smaller scale. At the nanoscale, there rapidly

becomes many advantageous properties such as a high specific surface area, greater mechanical properties and controllable surface functionalization (Zhang et al., 2005). Electrospinning is able to manufacture fibrous membranes as small as 10 nm in diameter from a wide variety of materials (Li et al., 2013). For this reason and many more to be discussed, there has been plenty of recent progress in the development of biosensors involving electrospinning, as will be elucidated to in this review.

## 2 | ELECTROSPINNING

Although iterations of this technology have existed since 1900, electrospinning has seen a more recent spike in attention regarding its ability to produce fibres which are highly suited for a number of key

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biomedical applications such as in biomaterials, tissue engineering, drug delivery and as biosensors (Sill & von Recum, 2008; Tucker et al., 2012; Wang et al., 2009; Zhang et al., 2009). Electrohydrodynamics describes the basis behind the technology where typically a polymer solution (the hydro- portion of the name) is introduced to a high-voltage electric field (the electro-portion), where it interacts to eventually produce micro-scaled polymeric products such as particles, beads and fibres (Castellanos, 1998). Electrohydrodynamics can be further separated into two distinct techniques, electrospraying, where particles and beads are produced, and electrospinning, where fibres are produced (Anu Bhushani & Anandharamakrishnan, 2014).

## 2.1 | Principles of electrospinning

Electrospinning is popular no doubt to its many advantageous benefits it flaunts over other conventional fibre production methods such as melt spinning and spinneret extrusion; this includes the superior control over fibre morphology, ability to produce finer fibres and the increased range of compatible materials (Luo et al., 2012). Furthermore, electrospinning is a low-cost, single-step and easy-to-use process that has recently seen automation and commercial scale-up (He et al., 2009; Mussa Farkhani & Valizadeh, 2014; Vass et al., 2020).

The production of fibres via electrospinning revolves around the specialist apparatus. The set-up consists of a metallic small-diameter

nozzle which is wired to a high-voltage power supply, the nozzle supplies a mass flow of polymer solution via chemically inert tubing and a highly accurate syringe pump device (Figure 1) (Agarwal & Jiang, 2014; Ahmed et al., 2018). There is also a collector which gathers the fibres, as the collector is grounded, the fibres are drawn to it due to the large potential difference between the nozzle and itself (Bhardwaj & Kundu, 2010). In order to produce polymeric structures, a polymer solution is created which consists of a polymer dissolved into a solution with a suitable solvent (Bosworth & Downes, 2012). Composite materials can also effortlessly be created via this technology, this is usually doped within the polymer, which typically acts as a very efficient carrier (Li & Xia, 2004; Simotwo et al., 2016; Zhang et al., 2017).

As electrospinning uses the same set-up as electrospraying, it is important to note that the difference in product formation lies in the polymer solution; low polymer chain entanglement of the solution leads to electrospraying, whilst higher polymer chain entanglement leads to electrospinning (Husain et al., 2016). As the polymer solution is fed into the nozzle, it comes in contact with the high-voltage electric field, this leads to a notable series of events which defines the technology. Firstly, the exiting droplet of the solution undergoes electrostatic repulsion which opposes and overcomes its surface tension, leading to a balance of several interfacial forces (Figure 1), the droplet morphs into a conical shape known as a Taylor cone (Garg & Bowlin, 2011; Stanger et al., 2009; Yarin et al., 2001). A polymer jet

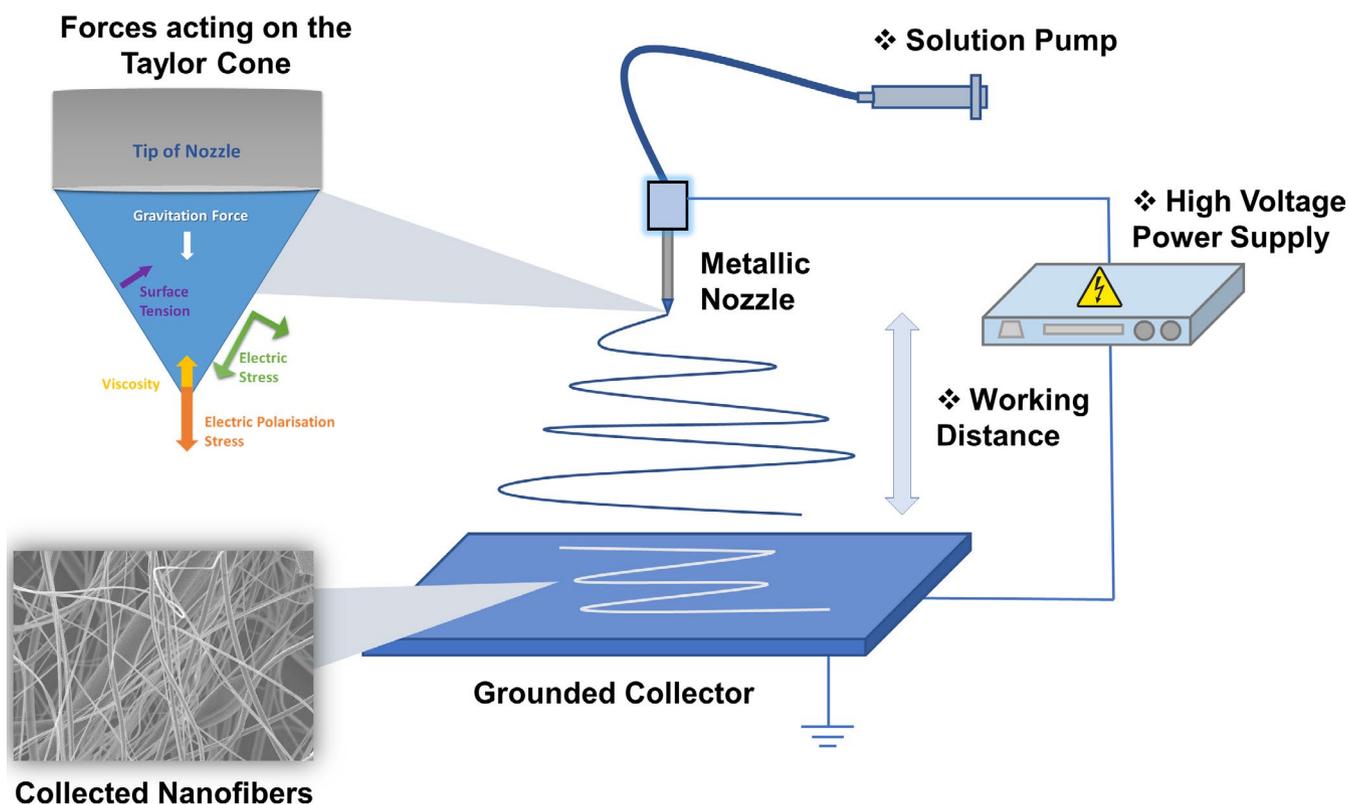


FIGURE 1 Schematic representation of the basic electrospinning set-up, with a diagrammatic insert showing the various forces present in the emerging Taylor cone. Working parameters that can be altered during production and are denoted with a bullet point.

then erupts from the apex of the cone, as this jet dries via evaporation of the solvent, it is drawn to the grounded collector and uniform fibres are thus produced (Guerrero et al., 2014).

## 2.2 | Working parameters

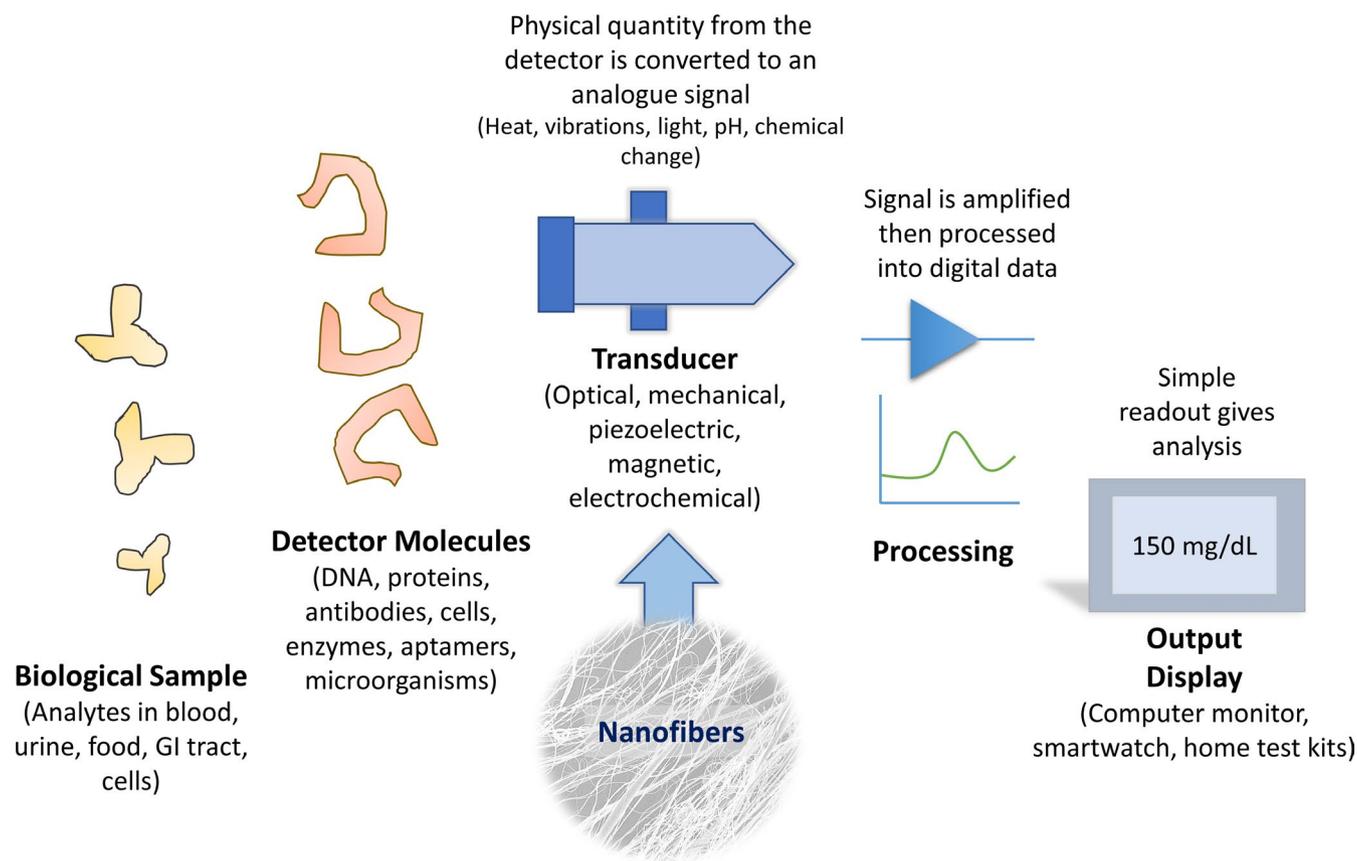
The ability to customize the fibre morphology ultimately is down to the working parameters that can be altered during production. Although not a working parameter, the choice of material (polymer) and the solvents plays a crucial role in the final product morphology.

**TABLE 1** Overview of major electrospinning processing parameters and their typical effects on fibre morphology.

Processing parameter (increasing ↑)	Effect on fibre morphology
Polymer viscosity	Increases fibre diameter
Solvent volatility	Reduced fibre diameter (more jet thinning)
Applied voltage	Reduces fibre diameter (up to 1.2 kV/cm)
Solution flow rate	Increases fibre diameter
Working distance	Reduces fibre diameter (more jet thinning)

One of the most influential factors is the molecular weight (Mw) and concentration of the polymer (Chakraborty et al., 2009). A higher molecular weight variant of the same polymer can increase its viscosity which leads to thicker fibres being produced (Koski et al., 2004). A similar trend is seen with a higher polymer chain entanglement polymer (Shenoy et al., 2005). The surface tension of a polymer solution, typically governed by the utilized solvent, can determine the range of voltage required for fibre formation (Geng et al., 2005). The volatility of the solvent additionally effects the evaporation time and the formation of surface pores (Zheng et al., 2006).

The main working parameters of electrospinning is the magnitude of applied voltage, the polymer feed rate and the collection distance. Being the main driving force in the production process, the applied voltage is required to overcome the surface tension of the solution in order to form uniform products. Fibres are typically seen after exceeding 0.3 kV/cm, and a small window of voltage exists which is specific to the solution properties of the polymer, outside this window, the Taylor cone does not appear and fibre generation is not reliable (Fallahi et al., 2008; Katti et al., 2004). The flow rate of the solution can be adjusted to suit the application; it should be adequate enough to maintain mass flow for the cone, by replacing lost solution ejected as a jet. Therefore, the flow rate can affect



**FIGURE 2** Diagram showing the key makeup of a biosensor, examples of each step are given and the potential for nanofibers for use as transducers are shown in the process.

the fibre diameter and give rise to differing morphologies (Zargham et al., 2012).

The distance between the tip of the nozzle and the collection unit further influences fibre morphology. By increasing the collection distance, evaporation time increases, allowing the polymer jet to thin further before being deposited. The chief working processing parameters and their effects are summarized in (Table 1).

### 3 | PRODUCTION OF BIOSENSORS

#### 3.1 | Biosensors

A biosensor is a synthetic functional analog based on an immobilized biological compound; it is wired into a signal transducer and then into an amplifier (Chaubey & Malhotra, 2002). When in contact with the specific analyte used for testing, the transducer converts a change in the physiochemical environment into an electrical signal that is used for detection. Biosensors typically detect change in chemical composition, heat, light and vibrations (Clark et al., 2020; Mehrotra, 2016; Shittu et al., 2020). Therefore, given the high specificity of biosensors, they require several key properties to function correctly; biosensors should be independent from their physical parameters such as having chemical resistance to the detection environment, they should be reusable, and they should be designed to give maximum output to improve detection sensitivity. Figure 2 shows the process in which biosensors operate. Transducer manufacturing at a smaller size allows for superior signal detection and higher sensitivity which can be utilized with novel user interfaces such as portable smart devices (Honjol et al., 2020; Lopez et al., 2020).

#### 3.2 | Electrospinning biosensors

Being able to produce nanofibres with unique morphologies gives electrospinning great suitability in biosensor fabrication. Electrospun fibres are highly matched for use in biosensors due to their one-dimensional confinement characteristics and high porosities, highly uniaxially orientated fibres can control electrical diffusion (Agarwal et al., 2013). Furthermore, chemical biosensors based on polymeric fibres can be generated with predictable pore geometries, large interconnectivity of pores, high specific surface area, can be made of composite materials which increase sensitivity to analytes, made of important polymer blends and made with active surface nanoparticles (Bala et al., 2016; Macagnano et al., 2015; Wang et al., 2013; Xia et al., 2010).

Achieving thinner diameter fibres leads to a high specific surface area, this improves the reactivity allowing adsorption and release to be more rapid, further increasing the number of available sites for interaction between the catalysts and other reactive substances (Mercante et al., 2017). Porosity provides extra three-dimensional space which increases the nanofibre's resistance to mass transport. It is believed that nanofibrous electrodes with high porosities can

be used to prevent a diffusion barrier of the analyte towards the electrode's surface (Agarwal et al., 2013). It is the ability to functionalize and highly customize the produced nanofibres to fulfil a wide variety of biosensing requirements which makes electrospinning an excellent fabrication process. In the next section, recent progress of fibrous biosensor designs is discussed with their unique advantages relating to specific applications and analytes.

### 4 | PROGRESS OF FIBROUS BIOSENSORS

#### 4.1 | Electrospinning

Early cases of using nanofibrous membranes for optical sensors began to appear in 2002, when electrospinning was used alongside a poly(acrylic acid) and poly(pyrene methanol) fluorescent polymer (Wang et al., 2002). Prior to electrospinning, similar fluorescence sensing was carried out on thin films, using an electrostatic layer-by-layer assembly technique (Lee et al., 2000). The produced biosensor would operate in an optical transducer mode, using pyrene methanol as the fluorescent indicator as it has a large high quantum yield, strong absorbance, large Stokes shift and has low toxicity (Thiel, 2001). The highly responsive quenching-based sensors can detect pH, iron and mercury ions, as well as 2,4-dinitrotoluene (DNT). Compared to the sensors produced as thin films, the sensitivity of the electrospun membranes in detecting DNT and metal ions where over two orders of magnitude higher, this is thanks to the highly increased surface area provided by the thin electrospun fibres. The polymer solution consisted of an 18 wt% solution of a poly(acrylic acid)-poly(pyrene methanol) and cross-linkable polyurethane latex, which was dissolved in dimethylformamide. For this work, the applied voltage was in the range of 15–20 kV, the produced electrospun fibres showed random fibre orientation and had a fibre size between 100 and 400 nm, which was evenly distributed. The distance between the fibres, the porosity, provides a surface area to volume ratio up to two orders of magnitude higher than for continuous thin films (Brezesinski et al., 2010). The study also states that additional increases in surface area may have been achieved by further optimizing conditions and working parameters such as solvent, polymer concentration and collection distances.

Electrospinning has also been used to produce biocomposite membranes for amperometric biosensors, in this work, poly(vinyl alcohol) and glucose oxidase were electrospun onto a gold electrode (Ren et al., 2006). This novel technique showed that enzymes were successfully immobilized inside the poly(vinyl alcohol) membranes, having high efficacy due to the high porosity and specific area afforded by the fabrication technique. The produced biosensor operates on the electrochemical transducer mode with an enzyme-based bioreceptor complex. Glucose oxidase was used as the enzyme bioreceptor which could detect changes in blood-glucose levels, its sensitivity depends on the change in enzyme structure following immobilization. Poly(vinyl alcohol) was used as the immobilization



matrix because of its biocompatibility, high thermal stability, elasticity and its ability to swell in aqueous environments which makes it a suitable matrix for enzyme immobilization (Djennad et al., 2003). The electrospinning process involved an operating voltage of 10 kV, and the produced fibres had a diameter range between 70 and 250 nm. It is not possible to form glucose oxidase fibres purely; therefore, electrospinning allows for facile manufacture of composite materials, in this case using poly(vinyl alcohol) to interrupt the complex three-dimensional structure consisting of strong inter- and intra-molecular forces. Absorption is more rapid given the large surface area which reduces the response time of the sensor; this allows for additional loading of enzyme into the fibres and provides the enzymes with additional time to react. Together, these improvements can increase the response current, increasing sensitivity by lowering the detection limit. Biocomposite biosensors such as this are useful in analysing blood-sugar levels.

Cyclooxygenase-2 is an important enzyme in pain, inflammation and cancer cell proliferation, it can act as a biomarker, and therefore, its reliable detection is very beneficial in healthcare (Plummer et al., 2001; Seibert et al., 1994). Highly porous nanofibres have been developed for cyclooxygenase-2 detection (Asmatulu et al., 2019). The completed biosensor device would operate in an electrochemical transducer mode which would be able to detect and quantify the expression of cyclooxygenase-2 enzyme, where overexpression is linked to several forms of cancer (Williams et al., 1999). The detection process involves the immobilization of the biomarker enzyme (cyclooxygenase-2) by polyaniline, an intrinsically conductive polymer which is easy to produce, low cost, easy to combine with other polymers and has good environmental stability. In this work, polyaniline and polystyrene were used as the main polymers, dissolved in chloroform and camphor sulfonic acid. A high operating voltage of 25 kV was necessary; four differing polymer flow rates were used so that there would be fibres of varying diameters. The smallest average fibre diameter of 256 nm was achieved using the lowest flow rate, by systematically increasing the flow rate, average fibre diameter increased. The thinnest fibre sample showed higher percentage impedance change and could detect the target antigen at a minimum of 0.01 pg/ml; this was significantly more sensitive than the control biosensor which managed detection at a minimum concentration of 100 pg/ml. It was noted that the drawback of the fabrication technique involved lack of control over fibre structure and uniformity, something that can be addressed using advanced collection and solvent optimization techniques (Ahmad et al., 2019; Katta et al., 2004; Yang et al., 2004).

## 4.2 | Other techniques

Other methods of nanofibre fabrication have been introduced since electrospinning. Centrifugal spinning and pressurized gyration are able to produce masses of fibrous membranes at a rate far superior than with electrospinning alone (Heseltine et al., 2018; Weitz et al., 2008). Optical fibres for light propagative biosensing applications

have been produced by centrifugal spinning (Zhang et al., 2015). In this work, indium tin oxide coated Polyvinylpyrrolidone nanofibres were produced using a 2500 rpm spinneret and a rotating collector which used 5 kV voltage to optimally collect the fibres. The average diameter of the produced fibres was 423 nm but reduced to 210 nm after calcination. The optical fibres demonstrated optical transmittance of about 85% and demonstrated this technique to be a viable means of production for biosensor components.

Pressurized gyration is also capable of mass-producing useful fibrous membranes (Ahmed et al., 2019; Matharu et al., 2020). Gold nanoparticles can be directly integrated into the fibrous matrix, allowing it to be used in the detection of crucial proteins. The gold-binding peptide Au-BP2 was integrated into nanofibres using a pressurized gyration based technology which had a diameter range of 117–216 nm (Zhang, et al., 2015). The fibres consisted of polyethylene oxide; water was used to dissolve the peptide constituents. In this work, it is shown how simply gold can be loaded onto the polymer matrix, forming composite nanofibrous membranes with selectivity to certain biological markers such as  $\text{Cu}^{2+}$ . The technology allows for easy incorporation of bioreceptors to biosensor designs. Furthermore, pressurized gyration is capable of producing gas-filled microbubbles that have biosensing capabilities; poly(vinyl alcohol) and lysozyme microbubbles in the size range of 10–250  $\mu\text{m}$  were produced with and without the incorporation of gold nanoparticles (Mahalingam et al., 2015). The addition of gold nanoparticles led to greater optical extinction values, and all the microbubbles demonstrated antibacterial activity against *Escherichia coli*. The bubbles were able to successfully detect the pesticide paraoxon in an aqueous solution.

## 5 | FUTURE PERSPECTIVES AND FINAL THOUGHTS

Electrospinning has exhibited great maturation in the progress of producing small polymeric products with boundless potential in the biomedical field. With its progress, advancements have been made to improve its yield, variety of materials it can process, reduction of product size and increase in commercial viability. Electrospinning thus brings a highly viable means to produce biosensor parts with extremely high surface area materials which rival other production methods to bring higher sensitivity and selectivity to the medical and biosensor industry. Furthermore, the facile nature of the technology allows for easy incorporation of nanoparticles which can increase sensitivity and reduce costs associated with expensive bulk materials.

With its great success and much well-earned attention, electrospinning has ushered in a new era of alternative nanofibre techniques which brings greater choice in the manufacture of small-diameter products. These newer technologies learn from the accomplishment of electrospinning and provide both scientists and industries with a greater range of options that they can choose to utilize for specific applications. Each technology will have its drawbacks when compared to another, but the future could be a hybridization of technologies available now that permit the mass production of nano-scaled

components which are far superior to their larger counterparts. As technology and medical requirements grow more and more advanced, improvements in processing and sensitivity can only be gained from discovering materials with greater conductive properties or processing them into smaller sizes using novel manufacturing processes.

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