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A review on designing nanofibers with high porous and rough surface via electrospinning technology for rapid detection of food quality and safety attributes

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ABSTRACT

Background: Agricultural and food products are highly perishable, which require advanced detection methods to monitor their quality. Electrospinning technology is a novel fiber production technique using electric force to draw charged threads of polymer solutions, leading to the elongation and thinning of the fiber and forming uniform fibers with nanometer-scale diameters. Electrospun nanofibers have unique surface properties such as high specific surface area and porosity, which can increase their potential for use in sensors for rapid detection with high accuracy and sensitivity.
Scope and approach: This review summarizes recent advances in using electrospinning technology to synthesize nanofibers with high porous and rough surfaces. The nanofibers can be used in rapid sensing methods to monitor food quality and safety attributes, including contaminants such as pesticide residues. The morphological control of electrospun nanofibers to increase the resolution of rapid detection sensors was discussed. Some recent achievements in identifying different analytes using electrospinning technology are elaborated, including SERS, aptamer-based biosensors, surface plasmon resonance sensors, and direct electron transfer method.
Key findings and conclusions: Electrospinning technology can be used to control the surface properties of nanofibers to design sensors and substrates for the rapid detection of various contaminants. Recent studies showed an improvement in the performance of sensors, such as increasing accuracy, reducing the limit of detection, and increasing the resolution due to the use of electrospun scaffolds in the design of sensors. The combination of electrospinning technology and sensors has great potential for applications in the food industry.

1. Introduction

In today's world, technology has penetrated all industries, including the agriculture and food industry. Many scientific advances have been made to increase the productivity in various sectors of the food industry and improve food quality and safety. However, one of the main challenges is the monitoring of food quality and safety attributes. Changes or declines in food quality can be monitored through a variety of indicators, including the microbial count of foods (Shaibani et al., 2018), pH changes (Aghaei, Emadzadeh, Ghorani, & Kadkhodaee, 2018), the presence of plant or microbial toxins (L. Luo, Liu, Ma, Li, & You, 2020), heavy metals (Senthamizhan, Celebioglu, & Uyar, 2014), and residues of pesticides and herbicides (Nguyen & Jang, 2021).

The advent of biosensors in recent decades can help solve problems in the food industry about detecting food contaminants. In general, a biosensor consists of three main parts: transducer, amplifier, and processor, which together catch a recognizable response (Xie et al., 2021). Usually, the transducer's role in a biosensor is to convert the recognition event into measurable signals. Then, the received electronic signals are amplified in the amplifier and transmitted to the processor for processing. After processing, the signals are converted into visible responses and then transferred to the display section for observation (Xie et al., 2021). What distinguishes a biosensor from other sensors is the use of biological reactions or components to detect and identify analytes. Numerous studies have been performed on the use of antibodies (Y. Luo et al., 2012), enzymes (Kant, Tabassum, & Gupta, 2018), nucleic acids (Huo, Hu, Gao, & Li, 2021), DNA (Balbinot, Srivastav, Vidic, Abdulhalim, & Manzano, 2021), metal ions (Senthamizhan, Celebioglu, Balusamy, & Uyar, 2015), microorganisms (Yang et al., 2018), cells (L. Zhao et al., 2015), and even tissues (Rahmati et al., 2021), to detect and

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identify analytes. In particular, immobilization of biological materials and molecules can be performed on specific substrates and reacts upon contact with the target analyte. By increasing the specific surface area of the substrate, more molecules can be immobilized on its surface, which significantly increases the efficiency of the detection in various fields, including biosensors (Matlock-Colangelo, Coon, Pitner, Frey, & Baeumner, 2016), indicators (Aghaei et al., 2018), and rapid detection kits (Asmatulu, Veisi, Uddin, & Mahapatro, 2019).

Some previous studies have been conducted on the applications of nanotechnology in the design of porous and rough substrates for their use in various detection techniques, including surface-enhanced Raman spectroscopy (SERS) (Liou, Nayigiziki, Kong, Mustapha, & Lin, 2017), aptamer-sensing technologies (Aptasensor) (Xie et al., 2021), surface plasmon resonance (SPR) sensor (Kant et al., 2018), and so on. The results of studies in the field of electrochemical sensors show that increasing the surface roughness and the contact surface between the analyte and the reactant by nanotechnology significantly increases the signal resolution of these electrochemical sensors (Wu et al., 2020).

Electrospinning is a production technique for micro- and nanoscale fibers. The beginning of this technology dates back to the early 20th century and was used for many years in the filtration industry (Tucker, Stanger, Staiger, Razzaq, & Hofman, 2012). Electrospinning is a simple, fast, and high-performance technology for the production of nanofibers from various bio- and petroleum-based polymers (Aman Mohammadi, Hosseini, & Yousefi, 2020). This technology ionizes a polymer solution by applying a strong electromagnetic force of about 20–30 kV. By placing the opposite charge on the other side of the device, the polymer solution tends to move toward its opposite charge, causing a narrow jet of polymer solution called the Taylor cone to form (Aman Mohammadi et al., 2020). This jet of the polymer solution can be narrowed to the nanoscale due to the many twists during spinning. The resulting nanofibers have a high potential for use as a substrate in the rapid detection technique because electrospun scaffolds with rough surfaces and high porosity can be designed in various ways (Aman Mohammadi et al., 2020). Several variables such as polymer solution, device variables, and environmental variables can affect the properties of electrospun fibers, such as morphology and fiber diameter. Therefore, by controlling these variables, fibers with ideal properties can be designed (Zaarour & Alhinnawi,). Today, these unique properties of electrospinning nanofibers have led to the introduction of this technology in many industries. For example, electrospun fibers were widely used for medical applications (Y. Luo et al., 2012), tissue engineering (Rahmati et al., 2021), wound dressing (Hajikhani, Emam Djomeh, & Askari, 2021), drug delivery (Balbinot et al., 2021), active packaging to keep food fresh (Aghaei et al., 2018; Topuz & Uyar, 2020), biosensors (N. Zhang et al., 2017), and energy harvesting (Zaarour, Zhu, & Jin, 2020a). Recent studies show the use of electrospun scaffolds as rough substrates in rapid detection applications, such as designing polyaniline nanofiber for cyclooxygenase enzyme detection (Asmatulu et al., 2019), polyaniline-based scaffolds as chemoresistive sensor for gaseous ammonia detection (Kumar et al., 2020), carbon-based electrospun nanofibers for tramadol detection (Jahromi, Mirzaei, Savardashtaki, Afzali, & Afzali, 2020), poly(vinylbenzyl chloride) electrospun nanofiber-based colorimetric probe for detection of Fe^{2+} in water (Ondigo, Tshentu, & Torto, 2013a), and designing poly(vinyl alcohol) with acetylcholinesterase (AChE) and indolyl acetate electrospun nanofibers for the analysis of pesticide residues (Zhai, Feng, Hu, Zong, & Wu, 2020). Nevertheless, these studies show only a small part of the various applications of electrospun nanofibers. Recent observations show growing applications of electrospinning in the sensor industry. Increasing the specific surface area and decreasing the fiber diameter will collectively lead to increased sensor performance in analytical chemistry.

This review article introduces new electrospinning techniques for the design of electrospun nanofibers with the aim of using them as a substrate or sensor in rapid detection. It also presents recent achievements in increasing the sensor's resolution by increasing the porosity and roughness of electrospun nanofibers that act as scaffolds.

2. Basic principles of electrospinning

The fibers produced by electrospinning technology have a great variety, making this technology applicable in various industries. In general, this device consists of three parts: a high voltage power supply, a syringe pump, and a collector (Fig. 1). The collector may come in various types that can move or be stationary (Aman Mohammadi et al., 2020). The types of movable collectors include rotating drum, rotating drum containing wire, rotating wire drum, rotating drum with sharp pin inside, rotating drum with knife edge electrode, rotating drum with multiple knife edge electrodes, rotating disk, and water bath collector (Subrahmanya et al., 2021). In the case of stationary collectors, plate collector, parallel ring collector, blade electrodes in-line, parallel electrodes, and array of counter electrode are common collectors in this category (Alfaro De Prá, Ribeiro-do-Valle, Maraschin, & Veleirinho, 2017; Subrahmanya et al., 2021). Different collectors are used to design fibers in different shapes and for different purposes (Alfaro De Prá et al., 2017). In general, electrospinning technology requires both poles of the electromagnetic current to orient the fibers during the process. The direction of motion is often from the positive pole to the negative pole, and the process may be vertical or horizontal (Alfaro De Prá et al., 2017). The common form of the device is needle electrospinning, but other surface-free methods have been developed that do not require a needle. A thin layer containing a polymer solution is used to form a large number of Taylor cones. This minimizes the length of the process (Alfaro De Prá et al., 2017). Various variables affect the performance of the device are illustrated in Fig. 1, including relative humidity, temperature, viscosity, electrical conductivity, surface tension, and others. These variables can be classified into three categories: polymer solution variables, instrumental variables, and ambient variables (Rostamabadi, Assadpour, Tabarestani, Falsafi, & Jafari, 2020). By controlling these variables, the properties of the fibers produced can be controlled according to the type of consumption.

2.1. Polymer solution variables

The first group of variables affecting the fibers' properties are the polymer solution variables, including concentration, viscosity, electrical conductivity, and surface tension of the solution. The polymer concentration in an electrospinning solution is an important factor in the formation of uniform and bead-free fibers (Zaarour, Zhang, Zhu, Jin, & Huang, 2019). The polymer concentration is usually expressed as a percentage (w/v%) and a sufficient polymer concentration is required to form the fibers. The minimum concentration for each polymer varies, depending on its properties, which has been reported in recent studies from 5% to over 40% (Hajikhani, Emam-Djomeh, & Askari, 2021). The polymer concentration can affect other variables. It is impossible to determine a specific concentration for all polymers. However, if the polymer concentration is not sufficient, it will cause the jet to break and lead to bead formation (Zaitoon & Lim, 2020). Excessive reduction of the polymer concentration can lead to the production of particles instead of fibers.

The viscosity of the solution is another important variable that can be affected by the polymer concentration, surfactants, and polymer molecular weight (Rostamabadi et al., 2020). A sufficient viscosity is required to form a Taylor cone and polymer spin, but excessive viscosity prevents the polymer solution from spinning (Aman Mohammadi et al., 2020). Therefore, viscosity is a limiting factor for selecting the concentration of polymer solution and increasing the concentration is allowed as long as the viscosity of the solution is acceptable. To reduce the viscosity, polymers with lower molecular weight or a more linear structure can be used (Wen, Xiong, Lei, Wang, & Qin, 2022). Recent

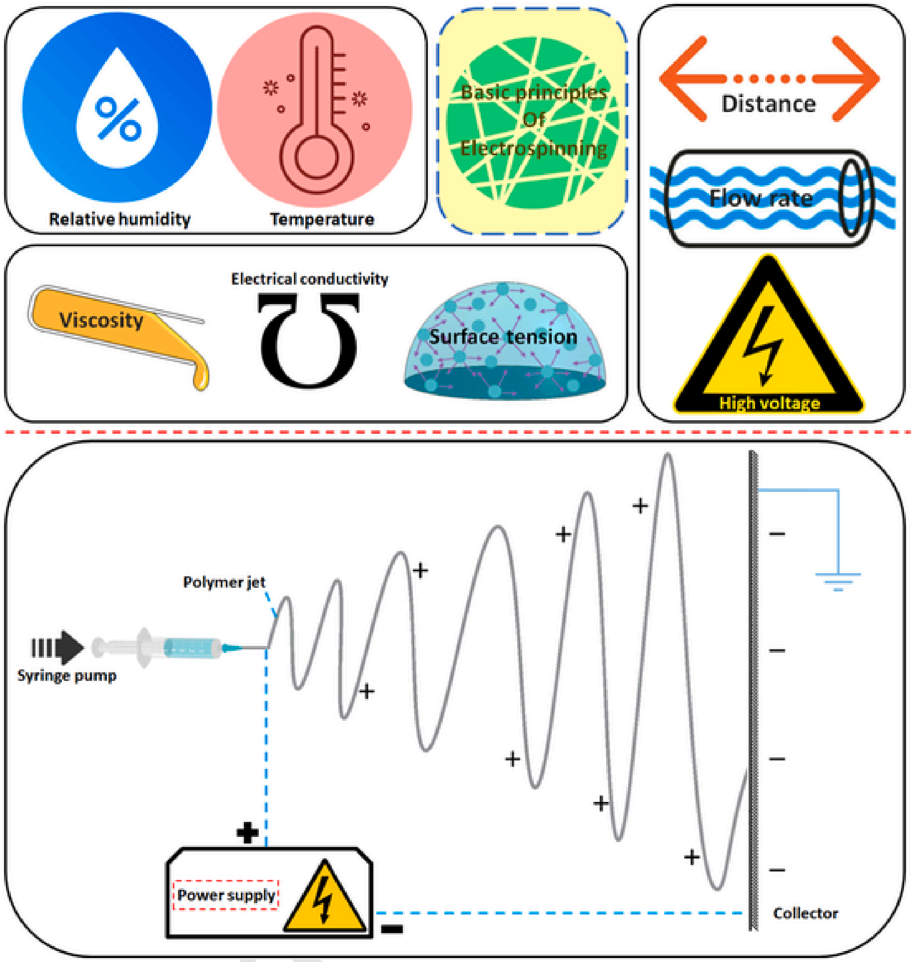

Fig. 1. An overview of the various variables affecting the properties of electrospun fibers (top) and the general setup of the electrospinning technique (bottom).

studies show that the use of some common surfactants can increase the viscosity by reducing the surface tension between the polymer and the solvent (Wen et al., 2022). However, a decrease in viscosity has been observed with the addition of surfactants (Fang, Yang, Yuan, Charlton, & Sun, 2017).

Surface tension is known as another influential variable in fiber properties. The increase in surface tension of an electrospinning solution can be due to the polymer-polymer or polymer-solvent behaviors (Ewaldz, Randrup, & Brettmann, 2021). To control the surface tension, ionic or non-ionic surfactants can be used in a solution to reduce the surface tension between polymers or polymer-solvent by creating intermediate properties. A high level of surface tension prevents the formation of stable jets during spinning, so the surface tension must be measured like any other effective parameters (Zaitoon & Lim, 2020).

Electrical conductivity is another critical variable for the electrospinning of polymer solution. The basis of electrospinning is spinning with the help of electromagnetic force, so the electrical conductivity in a polymer solution is one of the basic requirements. Organic solvents are commonly used in the preparation of electrospinning solutions, which have poor electrical conductivity and require high voltages to spin (K. Zhang, Hong, et al., 2018). Saline solutions containing sodium chloride can be used to increase the electrical conductivity (K. Zhang, Hong, et al., 2018). The reason for using organic solvents is their low boiling point compared to the aqueous solvents because they must be completely evaporated during the spinning of the polymer. Since salts are generally soluble in polar solutions such as water, aqueous solutions containing salt can be used to increase the electrical conductivity of a polymer solution, but excessive use can cause water to remain in the electrospun fibers. To solve the problem, high salt concentration in saline solution can be used to minimize the consumption of saline solution. Polar organic solvents can also be used to dissolve salts and disperse in a polymer solution.

2.2. Processing variables

Instrumental variables generally consist of three items: the flow rate of the polymer solution, the applied voltage, and the distance from the needle tip to the collector surface. If a rotating drum were used as the collector, the speed of drum rotation would also be an important variable. The flow rate is defined as the amount of injected polymer solution per unit time by the syringe pump. The flow rate of the solution in this technique is very low, which can be one of its weaknesses. The flow rate must be adjusted to maintain the formed Taylor cone. In other words, if the flow rate is not high enough, the Taylor cone will disappear and the jet will break. Increasing the flow rate can also cause the formation of a polymer drop at the tip of the needle, which can be thrown on the formed fibers as the drop enlarges (Ewaldz et al., 2021).

The distance from the needle tip to the collector surface is another important variable, which affects the diameter of the fibers because the polymer spins and becomes narrower as it passes through this path. Therefore, the greater the distance, the more the fibers are spun and the smaller the diameter of the fibers (Joy, Anuraj, Viravalli, Dixit, & Samavedi, 2021). Excessive distance causes the electromagnetic field to be interrupted and the fibers to be reluctant to settle on the collector surface (Joy et al., 2021).

The electric force is the driving force that causes the polymer to move and spin. One of the characteristics of the electromagnetic force is low current with high voltage (Li et al., 2017). To create strong electromagnetic fields, a high voltage of about 30 kV must be used. Electric force must be sufficiently applied to the polymer to form a Taylor cone. This sufficiency limit is affected by the flow rate and the distance to the collector. The less flow rate and shorter distance to the collector, the less electric force is required (Ahmed, 2021). In general, as the applied force decreases, the polymers are pulled towards the collector less quickly and the polymer flight time will be longer, making the fibers smaller in diameter. To produce fibers with the smallest possible diameter, the least possible electric force must be used with the greatest possible distance to the collector (Joy et al., 2021). By increasing the electric force, the electromagnetic field is strengthened and the distance to the collector can be increased. Also, by increasing the electric force, the polymer spins faster, so a higher flow rate can be used without droplets forming at the tip of the needle.

Some electrospinning devices have a movable collector, such as a rotating drum. Recent studies have shown that the use of a rotating drum during the process causes the fibers to be more elongated, so that fibers can be formed with a smaller diameter and higher uniformity (Pham Le, Uspenskaya, Olekhnovich, & Baranov, 2021).

2.3. Ambient variables

Ambient variables are critical to the production of electrospun fibers, including relative humidity and ambient temperature. During the electrospinning process, since the evaporation rate of the solvent from the polymer solution can be affected by the relative humidity of the ambient, this variable must be controlled during the process. High relative humidity can cause the solvent to evaporate insufficiently and deform the fibers because the formed nanofibers have a high specific surface area and tend to dissolve in the solvent (Park & Um, 2021). Low evaporation of the solvent can cause the fibers to re-dissolve. In some studies, due to the high tendency of some biopolymers to absorb moisture, the high relative humidity of the ambient causes water to be absorbed by the fibers after spinning. Therefore, the relative humidity should be controlled to less than a certain limit. However, high relative humidity due to the condensate of water droplets on the fiber surface can cause microscopic cavities and increase the porosity of the fibers (T. Min et al., 2021).

On the other hand, ambient temperature can also have significant effects on the production of electrospun fibers because the temperature can directly affect solvent evaporation. This is of little concern for organic solvents because most organic solvents have a low boiling point and are easily evaporated at room temperature during the electrospinning process. But in the case of aqueous solvents such as water, increasing the temperature can help the solvent evaporate. Temperature can also affect the properties of the polymer solution. Increasing the temperature reduces the viscosity of the solution. As the viscosity decreases, the mobility of ions in the polymer solution increases, so the electrical conductivity in the polymer solution must also increase. Thus, a smaller electrical force was used to spin, which eventually led to the production of fibers with a smaller diameter (Park & Um, 2021; Zhu, Zaarour, & Jin, 2020).

3. Recent achievement about designing nanofibers with high porosity and roughness

Electrospinning technology produces fibers with different porosity under the influence of many variables. Careful control of these variables can enable this technology to design nanofibers with porous and rough surfaces (Zaarour, Zhu, & Jin, 2020b).

Electric current is generally classified into two types of direct current (DC) and alternating current (AC). In electrospinning technology, the use of DC is more common because it carries fewer potential risks. However, the study on the effect of applying AC or DC current on the fabrication of polyurethane nanofibers showed that the morphology of the fiber's changes under the influence of the type of electric current. The use of AC current led to the production of fibers with more irregularity and porosity. However, the diameter of electrospun fibers with DC current was lower compared to AC current. This study introduced AC current as a suitable way to produce fibers with higher porosity properties. However, the big problem with using AC current is the safety, which is much lower than the DC current method (Havlíček, Svobodová, Bakalova, & Lederer, 2020).

Mixing different polymers with different polar properties can be effective in increasing the fiber porosity (Hajikhani, Emam-Djomeh, & Askari, 2021). Because these polymers have little tendency to mix with each other, they begin to separate during the spinning process. This phenomenon can be considered a problem to fibers and can be easily solved with the help of suitable surfactants. This technique can be used to increase the fiber's porosity by causing microscopic cracks on the fibers (Philip, Tomlal Jose, Chacko, Philip, & Thomas, 2019). Rapid evaporation of the solvent during spinning does not allow the polymers to separate completely, but it does cause irregularities and surface cracks that can significantly reduce the scaffold strength. Difference in polymers' chemical properties can directly affect fibers' surface properties, so the roughness and porosity of the fibers can be controlled (Hajikhani, Emam Djomeh, & Askari, 2021). The use of polymer mixtures with different solubilities is suggested as another approach. Removing some of the fiber-forming polymers from the electrospun scaffolds can be effective in designing high-porosity fibers. In this method, super hydrophilic polymers are usually used as copolymers along with the main polymer. The main polymer must be insoluble in water so that it is not degraded by the treatment. Polyvinylpyrrolidone (PVP) or polyvinyl alcohol (PVA) polymers are used as hydrophilic polymers in this method because they are easily soluble in water. A recent study showed that applying cellulose acetate/PVP nanofiber treatment with water at 60 °C for 3 h can wash PVP from the nanofibers and greatly increase the fiber's porosity (H. Lee, Nishino, Sohn, Lee, & Kim, 2018). The use of alkaline digestion by sodium hydroxide to remove SiO_2 and ZnO residues from electrospun nanofibers has been performed in recent studies. Removal of SiO_2 or ZnO led to the formation of fibers with high porosity (Aghasiloo, Yousefzadeh, Latifi, & Jose, 2019).

The influence of relative humidity on the production process of electrospun TiO_2/ZnO/PVP nanofibers was investigated (Aghasiloo et al., 2019). The study showed that increasing the relative humidity from 50% to 65% reduced the fiber diameter. This reduction was due to the dilution of the polymer solution during spinning and the increase in the electrospinning jet elongation, which increased the specific surface area of the fibers. With the relative humidity increased up to 80%, a decrease in fiber's diameter was observed because the excess water molecules were located between the tip of the needle and the collector, absorbing the extra charges of the electrospinning jet. This molecular polarization weakened the magnetic field and increased the fiber's diameter. Increasing the relative humidity also prevented the solvent from evaporating during the process, leading to an increase in fibers' diameter and specific surface area (Aghasiloo et al., 2019).

Increasing the specific surface area can also be achieved by reducing the fiber diameter. There are several ways to reduce the fiber diameter by optimizing various variables mentioned earlier. The use of different ratios of solvents in the polymer solution can change the physical properties of the solution, leading to the production of thinner fibers. For example, the addition of dimethylformamide (DMF) in a solution of polylactic acid (PLA) containing dichloroethane (DCM) can significantly reduce the fiber's diameter (Hajikhani, Emam Djomeh, & Askari, 2021). Increasing the DMF to DCM ratio further reduced the fiber's diameter. Interestingly, PLA is insoluble in DMF and excessive increase of this solvent causes PLA to precipitate in the polymer solution. DMF increases jet elongation and produces thinner fibers by increasing the electrical conductivity, vapor pressure, and dielectric constant of the polymer solution (Du, Xu, Zhang, & Zou, 2016).

The coaxial electrospinning technique is a common method for making two- or more-layered fibers. In recent years, this method has been used to design hollow fibers with high porosity. In this method, the solvent was used in the central layer and the polymer solution was used only in the outer layer. The resulting fibers were formed in a hollow form and had a high specific surface area. Studies have shown that the use of high volatility solvents can lead to rapid solidification of the polymer and the formation of porous and rough surfaces (Zaarour et al., 2020b). The use of different copolymers and the removal of them by different methods of digestion can lead to the formation of fibers with high porosity. However, the problem with this method is the possible fiber degradation during the treatment to remove the copolymer because the formed scaffolds have low mechanical strength (Hou et al., 2015).

Thermal degradation is one of the common treatments to increase the fiber's porosity. In this method, the prepared fibers are placed under a high temperature in an electric furnace. In this way, the organic part of the fibers or compounds with lower thermal stability decomposes and leads to the formation of porous structures in the fibers. In a recent study, porous carbon fiber was fabricated using heat treatment of electrospun polyacrylonitrile (PAN) and poly-(methyl methacrylate) (PMMA) fibers. Because PMMA has a lower thermal stability than PAN, it degraded during heat treatment and created cavities in the fiber structures (Gan & Gan, 2020).

Using the combined effect of variables can be useful in increasing the fiber's porosity. Using a high percentage of DCM as an organic solvent in the preparation of PLA fibers in the presence of a high ratio of relative humidity showed positive results. DCM evaporates rapidly due to increased volatility and the fibers take on a solid structure. The high ratio of relative humidity leads to the formation of condensed water droplets on the fibers and the pore templates remain on the surface of the PLA fibers. The high moisture content of the fibers leads to the design of the drying stage after spinning (T. Min et al., 2021).

4. The application of electrospinning technology in the rapid detection of food contaminants

Electrospinning techniques have been used to detect microbial contaminants, toxins, metal impurities, pesticide residues, and so on (Table 1). Electrospinning technology has the potential to increase the efficiency of rapid detection methods. The design of porous substrates to add analyte or immobilize intermediate materials has been considered in recent years. The high specific surface area of electrospun fibers can increase the resolution in rapid detection. In a previous study (Y. Luo et al., 2012), polyclonal antibodies related to the target organism were immobilized on the surface of electrospun nanofibers. The high porosity of the electrospun scaffold resulted in better surface functionalization. Cellulose nitrate nanofibers had an average diameter of 150 nm and were conjugated by glutaraldehyde as a crosslinker to the antibody. Porous and functionalized nanofibers can absorb *Escherichia coli* O157:H7 cells. The designed substrate increased the sensor's sensitivity to 67 CFU mL^{-1} and the linear range of the sensor response was calculated from 10^5 to 10^7 CFU mL^{-1}. By immobilizing various antibodies, it is possible to use this substrate to identify a wide range of bacteria. Increasing the porosity of nanofibers can also increase the specific surface area for antibody immobilizers (Y. Luo et al., 2012). In a similar study (L. Zhao et al., 2015), polystyrene-*co*-maleic anhydride (PSMA) electrospun nanofibers were used to identify *E. coli*. Tetraphenylethylene (TPE) and mannose were conjugated to nanofibers using poly(ethylene imine) spacer. The basis of this study is the tendency of TPE to bind to FimH proteins in *E. coli* fimbriae. The high specific surface area of electrospun nanofibers traps bacterial cells, which leads to greater binding and increased sensitivity in detection. *E. coli* was detected using fluorescent sensing. The more intense the bacterial load, the higher the fluorescent. The sensitivity of the sensor in this method was estimated to be 10^2 CFU $mL^{-1}$and it was able to detect *E. coli* at concentrations of 10^2 and 10^5 CFU mL^{-1} (L. Zhao et al., 2015).

Electrospinning nanofibers have been developed to detect chemical residues. Organophosphorus (OPs) and carbamates (CMs) two common pesticides that are difficult and costly to identify by conventional methods. Indole acetic acid (IA) is a plant hormone that can be hydrolyzed by the AChE enzyme to release indole phenol. Oxidation can lead to the conversion of this substance to blue indigo, but the presence of residual OPs and CMs interfere with AChE's functions. Thus, the presence of pesticide residues can be detected by color changes. In one study (Zhai et al., 2020), PVA nanofibers mixed with IA and AChE were used as a substrate for the rapid detection of pesticides. The detection time was less than 10 min and no color change in the sample indicated the presence of pesticide residues, while the control samples changed color. Thus, electrospinning was established as a promising method for designing rapid detection substrates for pesticide residues with high speed and accuracy (Zhai et al., 2020). The important point about food safety is not only the identification of potential contaminants in food, but also the use of new methods to maintain the quality of food for longer periods of time. Extensive capabilities of electrospinning can be used in the design of active packaging or patches for keeping food fresh. Electrospinning microstructures can be used to control permeability in packages (Topuz & Uyar, 2020). In addition, encapsulation of antimicrobial and antioxidant compounds can delay food spoilage by controlling the release of bioactive substances (Topuz & Uyar, 2020; Zhu, Zaarour, & Jin, 2019).

Heavy metals are common food contaminants and, especially, can be found in industrial wastewater. There are several methods used today for the rapid detection of heavy metals, including chemosensors. This method requires minimal equipment and can be useful in reducing time and cost. The unique properties of electrospun nanofibers have led to the use of electrospun scaffolds in substrate design for chemosensors and receptors. Chemosensor methods for detection of heavy metals are usually based on colorimetric, fluorimetric, and electrochemical detection. The basis of this measurement is the specific binding of an analyte to receptors, so by immobilizing the receptor on the surface of nanofibers, the performance of chemosensors increases. Receptors in electrochemical sensors are often based on alloys and amalgams, but organic and biological receptors are also used in other types of chemosensors. The binding of the target chemical to the substrate can induce signal changes received by the transducer. Some sensors use electron-rich donors. The binding of the receptor to the target chemicals leads to the intramolecular charge transfer and creates a color transition. Electrospinning technology can be used in the design of nanomaterial substrate-immobilized chemosensors or nanotreated transducer-based chemosensors. Nanomaterial substrate-immobilized chemosensors are created by physical adsorption or chemical bonding of chemosensors with electrospun fibers and is commonly used in optical detection. Nanotreated transducer-based chemosensors uses the physical adsorption or chemical bonding of receptors with electrospun fibers which often used for electrochemical and mass detection (S. Zhang, Jia, Liu, Wei, & Su, 2019).

In general, there are four ways to prepare electrospun fibers immobilized with chemosensors/receptors: the blending method to blend chemosensors/receptors with a suitable polymer to prepare fibers, the cross-linking of chemosensors/receptors with a suitable polymer, the physical adsorption method of attaching a suitable polymer and the chemosensors/receptors to the scaffold, and the chemical treatment of connecting chemosensors/receptors to fibers. The first two methods contain chemosensors/receptors inside and on the fiber surface. While in the third and fourth method, chemosensors/receptors are present only on the fiber surface (N. Zhang et al., 2017).

For example, N-allyl-4,5-di [(2-picolyl) amino]-1,8-naphthalimide (NAAP) was synthesized and copolymerized with methyl methacrylate

Table 1
A summary about recent applications of electrospinning technology for rapid detection in the food industry.

Detection group	Nanofiber Composition	Analyte/Organism	Limit of detection	Application/Description	References
Food pathogens	Poly(vinyl alcohol)/poly(acrylic acid)	*Escherichia coli*	10^2 CFU mL^{-1}	Rapid *Escherichia coli* detection in orange juice	Shaibani et al. (2018)
	poly-3-hydroxybutyrate	*E. coli* O157:H7	10^5 CFU mL^{-1}	Detection of *E. coli* O157:H7 cells in both milk and tryptic soy broth	Chen, Harrison, Ng, Sauvageau, and Elias (2021)
	SU-8 photoresist	*Salmonella* Typhimurium	10 CFU mL^{-1}	Sensitive and specific *Salmonella Typhimurium* whole cell detection	Thiha et al. (2018)
	Cellulose nitrate	*E. coli* O157:H7	64 CFU mL^{-1}	Immunobiosensor developed for rapid detection of *E. coli* O157:H7	(Y. Luo et al., 2012)
	Polydiacetylene/poly(ethylene oxide)/ polyurethane	*E. coli* ATCC25922	Not mentioned	Colorimetric sensor responding to *E. coli* and pH	Yapor et al. (2017)
Chemicals	Polyamide 6/Poly(allylamine hydrochloride)	Dopamine	0.15 μmol L^{-1}	Functionalized electrospun nanofibers as a differential pulse voltammetry electrode.	Mercante et al. (2015)
	Cellulose acetate	Biogenic amines	0.009, 0.003, and 0.006 mM using electrospun mat with 15, 30, and 60 min processing, respectively	Functional electrospun nanofibers for sensitive detection of biogenic amines for recognizing freshness of food products	Yurova et al. (2018)
	Polyacrylonitrile	Baseous ammonia	Less than 1 ppm by the naked eyes	Designing colorimetric ammonia gas sensor as hazardous air pollutant indicator	Hoang, Cho, Park, Yang, and Kim (2016)
	Cellulose acetate	Volatile basic nitrogen	Not mentioned	Designing colorimetric sensor as fish spoilage indicator	Aghaei et al. (2018)
	Chitosan/poly (vinyl alcohol)	Pirimiphos-methyl	0.2 nM	Designing electrochemical biosensor for rapid detection of pirimiphos-methyl in olive oil	El-Moghazy et al. (2016)
Pesticide residues	Poly(vinyl alcohol) with acetylcholinesterase and indolyl acetate	Omethoate, malathion, carbaryl and carbofuran	0.5 mg L^{-1} for omethoate, 1.5 mg L^{-1} for malathion, 0.1 mg L^{-1} for carbaryl and 0.02 mg L–[1] for carbofuran	Designing electrospun card for detecting pesticide residues in real food samples	Zhai et al. (2020)
	Poly(ε-caprolactone), indolyl acetate and acetylcholinesterase	Malathion	5 μg mL^{-1}	Design of pesticide detection cards via nano/micro-structured electrospun	Feng et al. (2021)
	Nitrocellulose	Triazophos	20 μg mL^{-1}	Electrospun membrane with an imprinted polymer to fix the recognition element for rapid detection of pesticide residues	He et al. (2020)
Heavy metals	Cellulose nanowhiskers	Pb^{2+}	10 nmol L^{-1}	Designing ternary nanocomposite sensor for monitoring metal ions in water samples	Teodoro, Shimizu, Scagion, and Correa (2019)
	Poly(1-pyrenemethylmethacrylate)-*block*-poly(N-isopropylacrylamide)-*block*-poly(N-methylolacrylamide)	Fe^{3+}	10^{-5} M	Designing multifunctional triblock copolymers nanofibers for metal-ion sensing	(J.-T. Wang et al., 2015)
	Poly(ether sulfones)	Cu^{2+}	1.1×10^{-9} M	Highly sensitive sensor based rhodamine dye doped poly(ether sulfones) electrospun nanofibers designed for detection of Cu^{2+}	(M. Min et al., 2013)
	Poly(N-isopropylacrylamide-*co*-N-hydroxymethyl-acrylamide)	Cu^{2+}	10^{-4} M	Fabrication thermo-responsive electrospun nanofibers for materials detection	Lin and Chen (2016)
	Poly(vinyl alcohol)	Fe^{3+}, Cr^{3+}, and Hg^{2+}	10^{-6} M for Fe^{2+} and Cr^{3+} 5×10^{-7} M for Hg^{2+}	Chemosensor based electrospun-immobilized for heavy metal removal in water purification with good recycle ability	Wei et al. (2014)
	Polycaprolactone	Hg^{2+}	50 ppt	Chemosensor designed by physical-immobilized fluorescent gold nanocluster on electrospun nanofiber for detection of heavy metal.	Senthamizhan, Celebioglu, and Uyar (2015)

polymer. The final scaffold was used as optical chemosensors to analyze Cu^{2+}. The addition of a copper-containing analyte shifted the fluorescence spectra up to 48 nm. The detection limit of the chemosensors was 20×10^{-6} M (W. Wang et al., 2011). The cross-linking method using polymer solutions was used to detect Zn^{2+} and Fe^{3+} (N. Zhang et al., 2017). The detection limits of the chemosensors were 10^{-8} and 10^{-5} M for Zn^{2+} and Fe^{3+}, respectively.

Among various methods, the physical blending method is the most widely used due to its simplicity. In one study (Saithongdee, Praphairaksit, & Imyim, 2014), the color change of curcumin in the presence of iron was used to detect Fe^{3+}. Zein electrospinning nanofibers containing curcumin were prepared by a mixed method. The chemosensors changed yellow to brown in the presence of Fe^{3+} with a detection limit of 1 mg mL^{-1}. Detection of Fe^{3+} by this method showed great accuracy in the presence of other ions (Saithongdee et al., 2014).

In the chemical bonding method with chemosensors/precursors, first, a polymer scaffold is prepared and then the fiber's surface is chemically functionalized with chemosensors/precursors. For example, poly (vinylbenzyl chloride) nanofibers were post-functionalized with imidazole-derived dye (Ondigo, Tshentu, & Torto, 2013b). In this study, the (2-(2′-pyridyl)imidazole) molecule was cross-linked on the surface of electrospun nanofibers to detect Fe^{2+}. The presence of Fe^{2+} ions led to a color change from yellow to red-orange, which can be seen with the naked eyes and the detection limit was 2 μg mL^{-1} (Ondigo et al., 2013b).

The physical adsorption method is introduced as another fiber preparation technique, which is very simple and does not require complex chemical synthesis or chemical post-functionalization. But unlike previous methods, it is not possible to use precursors in this physical adsorption method because these molecules are often dissolved in aqueous solutions and lose their efficiency in detecting heavy metals. The same technology was used to detect lead by physically adsorbed L-glutathione-conjugated gold nanoparticles on an electrospun Nylon-6/polyvinylidene fluoride scaffold. The presence of Pb^{2+} led to a change in color from pink to purple, and the detection limit was 10 μg dL^{-1} by naked eyes (N. Zhang et al., 2017). Other similar studies have used this technique to identify Hg^{2+} (N. Zhang et al., 2017) and Cu^{2+} (Senthamizhan, Celebioglu, Balusamy, & Uyar, 2015).

5. Applications of electrospinning technology in the design of sensors for rapid detection

Electrospinning technology has some advantages that make it ideal for the design of sensors, including the ability to use a wide range of polymers, control surface properties, and immobilize various receptors, as well as the unique properties of electrospun fibers. Articles published in recent years demonstrate that the future of the sensor industry will be closely related to electrospinning technology. This section introduces some of the applications of this technology in the design of sensors based on the latest studies.

5.1. SERS

SERS is classified as a subset of optical sensors (Fig. 2). This technique is more sensitive in diagnosis than the normal Raman method. The mechanism of action of SERS is to increase the sensitivity in detection via electromagnetic field enhancement or chemical enhancement (Asgari, Wu, Aghvami, Zhang, & Lin, 2021). This is done with the help of metal nanomaterials that are loaded on special substrates. Increasing the specific surface area by increasing the porosity and roughness in SERS substrates further increases the efficiency of this technique. The electrospinning technique has been introduced as a suitable candidate for designing SERS substrates with this capability (X. Zhao et al., 2019). Today, SERS is increasingly being used in the food industry to detect various analytes. A study on a substrate made of anatase TiO_2 nanofibers containing Ag nanoparticles showed that this substrate could detect organisms such as *E. coli* and *Staphylococcus aureus* without aptamer conjugation (Yang et al., 2018). The electrospun substrate also has a high content of Ag nanoparticles at the nanofiber surface due to its high surface porosity, which led to increased detection efficiency as well as high antimicrobial properties against both Gram-positive and Gram-negative bacteria. Finally, the Ag@TiO_2 substrate was introduced as a suitable option in the design of sensors to detect microorganisms (Yang et al., 2018). Another study developed a SERS substrate by electrostatic bonding of Ag to electrospun polyvinyl alcohol (PVA) substrate (X. Zhao et al., 2019). This substrate was highly sensitive in detecting crystal violet and malachite green, leading to accurate and *in-situ* detection of complex geometric structures. This method is an efficient approach to the design of biochemical sensors (X. Zhao et al., 2019). In a similar study (R. Zhang, Hong, et al., 2018), treated cellulose fibers were used for bacterial assay in SERS substrate design. The Ag nanoshell was deposited on the surface of electrospun cellulose nanofibers, and then the cellulose fibers were removed from the SERS substrate by aqueous treatment. The Ag-formed networks generated reproducible signals to detect molecular monolayers and bacterial cells, allowing different bacterial strains to be distinguished (R. Zhang, Hong, et al., 2018).

5.2. Aptamer-based biosensors

Aptamers are oligonucleotides or single-stranded DNAs that can bind to specific target molecules (Fig. 3). They can be isolated from a wide range of small to large molecules, whole cells, or even bacterial cells. The specific function of aptamers has led to their use in the design of biosensors due to their high sensitivity in detection. The combination of electrospun nanofiber properties and the specific performance of aptamers has led to the design of a new generation of biosensors with great sensitivity (Xie et al., 2021) and the use of aptasensor in detecting pesticides (Nguyen & Jang, 2021) and mycotoxins (L. Luo et al., 2020). Carbon scaffolds containing titanium nanoparticles were prepared by electrospinning technology (Xie et al., 2021). The surface of carbon nanofibers was modified with carbon ionic liquid electrode to be decorated with gold nanoparticles. Aptamer was stabilized at the nanofiber surface by binding thiol to gold ions (Au–S bond). The probe was designed to detect mercury and a linear performance range was between

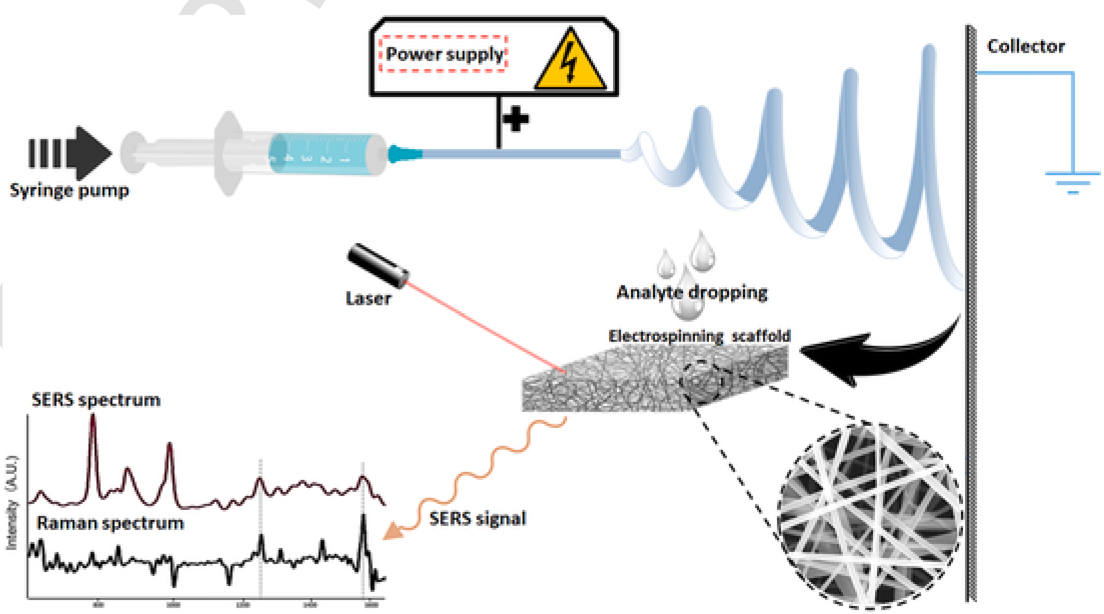

Fig. 2. An overview of the application of electrospinning in SERS-based rapid detection methods.

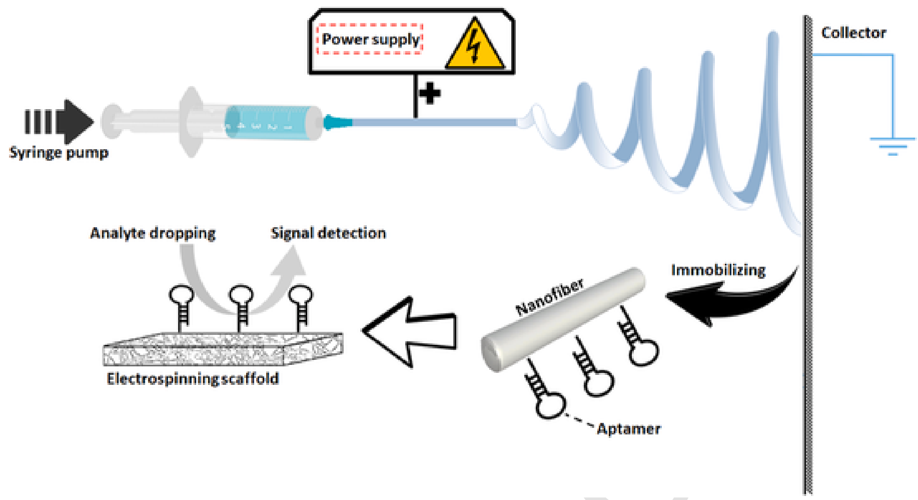

Fig. 3. An overview of the application of electrospinning in aptamer-based rapid detection methods.

1.0×10^{-15} to 1.0×10^{-6} mol L^{-1} with a detection limit of 3.33×10^{-16} (Xie et al., 2021). Functionalization of electrospun fibers was used to design protein-sensitive aptasensor (S. J. Lee, Tatavarty, & Gu, 2012). In this study, two thrombin-binding aptamers were immobilized to polystyrene-poly (styrene-*co*-maleic anhydride) fibers to detect thrombin. Aptamers were labeled with fluorescent dyes to be detected and measured by microscopic and spectroscopic methods. The results showed that thrombin was bound to the nanofiber surface and the minimum detectable concentration was 1 nM with a dynamic range of 1–200 nM. Labeling the aptasensor with quantum dots significantly reduced the detection limit to 10 pM with a dynamic range of 0.1–50 nM. Nanofiber-based aptasensor showed a sensitivity up to 2500 times higher than the 96-microwell plate method (S. J. Lee et al., 2012).

5.3. Surface plasmon resonance sensors

Surface plasmon resonance (SPR) is an optical method involving electromagnetic waves. The SPR method can detect biomaterials at very low concentrations by measuring the refractive index changes of surface propagation plasmons at the metal-dielectric interface (Fig. 4). This method is very sensitive to changes in the refractive index, allowing us to examine ultrathin films grown very close to the metal interface. This method has been developed to detect biomolecules because of its high accuracy, sensitivity, efficiency, and not requiring labeling. Sensor design based on this technology has many applications in rapid detection techniques (Barkat Rezaei, Rastegarzadeh, & Kiasat, 2018). In this section, the application of electrospinning in improving the performance of designed SPR sensors is introduced.

SPR has been used to detect environmental pollutants such as volatile organic compounds that are harmful to humans. Cadmium selenide/cadmium sulfide quantum dots along with silver nanoparticles were added to the polymer solution and the sensor glass substrate was prepared by the electrospinning method (Wu et al., 2020). UV-ozone treatment was used to etch the electrospun substrate and improve the specific surface area of electrospun fibers, leading to increased harvesting of volatile organic compounds. The presence of Ag^{2+} on the surface of the electrospun scaffold demonstrated detectability through polarization and surface plasmon resonance. The designed sensor showed detection limits of 100 and 500 ppm for butanol and chlorobenzene, respectively (Wu et al., 2020). In another study (Netsuwan et al., 2014), an immunobiosensor substrate for the detection of human immunoglobulin G was fabricated using electrospinning technology. A long-range surface plasmon resonance substrate was designed by electrospinning poly(acrylic acid) (PAA) fibers on a thin surface of the gold film. Increasing the specific surface area of nanofibers in LR-SPR can enhance the biosensor's efficiency. The electrospun scaffold was crosslinked with β-cyclodextrin to enhance water resistance and then heat-treated at 150 °C for 40 min. Increased water resistance increased

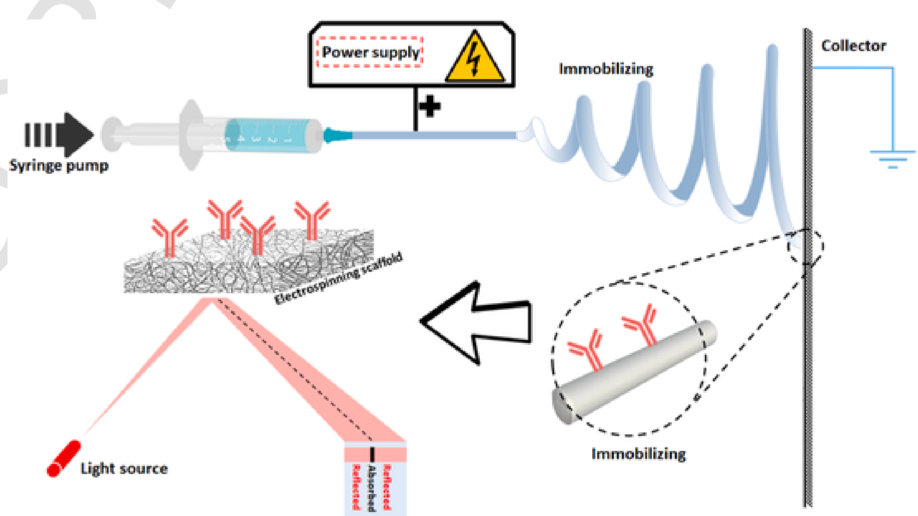

Fig. 4. An overview of the application of electrospinning in SPR-based rapid detection methods.

the number of active carboxylic acid groups in PAA, which enhanced the biosensor efficiency in detection (Netsuwan et al., 2014).

5.4. Direct electron transfer (DET)

Direct electron transfer (DET) is a new generation of biosensors based on the ability of an immobilized redox active protein to exchange electrons with an electrode during contact with analytes. Because electrons act as the second substrate of the enzymatic reaction that can displace the electrode potential, the measured current is correlated to the concentration of the analyte, which is often hydrogen peroxide (H_2O_2). For example, the activity of the oxidoreductase enzyme, which donates electrons from the oxidation reaction directly to the electrode (Schachinger, Chang, Scheiblbrandner, & Ludwig, 2021). Today, electrospinning technology is used to design DET electrodes that enhance electron conduction and biosensor performance due to the high active surface area of the nanofibers.

In a similar study (Alim, Kafi, Jose, Yusoff, & Vejayan, 2018), a biosensor electrode was fabricated by electrospinning technology for identifying H_2O_2 based on using carbon nanotube fibers and tin oxide. The fibers were prepared from PVP polymer with a tin precursor, then the redox proteins were immobilized on the fiber surface. The fibers showed high electrical conductivity due to the high porosity of the scaffold and resulted in a high load coefficient of biological molecules. The results showed the linear performance of the sensor at 1.0×10^{-6}–1.4×10^{-4} M of concentrations. Also, the detection limit of the developed electrode was 30 nM. The designed biosensor showed good stability and a rapid response to H_2O_2 (Alim et al., 2018).

6. Conclusion

Electrospinning is a simple, fast, cheap, and efficient option for developing sensors and rapid detection technologies. The flexibility of this technology in designing fibers with different surface properties can solve major challenges in the sensor industry. This review article provides an overview of the applications of electrospinning technology in sensors and summarizes the latest achievements in controlling fiber properties (morphology, diameter, and porosity). The study of controlling and increasing the porosity of electrospun fibers is critical in designing sensors for rapid detection because their sensitivity can be improved by increasing the specific surface area of the nanofibers. Some recent studies on electrospinning applications in the design of sensors for quality control in the food industry were discussed. In addition, some common methods improved by electrospinning technology for detecting food contaminants were elaborated. The combination of electrospinning technology with sensors is a big step toward increasing the detection power of various analytes such as microorganisms, toxins, and environmental pollutants. This combination has great potential in the food industry in controlling the quality and safety of food products by introducing some unique capabilities to increase the efficiency of sensors.

Data availability

No data was used for the research described in the article.

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