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Facile fabrication of foldable electrospun nanofiber electrodes for electrodes for electronic biosensing

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This study presents a rapid, sensitive and selective sensor based on electrospun nanofibers on laser-patterned electrodes, in which the laser micromachining process can directly fabricate the graphene electrode device for the electronical detection of glucose molecule. The layer of graphene film was formed on the glass substrate by a screen printing technique, and then the graphene electrodes can be fabricated by the ultraviolet (UV) nanosecond laser process with the wavelength of 355 nm. Based on the controlled the laser fluence and pulses, the electrode gap of 60 μm within the device can be fabricated. The polyvinyl alcohol (PVA) and glucose oxidase (GOD) composite nanofiber with weight percentage of 8% can be used by the electrospun process with used Glutaraldehyde(GA) steam for cross linking process. After that, it is blended with the conductivity of poly(3,4-ethylenedioxythiophene): poly (styrene sulfonate) (PEDOT:PSS), and the foldable nanofiber structures was made by electrospun process on the graphene electrode device. Finally, the electrical detection of graphene electrode device with the nanofibers at different concentrations of glucose can be measured, resulting in the linear variation is detecting the glucose concentration ranging form 0.01 to 3.12 mM. This work indicated that the low concentration and highly sensitive can be used for electronic biosensors.

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1. Introduction

In the past several years, the rapid advance in nanotechnology has opened up new possibilities of process to cultivate the next generation sensors for biosensing applications. In comparison to conventional nano-scale devices prepared by using semiconductor lithography, a dustless environment is required, and the manufacturing procedure is very complicated. The pulsed laser processing technology has potential advantages such as being maskless and being a programmable design for an electrode pattern structure and direct material removal in making screen-printed graphene film electrode structures.

Wakaya et al.[1] used a nanosecond laser for multilayer graphene material to study multilayer graphene processing with experimental parameters of wavelength 248 nm and pulse width 20 ns, further employing the Raman spectrum to analyze the observed changes in a structural out-of-sequence I_D/I_G peak intensity ratio. The findings showed that when the laser fluence was higher than 3 MW/cm^2, the I_G peak of the I_D/I_G peak intensity ratio fell and the I_D peak rose, because the laser processing increased the defects in the graphene thin layer. When the laser fluence was 1.4 MW/cm^2, the electrical variation was studied by using a single-shot pulsed laser and two-shot pulsed laser, respectively; the result showed that the resistivity of the electrode changed distinctly. This result provides a feasible method to make multilayer graphene film electrodes under atmospheric pressure.

Sahin et al.[2] used a femtosecond laser to process a single-layer graphene SiO_2/Si substrate with experimental parameters of wavelength 1030 nm, pulse width 550 fs, energy 100 nJ, and scanning speed 300 mm/s. The graphene layer was removed by using lower laser energy and increasing the laser scanning speed without damaging the SiO_2/Si substrate. When the speed was increased with lower energy, each shot of the laser only had a little overlapping, and thus the energy is absorbed only by the graphene layer and removed, the residual energy has not reached the ablation threshold of substrate, and the substrate will not be damaged. However, as this method uses femtosecond laser processing and uses a silicon wafer as the graphene substrate, its production is expensive and unfavorable for manufacturing low-cost and highly sensitive electronic biosensors. It is worth mentioning about the technology of laser-induced graphene (LIG), which has been successfully used in substrates with various carbon source contents. For example from organic films, plants, textiles and paper. Due to its advantages of fast fabrication, non-mask, and low-cost customizable fabrication. In recent years, LIG sensors it has been widely used in wearable electronics and smart biosensors that require high flexibility and versatility. [3-4]

The literature in recent years has paid extensive attention to the nanofiber preparation technique, as the electrospun technique is characterized by short preparation time and non-vacuum operation, and so it is extensively used for preparing fibers with a diameter range from several micrometers to tens of nanometers.[5-6] Liu et al.[7] used PVA-PEDOT:PSS to make an electroconductivity controllable nano-fibrous membrane for strain gauge. The addition of sulfuric acid or DMSO solution could remove resonium to increase the conductivity from 4.8×10^{-8} S/cm to 1.7×10^{-5} S/cm, and there was good Ohmic contact between the nanofiber and electrode. Chao et al.[8] employed PVA-PEDOT:PSS to make conducting nanofiber in linewidth of 100~300 nm on the micro-array electrode and performed cross-linking with the Glutaraldehyde (GA) solution. When the cross-linking concentration ratio was 1:50, a water insoluble nanofiber structure could be made, and the conductivity was 4~8 S/m. Ren et al.[9] used an electrospun technique to make the PVA-GOD nano-fibrous membrane for a glucose test. The findings showed there was a linear relationship when the glucose concentration was 0.1~1 mM.

This study utilized the pulsed laser ablation processing method to make a screen-printed multilayer graphene film electrode structure[10-11] and used the electrospun technique to make nanofiber as the sensing area. A nanofiber electronic biosensor is made by using a microchannel[12], and then the qualitative glucose solution concentration test is performed. The test is free of a dustless environment, in comparison to the present testing machine used in hospitals. A fast accurate testing method is thus provided, contributing to care preventive medicine.

2. Experimental

2.1. Experimental method

This experiment is characterized by using a non-vacuum process method to make a nanofiber graphene film electrode for electronic biosensors, with the purpose for testing the glucose solution concentration. The screen-printed multilayer graphene structure is processed by using a pulsed laser to make a film electrode.[13-14] The nanofibers of consistent wire size are then formed by using an electrospun technique and spray coated on the electrode structure to form a uniform porous nano-fibrous membrane as the sensing area, so as to develop an electronic biosensor. Finally, the microchannel is used for the qualitative electrical test for glucose concentration, so as to obtain an electrical measurement result with a linear relationship. The overall sensor fabrication design are shown in Figure 1.

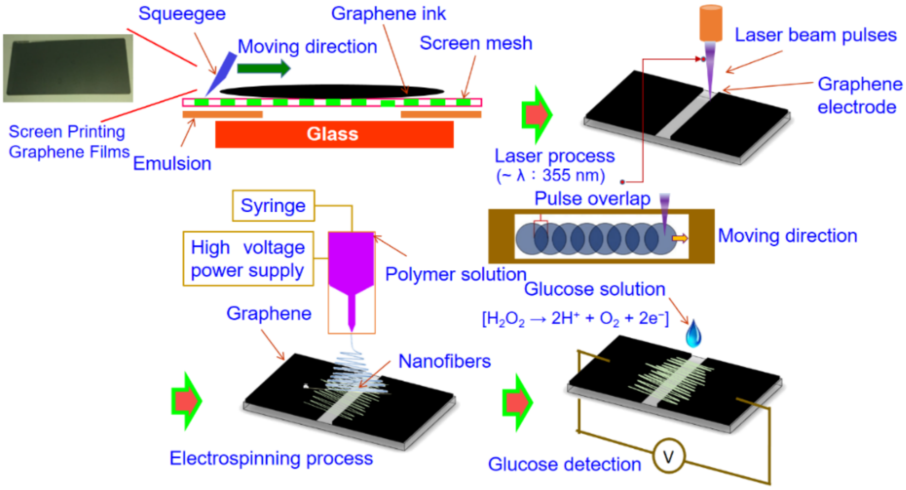


Fig. 1. Electronic biological detection electrode fabrication flowchart.

2.2. Fabrication of screen-printed graphene film electrode

The graphene ink is first transferred to the substrate by using a screen-printing technique. The graphene ink is instilled uniformly above the mesh, the glass substrate is placed under the mesh, and the ink is scraped with a squeegee, so that the ink distributes over the glass substrate uniformly through the mesh. The screen-printed graphene sample fabrication is then completed after drying.[15] The nanosecond UV pulsed laser of Neodymium-doped Yttrium Orthovanadate (Nd: YVO4) crystal in wavelength of 355 nm is used for processing. As an obvious heat affected zone will occur in nanosecond pulsed laser processing, three electrodes of 60, 120, and 180 μm gaps are fabricated in the electrode gap parameter design. Finally, the resistivity is measured to find out the electrode gap of the optimum electrical property. The schematic diagram of the experimental setup for the fabrication of graphene thin film electrodes by nanosecond UV pulsed laser processing and the fabrication of graphene electrodes with different sizes spacing are shown in Figure 2.

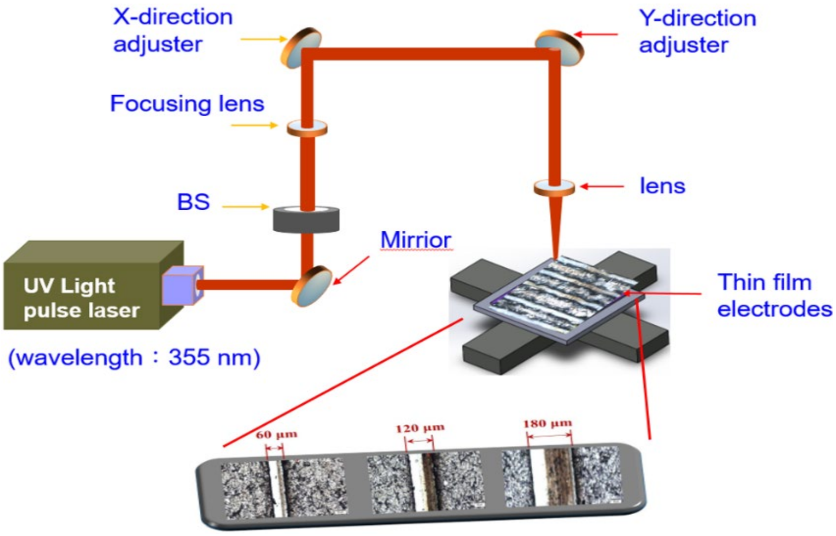


Fig. 2. Shows the nanosecond ultraviolet pulse laser processing to fabricate a graphene thin films electrode device and graphene electrodes with different spacing size.

The laser fluence value, laser pulse overlap rate, and energy ablation threshold shall be considered first in the laser process, so as to obtain appropriate working parameters. First, the energy values corresponding to different current percentages are corrected by a laser power meter, and the laser fluence value is calculated by Eq. (1).

$$Fluence = E / \pi D^2 \tag{1}$$

Here, E is the energy, J, and D is the laser light spot diameter, μm.

When the laser scanning speed is 100 mm/s and the laser repetition frequency is 100 kHz, the current percentage is modulated to record the relationships of laser energy to graphene thin layer removal width and depth. Starting from 15% of current and increasing by 2.5% each time until 40%, the 3D laser confocal microscope is used to observe and measure the depth and width of the processing line.

In the experiment on the laser pulse overlap rate, when the laser fluence is 1.2 J/cm^2 and the laser repetition frequency is 100 kHz, the stripe is processed at different scanning speeds. The speed starts from 100 mm/s, increased by 200 mm/s each time until 1500 mm/s for the experiment, and then the condition that leads to better removal efficiency is discussed. The pulsed overlap value is expressed as Eq. (2).[16]

$$PO = (1 - V / D \times f) \times 100\% \tag{2}$$

Here, V is the laser scanning speed, mm/s, D is the laser spot size, μm, and f is the pulse frequency repetition frequency, Hz.

Laser ablation is next performed, the purpose of which is to obtain the ablation threshold of the graphene film electrode layer after pulsed laser processing. The experimental energy thresholds at six points are measured using and optical microscope and then averaged. The theoretical value of the screen-printed graphene layer processing line width of different energies can be deduced from this parameter value by using Eq. (3) and compared with the actual line width value of experimental processing. The ablation threshold refers to the minimum fluence threshold of a nanosecond pulsed laser for removing material, expressed as Eq. (3).[17]

$$W^2 = 2\omega_0{}^2 \ln(F_0 / F_{th}) \tag{3}$$

Here, W is the laser processing line width, μm, ω_0 is the beam waist radius, μm, F_0 is the laser fluence, and F_{th} is the laser ablation fluence threshold.

2.3. Using an electrospun technique to fabricate nanofiber

This study used self-assembled electrospun equipment, in which the injection syringe is driven by a syringe pump, and the high-voltage power supply is connected to the constant voltage regulator to keep the working voltage at 16 KV. The output positive terminal of the high voltage meter is connected to the syringe needle tip, and the ground terminal is connected to the collector. The voltage, the distance from the syringe needle to collector, and the solution flow rate are adjusted to make the foldable nanofiber structure. The innoxious highly biocompatible PVA material with good chemical thermal stability (Mw=89000 g/mol) is selected as the main electrospun nanofiber polymer solution.[18-19] The PVA powder is mixed with DI water to prepare PVA solutions of five concentration weight percentages, which are 4%, 6%, 8%, 10%, and 12%, for electrospinning to fabricate nanofiber, and then it is blended with 1:1, 3:1, and 6:1 PEDOT:PSS solutions, and the optimum proportioning parameter is selected to enhance the electrical conductivity of nanofiber.

2.4. Electrical measurement method for glucose concentration of nanofiber film electrode

The graphene electrode sample with foldable nanofiber structures distributed uniformly on its surface is used for electrical measurement of glucose solutions at different concentrations, So as to observe the response curve variation. In terms of operating method, the GOD enzyme is fixed to the PVA/PEDOT:PSS composite nanofiber.[20] Afterwards, the glucose powder is mixed with phosphate buffer saline (PBS, pH ~ 7.4) to prepare a glucose solution. The electrical measuring instrument is connected to the nanofiber electrode and mixed with glucose solutions at different concentrations at fixed intervals for electrical measurement, So as to obtain the electrical values at those different glucose solution concentrations.

3. Results and discussion

3.1. Screen-printed graphene on laser-patterned electrode device

The Raman spectrometer in optical wavelength of 514 nm (Hitachi Corp., model: U-4001, Japan) is used to test the material characteristics of the screen-printed graphene thin layer. Three Raman intrinsic spectrum peaks are obtained. As shown in Figure 3, they are I_D peak (level of defect in lattice), I_G peak (degree of graphitization), and I_{2D} peak (stacking degree) at 1354, 1581, and 2717 cm^{-1}, respectively, but the I_{2D} peak intensity is lower than I_G peak intensity, and it is supposed to be multilayer graphene material.

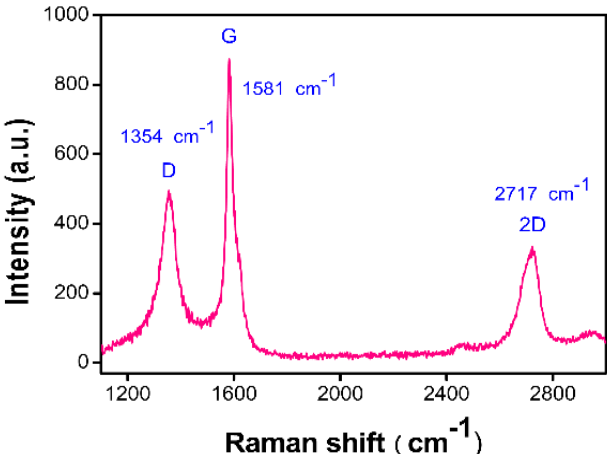


Fig. 3. Raman spectrogram of screen-printed graphene thin layer.

The screen-printed graphene layer surface morphology and section thickness are analyzed through an atomic force microscope (AFM) (Veeco Bruker DI Dimension, model: 3200, Germany) and a scanning electron microscope (SEM) (Hitachi Corp., model: S-3400N-II, Japan). As shown in Figures 4(a)–(c), the image of SEM shows that the screen-printed graphene surface is very uniform.

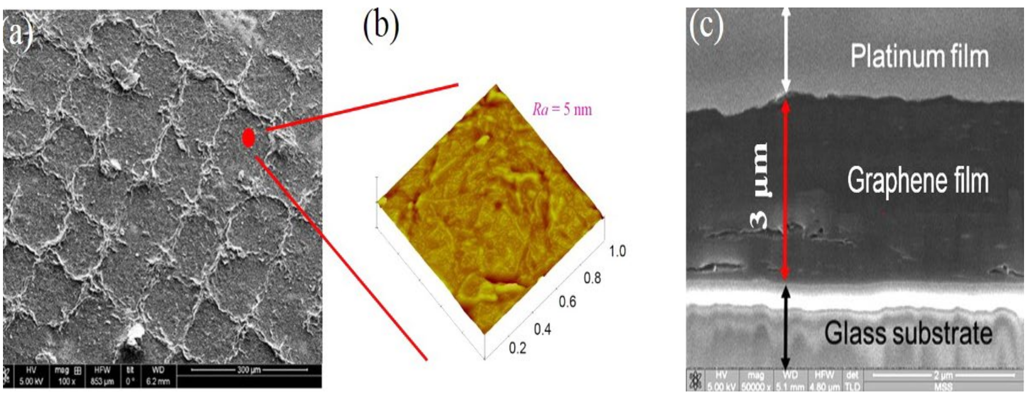


Fig. 4. (a) Screen-printed graphene sample surface roughness analysis, (b) enlarged view of regional image, and (c) section thickness.

The film thickness is about 3μm. The AFM measured surface roughness (Ra) is about 5nm. According to the optical equation absorptivity (A) + reflectivity (R) + transmittance (T) = 1, the spectrophotometer is used to measure optical characteristics, and reflectivity (R) and transmittance (T) of the front side and back side of sample are measured. The reflectivity is lower than 3% in the range from UV band to infrared band, and the transmittance is almost 0%. The energy absorptivity of a screen-printed graphene layer is higher than 97% in the range from UV band to infrared band. Therefore, this is favorable for subsequent electrode fabrication by laser processing. The laser wavelength is 355 nm for the experiment, because the experimental values show that the maximum energy absorptivity value occurs in this band, an accurate line width may be obtained in laser processing, and the electrode gap control will be more accurate.

The two working parameters of laser fluence and pulsed overlap are important parameter values that influence the processing depth and line width changes in the fabrication of a screen-printed graphene film electrode sample. The average energy threshold is 0.3 J/ cm^2 after ablation processing, and the ablation threshold after laser processing of the screen-printed graphene film electrode sample is deduced from Eq. (3). Figure 5 (a) shows the line width and depth values of the screen-printed graphene film electrode processed by different pulsed laser energies when the pulsed laser fluence is 0.6 to 4.7 J/ cm^2. The fluence 4.7 J/ cm^2 is selected as the parameter of laser ablation, the processing line depth is about 2.6 μm, the width is about 60μm, and the working electrode line width edge is relatively flat. However, if the fluence is increased continuously, then the graphene thin layer will be ablated up by the laser, damaging the glass substrate. Therefore, this experiment selects fluence value 4.7 J/ cm^2 as the experimental parameter value of optimal laser processing to fabricate the graphene film electrode. Figure 5(b) shows the ablation processing and heat affected zone when the pulsed laser fluence value is 4.7 J/ cm^2.

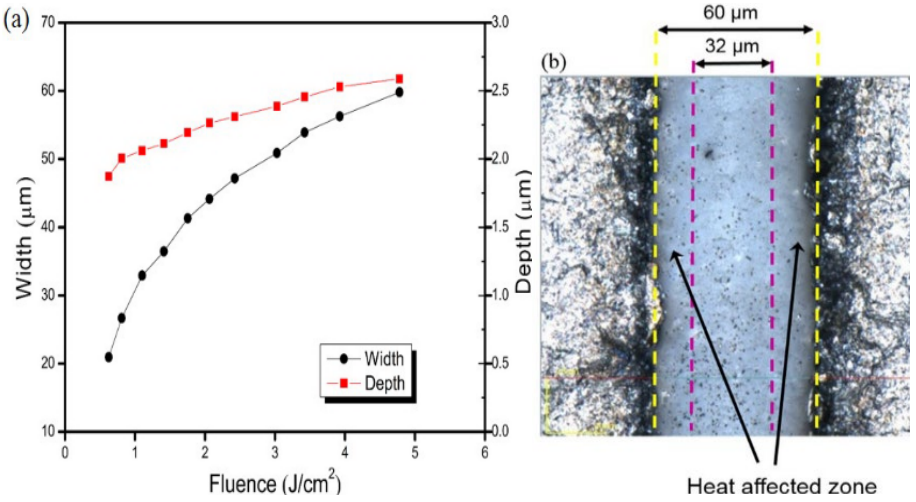
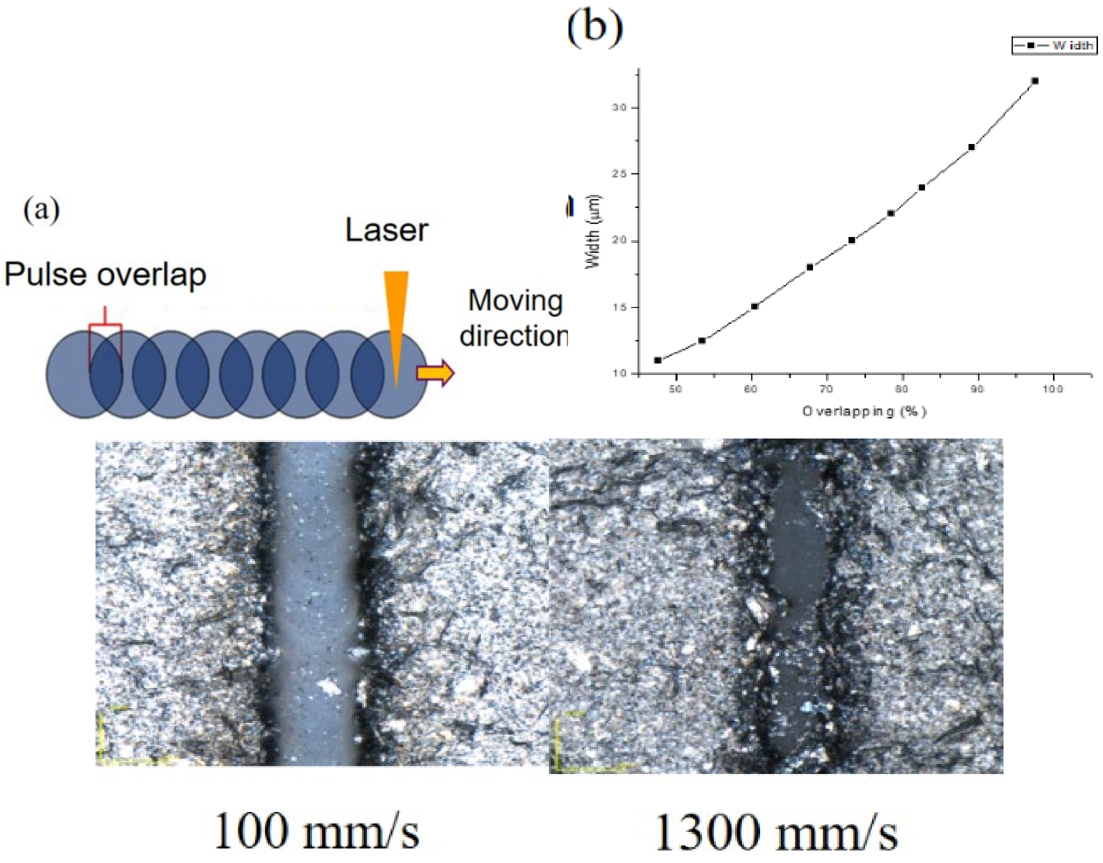


Fig. 5. (a) line width and depth of screen-printed graphene thin layer processed by different pulsed laser energies, (b) ablation processing and heat affected zone of pulsed laser fluence value 4.7 J/ cm^2.

When the laser fluence increases, the actual processing line width deviates from the theoretically calculated line width. The heat transfer time of phonon in the lattice is about 1ps, whereas the residence time of each shot of energy of the nanosecond pulsed laser is about several nanoseconds, or much lower than the heat transfer rate of phonon. Therefore, using nanosecond pulsed laser ablation to remove material will result in a heat affected zone. As the laser fluence increases, the heat affected zone resulting from the increase of heat absorption is enlarged. The experimental results show that accurate electrode width and depth can be obtained only if an appropriate pulsed laser fluence value is selected for electrode machining.

In terms of the pulsed overlap working parameter setting, Figure 6(a) shows the laser pulse overlap rate processing, while Figure 6(b) shows the relation between laser pulse overlap rate and processing line width. When the laser scanning speed is increased by 200 mm/s each time from 100 mm/s to 1500 mm/s, the line width decreases from 32μm to 14μm as the pulsed overlap decreases; and the pulsed overlap value decreases from 96.6% to 49.5%, meaning the processing line width and laser pulse overlap rate decrease as the laser scanning speed increases. The removed line width edge will be more uneven and rough, and electrode accuracy declines. Figure 6(c) shows the graphene film electrode line width processing when the pulsed laser scanning speed is 100 mm/s and the scanning speed is 1500 mm/s. When the scanning speed is 100 mm/s for processing, the laser pulse overlap rate is high, and the processed line width edge is relatively even. Therefore, the experimental processing parameters are laser scanning speed 100 mm/s and pulsed overlap 96.6% for electrode machining. The electrodes of 60, 120, and 180μm line widths are fabricated by pulsed laser processing, and the resistivity values measured by an electric meter at fixed range are 560Ω, 620Ω, and 660Ω. The measured values show that the electrical property is the best when the electrode width is 60μm. It is used as the experimental value for making electronic biosensors.



100 mm/s 1300 mm/s

Fig. 6. (a) Schematic diagram of laser pulse overlap rate processing, (b) line width variation of different laser pulse overlap rates, and (c) conditions of scanning speeds 100 mm/s and 1500 mm/s.

3.2. Composite nanofiber cross-linking blending

After the laser processing of screen-printed graphene film electrode is completed, the foldable nanofiber is made by using an electrospun technique. According to the nanofiber material characteristics, a good nano-fibrous membrane depends on such physical parameters as whether there is high specific surface area or not, whether the fiber line diameter is consistent or not, and if the membrane material structure has high porosity. In the biological detection experiment, the nanofiber made by the electrospinning technique shall contact an aqueous solution, so that the nanofiber is dissolved, and the sensor fails. The cross-linking process is then required,[21] so as to make the nanofiber resistant to water and have stable GOD activity. This experiment uses an ultrasonic atomizer to atomize glutaraldehyde vapor for about 30 min of PVA/GOD cross-linking. The PVA nanofibers electrospun at five weight percentage concentrations, which are 4%, 6%, 8%, 10%, and 12%, are then analyzed. This experiment uses PVA nanofiber at weight percentage concentration of 8% and GOD for cross-linking. In comparison to the problems resulting from experimental parameters of the other weight percentage concentrations, e.g. forming a part of coarse and flat nanofibers, the electrospun nanofiber line width increases with concentration and the fibers form into beads; thus, it is better than the PVA nanofibers at the other weight percentage concentrations. Figure 7(a) shows the nanofiber generated by using the electrospun technique; the distance between electrode head and collector is 10 cm, and the spray coating speed is 10 μL/min. Figure 7(b) shows the PVA nanofiber at weight percentage concentration of 8% in the electrostatic scanning electron micrograph. Its diameter is 120~160 nm, the wire size is uniform, the wire size error is small, and the specific surface area is high, thus enhancing the sensitivity and stability of detection, and so it is selected as the subsequent blending parameter.

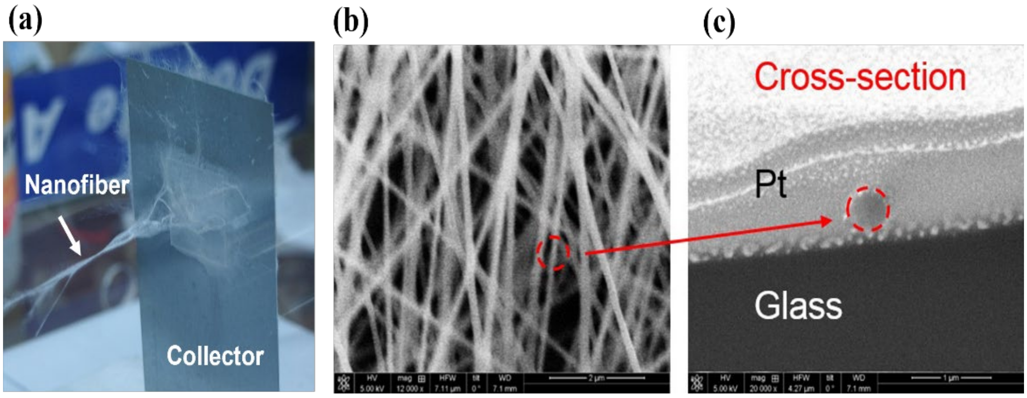


Fig. 7. (a). The electrospinning nanofiber structure is produced work distance between needle head and collector plate is 8 cm with 10 μL/min velocity, (b) nanofiber generated by using electrospun technique, (c) scanning electron micrograph of PVA/GOD nano-fibrous membrane at weight percentage concentration of 8%.

The PEDOT:PSS is blended with the PVA/GOD nanofiber after cross-linking, for the purpose of enhancing the electrical response and environmental stability of the nanofiber. Figures 8(a)-(c) show different proportions of PVA/GOD nanofiber and PEDOT:PSS. When the blending ratio is 1:1, as shown in Figure 8(a), the line diameter of the nanofiber remains uniform. When the blending ratio is 3:1 and 6:1 respectively, as shown in Figures 8(b)-(c), the nanofiber structure diffusing from the center like neurons or inhomogeneous line diameter is observed locally, making it inapplicable for sensing the nanofiber structure. Thus, there will be good electrical performance when the blending ratio 1:1 is selected as the parameter condition for the biological detection experiment.

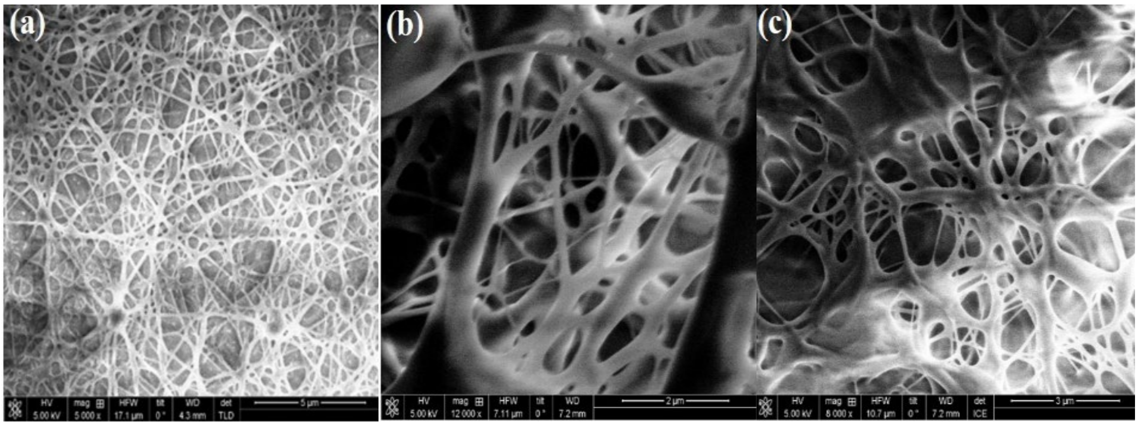


Fig. 8. SEM image of blending of PVA/GOD nanofiber after cross-linking and PEDOT:PSS; the blending solution ratios are (a) 1:1, (b) 3:1, and (c) 6:1.

3.3 Foldable nanofibers on graphene electrode device for glucose detection

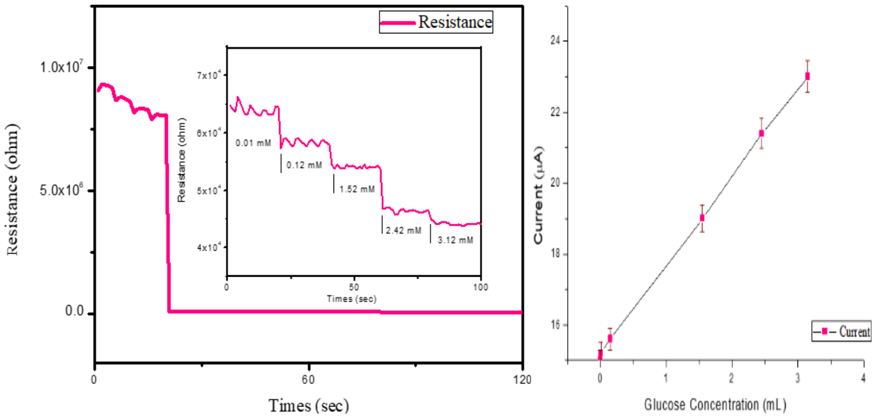
The PVA/GOD / PEDOT:PSS composite nanofiber after cross-linking and blending will form a foldable nano-fibrous membrane on the screen-printed graphene film electrode. According to the foldable characteristic of a fiber, it can distribute over the electrode uniformly, and there are high adsorbability and high porosity. The glucose solution is instilled in the foldable nanofiber region on the screen-printed graphene electrode for electronic biological electrical testing. The instrument is a nano component electrical parameter measurement system (Agilent, model: B1542A; HP, model: 81110A; and Agilent, model: 4284A, U.S.A.). The glucose bioassay mechanism uses GOD and glucose solution for a specific enzyme reaction. The GOD comprises two proteins, whereby each protein contains one flavine adenine dinucleotide (FAD), which is a cofactor. When the glucose molecule contacts GOD, the dehydrogenation reaction arises to form gluconolactone. When it contacts oxygen, the GOD- $FADH_2$ is oxidized to generate H_2O_2. The H_2O_2 exhibits an auto redox reaction on the electrode to generate electrons, so that the electrical property can be measured. The measured resistivity is inversely proportional to the quantity of H_2O_2, and the current is proportional to the quantity of H_2O_2, so that the concentration of glucose solution can be quantified indirectly. The reaction equation is expressed as:[22]

$$\text{GOD-FAD} + \text{glucose} \rightarrow \text{GOD-FADH}_2 + \text{gluconolactone} \quad (1)$$

$$\text{GOD-FADH}_2 + O_2 \rightarrow \text{GOD-FAD} + H_2O_2 \quad (2)$$

$$H_2O_2 \rightarrow 2H^+ + O_2 + 2e^- \quad (3)$$

The left picture of Figure 9 shows the resistivity response values measured at different glucose concentrations by the PVA/GOD / PEDOT:PSS composite nanofiber electrode after blending.



Fig

. 9. Left picture shows the resistivity changes of PVA/GOD/PEDOT:PSS composite nanofiber graphene electrode at different glucose concentrations. Right picture shows the response relation between different glucose concentrations and current.

As the water molecules in the glucose solution are electrolyzed when the voltage is higher than 1.2 V, the measured values may contain the electrons of water molecule electrolyzed, leading to errors in the measured values; as such, the given voltage for the test is set as 1 V. It is observed in the left picture that the resistivity changes drastically with time before and after the glucose solution is applied. It may because the foldable nanofiber graphene electrode sample has good sensitivity. Afterwards, the resistivity response changes that result from instilling glucose solutions at different concentrations in the duration are presented.

The inset shows the relationships among the glucose solution concentrations of 0.01~3.15 mM, resistivity, and time. It is observed that the resistivity of sensing element decreases as the glucose solution concentration increases. Because the glucose solution molecules and the PVA/GOD / PEDOT:PSS composite nanofibers bond together to perform a chemical reaction and generate H_2O_2, which contacts the electrode to perform self oxidation reduction reaction and to generate electrons, the measured resistivity decreases. In addition, as shown in the right picture of Figure 9, the magnitude of the current is approximately linear when the measured glucose concentrations are 0.01, 0.12, 1.52, 2.42, and 3.12 mM, meaning it is applicable to a low concentration high sensitivity glucose test.

4. Conclusion

This study's experiment successfully prepared a screen-printed nanofiber graphene film electrode with minimum electrode gap of 60μm by combining nanosecond pulsed laser processing with an electrospun technique, which can be used as an electronic biosensor detection device. The pulsed laser processing technique is characterized by maskless direct write and accelerated electrode fabrication. The experimental values of UV pulsed laser wavelength 355 nm, fluence 4.7 J/ cm^2, and laser pulse overlap rate 96.6 % are used as the optimal parameters for making the electrode structure. In order to obtain high conductivity, the electrospun technique can be used for spray coating to form foldable nanofibers on the surface of the electrode structure. As the nano-fibrous membrane structure has high porosity, high adsorption biocompatibility, and higher specific surface area, the detection sensitivity can be enhanced greatly. In the glucose biomolecule test, the glucose solutions at different concentrations are added to the electrode respectively. The resistivity distinctly changes when 0.01 mM glucose solution is applied, and the current response has a linear relationship at 0.01~3.12 mM. This range is applicable to a low concentration and high sensitivity glucose solution test. This study thus provides a method to rapidly fabricate a graphene nanofiber film electrode and confirms the feasibility for direct testing of biological samples.

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