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Transparent and ultra-flexible PEDOT:PSS/ITO/Ag/ITO on Parylene thin films with tunable properties

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ABSTRACT

Transparent and flexible conductive materials are critical components in many optoelectronic devices, such as wearable electronics, biosensors, displays, etc. Conventional transparent electrodes made of a single material, such as indium tin oxide (ITO), ultrathin metals, graphene and poly-(3, 4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS) have limitations and hardly possess the desired combination of broadband transmittance, low electrical resistivity, mechanical flexibility, and biocompatibility. Herein, we designed and constructed an ultra-flexible, conductive, transparent thin film using a PEDOT:PSS/ITO/Ag/ITO multilayer structure on Parylene C. The multilayer assembly was optimized to achieve the lowest theoretical reflectance by simulating the coatings admittance loci under the preferred reference wavelength. ITO and Ag were deposited consecutively using RF magnetron sputtering at room temperature, followed by spin-coating of PEDOT:PSS. The sputtering deposition temperatures were tuned to achieve the optimal optical and electrical properties. Compared to a single-layer ITO film of equivalent thickness, the multilayer films exhibited significantly decreased sheet resistance, reduced electrochemical impedance, remarkable transmittance, large Young's modulus values, and superior stability in air and saline. The multilayer films also showed strong adhesion to the Parylene C substrate and subsequently excellent bending tolerance. Moreover, the peak transmittance of our multilayer flexible thin films could be tailored to a specific wavelength for particular applications, such as optogenetics that utilizes light of different wavelengths to excite or inhibit the activity of genetically targeted neurons or intracellular signaling pathways.

Keywords: flexibility, oxide/metal/oxide, transparent electrode, ultrathin metal, optogenetics, indium-tin-oxide, silver

1. INTRODUCTION

Transparent and flexible conductive thin films are critical components in optoelectronics, such as wearable electronics, biosensors, displays, etc. [1]. A single layer of ITO, ultrathin metals, graphene or PEDOT:PSS, as conventional transparent electrode materials, have been widely used. Although they have some dramatic properties, they all have their own limitations. ITO has been applied because of its transparency over the entire visible spectrum [2], biocompatibility [3], and reasonably good electrical conductivity. However, the mechanical flexibility of ITO still needs to be concerned due to its relatively brittle property [4], especially when thicker ITO (~100 nm) is required for achieving the high transmittance and feasible conductivity. In addition, the electrical conductivity of ITO cannot achieve as low as the ultrathin metal. One of the problems of the ultrathin metal is the low ~30-70 % transmittance with ~3-7 nm thickness [5]. The transmittance is relatively low compared with other optical materials, making it difficult to implement the high transparency and high conductivity at the same time [6]. Biocompatibility and stability also vary depending on the different metal materials. For example, Au thin films have better biocompatibility and stability than Ag. Recently, graphene has been considered as a popular material for making transparent microelectrodes due to its broad-spectrum transparency, excellent conductivity, good biocompatibility, and flexibility [7, 8]. However, graphene still needs to face serious challenges that high-quality graphene over large areas requires either high temperatures of over 1000^{o}C or specific substrate materials, which are incompatible with polymer materials. To solve these challenges, a transferring method is utilized to obtain graphene-on-polymer thin films, which, however, requires complicated experiment techniques and may reduce the yield of production. PEDOT:PSS as an attractive conductive polymer succeeds to be applied in the microelectrodes for neural interface applications because of its outstanding biocompatibility, excellent stability, transparency, and flexibility [9-11]. However, it has limited electrical conductivity, making it unsuitable for miniaturized electrodes.

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In this paper, we reported, for the first time, an ultra-flexible, conductive, and transparent thin film using a PEDOT:PSS/ITO/Ag/ITO multilayer structure on Parylene C for achieving simultaneously enhanced sheet conductivity, reduced electrochemical impedance, remarkable transmittance, excellent adhesion, stability, and flexibility. The multilayer assembly was optimized to achieve the lowest theoretical reflectance by simulating the coatings admittance loci under the preferred reference wavelength. ITO and Ag were sputtered consecutively at room temperature, followed by spin-coating of PEDOT:PSS. The room temperature was chosen depending on our systematic study of the deposition temperature on the film transmittance and sheet resistance by depositing 100 nm ITO films on glass slides and 10 μm Parylene C coated glass slides at 22°C, 69°C, 92°C, 116°C and 140°C, using RF magnetron sputtering system. The transmittances and sheet resistances of the combined films and single-layer ITO of equivalent thickness were measured by utilizing a Filmetrics thin film analyzer and a four-point probe instrument, respectively. Tunable peak transmittances were confirmed by depositing different thicknesses of each layer on the Parylene C substrate after the theoretical admittance loci stimulation under selected wavelengths of 470 nm, 550 nm, and 630 nm. The electrochemical impedance spectroscopy (EIS) was used to measure the electrochemical impedance of different coatings in a 0.9% NaCl (saline) solution, proving the reduced electrochemical impedance of the combined film. The adhesion between ITO and Parylene C was evaluated using Scotch tape tests, where no film delamination was observed after 50 times peeling. The bending tests were conducted to demonstrate the mechanical flexibility and robustness of the combined thin film on the Parylene C substrate. The Young's modulus of the combined thin film was also measured quantitatively using a MTS nano-indenter and compared with the theoretical values. Finally, the combined thin films were placed in air and in saline individually for up to five weeks to evaluate their stability.

2. METHOD

2.1 Fabrication

To study the temperature effect of ITO deposition, 100 nm ITO films were sputtered at 22°C, 69°C, 92°C, 116°C, and 140°C, respectively. Before ITO sputtering, glass slide substrates were ultrasonically cleaned in an acetone bath for removing organic contaminants and particles, followed by cleaning with ethanol, and deionized (DI) water. Then, the glass slides were dried in an N_2 stream and baked on a hotplate at 100°C for 30 mins. For some cleaned glass slides, 10 μm Parylene C was deposited (PDS 2010, Specialty Coating System, Inc) as the substrate, which could be peeled off to form a flexible film. The sputtering process was carried out in an RF magnetron sputtering system (Denton Explorer-14, Denton Vacuum, Inc) with a 4-inch diameter ITO target (99.99% purity), which is composed of 90% indium oxide and 10% tin oxide. Before the introduction of argon, the base pressure inside the chamber was reduced below 2×10^{-6} torr. During the pumping procedure, the heater was turned on to allow sufficient time for the substrate temperature to increase to the desired value. Then, 60 sccm argon was introduced into the chamber to achieve 3×10^{-3} torr sputtering pressure. Substrate rotating speed was set at 20% to ensure the uniformity of the sputtered film and pre-sputtering was ran for 10 mins to get rid of the contaminants on the target surface. During the deposition, the sputtering RF power was set at 100 W with the deposition duration of 15 mins and 40 secs for 100 nm ITO. After the sputtering, the heater was turned off immediately and the samples were kept in the chamber until the temperature decreased to the room temperature.

For the PEDOT:PSS/ITO/Ag/ITO samples, the admittance loci coating stimulation was accomplished first to calculate the optimized thickness of each layer with the lowest theoretical reflectance under the preferred wavelengths: 470 nm, 550 nm, and 630 nm. Then we studied the transparency, conductivity, electrochemical properties, mechanical properties and stability of the combined PEDOT:PSS/ITO/Ag/ITO film under 550 nm specifically. Therefore, the following fabrication steps will be illustrated focusing on the combined PEDOT:PSS (30 nm) /ITO (24 nm) /Ag (9.5 nm) /ITO (20 nm) thin films under the 550 nm reference wavelength. Both two layers of ITO were sputtered with the same steps described above. However, the deposition time was reduced to 2 mins 30 secs and 3 mins 6 secs for obtaining the thicknesses of 20 nm and 24 nm, respectively. For the Ag deposition, a 4-inch diameter Ag target (99.99% purity) was installed as another cathode in the Denton system. The base pressure, sputtering pressure, and sputtering RF power were kept the same as the ITO sputtering, while the deposition time was changed to 22 secs for 9.5-nm-thick Ag and the substrate rotating speed increased to 70%. ITO/Ag/ITO deposition was finished step by step consecutively without breaking the vacuum. Last, 0.55% PEDOT:PSS, which was diluted from 1.1% PEDOT:PSS (768642, Sigma-Aldrich), was spun on the top of the substrate with 500 rpm spin speed for 5 secs and then 4000 rpm for 120 secs, followed by baking on a hotplate at 100°C for 30 mins. For comparison, 53.5 nm single layer ITO which had the equivalent thickness with the ITO/Ag/ITO structure was sputtered under the same recipe.

2.2 Admittance loci coating stimulation

Admittance loci coating stimulation was performed before the multilayered PEDOT:PSS/ITO/Ag/ITO film fabrication for optimizing each layer with different thicknesses and achieving the highest theoretical transmittance under the preferred wavelength. Reflective index n and k of PEDOT:PSS, ITO, Ag, glass and Parylene C substrate were obtained respectively from the reliable database (Filmetrics). Then, the specific thicknesses of PEDOT:PSS, Ag, and two layers ITO were decided according to the final optimized admittance loci, where the end of the loci should reach to the point (1, 0) on the horizontal axis. The transmittance results depending upon theoretical stimulation thicknesses might differ with the experimental results because of the different quality and brand of materials as well as different experimental deposition conditions, as compared to the data from the database. The thickness of each layer needs to be adjusted based on the experimental results in order to attain the final experimental transmittance plot under the preferred wavelength.

2.3 Testing methods

To fully understand the temperature impact on the ITO quality, the Filmetrics thin film analyzer (F20-UVX, Filmetrics, Inc) was utilized to measure the transmittance of the ITO samples prepared under different temperatures in a wavelength range of 300-800 nm. The four-point probe instrument (SRM-232, Bridge Technology, Inc) was applied to measure the sheet resistance of these ITO samples. For the multilayer combined PEDOT:PSS/ITO/Ag/ITO films, besides the transmittance and sheet resistance measurements, EIS measurements were performed using a potentiostat (Electrochemical Analyzer, CH Instruments, Inc.) to analyze the electrochemical impedance in a three-electrode cell, with the ITO or PEDOT:PSS coated ITO microelectrode as the working electrode (WE), an Ag/AgCl electrode as the reference electrode (RE), and a platinum electrode as the counter electrode (CE). The tests were conducted in physiological saline solution at room temperature. The electrochemical impedance of the microelectrode was measured from 0.1 Hz to 100 kHz when a 5 mV RMS sinusoid waveform was applied to the WE. Long-term humidity and temperature tests were performed in an incubator with a humidity of 60% at 37^{o}C. To confirm the stability of the combined films, the samples were placed in air and in saline for up to 5 weeks, and their sheet resistances were measured every week to record the changes in sheet resistances. To evaluate the adhesion between ITO and Parylene C, Scotch tape test was conducted by applying the Scotch No. 810 pressure-type tape on the ITO-Parylene C samples with a blade-scratched 10 $\times$ 10 array of 1 mm^2 squares. The peeling test was repeated up to 50 times with 180^{o} peel-off angel to inspect whether any ITO was delaminated from the Parylene C substrate. Furthermore, the flexibility of the combined films was studied by conducting the bending tests under a bending curvature of 3 mm and 6 mm in diameter, respectively. The sheet resistances of the combined films with different total ITO thickness were measurement every 100 bending cycles within the total 600 bending cycles. Last, the Young's modulus of the combined films was measured using the MTS Nanoindenter XP system, and the experimental value was compared with the theoretical values.

3. RESULTS AND DISCUSSION

3.1 Temperature study of ITO

Fig. 1 (a) shows the transmittances of five 100 nm ITO samples on the glass slides and five 100 nm ITO samples on the Parylene C substrates, which was sputtered at five different temperatures of 22°C, 69°C, 92°C, 116°C and 140°C. The sputtering temperatures of below 150°C were chosen carefully because the glass transition temperature of Parylene C is 80-100°C [12]. The transmittances of ITO on the glass slides were similar throughout the wavelength of 300-800 nm when the sputtering temperature was lower than 92°C. However, when the temperature went over 116°C and reached 140°C, the transmittances before ~440 nm were increased, resulting in a down shift of the peak transmittance. This down shift was caused by the improved crystallinity of ITO, which is closely related to the increased temperature [13]. Fig. 1 (b) shows the opposite experiment results under the Parylene C substrate. At a temperature of over 92°C, the transmittances between ~400 nm and ~600 nm dropped gradually, and the transmittance decreased by 7% from ~85% to ~78% under the 500 nm wavelength. This reduction in transmittance is mainly attributed to the changed in the Parylene C polymer properties at the sputtering temperature of above the glass transition temperature of Parylene C. The periodic waves appeared in Fig. 1 (b) were due to the light interference with the thin Parylene C substrate.

Fig. 1 (c) shows the average sheet resistances (n=5) of 100 nm ITO on the glass and Parylene C substrates under the different sputtering temperatures, respectively. The sheet resistances decreased from 60.96 Ω/sq to 24.23 Ω/sq on the glass substrate, while the sheet resistances reduced from 61.82 Ω/sq to 32.75 Ω/sq on the Parylene C substrate when the temperature increased from 22°C to 140°C. The high temperature deposited ITO has an improved degree of crystallinity

[13], resulting in better transparency and conductivity compared with the room temperature deposited amorphous ITO. However, the reduction in sheet resistance differs between the two substrates under the same sputtering temperature increase, which was caused by the low glass transition temperature of Parylene C.

The total sheet resistance of PEDOT:PSS/ITO/Ag/ITO multilayer thin films was a result of a parallel combination of the four individual layers. For the single layer of 30 nm PEDOT:PSS, 24 nm ITO or 20 nm ITO, the sheet resistance was in the range of hundreds of Ω /sq. However, for 9.5 nm Ag, the sheet resistance was as low as 10 Ω/sq. Therefore, the conductivity of the Ag would dominate the whole conductivity of this multilayered PEDOT:PSS/ITO/Ag/ITO thin films and the conductivity of PEDOT:PSS and ITO could be neglected. It is also noted that the sputtering temperature did not play an important role in improving the overall conductivity of the multilayered thin films. In fact, for the combined films deposited on the Parylene C substrate, the higher temperature resulted in the lower transmittance of the films, especially throughout the wavelength between ~400 nm and ~600 nm. Moreover, we observed darker Ag when the sputtering temperature increased, leading to lower transmittance and higher resistivity of the combined ITO/Ag/ITO films. This is mainly due to the oxidation of Ag at elevated temperatures. For all the reasons above, the room temperature was preferred during the ITO/Ag/ITO sputtering deposition.

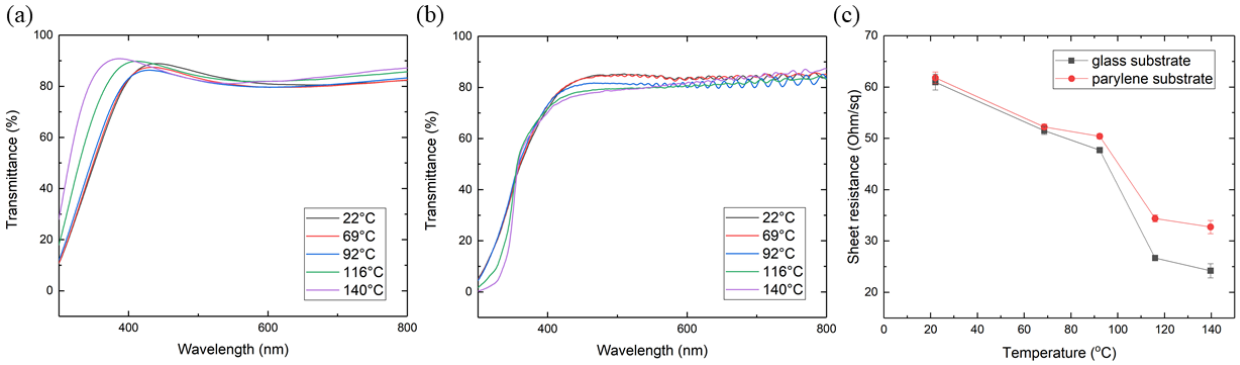

Figure 1. (a) Transmittances of 100 nm ITO deposited under different sputtering temperatures of 22°C, 69°C, 92°C, 116°C, and 140°C on (a) pure glass slides and (b) Parylene C coated glass slides. (c) Sheet resistances of 100 nm ITO deposited under different sputtering temperatures of 22°C, 69°C, 92°C, 116°C, and 140°C on pure glass slides (grey) and Parylene C coated glass slides (red), respectively.

3.2 Combined thin films

After the deposition of PEDOT:PSS (30 nm) /ITO (24 nm) /Ag (9.5 nm) /ITO (20 nm) thin films on the 10 μm Parylene C substrate, the transparent and flexible films were released from the glass substrate, as shown in Fig. 2 (a) and (b). The transmittances of the combined films were measured and illustrated in Fig. 2 (c). It can be seen that the combined PEDOT:PSS/ITO/Ag/ITO had a significant increase in transmittance throughout the visible spectrum from 300 nm to 700 nm on both Parylene C substrate and glass substrate, compared to 53.5 nm single layer ITO with the equivalent thickness. Under the 550 nm wavelength, the combined films show an improvement in transmittance by ~7% from ~78% to ~85% on Parlylene C, and by ~14% from ~75% to ~89% by ~14% on glass. The sheet resistance of the combined film was significantly reduced from 147.5 Ω/sq to 8.81 Ω/sq, as shown in Fig. 2 (e). There was no change for the sheet resistance of the combined films before and after adding PEDOT:PSS, as confirmed by the four-point probe. In addition, the uniformity of the deposited thin films was evidenced by the standard deviation of the sheet resistance measurements over the entire substrate (n=5), which was improved from 1.53 to 0. Fig. 2 (d) shows that the combined films on the Parylene C substrate with different thicknesses after the admittance loci coating stimulation could achieve wavelength-dependent transmittance peaks at 470 nm, 550 nm, and 630 nm. This result confirmed that these combine films could be specifically designed and optimized for different applications. For example, blue light with 470 nm wavelength has always been used in the field of optogenetics to activate channelrhodopsin (Ch2R), a light-sensitive ion channel protein for neuron excitation [14]. Similarly, yellow light under 580 nm wavelength could be utilized to activate halorhodopsin (NpHR) ion pumps that inhibit the activity of the targeted neurons, while another protein, OptoXR, allows for receptor-mediated intracellular signaling in response to green light stimulus under 530 nm wavelength [14].

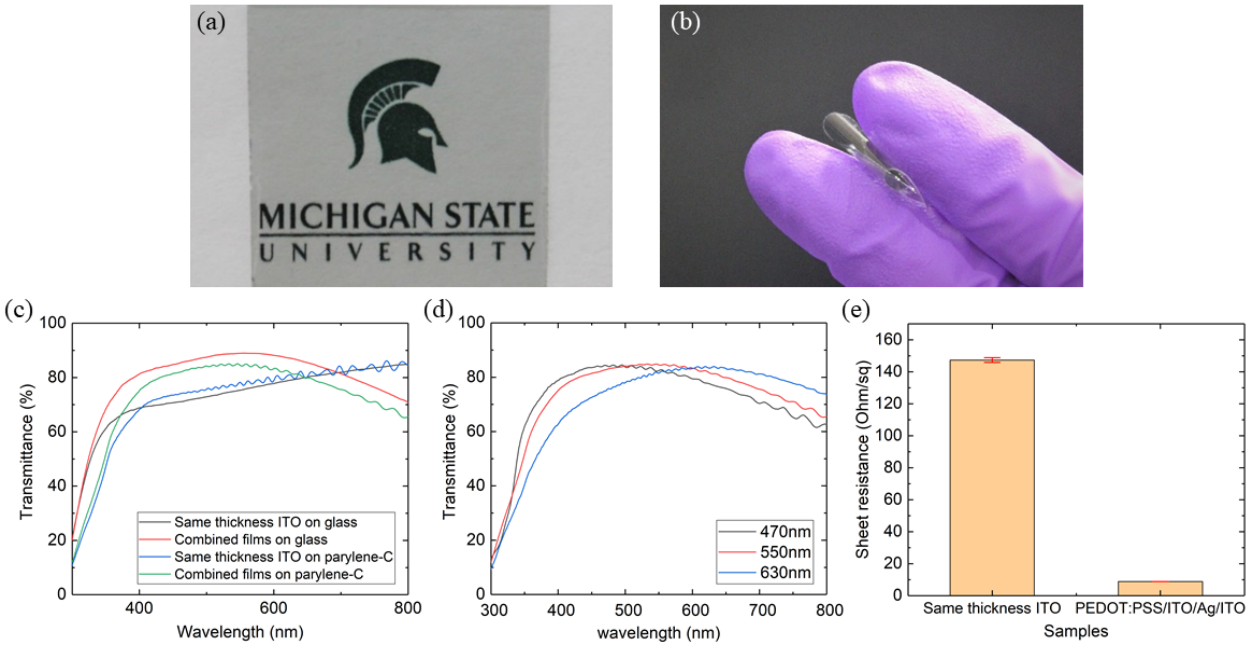

Figure 2. Thin films of PEDOT:PSS/ITO/Ag/ITO on 10 μm Parylene C, showing (a) excellent transparency and (b) ultra-flexibility. (c) Transmittances of the combined PEDOT:PSS/ITO/Ag/ITO films compared with pure ITO of equivalent thickness on the glass and Parylene C substrates, respectively. (d) The peak transmittances of the combined PEDOT:PSS/ITO/Ag/ITO films on Parylene C at wavelengths of 470 nm, 550 nm, and 630 nm, respectively, to confirm the tunable peak transmittance by stimulating the coatings admittance loci. (e) Average sheet resistances (n=5) of PEDOT:PSS/ITO/Ag/ITO and the equivalent thickness ITO on the Parylene C substrate with neglectable standard deviation to confirm the uniformity of the combined films using the four-point probe measurements.

3.3 Flexibility

Bending tests were performed to validate the flexibility of the ITO/Ag/ITO combined film on the Parylene C substrate with different ITO thicknesses. Three samples were tested, including 37 nm ITO/ 10 nm Ag/ 33 nm ITO, 15 nm ITO/ 10 nm Ag/ 38.5 nm ITO, and 20 nm ITO/ 9.5 nm Ag/ 24 nm ITO, corresponding to total ITO thicknesses of 70 nm, 53.5 nm, and 44 nm, respectively. The changes in the sheet resistance of the flexible ITO/Ag/ITO thin film were measured by the four-point probe and expressed as ΔR/R, where R was the initially measured sheet resistance and ΔR was the difference in the sheet resistance before and after bending cycles. Bending curvatures with diameters of 3 mm and 6 mm were tested individually in order to check how the sheet resistance changed with different degree of bending. From the results shown in Fig. 3 (a) and (b), we found that the films with thinner ITO coating are more robust to withstand the bending. From previous research results, a single layer of ITO was identified as a brittle material with a large Young's modulus [4]. However, when it came to the ITO/Ag/ITO structure, the tolerance to bending cycles was improved significantly and thinner ITO coating in the combined structure would help to further improve the bending tolerance. What's more, the results indicated that for smaller bending diameter, the changes of sheet resistance were more noticeable but still within a good tolerance range because the changes of sheet resistance were less than 100%.

After averaging all 36 indents from the nano-indenter, the measured Young's modulus of the pure Parylene C film and the combined film on Parylene C was 4.049 GPa and 4.064 GPa, respectively. Theoretical calculations were carried out via the equation for less mixture of particle and polymer:

$$\frac{1}{E_{composite}} = \frac{V_{ITO}}{E_{ITO}} + \frac{V_{Ag}}{E_{Ag}} + \frac{V_{ITO}}{E_{ITO}} + \frac{V_{Parylene}}{E_{Parylene}}$$

where E is the Young's modulus and V is the volume fraction of each layer. During the calculation, the volume fraction was replaced by the layer thickness by assuming the same cross-sectional area. The Young's moduli of Parylene C, ITO, and Ag are GPa [15], 116 GPa [16], and 85 GPa, and the thicknesses were 10 μm, 44 nm, and 9.5 nm, respectively, resulting in a calculated $E_{composite}$ of 4.021 GPa. The measured Young's moduli were close to the theoretical values, confirming the mechanical properties of our fabricated combined films.

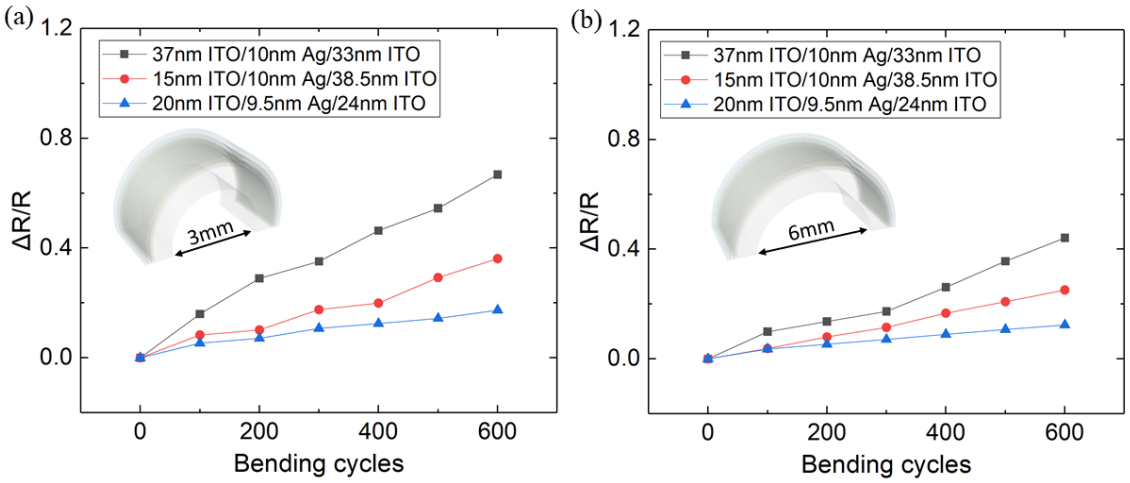

Figure 3. Sheet resistances of different combined films with different total ITO thickness were measurement every 100 bending cycles within the total 600 bending cycles under (a) 3 mm bending diameters and (b) 6 mm bending diameters, respectively.

3.4 Other properties

For the EIS measurements, the Bode plots in Fig. 4 (a) show the electrode impedance magnitudes of the bare ITO, ITO/Ag/ITO, and PEDOT:PSS/ITO/Ag/ITO versus frequency. The ITO/Ag/ITO structure effectively reduced the overall electrical impedance over a wide frequency range of 0.1 Hz - 100 kHz. And the coating of PEDOT:PSS further decreased the electrochemical impedance in a wide frequency spectrum, in consistency with other reports [17]. The electrochemical impedance of the combined film at 1 kHz decreased by at least one order of magnitude from 243.85 Ω/cm^2 to 19.88 Ω/cm^2 compared with the pure ITO film. This impedance reduction was mainly attributed to the improved conductivity of the film and the increased surface roughness of the electrode resulted from the PEDOT:PSS coating, as confirmed in our previous work [18]. The rough surface provided a large effective surface area of the electrode when exposed to the electrolyte, allowing more charge to flow across the electrode-electrolyte interface.

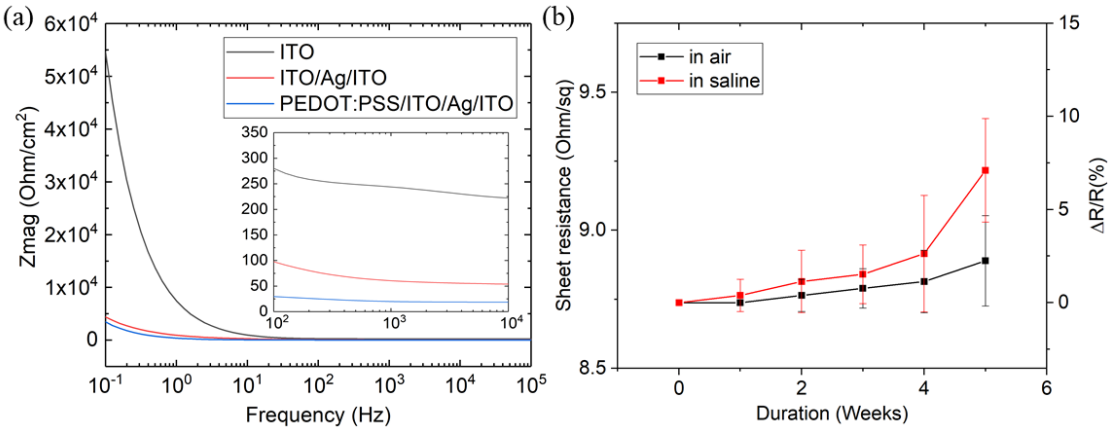

Figure 4. (a) Electrochemical impedance magnitudes of the bare ITO, ITO/Ag/ITO, and PEDOT:PSS/ITO/Ag/ITO versus the frequency. (b) Sheet resistance changes (n=5) of the combined films in air and in saline at 37°C within five weeks.

To evaluate the stability of the microelectrodes, the PEDOT:PSS/ITO/Ag/ITO combined films were stored at 37°C in both air and saline environments for up to 5 weeks, and their sheet resistances were monitored weekly and plotted in Fig. 4 (b). Our results show that the combined films exhibited excellent stability with a slight resistance increase of less than 1.25% and 2.75% after four-week exposure in air and saline at 37°C. After five weeks, the combined films stored in air still exhibited good stability with an overall resistance increase of less than 2.5%, while the samples stored in saline showed a resistance increase of less than 7.5%. As the initial sheet resistance of our combined films was as low as 8.81 Ω/sq, the 7.5% increase only raised sheet resistance to 9.25 Ω/sq after 5 weeks, which was still very promising for use as

transparent electrodes in short-term neural interface applications. The increase in the sheet resistance is mainly due to the diffusion of ionic solution into the multilayered film through defects or pinholes in the top-most ITO layer, which could accelerate the corrosion and oxidation of the Ag coating. One of our future goals is to refine the substrate cleaning and sputtering recipes to eliminate these residual particles as much as possible, which will further reduce the pinhole and defects in the ITO coating.

According to peeling tests, no ITO film delaminated from the Parylene-C for all 5 samples after 50 times peel-off tests. However, for some samples, Parylene C delaminated from the glass surface because of poor adhesion of Parylene C with glass [19]. Our peel-off tests result also confirmed that the adhesion of ITO-Parylene C interface had better adhesion compared with glass-Parylene C interface.

4. CONCLUSION

This paper, for the first time, reported an ultra-flexible, conductive, transparent thin film using PEDOT:PSS/ITO/Ag/ITO multilayer structure on Parylene C, which exhibited significantly reduced sheet conductivity, decreased electrochemical impedance, tunable peak transmittance with higher transmittance over the pure ITO film, excellent adhesion, stability, and flexibility, suitable for use in many different biomedical applications as optoelectronics. The effects of the sputtering temperature and layer thickness on the properties of the combined thin films were studied theoretically and experimentally to achieve the desired film properties. Our combined films also show remarkably improved conductivity and transmittance as compared to the pure ITO thin film. In addition, the combined thin films remained stable over 5 weeks in both air and saline environments at 37°C. Scotch tape peeling tests confirmed the strong adhesion strength between the ITO/Ag/ITO structure and the Parylene C substrate. Finally, bending tests results validate the excellent flexibility of the combined thin films and suggest that the thinner ITO coating in the multilayered structure results in better bending tolerance. Future work will focus on refining the processing parameters to optimize the film properties, and exploring the applications of this combined transparent thin film in optogenetic neuro interfaces.

REFERENCES

[1] Yokota, T., Zalar, P., Kaltenbrunner, M., Jinno, H., Matsuhisa, N., Kitanosako, H., Tachibana, Y., Yukita, W., Koizumi, M. and Someya, T., "Ultraflexible organic photonic skin," Sci. Adv. 2(4), e1501856 (2016).

[2] Tuna, O., Selamet, Y., Aygun, G. and Ozyuzer, L., "High quality ITO thin films grown by dc and RF sputtering without oxygen," J. Phys. D: Appl. Phys. 43(5), 055402 (2010).

[3] Ahani, A., Saadati-Fard, L., Sodagar, A. M. and Boroumad, F. A., "Flexible PET/ITO electrode array for implantable biomedical applications," Conf. Proc. IEEE Eng. Med. Biol. Soc., 2878-2881 (2011).

[4] Boehme, M. and Charton, C., "Properties of ITO on PET film in dependence on the coating conditions and thermal processing," Surf. Coat. Technol. 200 (1-4), 932-935 (2005).

[5] Kossoy, A., Merk, V., Simakov, D., Leosson, K., Kéna□Cohen, S. and Maier, S. A., "Optical and structural properties of ultra-thin gold films," Adv. Opt. Mater. 3(1), 71-77 (2015).

[6] Lee, W., Kim, D., Matsuhisa, N., Nagase, M., Sekino, M., Malliaras, G. G., Yokota, T. and Someya T., "Transparent, conformable, active multielectrode array using organic electrochemical transistors," Proc. Natl. Acad. Sci., 201703886 (2017).

[7] Kuzum, D., Takano, H., Shim, E., Reed, J. C., Juul, H., Richardson, A. G., De Vries, J., Bink, H., Dichter, M. A., Lucas, T. H. and Coulter, D. A., "Transparent and flexible low noise graphene electrodes for simultaneous electrophysiology and neuroimaging," Nat. Commun. 5, 5259 (2014).

[8] Park, D. W., Schendel, A. A., Mikael, S., Brodnick, S. K., Richner, T. J., Ness, J. P., Hayat, M. R., Atry, F., Frye, S. T., Pashaie, R. and Thongpang, S., "Graphene-based carbon-layered electrode array technology for neural imaging and optogenetic applications," Nat. Commun. 5, 6258 (2014).

[9] Greco, F., Zucca, A., Taccola, S., Menciassi, A., Fujie, T., Haniuda, H., Takeoka, S., Dario, P. and Mattoli, V., "Ultra-thin conductive free-standing PEDOT/PSS nanofilms," Soft Matter 7 (22), 10642-10650 (2011).

[10] Cho, C. K., Hwang, W. J., Eun, K., Choa, S. H., Na, S. I. and Kim, H. K., "Mechanical flexibility of transparent PEDOT: PSS electrodes prepared by gravure printing for flexible organic solar cells," Sol. Energy Mater. Sol. Cells 95 (12), 3269-3275 (2011).

[11] Abidian, M. R., Ludwig, K. A., Marzullo, T. C., Martin, D. C. and Kipke, D. R., "Interfacing conducting polymer nanotubes with the central nervous system: chronic neural recording using poly (3, 4 - ethylenedioxythiophene) nanotubes," Adv. Mater. 21 (37), 3764-3770 (2009).
[12] Goda, T., Konno, T., Takai, M. and Ishihara, K., "Photoinduced phospholipid polymer grafting on Parylene film: Advanced lubrication and antibiofouling properties," Colloids Surf. B 54(1), 67-73 (2007).
[13] Hu, Y., Diao, X., Wang, C., Hao, W. and Wang, T., "Effects of heat treatment on properties of ITO films prepared by RF magnetron sputtering," Vacuum 75(2), 183-188 (2004).
[14] Pama, E. A., Colzato L. S. and Hommel, B., "Optogenetics as a neuromodulation tool in cognitive neuroscience," Front. Psychol. 4, 610 (2013).
[15] Ejserholm, F., Stegmayr, J., Bauer, P., Johansson, F., Wallman, L., Bengtsson, M. and Oredsson, S., "Biocompatibility of a polymer based on off-stoichiometry thiol-enes+ epoxy (OSTE+) for neural implants," Biomaterials Res. 19(1), 19 (2015).
[16] Wittkowski, T., Jorzick, J., Seitz, H., Schröder, B., Jung, K. and Hillebrands, B., "Elastic properties of indium tin oxide films," Thin Solid Films 398, 465-470 (2001).
[17] Guex, A. A., Vachicouras, N., Hight, A. E., Brown, M. C., Lee, D. J. and Lacour, S. P., "Conducting polymer electrodes for auditory brainstem implants," J. Mater. Chem. B 3(25), 5021-5027 (2015).
[18] Yang, W., Broski, A., Wu, J., Fan, Q. H. and Li, W., "Characteristics of transparent, PEDOT: PSS-coated indium-tin-oxide (ITO) microelectrodes," IEEE Trans. Nanotechnol. 17(4), 701-704 (2018).
[19] Tsai, F. T., Chuang, C. T., Li, T.C. and Yu, P. C., "Study of parylene-C thin film deposited on flat substrates," Appl. Mech. Mater. 217, 1077-1082 (2012).