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One-step rod coating of high-performance silver nanowires-PEDOT:PSS flexible electrodes with enhanced adhesion after sulfuric acid post-treatment

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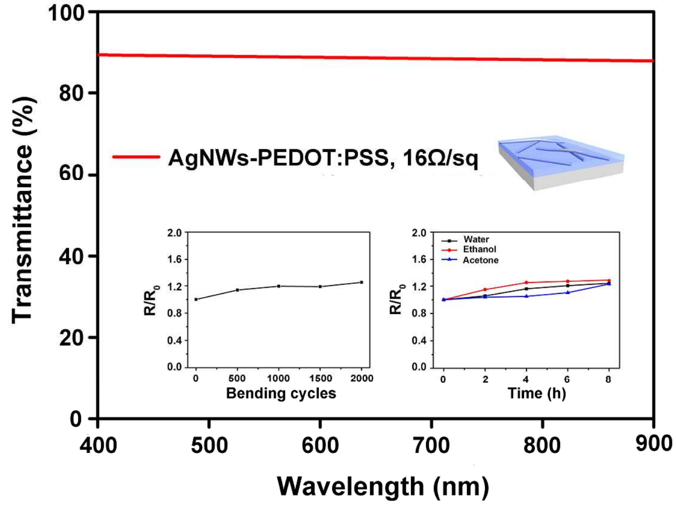

The AgNWs-PEDOT:PSS film treated with H_2SO_4 exhibited both excellent optoelectric and adhesive performance.

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ARTICLE

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Excellent optoelectric and adhesive properties are two crucial parts for the application of transparent conductive electrodes (TCEs). In this work, AgNWs are composited with PEDOT:PSS to prepare high-performance TCEs using a one-step rod-coating approach, where AgNWs are buried in PEDOT:PSS resulted in largely weakened roughness and lowered sheet resistance from 164 Ω/sq to 16Ω/sq at 90.4% optical transmittance. Besides, the addition of Zonyl improved the wettability of hydrophilic PEDOT:PSS on hydrophobic substrates, which enhanced the adhesion of AgNWs-PEDOT:PSS film. Sulfuric acid post-process further strengthened the interaction between PEDOT:PSS and organic substrate contributing improved adhesion of AgNWs-PEDOT:PSS composite film. The as-prepared film exhibits favorable stability in both cyclic deformations and solvent environment. The solution-processed fabrication, outstanding photoelectric performance and strong adhesion imparts the AgNWs-PEDOT:PSS tremendous potential in the application of flexible electronics.

Introduction

As a key component of rapidly developing optoelectronics, transparent conductive electrodes (TCEs) have been extensively studied in the applications of organic light-emitting diodes, solar cells, and touch panels.[1-4] Featuring high transparency and conductivity, indium tin oxide (ITO) is considered as the most common material to fabricate TCEs. However, ITO-based TCEs are limited to scarcity of indium and expensive sputtering process. What's more, the brittleness of ITO has severely restricted its application in future flexible electronics.[5] Therefore, several alternative materials have been developed to replace ITO such as carbon nanotubes (CNTs),[6-8] graphene,[9-13] conductive polymer[14-18] as well as silver nanowires (AgNWs).[19-22] Among the various candidates, only graphene grown by chemical vapor deposition (CVD) and AgNWs exhibit comparable optoelectronic performance to ITO. Due to the complex preparation process of CVD graphene, AgNWs are considered as the most promising one for replacing ITO. Besides, percolation network of AgNWs possesses superior flexibility and TCEs capable of bending can be obtained by coating AgNWs on flexible substrates.

To perform well in the application of flexible optoelectronics, low sheet resistance at high optical transmittance and strong adhesion are two significant properties for TCEs. However, when integrated into commercial devices, AgNWs conductive films sustained several problems such as high contact resistance, rough surface as well as poor adhesion to substrates. Though thermal annealing, mechanical press as well as plasmonic welding have been applied to overcome these problems, the high-temperature or high-pressure processes are not suitable for polymer substrates.[23-26] To composite with a second electrically conductive component is another approach to solve the problems, which compensates each other and leads to a higher-performance product. CVD graphene,[27] single wall CNT[28] as well as poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)[29-31] were separately composited with AgNWs imparting the electrodes with low sheet resistance at relatively high optical transmittance. However, the preparation usually requires a vacuum filtration, transfer process or repeated coating, which greatly restricted the large-scale manufacture of AgNWs-based TCEs. What's more, the adhesive performance is rarely mentioned. Notwithstanding some effort has been

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devoted to enhance the adhesion, the introduction of insulated binder usually goes with reduced electrical property. For instance, dopamine was used as an adhesive layer to increase the interaction between PDMS and AgNWs, but the sheet resistance is 35 Ω/sq at relatively low optical transmittance of 80%.[32] Though AgNWs film coated with GO and PDDA shows both enhanced adhesion and low sheet resistance, the process needs seven times of repeated coating, which limited its actual application.[33] Hence, the large-scale preparation of TCEs with both excellent optoelectric and adhesive performance of TCEs shows huge potential.

In this work, we report a one-step approach to manufacture AgNWs-PEDOT:PSS composite films, by rod coating of a mixture consisted of AgNWs, PEDOT:PSS doped by dimethyl sulfoxide (DMSO) as well as Zonyl (a surfactant). Here, Zonyl serves as a wetting agent of PEDOT:PSS with which the mixed solution contained PEDOT:PSS could be deposited on hydrophobic substrates; it also improves the adhesive performance of AgNWs-PEDOT:PSS composite film. Besides, AgNWs were buried in the highly conductive polymer doped with DMSO, which not only eliminated the rough surface but also increased the electrical conductivity of AgNWs film without sacrificing its optical property. By adjusting the density of AgNWs solution and mass ratio between AgNWs and PEDOT:PSS, the composite film exhibits a 16 Ω/sq sheet resistance at 90.4% optical transmittance, which is even better than an ITO film. When remaining the content of AgNWs constant, the sheet resistance of AgNWs-PEDOT:PSS film is 66 times smaller than that of the pristine AgNWs film under the similar optical transmittance. In addition, when the composite films were treated with sulfuric acid (H_2SO_4), the adhesive performance was further improved without any damage to the film. The as-prepared flexible electrode is capable of withstanding cyclic bending tests for 2000 times and solvent environments with slightly decreased electrical conductivity. The favourable optoelectronic properties and adhesion for the simple, cost effective and solution-compatible approach makes our AgNWs-PEDOT:PSS films good candidates of ITO in the application of the next-generation flexible electronics.

Experimental

AgNWs were provided by Blue Nano as suspensions in isopropyl alcohol with a concentration of 10 mg/ml. The average diameter and length of the AgNWs were respectively 30 nm and 10 μm. PEDOT:PSS solution (Clevios PH 1000) was purchased from Heraeus. Zonyl FS-300 (Zonyl) was purchased from Dupont and DMSO was provided by Aladdin. Dynol 607 was provided by Air Products and Chemicals. Sulfuric acid (H_2SO_4, 95%) was purchased from Shanghai Lingfeng Chemical Reagent Co., Ltd., China. All reagents were used as received.

The AgNWs-PEDOT:PSS composite films were prepared by using a one-step rod coating. First, AgNWs, PEDOT:PSS, DMSO (5 wt% of PEDOT:PSS) as well as Zonyl (0.1 wt% of PEDOT:PSS) were mixed and shook to from a uniform solution. IPA was added to control the mass fraction of AgNWs. Films with different mass ratio of AgNWs and PEDOT:PSS were prepared to explore the optimized experimental conditions. Then, the mixed solution was coated on a polyethylene terephthalate (PET) substrate with a Mayer rod followed by drying in an oven at 120 °C for 30 s to evaporate the solvents completely. As comparison, AgNWs films were also prepared by depositing pristine AgNWs solution with the same coating method and IPA was also used to adjust the concentration of AgNWs solution. To further enhance the adhesive attraction between conductive layers and substrates, the composite films were treated with H_2SO_4 solution. In this process, the composite films were immersed in 20 wt%, 40 wt%, 60 wt% and 80 wt% H_2SO_4 for 10 min followed by washed by deionized water for several times. And then, the films were dried in an oven at 120 °C for 15 min.

The surface morphology of the films was analysed by a field emission scanning electron microscopy (FE-SEM, Hitachi S-4800), operating at 15 kV. The sheet resistances were examined using a four-point probes (model RTS-8, Guangzhou 4Probes Tech Industrial Co., Ltd., Guangzhou, China) and the final values were obtained by averaging 20 positions on one sample. Optical transmission measurements were conducted on a UV/Vis/NIR Spectrophotometer from 400nm to 900nm and the transmittance at 550nm were reported. X-ray photoelectron spectroscopy (XPS) was conducted in a Kratos Axis Ultra DLD X-ray photoelectron spectrometer using an Al K α X-ray source and the C(1s) peak was set to 285 eV as an reference. A 3M scotch (19mm*30mm) was used to test the adhesive performance of the transparent electrodes by peeling the scotch from the films and the residue on substrates indicated the adhesion strength.

Results and discussion

The SEM images of pristine AgNWs film and AgNWs-PEDOT:PSS film are shown in Fig. 1. It can be seen from Fig. 1c and e, the pristine AgNWs film presents an exceedingly coarse surface where each nanowire is gently close to the other one and part of them are even out of contact of the substrate, which leads to poor electrical conductivity and weak adhesion of AgNWs film. On the contrary, for the composite film, AgNWs are buried in the conducting polymer exhibiting a comparatively smooth appearance (Fig. 1d and f). Besides, with the help of PEDOT:PSS, the whole conductive shell coated on the substrate totally with the largest contact area. Here, PEDOT:PSS acting as a soldering material reinforced the connections between

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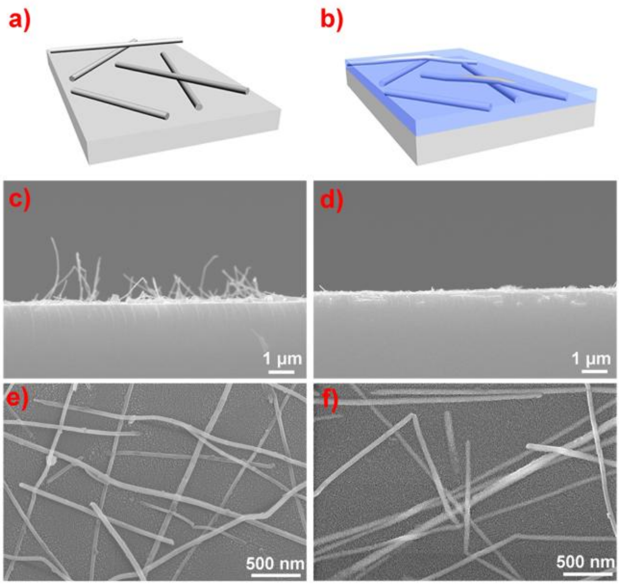

Fig. 1 Schematic diagrams of (a) pristine AgNWs film and (b) AgNWs-PEDOT:PSS film. Cross-section FE-SEM images of (c) pristine AgNWs film and (d) AgNWs-PEDOT:PSS composite film. Top view FE-SEM images of (e) pristine AgNWs film and (f) AgNWs-PEDOT:PSS composite film.

separate AgNWs and thus the electronic transmissions got enhanced. Pristine AgNWs films usually possess high sheet resistance and the lower the concentration of AgNWs solution is, the higher the sheet resistance is (Fig. 2b). For instance, the sheet resistance of AgNWs films with 0.6 wt% solute is as high as 1057 Ω/sq while that with 0.9 wt% is 164 Ω/sq. However, due to the welding effect, the addition of PEDOT:PSS could significantly decrease the sheet resistance of the AgNWs film. To obtain the most optimized condition, AgNWs and PEDOT:PSS were blended with different mass ratios. As shown in Fig. 2a, keeping the total solute mass fraction constant, the composite exhibits the lowest sheet resistance when the mass ratio of AgNWs and PEDOT:PSS is 2:1. Under this proportion, the sheet resistance of AgNWs-PEDOT:PSS film as a function of the content of solute was also investigated (Fig. 2b). When the solute mass ratio is 0.9 wt%, AgNWs-PEDOT:PSS film shows a lowest sheet resistance of 16 Ω/sq, 10 times smaller than pristine AgNWs film. For another, when remaining the AgNWs mass ratio of 0.6 wt%, the addition of PEDOT:PSS could result in a 66-fold decrease of sheet resistance.

Though the introduction of PEDOT:PSS has greatly enhanced the electrical property of pristine AgNWs film, the optical property was not affected. The optical transmittances of pristine AgNWs films with 0.6 wt% and 0.9 wt% AgNWs as well as the composite film (containing 0.6 wt% AgNWs and 0.3 wt% PEDOT:PSS) were measured at 550 nm wavelength. Pristine AgNWs with 0.6 wt% solute shows a transmittance of 90.8% while that of 0.9 wt% AgNWs film is 89.5% caused by increased AgNWs content. And the transmittance of

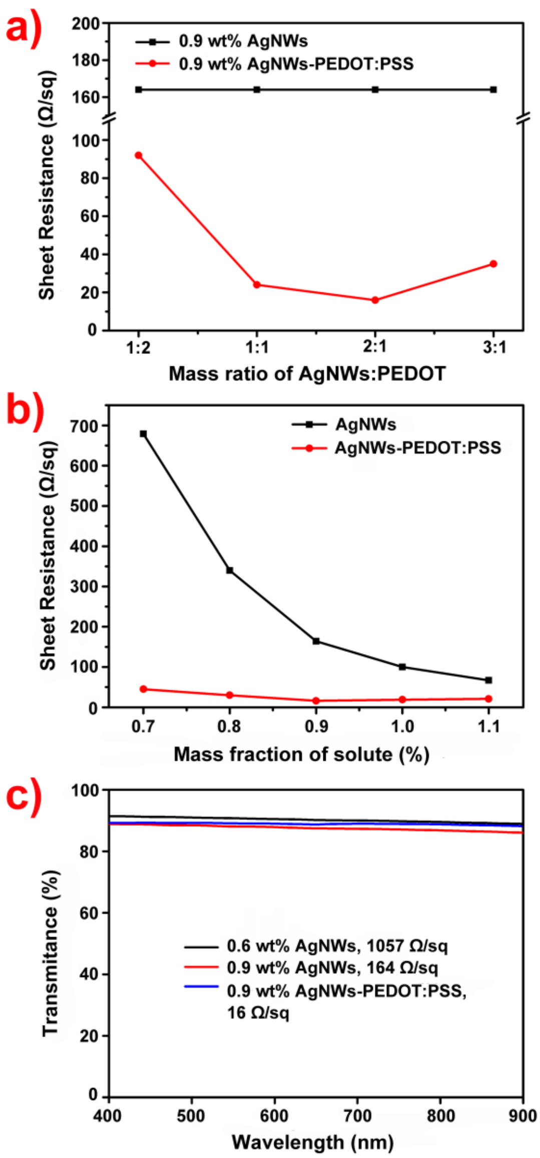

Fig. 2. (a) The sheet resistance functions as the mass ratio of AgNWs:PEDOT. (b) The sheet resistance of pristine AgNWs film and AgNWs-PEDOT:PSS composite film function as the mass fraction of AgNWs. c) Optical transmittance spectra of pristine AgNWs films with 0.6 wt% (black) and 0.9 wt% (red) solute as well as AgNWs-PEDOT:PSS composite film (blue) with 0.9 wt% solute.

AgNWs-PEDOT:PSS composite film is 90.4% which is comparable with 0.6 wt% AgNWs film indicating that the addition of PEDOT:PSS didn't reduce the transmittance of AgNWs film. What's more, with the same solute of 0.9 wt%, the transmittance of the composite film is even a little higher than a pristine AgNWs film. This is because pristine PEDOT:PSS film with 0.9 wt% solute shows a higher transmittance of 96.4% compared to 0.9 wt% AgNWs film (as shown in (Fig. S1, ESI†), and hence when part of AgNWs is replaced by PEDOT:PSS, the transmittance of AgNWs-PEDOT:PSS composite film is higher than pristine AgNWs film with the same solute. Overall, the introduction of PEDOT:PSS not only largely

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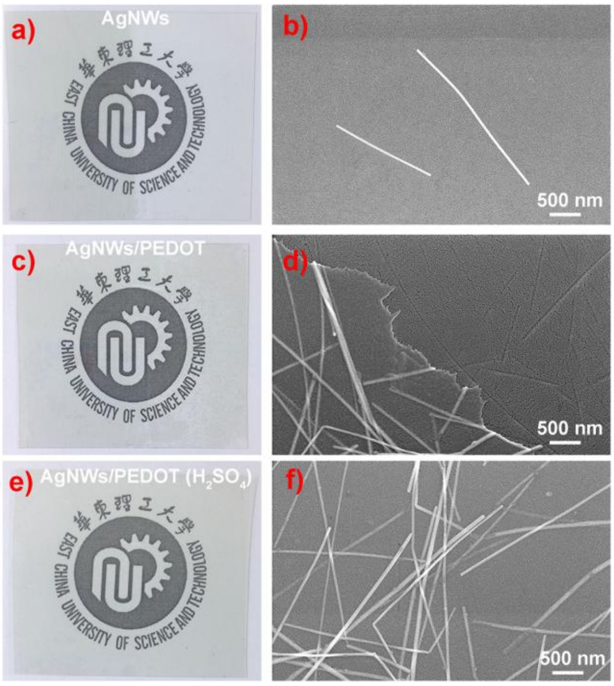

Fig. 3. Photographs before and after tape test and top view FE-SEM images after tape test of (a,b) AgNWs film, (c,d) AgNWs-PEDOT:PSS film and (e,f) AgNWs-PEDOT:PSS film (with H_2SO_4 post-treatment).

decreased the sheet resistance of AgNWs film but also maintained the high optical property resulting a high-performance with 16 Ω/sq at 90.4% optical transmittance.

Besides optical transmittance and sheet resistance, the adhesion between the film and substrate is another crucial part for high-performance TCEs, which shows great significance in their commercial applications. Pristine AgNWs films usually exhibit poor adhesive performance and they could be peeled off from PET substrate easily as shown in Fig. 3a and b. However, the addition of PEDOT:PSS (with 0.1 wt% Zonyl) is capable of improving the adhesion performance of the film, with which the film is partly peeled off and Fig. 3d shows the

Table 1. The adhesive performance of pristine AgNWs film, PEDOT:PSS film as well as AgNWs-PEDOT:PSS composite film on PET before and after H_2SO_4 treatment.

TCEs	Before	After
AgNWs	Yes	Yes
AgNWs (Zonyl)	Yes	Yes
AgNWs (607)	Yes	Yes
PEDOT:PSS (Zonyl)	Yes	No
PEDOT:PSS (607)	Yes	/
AgNWs-PEDOT:PSS (Zonyl)	Part	No
AgNWs-PEDOT:PSS (607)	Yes	No

(“Yes” and “No” repectively represent the film could be or not be peeled off after a tape test. “Part” represents the film can be partly peeled off. “/” means the film falled off during the process of washing with deionized water.)

boundary of removed and remained parts. To testify the role of Zonyl in improving the adhesion performance, another nonionic surfactant Dynol 607 was used to replace Zonyl, which was then added into pristine AgNWs film, PEDOT:PSS film as well as AgNWs-PEDOT:PSS film to investigate the effect of surfactant. As shown in Table 1, the addition of every surfactant has no effect on the adhesion of AgNWs. Though both of the two surfactants could make PEDOT:PSS wettable on hydrophobic substrate, the AgNWs-PEDOT:PSS film with Zonyl shows stronger effect on improving the adhesion between PEDOT:PSS and substrate resulting in strengthen interaction between AgNWs-PEDOT:PSS film and PET.

To further strengthen the interaction of the film and substrate, sulfuric acid (H_2SO_4) solution was used to post-process it. After immersing in H_2SO_4 solution for 10min, the adhesive performance of AgNWs-PEDOT:PSS film was further improved, which remained stable after the peel-off test (Fig. 3e and f). H_2SO_4 solutions with different mass fractions were used to post-process the film and it has demonstrated that the adhesive performance was all enhanced. Meanwhile, the sheet resistance and the transmittance of the film weren’t affected despite of the concentration of the H_2SO_4 solution. Even treated with 100 wt% H_2SO_4, the AgNWs-PEDOT:PSS composite film still shows the same transmittance as the untreated one (as shown in Fig. S2, ESI†). Actually, H_2SO_4 has been applied to increase the electrical conductivity of pure PEDOT:PSS film previously, which is caused by partial removal of insulated PSS and

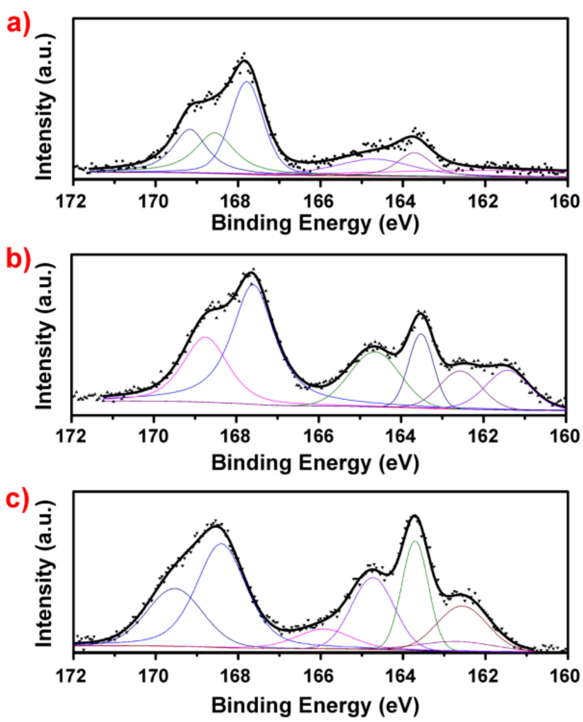

Fig. 4. S(2p) XPS spectra of AgNWs-PEDOT:PSS composite film (a) without treated by H_2SO_4, (b) treated by 20 wt% H_2SO_4 and (c) 100 wt% H_2SO_4.

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structural rearrangement of PEDOT.[16-17] Also, H_2SO_4 has been reported to chemically control the adhesion of PEDOT:PSS in order to realize its transfer from donor to soft target due to the strong van der waals (VDW) interaction between H_2SO_4-treated PEDOT:PSS and organic substrates.[18] Here H_2SO_4 is used to enhance the interaction between AgNWs-PEDOT:PSS conductive layer and PET substrate. As shown in Table 2, the post-process of H_2SO_4 shows no effect on pristine AgNWs film while it could significantly improve the adhesion property of pristine PEDOT:PSS film, which is presumably caused by the changed composition of PEDOT:PSS. X-ray photoelectron spectroscopy (XPS) was applied to analyse the compositional change of original AgNWs- PEDOT:PSS composite film and the films after treated with 20 wt%, 100 wt% H_2SO_4. It has been know that two bands of S(2p) in 171-167 eV and 167-163 eV are respectively the S atoms from sulfon acid group of PSS and thiophene ring of PEDOT. [18, 37] As shown in Fig. 4, after treated with H_2SO_4, the indensity ratio of PSS to PEDOT significantly descended and the more concentrated H_2SO_4 solution corresponds to lower ratio, which means that PSS chains are partly removed by H_2SO_4. Hence more PEDOT chains are exposed forming a dense network due to the π - π stacking effects and the intimate contact between H_2SO_4-treated PEDOT:PSS and soft substrate PET is enhanced resulting in stronger VDW interaction.[17,18] Besides, interdiffusion of polymer chains probably takes place on the interfaces, which may accompany established valence bond and makes a contribution to the improved interface interaction.[36] However, the weak VDW interaction between H_2SO_4-treated PEDOT:PSS and inorganic substrate makes PEDOT:PSS layer easy to be peeled off such as glass. Overall, both of Zonyl and H_2SO_4 enhanced the interaction of PEDOT:PSS and PET substrate and the improved adhesion of PEDOT:PSS leads to a stable AgNWs-PEDOT:PSS composite film.

Gnenerally, the performance of transparent conductive films is characterized by the ratio of DC conductivity to optical conductivity (σ_{DC}/σ_{Op}) which is expressed by the following equation

$$T(\lambda)=\left(1+\frac{188.5}{R_s}\frac{\sigma_{Op}(\lambda)}{\sigma_{DC}}\right)^{-2}$$

Where σ_{DC} is the DC conductivity, $\sigma_{Op}(\lambda)$ is the optical conductivity, T is optical transmittance (at 550 nm wavelength), and R_s is the sheet resistance. Since it was first reported by Hu et al, the value of σ_{DC}/σ_{Op} has been widely used to estimate the performance of various transparent electrodes.[38] The higher the value is, the greater performance of the electrodes exhibit, which usually corresponds to higher transmittance and lower resistance. The values of σ_{DC}/σ_{Op} of several AgNWs-based TCEs and their corresponding T and R_s are listed in Table 2. The σ_{DC}/σ_{Op} of our film is as high as 228 at 90.4% transmittance, which is higher than all previous AgNWs-PEDOT composite films.[30-32] Though AgNWs/clay composite film shows the highest σ_{DC}/σ_{Op}, the adhesive performance of AgNWs/clay was not investigated.[21] Besides, our film also possesses better optoelectronic property than AgNWs film with improved adhesion by biocompatible alginate gel.[35] The excellent optical and electrical property of our AgNWs-PEDOT:PSS film makes it a potential candidate in the applications of high-performance TCEs.

Table 2. Sheet resistance, transmittance and σDC /σOp of AgNWs-based TCEs.

AgNWs-based TCEs	R_s (Ω/sq) and T	σ_{DC}/σ_{Op}	Peel-off test
AgNWs transferred onto PEDOT:PSS[29]	17, 83%	114	No
Rod coated AgNWs/PEDOT:PSS[30]	25, 85%	89	Yes
Spray coated AgNWs/PEDOT:PSS[31]	10.8, 84.3%	197	No
AgNWs/clay[21]	10, 90%	349	No
GO-soldered AgNWs[34]	14, 88%	204	No
Ag nanoparticle/AgNWs[22]	14.9, 89.4%	220	No
AgNWs/CaAlg[35]	30.6, 92.5%	155	Yes
One-step coated AgNWs/PEDOT:PSS (this work)	16, 90.4%	228	Yes

("R" and "T" respectively represent sheet resistance and optical transmittance at 550 nm wavelength. "Yes" and "No" repectively represent whether the adhesive performance was tested or not.)

Limited by the rigidity of ITO, traditional transparent electrodes are fragile and their sheet resistance will significantly increase once undergoing deformation. However, the intrinsic flexiblility of AgNWs and conducting polymer imparts the as-prepared electrodes capability of deformation. Meanwhile, to realize the worthwhile application of the flexible transparent electrodes in optoelectronic devices, the long-term electromechanical stability is one of the necessary characterizations. The AgNWs-PEDOT:PSS composite film was bent with a bending radium of 3 mm and repeated for 2000 times. Here, a compressed and a tensile bending were regarded as one bending cycle and the sheet resistance was recorded each 100 times. As shown in Fig. 5a, after 2000 times of bending cycles, the sheet resistance of our composite film only slightly increased and reached 20 Ω/sq, which indicated that the AgNWs-PEDOT:PSS composite film possesses excellent endurance. Otherwise, the chemical stability is another crucial property for the practical applications of transparent electrodes, which allows them solution-

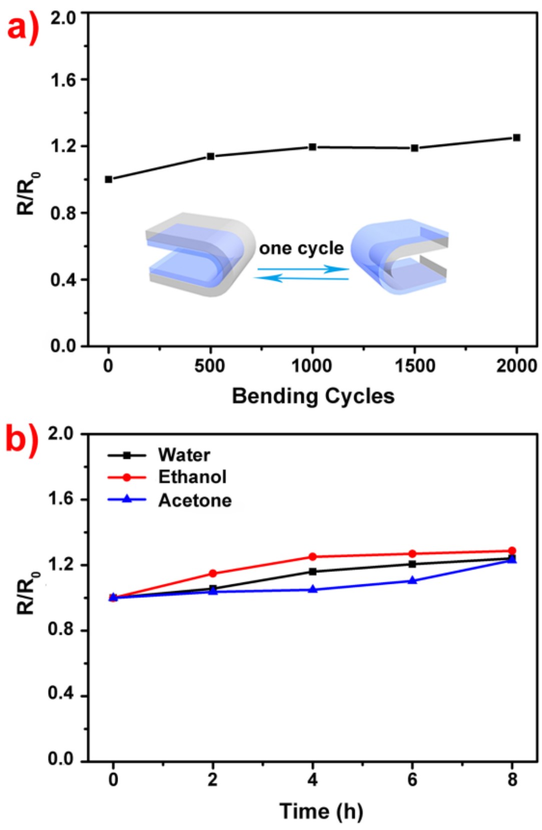

Fig. 5. (a) Sheet resistance of AgNWs-PEDOT:PSS composite film as a function of bending cycles. (b) Sheet resistance of AgNWs-PEDOT:PSS composite film as a function of the immersing time in water, ethanol as well as actone.

based preparation and keeps them from chemical damage. The AgNWs-PEDOT:PSS composite films were respectively immersed in three solvents: water, ethanol as well as acetone for 8h. As shown in Fig. 5b, the variations of the sheet resistance are all less than 30% conforming to the reported standard.[20] Above all, the enhanced adhesion resulted in both mechanical and chemical stability demonstrating the high-performance of our film though with a relative simple process.

Conclusions

In summary, high-performance AgNWs-PEDOT:PSS composite TCEs are prepared by a one-step rod-coating method. AgNWs are buried in the conducting polymer resulting in largely reduced roughness and sheet resistance without lowering optical transmittance. Consequently, flexible electrodes with sheet resistance of 16 Ω/sq at 90.4% optical transmittance was obtained and the σ_{DC}/σ_{Op} is as high as 228. The addition of Zonyl imparts PEDOT:PSS wettability on hydrophobic substrate, which, therefore, improved the adhesion of AgNWs-PEDOT:PSS composite film. Furthermore, the adhesive performance was enhanced after H_2SO_4 post-treatment by increasing the interaction between the conductive layer and the substrate. The as-prepared highly stable film performs well in cyclic deformation and long-time chemical environments. Briefly, the simple preparation and fascinating performance endow the AgNWs-PEDOT:PSS electrodes great potential in the application of future flexible optoelectronics.

Acknowledgements

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References

1 J. H. Du, S. F. Pei, L. P. Ma, H. M. Cheng. Adv. Mater., 2014, 26, 1958–1991.
2 S. R. Ye, A. R. Rathmell, Z. F. Chen, I. E. Stewart, B. J. Wiley. Adv. Mater., 2014, 26, 6670-6687.
3 M. Song, D. S. You, K. Lim, S. Park, S. Jung, C. S. Kim, D.-H. Kim, D.-G. Kim, J.-K. Kim, J. Paek, Y.-C. Kang, J. Heo, S.-H. Jin, J. H. Park, J.-W. Kang. Adv. Funct. Mater., 2013, 23, 4177-4184.
4 T. Y. Kim, Y. W. Kim, H. S. Lee, H. Kim, W. S. Yang, K. S. Suh. Adv. Mater., 2013, 23, 1250-1255.
5 D. R. Caims, R. P. Witte, D. K. Sparacin, S. M. Sachsman, D. C. Paine, G. P. Crawford, R. R. Newton. Appl. Phys. Lett. 2000, 76, 1425-1427.
6 C. Feng, K. Liu, J. S. Wu, L. Liu, J. S. Cheng, Y. Y. Zhang, Y. H. Sun, Q. Q. Li, S. S. Fan, K. L. Jiang. Adv. Funct. Mater., 2010, 20, 885-891.
7 F. Mirri, A. W. K. Ma, T. T. Hsu, N. Behabtu, S. L. Eichman, C. C. Young, D. E. Tsentalovich, M. Pasquali. ACS Nano, 2012, 6, 9737-9744.
8 N. Fukaya, D. Y. Kim, S. Kishimoto, S. Noda, Y. Ohno. ACS Nano, 2014, 8, 3285-3293.
9 H. X. Chang, G. F. Wang, A. Yang, X. M. Tao, X. Q. Liu, Y. D. Shen, Z. J. Zheng, Adv. Funct. Mater., 2010, 20, 2893-2902.
10 J. Kang, H. Kim, K. S. Kim, S.-K. Lee, S. Bae, J.-H. Ahn, Y.-J. Kim, J.-B. Choi, B. H. Hong. Nano Lett., 2011, 11, 5154-5158.
11 Y. Zhu, Z. Z. Sun, Z. Yan, Z. Jin, J. M. Tour. ACS Nano, 2011, 5, 6472-6479.
12 J. Wang, M. H. Liang, Y. Fang, T. F. Qiu, J. Zhang, L. J. Zhi. Adv. Mater., 2012, 24, 2874-2878.
13 L. G. D. Acro, Y. Zhang, C. W. Schlenker, K. Ryu, M. E. Thompson, C. W. Zhou. ACS Nano, 2010, 4, 2865-2873.
14 M. Vosgueritchian, D. J. Lipomi, Z. N. Bao. Adv. Funct. Mater., 2012, 22, 421-428.
15 J. Y. Oh, M. Shin, J. B. Lee, J.-H. Ahn, H. K. Baik, U. Jeong, ACS Appl. Mater. Interfaces, 2014, 6, 6954-6961.
16 Y. J. Xia, K. Sun, J. Y. Ouyang. Adv. Mater., 2012, 24, 2436-2440.
17 N. Kim, S. Kee, S. H. Lee, B. H. Lee, Y. H. Kahng, Y.-R. Jo, B.-J. Kim, K. Lee. Adv. Mater., 2014, 26, 2268-2272.
18 N. Kim, H. Kang, J.-K. Lee, S. Kee, S. H. Lee, K. Lee. Adv. Mater., 2015, 27, 2317-2323.
19 S. De, T. M. Higgins, P. E. Lyons, E. M. Doherty, P. N. Nirmalraj, W. J. Blau, J. J. Boland, J. N. Coleman. ACS Nano, 2009, 3, 1767-1774.

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20 L. B. Hu, H. S. Kim, J.-Y. Lee, P. Peumans, Y. Cui. ACS Nano, 2010, 4, 2955-2963.
21 T. Kim, Y. W. Kim, H. S. Lee, H. Kim, W. S. Yang, K. S. Suh. Adv. Funct. Mater., 2013, 23, 1250-1255.
22 H. F. Lu, D. Zhang, X. G. Ren, J. Liu, W. C. H. Choy. ACS Nano, 2014, 8, 10980-10987.
23 J.-Y. Lee, S. T. Connor, Y. Cui, P. Peumans. Nano Lett., 2008, 8, 689-692.
24 T. Tokuno, M. Nogi, M. Karakawa, J. T. Jiu, T. T. Nge, Y. Aso, K. Suganuma. Nano Res., 2011, 4, 1215-1222.
25 J. T. Jiu, M. Nogi, T. Sugahara, T. Tokuno, T. Araki, N. Komoda, K. Suganuma, H. Uchida, K. Shinozaki. J. Mater. Chem., 2012, 22, 23561-23567.
26 E. C. Garnett, W. Cai, J. J. Cha, F. Mahmood, S. T. Connor, M. Greyson Christoforo, Y. Cui, M. D. McGehee, M. L. Brongersma. Nat. Mater., 2012, 11, 241-249.
27 M.-S. Lee, K. Lee, S.-Y. Kim, H. Lee, J. Park, K.-H. Choi, H.-K. Kim, D.-G. Kim, D.-Y. Lee, S. Nam, J.-U. Park. Nano Lett., 2013, 13, 2814-2821.
28 P. Lee, J. Ham, J. Lee, S. Hong, S. Han, Y. D. Suh, S. E. Lee, J. Yeo, S. S. Lee, D. Lee, S. H. Ko. Adv. Funct. Mater., 2014, 24, 5671-5678.
29 W. Gaynor, G. F. Burkhard, M. D. McGehee, P. Peumans. Adv. Mater., 2011, 23, 2905-2910.
30 J. Lee, P. Lee, H. B. Lee, S. Hong, I. Lee, J. Yeo, S. S. Lee, T.-S. Kim, D. Lee, S. H. Ko. Adv. Funct. Mater., 2013, 23, 4171-4176.
31 D. Y. Choi, H. W. Kang, H. J. Sung, S. S. Kim. Nanoscale, 2013, 5, 977-983.
32 T. Akter, W. S. Kim. ACS Appl. Mater. Interfaces, 2012, 4, 1855−1859.
33 Y. Li, P. Cui, L. Y. Wang, H. Lee, K. Lee, H. Lee. ACS Appl. Mater. Interfaces, 2013, 5, 9155−9160.
34 J. J. Liang, L. Li, K. Tong, Z. Ren, W. Hu, X. F. Niu, Y. S. Chen, Q. B. Pei. ACS Nano, 2014, 8, 1590-1600.
35 Y. X. Jin, L. Li, Y. R. Cheng, L. Q. Kong, Q. B. Pei, F. Xiao. Adv. Mater., 2015, 25, 1581-1587.
36 B. Bhushan, J. Vac. Sci. Technol. B, 2003, 21, 2262-2296.
37 G. Greczynski, T. Kugler, W. R. Salaneck. Thin solid films, 1999, 354, 129-135.
38 L. Hu, D. S. Hecht, G. Grüner, Nano Lett., 2004, 4, 2513-2517.