Carbon Nanotubes with Carbon Blacks as Cofillers to Improve Conductivity and Stability

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Supporting Information

ABSTRACT: In this study, a simple solution-mixing method is used to develop a kind of excellent flexible, electrically conductive adhesives (ECAs). Carbon nanotubes (CNTs) and carbon blacks (CBs) as cofillers were added into Ag-based pastes. The use of the two fillers is due to the consideration that these two materials may provide positive synergetic effects for improving the conductivity of ECAs. The conductivity, flexibility, cyclability, and oxidation resistance of ECAs with different contents of carbon fillers were studied. It was found that a small amount of CNTs or CBs can dramatically improve the ECAs’ conductivity. Solution-mixing method brings excellent carbon nanofiller dispersion in polymer matrix. Highly dispersed CNTs and CBs among the Ag flakes formed three-dimensional conducting networks to improve the conductivity of ECAs. The conductivity of ternary hybrid ECAs (with addition of 3 wt % CNTs and 2 wt % CBs) with a low content of 55 wt % Ag flakes is higher than that of the ECAs filled with only the Ag content over 65 wt %. Meanwhile, by selecting thermoplastic polyurethane resin as the matrix, the ECAs exhibited excellent mechanical compliance. The resistivity did not change when the ECAs were bended at a 60% flexural strain or pressed under 1200 kPa. Additionally, the adhesion strength of the new composited ECAs is better than that of a commercial ECA (Abletherm 3188). Further, no obvious conductivity change was observed when the sample was stored in ambient air condition at 80 °C and 60% relative humidity (60%) for 15 days.

INTRODUCTION

Ever evolving advances in electrically conductive adhesives (ECAs) have encouraged many developments in microelectronics.1 ECAs were regarded as one kind of most promising substitutions to replace traditional joining materials such as Sn–Pb solders in applications for printed circuits, coupled resonators, thin-film devices, and flexible electronics.2–5 Compared with traditional Sn–Pb solders, ECAs have many advantages such as more environmental friendly, easy processing, lower processing temperatures, and allowing higher resolution for fine-pitch interconnection capability.6–8 Although ECAs have many advantages, there are still many challenges in achieving the replacement of traditional solders and fulfillment of rigid requirements for interconnecting materials. The use of significant amount of metal fillers for the current ECAs limits their widespread applications in the field of flexible electronics.9–11 It is highly advantageous and challenging to reduce the cost of metal fillers while maintaining excellent electrical conductivity. Reducing high cost of metal fillers is closely associated with a few approaches including engineering filler surfaces,12,13 adding additional conductive agents,14–16 or sintering nano-
ductivity, more than one type of filler with different dimensions is necessary to prepare ECAs. These materials include zero-dimensional atomic particles (e.g., nanocarbon black (nano-CBs) and silica), 1D rod-like nanofillers (e.g., carbon nanotubes and silver nanowires (NWs)), and two-dimensional (2D) layered nanofillers (e.g., clay platelets, and graphene).

In fact, because of their differences in shapes and element components, each kind of nanomaterial has its own unique ability. Positive synergistic effects of these nanoparticles on improving the electrical and other properties of polymer matrix are expected.

Therefore, some researchers turned to prepare three-dimensional (3D) hierarchical conductive networks with the hybrid of two or three conductive agents of different shapes and sizes.

Tang et al. reported a simple method to build a 3D foam-like conductive network structure by using CNTs and graphene sheets as the 1D and 2D nanoconstituents and latex-polystyrene microspheres as cofillers. The introduction of a second conductive filler of graphene significantly changed the conductive network morphology of conductive polymer composites filled with CNTs. Luo et al. fabricated a kind of adhesives with CNTs and Ag flakes. Their results showed that CNTs used there as conductivity fillers could bridge the neighboring silver flakes to accelerate the electron transportation. Fu et al. also studied the ECAs based on the ternary hybrid of Ag microflakes, Ag nanospheres (Ag-NSs), and Ag nanowires (Ag-NWs) and demonstrated that a small amount of Ag-NSs or Ag-NWs could dramatically improve the conductivity of the ECAs filled with single Ag-microflakes (MFs). The Ag-NSs and Ag-NWs with appropriate contents have a synergistic effect in improving the conductivity of the ECAs.

In this research, we focused on combining the merits of the properties of CNTs and the extremely low cost of activated carbon black (CBs). The goal of this research is to achieve high conductivity of ECAs by adding a small amount of carbon nanomaterials with difference dimensions. We used activated CNTs and CBs as additional conductive nanofillers for electrically conductive adhesives. Generally, typical ECAs contain polymer matrices and conductive fillers. To reduce the use of metal contents, some of the studies have reported the use of various nanomaterials included a small amount of metal nanoparticles/nanowire, nanowires, or carbon nanomaterials, and even conductive nanopolymer\textsuperscript{34–36} as additional conductive agents. However, adopting CNTs and CBs as cofillers to achieve low cost and high conductivity due to their unique positive synergistic effects has still not been reported.

For the prevention of their agglomeration, the CNTs used herein were functionalized with carboxylic groups by an acid treatment process and were called active CNTs. Small amount of activated CNTs and CBs were dispersed in the thermoplastic polyurethane (TPU) as flexible conductive adhesives. These different nanostructured carbon materials were introduced to significantly change the conductive network morphologies of ECAs filled with Ag flakes. The electrical properties of ECAs with hierarchical structures were better than those of ECAs with CBs or CNTs alone. The enhanced properties can be attributed to the synergy between CBs, CNTs, and Ag flakes in forming a 3D conducting networks with high quality.

### RESULTS AND DISCUSSION

#### Preparation of ECAs

The detailed preparation of the ECAs is described in detail in the Experimental Section. Briefly, acid-treated CNTs (Figure S1) prepared according to known literature\textsuperscript{37} and carbon blacks were mixed homogeneously in 1,4-dioxane. Polyurethanes (Figure S2) particles (TPU resin) were added to the mixture. And then, Ag flakes were put into the mixture. The obtained mixture was dried under vacuum to form the ECAs. Detailed compositions for the ECAs are given in Table 1.

#### Characterizations of Acid-Treated CNTs

Fourier transform infrared (FT-IR) spectra of CNTs before and after acid treatment are presented in Figure 1a. The peaks appearing at around 3436, 1720, and 1213 cm\textsuperscript{−1} were assigned to −OH, COO, and C–C–O stretching present in carboxylic groups (−COOH), respectively.\textsuperscript{38} Meanwhile, the intensities of peaks around 3436 and 1720 cm\textsuperscript{−1} considerably increased after acid treatment. The acid-treated CNTs were further investigated by Raman spectra and transmission electron microscope (TEM) (Figure S3). A detailed description of the Raman spectrum characteristics is given in Figure S3. These characterizations demonstrated the successful oxidation of CNTs for the generation of acid-functionalized CNTs.

The pictures of the dispersion of pristine CNTs and acid-treated CNTs at a concentration of 25 mg mL\textsuperscript{−1} are shown in Figure 1b. The pristine CNTs could not be well-dispersed in 1,4-dioxane at a concentration of 25 mg mL\textsuperscript{−1} after 24 h of bath sonication. These pristine CNTs precipitated after sonication. However, the acid-treated CNTs were homogeneously dispersed in 1,4-dioxane after sonication under similar conditions as those for pristine CNTs, and the dispersion is stable even after standing for 24 h without visible agglomerations. In contrast, CBs can be dispersed easily in 1,4-dioxane by facile sonication method. After sonication, no visible particles were recognized in CBs 1,4-dioxane suspension (Figure S4).

#### Bulk Resistivity and Conduction Mechanism

The bulk resistivity of a series of ternary hybrids is shown in Figure 2a, and the obtained resistivity could be as low as 32.84 × 10\textsuperscript{−5} Ω cm at 55 wt % silver loading after curing at 180 °C and cooling to room temperature for measurement. The resistivity of TPU–ECAs (2 wt % CB, 3 wt % CNTs, and 55 wt % Ag flakes) is one-tenth that of TPU–ECAs with 55 wt % Ag flakes only, which is even less than that of TPU–ECAs filled with 65% Ag flakes only. These results showed significant contribution of the addition of CNTs and CBs into the traditional Ag flakes-based ECAs in improving their conductivity.

In the case of ternary hybrid adhesives, high electrical conductivity was obtained by simply adding carbon materials instead of using a lot of expensive Ag flakes. Two main key factors can contribute to the high conductivity of the ternary
hybrid adhesives. One factor is that more than one type of conductive fillers with different dimensions have positive synergistic effects on improving the conductivity of ternary hybrid adhesives. The introduction of second and third conductive fillers can significantly change the conductive network morphologies of the adhesives. Thus, carbon-fillers are used to construct 3D continuous conductive networks with high-quality interconnections in the polymer matrix. Figure 2b shows that electrical properties of the adhesives with such 3D hierarchical structures are much better than those of binary adhesive with CBs or CNTs alone. The enhanced properties are attributed to the synergy effect of CBs and CNTs in forming 3D conducting networks with appropriate CBs/CNTs ratios. In Figure 2b, ternary hybrid adhesives (55 wt % silver fillers containing PU–ECAs as a function of different carbon materials loading cured at 180 °C for 120 min. (c) Bulk resistivity of 65 wt % silver fillers ECAs and C-4 PU–ECA cured at different temperatures for 120 min.

Another factor is the choice of solution-mixing method for adhesive preparation. In conductive adhesives, charge carriers are transported along the interconnected conductive fillers embedded in the polymer matrix. Thus, the construction of highly dispersed conductive network plays a crucial role in the fabrication of conductive adhesives with superior electrical properties. Compared to the results obtained from melt blending, which only realizes the macroscopic dispersion of fillers due to high viscosities of the composites, solution-mixing method we used here can be considered to obtain better fillers dispersion in a diluted solution. This solution-mixing process comprises three main steps: (1) dispersing nanofillers in a suitable suspension solvent; (2) mixing the resulting suspension with the polymer; and (3) forming target adhesives after solvent evaporation. We dispersed carbon nanofillers in 1,4-dioxane for a suspension and mixed the resultant suspension with TPU pellets by mechanical stirring and high-power ultrasonication to promote filler dispersion. This leads to form 3D continuous interconnected fillers distribution in the polymer matrix.

It is important to note that ternary hybrid adhesives (TH-ECAs) exhibit high electrical conductivity even when prepared at low curing temperature. Figure 2c shows that ternary hybrid adhesives, which was prepared by curing at 120 °C, with 55 wt % Ag flakes (named as C-4, Table 1) had a bulk resistivity of $5.8 \times 10^{-5} \Omega \cdot \text{cm}$. For comparison, the bulk resistivity of PU–ECA with Ag flakes (65 wt %) only is about 8-fold of that of C-4 when prepared at the same curing temperature of 120 °C. It was found that even a trace amount of carbon materials could reduce the resistivity significantly. Although using higher curing temperature could also bring about lower electrical resistivity (from $4.7 \times 10^{-5}$ to $2.8 \times 10^{-5} \Omega \cdot \text{cm}$ for the 65 wt % Ag-based ECAs) when curing temperature was increased from 120 to 200 °C without the addition of carbon materials, higher curing temperature is harsh for temperature-sensitive devices with high cost for processing the adhesives. The scanning electron microscope (SEM) images gave the morphologies of the dispersion of silver flakes and hybrid carbon materials in C-4 TH-ECAs (Figure S5). The results showed that hybrid carbon materials have obvious aggregations in the TPU resin matrix. Carbon materials dispersed well to bridge the neighboring silver flakes and build 3D hierarchical structures for TH-ECAs.

Conductivity under Mechanical Deformation. Recently, flexible electronics have attracted scientists’ attention significantly. For application in flexible devices, the ECAs are required to keep their conductivity under mechanical deformation. Figure 3 shows the resistivity change of TH-
ECAs films under flexural forms of mechanical deformation. The bending deformation of a free-standing TH-ECAs film was tested using a custom-made stretching machine. The testing approach with a bending angle of a custom-made stretching machine is illustrated in Figure 3a. Figure 3b gives the TH-ECAs’ resistance change as a function of flexural strain. Only very small resistance changes (within 10%) were observed even after bending at a flexural strain up to 60%. Moreover, the normalized resistance change is less than 20% even after 500 cycles of bending (Figure 3c).

Besides the above properties, the resistivity of the TH-ECAs was also stable under pressure. There is only less than 15% resistivity change after the film was pressured under 1200 kPa (~12 atmosphere pressure) (Figure 4b), showing excellent electrical properties under deformation.

Conductivity upon Continuous Thermal Treatment. We performed 2 weeks aging experiments at 80 °C with relative humidity of 60% (60% rh) to test the time-dependent resistivity changes (Figure 5a). The results were obtained by averaging 12 measurements from different areas of each tested sample. For TH-ECAs annealed at 80 °C, the bulk resistivity maintains basic stability, showing good thermal stability of the ECAs. In contrast, the resistivity of conventional PU–ECAs with 55 wt % Ag content only while without the carbon fillers increased by 40%.

For understanding the possible reason for this performance, we studied the surface properties of the ECAs. The surface wettability is shown in Figure 5b. The contact angle between distilled water and TH-ECAs (C-4) substrate was 107.2° (Figure 5b). However, the contact angle between distilled water and the common TPU–ECAs substrate (with 55 wt % Ag) without the addition of fillers of CNTs and CBs was only 79.90° (Figure 5c). This change might be attributed to the addition of hydrophobic fillers. This comparison demonstrates that TH-ECAs have excellent moisture resistance, which might be favorable to increase the oxidation resistance to result in good thermal stability.

Adhesion Strength. The adhesion properties of ECAs on aluminum surface were investigated by a lap shear test elaborated in Figure 6a. The adhesion properties of TH-ECAs (C-4), pure TPU, and PU-based ECA without the addition of carbon materials were assessed. It was found that the adhesion strength of TH-ECAs (C-4) was 0.10 kg mm−2, which is lower than that of pure PU resin (0.16 kg mm−2, Figure 6b). This indicated that a high inorganic filler content will lead to a low adhesive property. However, this value is still higher than that of a commercially available PU-based ECA (Abletherm 3188 with 0.06 kg mm−2) without the carbon material. This might be because the obtained ternary Ag–TPU adhesive with a low Ag content of 55 wt % is superior to those with a much higher Ag-MF content of over 65 wt %.

Demonstration of Applications of C-4 ECAs. Light-emitting-diode (LED) chips were attached to TH-ECAs on a paper substrate as a demonstration of the new type adhesive application. The brightness of these LED chips was constant for not only a flat device (Figure 7a) but also on a device under high flexural state (Figure 7b), indicating excellent conductivity of the ECA circuits under mechanical deformation. This might provide a potential alternative for flexible and printed electronics.

Comparison of Conductivities of This Work and Few Recent Works. Table 2 gives a comparison of the conductivities of the above-studied ECAs and those given in some other works. It was found that our TH-ECAs (C-4), which contains only 55 wt % silver filler, has a very low electrical resistivity of 32.84 × 10−5 Ω cm. By comparing our results to those of other works reported in Table 2, we concluded that high conductivity of conductive adhesives with lower silver content can be achieved by our approach. These properties may be beneficial for the goal of increasing electrical conductivity while reducing the use of expensive metal fillers.

CONCLUSIONS

In this study, new TPU–ECAs have been prepared. These TPU–ECAs offered high electrical conductivity by adding activated CNTs and CBs as additional conductive agents. Highly dispersed CNTs and CBs provide positive synergistic effects to reduce the resistivity of the TPU–ECAs. The electrical resistivity reached as low as 32.84 × 10−5 Ω cm using only 55 wt % silver loading and remained quite stable at flexural strain or under high pressure (1200 kPa). By grafting a

![Figure 3](image-url)

![Figure 4](image-url)

![Figure 5](image-url)

![Figure 6](image-url)

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small amount of hydrophobic CNTs, good thermal stability was achieved. The adhesives also exhibited excellent adhesion due to the use of relatively small amount of silver contents. The combination of these advantages are expected to enable...
the TPU–ECAs to have a very bright future as flexible and printed electronics.

## EXPERIMENTAL SECTION

**Materials.** Nonfunctionalyzed multiwalled carbon nanotubes (CM-95) were obtained from Hanwha Nanotech (Beijing, China). Conductive carbon black (Printex XE2B) was purchased from Evonik Degussa GmbH (Germany). 1,4-Dioxane and thermoplastic polyurethane (TPU) were obtained from Adamas Reagent Co., Ltd (Shanghai, China). Ag flakes with average diameters of 8–10 mm were purchased from Bolong Silver Industry (Changsha, China).

**Preparation of Acid-Functionalized CNTs.** The CNTs were acid-treated with H$_2$SO$_4$ and HNO$_3$. CNTs, 0.2 g, were treated with 75 mL of H$_2$SO$_4$ and 25 mL of HNO$_3$ to generate functional groups on the surfaces of the CNTs. After 12 h sonication process in the acid, acid-functionalized CNTs (named O-CNT) were centrifuged and washed for neutralization.

**Preparation of ECAs.** First, acid-treated CNTs and carbon blacks were mixed homogeneously in 1,4-dioxane. Second, TPU particles (the structure of polyurethane is given in Figure S2) were added to the mixture. Then, Ag flakes were put into the mixture under stirring at 600 rpm for 3 h. The obtained mixture was dried under vacuum (about 20 Pa) for 24 h to remove the solvent. Detailed compositions for the ECAs are provided in Table 1. The TUP/carbon nanocomposite ECAs films were prepared via the solvent-evaporation method (Figure S1).

**Characterization and Measurements.** The morphologies of carbon fillers, Ag flakes, and ECAs were observed using a scanning electron microscope (SEM, Hitachi S-4300, Japan). Transmission electron microscope (TEM) was operated at 200 kV using a JEOL-2100F microscope (Tokyo, Japan). For the observation of CNTs only, the sample for TEM was prepared by dispersing carbon materials in 1,4-dioxane by ultrasonication.

The samples for volume resistivity measurements were prepared according to a four-point probe system (RTS-9, 4 PROBES TECH, Guangzhou, China). First, two parallel strips of 3 meters of tape were placed 2.54 mm apart along the length of a standard glass slide of 25.4 mm × 76.2 mm. Second, a small amount of ECA was added in the space between the 3 meter tape. Then, a scalpel was used manually to squeeze the ECAs into the space between the two 3 meter tapes. After the samples were heated at various temperatures for 2 h, the volume resistivity of the ECAs was measured by a four-point probe and the volume resistivity ($\rho$) was calculated from eq 1.

$$\rho = RA$$

where $R$ is the sheet resistivity of the sample and $A$ is the thickness of the sample.

The samples for adhesion test were prepared according to Liu’s previous work. Briefly, an aluminum sheet with a dimension of 100 mm × 25 mm × 2 mm was polished to prepare the adhesion test. Afterward, two rectangular aluminum alloy sheets with an overlap length of 10 mm were fixed with the as-prepared adhesives. Then, the samples were heated at 180 °C for 2 h. Measurement of the adhesion of the ECAs was conducted by an electronic tensile testing machine (Instron 2367, Norwood). The adhesion can be calculated from eq 2.

$$\tau = F/WL$$

where $W$ and $L$ are the length and the width of the bonding parts of the sample and $F$ is the force applied to break the bond.

The bending of the adhesive sample was carried out using a custom-made stretching machine, which is illustrated in the Supporting Information (Figure S6). The flexural strain can be calculated from eq 3.

$$\text{flexural strain(%) span length/sample length}$$

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.8b03684.

Preparation of silver–PU–ECAs adhesives, the structure of polyurethane used in this study, Raman spectra of raw and oxidized CNTs and TEM image of acid-treated CNTs, dispersion test of CBs in 1,4-dioxane, SEM image depicting 3D hierarchical structures of TH-ECAs, and a custom stretching machine for bending the adhesive samples (PDF)

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**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

Authors would like to thank the start-up fund of SUSTech (Y01256009) and the high-level university construction fund for SUSTech (G01256018).

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