

## Stable Aqueous Dispersion of Reduced Graphene Nanosheets via Non-Covalent Functionalization with Conducting Polymers and Application in Transparent Electrodes

Kiyoung Jo,<sup>†</sup> Taemin Lee,<sup>†</sup> Hyun Jung Choi, Ju Hyun Park, Dong Jun Lee, Dong Wook Lee, and Byeong-Su Kim\*

*Interdisciplinary School of Green Energy and School of NanoBioscience and Chemical Engineering, Ulsan National Institute of Science and Technology (UNIST), Ulsan 689-798, Korea.*

<sup>†</sup>These authors contributed equally to this work.

Received November 6, 2010. Revised Manuscript Received December 19, 2010

We developed a simple and facile method of producing a stable aqueous suspension of reduced graphene oxide (RGO) nanosheets through the chemical reduction of graphene oxide in the presence of a conducting polymer dispersant, poly(3,4-ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS). This approach involves the cooperative interactions of strong  $\pi$ - $\pi$  interactions between a two-dimensional graphene sheet and a rigid backbone of PEDOT and the intermolecular electrostatic repulsions between negatively charged PSS bound on the RGO sheets, which impart the colloidal stability of the resulting hybrid nanocomposite of RGO/PEDOT. Moreover, our one-step solution-based method allows preserving the intrinsic chemical and electronic properties of both components, yielding a hybrid film of RGO nanosheets of high conductivity of 2.3 k $\Omega$ /sq with a transmittance of 80%. By taking advantage of conducting network structure of conducting polymers which provides an additional flexibility and mechanical stability of RGO nanosheets, we demonstrate the potential application of hybrid RGO/PEDOT as highly flexible and transparent electrodes.

### Introduction

Graphene, a single layer of two-dimensional carbon lattice, has recently attracted enormous interest with its remarkable electrical, thermal, and mechanical properties.<sup>1-3</sup> While earlier fundamental researches have been initiated from the micromechanical cleavage of graphite for high-quality graphene sheets,<sup>4</sup> recent efforts are geared toward producing the graphene sheets in a controlled and scalable manner, which are prerequisites for its wide applications.<sup>5-7</sup> In particular, chemical exfoliation methods for preparing stable graphene sheets have recently been the most popular protocols in achieving stable suspensions in various solvents.<sup>8</sup> These chemical exfoliation methods are efficient and scalable; however, they inevitably introduce the surface defects, which hamper the conductivity of the resulting graphene sheets. To restore the structure and electrical conductivity of the resulting graphene sheets, a number of approaches have been reported to reduce graphene oxide, including chemical reactions with reducing agents such as hydrazine,<sup>9,10</sup> hydriodic acid,<sup>11</sup> or pyrolysis at

high temperatures.<sup>12</sup> However, the reduced graphene oxide (RGO) still retains the structural defects which deteriorate the conductivity and often experiences an irreversible agglomeration and precipitation without proper additives during the chemical reduction process; as such, polymers,<sup>9,13-15</sup> surfactants,<sup>16,17</sup> and small molecules<sup>18,19</sup> have been widely used as additives during the chemical reduction. As a seminal example, Ruoff and co-workers have demonstrated the successful formation of stable suspensions of RGO in the presence of strong polyelectrolyte poly(styrene sulfonate) (PSS).<sup>9</sup> However, it is still highly desirable to prepare stable suspensions of RGO with a conductive dispersant for enhanced chemical stability and electrical conductivity necessary for advanced optoelectronic applications.<sup>14,15</sup>

A derivative of conducting polymer polythiophene, poly(3,4-ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS), has a wide utility as an interfacial modification layer in organic optoelectronic devices with its high conductivity, stability, and aqueous solubility.<sup>20</sup> The rigid backbone of PEDOT is doped by the PSS moiety, while the remaining unassociated sulfonic acid

\*To whom correspondence should be addressed. E-mail: bskim19@unist.ac.kr.

- (1) Geim, A. K.; Novoselov, K. S. *Nat. Mater.* **2007**, *6*, 183.
- (2) Novoselov, K. S.; Geim, A. K.; Morozov, S. V.; Jiang, D.; Zhang, Y.; Dubonos, S. V.; Grigorieva, I. V.; Firsov, A. A. *Science* **2004**, *306*, 666.
- (3) Lee, C.; Wei, X. D.; Kysar, J. W.; Hone, J. *Science* **2008**, *321*, 385.
- (4) Novoselov, K. S.; Jiang, D.; Schedin, F.; Booth, T. J.; Khotkevich, V. V.; Morozov, S. V.; Geim, A. K. *Proc. Natl. Acad. Sci. U.S.A.* **2005**, *102*, 10451.
- (5) Eda, G.; Fanchini, G.; Chhowalla, M. *Nat. Nanotechnol.* **2008**, *3*, 270.
- (6) Park, S.; Ruoff, R. S. *Nat. Nanotechnol.* **2009**, *4*, 217.
- (7) Kim, K. S.; Zhao, Y.; Jang, H.; Lee, S. Y.; Kim, J. M.; Kim, K. S.; Ahn, J. H.; Kim, P.; Choi, J. Y.; Hong, B. H. *Nature* **2009**, *457*, 706.
- (8) Dreyer, D. R.; Park, S.; Bielawski, C. W.; Ruoff, R. S. *Chem. Soc. Rev.* **2010**, *39*, 228.
- (9) Stankovich, S.; Piner, R. D.; Chen, X. Q.; Wu, N. Q.; Nguyen, S. T.; Ruoff, R. S. *J. Mater. Chem.* **2006**, *16*, 155.
- (10) Li, D.; Muller, M. B.; Gilje, S.; Kaner, R. B.; Wallace, G. G. *Nat. Nanotechnol.* **2008**, *3*, 101.
- (11) Moon, I. K.; Lee, J.; Ruoff, R. S.; Lee, H. *Nat. Commun.* **2010**, *1*, 73.

- (12) Li, X. L.; Zhang, G. Y.; Bai, X. D.; Sun, X. M.; Wang, X. R.; Wang, E.; Dai, H. J. *Nat. Nanotechnol.* **2008**, *3*, 538.
- (13) Bai, H.; Xu, Y. X.; Zhao, L.; Li, C.; Shi, G. Q. *Chem. Commun.* **2009**, 1667.
- (14) Liu, Y. Q.; Gao, L.; Sun, J.; Wang, Y.; Zhang, J. *Nanotechnology* **2009**, *20*, 465605.
- (15) Xu, Y. F.; Wang, Y.; Liang, J. J.; Huang, Y.; Ma, Y. F.; Wan, X. J.; Chen, Y. S. *Nano Res.* **2009**, *2*, 343.
- (16) Park, S.; Mohanty, N.; Suk, J. W.; Nagaraja, A.; An, J. H.; Piner, R. D.; Cai, W. W.; Dreyer, D. R.; Berry, V.; Ruoff, R. S. *Adv. Mater.* **2010**, *22*, 1736.
- (17) Green, A. A.; Hersam, M. C. *Nano Lett.* **2009**, *9*, 4031.
- (18) Xu, Y. X.; Bai, H.; Lu, G. W.; Li, C.; Shi, G. Q. *J. Am. Chem. Soc.* **2008**, *130*, 5856.
- (19) Hernandez, Y.; Nicolosi, V.; Lotya, M.; Blighe, F. M.; Sun, Z. Y.; De, S.; McGovern, I. T.; Holland, B.; Byrne, M.; Gun'ko, Y. K.; Boland, J. J.; Niraj, P.; Duesberg, G.; Krishnamurthy, S.; Goodhue, R.; Hutchison, J.; Scardaci, V.; Ferrari, A. C.; Coleman, J. N. *Nat. Nanotechnol.* **2008**, *3*, 563.
- (20) Groenendaal, B. L.; Jonas, F.; Freitag, D.; Pielartzik, H.; Reynolds, J. R. *Adv. Mater.* **2000**, *12*, 481.

group in PSS provides optimum aqueous solubility of the PEDOT:PSS.

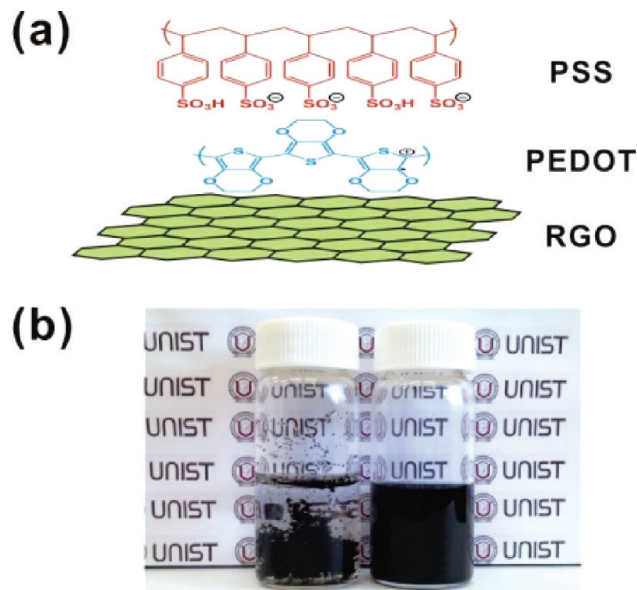
In this article, we present a simple and facile approach of preparing a stable aqueous dispersion of RGO nanosheets functionalized with conducting polymer, PEDOT:PSS (RGO/PEDOT hereafter). Our one-step solution-based method allows us to preserve the intrinsic chemical and electronic property of both components, yielding a hybrid film of RGO nanosheets of high conductivity of 2.3 k $\Omega$ /sq at a 80% transmittance (at 550 nm). We demonstrate that the composite RGO/PEDOT can be utilized as a potential transparent conducting electrode possessing high electrical conductivity with tunable transparency.

### Experimental Section

**Preparation of Graphene Oxide (GO) Suspension.** Graphite oxide was synthesized from graphite powder (Aldrich, < 20  $\mu$ m) by the modified Hummers method and exfoliated to give a brown dispersion of graphene oxide (GO) under ultrasonication.<sup>10</sup> Briefly, graphite powders (1.0 g), K<sub>2</sub>S<sub>2</sub>O<sub>8</sub> (0.50 g) and P<sub>2</sub>O<sub>5</sub> (0.50 g), were added to 3.0 mL of conc. H<sub>2</sub>SO<sub>4</sub> with stirring until the reactants are completely dissolved. The mixture is kept at 80 °C for 4.5 h, after which the heating is stopped and the mixture diluted with 1.0 L of Millipore water. The mixture is filtered and washed to remove all traces of acid. For the oxidation step of the synthesis, the pretreated graphite is added to the 26 mL of H<sub>2</sub>SO<sub>4</sub> and stirred. To this reaction mixture, 3.0 g of KMnO<sub>4</sub> was added slowly in an ice bath to ensure that the temperature remained below 10 °C. Then, this mixture reacts at 35 °C for 2 h after which 46 mL of distilled water is added under an ice bath. This mixture is stirred for 2 h at 35 °C, after which the heating is stopped and the mixture diluted with 140 mL of water and 2.5 mL of 30% H<sub>2</sub>O<sub>2</sub> is added to the mixture resulting in a yellow color along with bubbling. The mixture is allowed to settle for at least a day after which the clear supernatant is decanted. The remaining mixture is filtered and washed with a 1.0 L of 10% HCl solution. The resulting solid is dried in air and diluted in distilled water that is put through dialysis for 2 weeks to remove any remaining materials and residues, after which the product was centrifuged and washed several times with Millipore water to neutralization and remove residual species. Finally, the dark brown GO powders were obtained through drying at 50 °C in a vacuum oven for a day. The GO powder dissolved in a known volume of water is subjected to ultrasonication for 40 min to give a stable suspension of GO (typically conc. 0.50 mg/mL) and then centrifuged at 4000 rpm for 10 min to remove any aggregates remained in the suspension.

**Preparation of Reduced Graphene Oxide (RGO) with Hydrazine in the Presence of PEDOT:PSS.** The resulting GO suspension (20.0 mL, 0.50 mg/mL) was mixed with 7.70 mL of PEDOT:PSS solution (1:10 w/w ratio vs GO, total 1.3 wt % dispersion in water, 0.50 wt % of PEDOT, and 0.80 wt % of PSS, Aldrich). To this solution, 60.0  $\mu$ L of hydrazine solution (35 wt % in water, Aldrich) was slowly added, and the reaction mixture was heated to 95 °C for 3 h to afford a stable suspension of RGO nanosheets functionalized with PEDOT:PSS (RGO/PEDOT). After the reaction, the suspension was filtered with poly(ether sulfone) (PES) membrane filter (pore size of 0.22  $\mu$ m, Nalgene) and washed extensively with Millipore water to remove any residual hydrazine and unbound PEDOT:PSS polymer, yielding a thin film of RGO/PEDOT nanocomposite. After drying under ambient conditions, this hybrid thin film is redispersed with a known volume of water at a concentration of 0.40 mg/mL. The prepared suspension was spin-coated onto a silicon wafer to investigate the structure of the RGO/PEDOT using atomic force microscopy (AFM).

**Preparation of RGO/PEDOT Film.** A dilute suspension of RGO/PEDOT (conc. 0.40 mg/mL) was vacuum filtrated using a mixed cellulose ester membrane (pore size of 25 nm, Millipore) with an extensive rinsing with Millipore water (18 M $\Omega$ ·cm). The RGO/PEDOT film on the membrane filter was then transferred



**Figure 1.** (a) Schematic representation of reduced graphene oxide (RGO) nanosheet with PEDOT:PSS. (b) Images of aqueous dispersions of chemically reduced GO (left) without PEDOT:PSS and (right) with PEDOT:PSS suspension. Note the PEDOT:PSS polymers in (a) are actually surrounding both sides of the graphene sheets.

onto a desired substrate such as a flexible poly(ethylene terephthalate) (PET) or a quartz slide after wetting a drop of water on the surface of O<sub>2</sub>-plasma treated receiving substrate prior to transfer and pressed against the substrate with a 1 kg weight. After 12 h, the membrane was removed by a fresh acetone solution to leave RGO/PEDOT film on the substrate. For chemical doping with SOCl<sub>2</sub> solution, the RGO/PEDOT film on a quartz slide (2.5  $\times$  2.5 cm<sup>2</sup>) was subjected to the solution of SOCl<sub>2</sub> for 1 h. The RGO/PEDOT films were left under ambient conditions for several hours to remove any excess SOCl<sub>2</sub> before the measurement of transmittance and sheet resistance.

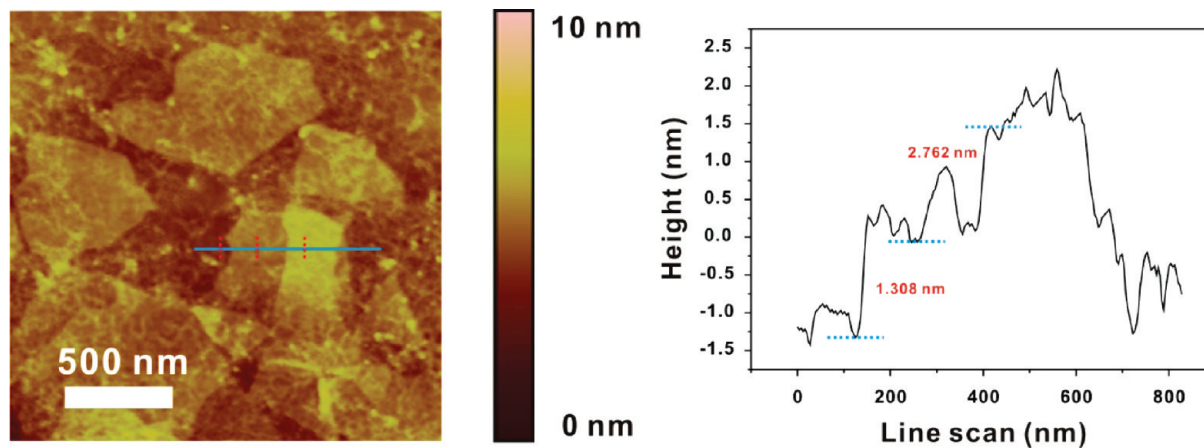
**Characterizations.** The  $\zeta$ -potential of colloidal suspensions was measured using a  $\zeta$ -potential analyzer (Malvern, Zetasizer nano zs). The transmittance of the hybrid films was characterized by using ultraviolet–visible (UV–vis) spectroscopy (VARIAN, Cary 5000). Thermogravimetric analysis was conducted in an N<sub>2</sub> atmosphere at a heating rate of 10 °C/min using a thermogravimetric analyzer (TA Hi-Res TGA 2950). Elemental analysis was conducted with Thermo Scientific Flash 2000. The thickness of the hybrid film on a silicon substrate was measured using ellipsometry (EC-400 and M-2000 V, J. A. Woollam Co., Inc.). The surface morphology of the samples were investigated using AFM (Nanoscope V, Veeco) via a tapping-mode and scanning electron microscopy (SEM, FEI, NOVA NANOSEM 230). Hybrid RGO/PEDOT was also characterized by X-ray photoelectron spectroscopy (XPS, Thermo Fisher, K-alpha). Raman spectra were obtained, and the D/G area ratio was fitted after baseline correction (WITEC, alpha-300M). The sheet resistance of the hybrid films was measured by using a four-point probe method (Advanced Instrument Technology, CMT-SR1000N).

### Results and Discussion

According to the modified Hummers method, graphite oxide (GO) suspension was initially prepared from a commercial graphite powder (see Experimental section).<sup>10,21,22</sup> Sonication of

(21) Hummers, W. S.; Offeman, R. E. *J. Am. Chem. Soc.* **1958**, *80*, 1339.

(22) Hong, T. K.; Lee, D. W.; Choi, H. J.; Shin, H. S.; Kim, B.-S. *ACS Nano* **2010**, *4*, 3861.



**Figure 2.** Height-mode AFM image of the RGO/PEDOT with a corresponding line scan. The image shows well-dispersed graphene sheets of monolayer and bilayer structures. All graphene sheets are covered with a polymeric PEDOT:PSS with an average thickness of  $1.28 \pm 0.11$  nm. The scale of the image is  $2 \times 2 \mu\text{m}^2$ .

the resulting graphite oxide powder and subsequent reduction of GO with hydrazine in the presence of PEDOT:PSS (1:10 w/w ratio vs GO) affords a stable suspension of RGO with a dark black color. This suspension was subjected to filtration through a PES membrane ( $0.22 \mu\text{m}$ ) with extensive rinsing to remove the unbound free polymers and chemical reductant, eventually yielding a thin nanocomposite film of RGO/PEDOT. This hybrid film can be readily redispersed at a concentration as high as  $1.0 \text{ mg/mL}$  in water. The dispersion was stable for several months without noticeable aggregates (more than 6 months).

We also found that there is a minimum concentration of PEDOT:PSS required to effectively disperse the RGO nanosheet (more than 1:5 w/w ratio vs GO), which supports that the sufficient surface coverage of RGO sheet by the conducting dispersants is crucial in the formation of stable RGO/PEDOT suspension. We postulate that the PEDOT:PSS can disperse RGO sheets via the interplay of two independent mechanisms, such that strong  $\pi$ - $\pi$  interactions between a graphene sheet and a rigid planar backbone of the conducting polymer PEDOT and the intermolecular electrostatic repulsions between negatively charged PSS bound on RGO sheets impart the colloidal stability of the resulting composite RGO/PEDOT ( $\zeta$ -potential of  $-57 \text{ mV}$ ). The control experiment of reducing GO with hydrazine in the absence of PEDOT:PSS typically yields an aggregate of graphene platelets due to irreversible aggregation with the loss of surface functional groups upon chemical reduction, as similarly observed in the other report (Figure 1b).<sup>9</sup>

As observed by AFM, the homogeneous suspension of RGO/PEDOT contained mostly single sheets of graphene covered with a thin granular structure of PEDOT:PSS colloid with a lateral sheet size ranging from  $0.5$  to  $1 \mu\text{m}$  (Figure 2). The surface profile displayed that the average height of the PEDOT:PSS covered graphene sheet is  $1.28 \pm 0.11 \text{ nm}$ , which is higher than the average thickness of uncoated pristine GO (ca.  $0.70 \text{ nm}$ ). This observation could indicate the polymer coating around the surface of RGO nanosheets.

Thermogravimetric analysis (TGA) was employed to provide the relative composition of polymer within the hybrid RGO/PEDOT. As shown in the TGA curve, an initial mass loss of 5% around  $100 \text{ }^\circ\text{C}$  is observed, which is attributed to the water contained in the composite film (see Supporting Information). Followed by the first thermal transition, the RGO/PEDOT composite exhibits additional mass loss (35% relative to starting materials) around  $300$ – $400 \text{ }^\circ\text{C}$  resulting from the decomposition

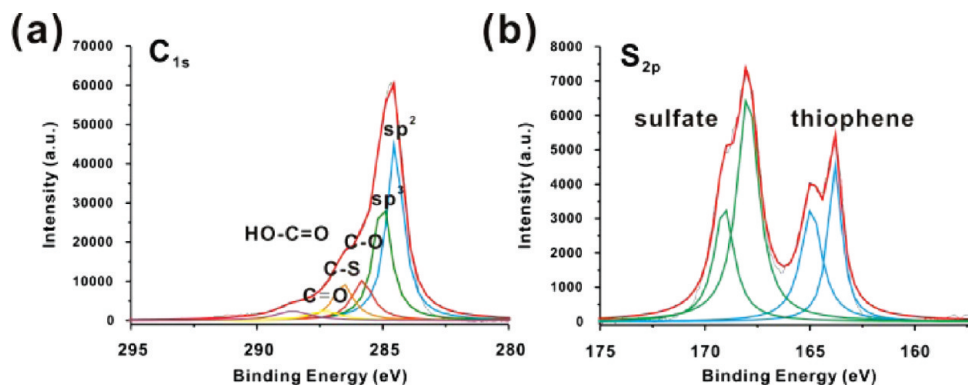
of PEDOT:PSS together with the surface functional groups on RGO nanosheets. The onset of the degradation temperature is higher than that of pure PEDOT:PSS, suggesting the enhanced thermal stability of RGO/PEDOT hybrid. However, it was difficult to estimate the relative composition of RGO within the composite, as the labile surface functional groups present on the RGO degrades concomitantly with PEDOT:PSS at this temperature range. Independent of the TGA result, the elemental analysis determined the elemental weight fraction of C, H, N, O, and S of RGO/PEDOT to be 44.0%, 4.5%, 5.4%, 18.3%, and 14.9%, respectively (average of three different batches). On the basis of the sulfur content, we can calculate that approximately 30% of RGO is present in the hybrid RGO/PEDOT composite.

According to the XPS measurement in Figure 3, there are six distinct types of carbon bonds (C1s) present in the RGO/PEDOT, including  $\text{sp}^2$  hybridized graphitic carbons ( $284.5 \text{ eV}$ ),  $\text{sp}^3$  hybridized saturated carbons ( $285.0 \text{ eV}$ ), C—O ( $285.8 \text{ eV}$ ), C—S ( $286.6 \text{ eV}$ ), C=O ( $287.3 \text{ eV}$ ), and carboxyl groups ( $288.6 \text{ eV}$ ), which are all in good agreement with previous works. The intensity of carboxyl groups decreases considerably upon chemical reduction, suggesting that the successful reduction of GO nanosheets has taken place (see Supporting Information). Moreover, the RGO/PEDOT suspension exhibits the characteristic peaks of spin-split doublet  $\text{S}_{2p}$  at  $169.1$ ,  $168.0 \text{ eV}$  and  $164.9$ ,  $163.8 \text{ eV}$ , corresponding to sulfonate from PSS and thiophene from PEDOT, respectively. Taken together, these XPS spectra confirm the presence of both components in the hybrid RGO/PEDOT.

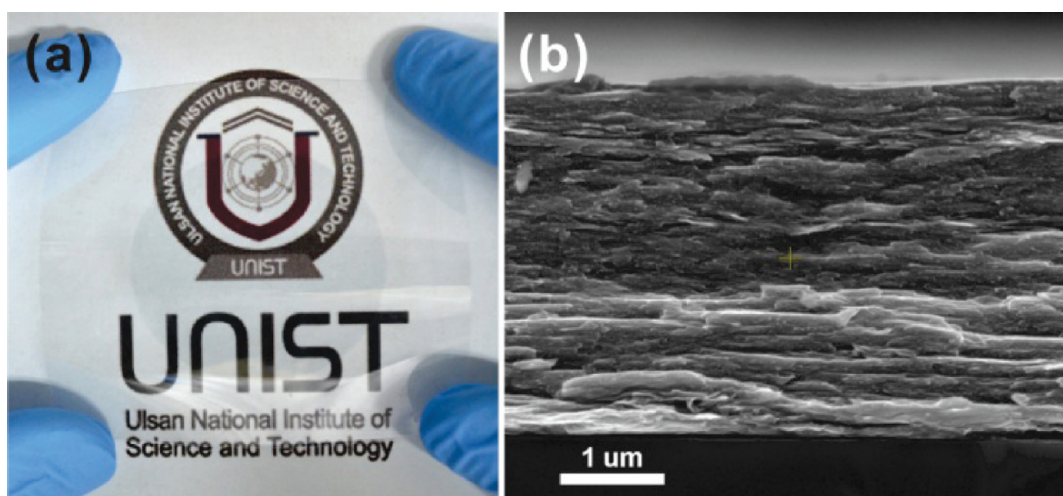
Furthermore, we have demonstrated that the filtration of aqueous suspension of RGO/PEDOT on a cellulose ester membrane yielded a flexible and free-standing film (Figure 4). As reported in other vacuum filtration assisted methods,<sup>5,12,18,23</sup> the film thickness can be easily tailored by the volume and the concentration of RGO/PEDOT suspension. Moreover, the RGO/PEDOT can be transferred onto a desired substrate such as a flexible PET or a quartz slide by dissolving away the membrane with acetone after the close contact between the graphene sheet and the substrate is established.<sup>5</sup> We found that the quality of transfer dramatically improved by addition of a drop of water on the surface of receiving substrate prior to transfer, to enhance the flexibility of the RGO/PEDOT film and the contact between the film and the substrate. Moreover, a slight pressure of a few bars

(23) Dikin, D. A.; Stankovich, S.; Zimney, E. J.; Piner, R. D.; Dommett, G. H. B.; Evmenenko, G.; Nguyen, S. T.; Ruoff, R. S. *Nature* **2007**, *448*, 457.

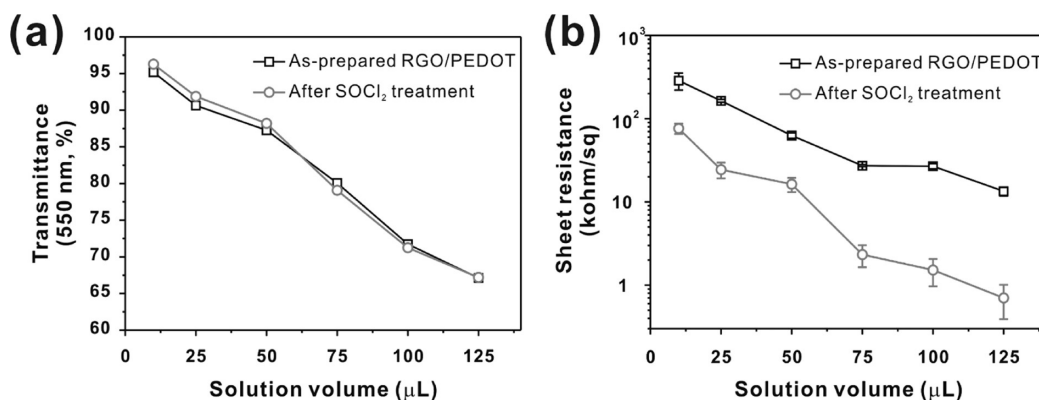




**Figure 3.** Deconvoluted high-resolution (a)  $C_{1s}$  and (b)  $S_{2p}$  XPS spectra of RGO/PEDOT.



**Figure 4.** (a) A photo of thin film of RGO/PEDOT deposited on a flexible PET substrate with a transmittance of 90% and (b) cross-sectional SEM image of a 3.8 μm thick RGO/PEDOT film on a silicon wafer.



**Figure 5.** (a) Transmittance and (b) sheet resistance of RGO/PEDOT film before and after the  $SOCl_2$  treatment. An exposure of  $SOCl_2$  for 1 h significantly reduced the sheet resistance for all transferred RGO/PEDOT film without the sacrifice of transmittance. All data are the average of five individual experiments.

(1 kg mass) was essential for conformational contact during the transfer. Insufficient time or pressure during the transfer leads to a partial transfer of the RGO/PEDOT film. When a large volume of filtrate was used, we could also produce a thick hybrid film of RGO/PEDOT as shown in Figure 4b. The cross-sectional SEM view of the hybrid film clearly indicates the well-stacked multilayers of graphene sheets covered with a thick polymer matrix.

The successful reduction of GO in the presence of PEDOT:PSS can be also demonstrated by measuring the sheet resistance of the

transferred film. As in Figure 5, the sheet resistances as well as the transmittance of the film are decreasing with the increase of volume of filtrate. The transferred RGO/PEDOT hybrid film on a quartz substrate exhibits the sheet resistance of 27.3 kΩ/sq at a transparency of 81%. In contrast, the control set of pure PSS (no PEDOT) coated RGO film with a sheet resistance of 40 MΩ/sq (ca. 1500 times higher in sheet resistance).<sup>9</sup> It clearly demonstrates the role of conducting polymer in enhancing the overall conductivity of the RGO/PEDOT hybrid. It is also of note that the

obtained sheet resistance is comparable to the recently reported composite of Nafion with RGO with  $80 \text{ k}\Omega/\text{sq}$  at a transmittance of 81%.<sup>14</sup> Moreover, a unique feature of these hybrid films on a flexible substrate can be further demonstrated by use of a flexible substrate such as PET. We compared the sheet resistance changes of our hybrid RGO/PEDOT film on a PET under forced bending experiments. We found that the hybrid RGO/PEDOT film was maintained on the substrate under excessively bending conditions and retained the initial sheet resistance values without a significant loss over multiple cycles. For example, the sheet resistance of a RGO/PEDOT film changed from  $42.95 \pm 12.90$  to  $42.24 \pm 13.65 \text{ k}\Omega/\text{sq}$  after 100 bending cycles.

As reported previously, the choice of proper dopant is critical in increasing the conductivity of the carbon nanomaterials.<sup>24,25</sup> Here, we have introduced the chemical dopant  $\text{SOCl}_2$  to further increase the conductivity of hybrid RGO/PEDOT film by dipping the transferred film in a solution of  $\text{SOCl}_2$  for 1 h. Interestingly, we found that all of sheet resistance values are significantly reduced by an order of magnitude upon treatment with  $\text{SOCl}_2$  without the sacrifice of transmittance of the composite RGO/PEDOT film. For instance, the sheet resistance value of RGO/PEDOT film prepared from  $75 \mu\text{L}$  of solution dropped from 27.3 to  $2.3 \text{ k}\Omega/\text{sq}$  with a subtle change of transmittance ( $\sim 1\%$ ). In the present study, the lowest sheet resistance is  $0.71 \text{ k}\Omega/\text{sq}$  at 68% transmittance; however, a much lower sheet resistance can be

(24) Dettlaff-Weglikowska, U.; Skakalova, V.; Graupner, R.; Jhang, S. H.; Kim, B. H.; Lee, H. J.; Ley, L.; Park, Y. W.; Berber, S.; Tomanek, D.; Roth, S. *J. Am. Chem. Soc.* **2005**, *127*, 5125.

(25) Tung, V. C.; Allen, M. J.; Yang, Y.; Kaner, R. B. *Nat. Nanotechnol.* **2009**, *4*, 25.

(26) Eda, G.; Lin, Y. Y.; Miller, S.; Chen, C. W.; Su, W. F.; Chhowalla, M. *Appl. Phys. Lett.* **2008**, *92*, 233305.

obtained with increasing the volume of RGO/PEDOT suspension, albeit at a lower transmittance. The enhanced conductivity of the film by doping with  $\text{SOCl}_2$  is likely to introduce  $\text{Cl}^-$  as a p-type dopant as similarly employed in other carbon-based nanomaterials.<sup>24–26</sup> This finding suggests that, through further process optimization, the RGO/PEDOT hybrid film may become an attractive candidate for a large-scale flexible transparent conducting film.

## Conclusion

In conclusion, we have prepared the RGO nanosheet via noncovalent functionalization with a conducting polymer dispersant, PEDOT:PSS. The resulting RGO/PEDOT suspension retains a fairly good colloidal stability in aqueous medium. A large-scale flexible thin film of RGO/PEDOT hybrid can be prepared by filtration, followed by transfer onto a receiving substrate, such as a flexible PET or a quartz slide. The hybrid RGO/PEDOT film exhibits high conductivity with a controllable transmittance, which will find its potential application for a large-scale transparent, conducting thin film.

**Acknowledgment.** This research was supported by WCU (World Class University) program through the Korea Science and Engineering Foundation funded by the Ministry of Education, Science and Technology (R31-2008-000-20012-0) and also by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MEST) (2010-0003219).

**Supporting Information Available:** Additional TGA, SEM, and XPS data. This material is available free of charge via the Internet at <http://pubs.acs.org>.