

High Throughput Preparation of Large Area Transparent Electrodes Using Non-Functionalized Graphene Nanoribbons

Yu Zhu, Wei Lu, Zhengzong Sun, Dmitry V. Kosynkin, Jun Yao, and James M. Tour*

Department of Chemistry, Department of Mechanical Engineering and Materials Science, and the Smalley Institute for Nanoscale Science and Technology, Rice University, MS 222, 6100 Main Street, Houston, Texas 77005, United States

Received July 15, 2010. Revised Manuscript Received December 14, 2010

Transparent conducting films were prepared by using non-functionalized, non-oxidized graphene nanoribbons. Two different dispersion methods were compared. The produced films show sheet resistance as low as 800 Ω /sq when the transmittance at 550 nm is 78%. The performance of the films were close to the entry level for transparent electrode applications and is comparable to those of graphitized chemically converted graphene films, but in this case, no high temperature annealing step was required, thereby rendering this a technique suitable for use on thermally sensitive materials.

Introduction

Theoretical and experimental studies of graphene show that it is a promising material for use in transparent electrodes.¹ As a single-atom layered material, graphene absorbs 2.3% of visible light.¹⁶ Pristine graphene can be produced by mechanical exfoliation of highly ordered pyrolytic graphite (“Scotch tape” graphene),² epitaxial growth on silicon carbide³ and chemical vapor deposition on metals such as Ni⁴ and Cu.⁵ These methods produce high quality graphene and are widely used for understanding the intrinsic properties of graphene. However, the limited throughput makes these processes difficult to use for the preparation of large area transparent electrodes.

Recently, reports^{4a,b,6} have demonstrated transparent electrodes made using CVD graphene, and a large-scale transfer method by Bae et al.⁷ seems particularly attractive for flexible substrates; nevertheless, the CVD graphene transfer process still needs to be improved for rigid substrates such as glass. Another type of graphene derivative that researchers use frequently is graphene oxide (GO).⁸ Unlike pristine graphene, GO is readily dispersible in water as a single layer because of the presence of hydrophilic oxygen groups on the basal planes and edges. However, the defects and oxygen-containing side groups introduced by the oxidative exfoliation reaction also render GO poorly conductive. Reduction by chemical,^{8e,9} thermal¹⁰ or electrochemical¹¹ treatment results in partial recovery of the graphitic character, producing what is

*To whom correspondence should be addressed. E-mail: tour@rice.edu.

- (1) (a) Blake, P.; Brimicombe, P. D.; Nair, R. R.; Booth, T. J.; Jiang, D.; Schedin, F.; Ponomarenko, L. A.; Morozov, S. V.; Gleeson, H. F.; Hill, E. W.; Geim, A. K.; Novoselov, K. S. *Nano Lett.* **2008**, *8*, 1704–1708. (b) Geim, A. K. *Science* **2009**, *324*, 1530–1534. (c) Geim, A. K.; Novoselov, K. S. *Nat. Mater.* **2007**, *6*, 183–191. (d) Li, X.; Zhang, G.; Bai, X.; Sun, X.; Wang, X.; Wang, E.; Dai, H. *Nat. Nanotechnol.* **2008**, *3*, 538–542. (e) Nair, R. R.; Blake, P.; Grigorenko, A. N.; Novoselov, K. S.; Booth, T. J.; Stauber, T.; Peres, N. M. R.; Geim, A. K. *Science* **2008**, *320*, 1308. (f) Novoselov, K. S.; Geim, A. K.; Morozov, S. V.; Jiang, D.; Katsnelson, M. I.; Grigorieva, I. V.; Dubonos, S. V.; Firsov, A. A. *Nature* **2005**, *438*, 197–200. (g) Zhang, Y. B.; Tan, Y. W.; Stormer, H. L.; Kim, P. *Nature* **2005**, *438*, 201–204.
- (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*, 666–669.
- (3) Emtsev, K. V.; Bostwick, A.; Horn, K.; Jobst, J.; Kellogg, G. L.; Ley, L.; McChesney, J. L.; Ohta, T.; Reshanov, S. A.; Rohrl, J.; Rotenberg, E.; Schmid, A. K.; Waldmann, D.; Weber, H. B.; Seyller, T. *Nat. Mater.* **2009**, *8*, 203–207.
- (4) (a) 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*, 706–710. (b) Reina, A.; Jia, X.; Ho, J.; Nezich, D.; Son, H.; Bulovic, V.; Dresselhaus, M. S.; Kong, J. *Nano Lett.* **2008**, *9*, 30–35. (c) Yu, Q. K.; Lian, J.; Siriponglert, S.; Li, H.; Chen, Y. P.; Pei, S. S. *Appl. Phys. Lett.* **2008**, *93*, 113103.
- (5) Li, X.; Cai, W.; An, J.; Kim, S.; Nah, J.; Yang, D.; Piner, R.; Velamakanni, A.; Jung, I.; Tutuc, E.; Banerjee, S. K.; Colombo, L.; Ruoff, R. S. *Science* **2009**, *324*, 1312–1314.
- (6) Li, X.; Zhu, Y.; Cai, W.; Borysiak, M.; Han, B.; Chen, D.; Piner, R. D.; Colombo, L.; Ruoff, R. S. *Nano Lett.* **2009**, *9*, 4359–4363.
- (7) Bae, S.; Kim, H.; Lee, Y.; Xu, X.; Park, J.-S.; Zheng, Y.; Balakrishnan, J.; Lei, T.; Ri Kim, H.; Song, Y. I.; Kim, Y.-J.; Kim, K. S.; Ozyilmaz, B.; Ahn, J.-H.; Hong, B. H.; Iijima, S. *Nat. Nanotechnol.* **2010**, *5*, 574–578.
- (8) (a) 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*, 457–460. (b) Higginbotham, A. L.; Kosynkin, D. V.; Sinitskii, A.; Sun, Z.; Tour, J. M. *ACS Nano* **2010**, *4*, 2059–2069. (c) Hummers, W. S.; Offeman, R. E. *J. Am. Chem. Soc.* **1958**, *80*, 1339–1339. (d) Kosynkin, D. V.; Higginbotham, A. L.; Sinitskii, A.; Lomed, J. R.; Dimiev, A.; Price, B. K.; Tour, J. M. *Nature* **2009**, *458*, 872–876. (e) Stankovich, S.; Dikin, D. A.; Piner, R. D.; Kohlhaas, K. A.; Kleinhammes, A.; Jia, Y.; Wu, Y.; Nguyen, S. T.; Ruoff, R. S. *Carbon* **2007**, *45*, 1558–1565.
- (9) (a) Bourlinos, A. B.; Gournis, D.; Petridis, D.; Szabó, T.; Szeri, A.; Dékány, I. *Langmuir* **2003**, *19*, 6050–6055. (b) Stankovich, S.; Piner, R. D.; Chen, X.; Wu, N.; Nguyen, S. T.; Ruoff, R. S. *J. Mater. Chem.* **2006**, *16*, 155–158.
- (10) (a) McAllister, M. J.; Li, J. L.; Adamson, D. H.; Schniepp, H. C.; Abdala, A. A.; Liu, J.; Herrera-Alonso, M.; Milius, D. L.; Car, R.; Prud'homme, R. K.; Aksay, I. A. *J. Phys. Chem. B* **2007**, *111*, 4396–4404. (b) Schniepp, H. C.; Li, J. L.; McAllister, M. J.; Sai, H.; Herrera-Alonso, M.; Adamson, D. H.; Prud'homme, R. K.; Car, R.; Saville, D. A.; Aksay, I. A. *J. Phys. Chem. B* **2006**, *110*, 8535–8539.
- (11) Voloshin, A. G.; Kolesnikova, I. P. *Elektrokhimiya* **1980**, *16*, 270.

called chemically converted graphene (CCG). Large transparent GO films can be prepared by spin-coating,¹² filtration,¹³ dip-coating,¹⁴ and Langmuir–Blodgett assembly.^{1d} After reduction using, for example, hydrazine^{8d,12b,13–15} or sodium boron hydride,¹⁶ the films become conductive. After further treatment at high temperature, typically, 1000–1200 °C, the sheet resistance of the film can be 1 K Ω /sq with a transmittance of 80% at 550 nm. This value is the entry level at which the material's properties become valuable for some transparent electrode applications.¹⁷ However, only limited substrates can withstand the graphitization temperature, which make practical applications of CCG-based films difficult.

Recently we reported that high aspect ratio, non-oxidized graphene nanoribbons (GNRs) can be synthesized by splitting multiwall carbon nanotubes with potassium vapor.¹⁸ The potassium produced GNRs (K-GNRs) are only 3 to 5 graphene layers. The width of the ribbons is between 100 and 500 nm, and the length is over 5 μ m. The new GNRs are promising transparent electrode materials because they combine the merits of both graphene sheets and carbon nanotubes: they are as transparent as few layered graphene and they can form percolation networks like carbon nanotubes, so complete surface area coverage is not essential. In addition, since they are free of oxygen containing groups, K-GNRs are highly conductive even without annealing. Single K-GNRs exhibit conductivities of 80,000 S/m. Although the intrinsic pristine graphene-like structure also renders K-GNRs poorly soluble in common solvents, they can be dispersed at 1 mg/mL in aqueous sodium dodecyl sulfate (SDS) or in chlorosulfonic acid, both of which are used to disperse carbon nanotubes.¹⁹ By using K-GNR solutions, a high-throughput procedure for preparing transparent GNR films on glass substrates is reported in this work. The produced film is transparent and has a sheet resistance comparable with the graphitized CCG films. The use of a low cost substrate and a scalable solution-based procedure renders this method an attractive procedure to prepare graphene-based transparent electrodes.

Experimental Procedures

Two kinds of K-GNRs solution were prepared: K-GNRs can be directly dispersed either in chlorosulfonic acid or in water

- (12) (a) Becerril, H. c. A.; Mao, J.; Liu, Z.; Stoltenberg, R. M.; Bao, Z.; Chen, Y. *ACS Nano* **2008**, *2*, 463–470. (b) Wu, J. B.; Becerril, H. A.; Bao, Z. N.; Liu, Z. F.; Chen, Y. S.; Peumans, P. *Appl. Phys. Lett.* **2008**, *92*, 263302.
- (13) Eda, G.; Lin, Y. Y.; Miller, S.; Chen, C. W.; Su, W. F.; Chhowalla, M. *Appl. Phys. Lett.* **2008**, *92*, 233305.
- (14) Wang, X.; Zhi, L.; Mullen, K. *Nano Lett.* **2007**, *8*, 323–327.
- (15) Zhu, Y. W.; Cai, W. W.; Piner, R. D.; Velamakanni, A.; Ruoff, R. S. *Appl. Phys. Lett.* **2009**, *95*, 103104.
- (16) Gao, W.; Alemany, L. B.; Ci, L.; Ajayan, P. M. *Nat. Chem.* **2009**, *1*, 403–408.
- (17) Kaempgen, M.; Duesberg, G. S.; Roth, S. *Appl. Surf. Sci.* **2005**, *252*, 425–429.
- (18) Kosynkin, D. V.; Lu, W.; Sinitskii, A.; Pera, G.; Marcano, D.; Tour, J. M. *ACS Nano* **2011**, DOI: 10.1021/nn102326c.
- (19) Davis, V. A.; Parra, V.; Green, M. J.; Rai, P. K.; Behabtu, N.; Prieto, V.; Booker, R. D.; Schmidt, J.; Kesselman, E.; Zhou, W.; Fan, H.; Adams, W. W.; Hauge, R. H.; Fischer, J. E.; Cohen, Y.; Talmon, Y.; Smalley, R. E.; Pasquali, M. *Nat. Nanotechnol.* **2009**, *4*, 830–834.

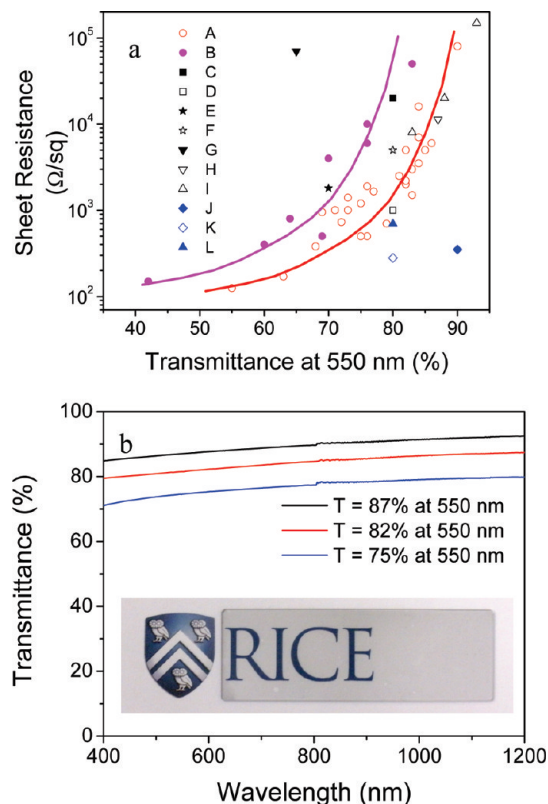


Figure 1. (a) Relationship of transmittance and sheet resistance of graphene-based thin films. (A) K-GNRs film prepared from chlorosulfonic solution in this work. (B) K-GNRs film prepared from SDS solution in this work. The red and magenta curves are descriptive curves of the $R_s \sim T$ % relationship for two K-GNRs films, respectively. (C) Graphene oxide film reduced by hydrazine and annealed at 400 °C from Becerril et al.^{12a} (D) Graphene oxide film reduced by hydrazine and annealed at 1100 °C from Becerril et al.^{12a} (E) Graphene oxide film reduced by hydrazine and annealed at 1100 °C from Wang et al.¹⁴ (F) Graphene oxide film reduced by hydrazine and annealed at 1100 °C from Wu et al.^{12b} (G) Graphene oxide film reduced by hydrazine and annealed at 200 °C from Eda et al.¹³ (H) Graphene oxide film reduced by hydrazine and annealed at 200 °C from Zhu et al.¹⁵ (I) Graphene oxide film reduced by hydrazine and annealed at 800 °C from Li et al.^{1d} (J) CVD grown graphene from Li et al.⁶ (K) CVD grown graphene from Kim et al.^{4a} (L) CVD grown graphene from Reina et al.^{4b} The transmittance values in panel a are measured at 550 nm, except for (I), which were measured at 1000 nm. (b) Full range transmittance of the K-GNRs films prepared in this work. Inset: Photo of a 1 \times 3 in. glass slide with a K-GNR film.

with SDS. The concentration of K-GNRs is 1 mg/mL in both cases. Normal glass substrates (Premiere Microscope slides 9101) were used as supports for the K-GNR thin films. Since high temperature annealing was not necessary, quartz was not required for this work. Several scalable solution-based film formation methods such as spin-coating, blade coating, and spray coating were tested. Spray coating was chosen because of the high quality of the films formed. Films were immediately formed upon the evaporation of the solvent from the preheated substrate. They were cleaned by washing procedures based on the solvents used.

Preparing Graphene Ribbons Film using Chlorosulfonic Acid.

Twenty milligrams of K-GNRs was dissolved in 20 mL of chlorosulfonic acid (Aldrich 98%). The mixture was sonicated in a bath sonicator (Cole Parmer, Model 08849–00) for 1 min and then centrifuged for 3 h at 10000 rpm. The supernatant, which had a concentration near that of the starting solution since very little precipitate was observed, was collected by decanting into a glass vial inside a glovebox. The clean glass substrates were placed on a hot plate and heated to \sim 350 °C. The glass vial

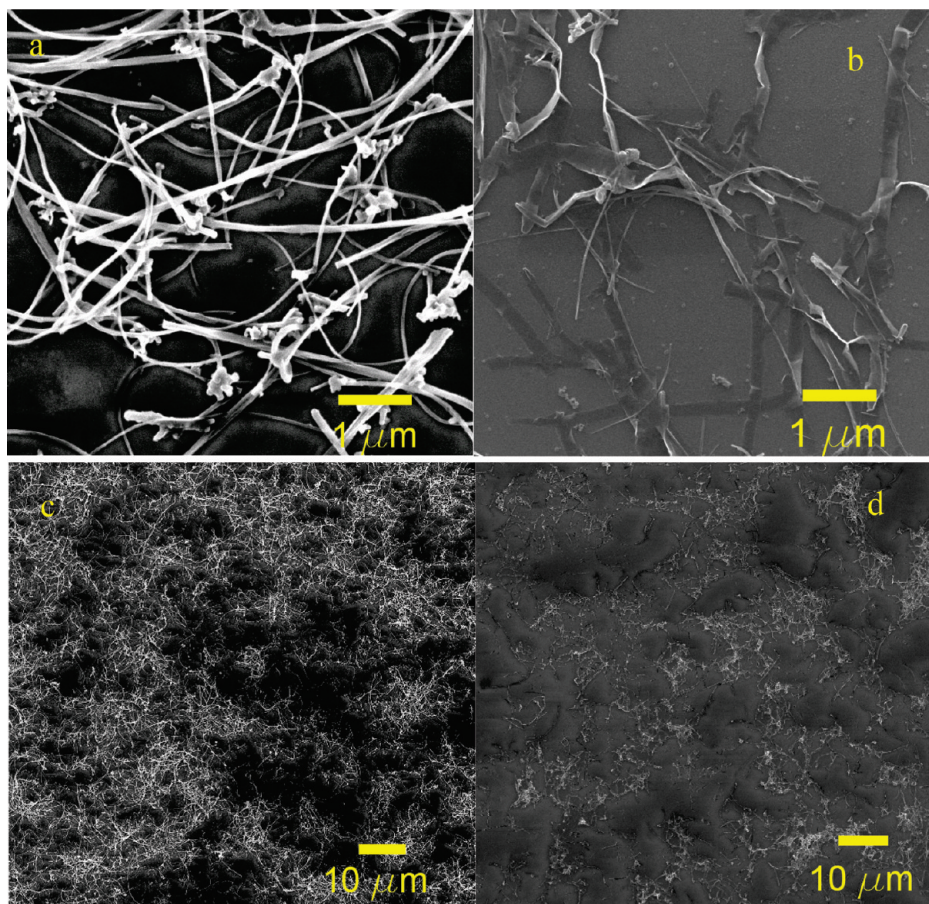


Figure 2. SEM images of graphene nanoribbons films on glass. a,b: films formed by spraying a surfactant K-GNR solution and chlorosulfonic acid K-GNR solution, respectively; c,d: the same films at lower magnification, respectively. The concentrations of the K-GNRs were 1.0 mg/mL in both cases. The concentration of surfactant was 0.5 mg/mL. The same volume of solution was sprayed and the films were cleaned as described in the Experimental Procedures. The images indicate that the GNR film from the chlorosulfonic acid solution is less aggregated.

containing K-GNRs solution was connected to an Iwata Eclipse HP-BCS airbrush by a homemade acid resistive junction. The solution was then sprayed onto the hot glass substrates. The acid evaporated quickly from the hot glass and a gray transparent K-GNRs film was immediately formed. The spraying must be carried out in a well ventilated fume hood. After the substrates cooled, they were immersed into hot ethanol (80 °C) and hot ethanol/water mixture (1:1 volume ratio, 80 °C) for 5 min to remove residual acid. The film was dried in a vacuum oven (75 mm Hg) at 80 °C for 24 h before the electrical measurements.

Preparing Graphene Ribbons Film Using Surfactant Solution. Twenty milligrams of K-GNRs was dissolved in 20 mL of 0.5 M sodium dodecylsulfate (SDS) aqueous solution. The mixture was sonicated using a tip sonicator (MISONIX sonicator 3000) for 3 h and then centrifuged for 30 min at 3000 rpm. The supernatant, which had a concentration near that of the starting mixture since very little precipitate was observed, was collected by decanting into a glass vial. The clean glass substrates were put on a hot plate and heated to 100 °C. The K-GNRs SDS solution was then sprayed on the hot glass substrates by using an Iwata Eclipse HP-CS airbrush. The water evaporated quickly from the hot glass, and a gray transparent K-GNRs/SDS film was immediately formed. After the substrates were cooled to room temperature, they were immersed into 50% H₂SO₄ for 5 s, (this short

exposure to sulfuric acid had proved to be a key step in removing surfactant²⁰), then soaked in hot ethanol/water mixture (80 °C, 1:1 volume ratio) for 1 h.^{20a} The film was dried in a vacuum oven (75 mm Hg) at 80 °C for 24 h before the electrical measurements.

Results and Discussion

The sheet resistance and transmittance of the cleaned transparent K-GNRs films were measured using a four-point probe and a UV/vis spectrophotometer, respectively. The relationship between sheet resistance and transmittance is shown in Figure 1a.

The films formed using the chlorosulfonic acid solution exhibit good electric properties (red circles in Figure 1a). The transmittance of the film (at 550 nm) reaches 83% when the sheet resistance is 1500 Ω/sq. At lower transmittance (78%), the sheet resistance of the film was 800 Ω/sq. The results of K-GNR films are better than or comparable to the published graphene oxide based films (data points C–I in Figure 1a), in which a high temperature graphitization was applied.^{1d,12,14} Note that glass cannot be used for high temperature graphitization, but works well with our procedure. The film is robust enough to be measured many times by the four point probe equipment. Although the K-GNR films are less conductive than CVD graphene (J–L in Figure 1a), the solution

(20) (a) Dan, B.; Irvin, G. C.; Pasquali, M. *ACS Nano* **2009**, *3*, 835–843. (b) Geng, H.-Z.; Kim, K. K.; So, K. P.; Lee, Y. S.; Chang, Y.; Lee, Y. H. *J. Am. Chem. Soc.* **2007**, *129*, 7758–7759.

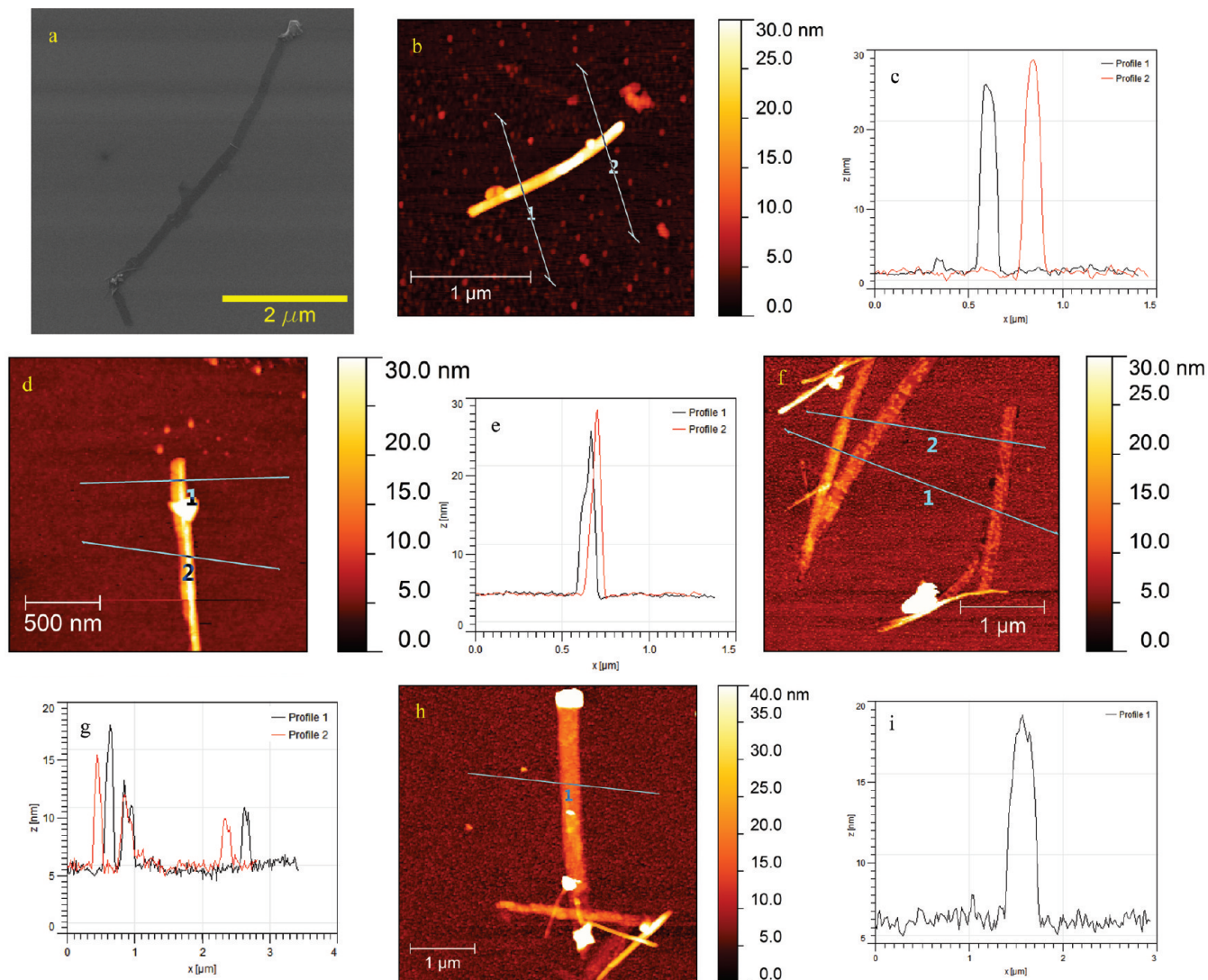


Figure 3. (a) SEM image of exfoliated K-GNRs ribbons. The sample was prepared by drop casting K-GNRs chlorosulfonic acid solution on a silicon chip with 300 nm thermal oxide. (b–e) AFM images of graphene nanoribbons on silicon chip with 300-nm-thick thermal oxide. The samples were prepared by drop-casting the surfactant K-GNR solution. b,d: AFM images. c,e: The height profiles along marked paths in b and d, respectively. The heights of the ribbons are 20–30 nm. (f–i) AFM images of graphene nanoribbons on silicon chips with 300-nm-thermal oxide. The samples were prepared by drop-casting the chlorosulfonic acid K-GNR solution. f,h: AFM images. g,i: The height profiles along marked paths in f and h, respectively. The heights of the ribbons are 5–12 nm.

processed K-GNR films provide transparent electrodes in a high-throughput procedure. By using an airbrush (Iwata Eclipse HP-CS and Iwata Eclipse HP-BCS), five 1×3 in. glass substrates can be coated with K-GNR films in 30 s. Figure 1b is the typical transmittance spectra of the sprayed films. The blank glass was used as background during the measurements. The transmittance of films slightly decreases when the wavelength decreases, but it maintains a variation smaller than 5% in the entire measured range from 400 to 1200 nm. The inset of Figure 1b shows a 1×3 in. glass slide coated with a K-GNR thin film having a transmittance of 80% at 550 nm.

When the surfactant dispersed K-GNR solution was used to form K-GNR films, the results are shown as magenta solid circles in Figure 1a. On average, these films are 10 times less conductive than the chlorosulfonic acid-prepared films at the same transmittance. The reason could be the higher aggregation states for the film prepared

from the surfactant solution, which was confirmed by our SEM experiments (see below). Additionally, because of the existence of surfactant, the as prepared K-GNRs films are almost non-conductive, implying the washing procedure is crucial for surfactant solution-prepared films. Acid dipping is an effective way to remove surfactant;²⁰ however, in some cases the films became detached from the substrate after acid dipping. The higher the transmittance of the film, the easier the film became detached. It was challenging to get films with transmittance over 80% by acid treatment. The reason for the easy detachment of the film after acid treatment is the existence of the surfactant in the solid film, which upon washing will be dissolved and cause the destruction of the film.

The SEM images of the thin films are shown in Figure 2. Figure 2a and 2b are the films formed by surfactant solution and chlorosulfonic acid solution, respectively. It is clear that percolation paths are formed in both films.

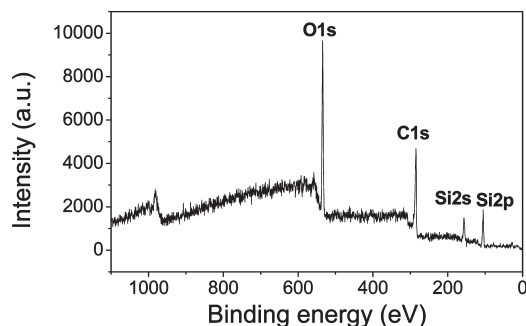


Figure 4. XPS spectra of a K-GNR thin film on glass substrate. The K-GNRs thin film was prepared by spraying K-GNR chlorosulfonic acid solution on heated glass substrates. The signals from the glass were also obtained (Si and O signals).

The K-GNRs are in contact with each other and there are many empty spaces for high transmittance. Figures 2c and 2d are SEM images of a larger area. The K-GNRs covered the substrate with entangled coils that appear to be evenly distributed. Furthermore, based in Figure 2a and 2b, the K-GNR ribbons were better exfoliated by using the chlorosulfonic acid procedure. When the surfactant solution was used to disperse graphene, the as-prepared films contained many open nanotubes consisting of many layers of stacked graphene ribbons (Figure 2a). However, in the films formed by chlorosulfonic acid dispersions, the major components are well exfoliated thinner ribbons. Recent reports^{19,21} have confirmed that chlorosulfonic acid exfoliates graphene and bundled nanotubes. In this work, we also found the thinner exfoliated ribbons (4–12 nm) (Figure 3) are the major form of K-GNR in chlorosulfonic acid solution. However, the thicker ribbons (20–30 nm) are the major form of K-GNRs produced from the surfactant solution (Figure 3). The conclusion that chlorosulfonic acid exfoliates K-GNRs is supported by the fact that centrifuging 1 mg/mL of K-GNRs in chlorosulfonic solution at 10000 rpm did not produce a visible precipitate, but the same treatment of 1 mg/mL surfactant K-GNR solution produced an obvious precipitate albeit minimal. The more highly exfoliated K-GNRs in the chlorosulfonic acid solution as compared to the surfactant solution explains the different sheet resistances of the films formed. The flat, exfoliated K-GNRs in Figure 2b should have smaller contact resistance between ribbons, which would lead to lower sheet resistance of the film.

To understand why the film was highly conductive even without doing any thermal annealing treatments, we analyzed the products using X-ray photoelectron spectroscopy (XPS); the spectra of the thin films are shown in Figures 4 (survey) and 5 (high resolution C1s). Previous results for

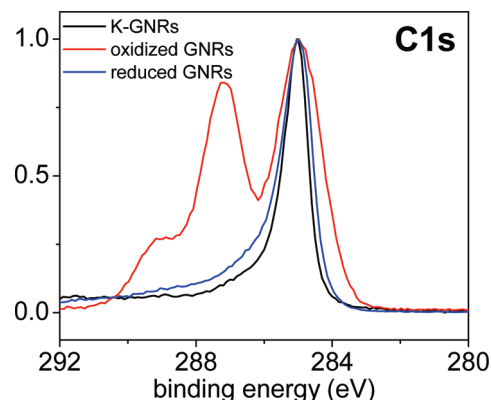


Figure 5. High-resolution XPS C1s spectra of oxidized GNRs (red), reduced GNRs (blue), and K-GNRs films (black). The fwhm of the major C1s peak are 1.67 eV, 1.13, and 0.87 eV for oxidized GNRs, reduced GNRs, and K-GNRs, respectively. The base pressure of the system was at 5×10^{-9} Torr. A monochromatic Al X-ray source at 100 W was used with a pass energy of 26 eV and with a 45° takeoff angle. The beam diameter was 100.0 μm . Binding energy values were referenced externally to a gold 4f peak at 84.00 eV and internally to a carbon 1s binding energy of 280.50 eV (NIST XPS Database).

the oxidized GNRs^{8d,22} and chemically reduced GNRs^{8d,22} are also shown in Figure 5. The high resolution XPS C1s spectrum of oxidized ribbons (red curve in Figure 5) show strong signals at 286–290 eV, indicating the existence of C–O and C=O functionalities. Those oxygen-containing functionalities were partially removed by reduction, as shown in the C1s spectrum of reduced ribbons (blue curve in Figure 5), which shows only one strong signal at 285 eV. The C1s spectrum of the K-GNR film (black curve in Figure 5) looks similar to that of reduced GNRs; however, the full width at half-maximum (fwhm) of the K-GNR film and reduced GNRs are 0.87 and 1.13 eV, respectively. The larger fwhm for the reduced GNRs indicates the presence of contributions from a variety of different carbon bonding configurations while the narrower peak for the K-GNR film indicates the carbon bonding of the K-GNRs is more homogeneous than other GNRs.

Conclusion

In summary, transparent conducting films are disclosed using non-functionalized, non-oxidized graphene nanoribbons. This solution-based process is suitable for forming large area transparent electrodes. No high temperature annealing under hydrogen of the film is required.

Acknowledgment. The authors thank N. Behabtu, J. Lomeda, and B. Chen for helpful discussions. The work was funded by the Office of Naval Research through the Graphene MURI program (#00006766), the AFOSR (FA9550-09-1-0581) and M-I SWACO, LLC.

(21) Behabtu, N.; Lomeda, J. R.; Green, M. J.; Higginbotham, A. L.; Sinitskii, A.; Kosynkin, D. V.; Tsentelovich, D.; Parra, V.; Schmidt, J.; Kesselman, E.; Cohen, Y.; Talmon, Y.; Tour, J. M.; Pasquali, M. *Nat. Nanotechnol.* **2010**, *5*, 406–411.

(22) Zhu, Y.; Higginbotham, A. L.; Tour, J. M. *Chem. Mater.* **2009**, *21*, 5284–5291.