AP Journal of Applied Physics

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Citation: J. Appl. Phys. **112**, 064309 (2012); doi: 10.1063/1.4752752 View online: http://dx.doi.org/10.1063/1.4752752 View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v112/i6 Published by the American Institute of Physics.

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## ADVERTISEMENT



## Direct laser-enabled graphene oxide–Reduced graphene oxide layered structures with micropatterning

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(Received 6 December 2011; accepted 22 August 2012; published online 19 September 2012)

We present a versatile technique to create three dimensional graphene oxide (GO)-reduced GO (rGO) stacked-layered structure with the added attribute that micropatterns could be defined in each layer. We used focused laser beam to convert GO to rGO and create micropatterns of rGO on a heated substrate with GO spin-coated on it. The sample was then sonicated. Either the (1) micropatterned rGO (laser scanned region) or (2) micropatterned GO (unscanned region) could be preferentially removed depending on laser power and temperature of the substrate. A new layer of GO was then spin-coated and thus achieving 3D multilayered and patterned structure. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4752752]

#### I. INTRODUCTION

Graphene, a single atomic layer of aromatic carbon atoms, has drawn much attention due to its unique electrical properties such as massless fermions, ballistic electronic transport, and ultrahigh electron mobility.<sup>1,2</sup> Hence, researchers have explored the applications of graphene in various interesting devices such as transistors, gas sensors, and electrodes in solar cells.<sup>3,4</sup> In order to facilitate the use of graphene in these devices, many synthesis methods have been developed. These methods ranged from the usage of chemical vapor deposition<sup>5,6</sup> (CVD) to solution phase methods that involve chemically exfoliating graphite.<sup>7</sup> Alternatively, the reduced form of graphene oxide (rGO) from graphene oxide (GO), which can be synthesized via the oxidative treatment of graphite, promises a low-cost, up-scalable, and solutionbased approach for graphene based device fabrication.<sup>8</sup>

Reduction of GO to rGO has been studied via a number of different approaches such as thermal,<sup>9</sup> chemical,<sup>10</sup> electrochemical,<sup>11</sup> and electron beam generated plasma.<sup>12</sup> As an indication of the degree of reduction,<sup>13</sup> conductivity of rGO (0.1-50 S/cm) has been demonstrated to increase significantly from insulating GO.<sup>14</sup> Mattevi *et al.* have also presented a method that combined thermal and chemical treatments to yield rGO with an impressive conductivity of ~1000 S/cm<sup>15</sup> even though it is still quite far from that of pristine graphene.<sup>16</sup> In our work, we employ the use of focused laser beam to reduce GO. Reduction via focused laser beam allows us to directly pattern conducting rGO in an insulating GO matrix, without the need for pre-patterned mask or substrates. Zhang *et al.* have also employed similar technique for high resolution microcircuit patterning of GO film.<sup>17</sup>

Previous efforts have demonstrated that graphene-based platelets can be assembled into a two-dimensional (2D) constructs such as thin films or papers. The ability to assemble these platelets into three-dimensional (3D) structures could result in carbon materials exhibiting novel physical and electronic properties for applications.<sup>18-20</sup> It was previously reported that 3D GO could be self-assembled into carbon films.<sup>21</sup> Furthermore, in order to achieve even higher degree of performance, the ability to create 3D GO-rGO multilayered structure with micropatterning in a hierarchical manner could further expand the range of potential applications for these materials.<sup>22</sup> For example, layered rGO-GO-rGO structure was found to exhibit good cyclic stability and energy storage capacities comparable to existing thin film supercapacitor.<sup>23</sup> Here, we report a simple and yet versatile method to fabricate micro-patterned multilayered 3D GO-rGO structures with hierarchical control through a series of steps involving (1) irradiation of focused laser beam (Suntech VD-IIIA DPSS Laser,  $\lambda = 532 \text{ nm}$ ) at elevated temperature on GO deposited on SiO<sub>2</sub>/Si substrate, (2) ultrasonicating in acetone-water mixture, and (3) controlled spincoating of GO solution on the resulting patterned substrate.

### **II. EXPERIMENT METHOD**

To prepare the sample for laser patterning, the substrate, silicon dioxide (SiO<sub>2</sub>), was first cleaned via ultra-sonication in ethanol and subsequently in isopropyl alcohol (IPA) for a period of 10 min. Next, it was placed in oxygen plasma chamber for further treatment. GO prepared via modified Hummer's method was then spin coated on the substrate for 20 s with a speed of 1000 revolutions per minute. The concentration of the GO solution used was approximately 0.9 mg/ml. The thickness of GO was about 65 nm–70 nm as determined by atomic force microscopy (AFM, Nanoman, DI300).

GO was irradiated with a focused laser beam (wavelength = 532 nm, Suntech VD-IIIA DPSS Laser) with the sample placed on a heating stage at an initial temperature of 22 °C. The focused beam spot size was approximately 1  $\mu$ m and the

0021-8979/2012/112(6)/064309/6/\$30.00

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objective lens magnification was  $100 \times$ . Laser power and the temperature of the stage were varied in this study. The experiment was repeated with increasing heating temperatures from 30°C to 110°C. At each temperature, the laser power was varied from 5 mW to 80 mW. The heating stage consisted of a heating copper coil wrapped around an aluminium rod. The heating coil was then connected to 236 Keithley current source. The temperature of the stage was constantly monitored by a thermocouple. The movement of the heating stage was controlled by Micos SMC-Basic micro-stage device and it was connected to a computer. The speed of the micro-stage movement was kept constant at 50  $\mu$ m/s. The spatial step size of the stage was  $0.5 \,\mu\text{m}$ . After laser irradiation, each of these samples was ultra-sonicated with water-acetone mixture of 1:1 by volume. The duration of sonication for each sample was fixed at 90 s.

The chemical composition on the surface of GO before and after thermal treatment was analyzed using Thermal Scientific theta probe x-ray spectroscopy. The XPS instrument was equipped with monochromatic Al K $\alpha$  source. The XPS spectral deconvolution was achieved by a curve fitting procedure using the manufacturer's standard software. Component peak shape and full-width-at-half-maximums (FWHMs) for a particular peak envelope were kept the same during curve fitting. To prepare the sample for x-ray photoelectron spectroscopy and Raman characterization, two samples of spincoated GO with different thicknesses of approximately 60 nm and 20 nm were prepared. The two samples were heated at 150 °C under ambient conditions.

### **III. RESULTS AND DISCUSSION**

When the focused laser beam ( $\lambda = 532 \text{ nm}$ ) was irradiated on GO on SiO<sub>2</sub>/Si placed on a heating stage, GO was readily converted to rGO.<sup>24–27</sup> The schematic diagram for the experimental setup is shown in Fig. 1(a). Laser induced micro-patterns on the GO was then sonicated in acetonewater mixture. Either rGO (laser scanned region) or GO (unscanned region) could be preferentially removed in the ultra-sonicating process by careful control of the laser power,



FIG. 1. (a) Schematic diagram showing the experimental setup. (b) Taiji logo with white portion being GO and light blue being rGO caused by laser reduction. (c) Positive development of 2D GO Taiji structure—laser scanned region is being removed by 90 s of sonication. (d) Negative development of 2D rGO Taiji structure—laser unscanned region is being removed by 90 s of sonication. temperature of the stage, proportions of the acetone-water mixture as well as the duration of sonication process. In this work, we shall denote positive development of the pattern as those where laser defined patterns (rGO) were removed after sonication. Similarly, we shall denote negative development of the pattern as those where laser defined patterns (rGO) were retained after sonication. This patterning process resembles that of a photoresist patterning, where positive resist has the light exposed region dissolved in the developer solution and vice versa. Fig. 1(b) shows a "Taiji" pattern with white region being GO and light blue region being rGO caused by laser reduction. First, we present the case of positive development. Fig. 1(c) shows the positive development of the GO patterns as the rGO (laser scanned region) was removed through ultra-sonication in the water-acetone mixture. The temperature of the stage and the laser power used during the laser irradiation were 80 °C and 13.4 mW, respectively. Next, Fig. 1(d) shows the negative development as the GO (unscanned region) was removed by ultra-sonication. In this case, the temperature of the stage and the laser power used during the laser irradiation were 40°C and 54 mW, respectively.

By varying the laser power and temperature in a series of experiments, a scientist's guide showing the laser power and temperature that were favorable for each type of micropattern development was constructed and shown in Fig. 2(a). Every single symbol as indicated in Fig. 2(a) corresponds to a set of experimental conditions explored in this work. The guide drawn was divided into four typical regions, namely, positive development, negative development, partial development, and unsuccessful development. Unsuccessful development of the pattern refers to two different scenarios. The first one corresponds to the complete removal of GO and rGO after 90 s of sonication. The second scenario corresponds to the case where both GO and rGO remained after 90s of sonication. At temperature below 50 °C and laser power below 19.2 mW, almost all GO and rGO were removed after sonication. However, when the laser power was above 25 mW, the laser defined pattern (rGO) remained after the sonication process, giving rise to negative development of the pattern. When the sample was exposed to laser beam with a power between 19 mW and 25 mW with sample heating temperature of 50 °C or below, we have partial development where only part of the GO was removed during the sonication. Positive development was favorable for the sample patterned when temperature of the stage was between 60 °C and 85 °C and laser power in the range of 10 mW to 20 mW. At high temperature (>60 °C) and high laser power (>35 mW), both rGO and GO became strongly adhered to the substrate and could not be removed after 90 s of sonication in water-acetone mixture.

We propose the following mechanism for the positive and negative micro-patterning processes. Controlling the temperature was crucial in enhancing the formation of bonds between the GO and the SiO<sub>2</sub> substrate. When the temperature of the heating stage was below 50 °C, there was insufficient formation of bonds between the GO and the substrate. As a result, the GO was readily removed during sonication. At 80 °C and above, the higher temperature facilitated the formation of strong adhesive force between the GO and the substrate. As a result, none of the GO was removed during the short duration of sonication. Heating the sample at a temperature range of 50 °C to 80 °C corresponded to the intermediate state and thus only part of GO was removed during sonication. This was denoted as partial development. It should be noted that if the time of sonication was set longer



FIG. 2. (a) Scientist guide showing the temperature and laser power favorable for positive and negative development of patterns on GO. The time of sonication was kept at 90 s. (b) Optical images of rGO with laser power (right) 15.6 mW and (left) 30.1 mW, respectively. The temperature of the stage was kept constant at 80 °C. (c) AFM measurements on the surface roughness of laser induced rGO with laser power.

than 90 s, all the GO would eventually be removed for the temperature range explored  $(22 \degree C-110 \degree C)$  in this work.

The addition of the focused laser beam provided a number of advantages. First, the focused laser beam served as a direct writing tool for localized micro-patterning in converting the exposed GO to reduced form of GO (rGO). Second, the localized heating caused by the focused laser beam served as a localized "welding" tool to further pin the GO/ rGO onto the substrate against the effect of sonication. Third, at a lower laser power, the focused laser beam helped to create more structural defect in the sample and rendered them to be more susceptible to removal by sonication. It was previously reported and shown that when GO is treated with laser power (~5 mW), large domains of stacked laser reduced graphene oxide are formed.<sup>28</sup> Thus, the focused laser presented an interesting and very useful option to achieve control developments on the substrate. As shown in the scientist's guide in Fig. 2(a), with sample heated at temperature below 50 °C, normally the GO would be removed after sonication. However, after the sample was exposed to focused laser beam with a power above 30 mW, we could achieve the negative development where laser defined region remained after sonication. In this case, we utilized the "welding" attribute of the laser beam. On the other hand, heating the substrate at a higher temperature ( $60 \degree C$  to  $85 \degree C$ ) facilitated stronger adhesion between the GO and the substrate. But the presence of a focused laser beam at moderate power (10-20 mW) provided the destructive effect with sample exposed to the laser beam became physically damaged. And when the sample was subsequently subjected to sonication treatment, these damaged regions became more readily removed by sonication thus giving rise to the positive development as illustrated in Fig. 1(c). Our group has previously reported that laser reduction of GO improves the conductivity to 38.3 S/cm.<sup>29</sup>

To further elucidate the effect of laser beam on the sample, we carried out more systematic characterizations of the sample after laser irradiation. Laser irradiation can create more defects on GO.<sup>30–32</sup> Fig. 2(b) shows the optical image of GO after irradiated with laser of power 15.6 mW and 30.1 mW, respectively. Note that the temperature of the sample stage was maintained at 80 °C during the focused laser treatment. At 15.6 mW, the optical shows many "spots" on the square box but at 30.1 mW such "spots" were significantly less visible. This optical image suggests that rGO caused by

laser reduction with power at 15.6 mW was rougher compared to situation where a higher laser power of 30.1 mW was used. Measurements of the roughness were carried out by AFM. Fig. 2(c) shows a plot of root mean square roughness of the laser treated sample against the power of the focused laser used during the experiment. The roughness of the sample increased with the laser power used initially. This was an indication of the physical damage caused by the moderate laser beam power. As the laser power increased further, the roughness of the sample decreased. This was an indication of the annealing contribution due to the higher laser power. A rougher sample could suggest that more defective sites were present in the sample and hence the rougher portion of the sample could be preferentially removed during sonication in the event of positive development. The AFM measurement shows that there was a large increase in the roughness of rGO using laser power of 10 mW to 20 mW. This implies that positive development of the pattern would be more likely to occur at this laser power range (as the rougher rGO would be easier to be removed) and this is in agreement with the scientist guide shown in Fig. 2(a).

The chemical composition on the surface of GO before and after thermal treatment was analyzed using XPS. Figs. 3(a)-3(d) show the XPS spectra obtained. Figs. 3(a) and 3(b)correspond to the XPS spectra of 60 nm thick GO before and after heated at  $150 \,^{\circ}$ C under ambient conditions and Figs. 3(c) and 3(d) correspond to the XPS spectra of 20 nm thick GO. The binding energy of C-C bond is assigned to be around 284.5 eV and C-O, C=O, and O=C-O have binding energies around 286.1 eV, 287.6 eV, and 288.9 eV, respectively. The error bound for the data obtained is 0.2 eV. As evident from XPS data, the relative peak area intensity of C–O to C–C for both samples decreases after thermal reduction. This is in agreement with the results previously reported.<sup>33</sup> XPS spectra obtained from 20 nm thick GO show Si peak around 101.8 eV and there is no peak shift before and after thermal reduction. In addition, 60 nm thick GO was too thick to pick up signal between Si and GO interface and it does not show Si peak in the XPS spectra. From the XPS data obtained, it does not suggest the possibility of charge transfer between GO and Si during thermal reduction process. Hence, the bond formed between the GO and the substrate is thus attributed to be physisorbed in nature. Each coating of the GO layers consists of many layers and Raman spectra do not show any difference at the thicker and the thinner layer regions.



FIG. 3. C1s XPS spectra of 60 nm thick GO (a) after reduction and (b) before reduction. C1s XPS spectra of 20 nm thick GO (c) after reduction and (d) before reduction.

We have demonstrated that it is possible to fabricate GO or rGO micro-patterns on the substrate with controlled positive or negative development. After the fabrication of the patterned layer, it is possible to deposit additional layer on top and the same patterning process can be repeated. As a result, we could create three-dimensional (3D) micropatterned multilayered structures and we could design and define a wide variety of micro-pattern in each layer en route to three-dimensional layered structures. As examples, we created layered and patterned 3D structures using the positive development technique. The

temperature of the heating stage and the laser power used to create the 3D structures were 80 °C and 12.6 mW, respectively. Fig. 4(a) shows the schematic diagram of the processes. First, a portion of GO was laser reduced to rGO. Next, the sample was sonicated for 90 s to remove rGO (laser irradiated portion). Third, GO was spin-coated on the sample. The thickness of this could be controlled by the concentration of GO and the speed at which the sample was rotated. Then, the sample was exposed to laser irradiation again and sonication followed after that. A series of laser irradiation, sonication, and



FIG. 4. (a) Schematic diagram for 3D patterning of GO. Positive development is used to develop the 3D patterns. (b) Checkered boxes after removing the scanned region through sonication. The sample was then spin coated with GO again to obtain (c), where yellowish region denotes a thicker GO region and light blue region denotes a thinner GO region. (d) "Staircase" like structure with two coatings of GO. AFM measurements show that each step was about 65–70 nm. (e) Taiji logo with one coating of GO (f) two coating of GO. (g) Two circular dots inside the logo are being removed through sonication after scanning with focus laser beam.

spin-coating could be repeated to create multilayered structure with the added attribute that micropatterns could be defined and fabricated in each layer. In this way, we created 3D layered structure with micro-patterning.

Fig. 4(b) shows the intermediate step of a checkeredbox GO pattern after sonication and Fig. 4(c) shows the final pattern after GO was spin-coated on the sample. Yellowish portion represents a thicker GO coating and light blue portion corresponds to the thinner coating. Fig. 4(d) shows a staircase structure with two coatings of GO. Optical image shows the color contrast of each step and the height of each step was measured by AFM. AFM characterization shows that each step height was about 60-70 nm. Figs. 4(e)–4(g) show the process of patterning a three-dimensional micropatterned logo with 3 layers. Fig. 4(e) shows one coating GO and Fig. 4(f) shows the sample with two different coatings. Laser was irradiated on the two small circular spots in Fig. 4(f) and finally removed through sonication for 90 s as shown in Fig. 4(g).

### **IV. CONCLUSION**

In conclusion, when laser was irradiated on GO, it can be converted to rGO. By tuning the laser power and temperature of the GO during the patterning process, rGO (positive development) or GO (negative development) can be preferentially removed by ultrasonication. Positive development of the patterns requires more careful experimental control as the conditions for this type of development work for a narrower range of laser power and temperature after which a new layer of GO can be spin-coated and the process can be repeated en route to 3D multilayered layered structure with the pre-defined micropatterns in each layer. The end product is akin to the stacked microelectronic devices except that, instead of Si based system, we have a complete GO-rGO system. Such 3D micropatterned and multilayered structure could offer more potential applications for graphene-based devices.

- <sup>1</sup>F. Wang, Y. Zhang, C. Tian, C. Girit, A. Zettl, M. Crommie, and Y. R. Shen, Science **320**, 206 (2008).
- <sup>2</sup>K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov, Nature 438, 197 (2005).
- <sup>3</sup>M. C. Lemme, T. J. Echtermeyer, M. Baus, and H. Kurz, IEEE Electron Device Lett. **28**, 282 (2007).
- <sup>4</sup>X. Wang *et al.*, Nano Lett. **8**(1), 323–327 (2008).
- <sup>5</sup>J. Coraux, A. T. N'Diaye, C. Busse, and T. Michely, Nano Lett. 8, 565 (2008).
- <sup>6</sup>P. W. Sutter, J. I. Flege, and E. A. Sutter, Nat. Mater. 7, 406 (2008).

- <sup>7</sup>S. Park and R. S. Ruoff, Nat. Nanotechnol. 4, 217 (2009).
- <sup>8</sup>S. A. Wang, P. K. Ang, Z. Q. Wang, A. L. L. Tang, J. T. L. Thong, and K. P. Loh, Nano Lett. **10**, 92 (2010).
- <sup>9</sup>C. Mattevi, G. Eda, S. Agnoli, S. Miller, K. Andre Mkhoyan, O. Celik, D. Mastrogiovanni, G. Granozzi, E. Garfunkel, and M. Chhowalla, Adv. Funct. Mater. **19**, 2577 (2009).
- <sup>10</sup>S. Stankovich, D. A. Dikin, R. D. Piner, K. A. Kohlhaas, A. Kleinhammes, Y. Jia, Y. Wu, S. T. Nguyen, and R. S. Ruoff, Carbon 45, 1558 (2007).
- <sup>11</sup>Y. Y. Shao, J. Wang, M. Engelhard, C. M. Wang, and Y. H. Lin, J. Mater. Chem. **20**, 743 (2010).
- <sup>12</sup>M. Baraket, S. G. Walton, Z. Wei, E. H. Lock, J. T. Robinson, and P. Sheehan, Carbon 48(12), 3382–3390 (2010).
- <sup>13</sup>I. Jung, D. A. Dikin, R. D. Piner, and R. S. Ruoff, Nano Lett. **8**, 4283 (2008).
- <sup>14</sup>I. Jung, D. Dikin, S. Park, W. Cai, S. L. Mielke, and R. S. Ruoff, J. Phys. Chem. C 112, 20264 (2008).
- <sup>15</sup>C. Mattevi, G. Eda, S. Agnoli, S. Miller, K. A. Mkhoyan, O. Celik, D. Mostrogiovanni, G. Granozzi, E. Garfunkel, and M. Chhowalla, Adv. Funct. Mater. **19**, 2577 (2009).
- <sup>16</sup>J. H. Chen, C. Jang, S. D. Xiao, M. Ishigami, and M. S. Fuhrer, Nat. Nanotechnol. 3, 206 (2008).
- <sup>17</sup>Y. L. Zhang, L. Guo, S. Wei, Y. Y. He, H. Xia, Q. D. Chen, H. B. Sun, and F. S. Xiao, Nanotoday 5, 15–20 (2010).
- <sup>18</sup>J. R. Lomeda, C. D. Doyle, D. V. Kosynkin, W. F. Hwang, and J. M. Tour, J Am. Chem. Soc. **130**, 16201 (2008).
- <sup>19</sup>X. An, T. Simmons, R. Shah, C. Wolfe, K. M. Lewis, M. Washington, S. K. Nayak, S. Talapatra, and S. Kar, Nano Lett. **10**(11), 4295 (2010).
- <sup>20</sup>E. Y. Choi, T. H. Han, J. Hong, J. E. Kim, S. H. Lee, H. W. Kim, and S. O. Kim, J. Mater. Chem. **20**, 1907 (2010).
- <sup>21</sup>S. H. Lee, H. W. Kim, J. O. Hwang, W. J. Lee, J. Kwon, C. W. Bielawski, R. S. Ruoff, and S. O. Kim, Angew. Chem., Int. Ed. 49, 10084–10088 (2010).
- <sup>22</sup>G. K. Dimitrakakis, E. Tylianakis, and G. E. Froudakis, Nano Lett. 8(10), 3166–3170 (2008).
- <sup>23</sup>W. Gao, N. Singh, L. Song, Z. Liu, A. Leela Mohana Reddy, L. Ci, R. Vajtai, Q. Zhang, B. Wei, and P. M. Ajayan, Nat. Nanotechnol. 6, 496–500 (2011).
- <sup>24</sup>Y. Zhou, Q. Bao, B. Varghese, L. A. L. Tang, C. K. Tan, C.-H. Sow, and K. P. Loh, Adv. Mater. **22**, 67–71 (2010).
- <sup>25</sup>D. A. Sokolov, K. R. Shepperd, and T. M. Orlando, J. Phys. Chem. Lett. 1, 2633–2636 (2010).
- <sup>26</sup>L. Huang, Y. Liu, L. C. Ji, Y. Q. Xie, T. Wang, and W. Z. Shi, Carbon 49, 2431–2436 (2011).
- <sup>27</sup>V. Abdelsayed, S. Moussa, H. M. Hassan, H. S. Aluri, M. M. Collinson, and M. S. El-Shall, J. Phys. Chem. Lett. 1, 2804–2809 (2010).
- <sup>28</sup>V. Strong, S. Dubin, M. F. El-Kady, A. Lech, Y. Wang, B. H. Weiller, and R. B. Kaner, ACS Nano 6(2), 1395–1403 (2012).
- <sup>29</sup>Y. Tao, B. Varghese, M. Jaiswal, S. Wang, Z. Zhang, B. Oezyilmaz, K. P. Loh, E. S. Tok, and C. H. Sow, Appl. Phys. A **106**(3), 523–531 (2012).
- <sup>30</sup>K. P. Loh, Q. Bao, G. Eda, and M. Chhowalla, Nat. Chem. 2, 1015–1024 (2010).
- <sup>31</sup>G. Eda, Y.-Y. Lin, C. Mattevi, H. Yamaguchi, H.-A. Chen, I.-S. Chen, C.-W. Chen, and M. Chhowalla, Adv. Mater. 22, 505–509 (2010).
- <sup>32</sup>A. Bagri *et al.*, "Structural evolution during the reduction of chemically derived graphene oxide," Nat. Chem. 2, 581–587 (2010).
- <sup>33</sup>D. Yang, A. Velamakanni, G. Bozoklu, S. Park, M. Stoller, R. D. Piner, S. Stankovich, I. Jung, D. A. Field, C. A. Ventrice, and R. S. Ruoff, Carbon 47, 145–152 (2009).