



## Optimization of Graphene Conductive Ink with 73 wt% Graphene Contents

Chang-Yan Xu\*, Xiao-Mei Shi, Lu Guo, Xi Wang, Xin-Yi Wang, and Jian-Yu Li

College of Materials and Science, Nanjing Forestry University, Nanjing, Jiangsu 210037, China

With the pace of development accelerating in printed electronics, the fabrication and application of conductive ink have been brought into sharp focus in recent years. The discovery of graphene also unfolded a vigorous research campaign. In this paper, we prepared graphene conductive ink and explored the feasibility of applying the ink to flexible paper-based circuit. Since experimental study concentrating upon ink formulation was insufficient, orthogonal test design was used in the optimization of preparation formula of conductive ink for the first time. The purpose of this study was to determine the effect of constituent dosage on conductivity of graphene conductive ink, so as to obtain the optimized formula and prepare graphene conductive ink with good conductivity. Characterization of optimized graphene conductive ink we fabricated showed good adhesion to substrate and good resistance to acid and water. The graphene concentration of the optimized ink reached 73.17 wt% solid content. Particle size distribution of graphene conductive ink was uniform, which was about 1940 nm. Static surface tension was 28.9 mN/m and equilibrium contact angle was 23°, demonstrating that conductive ink had good wettability. Scanning Electron Microscope (SEM) analysis was also investigated, moreover, the feasibility of lightening a light-emitting diode (LED) light was verified. The graphene conductive ink with optimized formula can be stored for almost eight months, which had potential applications in flexible paper-based circuit in the future.

Keywords: Reduced Graphene Oxide, Conductive Ink, Formula Optimization, Flexible Paper-Based Circuit.

## **1. INTRODUCTION**

Conductive ink and inkjet printing have been pushed forward by the progress of printed electronics and flexible electronics.<sup>1,2</sup> In recently years, numerous flexible electronics, such as printed circuit boards,<sup>3</sup> sensors,<sup>4,5</sup> RFID antenna<sup>6,7</sup> and transparent electrodes,<sup>8,9</sup> etc., have been fabricated by inkjet printing with conductive ink. Generally, the conductive material used involves metal nanoparticles,<sup>10–12</sup> carbon nanotubes<sup>13, 14</sup> and graphene.<sup>15-17</sup> As a cutting-edge material, graphene has many applications in supercapacitor,<sup>18</sup> solar cell<sup>19,20</sup> and transparent conductive film,<sup>21</sup> and is very suitable for fabricating inkjet-printing-conductive ink due to its intriguing conductivity and the potential for low-cost production.<sup>22</sup> So far, three kinds of graphene-related conductive ink have been fabricated, that is, pristine graphene (PG) ink (usually generated by liquid phase exfoliation of graphite in polar solvent<sup>23</sup> or exfoliating graphite

J. Nanosci. Nanotechnol. 2017, Vol. 17, No. xx

\*Author to whom correspondence should be addressed.

using ultrasound-assisted supercritical CO<sub>2</sub><sup>22</sup>), graphene oxide (GO) ink<sup>24</sup> (as a precursor of graphene conductive ink which needs subsequent reduction) and reduced graphene oxide (RGO) ink<sup>25</sup> (often prepared by the redox reaction of graphite). Each kind of ink has pros and cons. PG ink has prominent electrical conductivity but often needs to be exfoliated in toxic solvent or dealing with surface functionalization. The preparation of GO ink can be performed without stabilizer but limited to a post-printing treatment to regain poor conductivity of graphene and often with more defects. Their common disadvantage is the low graphene concentration (lower than 1 wt%).<sup>1</sup> Though RGO ink cannot rival PG ink in conductivity, it has the advantage of being prepared as a homogeneous suspension through versatile synthetic routes in large scale and can be used in inkjet printing technology.<sup>26</sup> Therefore, this research reported graphene conductive ink prepared by the redox reaction of graphite to take full advantage of RGO's large scale production and solvent processibility.

In general, conductive ink is composed of conductive phase with ink binder and additives in solvent. Among which, conductive phase plays the role of conducting current and precluding accumulation of electrostatic on substrate.27 Ink binder is dissolved in solvent to connect conductive phase while additives are to adjust the printability of conductive ink. Hoeng et al.<sup>28</sup> put forward that major issues need to be resolved in the formulation of conductive inks, and attention should not only be focusing on the synthesis of raw material but also the ink formulation for improving the stability, dispersion and physico-chemical properties of the ink. Various methods had been explored to prepare graphene-based ink. Dua et al.<sup>25</sup> printed RGO platelets by reducing GO with ascorbic acid and dispersed resulting RGO in water containing 1% polyethylene glycol (as a binder) and Triton-X100 nonionic surfactant (as a additive) to fabricate a rugged and flexible sensor. Lim et al.<sup>29</sup> produced a RGO/PVA colloidal suspension for inkjet printing, wherein PVA played an important role as binder and stabilizer as well. Lee et al.<sup>30</sup> prepared a graphene ink with high stability by the adsorption of sodium *n*-dodecyl sulphate (SDS) and adjusting the pH value to 10. The film they printed had a much lower conductivity of 6  $S \cdot m^{-1}$  and a resistivity of 16.68  $\Omega \cdot cm$  without any post-heating process. One reason was that the graphene they fabricated had defects, the other was that the lack of ink binder to fabricate conductive ink may led to the discontinuity of printed film. It is worthwhile to note that some studies used both binder and additives, and some just used one of them to prepare conductive ink. And to the best of our knowledge, insufficient attention has been paid on this issue. Therefore, optimal formula of conductive ink is badly in need of intensive and scientific study to obtain desirable printed products with high quality. In this paper, orthogonal test, which was helpful for systematical and efficient evaluation with minimum trials by means of orthogonal table,<sup>31,32</sup> was adopted to thoroughly investigate the effects of each component on the conductivity of resulting ink. Firstly, RGO was prepared by redox reaction process. Then, graphene conductive ink was fabricated with the obtained RGO, carboxymethylcellulose sodium (CMC) and sodium dodecylbenzenesulfonate (SDBS) as conductive phase, ink binder and additive, respectively. The solvent was the mixture of ethonal  $(C_2H_6O)$ , ethylene gly $col (C_2H_6O_2)$  and distilled water, which avoided the use of toxic solvents to expand the development of environmentally friendly graphene-based printing technology. The optimal combination of conductive ink formula was analyzed by orthogonal test and the property of the obtained optimized-formula graphene conductive ink was evaluated by SEM analysis, particle size analysis, dynamic surface tension and contact angle test and so on. At the same time, the practicablity of optimized graphene conductive ink applied in flexible paper-based conductive circuit was also performed.

## 2. EXPERIMENTAL DETAILS

#### 2.1. Materials and Methods

 $C_2H_6O$ ,  $C_2H_6O_2$ , hydrochloric acid (HCl) and sodium hydroxide (NaOH) were obtained from Nanjing Chemical Reagent Co., Ltd. L-ascorbic acid (L-AA) and CMC were purchased from Sinopharm Chemical Reagent Co., Ltd. SDBS was supported by Shanghai Lingfeng Chemical Reagent Co., Ltd. All chemicals were chemical pure as received.

#### 2.2. Preparation of RGO

RGO was obtained by reducing GO with L-AA as a reductant according to our previous report.<sup>33</sup> Firstly, GO was synthesized by modified Hummers' method.<sup>34</sup> Secondly, L-AA (300 mg) was added into GO suspension (300 mL, 1 mg  $\cdot$  mL<sup>-1</sup>), and the pH of the suspension was adjusted to 9–10 by adding 25% ammonia solution. Then the suspension was kept for 2 h at 95 °C with vigorous stirring, resulting in RGO. After being cooled to room temperature, the obtained RGO was washed with distilled water to neutral.

# 2.3. Fabrication of Graphene Conductive Ink 2.3.1. Formula Schedule

All associated factors, such as wettability, volatility and patterning, shall be considered to optimize the formula of graphene conductive ink. Herein, combined with RGO as conductive substance, CMC and SDBS were used as ink binder and addictive, respectively. Moreover, our previous study indicated that the optimal mixed solvent was ethanol (10): deionized water (9): ethylene glycol (1) (vol%). An orthogonal test schedule,  $L_9(3^4)$ , was chosen to investigate the dosage of such factors as RGO, CMC, SDBS and mixed solvent on conductive property of the resulting ink, shown in Table I. Each factor had three levels, regardless of interaction between each factor. The resistance value of prepared graphene conductive ink coating on photo paper substrate with different levels of factors was the test index. Variance analysis and significance analysis were used to analysis the results of different samples. After analysis, the optimized formula was used to prepare graphene conductive ink for subsequent characterization.

## 2.3.2. Preparation of Graphene Conductive Ink

For each experiment, certain amount of RGO, CMC and SDBS were mixed in compound solvent. Then the mixture was sonicated (XO-1200, Xianqu Biological Technology Co., Ltd., China) for at least 2 h under the condition of 20 kHz and 600 W. Through centrifugation (H-1650, Hunan Xiangyi Laboratory Instrument Development Co., Ltd., China) for 30 min at 8000 rpm, large graphene sheets were removed from the matrix, resulting in RGO conductive ink. The process of RGO conductive ink preparation was illustrated in Figure 1.

		Test index			
Test no.	A RGO (mg)	B CMC (mg)	C SDBS (mg)	D solvent (mL)	Y resistance value $(k\Omega)$
1	A <sub>1</sub> (100)	$B_1(100)$	$C_1(10)$	$D_{1}(50)$	11528
2	A <sub>1</sub>	$B_2(200)$	$C_2(30)$	D <sub>2</sub> (75)	14530
3	$A_1$	B <sub>3</sub> (300)	C <sub>3</sub> (50)	$D_3(100)$	28100
4	A <sub>2</sub> (200)	$\mathbf{B}_1$	$C_2$	$D_3$	272
5	A <sub>2</sub>	$B_2$	C <sub>3</sub>	$D_1$	57.6
6	$A_2$	$B_3$	$C_1$	$D_2$	2560
7	A <sub>3</sub> (300)	$\mathbf{B}_1$	C <sub>3</sub>	$D_2$	50.27
8	A <sub>3</sub>	$B_2$	$C_1$	$D_3$	65.67
9	A <sub>3</sub>	$B_3$	$C_2$	$\mathbf{D}_1$	86.43
$K_1$	54158.001	11850.27	14153.67	11672.031	
$K_2$	2889.6	14653.269	14888.43	17140.269	
$\overline{K_3}$	202.371	30746.43	28207.869	28437.669	
$k_1$	18052.667	3950.09	4717.89	3890.677	
$k_2$	963.2	4884.423	4962.81	5713.423	
$k_3$	67.457	10248.81	9402.623	9479.223	
R	17985.21	6298.72	4684.733	5588.546	

 Table I. Four factors with three levels and the results of resistance value after test.

## 2.3.3. Characterization

The conductive property of our graphene conductive ink was characterized by the resistance value of test sample. For the preparation of test sample, fixed volume of conductive ink was uniformly coated on a home-made mode made by photo paper substrate (see in Fig. 5), then the sample was dried at 100 °C to reach absolute dry condition. Each formula of graphene conductive ink coated on substrate was made to an individual test sample (S<sub>1</sub>~S<sub>9</sub>).



Graphene conductive ink



J. Nanosci. Nanotechnol. 17, 1–8, 2017

And the graphene conductive ink made by optimized formula was marked  $S_0$ . For the test of resistance value of one test sample by multimeter, three places were tested for average value.

SEM (S-4800, HITACHI, Japan) was used to observe the sectional and surface microstructure of oven-dried ink on the photo paper substrate ( $S_0$ , coated one time).

Particle size of graphene conductive ink was tested by Laser particle analyzer (Zetasizer Nano ZS) according to the principle of light scattering. 20 mL graphene conductive ink was underwent ultrasonication in the condition of 800 W for 20 min to reach fully dispersed station, and the measurement range was 0.3 to 10  $\mu$ m.

Dynamic surface tension of graphene conductive ink and common commercial ink was tested using SITA T15 dynamic surface tensiometer. Using bubble pressure generated in the testing process for dynamic monitoring, and bubble life is 15 ms-15 s, test temperature was 23 °C.

Contact angle is one of the most important parameters to evaluate wettability of solution. Measurement of contact angle is very important for further application of hydrophobic or hydrophilic coating, material protection, ink and so on. The smaller the contact angle is, the better wettability of liquid is obtained. Automatic OCA40 Micro single fiber contact angle measurement instrument equipped with SCA20-U software was used to measure the contact angle of graphene conductive ink. Photo paper substrate was placed evenly on the sample table, and 1  $\mu$ L droplet was persistently squeezed out from syringe each time.

Optimized graphene conductive ink was coated on a test sample with the size of  $6 \text{ cm} \times 1$  cm and then dried absolutely to test the resistance value for uniformity and continuity test. For uniformity test, test area was divided into six sections in length direction and the resistance value was tested for each centimeter. Then test length increased per centimeter for continuity test. All the data for each test were collected for three repetitions and the average values and standard deviation (error bar) were given.

Once coating, graphene conductive ink presented good wetting and permeating on photo paper surface, and when the solvent evaporated, CMC was assembled onto the surface and connecting RGO flakes, resulting in conductive paths. To test the adhesion force at the interface between graphene conductive ink and photo paper surface, a crosscut tester was applied to observe the test area before and after test.

Adhesion test was performed according to Standard Test Methods for Measuring Adhesion by Tape Test (ASTM D3359-02). Briefly, a lattice pattern with ten cuts in vertical and horizontal directions in graphene conductive ink to the substrate were made by cross cut tester (QFH-HG600, Dongguan Huaguo Precision Instrument Co., Ltd.), pressure-sensitive tape (3 M Scotch 600) was applied over the lattice and then been tore off at a minimum angle, and adhesion was evaluated by comparison Optimization of Graphene Conductive Ink with 73 wt% Graphene Contents

with descriptions and illustrations using an illuminated magnifier.

Graphene conductive ink can be applied in various fields and often exposed to acid, alkali and watery environments unavoidably. In this paper, NaOH, HCl and distilled water was chosen as reagents to assess the agent resistance of the optimized ink. The test was performed according to Graphic technology-Prints and printing inks-Assessment of resistance to various agents (ISO 2836: 2004 (E)). In brief, for acid resistance test, two pieces of filter paper (60 mm  $\times$  90 mm) were immersed in 5 vol% HCl, and the test sample (20 mm  $\times$  50 mm) was placed between filter paper when they were drained until no free solution drips formed. Then they were placed between two glass plates with 1 kg load to hold for 10 min. Afterwards, test sample was washed by deionized water to neutral, then followed by drying at 50 °C for 30 min. When it comes to alkali resistance test, the solution was prepared by 1% mass of NaOH in distilled water and the method was similar to acid resistance test but with a different drying temperature of 40 °C. Meanwhile, water resistance test was also performed with a holding time for 24 h. Assess agent resistance of test samples through observing any changes to them. Agent resistance of common commercial ink was also tested in a controlled experiment.

#### 3. RESULTS AND DISCUSSION

3.1. Optimal Combination of Conductive Ink Formula

The resistance values of prepared graphene conductive ink coating photo paper samples with different levels of factors were presented in Table I.

In range analysis in Table I,  $k_1$ ,  $k_2$  and  $k_3$  represent the average value of the resistance corresponding to different levels of each factor. For an conductive ink coating paper sample, a smaller resistance value means better conductive property. Herein, the smallest *k* corresponded to an optimal level. *R* represented the maximum variation of the resistance values and signified the importance degree of the factors. A larger *R* corresponded to a more significant factor. Conclusions can be drawn from Table I that the optimal combination was  $A_3B_1C_1D_1$  (RGO = 300 mg, CMC = 100 mg, SDBS = 10 mg, solvent = 50 mL), and the dosage of conductive phase (RGO) had more effect on the conductive property of resulting conductive ink than the dosage of ink binder (CMC), dispersant (SDBS) and solvent.

Meanwhile, in order to make up for the ignorance of error in range analysis, variance analysis was applied to estimate the significance of the factor, as shown in Table II. The factors of the dosage of SDBS (C) and the solvent (D) had smaller effect on the conductive property of graphene conductive ink, therefore they were set as error. The result of variance analysis showed that the factor of the dosage of conductive phase (RGO) had outstanding influence on the conductivity of ink, while the factors of the dosage of CMC, SDBS and solvent were insignificant. However,

Table II.	Variance	analysis	and	significance	analysis.
		~		<i>U</i>	~

Factors	Sum of square of deviations (DEVSQ)	Degree of freedom	<i>F</i> ratio	$F$ critical-value $(\alpha = 0.05)$	Significance
RGO	616320005.814	2	13.627	6.94	*
CMC	69323496.726	2	1.533	6.94	
SDBS	41718654.646	2	0.922	6.94	
Solvent	48735508.896	2	1.078	6.94	
Error	90454163.54	4			

the dosage of ink binder (CMC) had more influence on the conductivity of conductive ink than those of dispersant (SDBS) and solvent. In addition, we also found an exciting phenomenon in the course of the experiment that the optimized graphene conductive ink can be stored for almost eight months without any aggregation, which was directly related to the ink binder to effectively stabilize RGO flakes.

The resistance value of graphene conductive ink prepared by the optimal combination (RGO = 300 mg, CMC = 100 mg, SDBS = 10 mg, solvent = 50 mL) was 35.6 k $\Omega$  (±1.6 k $\Omega$ ), which was lower than those of other types of graphene conductive ink with different formulae. The graphene concentration of the optimized ink reached 73.17 wt% solid content. It was worth to note that optimized graphene conductive ink could be stored for nearly eight months without sedimentation, which can be seen in Figure 2. The optimal combination of conductive ink formula was used to prepare graphene conductive ink in our later research.

#### 3.2. SEM Analysis of Oven-Dried Ink on Photo Paper Substrate

Obvious lamellar structure can be seen in the bottom of Figure 3(a), which was RGO ink. The protrusion and the gap part (dotted part in Fig. 3(a)) was caused by permeating of ink into photo paper substrate. However, the adhesion between RGO ink and the photo paper was strengthened because the cohesive vander Waals forces



Figure 2. Optimized graphene conductive ink ((a) once prepared; (b) placed for eight months).

J. Nanosci. Nanotechnol. 17, 1-8, 2017

Xu et al.



Figure 3. Sectional (a) and surface (b) microstructure of oven-dried ink on photo paper substrate.

among RGO flakes intensified the bonding of RGO ink on photo paper substrate.<sup>35</sup> Figure 3(b) showed a smooth, homogeneous and continuous surface of oven-dried ink on the photo paper, illustrating that large RGO flakes were distributed uniformly on the substrate, and even further, that RGO could be dispersed and stabilized well in the conductive ink. This result was similar with that reported by Lee et al.<sup>36</sup> Therefore, a conclusion can be drawn that the good electronic conductivity of the optimal graphene conductive ink was attributed to the continuity and uniformity of conductive phase formed in the drying process.

#### 3.3. Particle Size Test

Figure 4 showed particle size distribution curve of graphene conductive ink, the result was given that average particle size was 1940 nm. The conductive ink contained ink binder (CMC) and additive (SDBS), and these components had effect on the particle size distribution of the conductive ink. Therefore, particle size distribution curve presented a asymmetry tail peak type, meaning that particle size distribution of graphene conductive ink was uniform.

#### 3.4. Dynamic Surface Tension

As shown in Figure 5, surface tension changed with time. It can be divided into three stages in the dynamic surface tension curve: sharp decline stage (within 0.5 s), gradually decline stage (0.5 s-1 s) and static surface tension stage. The faster dynamic surface tension decreased, the better wetting and spreading performance would be obtained. Therefore, graphene conductive ink showed a better wetting and spreading property when compared to commercial inkjet printing ink. The initial surface tension of graphene conductive ink was much lower than commercial inkjet



Figure 4. Particle size distribution of graphene conductive ink.

J. Nanosci. Nanotechnol. 17, 1-8, 2017



Figure 5. Dynamic surface tension of graphene conductive ink and commercial inkjet printing ink.

printing ink, and very fast (at the ms level) to achieve stable (minimum) static surface tension value of 28.9 mN/m. All these performances can prove its good wettability, and its static surface tension value is suitable for the range of inkjet printing surface tension (25 to 50 mN/m), so graphene conductive ink can be used in inkjet printing.

#### 3.5. Contact Angle Analysis

The interaction between substrate and conductive ink as well as the printing quality were mainly influenced by the wettability of conductive ink. And the wettability can be characterized by measuring the contact angle between substrate and conductive ink. The smaller size of contact angle represented good wettability of conductive ink. Figure 6 showed contact angle of graphene conductive ink varied with time, and the insert showed the contact angle of conductive ink at the initial stage, 0 s, 1 s, and 5 s. When conductive ink dripped on the substrate surface, penetration and spreading were accompanied by



Figure 6. Dynamic contact angle of graphene conductive ink.



Figure 7. Uniformity (a) and continuity (b) test of coating sample.

the formation of contact angle. The initial contact angle was 56°, and decreased to 34° after 1 s, demonstrating that ink was slowly wetting the substrate. After 5 s, contact angle decreased to 23° and remained stable to equilibrium contact angle, illustrating the wetting situation of conductive ink on the substrate had reached saturation. Contact angle decreased rapidly at the beginning of contact, then down to slow, and eventually achieved a relative balance. Combined with SEM picture, the conductive ink spread out on substrate first, and then further wetting the whole area, and part of ink penetrated the substrate. At the same time, combined with the dynamic surface tension analysis, conclusion can be drawn that graphene conductive ink had an excellent wettability on photo paper substrate.



Figure 9. Pictures of samples after agent resistance test.

## 3.6. Uniformity and Continuity Test

Figure 7 showed uniformity and continuity test of coating sample on photo paper by measuring the resistance value with same length (a) and with increasing length (b), respectively. Uniformity test (a) showed no obvious fluctuation in resistance value, indicating a potential application in conductive circuit taking advantage of the good uniformity. (b) showed a rectilineal trend (the resistance increased with increasing length) when verified the continuity of test area. Therefore, graphene conductive ink was well-distributed on photo paper showing electrical continuous and the electrical characteristic was reproducible.

#### 3.7. Adhesion on Substrate by Cross-Cut Tester

It was inevitable for graphene conductive ink coated on photo paper to be scraped when used in real package circulate, a good adhesion would extend service life of the package. During adhesion test, a homogeneous and continuous surface was cut in vertical and horizontal directions, and after 3 M tape been tore off, small RGO flakes were detached at intersection and less than 5% of the test area were affected. The performance of adhesion test reached 4B level in ASTM D3359-02. Therefore, the bond between graphene conductive ink and photo paper surface was strong. This result was in consistent with SEM analysis that part of the ink permeating substrate would strengthen adhesion force. Furthermore, the good adhesion force between ink and substrate was conducive to a good electrical conductivity.

#### 3.8. Agent Resistance of the Graphene Conductive Ink

The agent resistance test results were shown in Figure 9. After alkali resistance test, surface ink layer peeled when washed, indicating that our graphene conductive ink had undesirable resistance to alkali environment. COOH on the edge of RGO react with NaOH leading to bad resistance



Figure 8. Sample went through cross-cut tester.



Figure 10. Optical image of electrical circuit assembled by a graphene conductive ink coated strip on Epson photo paper. The inset is the magnified image of LED.

to alkali. However, when it comes to acid resistance test, a LED was still be lit in a closed circuit after test. And the test sample showed no changes after water resistance test. Hence, graphene conductive ink displayed good resistance to acid and water. In controlled experiment, the commercial ink had bad resistance to alkali and water, which is the universal problem of water-based ink. Therefore, the alkali-resistant modification of RGO conductive ink may be a challenge in its future development.

#### 3.9. Flexible Paper-Based Conductive Ink Circuit

It is worth noting that conductive ink can be applied to fabricate conductive patterns and devices by several methods, such as direct writing,37 printing.38 To further prove the applicability of our graphene conductive ink, a conductive strip with a size of 5 cm  $\times$  2 cm was coated on Epson photo paper using graphene conductive ink and then dried at 100 °C for 2 h. As demonstrated in Figure 10, the electrical circuit was composed of a battery (3.0 V), the coated strip and an LED. When the circuit was connected at two arbitrary contact points on the surface of the coated strip, the LED was illuminated and performed quite well. The coated strip was bent to display the good adhesion between graphene conductive ink and the photo paper substrate which we had discussed above, revealing that our graphene conductive ink can be used in flexible and bendable devices.

## 4. CONCLUSION

Graphene conductive ink with high stability had been successfully fabricated by adding CMC and SDBS in mixed solvents by optimizing the formula using orthogonal test design for the first time, which filled the vacancy in conductive ink formula design. Results showed that the dosage of RGO had significant effect on the conductive property of resulting conductive ink, while the dosage of CMC, SDBS and solvent had insignificant effect. We used the optimized formula to fabricated graphene conductive ink with 300 mg RGO, 100 mg CMC and 10 mg SDBS in 50 mL mixed solvent, which can be stored for almost

J. Nanosci. Nanotechnol. 17, 1-8, 2017

eight months without sedimentation. According to SEM analysis, RGO can be dispersed and stabilized well in conductive ink to form a homogeneous and continuous morphology. And part of ink permeating substrate will be helpful for a good adhesion between ink and substrate. Conductive samples were made by coating graphene conductive ink directly on photo paper. The particle size of graphene oxide was about 1,940 nm, and the ink showed good wettability to photo paper substrate. Graphene conductive ink we prepared had good resistance to acid and water and the ink can be used in flexible and bendable devices in the future.

**Acknowledgments:** This work was financially supported by "A Project Funded by the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD)," "Research and popularization of key techniques of functional electromagnetic shielding composites (Grant No. BC2013432)" and "Research and innovation project of the academic degree graduate students in Jiangsu Province (KYLX15\_0935)."

#### **References and Notes**

- 1. A. Kamyshny and S. Magdassi, Small 10, 3515 (2014).
- V. K. Rao, V. A. K. P. S. Karthik, and S. P. Singh, *RSC Advances* 5, 77760 (2015).
- J. Yang, J. H. Chu, Y. Yang, Y. B. Guo, X. G. Lou, and H. B. Li, Adv. Mater. 380, 129 (2012).
- S. Kim, B. Cook, T. Le, J. Cooper, H. Lee, V. Lakafosis, R. Vyas, R. Moro, M. Bozzi, A. Georgiadis, A. Collado, and M. M. Tentzeris, *IET Microwaves Antennas and Propagation* 7, 858 (2013).
- 5. C. Cho and Y. Ryuh, Sensors and Actuators A-Physical 237, 72 (2016).
- W. W. Li, L. H. Li, L. X. Mo, X. W. Hu, X. Leng, H. Fang, W. B. Li, and S. K. Li, *Advanced Materials Research* 380, 137 (2012).
- 7. Z. Q. Gao and X. P. Zhao, Applied Mechanics and Materials 469, 322 (2014).
- 8. K. Kim, S. I. Ahn, and K. C. Choi, Carbon66 172 (2014).
- 9. Z. Y. Liu, K. Parvez, R. J. Li, R. H. Dong, X. L. Feng, and K. Müllen, *Adv. Mater.* 27, 669 (2015).
- R. Dang, L. L. Song, W. J. Dong, C. R. Li, X. B. Zhang, G. Wang, and X. B. Chen, ACS Applied Materials and Interfaces 6, 622 (2014).
- A. Kosmala, R. Wright, Q. Zhang, and P. Kirby, *Mater. Chem. Phys.* 129, 1075 (2011).
- W. J. Cuia, W. S. Lua, Y. K. Zhang, G. H. Lin, T. X. Wei, L. Jiang, Colloids and Surfaces A-Physicochemical and Engineering Aspects 358, 35 (2010).
- 13. R. P. Tortorich and J. W. Choi, Nanomaterials 3, 453 (2013).
- 14. J. W. Han, B. Kim, J. Li, and M. Meyyappan, *Mater. Res. Bull.* 50, 249 (2014).
- D. Kong, L. T. Le, Y. Li, J. L. Zunino, and W. Lee, *Langmuir* 28, 13467 (2012).
- R. Giardi, S. Porro, A. Chiolerio, E. Celasco, and M. Sangermano, Journal of Materials Science 48, 1249 (2013).
- 17. P. Labroo and Y. Cui, Anal. Chim. Acta 813, 90 (2014).
- 18. Y. Chen, X. Zhang, D. C. Zhang, P. Yu, and Y. W. Ma, *Carbon* 49, 573 (2011).
- E. Singh and H. S. Nalwa, J. Nanosci. Nanotechnol. 15, 6237 (2015).

- E. Singh and H. S. Nalwa, Science of Advanced Materials 7, 1863 (2015).
- R. Karthick, M. Brindha, M. Selvaraj, and S. Ramu, J. Colloid Interface Sci. 406, 69 (2013).
- 22. Y. H. Gao, W. Shi, W. C. Wang, Y. P. Leng, and Y. P. Zhao, Ind. Eng. Chem. Res. 53, 16777 (2014).
- 23. F. Torrisi, T. Hasan, W. P. Wu, Z. P. Sun, A. Lombardo, T. S. Kulmala, G. W. Hsieh, S. Jung, F. Bonaccorso, P. J. Paul, D. P. Chu, and A. C. Ferrari, ACS Nano 6, 2992 (2012).
- 24. M. H. Ervin, L. T. Le, and W. Y. Lee, *Electrochim. Acta* 147, 610 (2014).
- 25. V. Dua, S. P. Surwade, S. Ammu, S. R. Agnihotra, S. Jain, K. E. Roberts, S. Park, R. S. Ruoff, and S. K. Manohar, *Angew. Chem. Int. Ed.* 49, 2154 (2010).
- 26. S. Lim, B. Kang, D. Kwak, W. H. Lee, J. A. Lim, and K. Cho, *Journal of Physical Chemical C* 116, 7520 (2012).
- X. W. Hu, L. H. Li, S. M. Zhao, and X. Leng, Advanced Materials Research 287, 577 (2011).
- 28. F. Hoeng, A. Denneulin, and J. Bras, Nanoscale 27, 13131 (2016).

- 29. S. Lim, B. Kang, D. Kwak, W. H. Lee, J. A. Lim, and K. Cho, *Journal of Physical Chemical C* 116, 7520 (2012).
- **30.** C. L. Lee, C. H. Chen, and C. W. Chen, *Chem. Eng. J.* 230, 296 (2013).
- X. Z. Rena, Y. Q. Liu, and Y. R. Jin, *Applied Mechanics and Materials* 201, 1190 (2012).
- 32. Z. Zhang, H. S. Fang, H. Yan, Z. M. Jiang, J. Zheng, and Z. Y. Gan, *Appl. Therm. Eng.* 91, 53 (2015).
- 33. C. Y. Xu, X. M. Shi, A. Ji, L. N. Shi, C. Zhou, and Y. Q. Cui, *PLoS ONE* 10, 0144842 (2015).
- 34. W. Hummers and R. Offeman, J. Amer. Chem. Soc. 80, 1339 (1958).
- 35. O. S. Kwon, H. Kim, H. Ko, J. Lee, B. Lee, C. H. Jung, J. H. Choi, and K. Shin, *Carbon58* 116 (2013).
- 36. C. L. Lee, C. H. Chen, and C. W. Chen, *Chem. Eng. J.* 230, 296 (2013).
- 37. L. Y. Xu, G. Y. Yang, H. Y. Jing, J. Wei, and Y. D. Han, Nanotechnology 25, 055201 (2014).
- 38. L. Huang, Y. Huang, J. J. Liang, X. J Wan, and Y. S. Chen, *Nano Research* 4, 675 (2011).

Received: 20 April 2017. Accepted: 21 April 2017.