

Inkjet printed flexible electronics on paper substrate with reduced graphene oxide/carbon black ink

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Abstract

A reduced graphene oxide (RGO) and carbon black (CB) ink was fabricated with a mixture of ethanol/ethanediol/propanetriol/deionized water as a solvent, and sodium carboxymethyl cellulose (CMC) as a binding and dispersant. The RGO was obtained by reducing graphene oxide using ascorbic acid as a green reductant at a mild temperature of 95 °C. The flexible paper-based electronic circuits were fabricated by inkjet printing the obtained ink on glossy photo paper substrate with an Epson piezoelectric printer. When the loads of RGO, CB, ethanol, ethylene glycol, glycerol, CMC and deionized water were 96 mg, 504 mg, 12 ml, 30 ml, 30 ml, 480 mg and 51 ml, the electrical conductivity, average particle size and viscosity of the ink were 122.4 μ s/cm, 1.966 μ m and 22.5 mPa s, respectively; and the ink exhibited good acid resistance. A continuous, dense and uniform conductive network was achieved when the printing pass number was 4 for a single circuit. The resistance at both ends of the aforementioned printed circuit (10×2×0.03338, length×width×thickness, mm) was 0.1 MΩ with a resistivity of 0.661 Ωm for the ink layer, and the circuits showed moderate uniformity, adhesion and mechanical flexibility. In the light-emitting diode operation, the three-dimensional conductive circuits also presented good electrical conductivity.

1 Introduction

Printed flexible electrical components have great application potential in consumer electronics and next-generation electronic systems [1, 2]. In terms of patterning process, cost, scalability and reproducibility, inkjet printing is a wellsuited technique for direct deposition of nanoparticles on flexible substrates [3]. It is a good alternative to the conventional fabrication techniques, such as etching, milling, and lithography for flexible electronics [4–6], including sensors

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² School of Packaging, Michigan State University, Packaging Building, 448 Wilson Road, East Lansing, MI 48824-1223, USA [4], electrodes [3, 5, 7], thin film transistors [6, 8], antennae [4], solar cells, displays, and smart packaging [9].

Conductive phase greatly affects the performance of printed circuits. Organometallic compounds, metallic nanoparticle suspensions, conductive polymers, nanocarbontubes and graphene have already been tested as conductive phases [1, 9, 10]. Although metalallic nanoparticles have higher electrical conductivity and air stability than conductive polymers and organometallic compounds [11], the metalallic nanoparticle ink needs to be sintered at temperatures generally too high for most flexible substrates, and the conductivity of the sintered nanoparticles is not as good as that of unsintered ones [12]. In addition, conductive polymers are also generally prone to oxidation in the air. Carbon nanotube is believed to be a good alternative because of its electrical heterogeneousness in nature [1], and tolerance to high current density and thermal conductivity [9]. Many electronic components have been fabricated by inkjet printing carbon nanotube inks on substrates [8, 13–15]. However, the electrical heterogeneousness of carbon nanotubes requires a selective growth and a sorting process for the separation to exploit in full its electronic properties [1]. In this context, graphene is expected to play an important role due to its high specific surface area, good chemical stability, electrical and thermal conductivity and high charge carrier mobility

[16, 17]. Inkjet printing of graphene based inks provides a new platform for printing electronics, including sensors, antennas, transparent conductors, thin film transistors and supercapacitors [9, 18-20]. The great challenge concerning inkjet printing of graphene is to find a environmentallyfriendly solvent that not only satisfies the requirements set by inkjet printers but also allows to prepare stable graphene based dispersion. In this respect, graphene oxide (GO) first attracted the attention of researchers. It is hydrophilic and can be readily dispersed in aqueous solvents due to its heavily oxygenated functional groups on the structure [21]. The traditional method for fabricating graphene electronics is to inkjet print GO onto substrates first, and then to retain the electrical performance of pristine graphite by reducing GO with a thermal or chemical treatment [19, 22], or UV light irradiation [10]. In this method, the electrical properties is degraded due to the presence of structural defects and residual oxygen in reduced GO [23]. Kim et al. offered a single-step process, reactive inkjet printing (RIP), to fabricate graphene films [3]. In RIP, the printer needs at least two nozzles for GO ink and reductant ink, which complicates the inkjet printing system; and furthermore, the substrate is glass slide, not flexible. It is desirable to formulate graphene inks with proper fluidic properties and high graphene concentration, and free from flake aggregation and solvent toxicity [20]. Most researches focus on solution-phase exfoliated graphene inks [18, 24] for electronics, such as thinfilm transistors [25] and graphene/MoS₂ nonasheets [26]. In order to increase the graphene concentration, and to mitigate the toxicity of such solvents as N-methyl pyrrolidone or dimethylformamide, ethyl cellulose (EC) has been used to stablize graphene in organic solvents [7, 18]. On this basis, Li et al. proposed a distillation-assisted solvent exchange technique to obtain high-concentration graphene dispersion [20]. Secor et al. [27] and Capasso et al. [1] went further. Instead of toxic solvents, they used water/ethanol mixtures to exfoliate graphite for fabricating inkjet printed electronics. The main drawback of this process, the added EC needs to be removed through annealing at 300-400 °C in air, which is too high for paper substrate.

In our work, a inkjet printable graphene ink was fabricated with a mixture of reduced graphene oxide (RGO) and carbon black (CB) as conductive filler, a compound of ethanol, ethanediol, propanetriol and deionized water as solvent, and sodium carboxymethyl cellulose (CMC) as binding and dispersant agent. The RGO was obtained by reducing GO using a non-toxic reducing agent, ascorbic acid at a mild temperature of 95 °C for 2 h. CB plays a role to avoid agglomeration of RGO, because the CB particles can distribute on the surface of RGO, and the RGO sheets can bridge CB particles and promote the formation of conductive paths in printed circuits [28, 29]. In the view of environment-friendly performance, porosity and the ease of being folded, unfolded and creased [10], glossy photo paper was selected as our substrate instead of the common used plastic. The effect of RGO load on the properties of inks, the influences of printing pass number on the conductivity, surface morphology, resistivity, uniformity, adhesion ability and bending property of the printed circuits were highly concerned. Light-emitting diode operation with the inkjet printed paper-based three-dimensional circuits was also investigated.

2 Materials and methods

2.1 Experimental materials

Natural graphite powder (40 µm) and CB (BP2000) were obtained from Qingdao Henglide Graphite Co., Ltd. (China) and CARBON Co., Ltd. (USA), respectively. Concentrated sulfuric acid (98 wt%), sodium nitrate, potassium persulfate, phosphorus pentoxide, potassium permanganate, hydrogen peroxide (30 wt%), ascorbic acid, ammonia (25 wt%), ethyl alcohol (boiling point, 78.3 °C; surface tension; 22.4 mN/m), ethylene glycol (boiling point, 197.8 °C; surface tension, 48.5 mN/m), glycerol (boiling point, 290.0 °C; surface tension, 63.3 mN/m), absolute ethyl alcohol, and CMC were analytically pure and purchased from Nanjing Chemical Reagent Co., Ltd. (China). The above boiling point and surface tension data were supported by the manufacturer. Homemade deionized water was used throughout the project.

2.2 Fabrication of RGO/CB Ink

GO was synthesized using a modified Hummers method [30, 31]. Natural graphite powder and corresponding GO suspension were shown in Fig. 1a, b.

RGO was produced by removing oxygen functionalities from GO with reducing agents. The common used reducing agents for RGO such as hydrazine and sodium borohydride are highly toxic. In this paper, ascorbic acid was used as a reducing agent [32]. Ascorbic acid was added into GO suspension with a quality ratio of 10.5:1 (ascorbic acid:GO suspension), and pH was adjusted to 9-10 by ammonia. Then, the mixture was stirred at 95 °C in a water-bath (HH-1, Nanjing Xianou Instrument Manufacturing Co., Ltd., China) for 2 h, and RGO suspension was obtained. RGO suspension was further filtered by a piece of microporous membrane (0.22 µm, Shanghai Mosu Science Equipment Co., Ltd., China) and washed with deionized water until neutral. RGO (Fig. 1c) was obtained by drying the residue in an oven (60 °C) (DZF-6090, Shanghai Jinghong Laboratory Equipment Co., Ltd., China) for 12 h, followed by being ground with an agate grinding bowl for 15 min.



Fig. 1 Photographs of a natural graphite powder, b GO suspension, and c RGO

The fabrication process of RGO/CB ink was divided into two steps (shown in Fig. 2).

The obtained RGO (96 mg), CB (504 mg), ethyl alcohol (12 ml), ethylene glycol (30 ml), glycerol (30 ml), CMC (480 mg) and deionized water (51 ml) were mixed together and been stirred for 10 min, followed by ultrasonication under 960 W by an ultrasonic plant cell shredder (XO-1200, Nanjing Xianou Instrument Manufacturing Co., Ltd., China) for 10 min. Then, RGO/CB ink was obtained by centrifuging the mixture (H-1650, Hunan Xiangyi Laboratory Instrument Development Co., Ltd., China) at 8000 rpm for 15 min. After 30 days of storage at room temperature, no agglomeration and precipitation (Fig. 3b) were observed in the ink, indicating that the obtained RGO/CB ink had good stability at room temperature [33].

2.3 Preparation of flexible conductive circuits

An Epson Piezoelectric Inkjet Printer with the resolution of 5760×1440 dpi (R330, Epson Co., Ltd., China) was selected to print RGO/CB ink based flexible conductive wires. The substrate was glossy photo papers coated with polyvinyl alcohol (PVA) (A6, C13S041860, Epson Co., Ltd., China). On-demand printing mode was chosen. The printer has 90×5 color nozzles (cyan, magenta, yellow, light cyan, light magenta). The size of each nozzle is 30 µm, and a minimum droplet volume is 1.5 pl.

A bitmap file was designed using Adobe Illustrator (CS6, Adobe systems Inc.), and the pattern was printed using the original ink in the printer (Fig. 4a). After thoroughly syringing out the original ink with deionized water



Fig. 2 Schematic diagram for producing conductive ink



Fig. 3 Photographs of newly prepared ink (a) and ink after 30 days of storage at room temperature (b)



Fig. 4 Inkjet printed patterns using original ink in the printer (a) and RGO/CB ink (b)

and ethanol, the RGO/CB ink (60 ml) was put into the Continue Ink Supply System. The parameters of the printer were set as high quality image mode, Epson high quality glossy paper, high quality and gray print. Figure 4b showed the inkjet printed pattern with RGO/CB ink. The pattern is characterized by smooth lines, high gloss and uniform tone, demonstrating that the RGO/CB ink is inkjet printable with an Epson Piezoelectric Inkjet Printer.

In printed electronics, it is of great interest to print homogeneous narrow lines [20]. It is crucial for inkjet printed lines with continuous, uniform and compact ink layer to optimize the number of printing pass. Five lines $(50 \times 3,$ length \times width, mm) designed by Adobe Illustrator were printed on a piece of photographic paper (A6) by inkjet printing. One line was removed on the bitmap file (from top to bottom) after each printing on the photographic paper. Thus, the number of printing pass for five printed flexible wires from top to bottom were 1, 2, 3, 4, and 5, respectively. Then, the photographic paper with printed wires were dried in oven at 100 °C for 30 min.

2.4 Characterization of RGO/CB ink

The realization of printable inks made of graphene is a very challenging task, since the various rheological properties, such as density and viscosity, along with the nozzle size have a strong effect on the printing process [1]. The conductivity of RGO/CB ink was tested with a conductivity meter (DDSJ, Shanghai Yidian Scientific Instrument Co., Ltd., China). 100 ml of ink sample was first sonicated with an ultrasonic plant cell shredder for 10 min under the condition of 960 W, and then centrifuged with a speed of 8000 rpm for 15 min. Supernatant was removed and residue was analyzed with a laser particle size analyzer (Nano ZS, Malvern Instruments Co., Ltd., Britain) for particle size of the ink.

Viscosity of RGO/CB ink was determined according to GB1723-1993 (Determination of Viscosity of Coatings) with a James-4 viscometer (XND-1, Suzhou Jiangdong Presion Instrument CO., Ltd) at ambient temperature. Triplicates were run for conductivity, particle size, and viscosity of RGO/CB ink.

The solid content of the ink was tested according to the China's nation standard, "Determination of non-volatile adhesive content" (GB/T 2793-1995). The ink sample was put into a beaker (its initial weight was M1, g) and weighed (total weight of the beaker and the ink was M2, g), after oven drying at 100 ± 2 °C to a constant weight (total weight of the beaker and the ink sample was M3, g). The solid content of the ink was calculated as follows.

$$X = \frac{(M_3 - M_1)}{M_2 - M_1} \times 100\%$$
(1)

where X is the solid content (%).

Reagent resistance of RGO/CB ink was assessed according to Graphic Technology-prints and Printing Inks-assessment of Resistance to Various Agents (GB/T 18724-2008, ISO 2836: 2004, IDT). A printed conductive wire was cut into a size of 27×2 (length × width, mm). The agents were deionized water, sodium hydroxide (5 wt%), acetic acid (5 wt%), hydrochloric acid (5 wt%) and sulfuric acid (5 wt%). Test conditions for different agents were demonstrated in Table 1. First of all, four pieces of filter paper (Whatman 10300211, Shanghai Bestest Biological Technology Co., Ltd.) were completely immersed into one type of agent and dried in air until no agent droplets left on the saturated filter paper. Immersing time for each agent was shown in Table 1. Next, the printed wire sample was placed between four pieces of saturated filter papers, with two on each side. The "sandwich" was then put between two pieces of glass sheets, and testing set was done. A weight of 1 kg was put on the testing set. After being taken out from the set, printed wire was washed to neutral and oven dried at 40 °C for 30 min. Color change of tested printed wire sample compared with the control (untested printed wire), and surface contamination of filter papers caused by contacting with printed wire was observed and recorded carefully.

2.5 Characterization of flexible paper-based circuits

An optical microscopy (BX51, Olympus Optical Co., Ltd., China) was used to observe the microcosmic morphology of dried inkjet printed wires with different number of printing pass. Surface morphology and thickness of the inkjet printed wire was further investigated with a scanning electron microscope (SEM) (S-4800, Hitachi Transport Co., Ltd., Japan). After being coated with a thin layer of gold for 50 s at room temperature with an accelerating voltage of 3 kV.

The electrical conductivity (ρ) of the printed wire with a dimension of 10 × 2 (length × width, mm) was calculated by following equation:

$$\rho = \frac{RS}{L} \tag{2}$$

where *R* is the resistance at both ends of the printed wire, *L* is the length of the printed wire (10 mm), and *S* is the section area of the printed wire ($S = 2 \text{ mm} \times \text{the thickness}$ of the printed wire). *R* was characterized by a multimeter (VC9802A+, Shenzhen Yisheng Victory Technology Co.,

Table 1 Testing conditions of different agents

Testing agent	Accepted surface	Immersing time	Contact condition
Deionized water	Filter paper	24 h	54 cm ² , 1 kg
Sodium hydrox- ide		10 min	
Acetic acid		30 min	
Hydrochloric acid		10 min	
Sulfuric acid		10 min	

Ltd., China), and the thickness of the dried printed wire was obtained from SEM.

Uniformity test was conducted using a multimeter on a printed wire with four printing passes and a dimension of 50×2 (length × width, mm). On each printed wire, the number of testing points was 6, and the spacing between two adjacent points was 4 mm. Four replicates were tested in this part.

Printed wires with four printing passes and a dimension of 50×2 (length × width, mm) were used for adhesion test. An adhesive tape with a dimension of 19×32.9 (length × width, mm) (3M 600[#], Shanghai Cenxin industrial Co., Ltd., China) was stuck flatly on the ink layer surface for 1 min, and then torn off. Ink layer's falling-off was observed, and the resistance values of the wire at different intervals, were tested. Four replicates were done in this test.

Folding performance of the printed wire was evaluated with the resistance change after folding according to the method shown in Fig. 5. Sample wires with a dimension of 40 × 1 (length × width, mm) were prepared for folding tests. Initial resistance of the sample, R_0 , was tested with a multimeter. Then, the sample wire was folded perpendicularly outward to 180° and put into a ziploc bag, then a 2 kg weight was placed over the fold line for 1 min. After being taken out of the bag, the sample wire was folded 180° again along the folding line to the opposite direction. Put the wire into the bag, and pressed with a weight of 2 kg on the folding line for 1 min again. Resistance of the wire after folding, R_1 , was test. Aforementioned steps were repeated for four more times, in order to get R_2 , R_3 , R_4 , and R_5 with 2, 3, 4, and 5 folding times, respectively.

Sample wires with a dimension of 50×1 (length × width, mm) were prepared for bending tests. Initial resistance of the sample, R₀, was tested with a multimeter. Then, the wire was manually curled for two laps and held for 1 min, as shown in Fig. 6. After being flatten, the wire was put into a plastic ziploc bag with a piece of glass plate outside on top. A 2 kg weight was placed at the center of the glass plate for 1 min. Then the resistance, R₁, of the wire was tested. Aforementioned steps were repeated for four more times, in order to get R₂, R₃, R₄, and R₅ with 2, 3, 4, and 5 bending times, respectively.

3 Results and discussion

3.1 RGO/CB ink analysis

3.1.1 Basic performances of the ink

For the ink with an the best-suited formula (RGO:CB:ethanol:ethylene glycol:glycerol:CMC:deionized water = 96 mg:504 mg:12 ml:30 ml:30 ml:480 mg:51 ml),



Fig. 5 Schematic diagram for folding test



Fig. 6 Printed wire sample of the bending test

the electrical conductivity, average particle size, viscosity and solid content is 122.4 μ s/cm, 1.966 μ m, 22.5 mPa s and 0.4 wt % (equivalent to 4.31 mg/ml, ignoring the amount of solvent lost in the centrifugal treatment), respectively.

3.1.2 Resistance to various agents

Figure 7 showed the result of solvent resistance tests toward ink. Color of the samples that had been immersed in water and sodium hydroxide was faint, and some ink was left on the filter surfaces, indicating bad resistance to water and alkali. This is an common problem for water-based ink. For the RGO/CB ink, the –COOH group in RGO can react with the –OH group with the help of NaOH, resulting in bad resistance to alkali. After acid immersing test, both inkjet printed samples and contacting filter surfaces presented no apparent changes compared with the control, suggesting that the RGO/CB ink had good acid resistance.



Fig. 7 Solvent resistance test of inkjet printed lines

3.2 Flexible conductive wires

3.2.1 Effect of the number of printing pass on conductive property and morphology

Figure 8 presented optical microphotographs of dried inkjet printed wires with different number of printing pass (magnification, 200). When the number of printing pass was 1, large gaps between ink droplets appeared for the wire with four printing passes, as shown in Fig. 8a. It is caused by the tunneling effect, which is formed among those conductive particles dispersing not homogeneously in the substrate. As the number of printing pass became from 1 to 4, the density and continuity of the combination of ink and paper substrate increased gradually, forming current paths and a conductive network.



Fig. 8 Microphotographs of inkjet printed wires with different number of printing pass (number of printing pass: a-1, b-2, c-3, d-4, e-5)

Figure 9 showed the SEM micrographs of inkjet printed wires under different magnification. In Fig. 9a, b, uniform CB particles contact compactly with each other, forming a relatively dense conductive network. Furthermore, those CB particles are evenly distributed around the fluffy, transparent gauze-like edges and on the surface of graphene sheets. When the number of printing pass increased from 1 to 4, the color of the ink layer became from light gray to black, and the resistance decreased from 1.43 to 0.1 M Ω , as shown in Fig. 10. The resistance further dropped to 0.09 M Ω with five printing passes, which demonstrated that a more continuous and stable conductive layer was formed by dense and uniform distribution of RGO/CB particles on the paper surface. However, the inkjet printed wire with five printing passes dried very slowly, just as found in the test. Thus, the authors believe that it is reasonable to set the number of printing pass to 4.

Figure 11 showed side SEM images of the printed wires with four printing passes (a) and paper itself without any ink layer (b). When the ink was inkjet printed onto the photo paper with a PVC coated layer (19.3 μ m), the ink would penetrate into the coating, expanding it and increasing its thickness (34.2 μ m for four printing passes). It was also found that the thickness of the ink layer was 33.38 μ m, and the resistance and resistivity at both ends of this ink layer (10 mm × 2 mm × 33.38 μ m, length × width × thickness)



Fig. 9 SEM edge micrographs of inkjet printed wire surface with four printing passes (folds. a—20 k, b—40 k, c—100 k)



Fig. 10 Effect of the number of printing pass on resistance of printed conductive wires

were $9.9 \times 10^4 \Omega$ and 0.661 Ω m, respectively. Both thickness increase per pass and resistivity are higher than the results of Romagnoli et al. [19]. The large thickness increase is mainly due to the high solid content of RGO/CB ink (4.31 mg/ml) and average particle size in RGO/CB ink (1.966 µm). The low conductivity of the RGO/CB ink layer can be attributed to the micro-porous photo paper substrate and the mild later heat treatment condition, 100 °C for 30 min.

3.2.2 Uniformity of ink in printed wires

Figure 12 showed the resistance at different points on an inkjet printed wires with four printing passes. When the distance from the start point (one end of the wire) was 10, 20, 30, 40 and 50 mm, the resistance was 0.107, 0.186, 0.262, 0.347 and 0.443 M Ω , respectively; the corresponding calculated electrical conductivity was 0.714, 0.621, 0.583, 0.579, 0.591 Ω ·m according to Eq. (2), respectively The resistance presented a linear relationship versus the

length, indicating that the conductive ink was evenly distributed in the printed wire. This phenomenon was supported by the SEM images of the wire cross section in Fig. 9, and was consistent with the result of Han [33].

3.2.3 Adhesion of the printed wires analysis

The adhesion resistance of conductive wires with four printing passes was shown in Table 2. After adhesion test, the ink layer on paper was intact, with no ink on tape. Furthermore, after adhesion test, the resistivity only increased 3.2, 3.2, 3.0, 2.9 and 2.8% for 10–50 mm, respectively. It indicates that the RGO/CB ink has good adhesion to meet the requirements of photo paper-based flexible printed wires. This is mainly due to the presence of CMC in the G/CB ink, which is not only an ink binder to ensure good bonding between the ink layer and the paper substrate surface, but also a graphene stabilizer to act as a bridge between graphene and the substrate. Moreover, the hydrophilic PVC layer on paper surface can enhance the adhesion of conductive ink to photographic paper.

3.2.4 Folding and bending of printed wires

Figure 13 gave the initial resistance of the inkjet printed wire with four printing passes before folding and the resistance values after different folding times. The initial resistance was 0.338 M Ω . With the increase of folding times, the resistance of the wire presented a slightly ascending trend. After five folding times, the printed wire showed a resistance of 0.588 M Ω , which was still a small value. And, in the bending test, the resistance of the wires presented little resistance increase after bending eight times, revealing a good mechanical flexibility of the wire. The good folding and bending performance of inkjet printed



Fig. 11 Side view FE-SEM images of inkjet printed wire with four printing passes (a) and the paper itself without ink deposition (b)



Fig. 12 Resistance change of the inkjet wire with the measured length

wires is also due to the presence of the binder, CMC, in RGO/CB ink, which can help conductive particles to fully contact with the substrate surface, and then promote the interface bonding between ink and paper substrate.

 $\label{eq:table_$

Length (mm)	Pre-adhesion resistance (M Ω)	After-adhesion resistance (MΩ)	Percentage of increase
10	0.31 ± 0.02	0.32 ± 0.01	3.2
20	0.31 ± 0.01	0.32 ± 0.02	3.2
30	0.33 ± 0.01	0.34 ± 0.01	3.0
40	0.34 ± 0.01	0.35 ± 0.01	2.9
50	0.36 ± 0.01	0.37 ± 0.01	2.8

Resistance value was reported as average ± standard deviation

3.2.5 Light-emitting operation with inkjet printed RGO/CB ink wires

Figure 14 exhibited an application example of the paperbased circuit with four printing passes. The system consisted of two metal wires, a 20×2 (length × width, mm) inkjet printed paper-based conductive wire, a LED



Fig. 13 Effect of folding times on the resistance of wires

(LMY-5MMB2R02, 20 mA, 3–3.4 V, Shenzhen City Green Margin Photoelectric Co., Ltd., China) and a power supply (3 V). When the circuit was closed, LED was lightened, stating clearly that the ink formed a conductive path on the paper substrate. The flexible paper-based three-dimensional conductive wires prepared in this paper showed a great application potential in electronics.

4 Conclusions

In this paper, RGO/CB ink with a high RGO concentration and its inkjet printed flexible electronic wires were prepared on glossy photo paper. Main results were as follows:

When the loads of RGO, CB, ethanol, ethylene glycol, glycerol, CMC and deionized water were 96 mg, 504 mg, 12 ml, 30 ml, 30 ml, 480 mg and 51 ml, the electrical conductivity, average particle size and viscosity of the RGO/CB ink was $122.4 \,\mu$ s/cm, $1.966 \,\mu$ m and $22.5 \,m$ Pa s, respectively. The ink has good stability, uniformity, and resistance to acid.

The resistance at both ends of the dried inkjet printed wire $(10 \text{ mm} \times 2 \text{ mm} \times 0.03338 \text{ mm})$ and the resistivity of the ink layer with four printing passes were 0.1 and 0.661 Ω m, respectively. The results of adhesion, folding and bending tests shows that the adhesion performance and mechanical flexibility of the inkjet printed wires are strong.

In the light-emitting operation, the inkjet printed flexible paper-based three-dimensional conductive circuits works well, showing a great application potential in electronics.



Fig. 14 Application example of flexible paper-based conductive circuit system (a-circuit open, b-circuit closed)

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References

- A. Capasso, A.E.D.R. Castillo et al., Solid State Commun. 224, 53–63 (2015)
- A.C. Siegel, S.T. Phillips, M.D. Dickey et al., Adv. Funct. Mater. 20, 28–35 (2010)
- 3. K. Kim, S.I. Ahn, K.C. Choi, Carbon 66, 172-177 (2014)
- S. Kim, B. Cook, T. Le et al., IET Microware Antennas Propag. 7, 858–868 (2013)
- M. Vaseem, K.M. Lee, A. Hong et al., ACS Appl. Mater. Interfaces 4, 3300–3307 (2012)
- 6. S. Hurch, H. Nolan, T. Hallam et al., Carbon **71**, 332–337 (2014)
- Y. Gao, W. Shi, W. Wang et al., Ind. Eng. Chem. Res. 53, 16777– 16784 (2014)
- T. Takenobu, N. Miura, S.Y. Lu et al., Appl. Phys. Express 2, 0255005 (2009)
- 9. G. Cummins, M.P.Y. Desmulliez, Circuit World **3**, 193–213 (2012)
- R. Giardi, S. Porro, A. Chiolerio et al., J. Mater. Sci. 48, 1249– 1255 (2013)
- 11. E.S. Snow, J.P. Novak, D. Park et al., Appl. Phys. Lett. 82, 2145 (2003)
- 12. A. Kamyshny, J. Steinke, S. Magdassi, Open Appl. Phys. J. 4, 19–36 (2011)
- 13. M. Ha, Y. Xia, A.A. Green, et al., ACS Nano 4, 4388–4395 (2010)
- M. Ha, J.T. Seo, P.L. Prabhumirashi et al., Nono Lett. 13, 954–960 (2013)

- B. Kim, S. Jang, P.L. Prabhumirashi et al., Appl. Phys. Lett. 103, 082119 (2013)
- V. Singh, D. Joung, L. Zhai, et al., Prog. Mater Sci. 56, 1178–1271 (2011)
- 17. P. Avouris, Z. Chen, Nat. Nanotechnol. 2, 605–615 (2007)
- 18. E.B. Secor, M.C. Hersam, J. Phys. Chem. Lett. 6, 620 (2015)
- M. Romagnoli, M.L. Gualtieri, M. Cannio et al., Mater. Chem. Phys. 182, 263–271 (2016)
- 20. J. Li, F. Ye, S. Vaziri et al., Adv. Mater. 25, 3985-3992 (2013)
- 21. A. Lerf, H. He, M. Forster et al., J. Phys. Chem. B **102**, 4477 (1988)
- 22. D. Kong, L.T. Le, Y. Li et al., Langmuir 28, 13467-13472 (2012)
- 23. M.J. Allen, V.C. Tung, R.B. Kaner, Chem. Rev. 110, 132-145
- (2010)
- 24. A.A. Green, M.C. Hersam, J. Phys. Chem. Lett. 1, 544–549 (2010)
- F. Torrisi, T. Hasan, W. Wu et al., ACS Nano 6, 2992–3006 (2012)
 D. Finn, M. Lotva, G. Cunningham et al., J. Mater. Chem. C 2.
- 26. D. Finn, M. Lotya, G. Cunningham et al., J. Mater. Chem. C 2, 925–932 (2014)
- 27. E.B. Secor, P.L. Prabhumirashi et al., J. Phys. Chem. Lett. 4, 1347–1351 (2013)
- S. Shukla, K. Domican, K. Karan et al., Electrochim. Acta 156, 289–300 (2015)
- 29. C. Svanberg, T. Pham, M.A. Malik et al., US Patent EP2374842 (2013)
- W.S. Hummers Jr, R.E. Offeman, J. Am. Chem. Soc. 80, 1339 (1958)
- Y. Xu, H. Bai, G. Lu et al., J. Am. Chem. Soc. 130, 5856–5857 (2008)
- 32. D. He, L. Shen, X. Zhang et al., AIChE J. 60, 2757-2764 (2014)
- 33. J.W. Han, B. Kim, J. Li et al., Mater. Res. Bull. **50**, 249–253 (2014)