Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

Graphene Oxide Scroll meshes Encapsulated Ag Nanoparticles for Humidity Sensing

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Experimental Section

1. Materials

Acetone, toluene, dichloromethane, concentrated sulfuric acid and nitric acid were obtained from Shanghai Ling Feng Chemical Reagent Co., Ltd. 99.99 % expanded graphite (32 mesh) and 99.9% potassium permanganate were bought from Zhong Nuo New Material Technology Co., Ltd. Silver nitrate was obtained from Xiya Reagent Co., Ltd. Octadecyltrimethoxysilane (OTS) was bought from Shanghai Aladdin biochemical Polytron Technologies Inc. Silicon wafers were purchased from Jing Xi Co., Ltd (Su Zhou, China).

2. Synthesis

2.1 Synthesis of large sized graphene oxide (GO) nanosheets

The GO nanosheets were prepared using modified Hummer's method.[1, 2] The GO-Ag NPs nanosheets were fabricated as previously reported.[3]

2.2 Preparation of OTS-Modified SiO_2 Substrates

Firstly, SiO_2/Si substrate was cleaned by ultrasonication in mixture of distilled water and acetone (1:1 in volume) for 30 min, followed by heating at 120 ℃ in mixture of H_2O_2 and H_2SO_4 (1:1 in volume) for 30 min. Then the substrate was dried by nitrogen and immersed in a 10 mM OTS toluene solution for 24 h, rinsed with ethanol and water, and dried under nitrogen.

2.3 Fabrication of GO and GO/Ag NPs scroll by molecular combing

The OTS-SiO_2/Si substrate was put on a filter paper. Then 5 μL GO-Ag NPs solution with a concentration at 0.3 mg/mL was deposited on one side of the OTS-SiO_2/Si substrate. A cover slip (22 ×22 mm^2) was used to drag GO-Ag NPs solution at an angle of 45°on the hydrophobic OTS-SiO_2/Si substrate from one side to another side. Excessive GO/Ag NPs solution was absorbed by a filter paper. The combing process took 3 seconds to complete on the OTS-SiO_2/Si substrate (1 ×1 cm^2). Thus, GO-Ag solution spread on the hydrophobic substrate to form the horizontally aligned GO-Ag scrolls due to the capillary interaction.[4] The vertically aligned GO-Ag scrolls arrays were obtained by rotating 90° of the substrate. The GO scroll was fabricated in the same process.

2.4 Fabrication of GO and GO-Ag scroll meshes

In brief, PMMA is spin-coated on the prepared OTS-SiO_2/Si substrate with horizontally aligned GO or GO-Ag scroll deposited on it. About 1 mm wide polymer strips at the edge of the OTS-SiO_2/Si substrate are then scratched off to expose the hydrophobic OTS-SiO_2/Si surface. Then a thick PDMS film is brought into conformal contact with the polymer film and the horizontally aligned GO-Ag or GO scrolls are left on the thick PDMS film by peeling off PDMS film from substrate. Next, the thick PDMS film with horizontally aligned GO-Ag or GO scrolls is brought into a conformal contact with another OTS-SiO_2/Si substrate where perpendicularly aligned GO-Ag or GO scrolls are pre-deposited. PMMA film with cross-stacked GO-Ag or GO scrolls is left on OTS-SiO_2/Si substrate by slowly peeling off PDMS film. Followed by dissolving PMMA film in dichloromethane (DCM) at 50℃ for several

seconds, GO or GO-Ag scroll meshes are left on the OTS-SiO_2/Si substrate.

2.5 Device fabrication

As-prepared GO or GO-Ag scroll meshes formed on OTS-SiO_2/Si substrate were reduced subsequently by hydrazine vapor at 80℃ overnight. Followed by thermal annealing at 300℃ for 2-3 h to further remove the PMMA residue, silver paste was deposited on the diagonal corners of GO or GO-Ag scroll meshes to obtain a device.

2.6 Humidity test

The real-time current change through the device at different humidity was performed by a Keithley 4200 semiconductor characterization system. Saturated LiCl, $MgCl_2$, $Mg(NO_3)_2$, NaCl and K_2SO_4 solutions were selected as standard humidity solution (11%, 33%, 52%, 75% and 97% RH, respectively) for humidity detecting at room temperature. The prepared devices were initially purged with compressed N_2 for about half an hour to get a stable baseline current. Later, rGO-Ag scroll meshes were exposed to humid air with different RH for ~1 min followed by ~1 min of compressed N_2 purging. While rGO scroll meshes were exposed to humid air with different RH for ~20 s followed by ~80 s of compressed N_2 purging.

3. Characterization

3.1 Atomic force microscopy (AFM) characterization

A commercial AFM instrument (Dimension ICON with Nanoscope V controller, Bruker) equipped with a scanner (90 × 90 μm^2) was used to image the samples in tapping mode in air.

3.2 Scanning electron microscopy (SEM) characterization

Morphology of GO and GO-Ag scrolls was captured by using a JEOL JSM-6700 field-emission scanning electron microanalyzer at accelerating voltage of 5 and 10 kV, respectively.

3.3 Transmission electron microscopy (TEM) characterization

GO and GO-Ag scrolls were imaged by using a JEM-1200EX transmission electron microscope (JEOL, Japan) operated at an accelerating voltage of 100 kV.

3.4 UV/vis spectrum characterization

The UV/vis spectrum (100–800 nm) was measured using a UV950 Lamda UV spectrophotometer. Aqueous solutions were used for UV measurement after ultrasonic processing. Pure millipore water was used as the background.

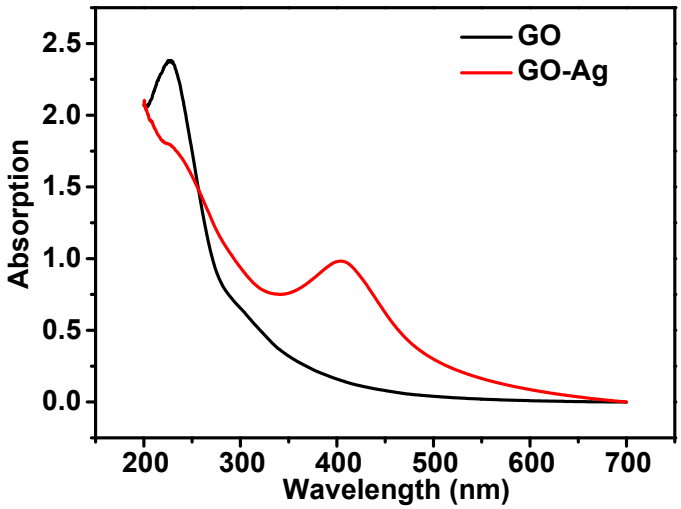

Figure S1. The UV/vis absorption spectra of aqueous solutions of GO (black line) and GO-Ag nanosheets (red line).

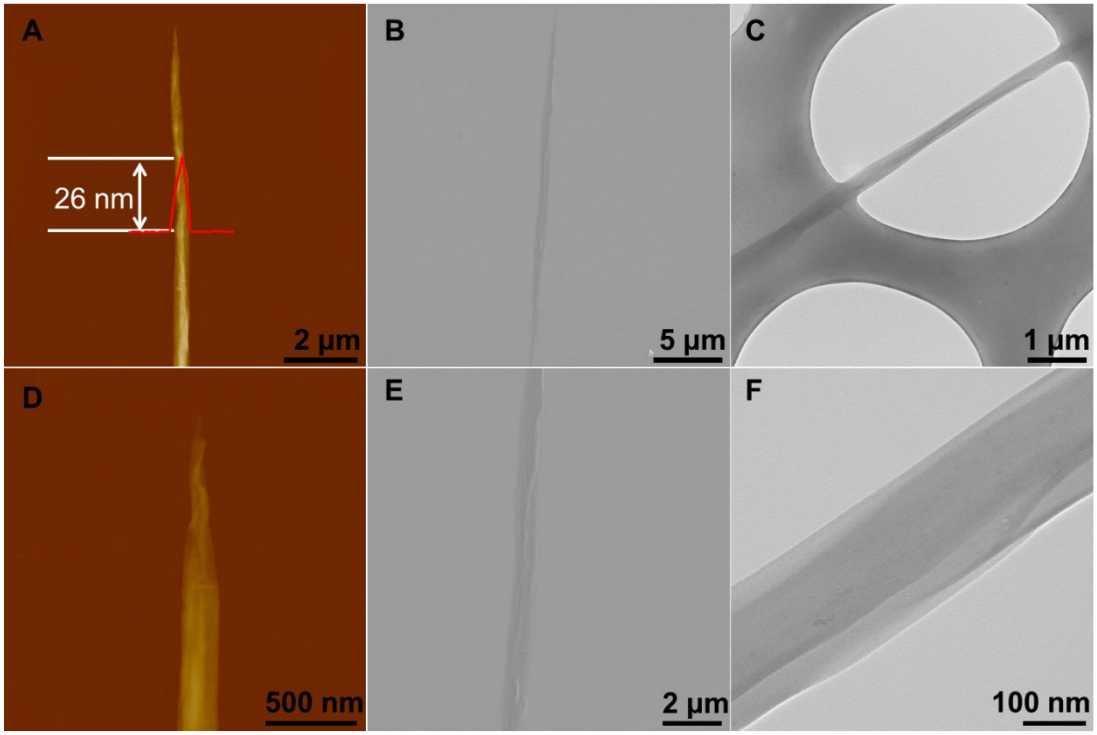

Figure S2. AFM (A, D), SEM images (B, E) and TEM images (D, F) of GO scroll prepared by molecular combing.

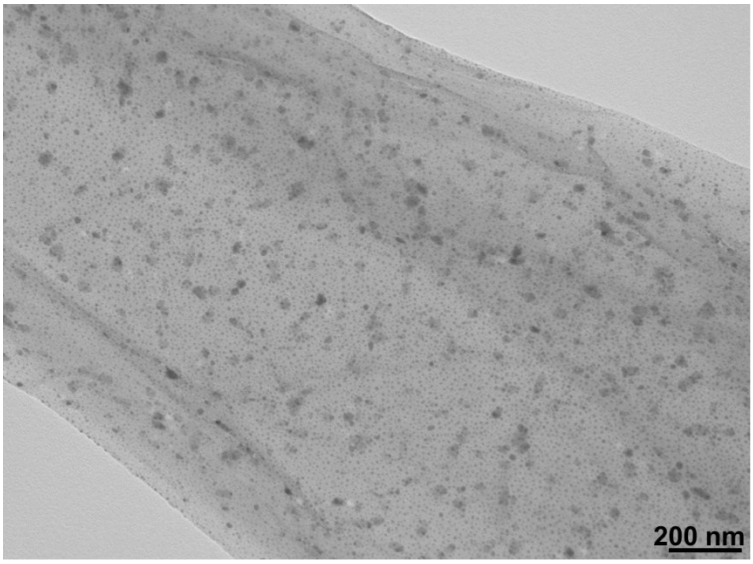

Figure S3. TEM image of dense Ag NPs homogeneously distributed on GO-Ag scroll.

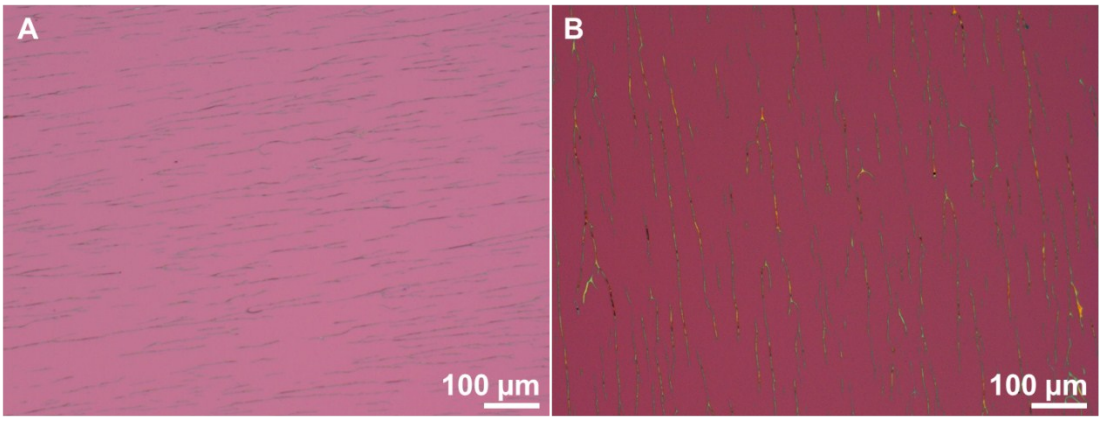

Figure S4. OM images of horizontally (A) and perpendicularly (B) aligned GO-Ag scrolls array by molecular combing.

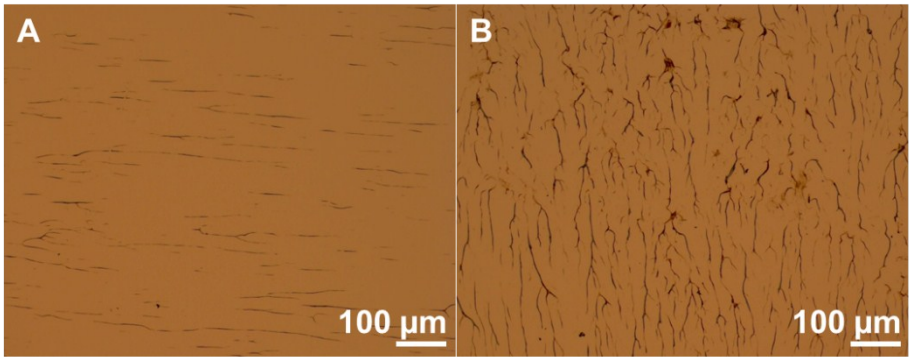

Figure S5. (A) Long straight GO-Ag scrolls array formed by molecular combing GO-Ag solution at a concentration of 0.08 mg/mL. (B) Dendritic GO-Ag scrolls array formed by molecular combing GO-Ag solution at a concentration of 0.3 mg/mL.

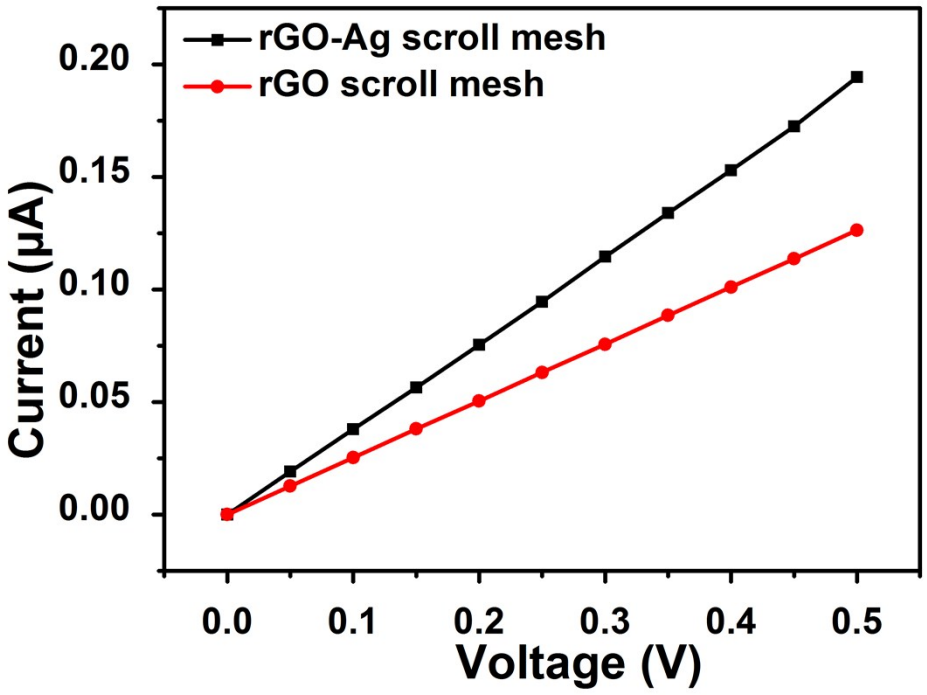

Figure S6. The conductivities of rGO and rGO-Ag scroll meshes-based devices.

1. C. Lee, X. D. Wei, J. W. Kysar and J. Hone, *Science*, 2008, **321**, 385-388.
2. X. L. Li, X. R. Wang, L. Zhang, S. W. Lee and H. J. Dai, *Science*, 2008, **319**, 1229-1232.
3. X. Z. Zhou, X. Huang, X. Y. Qi, S. X. Wu, C. Xue, F. Y. C. Boey, Q. Y. Yan, P. Chen and H. Zhang, *J Phys Chem C*, 2009, **113**, 10842-10846.
4. H. Li, J. Wu, X. Y. Qi, Q. Y. He, C. Liusman, G. Lu, X. Z. Zhou and H. Zhang, *Small*, 2013, **9**, 382-386.