Rolling up graphene oxide sheets into micro/nanotubes by nanoparticle aggregation

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Experimental Section

Materials: Graphite powder (500 mesh) was obtained from Beijing Chemical Reagents. $AgNO_3$, $KMnO_4$, $NaNO_3$, NaOH, concentrated H_2SO_4, concentrated HCl, H_2O_2 (30%) were all analytical-grade and purchased from Shanghai Sinopharm Chemical Reagent Co., Ltd. All aqueous solutions were prepared with ultrapure water (18 MΩ).

Preparation of GO: GO was produced by the modified Hummers method through acid oxidation of flake graphite. The primary product was suspended in water under sonication for one hour, followed by centrifugation at 4000 rpm for 30 min and dispersed in the water. The obtained yellow-brown aqueous suspension of GO was stored at room temperature for further characterization and chemical reduction.

Synthesis of Ag nanoparticle-decorated GO sheets: In a typical experiment, 5 mL $AgNO_3$ (10 Mm) aqueous solution was gradually added into 5 mL GO (0.5 mg/mL) aqueous solution under vigorously stirring at room temperature for 10 min. Subsequently, the mixture was kept statically under ambient conditions for 48 h. The resultant mixture was then separated from the solution by centrifugation at 6, 000 rpm for 20 min and washed with ultra-pure water twice.

Synthesis of Fe_3O_4 nanoparticle-decorated GO sheets: Fe_3O_4 nanoparticle-decorated GO sheets were prepared by co-precipitaion method with a ferrous complex in presence of GO. Firstly, $FeCl_2.4H_2O$ and $FeCl_3.6H_2O$ [Fe^{2+}: Fe^{3+}=1:2] were dissolved in 50 mL deionized water with

vigorously stirring at room temperature. Secondly, added 0.5 mg/mL aqueous solution of GO (5 mL) to the above solution, then heating solution to 80℃. Thirdly, this iron solution source was added drop-wise into NH_4OH under vigorously stirring for 30 minutes, and bubbling N_2 gas. Dark brown mixture of Fe_3O_4 nanoparticle-decorated GO sheets and Fe_3O_4 nanoparticles were collected by permanent magnet and cleaned by deionized water several times. Finally, Fe_3O_4 nanoparticle-decorated GO sheets were separated from the reaction mixture by filtration. Fe_3O_4 nanoparticle-decorated GO sheets were washed for three times with deionized water, and were dried finally at 40 ℃ for 24 hours under vacuum.

Rolling of inorganic nanoparticle-decorated GO sheet: The decorated GO sheets (in aqueous solution) were sonicated for 2 h using an ultrasonic reactor operated at a frequency of 33 kHz (100 W). The resultant solution was dried under reduced pressure for further characterization.

Microstructural characterizations: The AFM images of GO, Ag-decorated GO sheets, Ag-GO hybrid tubes were taken on a Multimode Nanoscope V scanning probe microscopy system (Veeco, USA). The commercially available AFM cantilever tips with a force constant of ~48 N/m and resonance vibration frequency of ~330 kHz were used. The SEM images were recorded on a ZEISS-ULTRA 55 SEM equipped with an X-ray EDS at an accelerating voltage of 20 KV. TEM images were acquired using a JEM-2010 TEM with an accelerating voltage of 200 kV.

Fig. S1 (a) SEM image of massive wire-like structures of Ag-GO hybrids. Inset is the magnification picture of individual one. (b) EDS analysis of the Ag-GO hybrids distributed on Si substrate. C, O and Ag elements are revealed, suggesting the co-existence of Ag and Graphene Oxide in the hybrid scrolls.