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One-Step and In-situ Decoration of High-Quality Graphene with Transition Metal Oxide Nanoparticles

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Graphene, as a new member of carbon allotropes family, has exhibited potential applications due to its unique electronic and mechanical properties [1-2]. In view of its highly specific surface area [1] and super electrical conductivity [2], graphene is being considered as an ideal template for loading transition metal oxides, which could manufacture various graphene-based composites for energy-related electrochemical devices [3]. In this abstract, monodispersed magnetic iron oxide nanoparticles (NPs) have been successfully deposited onto the surface of exfoliated graphene by a simple solvothermal process at a temperature of 180 °C, using iron (II) acetylacetonate and expanded graphite as precursor, and using anhydrous ethanol as the solvent. The transmission electron microscopy (TEM) micrographs show the graphene/Fe_3O_4 composites prepared by a solvothermal process at 180 °C for 4h, 8h, and 16h respectively, in Fig. 1a-c. The results indicate that the particle size and coating density can be tailored by varying reaction times. The high resolution EM image (HREM) confirmed that the attached Fe_3O_4 NPs are well crystallized, the lattice spacing was about 0.24 nm, 0.29 nm (Fig. 1d) and 0.29 nm (Fig. 1e) which are correspondent with the (-2-22), (2-20) and (220) plane in a cubic spinel Fe_3O_4. The correspondent selected area electron diffraction pattern (SAED) from the graphene/Fe_3O_4 composites shows the diffraction rings which are coincident with the graphene (002) lattice plane and magnetite Fe_3O_4 phase, (220), (311), (400), (511), (440), from inner to outside respectively, as shown in Fig. 1e (insert). It further confirms the formation of Fe_3O_4 on the graphene. Fig. 1f shows the typical Energy-dispersive X-ray spectroscopy (EDX) pattern of deposited Fe_3O_4 NPs.

Our results demonstrate that this simple and environmental-friendly chemical process, in general, can be used to combine graphene and other transition metal oxide NPs, such as Mn_3O_4 (Fig. 2) and CoO by changing manganese (II) acetylacetonate or cobalt (II) acetylacetonate as precursor. Fig. 2a-b shows the micrographs taken from the graphene/Mn_3O_4 composites prepared by a solvothermal process at 200 °C for 8h and 16h respectively. The results indicate that the particle size increase from 10 nm to 25 nm when reaction times extends. HREM images also confirmed that the deposited Mn_3O_4 NPs are well crystallized, the lattice spacing was about 0.25 nm, 0.28 nm (Fig. 2c) and 0.49 nm (Fig. 2d) which are correspondent with the (1-10) (200) and (112) plane in spinel Mn_3O_4. The inserted SAED image shows the diffraction rings, are from the graphitic carbon (002) plane and Hausmannite Mn_3O_4 phase, (112), (103), (211), (220), (105), (321) planes.

To investigate the potential application of graphene/Mn_3O_4 composite as the electrode material for a supercapacitor, as well as graphene/Fe_3O_4 and graphene/CoO composites in Lithium ion batteries, further work will be explored for measurement the cyclic voltammetry curves, galvanostatic charge/discharge curves and cycle stability.

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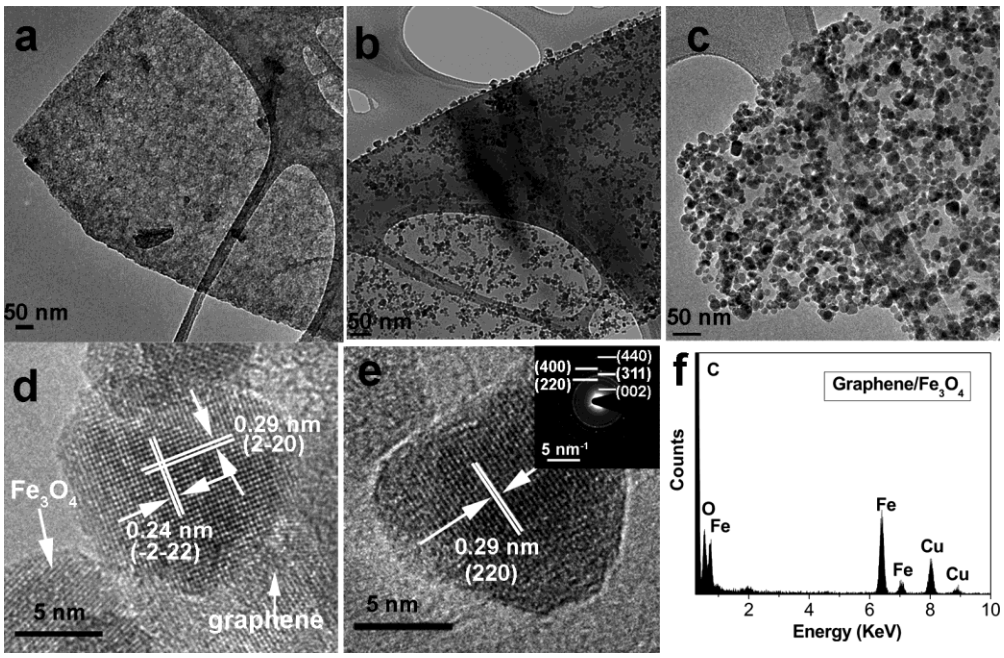

FIG. 1. TEM analysis of graphene/Fe_3O_4 composite prepared by a solvothermal process at 180 ℃ . (a)-(c) the reaction time for 4h, 8h, 16h respectively; (d)-(e) HREM micrographs of deposited Fe_3O_4 NPs and the corresponding SAED pattern (insert); (f) EDX pattern of the deposited Fe_3O_4 NPs.

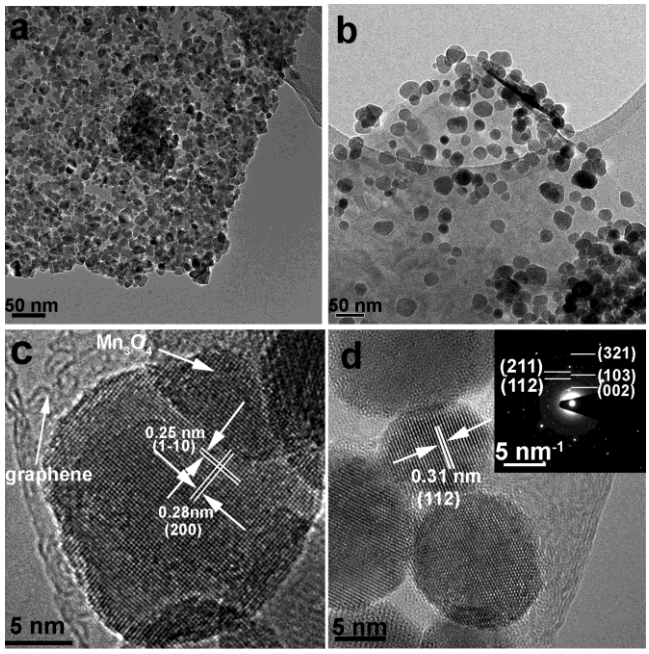

FIG. 2. TEM analysis of graphene/Mn_3O_4 composite prepared by a solvothermal process at 200 ℃ . (a)-(b) the reaction time for 8h and 16h respectively; (d)-(e) HREM micrographs of deposited Mn_3O_4 NPs and the corresponding SAED pattern (insert).

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