Carbon Nanotubes−Magnetite Nanocomposites from Solvothermal Processes: Formation, Characterization, and Enhanced Electrical Properties

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Received January 2, 2003. Revised Manuscript Received May 2, 2003

Carbon nanotubes (CNTs)−magnetite nanocomposite with 20−30-nm magnetite particles attached onto CNTs has been successfully prepared for the first time by in situ solvothermal synthesis from the precursor of Fe−urea coordination complex ($Fe[(NH_2)_2CO]_6(NO_3)_3$) and CNTs. The effects of CNTs and various processing parameters on ultimate composites characteristics were investigated. It has been found that pure CNTs−magnetite nanocomposite was obtained in an ethylenediamine medium at 200 °C for 50 h with the weight ratio of $Fe[(NH_2)_2CO]_6(NO_3)_3$:CNTs = 10:1. XRD, BET, TEM, EDS, and a Mössbauer spectrum were used to characterize the final product. A possible formation mechanism of the CNTs−magnetite nanocomposite was suggested. It has been concluded that a suitable amount of water introduced by CNTs is critical for preparing CNTs−magnetite nanocomposite. The addition of CNTs in the composite increased the electrical conductivity by about 32% from 1.9 to 2.5 S cm^{-1}, compared with the composite without CNTs. The relative density measurement and microstructure characterization performed by SEM showed that the percolation effect of CNTs and a good dispersion of CNTs in the matrix would lead to an effective and obvious improvement on the electrical conductivity.

Introduction

Since the discovery of CNTs in 1991 by Iijima,[1] their unique structural, electrical, mechanical, electro-mechanical, and chemical properties have triggered a worldwide research effort.[2−4] At present, CNTs are available in kilogram quantities with a price that is acceptable compared to that at the initial stage of their discovery. It costs about 2−20 U.S. dollars per gram for multiwalled carbon naotubes (MWCNTs), depending on the amount purchased. It makes the large-scale production of CNTs composites possible and practical. Theoretical and experimental results showed superior electrical properties of CNTs: electric-current-carrying capacity 1000 times higher than copper wires.[5] Therefore, it can be foreseen that CNTs could be used as additives to improve electrical properties of nanocomposite materials.

In recent years, a lot of research work on CNTs composites has concentrated on polymer−matrix materials for improved electrical conductivity,[6] enhanced optical properties,[7] and higher strength.[8] Andrews et al.[9] dispersed 5 wt % single-walled nanotubes (SWCNTs) in isotropic petroleum pitch and found the electrical conductivity increased by 340%. Other researchers[10−13] synthesized CNTs−metal or CNTs−reinforced ceramic matrix composites. Preliminary investigations by Chen et al.[14] focused on synthesis of CNTs−metal composites by electroless plating, and Xu et al.[15] showed slight improvements on the electrical conductivity of alumina with increasing nanotube volume fraction. Peigney et al.[16] synthesized CNTs−metal-oxide nanocomposites by hot-pressing the corresponding composite powders. It has been found that the composites were electrical conductors owing to the percolation of the nanotubes. In this study, CNTs−magnetite nanocomposite has been first prepared by in situ formation. It is expected that CNTs could offer tremendous opportunities for the

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10.1021/cm030007q CCC: $25.00

electrical property development of fundamentally novel CNTs−magnetite materials.

The effective utilization of CNTs in composite applications depends strongly on the homogeneous dispersion of CNTs throughout the matrix. CNTs−SiC composites have been prepared by Ma et al.,[17] who mixed MWCNTs with SiC powder and then hot-pressed the mixture. These authors reported an improvement of about 10% over monolithic SiC both in bending strength and fracture toughness. Peigney et al.[18,19] reported a novel catalytic route for the in situ formation, in a composite powder based on alumina and SWCNTs or MWCNTs. The microstructure and mechanical properties of dense CNTs−Fe−Al_2O_3 materials prepared by hot-pressing the nanocomposite powders have been investigated.[13,20] It has been shown that CNTs confer electrical conductivity to ceramic−matrix composites, which retain the mechanical properties of the ceramics. So in situ synthesis may be an effective way to obtain homogeneous composites.[10−13,16] Among the various methods of preparing magnetite powders, hydrothermal or solvothermal preparation is one very promising chemical controlling method, which is critical for controlling particle size, morphology, and size distribution in synthesizing and processing of the iron oxide particles.[21] In this paper, a novel simple solvothermal method is successfully employed to in situ synthesis of CNTs−magnetite nanocomposites from the precursor of Fe(III)−urea coordination compound and CNTs. The effects of the state of CNTs, various processing parameters, such as reaction time, reaction temperature, and reactant proportion, on ultimate nanocomposite characteristics are studied. In particular, the electrical properties, as well as the microstructures, of the nanocomposites have been systematically investigated.

Experimental Section

Multiwall carbon nanotubes prepared by the catalytic decomposition of CH_4 [22] were kindly provided from Shenzhen Nanoport Company. They were washed in concentrated acids to remove the catalytic metals effectively. Thus, the original CNTs supplied by the manufacturer are wet. Simultaneous thermogravimetry (TG)-differential scanning calorimetry (DSC) (STA 449C, NETZSCH, Selb, Germany) was used to determine the water content of the original CNTs under the nitrogen gas flow of 20 mL/min at a heating rate of 10 °C/min, which were shown in Figure 1. In the corresponding DSC curve, the endothermic peak appears around 115 °C due to the release of water. According to TG analysis, the original wet CNTs contain about 45 wt % water. After water removal, they have a purity >95% as claimed by the manufacturer. The density of the multiwall carbon nanotubes is approximately 2.1 g cm^{-3} according to the producer. Figure 2 shows the TEM and HRTEM images of CNTs,[23] which have lengths ranging from several to tens of micrometers (Figure 2a) with an inner diameter around 10 nm (Figure 2b). Figure 2c clearly shows that the nanotubes are multiwalled, with ≈25−30 graphene layers.

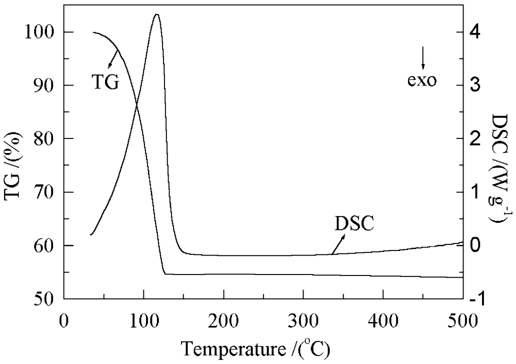

Figure 1. TG and DSC curves of the original wet CNTs.

In the most conventional synthesis of magnetite, the experiment would not succeed until three conditions are realized: deoxygenated protection, Fe(II)/Fe(III) molar ratio control, and basic conditions.[21,24,25] A novel simple solvothermal method introduced here synthesized magnetite from the precursor of Fe(III)−urea complex through CNTs-combined reaction, without respect to the above three conditions. Fe(III)−urea complexes were prepared on the basis of Worzala's method.[26] Fe(III) nitrate ($Fe(NO_3)_3 \cdot 9H_2O$, >98.5%) was dissolved in anhydrous ethanol to obtain concentrated solution. Then the solution was added dropwise into saturated urea/ethanol solution at 75−80 °C with stirring, until the final salt/urea molar ratio reached 1:9. The reaction gave water-soluble precipitation of Fe(III)−urea complex, which was separated by filtering and then dried at 80 °C. It was characterized without further purification. In a typical experiment, 0.2 g of CNTs were dispersed in 10 mL of ethylenediamine ($C_2H_8N_2$, ≥99.0%) by ultrasonication. Then the suspension and 2.0 g of complex were mixed together with 20 mL of ethylenediamine in a 40-mL Teflon-lined autoclave. The autoclave was maintained at 200 °C for 50 h and then cooled to room temperature naturally. A black precipitate was obtained by filtration. After further drying and grinding, CNTs−magnetite nanocomposite powders were obtained. For comparison, the same preparation processes were applied while varying the reaction time, reaction temperature, and complex/CNTs mass ratio.

The precursor of Fe(III)−urea complex was determined by chemical and elemental analysis (Vario El, Elementar, Germany). The structure was characterized by Fourier transformation infrared spectroscopy (FTIR, NEXUS, Nicolet, USA). TG-DSC measurement was also used to record the thermal decomposition of Fe(III)−urea nitrate under nitrogen gas flow of 20 mL/min at a heating rate of 10 °C/min.

The X-ray diffraction patterns of the synthesized composites were obtained by powder X-ray diffraction (XRD, D/Max 2550V, Rigaku, Japan), using Cu Kα radiation ($\lambda = 1.5418$ Å). Mössbauer spectroscopic studies (MS, Series 40 MCA, Canberra, USA) for the obtained composites were performed using a source of ^{57}Co in a Pd matrix at room temperature. Hyperfine interaction parameters were derived from the Mössbauer spectra using a least-squares method. The spectrometer was calibrated using a standard 25-μm α-Fe foil. The specific surface area of the nanocomposite powders was measured by nitrogen adsorption using a surface area analyzer (ASAP 2010, Micrometrics, USA). Transmission electron microscopy (TEM, JEM 2010, JEOL, Japan) was performed to observe the microstructure of the composites. Energy-dispersive spectrocopy (EDS) was also taken on the same apparatus.

The three different composite powders without and with CNTs were pressed into Φ20 × 1 mm pellets by applying a 20 N force and then sintered at 500 °C for 2 h in an Ar

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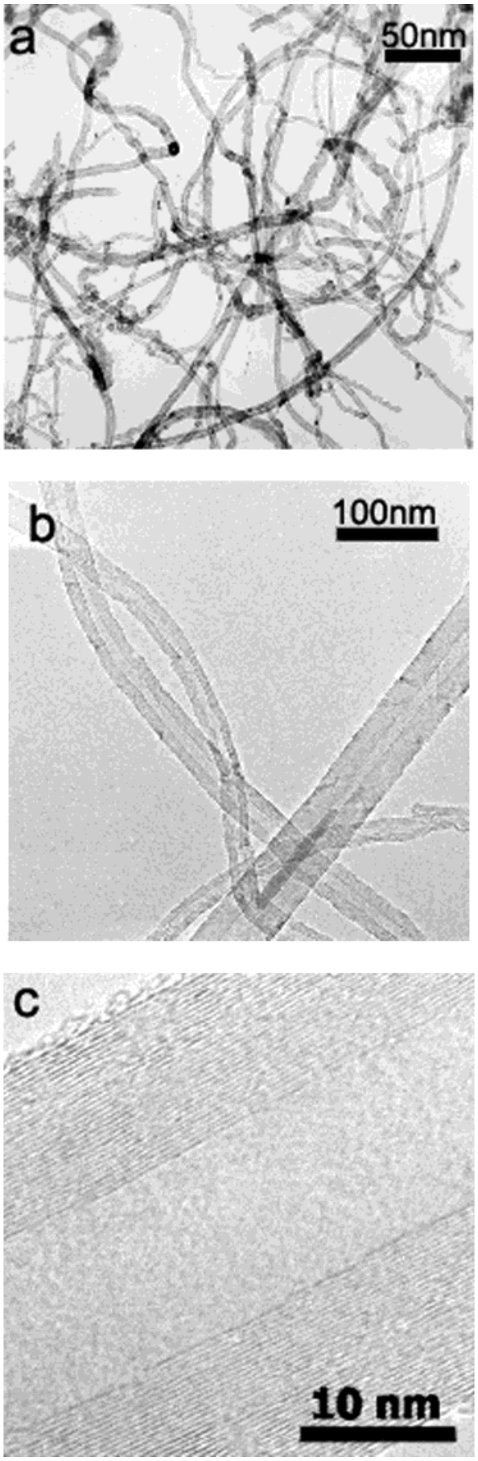

Figure 2. TEM and HRTEM images of CNTs.

atmosphere, respectively. The fracture surfaces of the composites were observed by a scanning electron microscope (SEM, JSM-6700F, JEOL, Japan). Archimedes' method was used to measure the composite density. Chemical analysis was also used to quantitatively provide the composition of the composite. The electrical conductivities of the composites were measured by the Hall effect measurement system (HL5500PC, U.K.).

Results and Discussion

The precursor of Fe(III)–urea complex $Fe[(NH_2)_2CO]_6(NO_3)_3$ is a water-soluble reddish brown powder. The chemical and elemental analysis found (calculated): Fe, 9.60 (9.27); C, 12.11 (11.97); N, 34.08 (34.89); H, 4.18 (4.02). The main bands of Fe(III)–urea complex and those of urea for comparison in the FTIR spectra are summarized in Table 1. Through this assignment, particularly by the absence of a carbonyl band at 1684 cm^{-1},[27] and by the decrease in wavenumber of ν(CO) + $\sigma(NH_2)$ vibration,[28] the formation of oxygen-to-metal coordinate bonds (C═O→Fe) in Fe–urea complex is determined.

Table 1. Absorption Maxima Observed for Urea and Fe–Urea Complex and Their Assignments[a]

	observed frequency (cm^{-1})	
type of vibration	urea	Fe–urea complex
$\nu_{as}(NH_2)$[28]	3435s	3460s
$\nu_s(NH_2)$[32]	3346s	3357s
third N–H band[28]	3259sh	3197m
ν(CO)[28,32]	1684s	/
$\nu(CN) + \delta(NH_2)$[32]	1628sh	1630s
$\nu(CO) + \delta(NH_2)$[28,32]	1600s	1572s
ν_{as}(CN)[28,29]	1468s	1498m
$\nu(NO_3^-)$	/	1385s
$\rho(NH_2)$ [28,29]	1155m	1163w
ν_s(CN) [28,29]	1057vw	1030w
$\nu(NO_3^-)$	/	825vw
τ(ONCN)[29]	787vw	766vw
δ(NCO)[28]	/	613w
δ(NCN)[28]	557s	544w

[a] Abbreviations: /, not found; s, strong; m, medium; w, weak; sh, shoulder. References for the assignment are given in brackets.

XRD patterns of the products obtained by solvothermal synthesis from the Fe(III)–urea complex through CNTs-combined reactions at various reaction temperatures for various reaction times are shown in Table 2. Figure 3 illustrates typical XRD patterns of magnetite–CNTs, magnetite–hematite–CNTs, and hematite–CNTs composites. Though the XRD patterns of magnetite are similar to those of maghematite, there are still slight differences between them.[29] In Figure 3c, the peaks denoted with a hollow pane can be well indexed with magnetite reflection. To further ascertain the phase of the obtained product, the Mössbauer spectrum (MS) of the sample shown in Figure 3c has been measured. Since the MS of magnetite, which is two sextet peaks,[30] obviously differs from that of maghematite, which is a sextet peak,[31] the form of the final product can be discriminated. Figure 4 shows the experimental and calculated MS of the as-prepared product. The interpretation of the spectrum is consistent with the well-established site assignment of magnetite. In this structure the Fe^{2+} ions reside in site B, whereas Fe^{3+} ions are distributed over sites A and B of the magnetite crystal structure. This spectrum is fitted by two sextets; for the sextet attributed to site A (Fe^{3+}) we obtain the hyperfine parameters $H_{eff} = 491$ kOe and I.S. = 0.4 mm s^{-1}. Corresponding to site B ($Fe^{3+} + Fe^{2+}$), the magnetic hyperfine field is $H_{eff} = 460$ kOe and I.S. = 0.79 mm s^{-1}. The data obtained are in accordance with the well-known hyperfine parameters for magne-

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Table 2. Product Phases Derived from the XRD Patterns and the Result of MS under Various Reaction Conditions

weight ratio $Fe[(NH_2)_2CO]_6(NO_3)_3$:CNTs	solvent	temp (°C)	time (h)	product containing CNTs
10:1	$C_2H_8N_2$	100	50	unreacted precursor
10:1	$C_2H_8N_2$	150	50	α-Fe_2O_3 + unreacted precursor (trace)
10:1	$C_2H_8N_2$	200	10	α-Fe_2O_3 + Fe_3O_4
10:1	$C_2H_8N_2$	200	25	α-Fe_2O_3(trace) + Fe_3O_4
10:1	$C_2H_8N_2$	200	50	Fe_3O_4
without CNTs	$C_2H_8N_2$	200	50	α-Fe_2O_3(trace) + Fe_3O_4 (without CNTs)
20:1	$C_2H_8N_2$	200	50	Fe_3O_4
5:1	$C_2H_8N_2$	200	50	α-Fe_2O_3(trace) + Fe_3O_4
2:1	$C_2H_8N_2$	200	50	α-Fe_2O_3(trace) + Fe_3O_4
1:1	$C_2H_8N_2$	200	50	α-Fe_2O_3 + Fe_3O_4
10:1	C_2H_5OH	200	50	α-Fe_2O_3
10:1	H_2O	200	50	α-Fe_2O_3
10:1 (baked CNTs)	$C_2H_8N_2$	200	10	α-Fe_2O_3 + Fe_3O_4
10:1 (baked CNTs)	$C_2H_8N_2$	200	25	α-Fe_2O_3 + Fe_3O_4
10:1(baked CNTs)	$C_2H_8N_2$	200	50	α-Fe_2O_3(trace) + Fe_3O_4

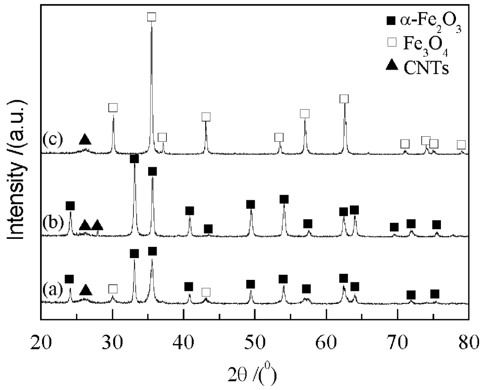

Figure 3. XRD patterns of the nanocomposites obtained by the solvothermal synthesis of precursor with CNTs (a) in a $C_2N_2H_8$ medium at 200 °C for 5 h, (b) in an aqueous medium at 200 °C for 50 h, and (c) in a $C_2N_2H_8$ medium at 200 °C for 50 h.

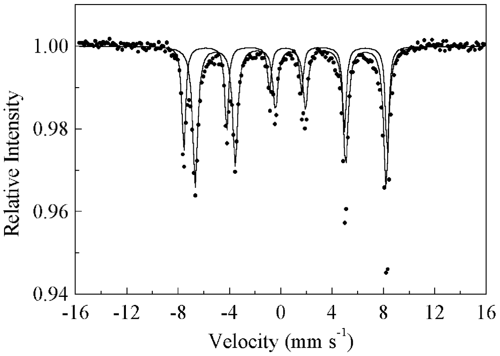

Figure 4. Mössbauer spectrum of the magnetite−CNTs nanocomposite powders measured at room temperature.

tite and provide conclusive evidence for the identity of the material as magnetite. EDS measurements also confirm the form of the pure magnetite−CNTs composite. The elemental ratio of Fe:O was calculated to be 3:4.1 and 3:4.0 at different crystals, which are very close to the stoichiometry of Fe_3O_4.

Fixing the weight ratio of Fe(urea)$_6$(NO$_3$)$_3$:CNTs at 10:1 and the reaction time for 50 h, the effect of reaction temperatures on the phase component was investigated in Table 2. XRD and MS analyses show the product phases varied with different reaction temperatures. At 100 °C, the precursor is not decomposed and remains in ethylenediamine even for 50 h, whereas hematite−CNTs composite containing some undecomposed precursor is obtained at 150 °C for 50 h. At 200 °C, the precursor is fully decomposed. This result corresponds to the TG-DSC curve of the precursor. Figure 5 shows the TG-DSC curve of the thermal decomposition of Fe(III)−urea complex in a N_2 atmosphere. In the corresponding DSC curve, the endothermic peak appears around 178 °C due to the decomposition of urea. The strong oxidation exothermic peak can be seen at 201 °C due to the presence of the vigorous oxidizing group NO_3^-. The weight loss is about 83.7%, consistent with the results of ref 32. Thus, the reaction temperature is chosen at 200 °C.

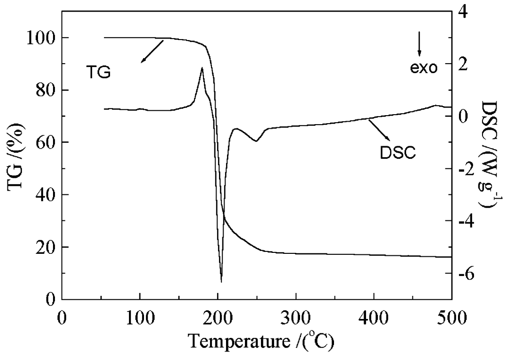

Figure 5. TG and DSC curves of $Fe[(NH_2)_2CO]_6(NO_3)_3$ precursor.

For studies on the reaction time, the solvothermal reaction was held at 200 °C for different times. With increasing reaction time for the weight ratio of Fe(urea)$_6$(NO$_3$)$_3$:CNTs at 10:1, the hematite phase transformed into pure magnetite gradually. After being heated for 50 h, pure magnetite−CNTs nanocomposite is obtained. TEM observation reveals that this nanocomposite consists of magnetite nanoparticles whose sizes are mostly 20−30 nm attached onto the CNTs, as shown in Figure 6. The specific surface area of pure magnetite−CNTs nanocomposite is 58.7 m^2 g^{-1}. The mixed density of the composite can be calculated according to the density of magnetite,[33] the density of CNTs, and the composition of the composite (Table 3). Thus, we obtain the equivalent particle diameter of magnetite of about 30 nm, which is consistent with the results of the TEM picture (Figure 6).

Results obtained in various solvents are also summarized in Table 2. Replacing ethylenediamine with alcohol or water only produces hematite−CNTs com-

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Table 3. Properties of the Investigated Materials

material	magnetite−CNTs[a]	magnetite−hematite−CNTs[b]	magnetite−hematite[c]	magnetite	CNTs
σ (S cm^{-1})	2.5	2.0	1.9	0.2[36]	10^2-10^{-4}[e]
magnetite content (wt %)[d]	67.05	54.19	100	/	/
CNTs content (wt %)	32.95	45.81	0	/	/
density (g cm^{-3})	2.51	2.27	3.69	5.18[33]	~2.1[e]
relative density (%)[d]	73	72	72	/	/

[a] Synthesized at 200 °C for 50 h from the mixture of the precursor and original wet CNTs at the weight ratio of 10:1. [b] Synthesized at 200 °C for 50 h from the mixture of the precursor and baked CNTs at the weight ratio of 10:1. [c] Synthesized at 200 °C for 50 h from the precursor without CNTs. [d] Ignoring the trace amount of hematite. [e] Claimed by the producers.

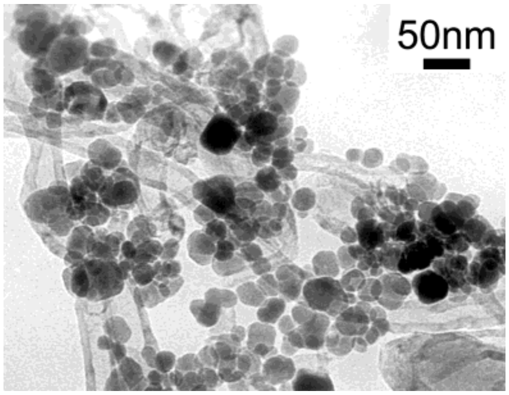

Figure 6. TEM micrograph of the magnetite−CNTs nanocomposite powders.

posite after reaction at 200 °C for 50 h. Kominami et al.[34] have shown that organic solvent, such as alcohols and hydrocarbons, produced magnetite after reaction at 300 °C. The solvent has a reducing effect on Fe^{3+}. With the increasing reaction time, hematite is transformed into magnetite gradually and hematite content in the composite decreases (Table 2). Combined with our experiment, it can be proposed that ethylenediamine used as the reaction medium presumably acts as the reducing agent of Fe^{3+}.

From Table 2, the composites of the products change with the content of CNTs. Without CNTs, the mixture of magnetite and trace hematite is produced after reaction at 200 °C for 50 h. For original wet CNTs, as the weight ratio of $Fe(urea)_6(NO_3)_3$:CNTs is below 10:1 (except the condition without CNTs), the pure magnetite−CNTs can be produced at 200 °C at 50 h. With the increasing original wet CNTs content, the composites contain more and more hematite. In other experiments, baked CNTs were also used to synthesize the composite. However, pure magnetite−CNTs composite would not be obtained under the same conditions (Table 2). It can be proposed that the state of CNTs used is very important for controlling the content of the final production. Previous studies[29,34] on the synthesis of magnetite in organic solvents have suggested that a suitable amount of water significantly affect the crystallization and reduction of iron oxides in organic media. In the presence of a small amount of water, hematite is first crystallized in this medium and then is reduced to magnetite.[34] Excessive water might lead to further oxidation of magnetite and transformation to hematite.[29] With the increasing original wet CNTs, hematite content in the composite increases due to further oxidation of magnetite and transformation to hematite by excessive water. In an aqueous medium, pure hematite−CNTs composite is obtained. For baked CNTs, a deficient amount of water results in the production of hematite as well as magnetite, which can be concluded from the experiments (Table 2). Therefore, the water contained by original wet CNTs significantly influences the formation of pure magnetite−CNTs composite.

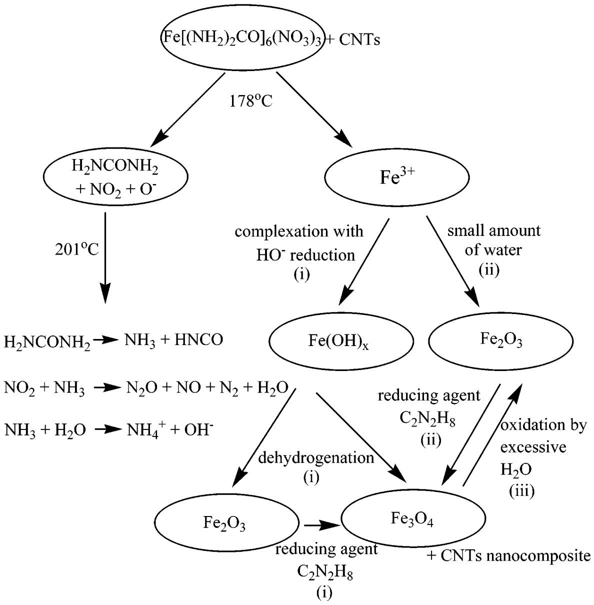

Figure 7. Reaction mechanism of the synthesized pure magnetite−CNTs nanocomposite from the $Fe[(NH_2)_2CO]_6(NO_3)_3$ precursor and the original CNTs.

To illustrate the reaction mechanism in detail, the mechanism scheme is proposed in Figure 7. At 178 °C, a simultaneous release of urea and NO_2 takes place. Subsequent urea degradation and ammonia oxidation by NO_2 occur at 201 °C. On the other hand, the thermal decomposition product medium Fe^{3+} coordinates with HO^- from the hydrolysis of NH_3 to form the $Fe(OH)_x$ complex.[28] On heating, the $Fe(OH)_x$ complex intermediates are converted into hematite and magnetite via a successive dehydrogenation process. Then hematite is further converted into magnetite by a reducing agent (Figure 5, route (i)).[28] Excessive water would oxidate magnetite into hematite (Figure 5, route (iii)).[29] In addition, a small amount of water causes the crystallization of hematite.[34] Then the hematite is reduced to magnetite under reducing agent $C_2N_2H_8$ (Figure 5, route (ii)).[34] Thus, an appropriate amount of water contained by original wet CNTs is very important for the formation of pure magnetite−CNTs nanocomposite. For the baked CNTs, a deficient amount of water retards the reaction of route (ii). For the excessive amount of original wet CNTs, magnetite is oxidated to hematite by excessive water.

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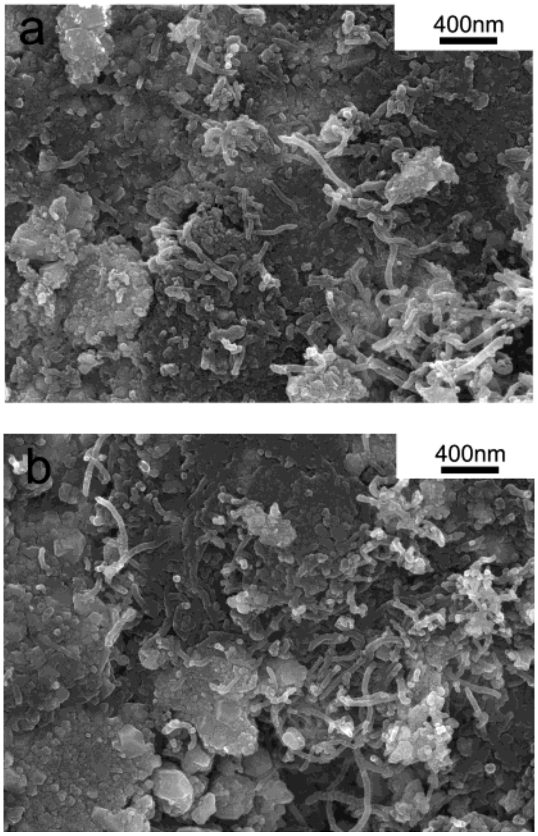

Figure 8. SEM micrograph of a fracture surface of (a) magnetite−CNTs nanocomposite and (b) hematite−magnetite−CNTs composite, respectively.

The composite powders synthesized at 200 °C for 50 h with original wet CNTs and without CNTs are sintered at 500 °C for 2 h in an Ar atmosphere, respectively. So we can compare the difference in the electrical conductivity between the pure magnetite−CNTs nanocomposite and the magnetite−hematite composite without CNTs. In addition, the magnetite−hematite−CNTs composite powders are also sintered under the same conditions. The values of the electrical conductivity, the composition of the nanocomposites, and the relative densities of the sintered materials are illustrated in Table 3. The electrical conductivity (σ) of pure magnetite−CNTs nanocomposite is 2.5 S cm^{-1}, and the magnetite−hematite composite synthesized under the same conditions without CNTs is only 1.9 S cm^{-1}. Morever, the σ value of magnetite−hematite−CNTs composite is 2.0 S cm^{-1}. The addition of CNTs in the nanocomposite increases the electrical conductivity by about 32% from 1.9 to 2.5 S cm^{-1}, compared with the composite without CNTs. The relative densities of the sintered materials are calculated from measurements obtained by Archimedes method, using the density of magnetite,[33] the density of CNTs, and the composition of the composites. A trace amount of hematite is ignored in the calculation of the relative densities. From Table 3, the relative density of the three different nanocomposites is approximately identical, which proposes that the effect of porosity on the measurement of the electric conductivity is the same for these nanocomposites.

To further investigate the fracture surface of the composite, SEM micrographs are shown in Figure 8. For magnetite−CNTs nanocomposite in Figure 8a, CNTs are dispersed in a magnetite matrix. Compared with the composite without CNTs, the increase of 32% on the electrical conductivity of magnetite−CNTs nanocomposite is owed to the introduction of conducting paths to the magnetite, viz., the percolation effect of CNTs,[16,35] considering the approximately equal relative densities of these three different nanocomposites. Figure 8b illustrates a SEM micrograph of hematite−magnetite−CNTs nanocomposite, the electrical conductivity of which is only 2.0 S cm^{-1}. It shows a slight improvement from 1.9 to 2.0 S cm^{-1}. However, it is much lower than the magnetite−CNTs nanocomposite. Comparing Figure 8a and 8b, it is obvious that the magnetite−CNTs nanocomposite leads to a better dispersion of CNTs bundles within the matrix, while the hematite−magnetite−CNTs composite shows serious aggregation of CNTs bundles in the matrix (Figure 8b). It has been found for CNTs−Fe−Al_2O_3 composite by Peigney et al.[16] that better dispersion of the CNTs bundles within the matrix would lead to a better connectivity of the CNTs in the composite, accounting for the higher electrical conductivity. On the other hand, probably because the electrical conductivity of hematite is lower than that of magnetite,[36] trace hematite would decrease the electrical conductivity of the composite in a way. Obviously, pure magnetite−CNTs nanocomposite would show a higher electrical conductivity than hematite−magnetite−CNTs composite. Consequently, the introduction of conducting paths by CNTs and a good dispersion of CNTs in the matrix would lead to an effective and obvious improvement in the electrical conductivity.

Conclusions

A novel simple solvothermal method is successfully employed to in situ synthesize CNTs−magnetite nanocomposite from the mixture of $Fe[(NH_2)_2CO]_6(NO_3)_3$ complex and CNTs. In the synthesis mechanism, the ethylenediamine medium presumably acts as the reducing agent of Fe^{3+}. A suitable amount of water introduced by CNTs significantly affects the crystallization and reduction of iron oxides in organic media to produce pure magnetite−CNTs nanocomposite. The addition of CNTs in the CNTs−magnetite nanocomposite shows an improvement of 32% in the electrical conductivity from 1.9 to 2.5 S cm^{-1}, compared with that of composite without CNTs. The introduction of conducting paths of CNTs and a good dispersion of CNTs in the matrix are possible mechanisms leading to the improvement of the electrical conductivity. Further work, including measurements of other important properties of CNTs−magnetite composites, a detailed investigation on the adhesion between CNTs and the matrix, the improvement of CNTs dispersion in the matrix, and the preparation of other CNTs−oxide composites, is in progress.

Acknowledgment. We thank Prof. J. S. Jiang of the Department of Chemistry and Physics at East China Normal University for his hearty assistance in acquiring MS data.

CM030007Q

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