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Electromagnetic properties of Nd^{3+} substituted iron oxide on graphite nanosheet

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ABSTRACT

Nd^{3+} substituted iron oxide (Fe_3O_4) was chemically precipitated on the surface of graphite nanosheet (NanoG) to get the composite $Nd_xFe_{3-x}O_4$/NanoG. The structure of the composite was characterized by scanning electron microscopy, energy dispersive spectroscopy and X-ray diffraction. Results show that under basic conditions, $FeCl_3.6H_2O$, $NdCl_3$ and $FeSO_4.7H_2O$ can carry out coprecipitation reaction to obtain $Nd_xFe_{3-x}O_4$ on the surface of NanoG. Measurement of electromagnetic parameters shows that when $x=0.06$ and the mass ratio of $Nd_xFe_{3-x}O_4$ to NanoG reaches 4:1, $Nd_{0.06}Fe_{2.94}O_4$/NanoG has good microwave absorbing properties ($R_{min}=-17.13$ dB at 11.90 GHz) in the X band (8.2–12.4 GHz).

1. Introduction

Iron ferrites Fe_3O_4 have reverse spinal crystal structure and are widely used as catalysts, magnetofluids, microwave devices and microwave absorbing materials [1–4]. But their heavy weight and easy oxidization restrain them from being wildly used. Graphite is a kind of material with light weight, low price and stable properties. By oxidation, rapid thermal treatment and ultrasonic dispersion, graphite nanosheet (NanoG) can be prepared from graphite [5,6]. By coprecipitation reaction, Fe_3O_4 can be chemically precipitated on the NanoG's surface.

To improve their magnetic properties and enlarge their application area, rare earth ions with larger ionic radius such as Pr^{3+} [7], Sm^{3+} [8], Gd^{3+} [9], La^{3+} [10], Nd^{3+} [11], Ho^{3+} [12], Dy^{3+} [13], Er^{3+} [14] are usually introduced into the ferrites. In this work, rare earth ion Nd^{3+} substituted Fe_3O_4 precipitating on the surface of NanoG is prepared. The structures of as prepared samples are characterized by scanning electron microscope (SEM), energy dispersive spectroscopy (EDS) and X-ray diffraction (XRD), respectively. Microwave absorbing properties of them are measured.

2. Experimental

NanoG was prepared as reference [15,16] and pretreated by KH550 in a flask. Then an amount of $FeCl_3.6H_2O$, $FeSO_4.7H_2O$ and $NdCl_3$ (mole ratio 2:1:x, $x=0.03$, 0.06, 0.09) were added into the flask with vigorous stirring and the solution was heated to 30 °C in a water bath. A mixture solution of $NH_3.H_2O$ with SDBS was then dropped into the solution and the pH was adjusted to 9–10. After that, the reaction was carried out for about 3 h at 70 °C with constant stirring. The products were isolated by magnetic decantation, washed, dried under vacuum at 80 °C for about 24 h and then annealed further at 600 °C for about 2 h.

The structures of $Nd_xFe_{3-x}O_4$ and $Nd_xFe_{3-x}O_4$/NanoG were characterized by using a scanning electron microscope (SEM; JSM-6390, HITACHI, Japan), energy dispersive spectroscopy analyzer (EDS; JED-2200 Series) and X-ray diffraction (XRD; PANalytical, Holland), respectively. The magnetic properties were measured by Lake Shore7307 vibrating sample magnetometer (VSM). The electromagnetic parameters of them were analyzed by using a HP8753D vector network analyzer.

3. Results and discussion

3.1. SEM images and EDS of $Nd_{0.06}Fe_{2.94}O_4$ and $Nd_{0.06}Fe_{2.94}O_4$/NanoG

By coprecipitation reaction, $Nd_xFe_{3-x}O_4$ can be obtained under basic condition. Fig. 1(a, b) show the structure of $Nd_{0.06}Fe_{2.94}O_4$ with magnification of 10,000 and 40,000, respectively. It can be seen that $Nd_{0.06}Fe_{2.94}O_4$ has cubic spinal crystal structure and the average diameter of it is around 100 nm. The structure of NanoG is shown in Fig. 1(c). It has a width about 1–20 μm and a thickness about 30–90 nm, indicating a large aspect ratio (300–500). Fig. 1(d) shows the structure of $Nd_{0.06}Fe_{2.94}O_4$/NanoG. It can be seen that $Nd_{0.06}Fe_{2.94}O_4$ particles are well dispersed on the surface of NanoG. The high aspect ratio of NanoG is beneficial to $Nd_{0.06}Fe_{2.94}O_4$ to be precipitated on it and can improve the dispersity of $Nd_{0.06}Fe_{2.94}O_4$.

Element contents of $Nd_{0.06}Fe_{2.94}O_4$ and $Nd_{0.06}Fe_{2.94}O_4$/NanoG are measured by EDS. Fig. 1(e, f) confirm the presence of O, Fe, and Nd

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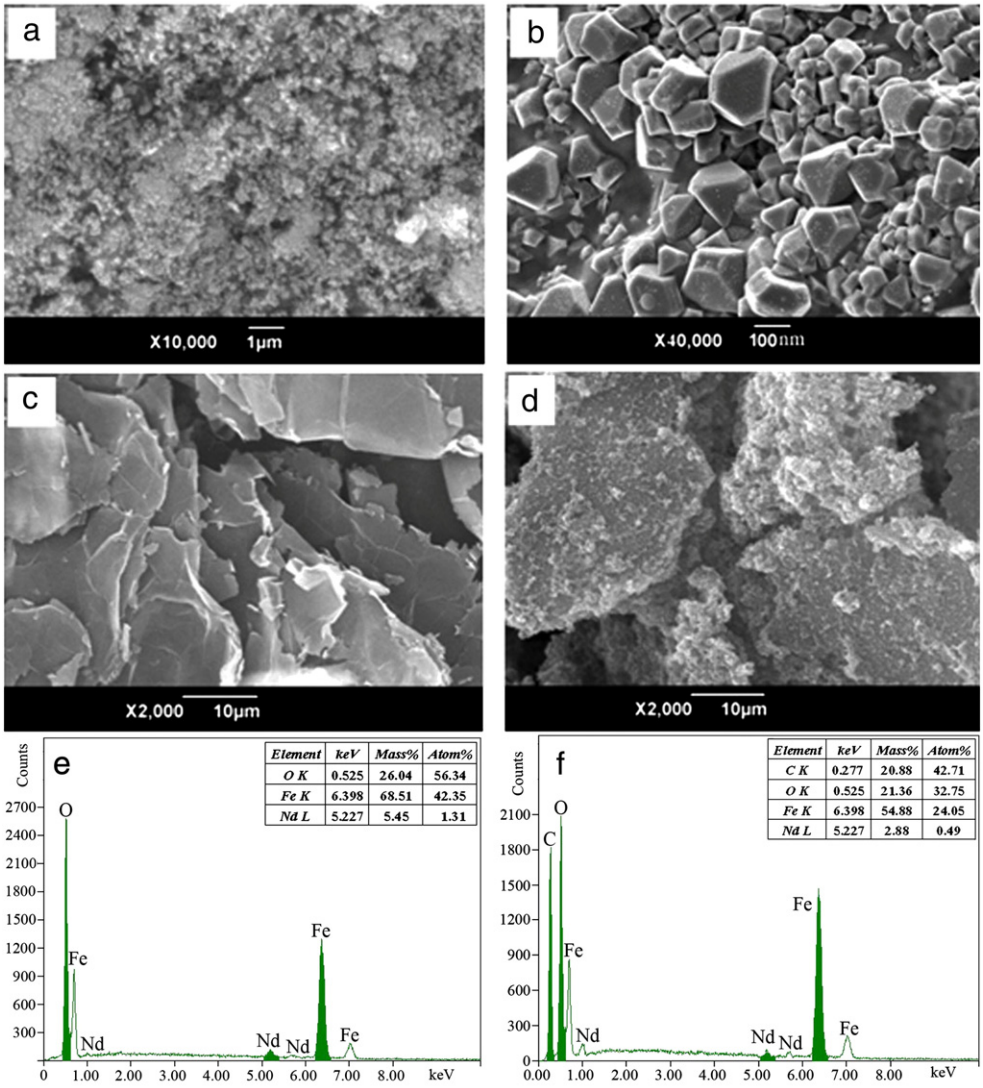

Fig. 1. SEM images of $Nd_{0.06}Fe_{2.94}O_4$ (a,b), NanoG (c), $Nd_{0.06}Fe_{2.94}O_4$/NanoG (d) and EDS of $Nd_{0.06}Fe_{2.94}O_4$ (e), $Nd_{0.06}Fe_{2.94}O_4$/NanoG (f).

in $Nd_{0.06}Fe_{2.94}O_4$ and C, O, Fe, and Nd in $Nd_{0.06}Fe_{2.94}O_4$/NanoG. The mass content of O, Fe, and Nd in $Nd_{0.06}Fe_{2.94}O_4$ are 26.04%, 68.51% and 5.45% while the atom contents of them are 56.34%, 42.35% and 1.31%, respectively. The mass content of C, O, Fe, and Nd in $Nd_{0.06}Fe_{2.94}O_4$/NanoG are 20.88%, 21.36%, 54.88% and 2.88% while the atom contents of them are 42.71%, 32.75%, 24.05% and 0.49% respectively. The experimental values are adjacent to the theoretical values. All these can further illustrate that $Nd_{0.06}Fe_{2.94}O_4$ is prepared on the NanoG's surface.

3.2. XRD of $Nd_{0.06}Fe_{2.94}O_4$ and $Nd_{0.06}Fe_{2.94}O_4$/NanoG

The crystal structure of $Nd_{0.06}Fe_{2.94}O_4$ and $Nd_{0.06}Fe_{2.94}O_4$/NanoG are characterized by X-ray diffraction and the results are shown in Fig. 2. Some diffraction peaks at 18.35°, 30.35°, 35.70°, 43.43°, 54.14°, 57.46° and 63.08° can be seen in Fig. 2(a), which relate to (111), (220), (311), (400), (422), (511) and (440) planes of reverse spinal crystal structure of ferrite [17–20]. The broader peak widths are an appreciable increase in the unit cell parameter *a* after the introduction of Nd^{3+} ion with larger radius compared to that of Fe^{3+} ion in the spinal network.

The typical diffraction peaks of NanoG (002), (110) and $Nd_{0.06}Fe_{2.94}O_4$ (220), (311), (400), (422), (511), and (440) can be seen in the diffraction peaks of $Nd_{0.06}Fe_{2.94}O_4$/NanoG in Fig. 2(b). All the corresponding peaks are relatively weaker than those of $Nd_{0.06}Fe_{2.94}O_4$ for the interaction between $Nd_{0.06}Fe_{2.94}O_4$ and NanoG.

3.3. Electromagnetic properties

To clarify the magnetic properties of the composites, the hysteresis loops of the samples are measured by using VSM. Fig. 3 shows that the composites are soft ferrites with high saturation magnetization. The *Ms* is decreased after the introduction of the nonmagnetic Nd^{3+} for the larger ionic radius of Nd^{3+} decrease the symmetry of the crystal structure (to Fe_3O_4, $Ms = 59.55$ emu/g while it is 50.20 emu/g to $Nd_{0.06}Fe_{2.94}O_4$). The introduction of NanoG can also lead to the decrease of *Ms* (to Fe_3O_4/NanoG, $Ms = 43.74$ emu/g while it is 39.45 emu/g to $Nd_{0.06}Fe_{2.94}O_4$/NanoG). But the conductivities of the composites increase and the electromagnetic parameters are anticipated to be matched well.

Microwave absorbing properties of the materials can be reflected by R-f relation diagram [21–23]. In our experiment, the reflection losses (*R*) of $Nd_xFe_{3-x}O_4$ with different Nd^{3+} contents and *R* of $Nd_xFe_{3-x}O_4$/NanoG with different $Nd_xFe_{3-x}O_4$ to NanoG mass ratios are discussed. Results are shown in Fig. 4. From Fig. 4(a) it can be seen that the reflection losses of $Nd_xFe_{3-x}O_4$ are firstly increased and then decreased with the increasing of Nd^{3+} content. When $x<0.06$, the low Nd^{3+} content can't clearly change the properties ($R_{min} = -11.94$ dB at $f = 11.92$ GHz) whereas