Broadband electromagnetic-wave absorption by FeCo/C nanocapsules

Z. Han, D. Li,^{a)} H. Wang, X. G. Liu, J. Li, D. Y. Geng, and Z. D. Zhang

Shenyang National Laboratory for Materials Science, Institute of Metal Research, and International Centre for Material Physics, Chinese Academy of Sciences, 72 Wenhua Road, Shenyang 110016,

People's Republic of China

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Electromagnetic-wave absorption by FeCo/C nanocapsules has been investigated. In contrast to earlier reported materials, including other nanocapsules, the absorption amplitude of FeCo/C nanocapsules is found not to decrease with increasing absorption-layer thickness. A reflection loss (RL) exceeding -20 dB can be obtained for all frequencies within the 2–18 GHz range by choosing an appropriate layer thickness between 1.6 and 8.5 mm. The broadest bandwidth (RL values exceeding -10 dB) from 10 to 18 GHz, covering half of the X-band and the whole K_u -band, is obtained for a 2 mm layer. © 2009 American Institute of Physics. [DOI: 10.1063/1.3177067]

The development of radar-absorbing materials for commercial and military applications has become a cutting edge issue.¹⁻⁷ Among these materials, the magnetic-core/ dielectric-shell structured nanocomposites are especially promising.^{5,6} FeCo nanocomposites and their self-assembled aggregates have been reported to have prominent magnetic properties with potential application in many fields, such as magnetic-resonance imaging (MRI) (Ref. 8) and electromagnetic (EM)-wave absorption.^{4,9} Carbon is used as shell material of nanocapsules for EM-wave absorption, because of its low price and typical dielectric character. FeCo nanocrystals coated with graphitic carbon, pyrolytic carbon, and carbon nanotubes have been synthesized.^{4,10,11} However, the EM properties of FeCo/C nanocapsules have not yet been investigated. In the present letter, we report on the loss behaviors of FeCo/C nanocapsules, in which the absorption peak strongly redshifts with increasing thickness of the absorbent, but always remain exceeding -20 dB.

The FeCo/C nanocapsules were prepared by a modified arc-discharge technique described elsewhere.¹² An ingot with an molar Fe:Co ratio of 55:45 was chosen because of its high saturation magnetization.¹³ The high-resolution transmission electron microscopy (TEM) images in Figs. 1(a) and 1(b) show that the nanocapsules are spherical and 10–40 nm in diameter and that the thickness of the C shells is about 2–3 nm. Figure 1(d) shows the energy dispersive spectrum of the dotted area in Fig. 1(c), which indicates that the cores consist of the elements Fe and Co. The Cu detected in Fig. 1(d) comes from the copper grid, which is used to mount the powder specimens.

The relative complex permittivity $(\varepsilon_r = \varepsilon' - j\varepsilon'')$ and the relative complex permeability $(\mu_r = \mu' - j\mu'')$ of the FeCo/C nanocapsules have been measured between 2 and 18 GHz as described elsewhere.⁵ The frequency dependencies of the real part (ε') and the imaginary part (ε'') of ε_r are shown in Fig. 2(a). Both ε' and ε'' are found to exhibit a tendency to decrease with increasing frequency. In both curves, the decrease with frequency is not strictly uniform and a few weak local maxima are found. Such loss phenomena can be attributed to various polarizations.¹⁴ Liu *et al.*⁷ reported a quite similar permittivity spectrum for the porous-C/Co system.

They showed the enhancement of the dielectric response due to the interfacial polarization relaxation loss and Ohmic loss in heterogenous systems. Such dielectric relaxation behavior has also been found for C-coated Cu.¹⁵

The frequency dependencies of the real part (μ') and the imaginary part (μ'') of μ_r are presented in Fig. 2(b). The μ' values decrease monotonously in the 2–18 GHz range. However, μ'' exhibits a broad maximum around about 10–11 GHz, with relative maxima at about 7, 11, and 17 GHz. These multiresonance peaks of μ'' have been suggested to result from single or multiple relaxation mechanisms,¹⁶ but there is no clear evidence about the exact origin of this fine-structure. The high ferromagnetic-resonance frequency around 10–11 GHz in the FeCo/C system may be attributed to a large damping coefficient, which comes from intrinsic damping, surface effects, and interparticle interactions.¹⁷ This high resonance frequencies in magnetic nanoparticles have been researched both experimentally and theoretically.^{1,5,17–19}

The absorption properties of the FeCo/C nanocapsules have been derived according to transmission-line theory.⁹



FIG. 1. (Color online) (a) TEM and (b) high-resolution-TEM image of FeCo/C nanocapsules, (c) scanning-TEM image of (a), and (d) energy dispersive spectrum of the dotted area in (c).

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^{a)}Electronic mail: dali@imr.ac.cn.



FIG. 2. (Color online) (a) Relative complex permittivity and (b) relative complex permeability of FeCo/C nanocapsules dispersed in paraffin as a function of frequency (open squares and triangles represent measured data, dashed lines of the smoothed curves of ε_r and μ_r).



FIG. 3. (Color online) Frequency dependence of the dielectric loss factor and the magnetic loss factor of FeCo/C nanocapsules dispersed in paraffin.

TABLE I. EM-wave absorption properties of some recently reported nanocomposites. The optimal RL means the largest absolute value of the reflection loss, namely, the best absorption. The absorbent thickness denotes the layer thickness (range) for which RL \leq -20 dB. The absorption bandwidth is the frequency range in which RL \leq -20 dB can be achieved.

Specimen	Particle size (nm)	Optimal RL values (dB)	Absorbent thickness (mm)	Absorption bandwidth (GHz)	Ref.
Ni/C	25-30	-32	2	12.5-13.5	1
Fe(Mn)/ferrite	20-30	-28	1.8-2	13-15	5
FeNi/C	10-60	-27	1.9-2.1	13-17	6
C-tubes/Co	10 µm	-40	$\sim 2.5 - 5$	$\sim 4 - 10$	7
FeCo/C	10-40	-29	1.6-8.5	2-18	This work



FIG. 4. (Color online) Frequency dependence of the dielectric- and magnetic-loss factors of FeCo/C nanocapsules dispersed in paraffin.



FIG. 5. (Color online) Three-dimensional representations of RL with (a) the measured ε_r and μ_r and (b) the smoothed ε_r and μ_r .

Figure 3 shows the obtained relationship between the reflection loss (RL) and the EM-wave frequency in the 2–18 GHz range for the paraffin-nanocapsules samples with various thicknesses. It is worth noting that the absorbent with a thickness of 2 mm has RL values exceeding -10 dB in the 10–18 GHz range, which covers half of the X-band (8–12 GHz) and the whole K_u -band (12–18 GHz). For all frequencies within the 2–18 GHz range, RL values exceeding -20 dB can be obtained by choosing an appropriate thickness of the absorbent layer between 1.6 and 8.5 mm.

Table I shows that the present FeCo/C system exhibits the broadest absorption bandwidth among recently reported materials. Unlike other nanocomposites,^{1,2,13,17} the RL peak of the present nanocapsules shifts to lower frequencies without any decrease in amplitude (Fig. 3). Most nanocapsules with a core/shell structure, such as Fe(Mn)/ferrite,⁵ (FeNi)/C,⁶ etc., exhibit narrow dielectric- and magnetic-losstangent peaks, which usually have an overlapping frequency range. This is not the case in the present system, where the dielectric-loss factor (tan $\delta_{\varepsilon} = \varepsilon'' / \varepsilon'$) and the magnetic-loss factor (tan $\delta_{\mu} = \mu''/\mu'$) exhibit maxima at low (2–4 GHz) and at high (11-18 GHz) frequencies, respectively (Fig. 4). The present samples were investigated with a set mass ratio of 50 wt % (thus a set volume fraction), and the intrinsic permittivity and permeability are assumed to be unchanged with varying the nanocapsules/paraffin layer thickness. We speculate that the enhancement of the microwave-absorption properties of FeCo/C nanocapsules results from an excellent synergistic effect of the magnetic and dielectric losses of the cores and shells. Compared with other nanocapsules,^{1,5–7} differences in the particles' size and the shell thickness lead to different ratio of core to shell volumes that might contribute differently to the microwave-absorption properties.

Since the details of the observed weak resonance peaks in the permittivity and the permeability curves are not quite understood, it is important to establish to which extent these fine structures affect the absorption properties. Figure 2 shows the ε_r and μ_r spectra smoothed by a fifth-order polynomial fitting. The RL results, obtained on the basis of these smoothed data, turn out to be strikingly similar to the results if the fine-structures are included. For comparison, the RL obtained from smoothed data for an absorbent thickness of 2 mm is also presented in Fig. 3 (dashed line). Threedimensional RL plots without and with smoothing the ε_r and μ_r spectra are shown in Fig. 5 which reveals in detail the very limited influences of the fine-structures on the absorption properties.

In conclusion, FeCo/C nanocapsules have been prepared by arc-discharging. Fine-structures in permittivity and permeability curves have been analyzed to have negligible influence on the derived wave-absorption properties. For all frequencies within the 2–18 GHz range, RL values exceeding

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-20 dB can be realized by choosing an appropriate thickness of the absorbent layer between 1.6 and 8.5 mm. Especially, for a 2 mm layer, RL values exceeding -10 dB are obtained in the broadest range, from 10 to 18 GHz, which covers half of the *X*-band and the whole K_u -band. FeCo/C nanocapsules seem very attractive for application in radar-wave absorption.

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