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A newly-designed magnetic/dielectric [Fe₃O₄/BaTiO₃@MWCNT] nanocomposite system for modern electromagnetic absorption applications

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ABSTRACT

Developments in electronic industries for telecommunications and demands for decreasing electromagnetic radiation pollution result in developing researches on microwave absorption materials. The target of the present study is to design materials with high absorption properties for electromagnetic waves in the 12-18 GHz range. Thus, Fe₃O₄ magnetic nanoparticles were syntheses through chemical coprecipitation reinforced by ultrasonic. Then, BaTiO₃ nanocrystalline powder was synthesized by the hydrothermal sol-gel method under atmospheric oxygen. Next, nano-particles of barium titanate were deposited on the multi-walled carbon nanotubes (BaTiO₃@CNT). It was concluded that a magneticdielectric nanocomposite has superior microwave absorption properties in comparison to individual magnetic or dielectric absorbers. Also, in order to obtain an optimum absorption in a wide frequency band, dielectric-CNT nanocomposites represents higher properties than magnetic-CNT composites. It is concluded that composites with more magnetic percentage showed better absorption in low frequency band (12 GHz), whereas composites with more dielectric percentage exhibited superior absorption for high frequency band (18 GHz). 80-93% absorption was obtained in the frequency range of 16.7-18 GHz by composite 40M.20F.40C (40% paraffin, 20% magnetite, 40% multi-walled carbon nanotubes). Also, composite 40M.20B.40B@C (40% paraffin, 20% barium titanate, 40% barium titanate deposited on multi-walled carbon nanotubes) showed the absorption of 80-90%.

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1. Introduction

In the recent years, considerable attention has been devoted to electromagnetic wave absorbing materials in 0–25 GHz range for solving the expanded electromagnetic interference (EMI) problems, including intense demands for reduction of electromagnetic (EM) pollution/radiation produced by cellular phones and related intensifying stations as well as shielding interference in the field of electric communication. Also, the use of electromagnetic interference shielding materials results in increasing the precision, reliability and longer life of wireless electronic devices. Also the development of thin and lightweight microwave absorptive coatings has led to indiscriminate use of this concept in stealth technology [1,2].

To address these issues, multifarious microwave absorbing materials, which can attenuate microwave by converting them into

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http://dx.doi.org/10.1016/j.jmmm.2017.05.074 0304-8853/© 2017 Elsevier B.V. All rights reserved. heat or dissipating them via interference, have been extensively investigated, including ferrites [3,4], magnetic metals [5,6], conductive macromolecules [7,8] and dielectric materials. For dielectric absorbers, the carbon-based materials with light weight, e.g. the carbon black, carbon fiber and nanotube, and graphite, have been extensively evaluated, which showed good microwave absorbing properties arising from the dielectric loss [9,10]. Additionally, many studies have focused on magnetic/dielectric composite systems to broaden further the microwave absorbing frequency band.

Among absorbing materials, magnetic ferrites have been widely used to absorb microwaves due to their high magnetic loss [11]. BaTiO₃ is also considered to be a good potential candidate for this application as a dielectric absorber, because of its high permittivity and a large propagation constant, which leads to a high attenuation constant [12]. Also, owing to their unique structure, multi-walled carbon nanotubes (MWCNTs) display interesting properties that make them appropriate potential in the wave absorbing complex material domain [13]. Additionally, because MWCNTs have a





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nanoscale structure, the reflectivity of electromagnetic waves with much larger wavelengths (such as those used for radar) is greatly reduced, which contributes to the stealth capabilities of the material. On the other hand, the high aspect ratio of MWCNTs provides a large interfacial area, which favors electron transport and electric conductivity.

However, pure Fe_3O_4 nanostructures show relatively low complex permittivity, resulting in poor impedance matching condition. These situations cause that the microwave absorption efficiency of pure Fe_3O_4 nanostructures is relatively low and subsequently larger matching thickness is needed. Therefore, combining Fe_3O_4 nanostructures with dielectric materials has been promised to be an effective route to enhance their microwave absorption properties [14–18]. Considering the above advantages of BaTiO₃ as a dielectric material, and MWCNTs, the combination of both may



Fig. 1. synthesis chart of BaTiO3 powder.

result in the development of high performance microwave absorbers. Therefore, the current study is conducted to evaluate the synergistic effect of newly-designed magnetic/dielectric [Fe₃O₄/ BaTiO₃@MWCNT] nanocomposite system on the mechanism of enhanced microwave absorption properties.

2. Experimental procedure

2.1. Synthesis of BaTiO₃

Barium titanate was prepared via precipitation reaction in aqueous solution, according to the following reaction [19]:

$$BaCl_{2}(aq) + TiCl_{4}(aq) + 6NaOH(aq)$$

$$\rightarrow BaTiO_{3}(s) + 6NaCl(aq) + 3H_{2}O$$
(1)

The details of approach by which the $BaTiO_3$ nanoparticles are produced (i.e., sol-gel-hydrothermal method) is shown by flow-chart in Fig. 1. TiCl₄ and $BaCl_2$ starting materials are used for the direct synthesize of nanocrystalline $BaTiO_3$ powder.

2.2. Synthesis of Fe₃O₄

 Fe_3O_4 nanoparticles co-precipitation preparation flowchart is shown in Fig. 2. $Fe(OH)_3$ precipitate was washed several times with de-ionized water. Then, $FeCl_3$ solution was obtained by $Fe(OH)_3$ precipitate dissolution with hydrochloric acid. By adding measured amount of $FeSO_4$ ·7H₂O, Fe^{3+} and Fe^{2+} molar ratio to $FeCl_3$, the solution was adjusted to 1.5:1. Under ultrasonic agitation, black precipitate was produced immediately by adding sodium hydroxide (NaOH). The principle reaction is:

$$Fe^{2+} + 2Fe^{3+} + 8OH^- \rightarrow Fe_3O_4 + 4H_2O$$
 (2)

Obtained Fe₃O₄ precipitate was aged at 65 °C for 30 min in ultrasonic water bath. To purify prepared Fe₃O₄ particles, the samples were washed repeatedly with de-ionized water and ethanol until pH level of 7 was reached. Particles were then dried at 74 °C in vacuum [20].



Fig. 2. synthesis chart of Fe₃O₄ powder.

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