Journal of Colloid and Interface Science 504 (2017) 479–484

Contents lists available at ScienceDirect

Journal of Colloid and Interface Science

journal homepage: www.elsevier.com/locate/jcis

Regular Article

Incorporation of the polarization point on the graphene aerogel to achieve strong dielectric loss behavior

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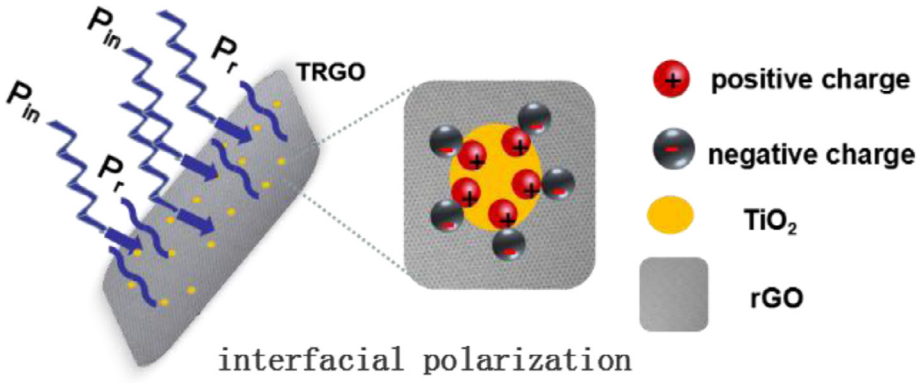
GRAPHICAL ABSTRACT

Taking full advantage of the interface is an efficient strategy to achieve good electromagnetic absorption performance. Therefore a simple strategy to improve the electromagnetic absorption performance is achieved by inserting the polarization point on the graphene aerogel. The obtained TRGO samples show the excellent absorbing property.



ARTICLE INFO

Article history:
Received 19 April 2017
Accepted 2 June 2017

Keywords:
Interfacial polarization
Polarization point
Contact area
Microwave absorption
Impedance matching
Lightweight

ABSTRACT

The preparation of nanocomposites of reduced graphene oxide with loaded TiO_2 nanoparticles (TRGO) by a facile one-step hydrothermal treatment is reported. We have successfully increased the contact area of TiO_2 and RGO to enhance polarization point, which is in favor of strengthening interfacial polarization. The interfaregioncial polarization has been regarded as an important role on the attenuation of high-frequency electromagnetic waves. Therefore, a good absorber is prepared by inserting the polarization point on the graphene aerogel, which shows excellent electromagnetic wave absorbing properties. In detail, the minimum reflection loss value at 2.1 mm is up to −27.2 dB for the TRGO-1.5 composite and the frequency bandwidth of 5.2 GHz can be obtained. Thus, it demonstrates that the adjustment of interface polarization would play a key role in the microwave-absorbing ability.

1. Introduction

In recent years, due to the rapid growth of the electronics industry and information technology, electromagnetic (EM) pollution or electromagnetic interference (EMI) has become a huge threat not

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http://dx.doi.org/10.1016/j.jcis.2017.06.004
0021-9797/

only on human health and other biological systems but also on the normal operation of the electronic equipments [1,2]. To solve this issue, more and more attentions have been paid on the EM wave absorbing materials (MAMS), which can transfer the incident EM wave energy into thermal energy and prevent strong reflection on the absorber surface [3,4]. It is well known that a kind of high-performance EM wave absorption materials must possess features including wide frequency bandwidth, strong absorption intensity, lightweight, and thin thickness [5]. Based on these requirements, graphene has been widely studied due to the good electrical properties, lightweight and high specific surface area [6,7]. Unfortunately, pure graphene usually exhibited bad absorption performance because of the limited attenuation mechanism, as well as its poor impedance matching characteristic [8].

Therefore, significant effort has been devoted to graphene-based composite materials. For instant, Qu and co-workers coupled hollow Fe_3O_4-Fe magnetic nanoparticles with highly conductive graphene sheets to improve the EM wave absorption properties [8]. Ding et al. prepared nanocomposites of reduced graphene oxide with embedded Fe_3O_4/Fe nanorings to functionalize rGO and got the outstanding microwave absorption [9]. Whereafter, they successfully synthesized a lightweight and highly efficient absorbing material for application in the high frequency band by combining Co_3O_4 nanosheets and RGO [10]. Wang et al. prepared surface modified $Fe_{50}Ni_{50}$ nanoparticles/RGO epoxy composites to regulate the impedance matching and microwave attenuation [11]. However, most of them only focus on the impedance matching by combining the magnetic materials with graphene. The research about the method of adjusting impedance matching by coupling dielectric materials with graphene is little. Moreover, previous report paid more attention on the impedance matching and too poor on the interfacial polarization. Hence, we inserted the polarization point of dielectric nanoparticles on the graphene aerogel. In addition, we also investigated the common effect of interfacial polarization and impedance matching on the absorption performance. TiO_2 is an important semiconductor and has been widely used to regulate complex permittivity of carbon materials due to its low density, thermally stability as well as the lower permittivity [12–14]. However, to the best of our knowledge, nanocomposites of TiO_2 nanoparticles and RGO for microwave absorption have scarcely reported.

Herein, we prepared TiO_2/RGO (TRGO) 3D aerogel by a facile one-step hydrothermal treatment. The introduction of TiO_2 not only improves the impedance matching but also strengthen interfacial polarization. More importantly, the interfacial polarization can be adjusted by changing the contact area of TiO_2 and graphene. When the amount of tetrabutyl titanate is up to 1.5 mL, the sample shows superior microwave absorption ability. The optimal RL value reached −27.2 dB at 14.8 GHz and the effective frequency range (RL below −10 dB) of 5.2 GHz (12.68–17.88 GHz) under 2.1 mm can be achieved. Our study indicates that TRGO shows improved performance due to both improved impedance matching and strong interfacial polarization.

2. Experimental section

2.1. Synthesis of TiO_2/RGO 3D aerogel

The TiO_2/RGO 3D aerogel were synthesized by a facile one-step hydrothermal treatment with different proportions of GO aqueous solution and TiO_2 nanosheets. Typically, 75 mL GO aqueous solution (2 mg/mL) and HF (0.2 mL) were added in an autoclave under stirring for 5 min. Then, tetrabutyl titanate (the precursor of TiO_2), in a different amount (1.0 mL, 1.5 mL, 2.0 mL) were added under magnetic stirring. The mixture was magnetic stirred for 72 h to improve the loading amount of the TiO_2 nanoparticles on graphene. After that, the autoclave was treated at 200 °C for 48 h. The obtained aerogel was washed several times with deionized water and freeze-dried. The samples were denoted as TRGO-1.0, TRGO-1.5, TRGO-2.0.

2.2. Characterization of the samples

X-ray diffraction (XRD, Bruker D8 ADVANCE) data were collected with 2θ range of 10–90° at 40 kV and 40 mA using Co-K*a* as the irradiation source (λ = 0.1789 nm) to analyze the crystal phase. Transmission electron microscopy as well as the high resolution transmission electron microscopy (TEM and HRTEM; JEOL JEM-2100) were used to characterize the morphology and size of the synthesized samples. Their microscopy images were obtained by the Field emission scanning electron (SEM; Hitachi S4800). In addition, Raman spectra were obtained on a Renishaw in Via 2000 Raman microscopes using 514 nm incident radiations. The content of each component were measured by X-ray energy spectrometer and thermo-gravimetric analysis (TG; NETZSCH STA 449F3). The vector network analyzer system (Agilent PNA N5224A, coaxial line method) were introduced to record the electromagnetic parameters of the samples in microwave frequency range. Before the measurement, the samples were prepared by homogeneously mixing paraffin wax with 20 wt% products and then pressing into toroidal-shaped samples (Φ_{out}: 7.0 mm, Φ_{in}: 3.04 mm). Afterward, the ε′, ε″, μ′, and μ″ values were calculated by a software, which has been installed in Agilent PNA. Finally, based on transmission line theory, the value of reflection loss (RL) with different thicknesses can be calculated according to the measured relative complex permittivity and permeability. Using the following equations [15,16].

$$Z_{\text{in}} = Z_0(\mu_r/\varepsilon_r)^{1/2}\tanh[j(2\pi fd/c)(\mu_r\varepsilon_r)^{1/2}] \quad (1)$$

$$RL = 20\log|(Z_{in} - Z_0)/(Z_{in} + Z_0)| \quad (2)$$

In which Z_0 and Z_{in} is the impedance of free space and the input impedance of the absorber, respectively, μ_r is the relative complex permeability, ε_r is the relative complex permittivity, f is frequency of microwaves, d is the thickness of the absorber, and c is the velocity of electromagnetic waves in free space.

3. Results and discussion

XRD was used to investigate the crystal structures of the as-obtained samples, and the results are presented in Fig. 1a. In the XRD pattern of the products, the diffraction peaks at 25.3°, 37.8°, 48.0°, 53.9°, 62.7° and 75.0° are well matched with (101), (004), (200), (105), (204) and (215) planes of anatase TiO_2 (JCPDS card No.: 21-1272). The diffraction peaks of RGO can not be detected due to the relatively low diffraction intensity [17]. By comparing the typical samples of TRGO-1.0, TRGO-1.5, TRGO-2.0, we can not find huge difference. Therefore one conclusion can be made that the amount of TiO_2 has no effect on the crystal structures of the samples. The degree of graphitization of TRGO-1.0, TRGO-1.5, TRGO-2.0 have been characterized by the Raman. Fig. 1b shows the Raman spectra of TRGO-1.0, TRGO-1.5, TRGO-2.0. And the two characteristic peaks appear at around 1347 and 1593 cm^{-1} are assigned to the typical D (A_{1g} breathing mode) and G (E_{2g} in-plane vibrational mode) bands of the carbon materials, respectively [18]. To investigate the disordered structure of the carbon-based materials, the intensity ratio of the D band to G band (I_D/I_G) were calculated. It is amazed to find that all the values of I_D/I_G of the samples are nearly same, suggesting that the degree of graphitization changes little for three samples. In this view,