Advanced Powder Technology xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Advanced Powder Technology

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

Original Research Paper

Fabrication of MoS_2-graphene modified with Fe_3O_4 particles and its enhanced microwave absorption performance

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ARTICLE INFO

Article history:
Received 29 July 2017
Received in revised form 14 December 2017
Accepted 19 December 2017
Available online xxxx

Keywords:
Graphene
MoS_2
Nanocomposites
Microwave absorption properties

ABSTRACT

To obtain high-performance microwave absorbers, the 3D hierarchical structure material (MoS_2/Fe_3O_4/graphene) has drawn much attention due to its superior structure and properties. Here, 3D architecture like flower structure of MoS_2/graphene was synthesized by one-pot hydrothermal. Fe_3O_4 particles were grown on the petals of MoS_2/graphene flower structure through co-precipitation route. Electromagnetic (EM) wave absorbing results of MoS_2/Fe_3O_4/graphene demonstrate the maximum reflection loss (R_L) is −45.8 dB at 5.9 GHz for a coating of 2.5 mm, which is significantly enhanced compared with MoS_2/graphene, in terms of absorption intensity and absorbing bandwidth. The absorption bandwidth less than −10 dB can achieve almost 7.4 GHz (4.6–9.7 GHz, 15.7–18 GHz) with a thickness of 2.5 mm. The excellent EM wave properties of MoS_2/Fe_3O_4/graphene are ascribed to the suitable impedance matching and multi-polarization. Consequently, it is believed that the 3D hierarchical structure of MoS_2/Fe_3O_4/graphene can serve as a potential EM wave absorber and can be used in practical applications.

1. Introduction

Currently, electromagnetic (EM) absorption materials have attracted more and more attention due to serious EM pollution, which results from increasing use of radar equipment and wireless system and has severely threatened human health [1–3]. The high-performance EM wave absorbers must satisfy the need for strong absorption, light weight, thin thickness and wide bandwidth on the condition that high impedance matching, excellent synergistic effect [4–6]. Traditionally, microwave absorbers are mainly comprised of metal oxides. Unfortunately, the traditional absorbers face lots of challenges, such as high density and low permittivity, which are barriers to prevent the development of high-performance EM wave absorber. Compared with metal oxides as magnetic materials, dielectric material has been developed to further improve microwave absorption performance because of its light-weight, higher permittivity [7,8].

Since graphene was extracted from graphite by an ordinary adhesive tape in 2004, it has developed rapidly in many fields such as capacitor [9,10], lithium-ion battery [11], photocatalyst [12,13], sensor [14,15]. Importantly, as a thinnest 2-D material, graphene is a candidate of EM wave absorption materials, which is ascribed to abundant defects and functional groups of its surface [16,17]. However, the high permittivity of graphene has limited the improving of EM wave absorption performances due to the mismatched impedance effect. Therefore, all kinds of dielectric and magnetic materials have been introduced into graphene surface to lower the permittivity, results in enhanced EM wave performances. For instance, Wang et al. reported that PANI nanorod arrays grew on the surface of N-doped graphene and the highest R_L could be up to −38.8 dB for a thickness of 3 mm [18]. The graphene@polyaniline fim composite was studied by Liu and the maximum R_L could achieve almost −41.4 dB, the absorption bandwidth below −10 dB was 4.2 GHz [19]. Fu et al. synthesized nanostructured graphene/$NiFe_2O_4$ nanorod composite and found that the composite reached −29.2 dB with a coating of 2 mm, which was better than that of pure graphene [20]. As a representative dielectric material, MoS_2 possesses multilayer structure like graphene and indirect energy band of 1.2 eV, which has received increasing attention and been used as an attractive 2-D material analogous to graphene [21]. Furthermore, MoS_2/graphene composite with 3-D architecture nanosheets can prevent the adjacent layers of graphene from restacking, which provides favourable properties [22]. Nowadays, the composite of MoS_2/graphene is widely used in the area of energy storage [23,24], hydrogen evolution [25], microwave

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https://doi.org/10.1016/j.apt.2017.12.016
0921-8831/

absorption [26]. As far as we know, the dielectric properties and microwave absorption performance of MoS_2/graphene composite modified with magnetic particles have never been reported. The introduction of magnetic particles can improve the equilibrium between permittivity and permeability, which is beneficial to enhanced EM wave absorption performances.

In this study, the 3-D architecture of MoS_2/Fe_3O_4/graphene composite was fabricated and its enhanced EM wave absorption mechanisms were investigated.

2. Experimental

Graphene oxide (GO) was produced according to the previous literature process [27]. The synthesis of MoS_2/graphene was carried out as follows. In detail, 2.19 g sodium molybdate (Na_2MoO_4) and 2.07 g thiourea were dissolved in 70 mL GO aqueous solution by stirring to form a homogeneous solution. Then, the mixture was placed to a 100 mL Teflon-lined steel autoclave and kept at 200 °C for 24h. After cooling down to room temperature naturally, the product (MoS_2/graphene) was washed with ethanol and deionized water, and dried at 60 °C overnight.

The fabrication of MoS_2/Fe_3O_4/graphene composite was prepared by one-pot co-precipitation process. First, 0.2 g MoS_2/-graphene composite was dispersed in 80 mL distilled water via ultrasound treatment for 20 min. Second, 0.14 g $FeCl_3{\cdot}6H_2O$ and 0.1032 g $FeCl_2{\cdot}4H_2O$ were added into the above solution and dissolved slowly. Then, 1 M NaOH solution was added dropwise to the above suspension until pH = 11, and the mixture was prepared by stirring at 80 °C for 3 h. Finally, the products (MoS_2/Fe_3O_4/-graphene) were washed with ethanol and deionized water, and dried at 60 °C for 24 h.

The crystal phase was tested by X-ray powder diffraction (German Bruker D8 using Cu-Kα radiation). The FESEM (Quanta 600FEG) and TEM (JTM-2100) were used to observe the morphology and nanostructure. The XPS spectrum was recorded in an ESCALAB 250 X-ray photoelectron spectrometer to explore the valence state of element. The magnetization hysteresis curves of the samples were tested to obtain coercive force and saturation magnetization by a vibrating sample magnetometer (VSM, Lake Shore7307). The electromagnetic parameters (ε′, ε″, μ′ and μ″ values) were recorded with coaxial-line method using a vector network analyzer (HP8720ES) from 2 GHz to 18 GHz, with a product to paraffin weight ratio of 3:7 and pressed into a ring shape (OD 7 mm and ID 3.04 mm).

3. Results and discussion

The crystal structures of MoS_2/graphene, MoS_2/Fe_3O_4/graphene composite were investigated by XRD measurements, as shown in Fig. 1. As for MoS_2/graphene shown in Fig. 1a, the diffraction peaks of 14.2°, 33.5°, 39.6° and 58.9° can be corresponded to (0 0 2), (1 0 0), (1 0 3) and (1 1 0) planes, suggesting the hexagonal phase MoS_2 (JCPDS No. 37-1492) [21,22]. For MoS_2/Fe_3O_4/graphene composite in Fig. 1b, it can be seen that except for the existence of MoS_2 peaks, the diffraction peaks of Fe_3O_4 at 30.3°, 35.5°, 43.4°, 57.2° and 62.9° are assigned to the (2 2 0), (3 1 1), (4 0 0), (5 1 1) and (4 4 0) planes, match well with Fe_3O_4 (JCPDS No. 19-0629) [28]. In particular, the peaks of graphene can be not observed in the two samples, which may be ascribed to the decreasing stacking of graphene sheets by the introduction of MoS_2 and Fe_3O_4 [29]. Furthermore, the impurity peaks are not detected, indicating the high purity of all the samples.

Fig. 2 shows XPS spectra of MoS_2/Fe_3O_4/graphene composite. From Fig. 2a, the wide scan XPS spectrum confirms the existence of S 2p, Mo 3d, C 1s, O 1s and Fe 2p, according with the experimental theoretical value. As demonstrated in Fig. 2b, the C1s spectrum can be separated into two peaks, originating form C—C/C = C (284.5 eV) in the aromatic rings, C—O (286.1 eV) in oxygen-containing bound [30]. The extremely weak peak of C—O suggests that the GO has been successfully reduced into graphene. The peaks at 711.4 and 725.3 eV (Fig. 2c) are attributed to Fe 2p3/2 and Fe 2p1/2, which illustrates the existence of Fe^{2+} and Fe^{3+}, proving the formation of Fe_3O_4. The O 1s peaks (Fig. 2e) at 530.1 and 531.4 eV correspond to the metal-oxygen bonds [31]. From the Mo 3d spectrum in Fig. 2d, the Mo 3d band can be divided into two peaks located at 229.1 and 232.3 eV, which corresponds to Mo 3d5/2 and Mo 3d3/2 [21,22], revealing the formation of Mo^{4+}. The peaks at 161.8 and 162.9 eV are ascribed to the S 2p3/2 and S 2p1/2 of divalent sulfide ions (S^{2-}), respectively, as shown in Fig. 2f [32]. The characteristic spectra in Fig. 2d and f prove the existence of MoS_2.

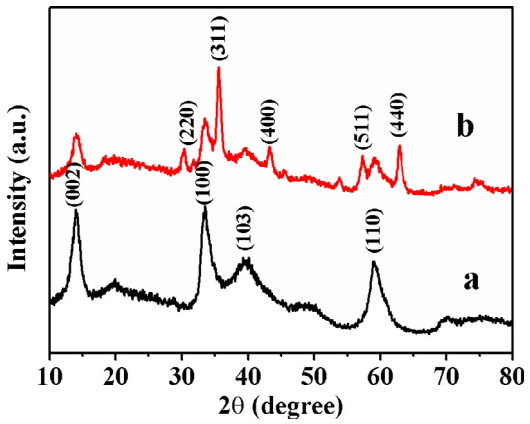

Fig. 1. XRD patterns of MoS_2/graphene (a) and MoS_2/Fe_3O_4/graphene (b).

FESEM and TEM were employed to observe the morphology and microstructure of the as-prepared samples. From Fig. 3(a, b), it is obvious that MoS_2/graphene composite displays a homogeneous flower-like structure, which is composed of intertwined sheet-like structure. The flower-like structure can enlarge the specific surface area and provide more absorption sites, which is beneficial to the microwave absorption performances. As can be observed in Fig. 3(c, d), during the co-precipitation route, Fe_3O_4 nanoparticles with a size of around 20–30 nm were grown in the cavity of MoS_2/-graphene. The adding of Fe_3O_4 particles does not destroy the 3D flower-like structure. TEM was used to further obtain microstructure information of the samples. From Fig. 4(a, b), MoS_2/graphene composite displays a hydrangea-like structure and the particle size agrees with FESEM analysis. It is found from Fig. 4c that the edge of MoS_2/graphene is wrinkled and transparent. As depicted in Fig. 4 (d–f), we can observe clearly that the transparent edge of MoS_2/-graphene is covered with Fe_3O_4 particles with a uniform distribution. Based on the above analysis, it can be inferred that the MoS_2/Fe_3O_4/graphene composite were successfully fabricated.

The magnetic properties are crucial to EM wave absorption performances and the magnetic parameters of Fe_3O_4, MoS_2/Fe_3O_4/-graphene were tested at room temperature by VSM. As shown in Fig. 5, both the two samples display a typical superparamagnetic behavior, which possess negligible remanence and coercivity. The saturation magnetization (M_S) values of Fe_3O_4, MoS_2/Fe_3O_4/-graphene are 84.1 emu/g and 33.4 emu/g, respectively. The great difference in M_S values of the two samples may be ascribed to the presence of graphene and MoS_2.

The EM wave absorption performances of the MoS_2/graphene, MoS_2/Fe_3O_4/graphene were investigated at 2–18 GHz and the

Please cite this article in press as: Y. Wang et al., Fabrication of MoS_2-graphene modified with Fe_3O_4 particles and its enhanced microwave absorption performance, Advanced Powder Technology (2017), https://doi.org/10.1016/j.apt.2017.12.016