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Preparation of SnO_2-coated carbonyl iron flaky composites with enhanced microwave absorption properties

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ABSTRACT

The SnO_2-coated carbonyl iron (CI) (SCCI) composites were synthesized by combining ball milling and mild hydrothermal treatment and their microwave absorption performance at 2–18 GHz was evaluated. The results showed that the microwave absorption properties of SCCI were improved remarkably due to the existence of the uniform coating of SnO_2 on the surface of the flattened CI particles. The strongest reflection loss (RL) of SCCI reached −57.8 dB with a thickness of 8.3 mm at 12.1 GHz, and strong absorption (RL < −10 dB) was obtained between 7.2 and 18 GHz with the thickness of 7.0–13.0 mm. Furthermore, at a low thickness of 2.0–2.9 mm, the SCCI still showed strong microwave absorption property and its maximum RL value reached −20.5 dB with a thickness of 2.3 mm at 15.7 GHz. This SCCI composites possessed excellent microwave absorption properties and could become a promising candidate for the application of microwave absorption.

1. Introduction

With the increasingly serious electromagnetic pollution problems, especially in gigahertz range, a growing and widespread research interest has been focused on developing new types of microwave-absorbing material. CI powder has traditionally been widely used as microwave-absorbing material due to its low cost, high magnetic losses and high specific saturated magnetization properties. However, low microwave permittivity value of monodispersed CI particles is an obvious disadvantage [1,2]. It has been reported that microwave absorption properties could be enhanced via the complementation of dielectric and magnetic loss [3–5]. Among the dielectric loss materials, tin oxide (SnO_2), an inexpensive large band gap (3.6 eV) semiconductor, has environmentally stable dielectric properties [6]. Although SnO_2 and its composites have been extensively studied and reported for their application of electrode, studies on their microwave absorption properties are scarce [7–9].

Generally, ball milling technique and chemical methods are used to fabricate coating materials. However, the ball milling technique does not provide subtle control over the coating layer, and the chemical methods are too complex to use [4,6]. Therefore it is essential for us to find a way around the defects of these common techniques. In our work, we utilized a combined method of ball milling and mild hydrothermal treatment, through which the CI particles became flaky-shaped and the in-situ growth of SnO_2 particle layer was achieved on the surface of CI particles. The structure and microwave absorption properties of the resultant SCCI were investigated.

2. Experimental

All reagents, including CI powder and tin (Sn) powder, were obtained from commercial suppliers and used without further purification. The commercial CI powder and 1 wt% Sn powder were initially milled together at 400 rpm for 5 h in a planetary ball mill under argon atmosphere. The product obtained was calcined at 200 °C for 5 h and then added to a 50 mL aqueous solution containing 1.0 mmol of $KBrO_3$ and 5.0 mmol of NaOH. After that, the mixed solution was transferred into a 60 mL Teflon-lined stainless steel autoclave, which was maintained at 140 °C for 6 h. After the reaction was done, the final product, i.e., SCCI was collected by a magnet, rinsed with deionized water to neutral, and dried in air at 40 °C.

The SCCI samples were characterized by X-ray powder diffraction (XRD, Rigaku D/MAX-2500, with Cu Kα) and Fourier transform infrared (FT-IR, Bruker TENSOR 27) spectroscopy. Scanning electron microscopy (SEM, FEI Nanosem 430) and high resolution transmission electron microscopy (HRTEM, FEI Tecnai G2 F20) were used for structure and morphology analyses. Energy dispersive spectroscopy (EDS) attached with the SEM was used for elemental analysis. The complex permittivity $\varepsilon_r = \varepsilon' - j\varepsilon''$ and permeability $\mu_r = \mu' - j\mu''$ of the composites were measured between 2 and 18 GHz on a vector network analyzer (Agilent E8363B), where ε' and ε'' are the real and imaginary part of

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permittivity, and μ' and μ'' are the real and imaginary part of permeability, respectively. The sample for electromagnetic parameter measurement was prepared by mixing CI/SCCI with wax in a mass ratio of 1:1 and compressing into a 2 mm-thick ring form of 7 mm outer and 3 mm inner diameters.

3. Results and discussion

All the diffraction peaks shown in Fig. 1a can be indexed as body-centered cubic iron (JCPDS: 65-4899). Due to low coating amount, no peak corresponding to SnO_2 is observed [10]. However, the FT-IR spectrum of SCCI shows a peak at around 560 cm^{-1} which refers to the vibration of O–Sn–O groups. This peak is an indication of the presence of SnO_2 [11]. Besides, both the CI and SCCI FT-IR spectra display peaks at 3457 and 1632 cm^{-1}, which are assigned to the hydroxyl bands [11–13]. The impurities of the commercial CI give rise to the unwanted absorption peaks that can be ignored.

Fig. 2(a) shows that the raw CI particles are spherical with smooth surface and aggregate easily. After being milled, they became flaky and de-aggregated (Fig. 2b). The SCCI morphology shown in Fig. 2(c) and (d) reveals that the flaky CI particles are coated well by SnO_2 particles with an average size of 200 nm.

EDS result (Fig. 2e) indicates the presence of Fe, O, Sn, C and Au elements (Au signal from sputter-coating for SEM observation). The HRTEM image (Fig. 2f) exhibits the clear fringes of *d*-spacing value of 0.34 nm, corresponding to (110) plane of the tetragonal phase SnO_2.

From the above findings, we can propose the formation of the SCCI. Sn coatings were formed on the flaky CI particles after ball milling [10,14]. During the hydrothermal process, the Sn particles

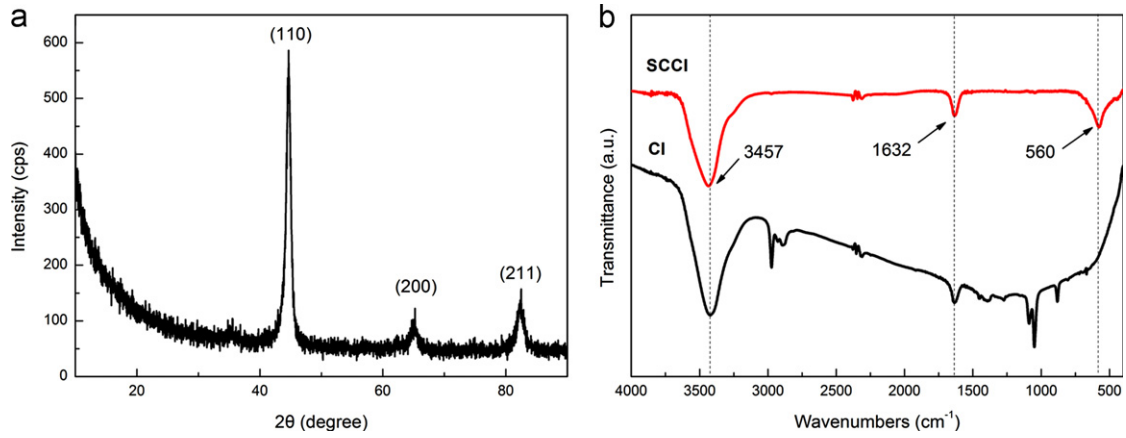

Fig. 1. (a) XRD pattern of the SCCI and (b) FT-IR spectra of CI and SCCI.

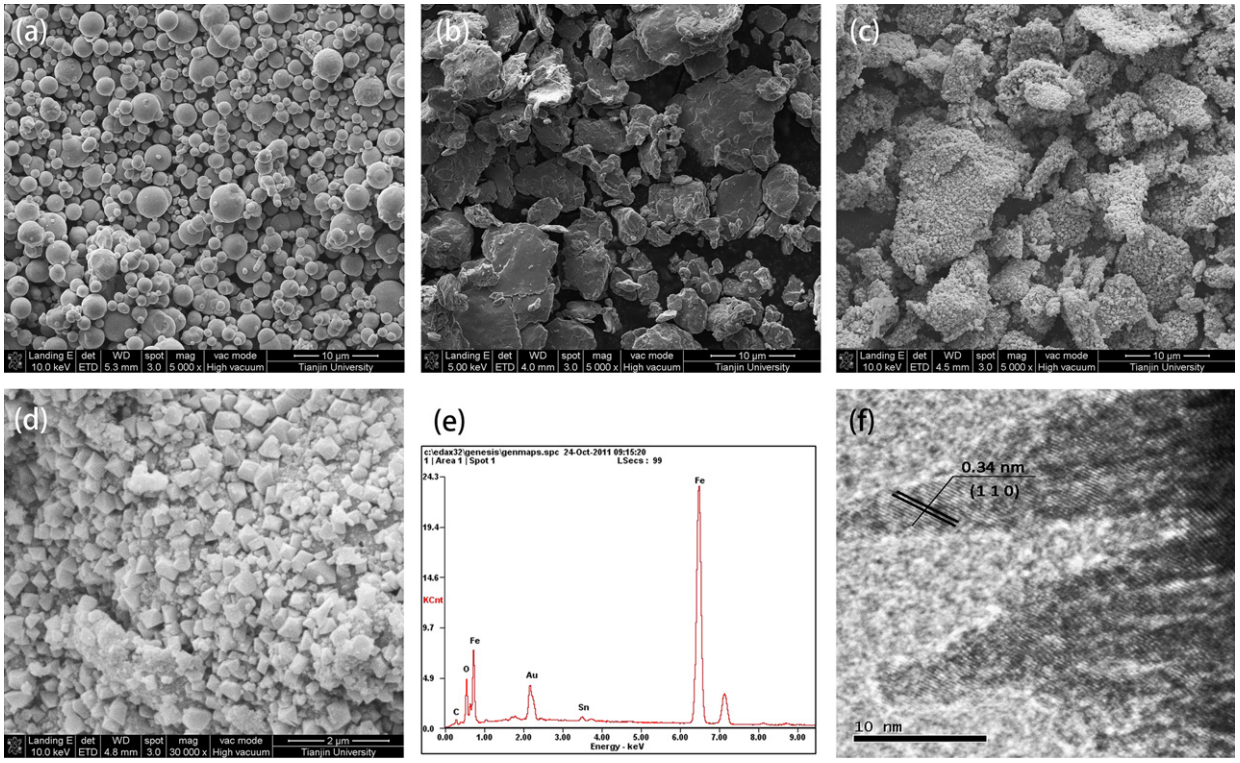

Fig. 2. SEM images of (a) CI and (b) as-milled CI; (c) low and (d) high magnification SEM images, (e) EDS graph and (f) HRTEM image of SCCI.