Journal of Magnetism and Magnetic Materials 340 (2013) 65–69

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

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

Microwave absorption and antioxidation properties of flaky carbonyl iron passivated with carbon dioxide

Chunlei Yin [a,b], Junmei Fan [a], Liuyang Bai [a], Fei Ding [a], Fangli Yuan [a,*]

[a] State Key Laboratory of Multi-phase Complex Systems, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, PR China
[b] University of Chinese Academy of Sciences, Beijing 100049, PR China

ARTICLE INFO

Article history:
Received 8 October 2012
Received in revised form
19 February 2013
Available online 8 April 2013

Keywords:
Carbonyl iron
Microwave absorption
Carbon dioxide
Passivation

ABSTRACT

Carbonyl iron particle is a typical magnetic microwave absorbing material. Because of the effect of particle shape, the flaky carbonyl iron has large values of microwave permeability and can be an excellent super-thin absorber. However, the flaky carbonyl iron prepared by high-energy ball milling has high activity and is apt to be oxidized. Based on a reverse reaction of the blast furnace iron-making process, this work developed a facile passivation method with carbon dioxide to improve the antioxidation property of the flaky carbonyl iron. The antioxidation property of the passivated carbonyl iron was improved greatly while the excellent microwave absorption property maintained.

1. Introduction

There has been a growing interest in absorbers of electromagnetic waves because of their widespread applications in the civil and military fields with the fast advancement of wireless communications [1–5]. Generally, there are two types of microwave absorption materials: dielectric materials and magnetic materials. Carbonyl iron particle, having the onion skin structure and the high purity, is a typical magnetic microwave absorbing material [6–8]. With the advantages of higher saturation magnetization than ferrites and low eddy-current loss due to the effect of particle shape, the flaky carbonyl iron particles could have large values of microwave permeability [9–12]. For magnetic absorbers, high values of permeability can benefit broader bandwidth, higher absorption and thinner thickness. So the flaky carbonyl iron particles have the potential to be developed as a super-thin (below 1 mm) microwave absorber [13]. High-energy ball milling is the most popular method to manufacture flaky metals [14]. However, the flaky carbonyl iron prepared by the method of high-energy ball milling has high activity and is apt to be oxidized or even to be spontaneously ignited. The oxidation of flaky carbonyl iron will decrease the property of absorption seriously. Controllable passivation is a useful method to prevent ultra-fine iron powder from exceeding oxidization or even spontaneous combustion by forming a shielding layer on the surface. Passivation of iron by oxidation in H_2O vapor or O_2/H_2O vapor mixtures, a traditional method to passivate high activity iron powder, has some disadvantages such as strict operation condition, secondary oxidation and high operating temperature [15].

This work presents a new facile passivation route to improve the antioxidation property of the flaky carbonyl iron, and the mechanism of passivation of flaky carbonyl iron is discussed.

2. Experimental

The raw material was sphere-shaped carbonyl iron powder, purchased from Tianyi Ultra-fine Metal Powder Co. Ltd., Jiangsu province, China. The sphere-shaped carbonyl iron powder and ethanol were loaded together with multiple diameter steel balls in a steel container. The weight ratio of powder and ball charge was 1:10. The mechanical milling was performed using a QM-QX4 planetary mill for 12 h at 400 rpm. The slurry and the balls were separated by a sieve. The as-milled powder i.e. the flaky carbonyl iron was filtered off and dried at room temperature. The passivated flaky carbonyl iron was prepared by bubbling carbon dioxide in the slurry at certain flow rate and certain time.

The crystalline phase of the samples was characterized by X-ray diffractometry (XRD, X'pert PRO, Panalytical, CuKα radiation) in a 2θ range from 5° to 90°. Their size and morphology were observed with field scanning electron microscopy (FESEM, JEOL JSM-6700F).

The prepared powders were homogenously dispersed in paraffin with a mass fraction of 80% (powder) and then formed into a cylindrical sample (7 mm in outer diameter and 3 mm in inner diameter) with a coaxial mold, respectively. The effective complex

* Correspondence to: No. 1 Zhongguancun North Second Street, Beijing 100190, PR China. Tel.: +86 10 82544974; fax: +86 10 62561822.
E-mail address: flyuan@home.ipe.ac.cn (F. Yuan).

0304-8853/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jmmm.2013.03.038

permittivity ($\varepsilon_r=\varepsilon'-j\varepsilon''$) and permeability ($\mu_r=\mu'-j\mu''$) of these samples in the frequency range from 2 to 18 GHz were measured using an HP-8722ES network analyzer. The reflection loss was calculated according to the transmission line theory [10], expressed as follows:

$$R = 20\log\left|\frac{Z_{in}-1}{Z_{in}+1}\right| \quad (1)$$

Z_{in} is the normalized input impedance of a metal-backed microwave absorbing layer.

$$Z_{in} = \sqrt{\frac{\mu_r}{\varepsilon_r}}\tanh\left[j\left(\frac{2\pi f d}{c}\right)\sqrt{\mu_r\varepsilon_r}\right] \quad (2)$$

where f is the frequency of the electromagnetic wave, d is the thickness of the absorber, c is the velocity of light.

The oxygen content of the passivated carbonyl iron sample was measured using an oxygen and nitrogen analyzer (Beijing NCS Testing Technology Co., Ltd., ON-3000). The antioxidation property of the samples was recorded using a thermal analyzer (Netzsch STA 449 TG-DTA/DSC) in air with the heating rate of 10 °C/min up to 800 °C.

3. Results and discussion

3.1. Characteristics of the samples

Fig. 1a shows the FESEM image of the initial carbonyl iron, i.e. the raw material. The sample is composed of well-dispersed spheres, and the average diameter is about 3–5 μm. Fig. 1b, c shows the FESEM image of the flaky carbonyl iron. The as-milled powders exhibit a typical microstructure of mechanically milled materials, that is, thin flake shape and a relatively broad distribution of particle size (the thickness is less than 100 nm and the length is smaller than 20 μm). Fig. 1d shows the FESEM image of the passivated carbonyl iron. It is observed that the morphology of the passivated carbonyl iron is similar to the flaky carbonyl iron, indicating that the passivation process has no apparent influence upon the microstructure of the flaky carbonyl iron.

Fig. 2 presents XRD patterns of the initial carbonyl iron sample, the flaky carbonyl iron sample and the passivated carbonyl iron sample, respectively. Compared with the initial carbonyl iron sample, the relative intensities of the diffraction peaks of the flaky carbonyl iron sample decrease, while the peaks are significantly broadened, demonstrating that large amounts of defects are introduced into the flaky carbonyl iron sample. The patterns of the flaky carbonyl iron sample and the passivated carbonyl iron sample are almost the duplicates of each other. It can be concluded that there is no difference in the inner structure between the flaky carbonyl iron sample and the passivated carbonyl iron sample.

3.2. Microwave absorption property of the samples

Fig. 3 depicts the complex permittivity and complex permeability spectra of the initial carbonyl iron sample, the flaky carbonyl iron sample and the passivated carbonyl iron sample, respectively. As shown in Fig. 3a and b, for the flaky carbonyl iron

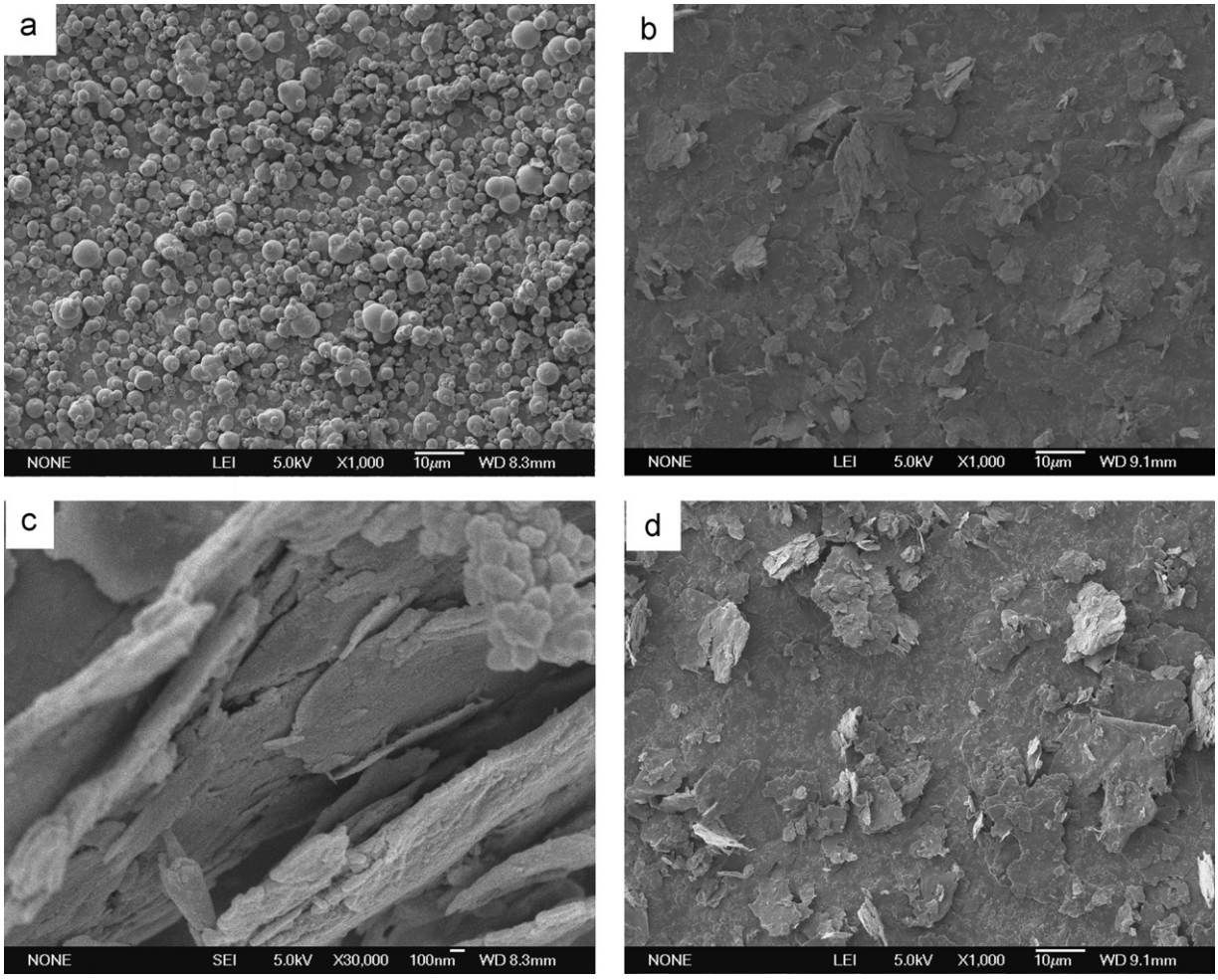

Fig. 1. FESEM image of samples (a) initial carbonyl iron; (b), (c) flaky carbonyl iron; (d) passivated carbonyl iron.