

One-step and green synthesis of metal doped spinel ferrite (Ni,Zn)Fe₂O₄ with high magnetic property from Zn-containing electric arc furnace dust

Min Guo*, Hui-gang Wang, Mei Zhang

School of Metallurgical and Ecological Engineering, University of Science and Technology Beijing, Beijing, 100083, China

Keywords: Zn-containing electric arc furnace dust, Solid state reaction method, Spinel ferrite (Ni,Zn)Fe₂O₄, Magnetic property

Presenting author email: guomin@ustb.edu.cn

Introduction

Zn-containing electric arc furnace dust (EAFD), a solid waste produced during steelmaking process, contains many valuable metals, such as Fe, Zn, Mn, Ca, etc. (Kukurugya et al., 2015; Wang et al., 2016b). And the dust is classified as hazardous wastes according to the US Environmental Protection Agency because of the presence of Cr, Pb, etc. (Ann M. Hagni et al., 1991; Wang et al., 2016a). Therefore, it is becoming increasingly important to find an environmentally friendly solution to treat the EAFD for sustaining a cleaner and greener environment.

Up to now, two main methods for treatment of Zn-containing EAFD include solidification/stabilization (S/S) process and recovery of valuable metals. However, the point of the former is the immobilization of hazardous elements, such as Cr, Pb, etc., and the later only pays attention to the pursuit of high added value. Little study has focused on the above two aspects simultaneously.

Spinel ferrites such as MFe₂O₄ (M: Ni, Zn, Mn, Ca, etc.) have attracted considerable interest and efforts due to their stability, novel magnetic/electric properties, and great applications in different fields (Srinivasan et al., 2009). Among them, Ni-Zn spinel ferrite, which has a high saturation magnetization (Ms) and low coercivity (Hc), has been widely used as soft magnetic materials, leading to many studies focused on obtaining high Ms and low Hc. Considering the fact that elements such as Fe, Zn, Cr, and Mn coexisted in the Zn-containing EAFD, it is reasonable to assume that by adjusting the components of the dust, the mole ratio of Fe (Fe, Cr, Al) to M of around 2.0 can be obtained, resulting in the direct synthesis of spinel ferrites from the dusts.

In this study, one-step solid state reaction method was proposed for the first time to realize green and simple synthesis of ferrite from Zn-containing EAFD through the addition of NiCl₂·6H₂O. Single-phase metal doped spinel ferrite (Ni,Zn)Fe₂O₄ was obtained after the calcined sample was washed by dilute HAc solution. The calcined temperature played an important role in determining the phase structure and magnetic properties of as-prepared MFe₂O₄. In addition, the hazardous elements were bound to the lattice of the ferrite, realizing the harmless treatment of the EAFD. This work may explore an environmentally friendly and simple pathway for the high value-added utilization of Zn-containing EAFD.

Results and discussion

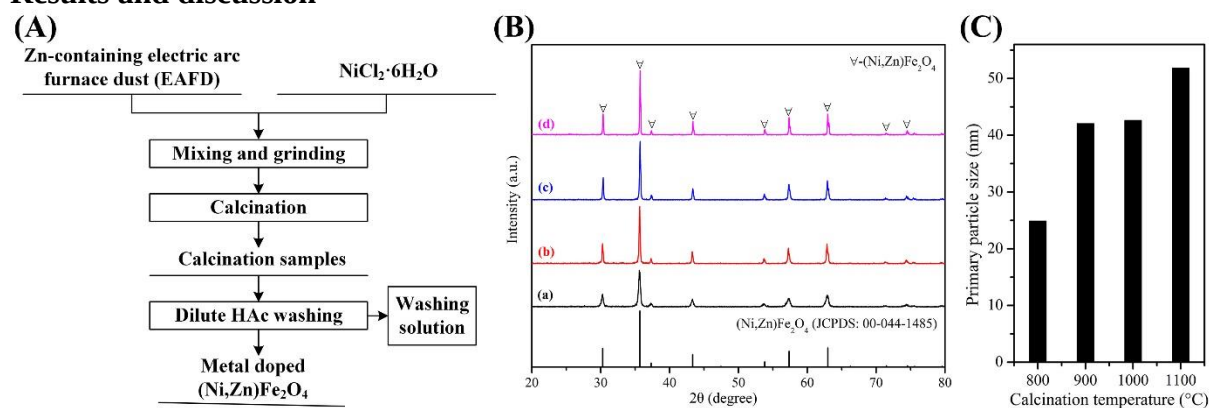


Fig. 1. (A) General flow sheet of the synthesis of (Ni,Zn)Fe₂O₄ from Zn-containing EAFD, (B) XRD patterns of the synthesized ferrites under different calcination temperatures (a) 800 °C, (b) 900 °C, (c) 1000 °C, (d) 1100 °C and (C) primary particle sizes calculated according to XRD results. (Other experiment conditions: R_{ZE/N} of 2:1.2, time for 2 h)

Based on theoretical calculation and pre-experiments, the mass ratio of Zn-containing EAFD to NiCl₂·6H₂O was controlled at 2:1.2 (R_{ZE/N}, g/g). Fig. 1(A) illustrates the general flow sheet of synthesizing (Ni,Zn)Fe₂O₄ from Zn-containing EAFD. Fig. 1(B) gives the XRD patterns of the synthesized ferrites under different calcination

* Corresponding author. Tel./Fax: +86 10-62334926
E-mail address: guomin@ustb.edu.cn (Min Guo)

temperatures. It can be seen from Fig. 1(B) that all of the diffraction peaks for each sample were well matched with the standard diffraction peaks of $(\text{Ni,Zn})\text{Fe}_2\text{O}_4$ with cubic spinel structure (JCPDS: 00-044-1485), and no diffraction peaks of any other phases were detected, suggesting that pure $(\text{Ni,Zn})\text{Fe}_2\text{O}_4$ were obtained under the suitable conditions. In addition, the inductively coupled plasma atomic emission spectrometer (ICP-AES) analysis results showed that elements Mn, Ca, Al, Cr, etc. besides Ni, Zn and Fe coexisted in the obtained $(\text{Ni,Zn})\text{Fe}_2\text{O}_4$, which confirmed that the pure $(\text{Ni,Zn})\text{Fe}_2\text{O}_4$ was indeed multi-metal co-doped ferrite MFe_2O_4 (Fe: Fe, Al, Cr; M: Ni, Zn, Mn, Ca, and etc.). Moreover, the primary particle size d of the samples increased rapidly from 24.88 to 51.84 nm with temperature increasing from 800 to 1100 °C as shown in Fig. 1(C).

The magnetic properties of the synthesized metal doped $(\text{Ni,Zn})\text{Fe}_2\text{O}_4$ are illustrated in Fig. 2. It can be seen that the M_s value increased from 46.67 to 61.76 emu/g with the calcination temperature from 800 to 1100 °C. According to Neel's two sublattice model, the enhanced magnetic property (higher M_s value) may be mainly ascribed to the more magnetic ions (Ni^{2+} ions etc.) migrating from A-site to B-site based on Raman results. In addition, due to the inverse relationship between particle size and H_c (Gharagozlou, 2009), with the temperature rising, the H_c value decreased from 81.72 to 34.43 Oe accordingly. Moreover, it should be noted that the obtained metal doped $(\text{Ni,Zn})\text{Fe}_2\text{O}_4$ exhibited better magnetic properties compared to those prepared by pure chemical reagents (A. Verma et al., 2000; Gabal et al., 2012). More importantly, the as-prepared ferrites were harmless according to the result of toxicity character leaching procedure (TCLP), realizing the green and high value-added utilization of the dust.

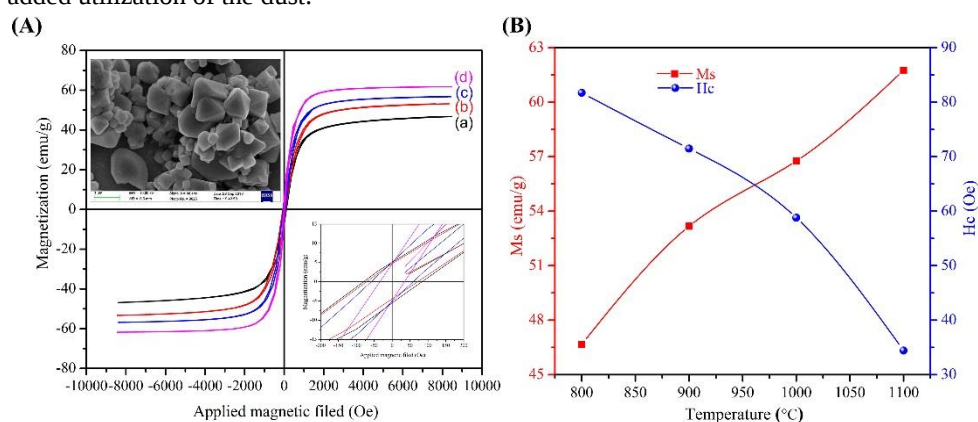


Fig. 2. (A) Room temperature hysteresis loops and (B) variation of the M_s and H_c values of synthesized ferrites under different calcination temperatures (a) 800 °C, (b) 900 °C, (C) 1000 °C, (d) 1100 °C (other experiment conditions: $R_{\text{Zn/N}}$ of 2:1.2, time for 2 h)

Acknowledgements

This work was financially supported by the National Basic Research Priorities Program of China (No. 2014CB643401, and No. 2013AA032003), the National Natural Science Foundation of China (No. 51672025, and No. 51372019), and Shanxi Collaborative Innovation Center of High Value-added Utilization of Coal-related Wastes.

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