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Characterization of Ni–Cu–Zn ferrite prepared from industrial wastes

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Abstract

We propose the distillation method to synthesize Ni–Cu–Zn ferrite powder and to recover nitric acid, using scrap iron and the waste solution of electroplating as the starting materials. It was found that the Ni–Cu–Zn ferrite powder prepared from industry wastes also showed the formation of cubic ferrite with a saturation magnetization (M_s) of 55,825 A m^2 g^{-1} and an intrinsic coercive force (Hci) of 579 A m^{-1}. For sintered Ni–Cu–Zn ferrite specimen, the toroidal specimen sintered at 950 °C for 2 h presented an maximum initial permeability (μ_i) of 176 at 28.3 MHz, a maximum quality factor (Q) of 32 at 3 MHz. The AC impedance measurements were performed by using impedance analyzer Solartron 1260. The semicircles in the impedance spectra shift to higher frequencies with increasing temperatures. The values of resistance (grain interior, grain boundary, and total) decreased with increasing temperatures. The semicircles of grain boundary and electrode are observed clearly. These data can be used to analyze typical the grain interior and the grain boundary resistance of Ni–Cu–Zn ferrite.

Keywords: A. Powder: chemical preparation; C. Magnetic properties; D. Ferrites; D. Spinels; E. Soft magnets

1. Introduction

Ni–Cu–Zn ferrite is usually used as magnetic material for multilayer chip inductors due to its lower sintering temperature and better properties at high frequency than Ni–Zn ferrite [1]. The conventional way of producing these materials is by the solid-state reaction of oxides/carbonates which are calcined at high temperatures [2,3]. Our group had investigated to synthesize Ni–Cu–Zn ferrite powder by hydrothermal process from the steel pickling liquor and the waste solution of electroplating for several years [4,5]. We attempted a new process (distillation method) to synthesize Ni–Cu–Zn ferrite powder and recovery nitric acid, using the scrap iron, electroplating waste solutions of nickel, zinc and copper as the starting materials. Nickel nitrate, copper nitrate and zinc nitrate waste solutions come from nickel, copper, and zinc electroplating plants, respectively. All these waste solutions must be treated to correspond to the environmental law and regulation in most industrialized countries. It involves high cost for the producers to treat these waste solutions. If we can effectively use recycled resources such as the scrap iron and the waste solutions of electroplating as the starting materials for Ni–Cu–Zn ferrite, then we can contribute to the protection of the earth and reduce the amount of industrial waste solutions.

2. Experimental procedures

The flow chart of experimental procedure is given in Fig. 1. Firstly, the concentration of cations in the electroplating waste solutions of nickel, copper, and zinc were analyzed by inductively coupled plasma (ICP). Secondly, the scrap irons and electroplating waste solutions were mixed together and heated until the scrape iron is completely dissolved in the mixed solution. The concentration of cations in the mixed solution was adjusted by adding the chemicals in accordance with ICP analysis. The distillation process involves heating the mixed solution consisting of the scrap iron, and electroplating waste solutions of nickel, zinc and copper. According to ion chromatography analysis, it was found that the condensate contained the nitrate ion. It is confirmed that these condensed liquid is nitric acid. Accordingly, we can recover the nitric acid from the distillation and recycle to use these nitric acids in other industry. During the distillation procedure, the great part of the mixed solution recovered in form of nitric acid and produced a

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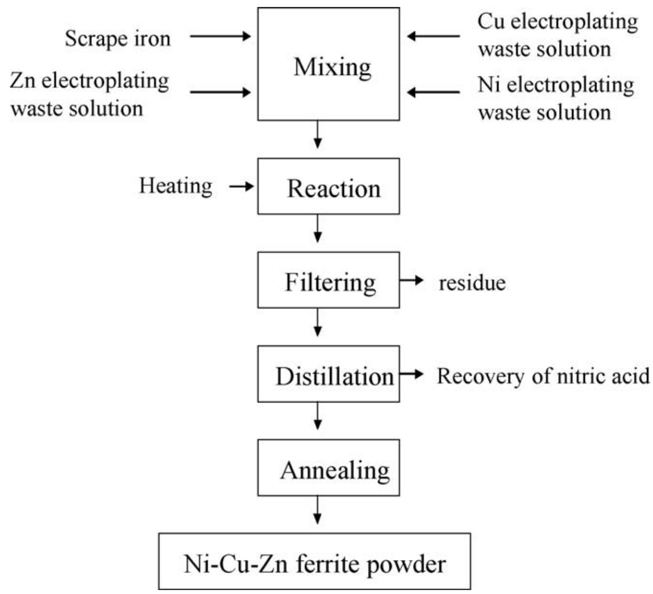

Fig. 1. Flow chart for preparation of Ni–Cu–Zn ferrite powder by distillation process from industrial wastes.

small part of residue. We can regard these residues as the precursor of Ni–Cu–Zn ferrite. Finally the Ni–Cu–Zn ferrite powder was obtained by annealing at 700 °C for 2 h. Then these ferrite powders were granulated and pressed into toroidal specimens under a uniaxial pressure of 1000 kg/cm^2. The toroidal specimens were then sintered at 950 °C for 2 h at a rate of 5 °C.

A computerized X-ray powder diffractometer (XRD) with Cu Kα radiation (XRD; Rigaku D/Max-II, Tokyo, Japan) was used to identify the crystalline phase. A vibrating sample magnetometer (VSM; Lake Shore 7407, Westerville, OH) was used to measure the saturation magnetization (M_s) and intrinsic coercivity (Hc) of Ni–Cu–Zn ferrite powder. Scanning electron microscopy (SEM; Hitachi S-3500H, Tokyo, Japan) was used to study the microstructure of Ni–Cu–Zn ferrite. The initial permeability (μ_i) and quality factor (Q) of sintered Ni–Cu–Zn ferrite were measured on an Hewlett-Packard 4194A impedance analyzer (Agilent, Santa Clara, CA) in the frequency range of 1 kHz–100 MHz; 15 turns of coil were wound around the sintered toroidal specimens. The impedance measurements were performed with an impedance analyzer SI 1260 (Solartron analytical, Hampshire, UK) over 1 Hz–10 MHz frequency range on isothermal plateaus half an hour long and the measurement temperature ranged from 250 °C to 350 °C with an increment of 50 °C.

3. Results and discussion

Fig. 2 shows the X-ray diffraction patterns of the annealed Ni–Cu–Zn ferrite powder and sintered Ni–Cu–Zn ferrite specimen. It revealed that annealed powders and sintered specimens contain only the spinel cubic ferrite. All the peaks in the pattern match well with the Joint Committee of Powder Diffraction Standard (JCPDS) card. No impurity phases can be detected in the X-ray pattern. The magnetization measurement

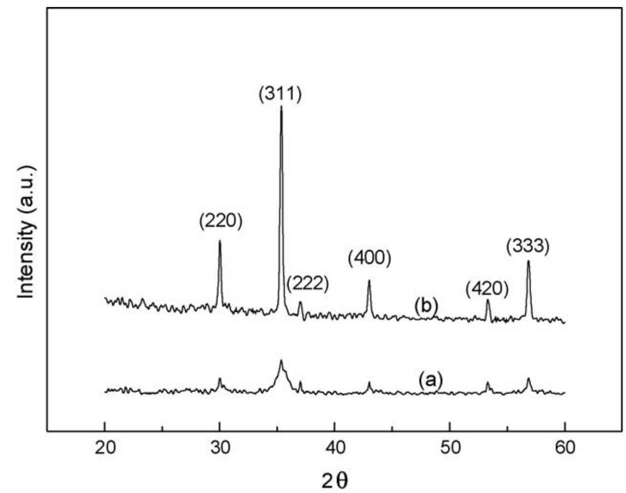

Fig. 2. XRD powder patterns of (a) annealed Ni–Cu–Zn ferrite powder, and (b) sintered Ni–Cu–Zn ferrite specimen.

for Ni–Cu–Zn ferrite powder prepared by distillation method was carried out using a vibrating sample magnetometer at room temperature with an applied magnetic field of 10 kOe to reach the saturation values. Fig. 3 shows hysteresis loops for annealed Ni–Cu–Zn ferrite powder. It indicates that the annealed Ni–Cu–Zn ferrite is a soft magnetic material, which reveals minimal hysteresis. The annealed Ni–Cu–Zn ferrite powder revealed the formation of cubic ferrite with a saturation magnetization (M_s) of 55825 Am^2 g^{-1} and an intrinsic coercive force (Hci) of 579 A m^{-1}.

It is well known that the permeability of spinel ferrite is strongly affected by saturation magnetization, crystal magnetization anisotropy, magnetostriction constant and internal stress. The equation of initial permeability is expressed as follows:

$$\mu_i = \frac{M_s^2}{aK + b\lambda\sigma}$$

where μ_i is initial permeability, M_s is saturation magnetization, K is crystal magnetic anisotropy, λ is magnetostriction constant,

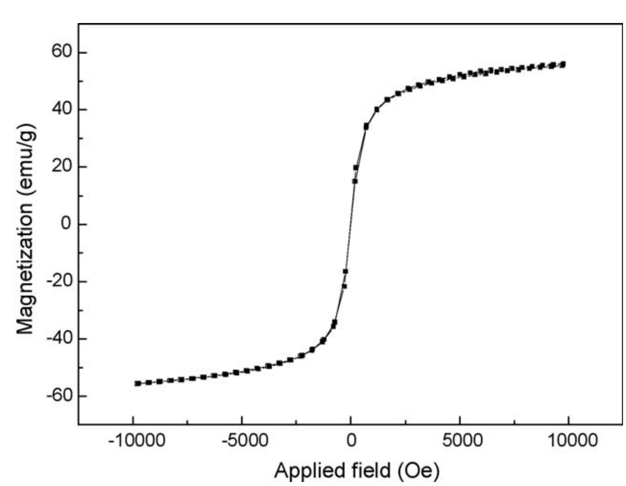

Fig. 3. Magnetization against applied field hysteresis curve for as-prepared Ni–Cu–Zn ferrite powder annealed at 700 °C for 2 h.