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Milling time and BPR dependence on permeability and losses of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ synthesized via mechanical alloying process

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$Ni_{0.5}Zn_{0.5}Fe_2O_4$ has been synthesized using mechanical alloying method with two variables (milling time and ball-to-powder weight ratio (BPR)) were varied in order to study its effect on the magnetic properties of the material. The effects of these two variables were studied using XRD, SEM, TEM and later by impedance analyzer with the frequency range from 1 MHz to 1.8 GHz. The results obtained however show that there are no significant trends to relate the milling time and BPR with the permeability and losses of the material studied. After being sintered at 1150 °C, all the effects of alloying process seem to diminish.

1. Introduction

Increasing interests in the study of soft ferrites such as NiZn ferrite, which is the most versatile magnetic materials and has been used for many years. NiZn ferrites are soft ferrimagnetic materials having low magnetic coercivity and high resistivity values [1] and little eddy current loss in high frequency operations (10–500 MHz) [2]. Magnetic properties of NiZn ferrites rely heavily on the chemical composition and are also sensitive to their microstructure [3,4]. There are a few factors that determine the microstructure of ferrites such as the quality of raw materials, the calcinations temperature, the milling procedure and the sintering regime [5]. Generally, the parameters of microstructure are the grain size, the pore size, the porosity and the density, intra-granular and inter-granular distribution of pores and grains. The quantity, size, shape and distribution of both crystal grains and pore of a ferrite will vary with different preparation conditions and techniques [6,7]. One of the various techniques for synthesizing magnetic materials is mechanical alloying which is used to reduce particle size, mix powder uniformly and make non-equilibrium structure materials, including nanocrystals, quasicrystals and amorphous alloys. Mechanical alloying via high-energy ball milling has now become one of the conventional methods for producing nano/non-crystalline materials. We intent to employ mechanical alloying process to work at reasonable time by varying the milling time and also the ball-to-powder weight ratio (BPR). These parameters are varied to study the useful result within time range of 24 h in order to achieve nanosize particle. However for research purpose, we extended the process up to 48 h. This time limit is useful if we want to make it relevant for practical industrial purposes. Bid and Pradhan [8] discovered nanocrystalline $Ni_{0.5}Zn_{0.5}Fe_2O_4$ synthesized via mechanical alloying just within 11 h with ball-to-powder weight ratio of 40:1. They also found that NiO has slower diffusion in Fe_2O_3 lattice than ZnO. The goal of this work is to study the effects of BPR and also the milling time variation on the permeability of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ synthesized via mechanical alloying. To the best of our knowledge, the BPR and milling time as a function of permeability of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ synthesized via mechanical alloying has not been reported yet especially within the frequency range from 1 MHz up to 1.8 GHz.

2. Experimental details

A composition of powder for $Ni_{0.5}Zn_{0.5}Fe_2O_4$ was prepared by mechanical alloying of a mixture of metallic oxides. The materials used were Fe_2O_3 (Alfa Aesar) (99.95%), NiO (Alfa Aesar) (99.99%) and ZnO (Alfa Aesar) (99.99%) weighed according to the composition formula. The chemicals were mixed with chosen molar ratio of 1:0.5:0.5. The ball-to-powder mass–charge ratio (BPR) were varied from 4:1, 6:1, 8:1, 10:1, 12:1, 14:1, 16:1, 18:1 and 20:1 with 4 h milling time for every BPR. High-energy milling was carried out in a SPEX 8000D shaker mill in ambient atmosphere

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for 1, 4, 8, 12, 16, 20, 24, 30, 36, 42 and 48 h and 10:1 BPR was chosen for every milling time carried out.

All the samples were examined with X-ray diffraction (Phillips Expert Pro PW3040) using CuKα. The Rietveld's powder structure refinement analysis of X-ray powder diffraction data was adopted in order to refine microstructural parameter such as crystallite size and micro-strain using X'pert Highscore Plus (PANalytical, Almelo, The Netherlands) using the full range of data. The profile was fitted with the Pearson VII function. The following parameters were fitted: zero shift, background, scaling factor, Pearson VII coefficients, lattice parameters and preferred orientation [9,10].

Two samples which were 12 and 24 h of milling were sent for Scanning Electron Microscope (SEM) images (JEOL 6400), and three samples which were 12, 14 and 48 h of milling were examined under a Transmission Electron Microscope (TEM) (LEO 912AB). The samples milled with BPR 4:1, 6:1, 8:1, 10:1, 12:1, 14:1, 16:1, 18:1 and 20:1 were formed into toroidal-shape samples without any binder aid. This was also done for samples milled for 1, 4, 8, 12, 16, 20, 24, 30, 36, 42 and 48 h. All powder handling, milling and subsequent pressing and sintering were performed under air environment. All samples with various BPR and milling time were employed and characterized using impedance analyzer (Agilent Model 4291B) from 1 MHz to 1.8 GHz. The two series of samples (various BPR and milling time) were later sintered at 1150 °C using a heating rate of 4 °C/min and once again were studied using impedance analyzer with the same setting as previously done.

3. Results and discussion

A closer look at the peaks in Fig. 1 showing that the effect of different BPR ratio via mechanical alloying caused to the broadening of X-ray diffraction peaks. The effect of XRD broadening increases with the increasing of BPR ratio. This shows that the formation of fine grain and a high density of defects caused by large local strains in the powder particles. The higher the BPR, the shorter is the time required [11]. At a high BPR, due to an increase in weight proportion of the steel balls, the number of collision per unit time increases and consequently more energy is transferred to the powder particles and so alloying takes place faster [11]. The crystallite size and micro-strain as a function of BPR were calculated using the Rietveld's method shown in Fig. 2. From the results it can be seen that the crystallite size decreases with the increasing of BPR. The early part of BPR variation shows the fracture and deformation process and the intermediate and final BPR variation continued with the cold welding process with crystalline sizes of 40.6, 38.0, 27.7, 20.9, 23.5, 17.0, 13.9, 14.6, 15.8 nm for BPR of 4:1, 6:1, 8:1, 10:1 12:1 14:1 16:1 18:1 and 20:1, respectively. Since mechanical alloying involves the synthesis of materials by high-energy ball milling, the repeated collision between balls and powders with very high impact velocity deform and work-harden the powder. In this repetitive cold welding and fracturing mechanism, cold welding of overlapping particles occurs between clean surfaces formed by prior fractures. The competing process of deformation, fracture and welding during milling produces a microstructural refinement and finally some composition changes. The cold welding effect as seen in Fig. 2 for 10:1 and above can be explained on the basis that at high BPR, the collision energy is high and therefore complete reduction to the pure metal is possible [11]. The micro-strain obtained from the samples in Fig. 2 shows the increasing trend with the increase of BPR, this shows that if we increase the total weight of the ball, it would induce more strain to the particles.

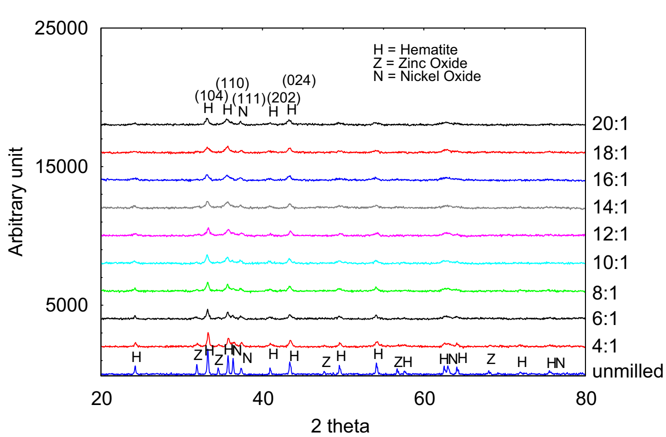

Fig. 1. XRD pattern of Ni–Zn ferrites, 4 h milling as a function of BPR.

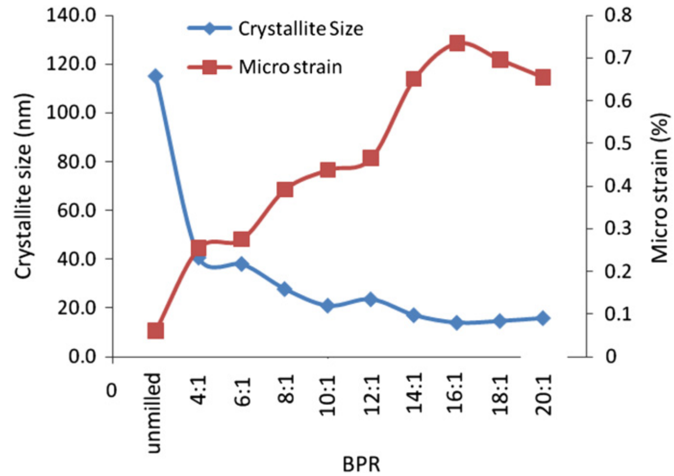

Fig. 2. Crystallite size and micro-strain as a function of BPR variation.

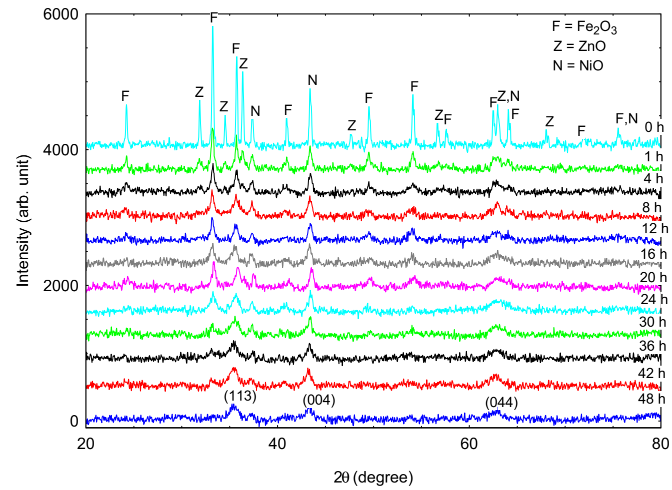

Fig. 3. XRD spectra of Ni–Zn-ferrites milled for various milling time.

Fig. 3 shows the XRD powder patterns recorded from unmilled and mechanically alloyed (BPR=10:1) homogeneous powder mixtures. From the spectra shown above, the unmilled powder mixture shows only individual reflections of NiO, ZnO and Fe_2O_3 phases. It is evident from the spectra that in the course of milling of these three individual powders, the Ni–Zn-ferrite phase was formed and its amount increased gradually with the increasing of milling time. A significant change after 1 h of milling is the reduction of the ZnO phase to a large extent compared to the NiO and Fe_2O_3 phases, which vanished after 4 h of milling.