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The Influence of the Preparation Conditions on Initial Permeability of $Cu_{0.5}Zn_{0.5}Fe_{2-x}R_xO_4$

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Samples with the chemical formula $Cu_{0.5}Zn_{0.5}Fe_{2-x}R_xO_4$ (x = 0.0 and 0.02; R = Sm and Gd) were prepared using a conventional ceramic method. x-ray analysis revealed the formation of a single spinel phase. The porosity decreases with an increase in the pressing pressure and the pre-sintering time during the preparation. The initial permeability, Curie temperature, and the homogeneity of the samples are increased with the pressing pressure and the pre-sintering time. This improvement of initial permeability is a novel result for different applications at a normal frequency range. The electrical resistivity is increased by a decrease of the pressing pressure. This is another novel result for the technology especially at a high-frequency range. The results are explained according to the increase of the stoichiometry of the samples.

Keywords Curie temperature, ferrite, inorganic compounds, magnetic properties, porosity, pressing pressure

1. Introduction

Ferrites have different important applications depending on their properties. The use of ferrites for certain applications depends on their magnetic and electrical properties. These properties are sensitive to the type and the amount of substitution as well as the preparation condition. Magnetic permeability and resistivity are mainly dependent on the pores in the ferrites (Ref 1, 2). Sattar et al. (Ref 3, 4) studied the electrical and the magnetic properties of $Cu_{0.5}Zn_{0.5}Fe_{2-x}R_xO_4$ ferrite; x = 0.0 and 0.1, R = La, Nd, Sm, Gd ions. They found that Sm sample has the highest initial permeability. These studies were followed by other articles to investigate Cu-Zn substituted with Sm of different concentrations and with different rare earth ions (Ref 5, 6). These reported studies showed that the sample with Sm = 0.02 has the highest initial permeability and Curie temperature. The initial permeability is improved by about 80% for Sm and Gd samples relative to the pure composition (Ref 6). Moreover, the electrical resistivity was increased with the increasing of Sm and Nd content (Ref 5–7). These results are very important for different applications. Therefore, this study is aimed to improve the initial permeability and the electrical resistivity by changing the preparation condition of Cu-Zn ferrite. In this study, the pressing pressure and the time of pre-sintering temperature are changed for $Cu_{0.5}Zn_{0.5}Fe_{2-x}R_xO_4$ (x = 0.0 and 0.02; R = Sm and Gd). The results are promising for technological applications at a low- and a high-frequency ranges.

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2. Experimental Details

Cu-Zn ferrite samples are prepared according to the following chemical formula $Cu_{0.5}Zn_{0.5}Fe_{2-x}R_xO_4$ (x = 0.0 and 0.02; R = Sm and Gd) using the standard ceramic method. Oxides Fe_2O_3, CuO, ZnO, and R_2O_3 with purity 99.99% are weighed in stoichiometric proportions. All oxides for each sample are mixed together and ground to a very fine powder. The mixture of the sample with x = 0.0 is pre-sintered at 900 °C for 30 h then cooled slowly in the furnace. The mixture is ground again and pressed into disks and toroid forms at pressures of 2.98, 3.98, and 4.98 (10^8 Pa). But, part of the mixture for the samples with x = 0.0 and 0.02 was pre-sintered at 900 °C for 12 h and the other part of the mixture with x = 0.02 was pre-sintered for 30 h then cooled slowly to the room temperature. The mixture of each sample is ground again and pressed into disks and toroid forms at a pressure of 2.98 (10^8 Pa). Then all prepared samples of the above chemical formula are sintered at 1000 °C for 6 h and then cooled slowly to the room temperature. For all samples, using a diffractometer type (X'Pert Graphics), x-ray diffraction patterns were performed and the data revealed no impurity phases.

The porosity percentage P (%) was calculated using the relation: $P = (1 - d/d_x)$ 100%, where d_x (g/cm^3) is theoretical x-ray density. It was calculated using the following formula: $d_x = 8\ M/Na^3$, where M is the molecular weight of each sample, N is Avogadro's number, and a is the average lattice parameter. The parameter d (g/cm^3) is the apparent density which was calculated using Archimedes' principle. The toroidal samples were used as transformer cores for measuring the initial permeability, μ_i. This measurement is based upon Faraday's law of induction. The initial permeability was measured as a function of temperature at constant frequency f = 10 kHz and low magnetizing current (I_p = 4 mA). The value of μ_i was calculated using Poltinnikov's formula (Ref 8) which is given by $V_s = K\mu_i$, where V_s is the induced voltage in the secondary coil and K is a constant and equal to: $K = (\mu_o n_p n_s I_p A\omega)/L$. This value depends on the magnetizing current, frequency, number of turns of the primary and secondary coils, and the dimensions of the sample. L is the

average path of the magnetic flux, $L = 2\pi r_m$, where r_m is the mean radius of the toroidal sample, $r_m = (r_o + r_i)/2$. The outer and inner radii of the toroid are about 0.8 and 0.4 cm, respectively, and the thickness of the sample is 0.3 cm. The Curie temperature T_c of each sample was determined by the intersection of the initial permeability curve with the temperature axis, x-axis. The temperature range is from room temperature to 560 K. The rate of heating was controlled to be very slow to get a homogeneous and stable temperature. The dc electrical resistivity was measured at room temperature using the two probe method. A uniform layer of In-Hg was formed on the two surfaces of the disk sample as a contact material and then inserted between two platinum electrodes. The resistance is obtained from the measurement of the potential difference across the sample and the current passing through it. The current did not exceed 10 mA to avoid the Joule heating effect. The dc resistivity, ρ is calculated from the relation $\rho = RA/t$, where t and A are the thickness and the cross section area of the sample, respectively.

3. Results and Discussion

3.1 The Effect of the Pressing Pressure for Cu-Zn Ferrite

Figure 1 shows the dependence of initial permeability, μ_i on the pressing pressure, P (Pa) at the room temperature for Cu-Zn ferrite. It is clear that the sample pressed at 4.98 (10^8 Pa) has the highest initial permeability value. It is known that, μ_i for ferrites is due to the domain wall displacement and the rotation of spins inside the domains (Ref 9, 10). The movement of domain walls and rotation of spins are affected mainly by inter- and intra-pores. Figure 2 indicates the dependence of porosity, P (%) on the pressing pressure for Cu-Zn ferrite. One notices the decrease of porosity with the increasing of the pressing pressure. The decrease of porosity affects μ_i and leads its value to increase. Accordingly, this result accounts for the highest value of μ_i for Cu-Zn sample pressed at 4.98 (10^8 Pa), μ_i is nearly improved 340% over that pressed at 2.98 (10^8 Pa). This improvement is very important for different applications in the normal operating frequency range. Furthermore, it is known that Fe^{2+} ions are formed during the preparation of ferrites, at the final sintering temperature of the samples (Ref 11). These ions increase the anisotropy field of the sample. This causes the initial permeability to decrease according to the reported relation $\mu_i \propto \sqrt{(M_s^2\, D/K)}$ (Ref 10). This relation indicated that μ_i depends on the saturation magnetization, M_s; grain size, D; and anisotropy field, K. Accordingly, the highest value of μ_i for the sample pressed at 4.98 (10^8 Pa) also can be attributed to the increase of its stoichiometry. This leads to the decrease of the formation of Fe^{2+} at final temperature and accordingly the decrease of the anisotropy field. Finally, the highest value of μ_i for the samples pressed at 4.98 (10^8 Pa) is due to its smallest value of porosity and the decrease of Fe^{2+} ions formation. This means that the initial permeability depends mainly on the preparation condition. From another point of view, this result makes the sample easier to magnetize.

The values of Curie temperature, T_c as a function of pressing pressure is shown in Fig. 3. These values are determined from the linear part of the experimental initial permeability against

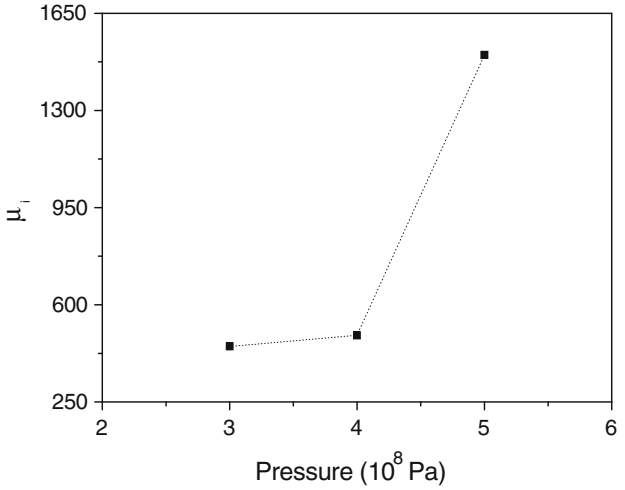

Fig. 1 The dependence of initial permeability, μ_i on pressing pressure for Cu-Zn ferrite

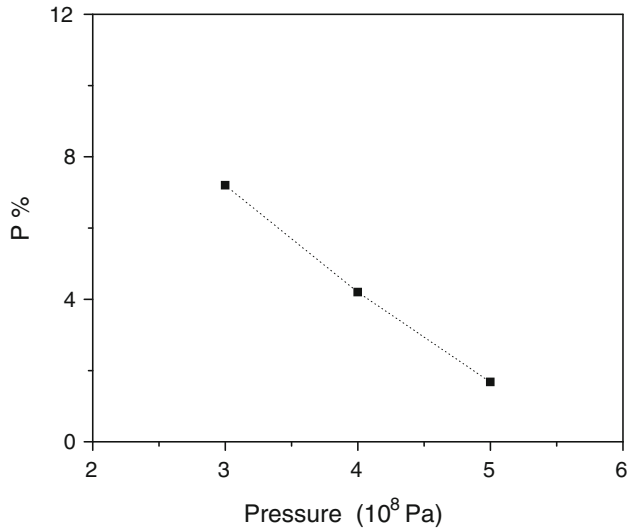

Fig. 2 The variation of porosity, P (%) with pressing pressure for Cu-Zn ferrite

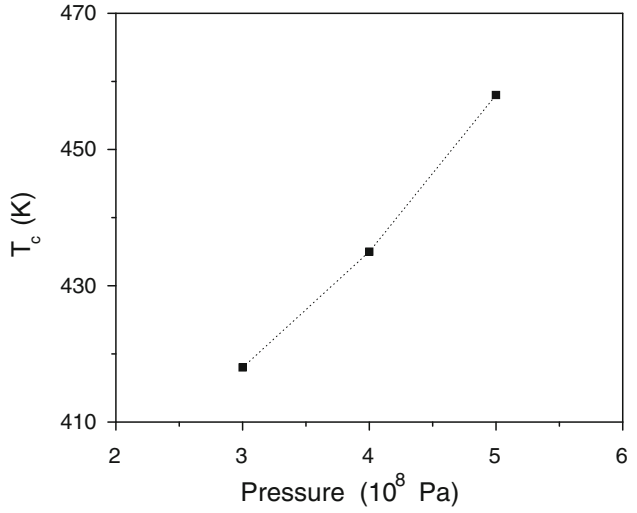

Fig. 3 The variation of Curie temperature, T_c with pressing pressure for Cu-Zn ferrite

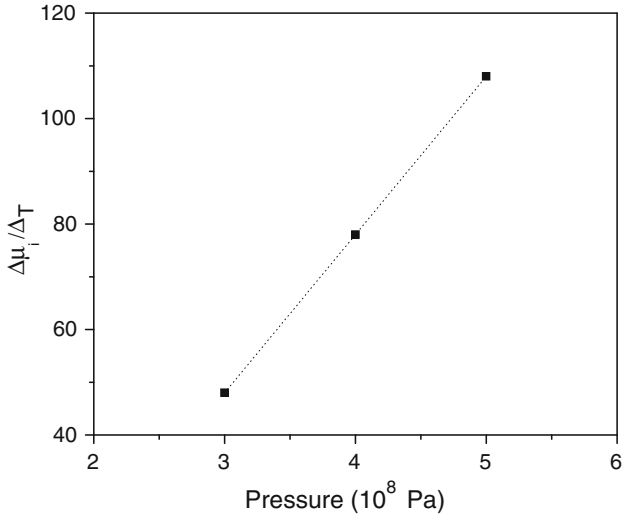

Fig. 4 The dependence of homogeneity on pressing pressure for Cu-Zn ferrite

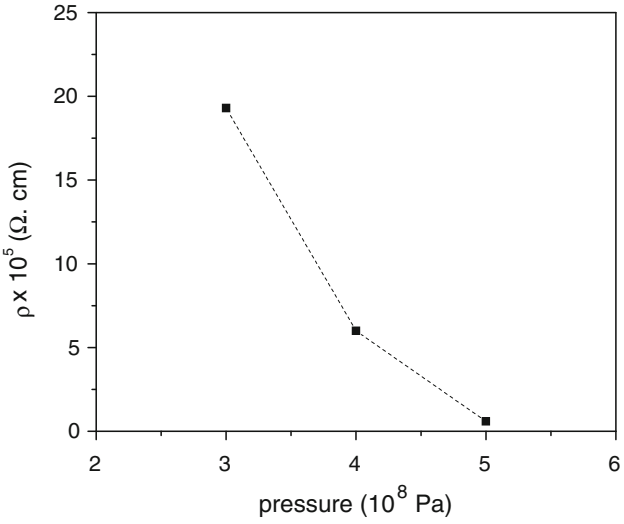

Fig. 5 The dependence of electrical resistivity on pressing pressure for Cu-Zn ferrite

the temperatures. There is an increase in the T_c with increasing of the pressing pressure which indicates the increase of super-exchange interaction between the cations distribution in A-sites and B-sites. As discussed above the formation of Fe^{2+} ions at final sintering temperature decrease with the increasing of the pressing pressure. These ions preferred the B-sites, the decrease of Fe^{2+} ions leads the net magnetic moment between B and A sites to increase, the magnetic moments of Fe^{2+} and Fe^{3+} ions are 4 μ_B and 5 μ_B, respectively. Accordingly, the magnetization will increase with the increasing of the pressing pressure and then the Curie temperature.

Figure 4 shows the dependence of homogeneity, $\Delta\mu_i/\Delta T$ on pressing pressure for Cu-Zn ferrite. It is reported that the value of $\Delta\mu_i/\Delta T$ gives a good indication of the homogeneity of the ferrite sample (Ref 12). The higher value of the slope, $\Delta\mu_i/\Delta T$, of the linear part at a sudden decrease of μ_i curve with increasing temperature corresponds to a more homogeneous sample. It is clear that the sample pressed at 4.98 (10^8 Pa) has the highest homogeneity value. This can attributed to the increase of the stoichiometry of the sample. The variation of the homogeneity has an inverse behavior to the porosity, Fig. 2.

Figure 5 illustrates the dependence of the electrical resistivity, ρ, on pressing pressure for Cu-Zn ferrite at room temperature. One notices that there is a decrease in the electrical resistivity with an increase of the pressing pressure. In other words, the electrical resistivity increases with a decrease in the pressing pressure. This can attributed to the increase of the porosity of the samples. This is a novel result for the technology especially at high-frequency range.

3.2 Effect of Varying the Pre-Sintering Temperature for $Cu_{0.5}Zn_{0.5}R_xFe_{2-x}O_4$ (x = 0.0 and 0.02; R = Sm and Gd), at Constant Pressure

Figure 6 shows the dependence of the initial permeability μ_i at constant temperature on the ionic radius of the rare earth ions for $Cu_{0.5}Zn_{0.5}R_xFe_{2-x}O_4$ (x = 0.0 and 0.02; R = Sm and Gd) samples. These samples are prepared at two different pre-sintering times 12 and 30 h. It is noticed that μ_i increases with the increasing of the pre-sintering time for each sample. Furthermore, it increases with the increasing of the ionic radius

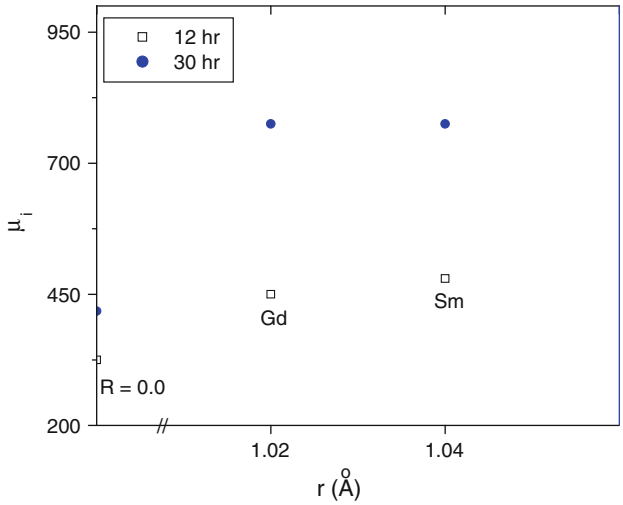

Fig. 6 The dependence of initial permeability, μ_i on of rare earth ions, at ionic radius two different pre-sintering times

of the rare earth ions relative to the pure Cu-Zn ferrite, at constant pre-sintering time. The increasing of pre-sintering time allows the sample to be more homogeneous, Fig. 7. Also, this condition leads to the slow growth of grains and a decrease in the total porosity of the sample. It is known that the total porosity, P (%) is the sum of inter-granular pores, P_{inter}, and intra-granular pores, P_{intra} (Ref 13). So, the decrease of the porosity may be due to the decrease in P_{intra}. This means that the porosity inside the grains decreases, Fig. 8. This facilitates the domain walls motion and causes the increase of the initial permeability.

Also, the increase in pre-sintering time allows the sample to be more stoichiometric. As discussed above, this leads to the decrease of the formation of Fe^{2+} ions and the anisotropy field. Accordingly, this causes μ_i to increase with pre-sintering time. The effect of the rare earth substitution causes Fe^{2+} ion formation to decrease and the increase of μ_i values. Although the percentage of Sm and Gd ions is the same, the Sm sample has higher values of μ_i than the Gd sample. This can be

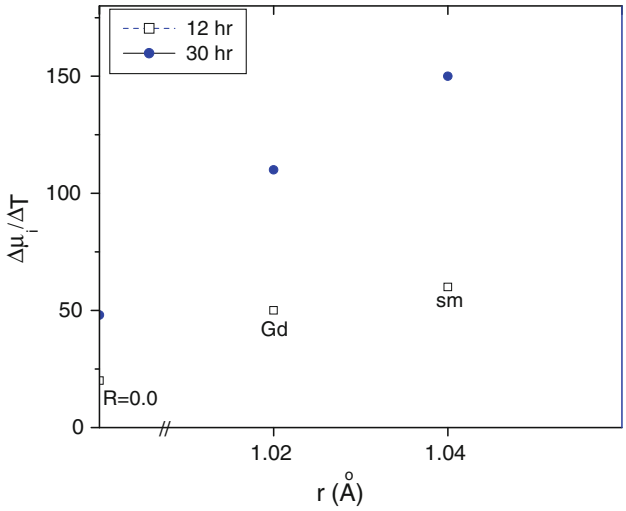

Fig. 7 The dependence of homogeneity on ionic radius of rare earth ions, at two different pre-sintering times

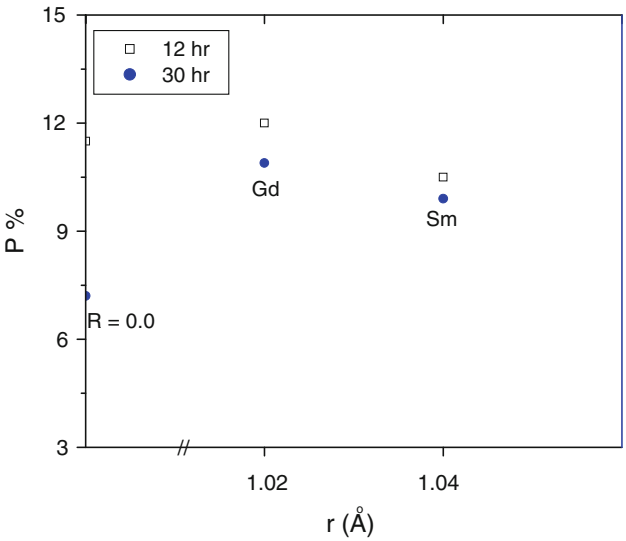

Fig. 8 The dependence of porosity on ionic radius of rare earth ions, at two different pre-sintering times

attributed to the decrease of porosity, P (%), for Sm sample relative to Gd sample.

Figure 9 shows the dependence of Curie temperature, T_c on the ionic radius of the rare earth ions, at two different pre-sintering times. The Curie temperature increases with the increasing of the pre-sintering time. This is attributed to a decrease in the formation of Fe^{2+} ions. Also, the decrease of Fe^{2+} ions due to the rare earth substitution, for Gd and Sm samples at certain pre-sintering time explains the increasing of T_c.

4. Conclusions

The highest value of μ_i in the Cu-Zn sample pressed at 4.98 (10^8 Pa) pressure is due to its smallest value of porosity and anisotropy field. The homogeneity increases with the increasing of the pressing pressure for Cu-Zn ferrite. The increase of pre-sintering time leads to increase in the initial permeability and

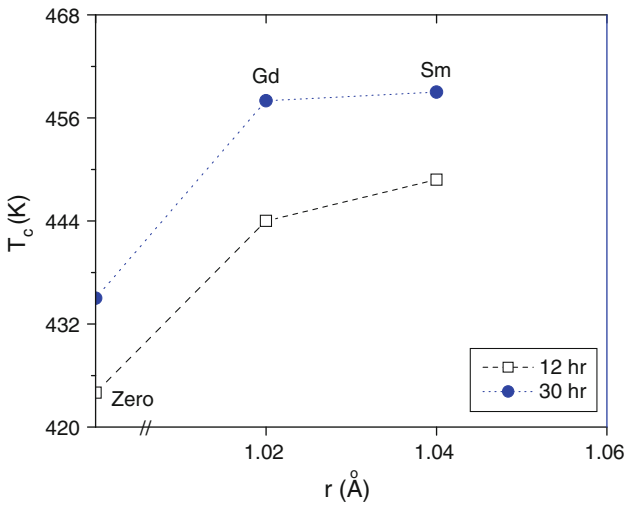

Fig. 9 The dependence of Curie temperature, T_c on ionic radius of rare earth ions, at two different pre-sintering times

homogeneity for each sample of $Cu_{0.5}Zn_{0.5}R_xFe_{2-x}O_4$ (x = 0.0 and 0.02; R = Sm and Gd), at constant pressure. However, the porosity decreases with the increasing of the pre-sintering time. A decrease in the pressing pressure leads to an increase in the electrical resistivity. The novel results, increasing the initial permeability and the electrical resistivity with variation in the preparation conditions is promising for technological applications in the low- and the high-frequency ranges, respectively.

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