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Effect of Sm substitution on the magnetic and electrical properties of Cu-Zn ferrite

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Samples of the chemical formula $Cu_{0.5}Zn_{0.5}Fe_{2-x}\ Sm_xO_4$ where $x = 0.0$, 0.02, 0.04, 0.06, 0.08 and 0.1 are studied. X-rays analysis indicated that all investigated samples are formed in a single cubic phase. The lattice parameter is found to increase relative to $x = 0.0$ except $x = 0.02$. The grain size is decreased for all samples relative to unsubstituted one except that with $x = 0.02$. It is noticed that the sample with $x = 0.02$ has the highest values of initial permeability, magnetization and Curie temperature, T_c. The homogeneity of the samples, the coercive field as well as T_c are found to decrease while the electrical resistivity increases with increasing Sm content. © *2002 Kluwer Academic Publishers*

1. Introduction

Ferrites are very important materials as they are successfully used in high frequency devices. Chemical composition and preparation condition influence the ferrite properties (porosity, resistivity, permeability and saturation magnetization) [1]. The effect of substitution with trivalent ions for iron has been investigated [2, 3]. The presence of multi-phase during substitution affects the actual internal physical properties. Later, Sattar *et al.* studied the electrical and magnetic properties of a single phase of Cu-Zn ferrites doped with different rare earth ions [4, 5]. Among the different substituted rare earth ions, Sm showed the highest initial permeability value. As this parameter plays an important role in technology, therefore we aimed to study how the initial permeability and related parameters as well as the electrical resistivity are affected by taking different Sm content. Both the magnetic and electrical measurements are reported.

2. Experimental techniques

Polycrystalline samples with the chemical formula $Cu_{0.5}Zn_{0.5}Fe_{2-x}\ Sm_xO_4$ where $x = 0.0$, 0.02, 0.04, 0.06, 0.08 and 0.1 were prepared by the usual ceramic technique. High purity oxides of CuO, ZnO, Fe_2O_3 and Sm_2O_3 were mixed together according to their molecular weights. The powders of each sample were ground to a fine powder and then presintered at 900°C for 12 h. The presintered powders were ground again until a particle size of $\approx 36\ \mu$m is obtained and then pressed at a pressure of 3 ton /cm^2 into two forms, discs and toroids. They were finally sintered at 1000°C for 6 h in two cycles and then slowly cooled to room temperature. X-rays diffraction patterns were performed using Philips diffractometer type Pw1373 with Cu K_α radiation. The porosity percentage P was calculated according to the relation, $P = 100\ (1 - d/d_x)$ where d_x is the theoretical x-ray density and d is the density of each sample which was measured in toluene using Archimed's principle. The scanning electron microscope (SEM), type JEOL JEM-1200 EXII was used to observe the microstructure of some polished and etched samples ($x = 0.0$, 0.02, 0.04 and 0.1). For magnetization measurements M(A/m), a toroidal sample was used as a transformer core. The initial permeability μ_i was measured as a function of temperature, at a frequency $f = 10$ KHz, to determine the Curie temperature. The value of μ_i was calculated using Poltinnikov's formula [6] 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 given by $K = \mu_o n_s n_p i_p A\omega/L$, where n_s and n_p are the number of turns of the secondary and primary coils respectively, i_p is the magnetizing current in the primary coil, A is the cross sectional area of the toroid and L is the average path of the magnetic flux $= 2\pi r_m$ where r_m is the mean radius of the toroid. Moreover, the coercive field was determined from the hysteresis loop. Disc samples were used for the electrical resistivity measurements. The sample is inserted between two platinum electrodes where In-Hg was used as a contact material. The current passing through the sample does not exceed 10 mA to avoid the joule heating effect. The temperature was measured using K-type thermocouple.

3. Results and discussion

X-ray diffraction patterns indicated that all investigated samples are formed in a single cubic phase. The average lattice parameters a (Å) for all samples are calculated and are given in Table I. It is clear that, there is an increase in the lattice parameter for all samples relative to $x = 0.0$ except $x = 0.02$. The porosity of the samples is given also in Table I. One notice that the porosity increases with increasing Sm content. It was reported that the porosity results from the formation of SmO_2 which favors the growth of grains with inner pores [7]. It is expected that SmO_2 concentration increase with increasing Sm content, which explains the increase of the porosity.

Fig. 1 shows the scanning electron micrographs for samples with $x = 0.0$, 0.02, 0.04 and 0.1. It is clear

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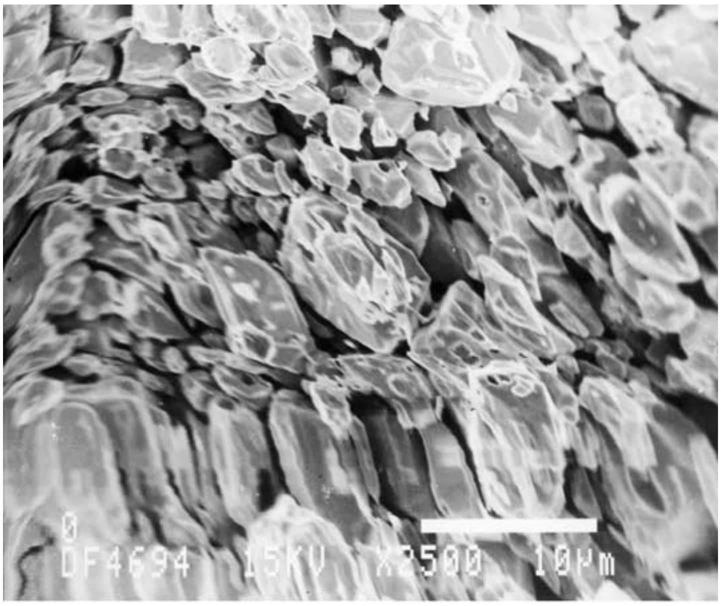
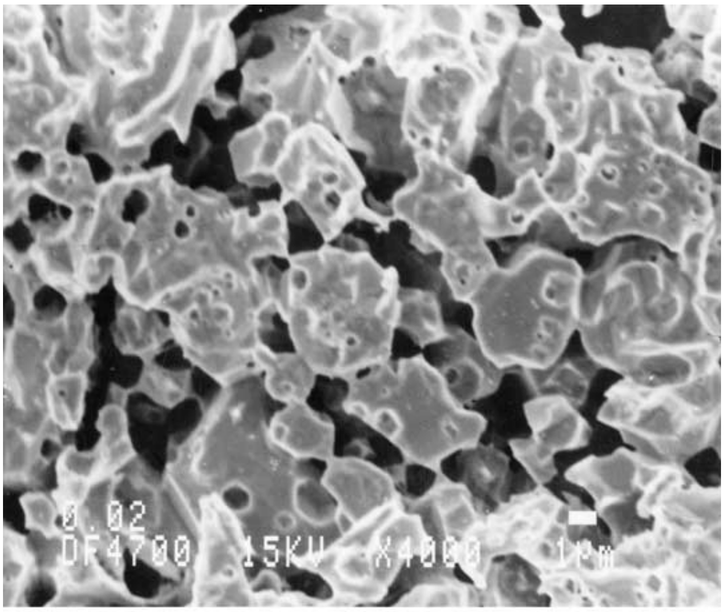
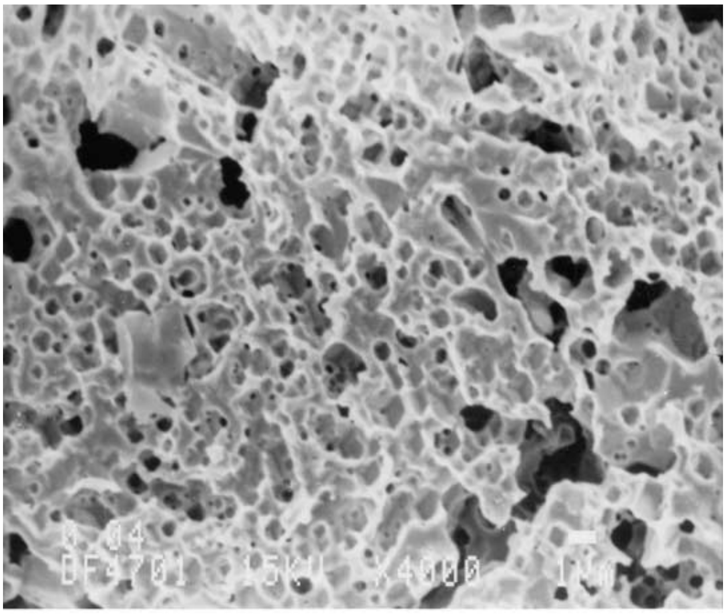


a) x= 0.0

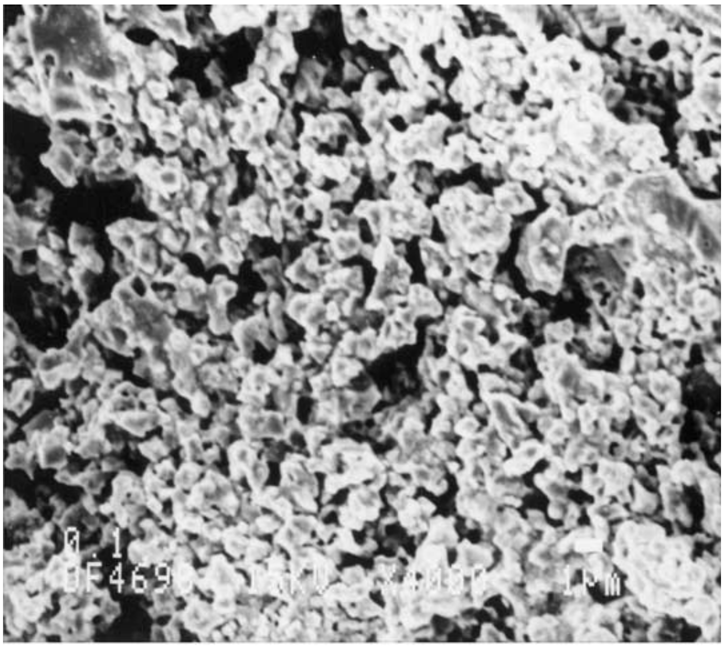


b) x=0.02



c) x=0.04

Figure 1 The scanning electron micrographs for (a) $x = 0.0$, (b) $x = 0.02$, (c) $x = 0.04$ and (d) $x = 0.1$. (*Continued.*)



d) x= 0.1

Figure 1 (*Continued*).

TABLE I Values of the average lattice parameter, porosity and Curie temperature for all samples

Sm content	a (Å)	P(%)	T_c (K)
0.00	8.407	07.2	452
0.02	8.346	10.15	456
0.04	8.428	12.60	445
0.06	8.432	12.75	435
0.08	8.468	13.50	430
0.10	8.412	13.75	423

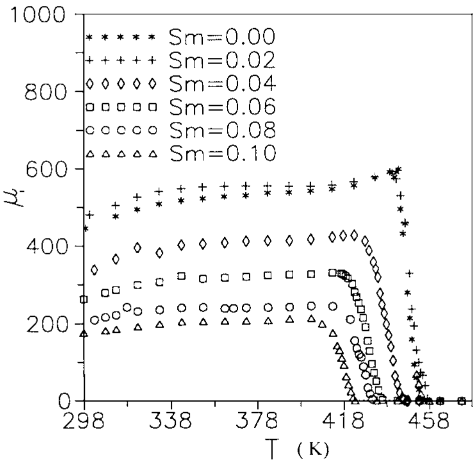


Figure 2 The variation of the initial permeability μ_i with temperature T (K).

that the sample with $x = 0.02$ has the largest grain size relative to other samples.

Fig. 2 shows the variation of the initial permeability μ_i with temperature. The intersection of the linear part of μ_i with the temperature axis determines the Curie temperature T_c. The variation of T_c with Sm content is shown in Table I. It is clear that T_c decreases with increasing Sm content except for $x = 0.02$. Also, it is clear that the sample with $x = 0.02$ has the highest value of μ_i relative to unsubstituted one. This means that there is a direct relation between the average grain size and μ_i which is the expected behavior and is in agreement with our previous work [8].

The decrease of T_c with increasing Sm content, except $x = 0.02$, could be explained as follows: On substitution of Sm in Cu-Zn ferrite, then Sm-Fe and Sm-Sm interactions will be weak than that of Fe-Fe. Moreover, the lattice parameters for samples with $x > 0.02$ are found to be larger than that for $x = 0.0$. Thus one expect T_c to decrease for $x > 0.02$. For $\text{Sm} = 0.02$, one note that the lattice parameter has the smallest value which leads the magnetic interaction to increase.

The curves of Fig. 2 show sharp maxima just before T_c for $x = 0.0$ and $x = 0.02$ only. It is known that the magnetization and the anisotropy field decrease with increasing temperature. However, the decrease of the anisotropy field with temperature is faster than that of the magnetization. This leads to the appearance of maximum in μ_i. One notice also that the drop of μ_i at T_c is sharp for $x = 0.0$ and such sharpness decreases with increasing Sm concentration. The sharpness, which can be expressed as $\Delta\mu_i/\Delta T$, at $T = T_c$ illustrates the homogeneity of the sample [9]. Fig. 3 shows that the homogeneity of the investigated samples decreases

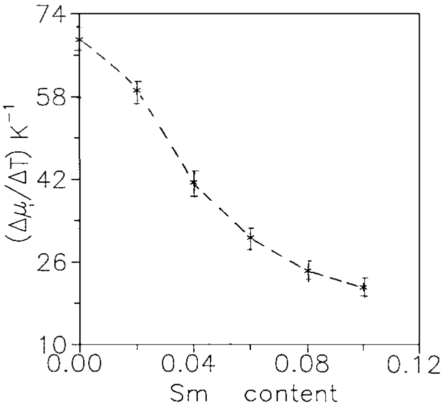


Figure 3 The homogeneity of the samples $(\Delta\mu_i/\Delta T)_{T_c}$ with Sm content.

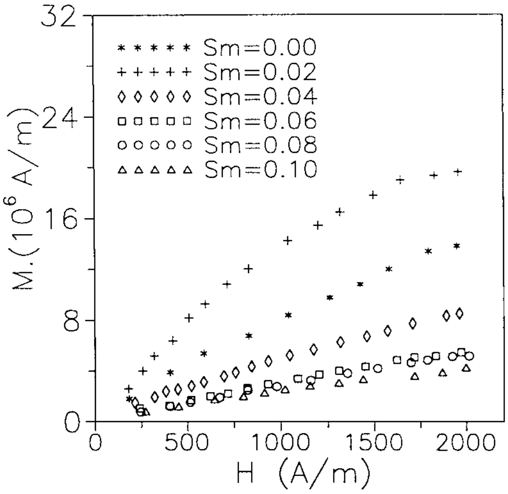


Figure 4 The variation of magnetization M (A/m) with the applied magnetic field H (A/m).

with increasing Sm content. This is in agreement with our assumption that SmO_2 concentration increases with increasing Sm content.

Fig. 4 shows the changes of magnetization, M (A/m), with the magnetizing field, H (A/m), for all samples. One note that the magnetization increases with the applied magnetic field. However, the saturation magnetization can not be reached for any sample as the applied field is small. It is clear that, relative to the sample with $x = 0.0$, the sample with $x = 0.02$ has the highest magnetization value while those with $x > 0.02$ have lower values. It is known that the value of the magnetization depends on the magnitude of the magnetic moment as well as the interaction between these moments. Such interactions are affected greatly by the separation between moments and also by the porosity. For $x = 0.02$, both the lattice parameter and the porosity are small such that the effect of the porosity is masked by the strong magnetic interaction. This explains the highest magnetization value for $x = 0.02$. For $x > 0.02$, both the lattice parameters and the porosity increase with x leading the magnetic interactions to decrease.

Fig. 5 shows the changes of the coercive field H_c (A/m) as a function of Sm content. It is clear that H_c has the highest value for $x = 0.0$ and 0.02. Further increase of Sm concentration leads H_c to decrease. Taking H_c as a function of the anisotropy field [10], then the anisotropy field decreases with increasing Sm content.

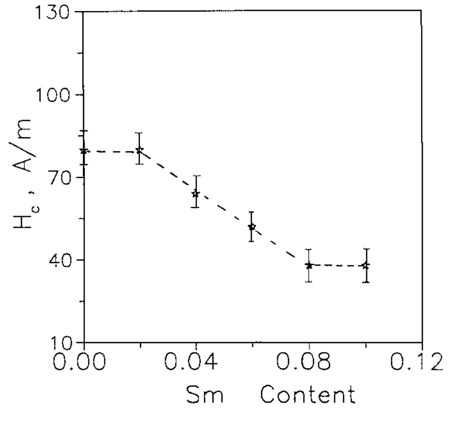


Figure 5 The variation of the coercive field H_c (A/m) with Sm content.

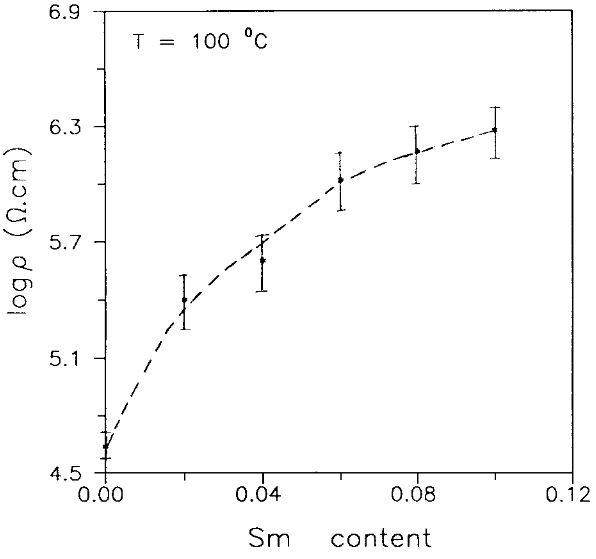


Figure 6 The dependence of the resistivity, log ρ, on Sm content.

This behavior could be explained as follows: With increasing Sm content, the probability of Fe^{2+} formation decreases. As the Fe^{2+} is responsible for the anisotropy in ferrites [11], hence H_c decreases with increasing Sm concentration.

Fig. 6 shows the dependence of the electrical resistivity, log ρ, on Sm concentration at 100°C. It is clear that with increasing Sm content, the electrical resistivity increases. As Sm content increases the probability of SmO_2 formation increases which hinders the motion of the charge carriers. This means that the mobility will be decreased. This accounts on the increase of the electrical resistivity with increasing Sm content.

4. Conclusion

In our samples, it is found that the lattice parameter increases relative to $x = 0.0$, except for $x = 0.02$. Meanwhile, the substitution with different concentration of Sm affects the grain size, lattice parameter, magnetization, initial permeability, porosity, Curie temperature T_c, the homogeneity of the samples and the electrical resistivity.

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