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To cite this version:
T. Murase, K. Igarashi, J. Sawai, T. Nomura. Relaxation Phenomena of MgZn Ferrites. Journal de Physique IV Colloque, 1997, 07 (C1), pp.C1-99-C1-100. <10.1051/jp4:1997128>. <jpa-00254992>

HAL Id: jpa-00254992
https://hal.archives-ouvertes.fr/jpa-00254992
Submitted on 1 Jan 1997

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Relaxation Phenomena of MgZn Ferrites

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Abstract. The decrease of initial permeability ($\mu_i$) with increasing frequency is enhanced by CuO addition to MgZn ferrite. The gradual deterioration of $\mu_i$ with frequency is due to a relaxation phenomenon. By measuring the temperature dependence of $\tan \delta$, the activation energy of the relaxation was calculated as approximately 0.43eV. From the results of time disaccommodation, the change in permeability for MgCuZn ferrite was found to be larger than that of MgZn ferrite. Furthermore, the deterioration of $\mu_i$ with time for both ferrites continued for more than 500 hours.

1. INTRODUCTION

Soft ferrites are widely used in electronic equipments, and recently, the demand for highly inductive materials has increased so as to lower prices. MgZn ferrite is used in large quantities because it is highly cost effective. In general, copper and manganese oxides are added to MgZn ferrites in order to achieve high quality. CuO addition is especially effective in lowering the sintering temperature and core loss at high frequency. It has, however, a deleterious effect on the stability of initial permeability ($\mu_i$). CuO addition also causes the deterioration of $\mu_i$ with increasing frequency. This gradual deterioration of $\mu_i$ with frequency is a relaxation phenomenon. The relaxation is presumed to be the delay of the domain wall motion with respect to change in the external magnetic field. It is inferred that the delay of the domain wall motion may be caused by electron migration, cation migration, impurities or by defects. The purpose of this paper is to focus on the relaxation phenomena in MgZn and MgCuZn ferrites, with special reference to the chemical composition and microstructure.

2. EXPERIMENTAL

Ferrite powders in the MgO-CuO-ZnO-Fe$_2$O$_3$ system were prepared by conventional powder processes. The starting raw materials are commercial grade powders, Mg(OH)$_2$, CuO, ZnO, Fe$_2$O$_3$. The above raw materials were weighed and then mixed in a ball-mill for 16 hours. Afterwards, the mixed powders were dried, calcined at 900°C for 3 hours and re-milled using a wet-method for 16 hours. The average particle size of the ferrite powders was about 1.5 μm. Then, the powders were granulated using 0.6wt% of PVA and pressed into T-10 toroids and disks (d =12mm, d=1.8mm). The green bodies were fired between 1100 and 1400°C in air, with heating and cooling rate of 300°C per hour in air. Afterwards, the toroids were wound with 20 turns of Cu wire and the disks were electroded using In-Ga or Pt on both faces. The electromagnetic properties were measured using LCR meters (HP 4192A, HP E2377A, YHP 4194A and YHP 4329A). Microstructures were observed using optical microscopy (OM) and transmission electron microscopy (TEM).

3. RESULTS AND DISCUSSION

The effect of CuO addition on the frequency dependence of $\mu_i$ is shown in Fig. 1.
The decrease of $\mu$ with increasing frequency is enhanced with CuO addition. These phenomena are similar to those seen in the case of MnO addition.[1, 2] The temperature dependence of $\tan \delta$ for MgZn and MgCuZn ferrites are shown in Fig. 2 and 3. Peaks for the MgZn ferrite are not observed in the temperature range measured but may exist at lower temperatures than -70°C. On the other hand, peaks for the MgCuZn ferrite appear within the tested temperature range. The peaks shift to a higher temperature region and the values decrease with increasing frequency. In general, $\tan \delta$ is expressed by the Curie-Weiss law in equation (1).

$$\tan \delta = \chi, \omega \tau [(1 + \chi) + \omega^2 \tau^2] \quad (\chi : \text{relative magnetic susceptibility}) \cdots (1)$$

$$\omega = 1 \cdots (2)$$

Consequently, the activation energy for the relaxation of the MgCuZn ferrite is calculated as about 0.43eV. This value is similar to that found for MnO addition to MgZn ferrite. The activation energy for electron transfer in ferrites, i.e. for $Fe^{2+} \rightarrow Fe^{3+}$, is known to be about 0.16eV[3], while that for carbon migration in Aramco iron is about 1.0eV[4]. As the value measured here falls between the two, it is reasonable to infer that it is also due to cation migration. Usually, the diffusion in the crystal occurs via point defects. It is suggested that the relaxation in the MgCuZn ferrite may be caused by the delay of domain wall motion due to point defects.

The relaxation time $\tau$ at a given temperature is calculated from the measured $\tan \delta$ value by the equation

$$\tau = \tau_c \exp \left( \frac{Q}{kT} \right) \quad \text{(frequency factor, } k \text{ : Boltzmann constant}) \cdots (3)$$

The relaxation time for our specimens is longer than that for MgZn ferrite, as observed in Fig. 4. As the relaxation in the MgCuZn ferrite is larger than that in MgZn ferrite, it is thought that the difference is due to the concentration of defects.

In this study, the effect of addition to MgZn ferrite. By examining the relationship between the temperature and the relaxation time, the activation energy for MgCuZn ferrite was found to be about 0.43eV. Furthermore, the relaxation time on disaccommodation for both compositions was longer than 500 hours.

4. CONCLUSION

In this study, the effect of CuO addition on relaxation in MgZn ferrite was investigated. The changes of permeability with frequency and time are enhanced by CuO addition to MgZn ferrite. By examining the relationship between the temperature and the relaxation time, the activation energy for MgCuZn ferrite was found to be about 0.43eV. Furthermore, the relaxation time on disaccommodation for both compositions was longer than 500 hours.

Reference