



HAL
open science

Relaxation Phenomena of MgZn Ferrites

T. Murase, K. Igarashi, J. Sawai, T. Nomura

► **To cite this version:**

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

HAL Id: jpa-00254992

<https://hal.science/jpa-00254992>

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Relaxation Phenomena of MgZn Ferrites

T. Murase, K. Igarashi, J. Sawai and T. Nomura

570-2 Aza-Matsugashita, Minami-Hatori, narita-shi, Chiba-ken 286, Japan

Abstract. The decrease of initial permeability (μ_i) with increasing frequency is enhanced by CuO addition to MgZn ferrite. The gradual deterioration of μ_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 μ_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 (μ_i). CuO addition also causes the deterioration of μ_i with increasing frequency. This gradual deterioration of μ_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-Fe₂O₃ system were prepared by conventional powder processes. The starting raw materials are commercial grade powders, Mg(OH)₂, CuO, ZnO, Fe₂O₃. 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 (φ = 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 μ_i is shown in Fig. 1.

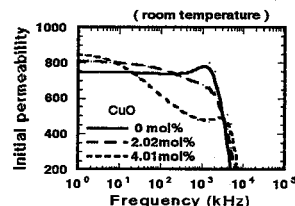


Fig. 1 Frequency dependence of initial permeability. (ZnO=21.2, Fe₂O₃=49.4mol%)

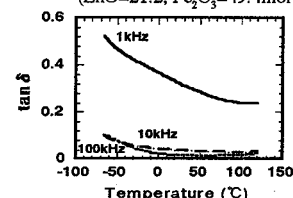


Fig. 2 Temperature dependence of $\tan \delta$ for MgZn ferrite. (MgO=29.2, ZnO=21.3, CuO=0, Fe₂O₃=49.5mol%)

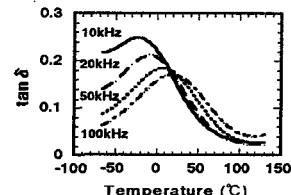


Fig. 3 Temperature dependence of $\tan \delta$ for MgCuZn ferrite. (MgO=25.2, ZnO=21.3, CuO=4.0, Fe₂O₃=49.5mol%)

The decrease of μ with increasing frequency is enhanced with CuO addition. These phenomena are similar to those seen in the case of Mn_2O_3 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). The frequency is expressed by equation (2) when $\tan \delta$ is maximum. On examining the relationship between the temperature at the $\tan \delta$ peak in Fig. 3 and the relaxation time (τ), good agreement was found with the Arrhenius equation (3).

$$\tan \delta = \chi_r \omega \tau / [(1 + \chi_r) + \omega^2 \tau^2] \quad (\chi_r : \text{relative magnetic susceptibility}) \dots (1)$$

$$\omega \tau = 1 \dots (2)$$

$$\tau = \tau_0 \exp (Q/kT) \quad (\tau_0 : \text{frequency factor, } k : \text{Boltzmann constant}) \dots (3)$$

Consequently, the activation energy of the relaxation for the MgCuZn ferrite is calculated as about 0.43eV. This value is similar to that found for Mn_2O_3 addition to MgZn ferrite. The activation energy for electron transfer in ferrites, i.e. for $Fe^{2+} \leftrightarrow Fe^{3+}$, is known to be about 0.1eV[3], while that for carbon migration in Armco 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 characterization technique for defects in semiconductors[5, 6], involving measurement of electric conductivity dependence on the oxygen pressure(P_{O_2}), was used in the present study on the MgZn and MgCuZn ferrites (Fig. 4). It was expected to recognize that the MgCuZn ferrite differed from MgZn ferrite in the type, concentration and arrangement of defects. In our experiment the specimens were kept at a given P_{O_2} at 700°C and were measured after 2 hours. Each of them was measured from high- P_{O_2} to low- P_{O_2} (the solid lines), and then from low- P_{O_2} to high- P_{O_2} (the dotted lines) using the same specimen in each case. In both MgZn and MgCuZn ferrites the values from the first measurement are not in agreement with those from the second measurement. It is supposed that the specimens formed different phases in the strongly reducing atmosphere and did not equilibrate sufficiently in 2 hours from low- P_{O_2} to high- P_{O_2} . At present, variations of time and temperature are being investigated to solve this problem.

In order to study the effect of microstructures on relaxation phenomena, OM and TEM were performed. Photo. 1 shows the microstructures of polished and hydrofluoric acid-etched surfaces of MgZn and MgCuZn ferrites sintered at 1250°C . It seems that CuO addition accelerates grain growth. The large grains of MgCuZn ferrite contain pores and etch pits. It is supposed that grain growth and pores are related to the decrease of μ_1 with frequency. Furthermore, it is believed that etch pits are initiated by defects. In order to find lattice defects, the nanostructure of these samples was observed using TEM. It was expected that the lattice fringes would be distorted by defects. However, no distortion was witnessed (Photo. 2).

The electric conductivity investigation and the TEM observation did not adequately characterize the defects for MgZn and MgCuZn ferrites. However, results of time disaccommodation of magnetic permeability for these ferrites confirm differences in the defect structures. The time dependence of the change of permeability is shown in Fig. 5. The change in permeability for MgCuZn ferrite is larger than that in MgZn ferrite. It is thought that the difference is due to the concentration of defects. Furthermore, from this figure it can be seen that the deterioration of μ for both ferrites continues for 500 hours or more. It is expected that the relaxation time for our specimens is longer than that for MgZn ferrite, as observed by Ohta[7].

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

[1] K. Igarashi, T. Murase and T. Nomura, Powder and Powder Metallurgy, **42** (1995) 22.
 [2] K. Igarashi, T. Murase and T. Nomura, Materials Engineering for Resources of Japan, **8** (1995) 61.
 [3] Y. Tawara, J. Phys. Soc. Japan, **17** (1962) 1711.
 [4] Y. Tomono, J. Phys. Soc. Japan, **7** (1952) 174.
 [5] F. A. Kroger and H. J. Vink, "Solid State Physics", **3**, F. Seitz, D. Turnbull(eds.), (1956) 313.
 [6] N. G. Eror and U. Balachandran, Journal of Solid State Chemistry, **42** (1982) 227-241.
 [7] K. Ohta, J. Phys. Soc. Japan, **16** (1961) 250.

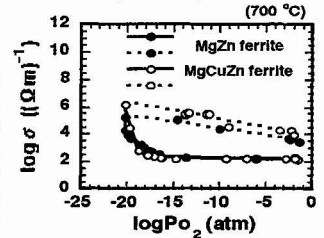


Fig. 4 Oxygen pressure dependence of conductivity.

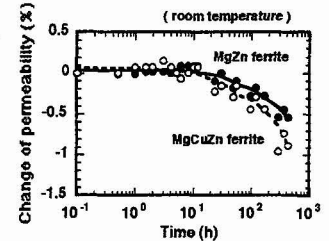


Fig. 5 The disaccommodation of the MgZn ferrite.

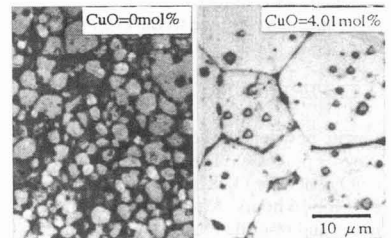


Photo. 1 Microstructures of the MgZn ferrites.

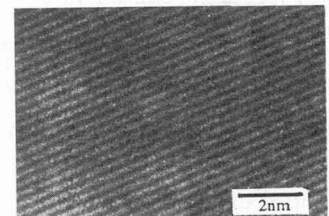


Photo. 2 TEM image of MgCuZn ferrite.