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RAPID PUBLICATION

Soft Magnetic Properties of Single Crystalline Mn-Zn Ferrites Containing Er₂O₃

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Soft magnetic ferrites in now use as magnetic devices have a disadvantage that the saturation magnetization is considerably lower than that of metallic soft magnetic materials. The low saturation magnetization prevents us to give a higher degree of performance for transformers and inductors and to cause an increase in the density of magnetic recordings.

The present authors found out the Mn-Zn ferrites containing Er₂O₃ with improved saturation magnetization and relative permeability.

When Er₂O₃ is added at the composition range of 0.005 ~ 0.5 mol% to Mn_{0.54}Zn_{0.36}Fe_{2.10}O₄ single crystal, saturation magnetization (σ_s) and relative permeability (μ_r') are increased by the addition of Er₂O₃, showed maximum values of 10.56×10^{-3} Wbkg⁻¹ and 640 at 5 MHz, respectively, at about 0.05 mol%Er₂O₃ and then decrease significantly with further increasing Er₂O₃ content.

At the composition range of 51 ~ 56 mol%Fe₂O₃, 24 ~ 34 mol%MnO and 13 ~ 21 mol%ZnO, the σ_s for the single crystals in Fe₂O₃-MnO-ZnO and Fe₂O₃-MnO-ZnO-0.05 mol%Er₂O₃ systems tends to increase with increasing Fe₂O₃ and MnO contents, but σ_s value of the ferrite containing Er₂O₃ increases with further increasing MnO content than that of Fe₂O₃-MnO-ZnO system. The compositional dependences of μ_r' value for single crystals in both Fe₂O₃-MnO-ZnO and Fe₂O₃-MnO-ZnO-0.05 mol%Er₂O₃ systems are nearly the same. However, the Er₂O₃-containing ferrites exhibit the μ_r' values higher than that of the ferrites without Er₂O₃ in the entire composition range. The effect of the Er₂O₃ addition on magnetic properties is closely related to the dissolve of Er³⁺ ions in the Mn-Zn ferrite.

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I. Introduction

Soft Magnetic ferrites have been used as core materials for transformers and inductors etc. by utilizing good soft magnetic properties and low core losses in high frequency range resulting from electrical resistivities higher than those for metallic soft magnetic materials. In addition, the ferrites are also known to be useful materials as a head materials for magnetic recording because of good wear resistance resulting from high hardness as well as good soft magnetic characteristics⁽¹⁾⁻⁽⁸⁾.

However, the ferrites have a disadvantage that the saturation magnetization is considerably lower than that of metallic magnetic materials. The low saturation magnetization prevents us to give a higher degree of performance for transformers and inductors and to cause an increase in the density of magnetic recordings⁽⁹⁾. For instance although the modification of composition in Mn-Zn ferrites causes a slight increase in magnetization,

it also results in a decrease in permeability⁽¹⁰⁾⁽¹¹⁾. It is therefore important for the development of a new type of soft magnetic ferrite to achieve a simultaneous enhancement of magnetization and permeability. The present authors examined the influence of the addition of various rare earth oxides on magnetic properties of single crystalline Mn-Zn ferrites which are presently in practical use as a magnetic head material for the field in a video cassette recorder (VCR). This paper is intended to present the data on the simultaneous increase in the saturation magnetization and permeability for the single crystalline Mn-Zn ferrites by the addition of Er₂O₃.

II. Experimental Method

After homogeneous mechanical mixing of Fe₂O₃, MnCO₃, ZnO and Er₂O₃ powders to desired compositions, the mixture was presintered for 4 h at 1573 K in order to produce a raw material for the formation of single crystalline Mn-Zn ferrites. The single crystal ingot was prepared in a platinum crucible by the Bridgeman method. The single crystallinity and orientation of the ingot prepared thus was examined by the light figure method. The ferrite single crystals were cut along a desired crystal orientation and then used as the sample

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for the measurement of magnetic properties. The compositions of the ingots were determined by the fluorescence X-ray analyses. The identification of the second phase was made by the electron probe microanalysis.

The measurements of magnetic properties were made at room temperature through the following procedure. The relative permeability (μ_r') was evaluated from the impedance values which were measured under a field of 0.4 A/m with a LCR meter (YHP4192A). The sample used in the evaluation of μ_r' value has a toroidal core shape with outer and inner diameters of 5 and 3 mm, respectively, and a thickness of 0.45 mm and the plane perpendicular to the toroidal axis lies along the (111) plane. It is known that the Mn-Zn ferrites with the crystal orientation have the highest μ_r' values⁽¹²⁾ and have been used as a head for the field in VCR. The saturation magnetization (σ_s) was measured for the disk-shaped sample with a dimension of 4 mm in diameter and 1 mm in thickness under an applied magnetic field of 800 KA/m by using a vibrating sample magnetometer. The surface of the disk also lies along the (111) plane.

III. Results and Discussion

Figure 1 shows the changes in the saturation magnetization (σ_s) and relative permeability (μ_r') as a function of additional amount of Er_2O_3 for the $\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ single crystal with the preferred orientation. The σ_s and μ_r' increase by the addition of Er_2O_3 , show maximum value of $10.56 \times 10^{-3} \text{ Wbkg}^{-1}$ and 640 at 5 MHz, respectively, at about 0.05 mol% Er_2O_3 and then decrease significantly with further increasing Er_2O_3 content.

In order to clarify a maximum solubility limit of Er_2O_3 into $\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ at room temperature, we examined the microstructure on the (110) plane of $\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ single crystals containing various

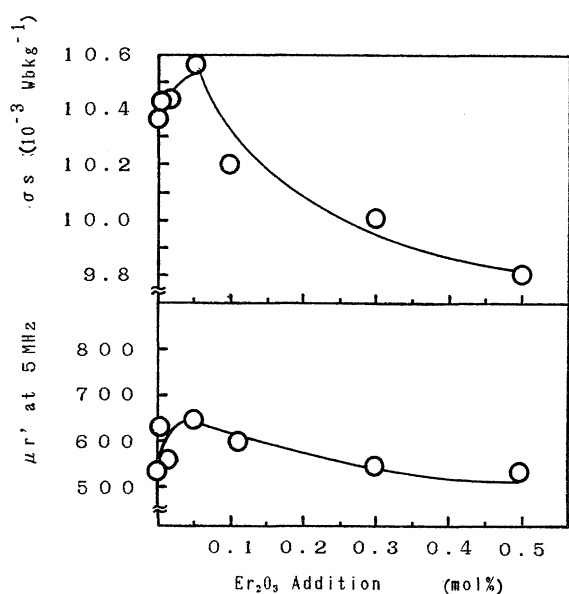


Fig. 1 Change in the saturation magnetization (σ_s) and permeability at 5 MHz (μ_r') of a $\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ single crystal by the addition of Er_2O_3 .

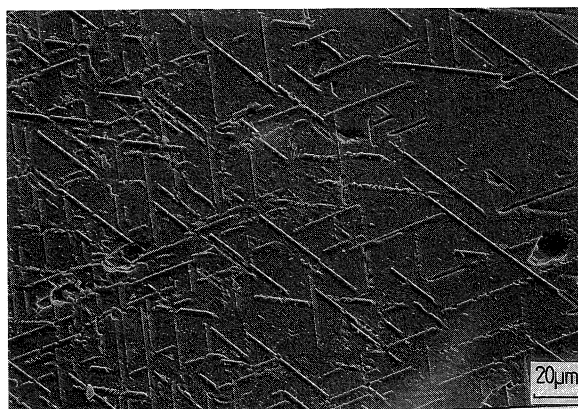


Fig. 2 Optical micrograph revealing the structure on the (110) plane of $\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ single crystal containing 0.5 mol% Er_2O_3 .

additional amounts of Er_2O_3 by scanning electron microscopy. Although no contrast revealing the existence of any precipitates is seen in the additional amount range below 0.4 mol% Er_2O_3 , the contrast of the acicular precipitates along the direction of $\langle 112 \rangle$ and $\langle 110 \rangle$ is observed for the Mn-Zn ferrite containing 0.5 mol% Er_2O_3 . Judging from the EPMA result that the Er element is detected only from the acicular precipitates, the precipitates appear to be an oxide containing Er. This result allows us to conclude that the maximum solubility limit of Er_2O_3 into $\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ lies in the concentration range between 0.4 and 0.5 mol%. It is therefore said that the additional Er_2O_3 has been dissolved in the Mn-Zn ferrite in the concentration range below 0.4 mol%. Consideration that the σ_s and μ_r' values exhibit maxima at 0.05 mol% Er_2O_3 which is much lower than the maximum solubility limit, there is no close relation between the dissolved Er_2O_3 content and σ_s or μ_r' . Accordingly, the detailed investigation on the dissolved state of Er_2O_3 will shed some light on the clarification of the reason for the effect of the Er_2O_3 addition on the magnetic properties.

Figure 3 shows the compositional dependence of σ_s for the single crystals in Fe_2O_3 -MnO-ZnO and Fe_2O_3 -MnO-ZnO-0.05mol% Er_2O_3 systems. The σ_s tends to increase with increasing Fe_2O_3 and MnO contents, but the tendency is independent of the existence of Er_2O_3 . Although the σ_s for both the systems is nearly the same in the range of 23 to 25 mol% MnO, the σ_s value of the ferrite containing Er_2O_3 increases with further increasing MnO content and the difference in σ_s reaches as large as about 10%. It is therefore said that the additional effect of Er_2O_3 is more significant in the higher MnO concentration range. It is known that an Er^{3+} ion has a larger saturation magnetization moment as compared with Mn^{2+} and Fe^{3+} ions⁽¹³⁾. However, the present additional amount of Er_2O_3 is as small as 0.05 mol% and hence no explanation on the increase in the σ_s by about 10% is given even in the assumption that the Er^{3+} ion can replace the Mn^{2+} and Fe^{3+} ions. And the Curie point (T_c) was not changed significantly by the addition of various amounts of Er_2O_3 . The reason for the extreme effectiveness of the Er_2O_3 addition is under investigation.

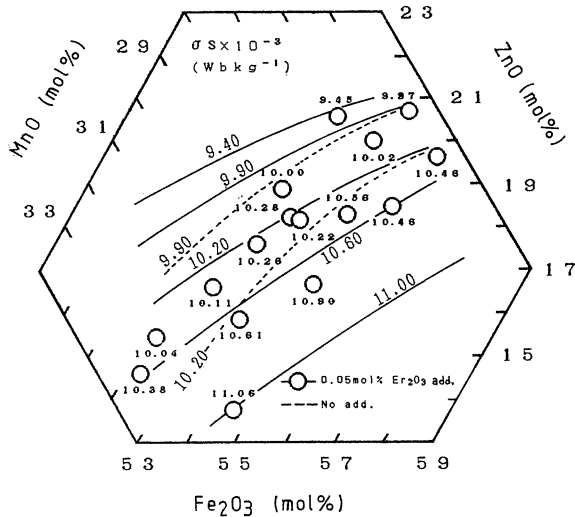


Fig. 3 Compositional dependence of σ_s for Fe_2O_3 -MnO-ZnO and Fe_2O_3 -MnO-ZnO-0.05 mol% Er_2O_3 single crystals.

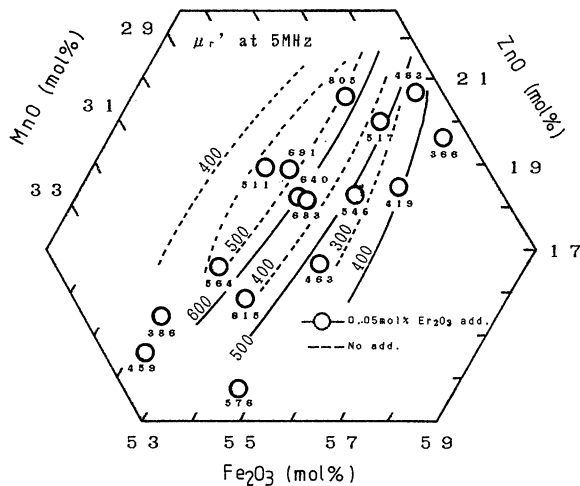


Fig. 4 Compositional dependence of μ_r' for Fe_2O_3 -MnO-ZnO and Fe_2O_3 -MnO-ZnO-0.05 mol% Er_2O_3 single crystals.

Figure 4 shows the compositional dependence of μ_r' value for the single crystals in Fe_2O_3 -MnO-ZnO and Fe_2O_3 -MnO-ZnO-0.05 mol% Er_2O_3 systems. The μ_r' value in both the systems shows a maximum around 52.5 mol% Fe_2O_3 , 29.0 mol%MnO and 18.5 mol%ZnO and decreases with a deviation of Fe_2O_3 from the composition. However, the Er_2O_3 -containing ferrites exhibit the μ_r' values higher than that of the ferrites without Er_2O_3 in the entire composition range.

On the temperature dependence of the μ_r' for $\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ and $\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ -0.05 mol% Er_2O_3 single crystals, 2nd peaks are appeared around 240 K and 250 K for the former and the latter respectively. It is considered that the shift of 2nd peak to the higher temperature with the addition of Er_2O_3 causes the μ_r' to increase at the room temperature.

It is shown in Figs. 3 and 4 that the σ_s and μ_r' values of the $\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ single crystal increase by the addition of 0.05 mol% Er_2O_3 . In order to clarify more clearly this point, the relation between μ_r' and σ_s for the

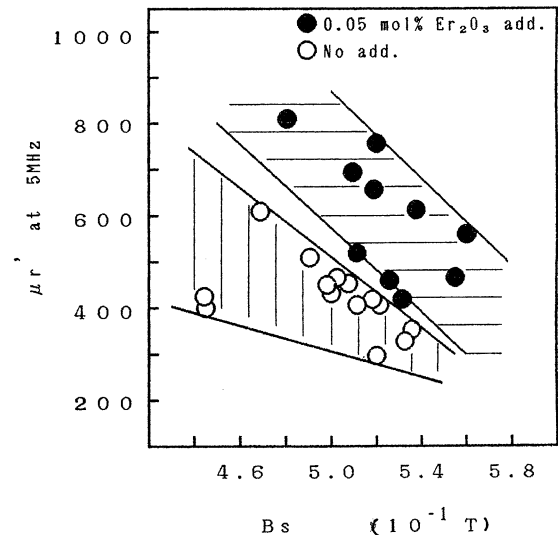


Fig. 5 Relation between σ_s and μ_r' for Fe_2O_3 -MnO-ZnO and Fe_2O_3 -MnO-ZnO-0.05 mol% Er_2O_3 single crystals.

$\text{Mn}_{0.54}\text{Zn}_{0.36}\text{Fe}_{2.10}\text{O}_4$ ferrites without Er_2O_3 and with 0.05 mol% Er_2O_3 is shown in Fig. 5. There is a clear tendency for both the systems that the μ_r' values decrease with increasing σ_s . However, the addition of 0.05 mol% Er_2O_3 causes a significant increase in both μ_r' and σ_s , indicating a significant effect of Er_2O_3 on the improvement of soft magnetic properties.

As described above, the Mn-Zn ferrites have presently been used as magnetic head materials for the field in VCR. Accordingly, the single crystalline ferrites in the Mn-Zn-Fe-O system containing about 0.05 mol% Er_2O_3 are expected to develop hereafter as a fundamental system for magnetic head materials for the field in VCR where a higher degree of performance and a higher density of magnetic recordings are required. Furthermore, there is a high possibility that the similar useful effect caused by the addition of Er_2O_3 appears for polycrystalline Mn-Zn ferrites as well as for the single crystalline Mn-Zn ferrites.

REFERENCES

- (1) Z. Igarashi: *Tekkoukai*, **139** (1989), 54.
- (2) T. Mitsui and A. Morita: *Electronic Engineering*, **129** (1987), 168.
- (3) D. Graetzerlayne and A. D. Giles: *J. Mag. Mag. Mater.*, **19** (1987), 91.
- (4) J. W. Chamberlayne and A. D. Giles: *IEE Conf. Publ. (Inst. Electr. Eng.)*, **142** (1976), 17.
- (5) K. Okutani: *Papers of Technical Meeting on Magnetics*, **MAG-89-141-155**, IEE Japan, (1989), 121.
- (6) H. Matsuo: *Electronics*, **134** (1989), 81.
- (7) E. Hirota and K. Kugimiya: *National Technical Report*, **22** (1976), 753.
- (8) H. Eiichi, K. Hirota and K. Kugimiya: *Proc. Inter. Conf. on Ferrite*, Univ. of Tokyo Press, (1980), p. 667.
- (9) T. Hiraga, K. Okutani and T. Ojima: *Ferrite*, Maruzen, (1986).
- (10) Y. Shichijo: *Trans. JIM*, **2** (1961), 204.
- (11) T. Imura, T. Shinohara and M. Kudo: *Proc. Inter. Conf. on Ferrite*, Univ. of Tokyo Press, (1980), p. 726.
- (12) R. Ishii, I. Sasaki and H. Goto: *IEIC Technical Report*, **MR74-8** (1974), 17.
- (13) For example, S. Chikazumi: *Physics of Magnetism*, John Wiley & Sons, Inc., New York, (1964).