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Effect of intragranular porosity of initial permeability and coercive force in a manganese zinc ferrite

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Initial permeability and coercive force are structure sensitive properties which depend upon intragranular porosity. It has been found that in a manganese zinc ferrite the initial permeability varies directly as the separation between intragranular pores and the coercive force varies inversely as the square root of this distance. Assuming reversible motion of the domain wall pinned at the pores, the behaviour of initial permeability with respect to the distance between the pores can be adequately explained. It is not possible to give an explanation for the observed relationship between coercive force and the distance between the pores.

1. Introduction

Ferrites have been used as inductor cores for high frequency use mainly because they have high resistivity compared to metallic cores. Depending upon the required use, different properties of the ferrite can be optimized by adjusting process parameters. Two of the more important properties of magnetic materials are initial permeability and coercive force. Both of these properties are dependent not only on the composition of the ferrite, but also on the microstructure of ferrite. This work deals with dependence of these two properties on the pore structure of the ferrite.

The initial permeability μ_i is mainly due to reversible motion of the domain walls under a very small magnetic field and contribution of the spin rotation is rather negligible as found by Globus [1]. Globus [1] has taken a model where domain walls are pinned at grain boundaries and they bulge under application of a magnetic field until a critical field, H_{cr} is reached when the wall gets unpinned. In such a model

$$\mu_i - 1 = \frac{3}{16}\,\frac{M_s^2 D}{\gamma}, \tag{1}$$

where M_s is the saturation magnetization, D the diameter of the grain, and γ the domain wall energy per unit area, since

$$\gamma \sim \sqrt{(AK_1)} \tag{2}$$

$$\mu_i \sim \frac{M_s^2 D}{\sqrt{K_1}}. \tag{3}$$

This agrees with the results of Enz [2] and Ohta [3] who showed that μ_i reaches a maximum when K_1 goes through zero.

Such a model can be extended to the case where the domain walls are pinned at intragranular pores and D can be interpreted as the distance between the intragranular pore, i.e. the span of the domain wall.

Kernsten [4] has also considered bulging of domain wall pinned between points and found that

$$\mu_i = \frac{5\,M_s^4 D}{\pi\,K_1^2 d_s}, \tag{4}$$

where d_s is the spacing between domain walls, and D is the span of the domain wall. In both these models $\mu_i \sim D$.

The effect of intragranular porosity has also been considered by Kernsten [5] and Dijkstra and Wert [6]. Kernsten considered only surface tension effects of pores derived the coercive force, H_c as

$$H_c \sim \frac{K_1}{M_s}\,Sp^{2/3} \tag{5}$$

where p is the intragranular porosity, and S a factor depending on the ratio of the domain wall width, δ, to pore diameter, d. The value of S is maximum when $d \approx \delta$. Dijkstra and Wert [6] considered spike formation at the pores and have arrived at similar conclusions although the functional dependence of H_c on p and d according to them is different, as given below:

$$H_c = \frac{3\,\gamma_{180^\circ}\,p\,L}{M_s\,d^2} \quad \text{for } d \gg \delta \qquad (6)$$

$$= 2.8\left(\frac{M_s\,d^{7/12}}{D\,\delta^{5/2}_{180^\circ}}\right) p^{1/2} \ln\frac{2D}{\delta_{180^\circ}} \quad \text{for } d \ll \delta,$$

where γ_{180° is the wall energy of 180° domain wall of width δ_{180° and L is the length of the spike.

2. Experimental procedure

The manganese zinc ferrite used in this work was prepared by the familiar ceramic process. Manganese carbonate, iron oxide and zinc oxide (all 99% pure or better) were mixed in a ball mill, calcinated at 1150° C for 2 h, and ground to a particle size less than 1 μm. Raw materials of commercial purity were intentionally used to obtain intragranular porosity. Toroids were pressed out of the calcinated powder and sintered in air between 1250 and 1350° C for different time durations. In some cases the toroids were soaked in nitrogen for at least 1 h of the sintering time and then cooled in nitrogen; in other cases, they were cooled in air. The rate of cooling was $\sim 100^\circ\,\mathrm{C\,h^{-1}}$ in all cases.

The coercive force was measured by the ballistic galvanometer method and the initial permeability was measured at 1 kHz using a Siemens Universal Bridge. The toroids were polished after grinding about 1 mm of the surface and etched in boiling hydrochloric acid for metallographic examination. The grain size was determined by the standard technique of counting the number of grain boundaries crossing a straight line and the distance between pores was determined by dividing the grain size with the square root of the average number of the pores inside the grain.

3. Results and discussion

In order to optimize the initial permeability, the composition of the Mn–Zn ferrite was so chosen that λ and K_1 would be small and the Curie temperature was $\sim 150^\circ$ C. The optimum initial permeability in air sintering was 850 ± 10%. The initial permeability could be increased to 1050 by cooling in nitrogen for obvious reasons. Because the raw materials were relatively impure and contained up to 1% of silica, discontinuous grain growth was favoured. Silica remains on the grain boundary and reduces the activation energy for grain growth. When the rate of growth is fast, the pores on the grain boundary can no longer move with it and hence become detatched and intragranular porosity results. Such pores are non-

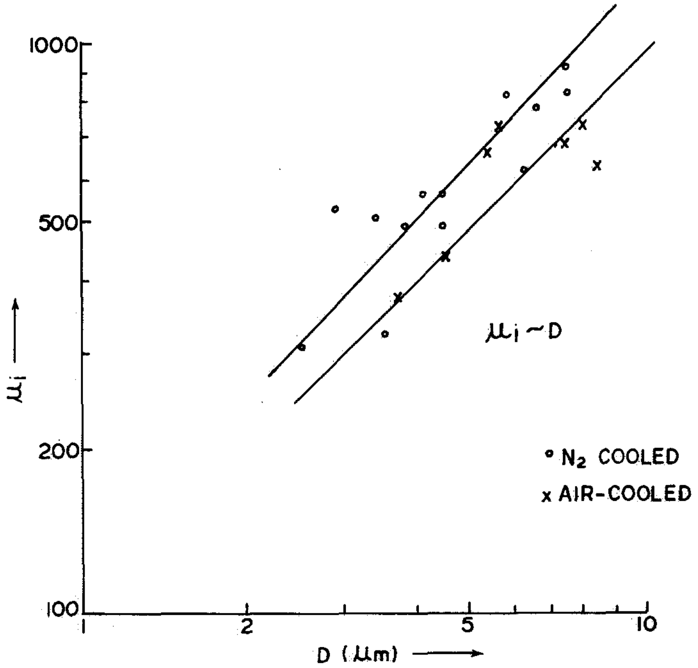

Figure 1 Variation of initial permeability (μ_i) of an Mn–Zn ferrite with distance between intragranular pores (D). $\mu_i \sim D$ and is higher for ferrites cooled under nitrogen.

magnetic inclusions and hence can pin down the domain wall thus restricting its motion.

Fig. 1 shows the dependence of initial permeability on pore-to-pore distance, D. The slope of the logarithmic plot is 1 and so the initial permeability is directly proportional to D. This follows the relationship arrived at by Kernsten [4] and Globus [1] for initial permeability by considering domain wall motion. Perduijn and Peloschek [7] and Roess [8] have shown that the initial permeability of Mn–Zn ferrites is linearly dependent on the average grain diameter D_m if the grains are pore-free. Similar relationship between μ_i and grain diameter of pore-free grains was also observed for YIG by Globus and Duplex [9]. In pore-free grains the domain walls are pinned at the grain boundaries and hence the distance between the pinned points is D_m, making $\mu_i \sim D_m$. These observations corroborate the findings of the present work.

It has already been discussed that the coercive force of a ferrite depends upon K_1, the distance between pores, D, and the diameter of the pores, d. For low coercive force both D and d should be large and K_1 should be small. The thickness of the domain wall is given by

$$\delta = \sqrt{\left(\frac{A}{K_1}\right)} \approx \sqrt{\left(\frac{KT_c}{aK_1}\right)} = \sqrt{\left(\frac{1.37 \times 10^{-23} \times 423}{3 \times 10^{-10} \times 10^{2}}\right)} \simeq 0.4\,\mu\text{m}, \tag{7}$$

where A is the exchange interaction energy density, a is the interionic distance and T_c is the curie temperature. Most of the pores observed optically in the course of this work were of the order of 1 μm diameter. It is possible that the measured size could be bigger than the actual size because etching makes it appear so and the resolution of the microscope is $\sim 1\,\mu$m. The pore diameters were slightly bigger than the domain wall width in this case and S was near the maximum. For this analysis it is assumed that the pore diameters were the same from sample to sample.

Fig. 2 gives the relationship between H_c and pore-to-pore distance, D. The slope on the logarithmic plot is -0.5 and hence $H_c \sim D^{-0.5}$. The coercive force could not be appreciably affected by maintaining a nitrogen atmosphere during cooling. This may presumably be due to relatively impure raw materials used.

Assuming that pore diameter does not change from sample to sample, $H_c \sim D^{-2}$ according to Kernsten [5], and $H_c \sim D^{-1}$ according to Dijkstra and Wert [6]. The difference between the observed relationship $H_c \sim D^{-0.5}$ and various theoretical relationships cannot be attributed to the assumption that pore diameters were the same from sample to sample. For large D, the pores were slightly larger, as expected under prolonged sintering, and hence the value of S of these samples would be lower. The magnitude of the slope of the observed logarithmic plot of H_c/S

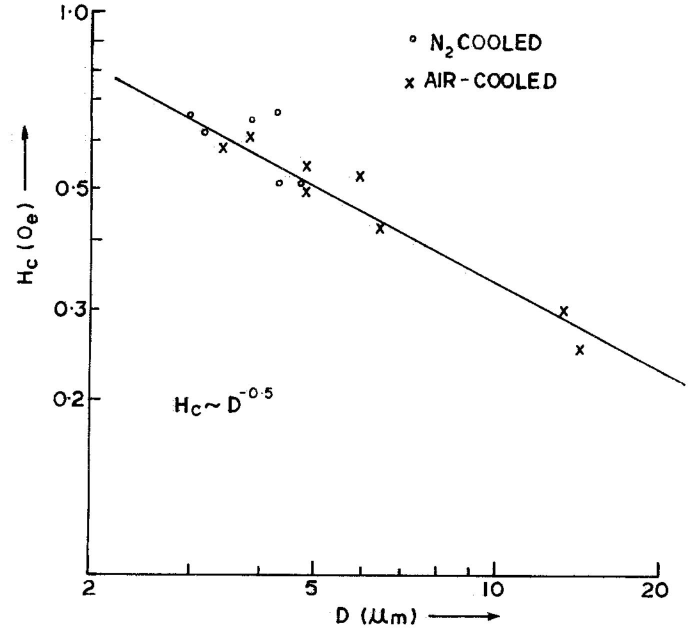

Figure 2 Variation of coercive force (H_c) with distance between intragranular pores (D). $H_c \sim D^{-0.5}$.

versus D will be less than 0.5 since at high values of D, S is smaller. S can also vary if the domain wall thickness changes due to a change in the effective anisotropy constant [1] caused by stress or by variation in ferrous ion content. However, such an effect will cause a random variation of H_c and should not systematically affect the exponent of the equation $H_c \sim D^{-n}$. Hence it can be concluded that although the pinning model of Kernsten and others can satisfactorily explain the dependence of μ_i on D, it cannot explain the dependence of H_c on D. This may be because the initial permeability is given by a small displacement from the pinned position whereas for H_c, the domain wall has to bow out considerably (the domain wall energy of Mn–Zn ferrite being quite low) before breaking away from the pores. Kernsten assumed the grain diameter to be much larger than D, but in the ferrite under consideration it is not; this grain sizes varying from 10 to 35 μm, D varying from 3 to 15 μm, and the grain boundaries, may all affect the bowing of the domain through the presence of closure domains near them, causing a high H_c which results in a lowering of the exponent n.

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