FORMATION OF MICROSTRUCTURE AND ITS EFFECTS ON THE PROPERTIES OF LITHIUM-CONTAINING RECTANGULAR HYSTERESIS-LOOP FERRITES

L. M. Letyuk

The rapid advances being made in computer technology impose more and more stringent requirements concerning the level of electromagnetic parameters of ferrites with a rectangular hysteresis loop (RHL), in particular their thermal stability and quick operation [1]. In this field of application, ferrites based on the system Li₂O-Fe₂O₃ show considerable promise. These ferrites are characterized by great thermal stability of electromagnetic parameters (their temperature coefficient of coercive force is < 0.15%/°C) and a wide range of coercive force values (1-10 Oe), and enable compositions to be formulated which are capable of very quick operation. One of the key factors in the formation of electromagnetic properties in lithium-containing RHL ferrites is their ceramic structure (microstructure) and its individual characteristics [2, 3].

In a magnetic material, the most likely sites of formation of magnetic polarity reversal nuclei are its grain boundaries. It follows, therefore, that certain microstructure parameters (mean grain size, grain shape, porosity, and grain size variation — so-called intraphase heterogeneities) can be expected to exert a strong influence upon the conditions of formation and displacement of the boundaries of reversed magnetization domains and largely determine the shape of the hysteresis loop [4-7]. In its turn, the formation of the microstructure of ferrites depends to a large extent upon the recrystallization processes occurring during the sintering of these materials [3].

In the present work, a study was made of the formation of microstructure and its effects upon the properties of the temperature-stable RHL ferrites of the general chemical formula Li₀.₅(l-x)MnxFe₂.₅₋₀.₅O₄ (where 0 ≤ x ≤ 0.3). To these ferrites were added, after precalcining (800°, 3 h), the low-melting-point compounds Bi₂O₃ and V₂O₅ in amounts ranging from 0.5 to 4 wt.%. Ferrite cores of two sizes, 5 × 3 × 2 and 0.6 × 0.4 × 0.13 mm, were investigated. The mean grain sizes of the materials and their grain-size variations were determined with the aid of photographs of microsections taken under an MIM-8 microscope; results of measurements were converted to give data characterizing the volume microstructure. In assessing grain-size variations, use was made of the ratio σ²/μ, where σ² is the mean square deviation in grain-size distribution and μ the mean grain size.

Examination of the microstructures of ferrites (Fig. 1) calcined at various temperatures in the presence of Bi₂O₃ and V₂O₅ additions, which form during sintering a eutectic-type liquid phase with a comparatively low melting point (about 600-700° [8]), and without them and also of the relationships illustrated in Fig. 2 shows that the microstructure-formation processes differ substantially in these two cases. Some of the salient features of the processes are:

1) A well-developed microstructure is obtained in these ferrites after calcining at temperatures as low as 750-800°C in the presence of low-melting-point additions and only after calcining at 1100°C without such additions. The process of grain growth in ferrites with fluxing additions can conveniently be divided into two stages — one of comparatively slow and the other of intense grain growth. The transition from the first to the second stage takes place over the temperature range 1100-1200°C. In ferrites without low-melting-point additions, the stage of slow growth is virtually absent; starting from a temperature of 1100°C, the grains at once begin to grow rapidly.


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2) The quantity $\Delta L/\Delta T$, which characterizes the rate of grain growth, is greater for ferrites with $\text{Bi}_2\text{O}_3$ and $\text{V}_2\text{O}_5$ up to calcining temperatures of $1000-1100^\circ\text{C}$, but above these temperatures the quantity $\Delta L/\Delta T$ is greater for ferrites without low-melting-point additions. At temperatures of $1250-1300^\circ\text{C}$, the values of $\Delta L/\Delta T$ for ferrites with and without additions become equalized.

3) The maximum grain-size variation is much less in the microstructure of ferrites sintered with $\text{Bi}_2\text{O}_3$ and $\text{V}_2\text{O}_5$ additions than in that of ferrites sintered without additions.

In the light of these findings and of the results of earlier investigations [9-12], the process of microstructure formation in ferrites calcined in the presence of fluxing additions can be represented as follows. The liquid phase forming at a temperature of about $650-700^\circ\text{C}$ apparently becomes evenly distributed among the particles of the starting powder. This reduces interparticle friction and enables the particles to move freely relative to one another. Apart from this, various protuberances on the solid particles dissolve in the liquid phase and the dissolved material is then deposited in concave parts, causing the particle shape to change from polygonal to spheroidal. This leads to reorientation and much closer packing of the particles, as a result of which the ferrite rapidly shrinks and its density increases. At a certain degree of interparticle contact, the particles begin to sinter together. They then cease to exist as separate structural constituents, and a monolithic polycrystalline solid is formed having a characteristic microstructure of its own. High-temperature grain growth in this solid is due to individual atoms diffusing through the liquid phase, and collective recrystallization can only occur within the boundaries of individual particles. Intergranular collective recrystallization is prevented by the presence of thin films of molten material enveloping the grains. Microstructure formation due to the diffusion of individual atoms through the liquid phase is characterized by a slow and uniform growth of crystallites. At high sintering temperatures, the liquid phase can partly evaporate or drain away, enabling intergranular collective recrystallization, characterized by a rapid and uneven growth of crystallites, to take place.

In the absence of a liquid phase, the sintering process is less intense, as a result of which the necessary degree of interparticle contact and hence the initial instant of grain growth are attained at temperatures some $250-300$ deg C higher than those characterizing the beginning of grain growth in ferrites sintered in the presence of a liquid phase. As ferrites without additions have no liquid phase preventing grain migration, crystallite growth in them is apparently due mainly to intergranular collective recrystallization, as a result of which their microstructure exhibits greater grain-size variation. The stage of slow crystallite growth in this case is practically nonexistent, and consequently the values of $\Delta L/\Delta T$ at calcining temperatures of up to $\sim 1050-1100^\circ\text{C}$ exceed those characteristic of ferrites without additions. Above