Abnormal Dielectric Behaviour of Li-Ti Ferrite Ceramics

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Abstract – Field dependences of Li-Ti ferrite ceramics polarization are obtained. When related to temperature, these dependences exhibit either non-linear behavior or non-linearity and hysteresis. Electric Barkhausen's pulses are revealed. Temperature dependences of the electric pulse heights are obtained for polarized by electric field ferrite specimens. The results provide evidence on the presence of ferroelectric properties in Li-Ti ferrite ceramics.

1. Introduction

The ferrite ceramics of the lithium group are materials whose properties have been extensively studied [1–3]. In [4, 5], it was shown that a ceramic material of the composition $Li_{0.649}Fe_{1.598}Ti_{0.5}Zn_{0.2}Mn_{0.051}O_4$ with an addition of 0.22% Bi_2O_3 can also possess ferroelectric properties.

We have obtained temperature dependences of the complex dielectric permittivity at the frequencies 10^2, 10^3, 10^4 and 10^6 Hz, which exhibited anomalies when the measurements were made at a high level test-signal (E_{test} = 120 V/cm) at a frequency of 10^2 Hz [4]. The anomalous behavior consisted in a sharp decrease in the values of the real part ε′ of dielectric permittivity in the vicinity of the magnetic Curie temperature (575 K). When these dependences were obtained at a low test-signal level (E_{test} = 2.3 V/cm) at a fixed-bias voltage we also observed anomalies characteristic of ferroelectric, manifested as sharp changes in the values of ε′ and ε″ in narrow temperature intervals [4, 5]. It was, therefore, suggested that the relaxation polarization is of ferroelectric nature [4, 5].

This work describes the experiments on determination of electric Barkhausen's pulses [6], non-linearity and hysteresis in the dependences $P = f(E_P)$ aimed at acquiring additional data on ferro- or anti-ferroelectric properties of Li-Ti ferrite. Here, P and E_P are the polarization vector and electric field strength values, respectively.

The sintering technology and the process of specimen preparation were the same as in [4]. The specimens were shaped as tablets measuring 13 mm in diameter, with silver electrode deposited onto their surface by thermal evaporation in vacuum. The electrode diameter and the specimen thickness were 5 and 0.24 mm, respectively.

2. Dielectric hysteresis

In order to identify non-linearity and hysteresis of dielectric characteristics, the dependence of C on E_P was measured at different temperatures, where C is the specimen capacity. The capacity measurements were made with an automatic LCR-meter E7–14. The dependence $P = f(E_P)$ was obtained by a graphic integration of the curves $C = f(E_P)$.

Shown in Fig. 1 are the field dependences $P = f(E_P)$. These curves were obtained in the following manner. A negative bias voltage value $-|U_{max}|$ was invariably set at the specimen, and the capacitance was measured for a low variable signal (i.e., differential capacitance). Measuring the bias voltage up to $+|U_{max}|$, we obtained the dependence of the differential capacitance C on the field strength E_P in a direct and then reverse run (from $+|U_{max}|$ to $-|U_{max}|$). The capacitance was measured at a frequency of 10 kHz using a E7–14 bridge. At elevated temperatures, the results of the first measurement cycle were different from the reproducible results of the second and further measurements. Thus, a few measurements cycles were made at these temperatures, and use was made of the data obtained in the third one. Each measurement was performed after a 1-minute tempering.

Using $C(U) = dq/dU$, $dq = C(U)dU = C(U)dE_PL$, where L is the specimen thickness, and integrating $C(U)$ in the direct and reverse runs, we obtain

$$\left(\int_{-|E_{P\max}|}^{E_P} C(E_P)dE_P + \text{const}\right)L = q(E_P) \quad (1)$$

$$\left(\int_{-|E_{P\max}|}^{+|E_{P\max}|} C(E_P)dE_P + \int_{+|E_{P\max}|}^{E_P} C(E_P)dE_P + \text{const}\right)L = q(E_P) \quad (2)$$

Here, Eqs. (1) and (2) are the direct and reverse runs of $q(E_P)$, and q and U are the charge and voltage of the capacity. Graphic integration was carried out using an ORIGIN program. In this manner, two branches were constructed $P(E_P) = q(E_P)/S$, where S is the area of the electric contact.

It was found out that at low temperatures ferrite behaves as a linear dielectric up to E_P = 5 kV/cm. While at T = 425, 455 and 475 K, non-linear dependences were observed $P(E_P)$, there is no hysteresis. At the temperatures 495 and 520 K (Fig. 1, *a*, *b*), non-linear dependences with a pronounced hysteresis are seen. Note that at still higher temperatures, there is also a hysteresis in the curve, with the initial polarization value in the direct branch differing from the final one in the reverse branch after multiple measurement cycles.

The data presented in Fig. 1 show vividly that the polycrystalline ferrite material under study exhibits special features characteristic of dielectric materials, which are likely to be due to the ferroelectric character of relaxation polarization.

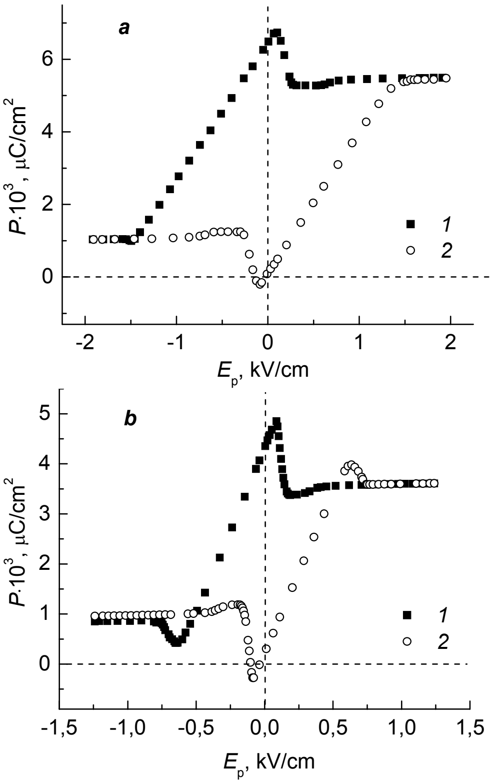

Fig. 1. Dependences of polarization on electric field strength at different temperatures 485 K (*a*) and 520 K (*b*). Direct run (from $-|E_{P\,\max}|$ to $+|E_{P\,\max}|$) (curve *1*), reverse run (from $+|E_{P\,\max}|$ to $-|E_{P\,\max}|$) (curve *2*)

3. Barkhausen's thermal effect

Observation of the electric Barkhausen's pulses was performed in a setup shown in Fig. 2, whose flow block is given in the insert to the figure. Measurements were made in the course of slow (2°/min) heating of the specimens within the temperature interval $T = 300$–600 K. A bias voltage was applied to the specimens. Detection of the electric pulses was done with a computer-controlled Velleman oscilloscope.

A typical pulse form is shown in Fig. 2. Figure 3 depicts the temperature dependence of the maximal Barkhausen's pulse heights for polarized by electric field ferrite specimens. It is obvious from Fig. 3 that the temperature interval where the pulses are detected are T=465–575 K, on the understanding that polarization electric field strength was E_p>80 V/cm. Over the range E_p<80 V/cm electromagnetic noise was observed with pulse amplitude as low as 100 mV.

One of the reasons for sharp decrease of the pulse height at the temperature 575 K might be decomposition of the domains resulting from the transition of the specimen from ferro-electric into paraphase state [7].

Thus, non-linearity and hysteresis of the field dependence of the ferrite polarization value have been observed, and Barkhausen's electric pulse generation has been detected for polarized by electric field ferrite specimens. These results vindicated earnestly our conjecture about ferroelectric properties by Li-Ti ferrite ceramics over the temperature range 475–575 K, induced by electric field.

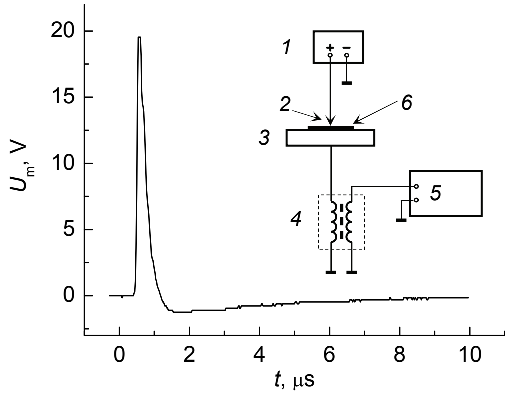

Fig. 2. Typical electric pulse form at $T = 510$ K, $E_P = 125$ V/cm. The insert shows the experimental schematics: direct current power supply – *1*, upper electrode – *2*, lower electrode with heater – *3*, transformer – *4*, Velleman oscilloscope – *5*, and tested specimen – *6*

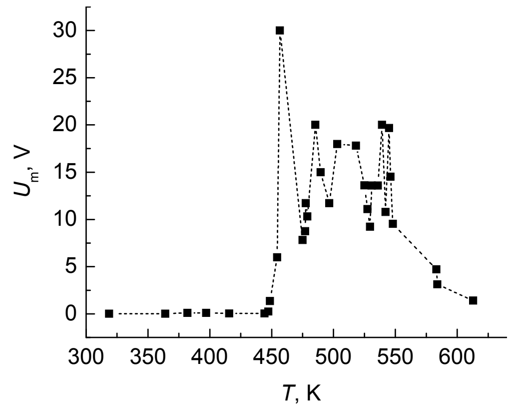

Fig. 3. Maximum Barkhausen's electric pulse height versus temperature for polarized by electric field ferrite specimens

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