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Influence of external electric field on relaxor behaviour of $(Pb_{0.75}Ba_{0.25})(Zr_{0.70}Ti_{0.30})O_3$ ceramics

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Abstract

The effects of the external d.c. and a.c. electric field on the dielectric response and relaxor behaviour of lead–barium–zirconate–titanate (PBZT) ceramics of the composition Ba/Zr/Ti 25/70/30 have been studied. The significant influence of the strength of the d.c. bias field and the amplitude of the a.c. field on the dielectric constant maximum and frequency dispersion was determined. The magnitude of the dielectric response strongly decreases under the bias field and increases with the increase of the a.c. field amplitude. All parameters describing the relaxor behaviour of the studied ceramics changed under the external d.c. and a.c. electric field. The experimental results can be explained in terms of existing models of relaxors.

Keywords: C. Dielectric properties; Relaxor ferroelectrics; Ceramics

1. Introduction

Dielectric properties of $(Pb_{0.75}Ba_{0.25})(Zr_{0.70}Ti_{0.30})O_3$ (PBZT 25/70/30) ceramics have been broadly investigated and the behaviour of these ceramics typical for the relaxor ferroelectrics was confirmed [1–4]. Relaxor ferroelectrics (RF) are characterized by the existence of a strong non-Debye frequency dispersion of the dielectric constant (ε'). The dielectric dispersion occurs in a broad temperature region of the diffuse dielectric constant maximum (ε'_{max}). The peak of $\varepsilon'(T)$ and the corresponding temperature T_m move towards higher temperatures with the increasing frequency of the measuring field. The dielectric behaviour of Pb-containing relaxors is generally explained in the literature in terms of small regions of local spontaneous polarisation (so-called polar regions) with a nanometre scale size [5,6] which appear on cooling at the temperature T_B much higher than T_m.

Recently published results indicate that the experimental investigations of the RF under high external field conditions could provide a basis for better understanding of the processes and mechanisms responsible for their particular macroscopic behaviour [7–10]. In the present paper, the effects of the external d.c. and a.c. fields on the dielectric response and relaxor behaviour in the vicinity of the diffuse ferroelectric–paraelectric phase transitions in PBZT 25/70/30 are studied. The weak field dielectric constant depends on measurement procedures reflecting different electrical and thermal history of the studied samples [7]. Therefore, the investigations were carried out for the different electrical states of the ceramic samples (field-heated after zero-field-cooled; zero-field-heated after field-cooled).

2. Ceramics preparation and experimental procedure

The $(Pb_{0.75}Ba_{0.25})(Zr_{0.70}Ti_{0.30})O_3$ ceramics were prepared using a conventional method of sintering. Thermal synthesis of the mixed and pressed PbO, $BaCO_3$, ZrO_2 and TiO_2 oxides was carried out at 925 °C for 2 h. The crushed,

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milled, and sieved material was pressed into cylindrical pellets and sintered again at 1250 °C for 4 h. This procedure was repeated before the final sintering at 1300 °C for 7 h. During sintering, the ceramics were placed inside a double crucible with some amount of PbO and ZrO_2 in order to preserve the predetermined composition, and especially to avoid the loss of PbO caused by its sublimation. The weight losses were smaller than 1%. Archimedes displacement method with distilled water was employed to evaluate sample density. Sample bulk density was 6.8 g/cm^3 (92% of the theoretical density). The grain structure and the atomic composition of the obtained ceramics were examined by a scanning electron microscope JSM-5410 equipped with an energy dispersive X-ray spectrometer. The average grain size was ca. 15 μm. The EDS analysis indicated a homogenous distribution of all the elements throughout the grains.

Samples 0.2–0.6 mm thick coated with sliver electrodes were used for measurements of the dielectric constant as a function of temperature. The weak field dielectric constant, ε', was measured using an HP4192A impedance analyzer in the frequency range 0.1–100 kHz, on heating from 50 to 300 °C, at bias levels of 0, 0.5, 1, 1.5, and 2 kV/cm. The effect of external a.c. field was investigated in the same temperature and frequency range using an HP4284A LCR meter. The amplitude of the a.c. measurement field was varied from 0.01 kV/cm (the field level normally used in the measurements of the weak field dielectric constant) up to 1 kV/cm. Before the measurements, the samples were rejuvenated by annealing for 30 min at 300 °C and then cooled down to 50 °C without applying the external electric field (zero-field-cooled state, ZFC). The weak field dielectric constant was also measured for the samples after initial polarisation in the d.c. field of strengths 1, 2, and 4 kV/cm, applied for 30 min at 250 °C and then cooled in the field to 50 °C (field-cooled state, FC). All the measurements were carried out on heating with constant rate 0.5 °C/min.

3. Results and discussion

The temperature dependencies of the weak a.c. measurement field (E_m = 0.01 kV/cm) dielectric constant and the loss factor measured at various frequencies are shown in Fig. 1. They demonstrate the typical relaxor behaviour with the magnitude of the dielectric constant decreasing with increasing frequency and the maximum shifting to higher temperatures.

The dependence of the 1 kHz dielectric response on various bias levels from 0.5 to 2 kV/cm (bias field-heated state, BFH) is illustrated in Fig. 2. It can be seen that the electric field very strongly reduces the dielectric constant and decreases the temperature T_m of its maximum (see the inserted plot in Fig. 2). The similar results were obtained for the other frequencies. The dependencies of $\Delta\varepsilon'_{max}$, defined as the difference between ε'_{max} measured at 0.1 and 100 kHz, and ΔT_m, defined in similar manner on the bias field are listed in Table 1. The increase of these parameters means that the mechanism responsible for the relaxor behaviour of the investigated ceramics strongly depends on the bias field.

The influence of an initial polarisation (field-cooled state, FC) on the dielectric response measured at 1 kHz is shown in Fig. 3. The polarising electric field strength effect on ε'_{max} is

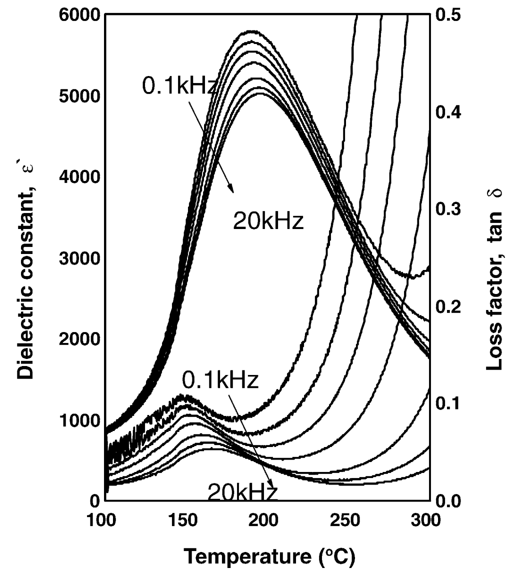

Fig. 1. Dielectric constant (ε') and loss factor (tan δ) as a function of temperature at frequencies of 0.1, 0.2, 0.4, 1, 4, 10, and 20 kHz for PBZT 25/70/30 ceramics. The amplitude of the a.c. measurement field, E_m = 0.01 kV/cm.

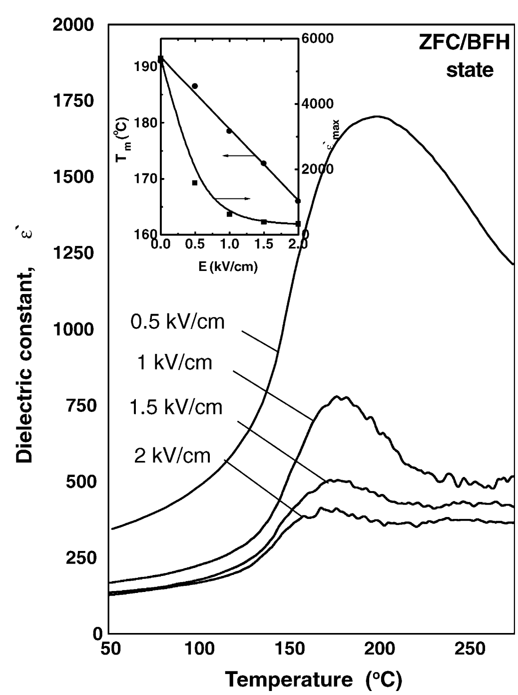

Fig. 2. Dielectric constant as a function of temperature, measured at 1 kHz for various the d.c. bias field.