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Dielectric behavior of samarium-doped $BaZr_{0.2}Ti_{0.8}O_3$ ceramics

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ABSTRACT

The dielectric properties and phase transition of Sm^{3+}-doped $BaZr_{0.2}Ti_{0.8}O_3$ (BZT20) ceramics were investigated. Room temperature X-ray diffraction study suggested that the compositions had single-phase cubic symmetry. Microstructure studies showed that the grain size decreased and that the Sm_2O_3 amount markedly affected the dielectric properties of BZT20. A dielectric constant of 5700 at 0.2 mol% Sm_2O_3 and a dissipation factor of only 0.0011 at 2 mol% Sm_2O_3 were observed, indicating that BZT20 had significant potential applications. Moreover, the dielectric constant, dissipation factor, phase-transition temperature, and maximum dielectric constant increased with increased Sm_2O_3 amount at $\leq$0.2 mol% Sm_2O_3 but decreased with increased Sm_2O_3 amount at $>$0.2 mol% Sm_2O_3.

1. Introduction

Barium titanate ($BaTiO_3$) has superior dielectric properties and is extensively used in electronic components [1–3]. To reduce the dissipation factor at low frequencies, ZrO_2 is doped into $BaTiO_3$ to substitute for Ti^{4+} ions on B sites and form $BaZr_xTi_{1-x}O_3$ (BZT) [4–7]. BZT materials are receiving increased attention in the field of materials science because their structure and physical properties significantly depend on the titanium and zirconium contents of the matrix. Most studies are focusing on the preparation, microstructure, and dielectric properties of BZT ceramics [8–10]. These properties can be modified by various possible substitutions of Ba^{2+} on A sites or of Ti^{4+} or Zr^{4+} on B sites independently or simultaneously in the perovskite structure [11,12]. These dopants can be isovalent or heterovalent.

The nature of ferroelectric phase transition at the transition temperature (T_m) of $BaZr_xTi_{1-x}O_3$ bulk ceramics is known to change strongly with the Zr content [4,5]. For Zr contents more than 0.08, BZT bulk ceramics show a broad curve of dielectric constant (ε) versus temperature (T) near T_m because of the inhomogeneous distribution of Zr ions on Ti sites and the mechanical stress in grains. With increased Zr content, the phase transition temperatures approach one another until only one phase transition exists at Zr contents of about 0.20. Proper doping can reportedly induce diffuse phase transition (DPT) [13–15], indicating that doping is a promising method of improving the dielectric properties of BZT ceramics. In the present study, a series of $BaZr_{0.2}Ti_{0.8}O_3$ (BZT20) + $x Sm_2O_3$ (x = 0.0–2 mol%) ceramics was prepared by conventional ceramic processing. The aim was to determine the effects of Sm_2O_3 content on the dielectric behavior of BZT20 ceramics.

2. Experimental

BZT20 ceramic specimens were prepared by a conventional solid-state reaction using reagent-grade $BaCO_3$, TiO_2, and ZrO_2 as starting materials. Different Sm_2O_3 amounts (0 mol%, 0.1 mol%, 0.2 mol%, 0.4 mol%, 0.8 mol%, and 2 mol%) were added to the calcined powder by wet mixing. The ball-milled raw mixture was calcined at 1150 °C for 2 h, and the pellets were sintered at 1260–1320 °C for 2 h in air. The sintered samples were cleaned in an ultrasonic bath and dried. For dielectric property measurements, both sides of each specimen were screened with Ag electrode paste and then fired at 530 °C for 10 min.

The crystal structure of the samples was analyzed by X-ray diffraction (XRD; GIRAKU D/MAX 2500 V/PC, Cu K_α) from 20° to 70°. The samples were also observed by scanning electron microscopy (ESEM; Philip XL 30 ESME). The bulk densities of the samples were measured by the Archimedes' method, and the results can be seen in Table 1. The densities of the samples were in the range of 95% of the theoretical of the specimens. The temperature dependence of the dielectric constant (ε) and

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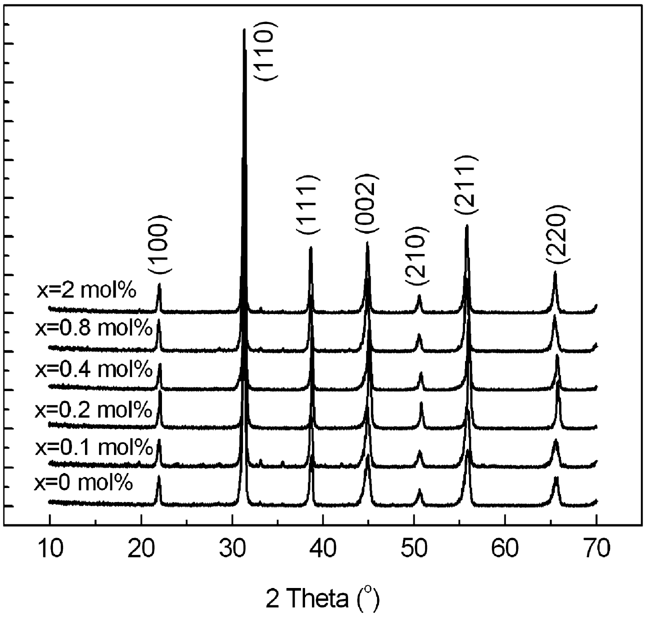

Fig. 1. XRD patterns of the Sm_2O_3 doped BZT20 ceramics.

dissipation factor (tan δ) were measured using a capacitance apparatus (Model YY 281 automatic LCR Meter 4225) at different frequencies (0.1–10 kHz) and temperatures (−30 °C to 125 °C). The phase-transition temperature (T_m) was determined from the temperature dependence of the dielectric constant.

Table 1
Bulk density, ε_{max} and T_m of BZT20 ceramics with different amount of Sm_2O_3.

x (mol%)	Bulk density (g/cm^3)	ε_{max}	T_m (°C)
0.0	5.67	7698	−22
0.1	5.74	8367	−20
0.2	5.79	8404	−15
0.4	5.76	9195	−19
0.8	5.80	7466	−27
2	5.72	–	<−30

3. Results

3.1. Crystal structure and microstructure

The XRD patterns of the samples with different Sm_2O_3 amounts sintered at 1260 °C for 2 h are shown in Fig. 1. The patterns indicate that all samples are in cubic perovskite phase. The peaks slightly shift toward higher angles at ≤0.2 mol% Sm_2O_3, indicating a decrease in the lattice parameter *a*. By contrast, the peaks slightly shift toward lower angles at >0.2 mol% Sm_2O_3, indicating an increase in the lattice parameter *a*. Fig. 2 shows the SEM images of Sm^{3+}-doped BZT20 ceramics. The diameter of pure BZT20 granule is approximately 6 μm. After doping with >0.1 mol% Sm_2O_3, the grain diameter decreases to approximately 4 μm [Fig. 2(b)–(e)] and approximately 3 μm [Fig. 2(f)].

3.2. Dielectric properties and DPT

Fig. 3 shows the dielectric constants of the samples at 1 kHz. The dielectric constants increase with increased Sm_2O_3 up to

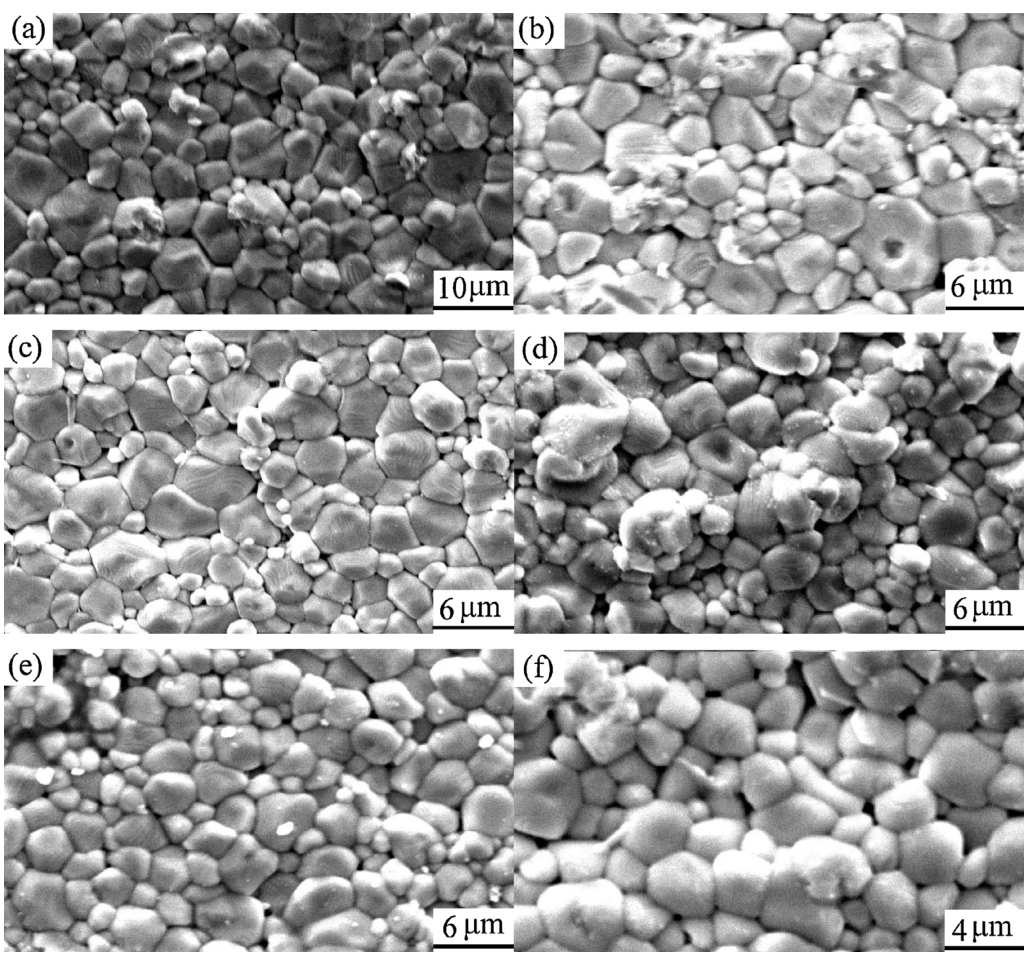

Fig. 2. Surface SEM images of the Sm_2O_3 doped BZT20 specimens: (a) 0.0 mol%, (b) 0.1 mol%, (c) 0.2 mol%, (d) 0.4 mol%, (e) 0.8 mol%, (f) 2.0 mol%.