Materials Letters 128 (2014) 263–266

Contents lists available at ScienceDirect

Materials Letters

journal homepage: www.elsevier.com/locate/matlet

Morphological evolution of tridymite crystal in $SrO–BaO–Nb_2O_5–CaO–SiO_2–B_2O_3$ ferroelectric glass-ceramic

Xiaoyan Liu, Yongping Pu*, Peikui Wang, Zijing Dong, Zixiong Sun, Yao Hu

School of Materials Science and Engineering, Shaanxi University of Science & Technology, Xi'an 710021, China

ARTICLE INFO

Article history:
Received 9 January 2014
Accepted 20 April 2014
Available online 26 April 2014

Keywords:
$SrO–BaO–Nb_2O_5–CaO–SiO_2–B_2O_3$
Tridymite
Morphological feature
Ferroelectrics
Crystal growth.

ABSTRACT

In order to increase the breakdown strength of ferroelectric glass-ceramic, morphological control of tridymite in $SrO–BaO–Nb_2O_5–CaO–SiO_2–B_2O_3$ glass-ceramic is achieved by adjustment of annealing temperature. Annealed at 600–650 °C, round tridymite crystal can be obtained using Cr_2O_3 as the nucleating agent, whereas flowerlike tridymite crystal would be formed after annealing at the temperatures of 700–800 °C. With XRD, SEM and EDS analyses, a crystallization model is used to explain the morphological feature change. Because of more SiO_2 keeping as amorphous phase, the breakdown strength of the ferroelectric glass-ceramic with round morphological feature of tridymite crystal can be increased. The results have indicated that the annealing processes have a great impact on the composition of the glass-ceramic hosting tridymite crystal and morphological feature of tridymite.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

The morphological feature of the crystal is a reflection of the whole processes of the crystal growth [1]. Tridymite crystal exhibits overall morphologies with a six-fold symmetry [2]. A transition from platelike to round structure is observed with the increase of undercooling, the platelike morphology is replaced by spherical crystals. Much effort has been devoted to the understanding of the dynamics of both pattern formation and selection in these systems [3]. A glass-ceramic material is usually comprised of one or several crystalline phases distributed in a glassy matrix. The crystalline phases and glass matrix each may have its own contribution to the ferroelectric properties of the glass-ceramics as a whole [4]. More SiO_2 kept as amorphous phase rather than tridymite crystal can increase the breakdown strength of glass-ceramic. At present, synergistically, the dielectric properties and structure observations for glass-ceramics still remain challenging.

In this work, morphological control of tridymite in $SrO–BaO–Nb_2O_5–CaO–SiO_2–B_2O_3$ glass-ceramic was achieved by adjustment of annealing temperature, in order to obtain the material with high breakdown strength. We focus on the effect of annealing temperature on the morphological feature of the tridymite crystal, qualitative study the changes in the morphology of the tridymite as a function of the habit of morphological feature and its growing environmental conditions.

2. Experimental procedure

$2SrO–12BaO–24Nb_2O_5–15CaO–14SiO_2–23B_2O_3–3Cr_2O_3$ (mol%) glass was prepared by melting $SrCO_3$, $BaCO_3$, Nb_2O_5, $CaCO_3$, SiO_2, H_3BO_3 and Cr_2O_3 in a quartz crucible and heating them at 1350 °C for 30 min. Quickly poured onto a thick copper plate and immediately quenched by pressing with another similar copper block, bulk glass with about 1 cm thickness was successfully obtained. Subsequently the glass samples were annealed at 600 °C, 650 °C, 700 °C, 750 °C and 800 °C for 10 h to remove residual stresses and were referred to as G0, G1, G2, G3 and G4, respectively. All samples with 0.1–0.2 mm were cut by a cutting machine. Glass-ceramics were examined by an X-ray diffractometer (D-MAX 2200 pc, Rigaku Co, Tokyo, Japan) at room temperature to investigate the phase evolution. The microstructure observation of the crystallized samples was performed using a scanning electron microscope (SEM; Model: JSM-5610LV, JEOL) with associated energy dispersive spectroscopy (EDS) (EDS-Energy Dispersive Spectroscopy JSM-6390A). For electrical measurements, these samples were polished to achieve parallel, smooth faces, and silver electrodes were sputtered on both faces. The polarization–electric field ($P–E$) ferroelectric hysteresis loops were measured using a ferroelectric tester (TF Analyzer 2000, aixACCT, Aachen, Germany). The

* Corresponding author. Tel.: +86 15102956142; fax: +86 2986168133.
E-mail addresses: liuxiaoyanood@163.com (X. Liu), 273835828@qq.com (Y. Pu).

http://dx.doi.org/10.1016/j.matlet.2014.04.104
0167-577X/© 2014 Elsevier B.V. All rights reserved.

specimens were used to measure the dielectric breakdown strength with a high-voltage source (Model 610E) using a voltage ramp rate of about 1 kV/s at room temperature until dielectric breakdown. At least 10 specimens were immersed in silicon oil to prevent flashover and corona discharge. The measurements of dielectric constant for glass-ceramics were performed using a precision multifunction LCR meter (E4980A, Agilent Tech, CA, U.S.).

3. Results and discussion

Fig. 1 shows the XRD patterns of the glass-ceramic samples annealed at temperatures of 600–800 °C. The intense diffraction peaks can be ascribed to the crystalline $(Sr_{0.5}Ba_{0.5})$ Nb_2O_6 phase and tridymite crystal. G0 exhibits the lowest intensity of quartz phase. The tridymite crystallization increases with the increase in annealing temperature (from G0 to G4). In short, the XRD results have indicated that the content of the tridymite crystal is significantly affected by annealing temperature.

The SEM images of glass-ceramics are shown in Fig. 2. The morphological features of the tridymite crystal and the surface layers of the whole glass-ceramics can be identified easily. The flowerlike morphology is found at temperatures of 700–800 °C, as shown in Fig. 2(a)–(c). With the decrease of annealing temperature (650 °C), the flowerlike morphology disappears and round morphology forms at a very high undercooling (Fig. 2(d)). Similar morphology also forms in the glass sample annealed at 600 °C. Similarly, both the flowerlike and round crystals in the glass-ceramics are homogeneous and uniform in shape and size.

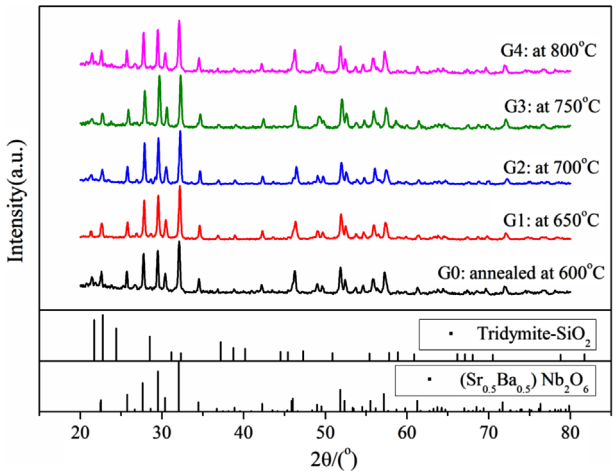

Fig. 1. XRD patterns of the glass-ceramics annealed at different temperatures.

Glasses annealed at 700 °C with flowerlike crystal and at 600 °C with round crystal are selected to perform EDS analysis (Fig. 3). In Fig. 3(A1) and (B1), EDS spectra from the center of the flowerlike crystal and round crystal are presented, respectively. Clearly it contains primarily Si, O elements with a little amount of Ca, Ba elements. This implies that the flowerlike and round grains are mainly composed of SiO_2 phase. In addition, Nb element is also detected. EDS (Fig. 3(A2) and (B2)) analysis shows the primary incorporation of Nb, Sr, Ba and O in the surface layer. Therefore, both the round and flowerlike crystals are tridymite crystal.

Crystal growth has two stages: the first nuclei formation at a relatively low temperature and the second crystals growth at a higher temperature by diffusion. Consequently, higher temperatures are required to maximize the growth rate [5,6]. In short, the temperature is a driving force for atomic movement. From 700 °C to 600 °C, annealing treatment gives crystalline with round structure. Because of lower speed of atomic movement, crystal growth is limited.

The polarization–electric field (P–E) hysteresis loops of the glass-ceramics are plotted in Fig. 4. The breakdown strength of the samples is deteriorated with the increase of annealing temperature, which is attributed to the increase of tridymite crystals remarkably. The sample G4 with flowerlike tridymite crystal annealed at 800 °C has an average breakdown strength of 429 kV/cm, and the breakdown strength of G3 and G2 are 451 kV/cm and 516 kV/cm respectively. Among all the measured samples, the sample G0 annealed at 600 °C shows the highest average breakdown strength of 687 kV/cm, which was 1.6 times higher than that of G4. Meanwhile, the intensity of polarization does not increase significantly, which is consistent with the relative dielectric constants (Fig. 4). All in all, the breakdown strength of the ferroelectric glass-ceramics with round morphological feature of tridymite which annealed at 600 °C and 650 °C is bigger than the others. So the breakdown strength of samples is indeed enhanced by adjusting the annealing temperature.

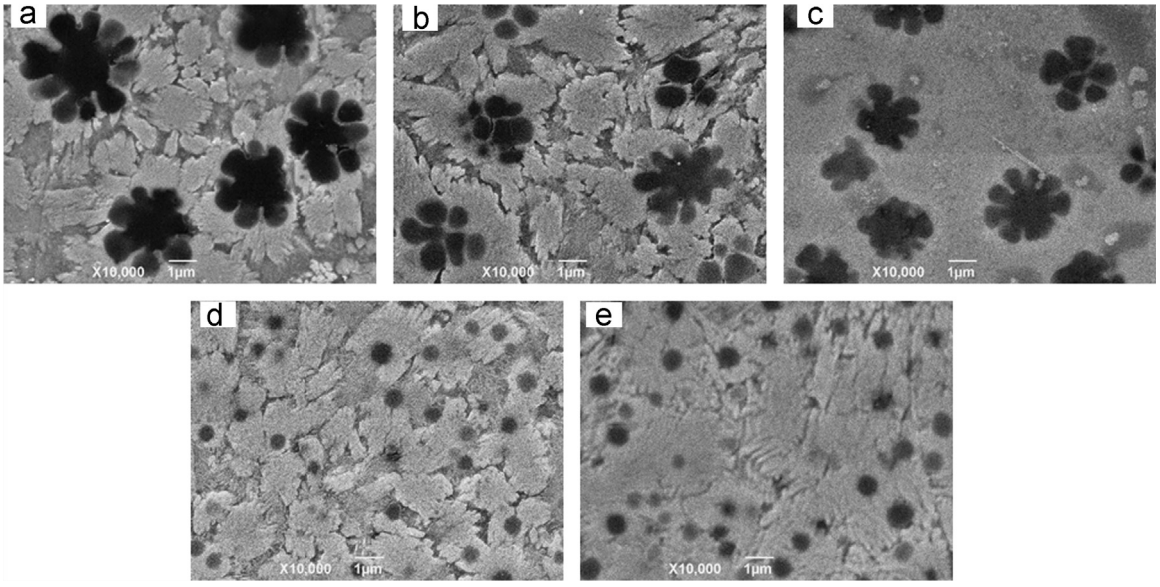

Fig. 2. SEM micrographs of glass-ceramics annealed at different temperatures: (a) 800 °C, (b) 750 °C, (c) 700 °C, (d) 650 °C, and (e) 600 °C.

Download English Version:

https://daneshyari.com/en/article/8020470

Download Persian Version:

https://daneshyari.com/article/8020470

Daneshyari.com