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Preparation and dielectric properties of ceramics in the $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ solid solution system

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Abstract

Ceramics in the $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ solid solution system were prepared by a solid state reaction process, and the dielectric characteristics were determined together with the microstructures. An increased dielectric constant and improved temperature coefficient of permittivity were achieved in this solid solution system, while the dielectric loss remained at the same level as that of $SrBi_2Ta_2O_9$.

Keywords: Dielectric properties; $SrBi_2(Ta,Nb)_2O_9$; Solid state reaction; Sintering

1. Introduction

Because of its low fatigue rate and leakage current, $SrBi_2Ta_2O_9$ has recently received much attention as ferroelectric thin films for nonvolatile random access memories (NVRAM),[1–3] and related investigations have been carried out on the $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ solid solution system.[4,5] In order to discuss the potential application of such ferroelectrics in bulk ceramic form, Yang and Chen[6] investigated the preparation and dielectric characteristics of $SrBi_2Ta_2O_9$. In the present work, ceramics in the $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ solid solution system are prepared and characterized together with the microstructures characterization.

2. Experimental procedure

The starting materials were reagent-grade $SrCO_3$, Bi_2O_3, Ta_2O_5 and Nb_2O_5. They were mixed in the appropriate molar ratios and ground for 24 h in ethanol using a ball mill, then dried and calcined at 850°C in air for 3 h. The calcined powders were re-ground for one day and dried. Then, 5 wt% organic binder was added to the mixed powder in an agate mortar to form a feed which was pressed into disks of 12.5 mm in diameter and 2–4 mm in thickness under a pressure of 98 MPa. The disks were sintered at 900–1150°C in air for 3 h. The density was measured by the dimensional method.

Microstructures of samples prepared in this way were characterized by scanning electron microscope (Hitachi S-570); X-ray diffraction using CuK_α radiation was carried out for phase identification.

Measurements of the dielectric constant and dielectric loss were conducted from 1 KHz to 1 MHz at room temperature by an HP4284A LCR meter. The temperature coefficient of capacitance τ_c was measured at 10 KHz using another LCR meter (WK4210) equipped with a thermostat in the range from room temperature to 85°C. Silver paste was used for the electrode.

3. Results and discussion

As shown in Fig. 1, the fine microstructures and nearly full densification of $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ ceramics can be achieved at suitable sintering temperatures.

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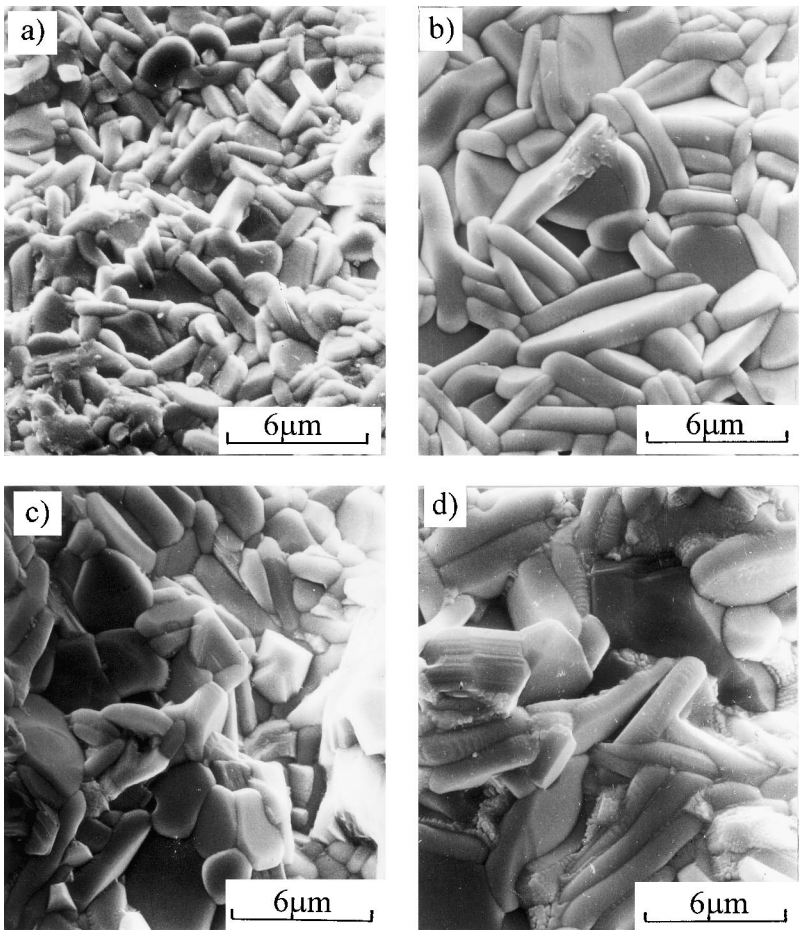

Fig. 1. SEM micrographs of $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ ceramics: (a) $x=0.2$, sintered at 950°C in air for 3 h; (b) $x=0.6$, sintered at 1050°C in air for 3 h; (c) $x=0.8$, sintered at 1050°C in air for 3 h; (d) $x=1.0$, sintered at 1100°C in air for 3 h.

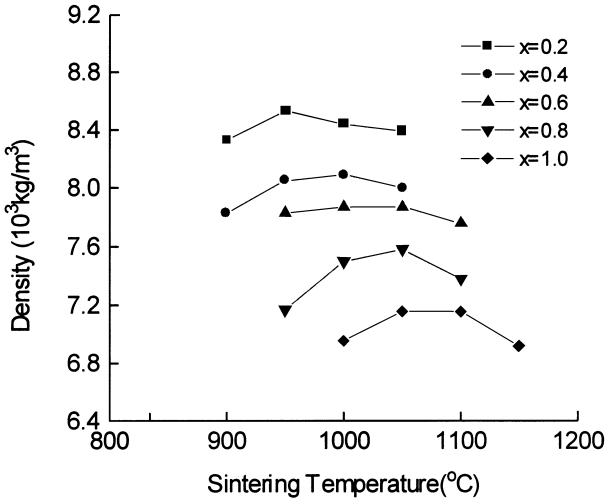

Fig. 2. Bulk density of $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ ceramics as functions of composition and sintering temperature.

The relationship between the bulk densities of the present ceramics with various compositions and the sintering temperature is given in Fig. 2, which indicates that the optimum sintering temperature increases with increasing x.

As shown in Fig. 3, the XRD pattern varies little with the composition in the present system ($x=0.2$–0.8). That is, the main peaks remain, the position and relative intensity varies slightly with the composition. These results confirm that $SrBi_2Nb_2O_9$ tends to form a continuous solid solution in $SrBi_2Ta_2O_9$ [4]. However, the XRD pattern for the $SrBi_2Nb_2O_9$ end member (Fig. 4) differs from the above situation.

Fig. 5 shows the room-temperature dielectric constant (at 10 kHz) of the present ceramics as a function of the composition and sintering temperature. Each composition has its own maximum dielectric constant corresponding to the optimum densification temperature. The room-temperature dielectric properties for the present ceramics sintered at the optimum densification temperatures are listed in Table 1 as a function of composition. With increasing x, the dielectric constant increases from 192.3 to 266.6, and the temperature coefficient of permittivity falls from $+5574$ to $+1850$ ppm/°C, while the dielectric loss remains at the same level, 0.025–0.028. The pronounced improvement in the temperature coefficient of permittivity is an advantage for application of these ceramics.

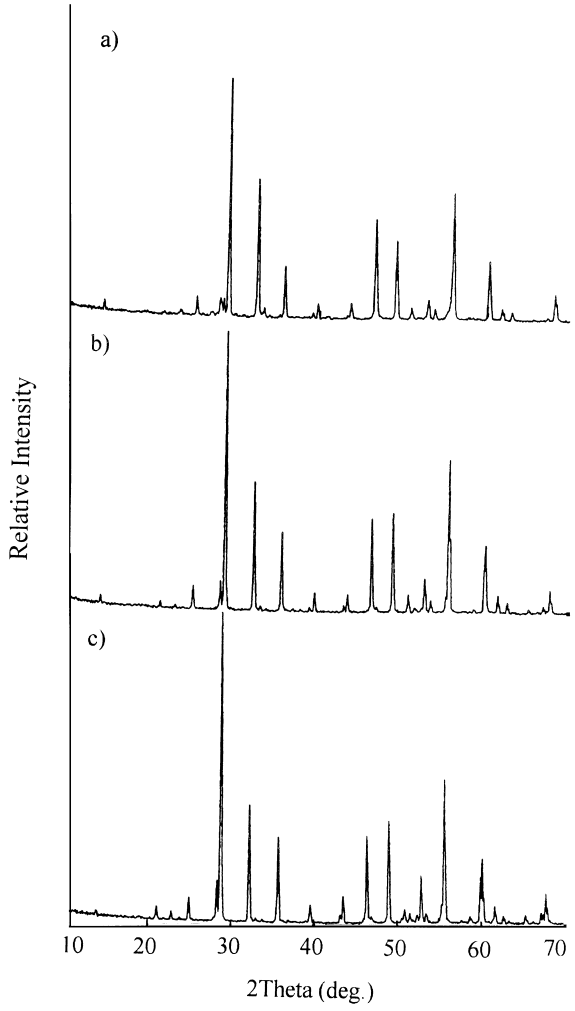

Fig. 3. XRD patterns of $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ ceramics: (a) $x=0.2$, sintered at 950°C in air for 3 h; (b) $x=0.4$, sintered at 1000°C in air for 3 h; (c) $x=0.8$, sintered at 1050°C in air for 3 h.

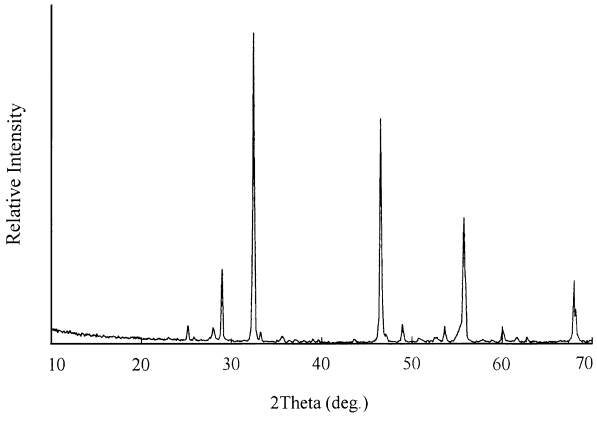

Fig. 4. XRD pattern of $SrBi_2Nb_2O_9$ ceramics sintered at 1100°C in air for 3 h.

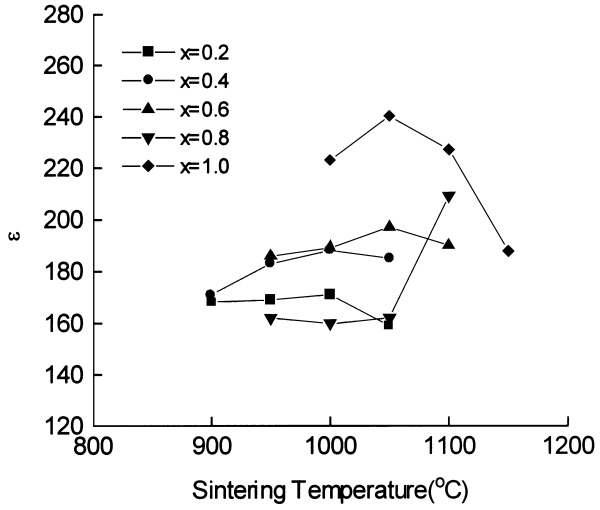

Fig. 5. Room-temperature dielectric constant (at 10 kHz) of $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ ceramics vs composition and sintering temperature.

Table 1
Room-temperature dielectric properties of $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ ceramics (at 10 kHz)

Composition	ε	tan δ	τ_ε [a] (pp m/°C)
$x=0.2$	169	0.0250	5574
$x=0.4$	188	0.0250	3984
$x=0.6$	197	0.0270	3376
$x=0.8$	209	0.0280	2514
$x=1.0$	240	0.0270	1850

[a] $\tau_\varepsilon = (\varepsilon_{80°C} - \varepsilon_{20°C})/(\varepsilon_{20°C}\Delta T)$.

4. Conclusions

Dense ceramics in the $SrBi_2(Ta_{1-x}Nb_x)_2O_9$ system were prepared by solid state reaction, and the continuous solid solution structure was confirmed. The dielectric constant of the $SrBi_2Ta_2O_9$ ceramic was increased by forming solid solution with $SrBi_2Nb_2O_9$, and the temperature coefficient of permittivity was significantly improved, while the dielectric loss remained at the same level. These solid solution dielectrics with increased dielectric constant and improved temperature coefficient are expected to receive more scientific attention than the respective end-members.

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References

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