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# Dielectric Properties of $\text{Ba}(\text{Mg}_{0.2/3}\text{Zn}_{0.8/3}\text{Nb}_{2/3})\text{O}_3$ and $\text{Ba}_{1-x}\text{Sr}_x(\text{Mg}_{0.2/3}\text{Zn}_{0.8/3}\text{Nb}_{2/3})\text{O}_3$ Microwave Ceramics

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**Keywords:** Microwave ceramics, Dielectric properties, Perovskite

**Abstract.** Microwave ceramics of  $\text{Ba}(\text{Mg}_{0.2/3}\text{Zn}_{0.8/3}\text{Nb}_{2/3})\text{O}_3$  and  $\text{Ba}_{1-x}\text{Sr}_x(\text{Mg}_{0.2/3}\text{Zn}_{0.8/3}\text{Nb}_{2/3})\text{O}_3$  were synthesized with conventional solid-state reaction method. Dielectric properties of the samples were studied as functions of compositions and sintering temperatures. Experimental results show that a higher  $Q \times f$  value is reached by substituting Zn ions with Mg ions and a near-zero temperature coefficient of resonant frequency is obtained by replacing Ba ions with Sr ions.

## Introduction

Microwave components have been rapidly developed for the application of dielectric resonators in wireless technologies. As a kind of material for resonator it is expected to behave high relative dielectric constant ( $\epsilon_r$ ), high quality factor ( $Q$ ) and low temperature coefficient of resonant frequency ( $\tau_f$ ) [1]. The perovskite oxides of  $\text{A}(\text{B}'_{1/3}\text{B}''_{2/3})\text{O}_3$ , here  $\text{A} = \text{Ba}$  or  $\text{Sr}$ ,  $\text{B}' = \text{Zn}$  or  $\text{Mg}$ ,  $\text{B}'' = \text{Nb}$  or  $\text{Ta}$ , display excellent dielectric properties including very low dielectric loss at high frequencies. Therefore, these kinds of oxides are of great importance for microwave application [2].  $\text{Ba}(\text{Zn}_{1/3}\text{Ta}_{2/3})\text{O}_3$  is a typical compound of them with perfect quality factor  $Q$  [3,4]. However, practical application is limited due to the expensive reagent of  $\text{Ta}_2\text{O}_5$  and high sintering temperature for ceramics. There has been considerable interest in the development of  $\text{Ba}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3$  system, i.e., BZN, with the advantage of cheap  $\text{Nb}_2\text{O}_5$  [1,5]. In this work, effect of A-site and B-site substitutions on the dielectric properties of BZN system was focused on, including ceramics of  $\text{Ba}(\text{Mg}_{0.2/3}\text{Zn}_{0.8/3}\text{Nb}_{2/3})\text{O}_3$ , i.e., BMZN, and,  $\text{Ba}_{1-x}\text{Sr}_x(\text{Mg}_{0.2/3}\text{Zn}_{0.8/3}\text{Nb}_{2/3})\text{O}_3$ , i.e., BSMZN.

## Experimental Procedures

Reagents of  $\text{BaCO}_3$ ,  $\text{SrCO}_3$ ,  $\text{MgO}$ ,  $\text{ZnO}$  and  $\text{Nb}_2\text{O}_5$  with high purity were weighed and ball-milled for 2 h. The dried powder was calcined at  $1200^\circ\text{C}$  for 2 h. After re-milling, the powder was dried and pressed into pellets with 5wt% polyvinyl alcohol (PVA) solution at 200 MPa. The green bodies were sintered at temperatures ranging from  $1280^\circ$  to  $1340^\circ\text{C}$  for 4 h in air. The dielectric properties of ceramics were measured at microwave frequencies by the Hakki-Coleman dielectric resonator method [6]. The temperature coefficients of resonant frequency for the samples were measured at micro-

wave frequencies from ambient to  $80^\circ\text{C}$ . The microstructure of the ceramics was observed by scanning electron microscopy (SEM). The crystalline phases of the sintered samples were identified by X-ray diffraction (XRD).

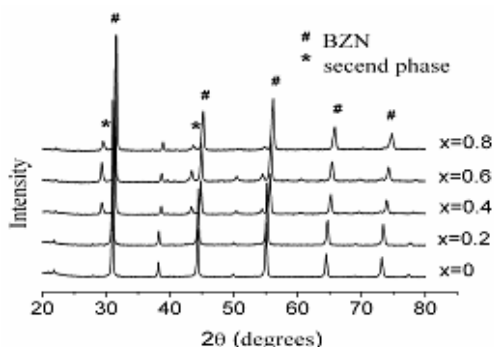


Fig. 1 XRD patterns of BSMZN ceramics with various  $x$  values.

## Results and Discussion

XRD patterns of BSMZN ceramics with  $x = 0 - 0.8$  sintered at  $1320^\circ\text{C}$  for 4 h are shown in Fig.1. It can be seen that major phase of the ceramics is BZN, meanwhile a few of weak peaks are also identified as second phase when  $x \geq 0.4$ . And there is a shift of peaks towards higher angle with the increasing of  $x$  value. This may be due to the sub-

Table 1 Lattice parameter for BSMZN ceramics

Composition x	0	0.2	0.4	0.6	0.8
$d_{110}$ (Å)	2.873	2.869	2.851	2.842	2.833

stitution of Ba ions with Sr ions. Ionic radius of  $Ba^{2+}$  is 0.142 nm and that of  $Sr^{2+}$  is 0.126 nm. Smaller size of  $Sr^{2+}$  leads to a decrease of lattice parameter. Therefore, d values become smaller with increasing of Sr ion concentration. For example, the values of  $d_{110}$  for the samples are listed in Table 1. According to the Bragg equation,

$$\lambda = 2d\sin\theta \quad (1)$$

the decrease of d value results in the increase of  $2\theta$  value. Thus the peaks shift to higher angles.

The plot of bulk densities for the samples versus different sintering temperature is illustrated in Fig.2. It is obvious that the densities of ceramics decreased with increasing of x value, which is attributed to the smaller relative atomic weight of Sr (87.62) than that of Ba (137.33).

In addition, the densities of the ceramics with  $x = 0$  change slightly when the sintering temperatures are above 1300 °C. However, the maximal value of density for each composition appears at higher temperature with x values ranging from 0.2 to 0.8. This may mean that the substitution of Ba ions with Sr ions leads to the increasing of sintering temperature for the samples.

The SEM micrographs of BSMZN ceramics with various x values are demonstrated in Fig.3. The grain sizes of the samples are around 1  $\mu m$ . For ceramics with  $x = 0$  microstructure is more homogeneous than that of other samples. And the grain sizes of samples with Sr ions substitution for Ba ions become smaller with the increasing of x values.

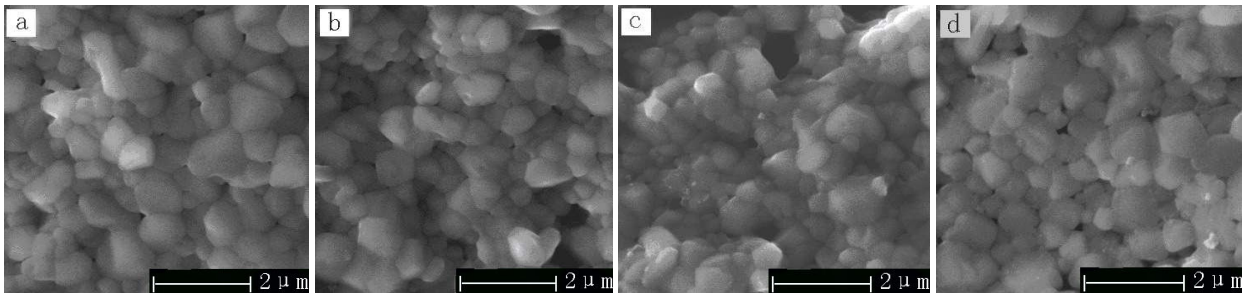


Fig. 3 SEM micrographs of BSMZN ceramics sintered at 1320 °C for 4 h with (a)  $x = 0$ , (b)  $x = 0.2$ , (c)  $x = 0.4$  and (d)  $x = 0.6$ , respectively.

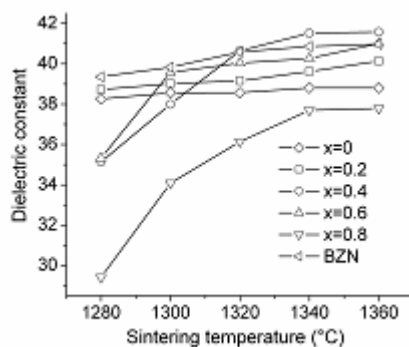


Fig. 4 Dielectric constants of samples as a function of sintering temperature.

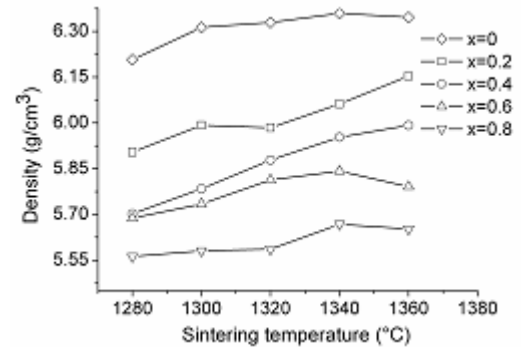


Fig. 2 Densities of samples as a function of sintering temperature.

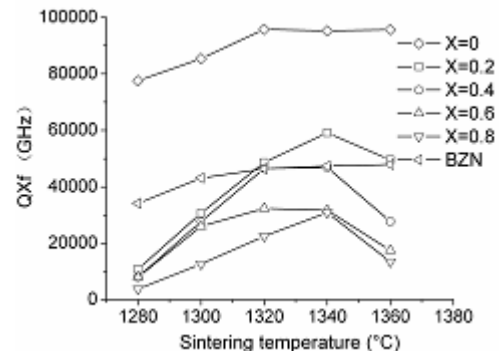


Fig. 5  $Q \times f$  values of samples as a function of sintering temperature.

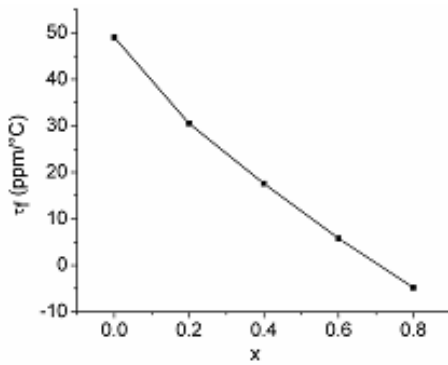


Fig. 6 Temperature coefficient of resonant frequency as a function of x value for BSMZN ceramics.

The dielectric constants ( $\epsilon_r$ ) of the BSMZN and BZN ceramics as a function of sintering temperature (Fig.4) show that the dielectric constants of the compositions with  $x = 0, 0.2$  and the samples of BZN are not significantly affected by sintering temperature. However, the samples with  $x = 0.4, 0.6$  and  $0.8$  can not be sintered densely when sintering temperature is below  $1320^\circ\text{C}$ , which results in lower  $\epsilon_r$  values for the samples sintered at  $1280^\circ\text{C}$  and  $1300^\circ\text{C}$ .

The x value for the ceramics has a similar effect on the value of  $Q \times f$  like that of dielectric constant. Figure 5 demonstrates the  $Q \times f$  values of the samples with different x values as a function of sintering temperature. The  $Q \times f$  values increase dramatically with the substitution of Mg ions for Zn ions. Meanwhile, replacement of Ba ions with Sr ions leads

to gradual reducing of  $Q \times f$  values.

Temperature coefficient of resonant frequency for the ceramics is another important microwave property, which is shown in Fig.6 as a function of x value. It is obvious that the  $\tau_f$  values of the samples decrease linearly with increasing of x value. When x value ranges from 0 to 0.8,  $\tau_f$  value varies from 49 to  $-4.8$  ppm/°C. Therefore a near-zero  $\tau_f$  value can be obtained within  $x = 0.6 - 0.7$ .

## Conclusions

Based on the experimental results, dielectric constants of BMZN and BSMZN ceramics from 30 to 40 are obtained. The value of  $Q \times f$  for BZN ceramics can be effectively improved by substitution of Zn ions with Mg ions, with maximal value of about 90,000 GHz. The value of  $\tau_f$  for the samples can be adjusted suitably by replacing Ba ions with Sr ions, and a near-zero  $\tau_f$  value is obtained for  $x = 0.6 - 0.7$  with a slight decrease of  $Q \times f$  value.

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## **High-Performance Ceramics IV**

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