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Origin of High *Q* for Microwave Complex Perovskite

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Keywords: microwave dielectrics, quality factor, high Q, ordering, order-disorder transition, high symmetry, crystal structure, complex perovskite, $Ba(Zn_{1/3}Ta_{2/3})O_3$ (BZT), $Ba(Zn_{1/3}Nb_{2/3})O_3$ (BZN), spark plasma sintering (SPS).

ABSTRACT

Origins of high *Q* are considered on intrinsic as high symmetry, ordering structure and high density crystal structure. It was concluded that the high symmetry brings high *Q* instead of ordering comparing some cases as follows: As if ordering ratio of $Ba(Zn_{1/3}Ta_{2/3})O_3$ (BZT) is high of about 80%, *Q* values are distributed from low to high *Q*. Disordered BZT ceramics with high density obtained for short sintering time by spark plasma sintering (SPS) showed high *Q*. $Ba(Zn_{1/3}Nb_{2/3})O_3$ (BZN) with order-disorder transition showed high *Q* at disorder form sintered over the transition temperature. And, the disordered BZN with high *Q* annealed at lower temperature changed to order structure without improvement of *Q*.

INTRODUCTION

Recently, the usage of radio frequency (RF) for microwave communication is expanding to high frequency because of the shortage of frequency area, and request of high speed and high data transfer rate. As microwave dielectrics are expected to have high quality factor *Q* (high *Q*) based on this background, we will clarify the origin of high *Q* that is one of three important properties [1,2]. *Q* is inverse of dielectric losses tan , other properties are dielectric constant $_r$ and temperature coefficients of resonant frequency $_f$. The $_r$ is expected to be small for more high frequency millimeterwave region, because of reducing the delay time of electronic signal transition and improvement of accuracy for production. The $_f$ is expected to be near zero ppm/^{o}C for receiving the RF signals in all places in the world. The *Q* is affected by intrinsic factors such as crystal structure and by extrinsic factors such as grain growth, impurity, and so on.

In this paper, we focus on the quality factor *Q* in the three properties based on the crystal structure. Origin of the *Q* will be clarified based on the results presented in the previous papers up to now as follows: complex perovskite $Ba(Zn_{1/3}Ta_{2/3})O_3$ (BZT) [3-6], complex perovskite $Ba(Zn_{1/3}Nb_{2/3})O_3$ (BZN) [7].

EXPERIMENTAL

Synthesis methods of each compound are refer to previous papers following: BZT and BZN by solid state reaction (SSR) [4], BZT by spark plasma sintering (SPS) [6]. Most all these compounds are synthesized by SSR with high purity raw materials. BZT was synthesized also by SPS which sintered high density ceramics for short time about 5 min.

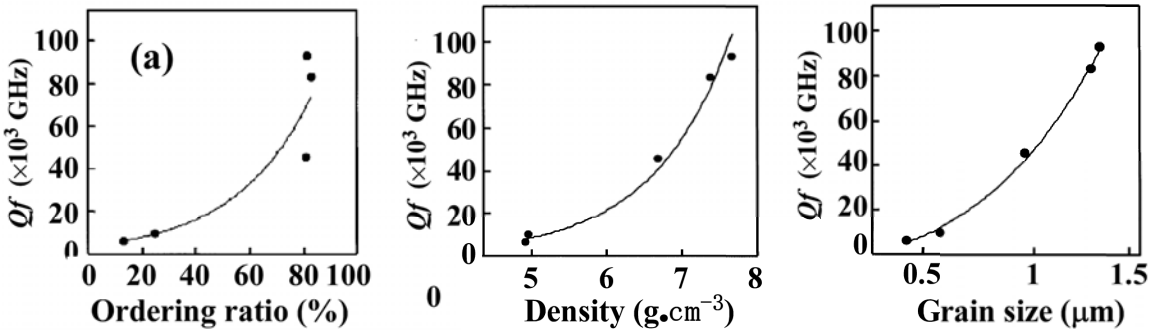

Fig. 1. *Qf* as a function of ordering ratio (a), density (b), and grain size (c) of BZT ceramics.

The characterizations of crystalline states also presented the previous papers. Identification of precipitated phases and the crystal structure analysis by Rietveld method (RIETAN-2000 program) [8] were performed by X-ray powder diffraction (XRPD). Densities of these compounds were measured by Archimedes method. Microwave dielectric properties were measured using Hakki and Colman method [9,10].

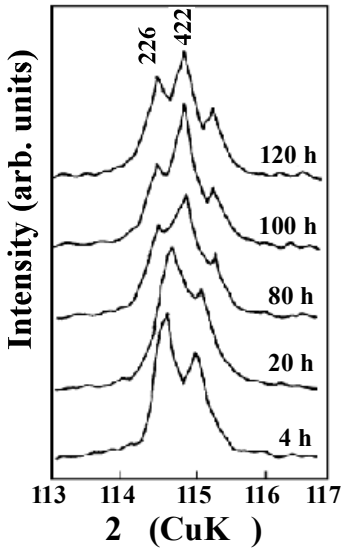

Fig. 2. XRPD patterns of BZT ceramics as a function of sintering hours at 1350°C.

RESULTS AND DISCUSSIONS

The ordering of atomic arrangements based on order-disorder transition has been believed to bring high quality factor *Q* on the RF resonance. We found some aspects for relationship between the ordering and *Q*, which disordering with high symmetry is prior to the ordering on the high *Q*. The first, we present about complex perovskite BZT prepared by solid state reaction (SSR). Fig. 1 presented by Koga *et al*. [3], shows *Qf* values as a function of ordering ratio, density, and grain size. The ordering ratios determined by Rietveld method on XRPD patterns are saturated about 80% as shown in Fig. 1a. The *Qf* values on the sample with the same ordering ratio are distributed from 40000 GHz to 100000 GHz. This result shows that as if showing high ordering ratio, does not always show high *Qf*. On the other hand, density (Fig. 1b) and grain size (Fig. 1c) show high *Qf* depending the values. Fig. 2 shows the XRPD patterns of BZT as a function of sintering time. The crystal structure changed from cubic with disordering to trigonal with ordering by long sintering more than 100 hours. This phenomenon presented by Kawashima *et al*. [11] shows that the ordering brings high *Q* as believe in generally.

Fig.3 shows the *Qf* as a function of densities on BZT fabricated by SSR and SPS. The samples obtained by SPS were disordered cubic type as shown in Fig. 4 with only 420 reflection, compared with the peak separation of 422 and 226 of ordered trigonal type sintered by SSR (1400°C 100 hours). The SPS samples with high densities were obtained by a very short sintering time 5 min. The short time sintering by SPS may bring the disordered BZT with high density. Regardless of the synthesis method, *Qf* strongly depended on density, and *Qf* values were improved with density as shown in Fig. 3. The highly crystallized dense BZT ceramics synthesized by SPS showed *Qf* (≒ 50000 GHz) similar to that of the ordered BZT sample with the same density (≒ 7.5 g/cm^3). The crystallization with densification of BZT ceramics should play a more important role in *Q* factor improvement in this BZT system than the structural ordering.

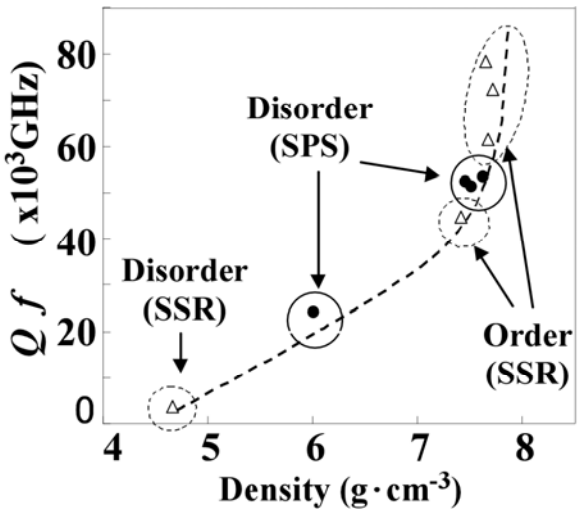

Fig.3. *Qf* as a function of density for BZT by solid state reaction (SSR) and spark plasma sintering (SPS). Order: ordered perovskite, Disorder: disordered perovskite.

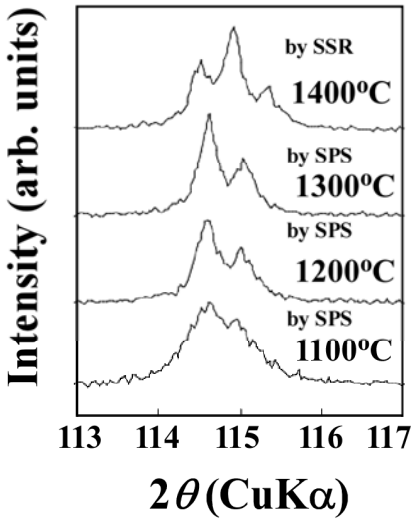

Fig. 4. XRPD patterns around 420 diffraction of BZT ceramics as a function of sintering temperature sintered by SPS for 5 min under 30 MPa.

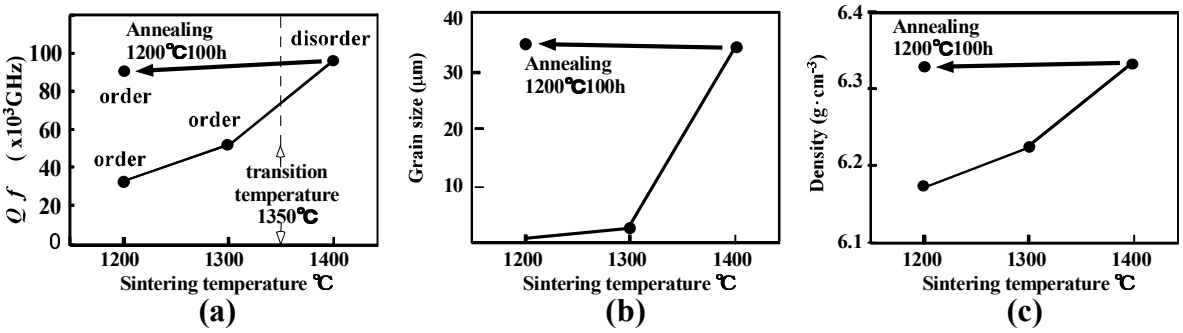

Fig. 5. *Qf* (a), grain size (b) and density (c) as a function of sintering temperature for BZN with transition temperature at 1350°C. And though the disorder one with high *Q* annealed at 1200°C 100 hours, the Qf was not improved.

Ordering based on the order-disorder transition brings low symmetry, and disordering brings high symmetry. Usually, high symmetry also brings high *Q* same as ordering. We will present an example that high symmetry brings high *Q* more than ordering. BZN shows clearly order-disorder transition temperature at 1350°C, though BMT and BZT is not clear the transition temperature. Fig. 5 shows *Qf* as a function of sintering temperature of BZN. Samples sintered at 1200 and 1300°C under the transition temperature show ordering and lower *Qf* than a sample with disordering sintered at 1400°C. Moreover, the disordered BZN annealed at 1200°C 100 hours transformed to order, and the *Qf* value was not improved. The grain size and densities also unchanged by the annealing as shown in Fig. 5(b) and (c). So, this case shows that high symmetry brings high *Q*.

CONCLUSION

In this paper, intrinsic origins of *Q* based on crystal structure are discussed. In complex perovskite BZT and BZN with order-disorder transition, high *Q* was brought by high symmetry instead of ordering based on following facts:

/ In the case of BZT synthesized by SSR, as if ordering ratio is high of about 80%, *Q* values does not always show high.
/ Disordered BZT with high density synthesized by SPS showed high *Q* same as ordered samples sintered by SSR.
/ In the case of BZN with order-disorder transition, disordered sample sintered at 1400°C higher than the transition temperature showed higher *Qf* value than ordered samples sintered at 1200 and 1300°C under the transition temperature.
/ Moreover, the disordered sample annealed at 1200°C 100 hours changed to ordered one, but the *Qf* value is not improved.

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