Quenching Effects for Electrical Properties on Lead-free $(Bi_{1/2}Na_{1/2})TiO_3$ and Related Solid Solution Ceramics

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Bismuth sodium titanate, $(Bi_{1/2}Na_{1/2})TiO_3$ (BNT), is a perovskite-structure ferroelectric ceramic with a rhombohedral symmetry (*R3c*) at room temperature (RT). BNT-based solid solutions have relatively high piezoelectric properties in lead-free piezoelectric materials and enable the easy preparation of dense ceramics, which is an advantage for manufacture; therefore, BNT-based solid solutions have attracted attention as lead-free piezoelectric ceramics. However, BNT has the problem of having a low depolarization temperature T_d of approximately 180 °C, resulting in a working temperature below T_d. In particular, the morphotropic phase boundaries (MPBs) of BNT-based solid solutions have a low T_d of approximately 100 °C, even though an MPB has excellent piezoelectric properties. That is, the piezoelectric constant and T_d show a trade-off relationship, so that the elevation of T_d without the deterioration of piezoelectric activities is strongly demanded for practical piezoelectric applications using BNT-based ceramics.

Recently, the effects of quenching on dielectric ageing behavior in relaxor ceramics such as PLZT and BNT-$BaTiO_3$ systems have been reported and they suggest that quenching could be a promising method to improve the dielectric ageing effect and other thermal behaviors in relaxor ferroelectrics. These results suggest that control of the cooling (quenching) process from a high temperature in preparing BNT ceramics could change their electrical properties including the depolarization temperature T_d. Therefore, in this study, pure BNT and BNT-based solid solution ceramics with MPB were quenched after sintering, and the effects of quenching on the electrical properties of BNT ceramics were investigated.

BNT and BNT-based solid solution ceramics were basically fabricated by using conventional ceramic fabrication process. After sintering at 1140 °C, these ceramics were cooled at a rate of -100 °C/h in the ordinary firing (OF) procedure, which is hereafter referred to as OF. In the case of rapid cooling, after sintering at 1140°C, the samples were removed out from the electric furnace set at a high temperature (600, 800, 900, 1000, or 1100 °C), exposed to room air and immediately cooled rapidly to RT using an air fan. The quenched samples were abbreviated to q(temp).

Figure 1 shows the T_d derived from the first peak of tanδ in the measurement of the temperature dependence of dielectric properties as a function of quench temperature for pure BNT ceramics. When the quenching temperature is below 800 °C, the T_d values are almost the same as those of OF-BNT ceramics. In contrast, a rapid increase in T_d is observed in the samples quenched above 800 °C. From the, T_d increased with increasing quench temperature. The T_d of a pure BNT sample quenched from 1100 °C was 223 °C, which was almost 50 °C higher than that prepared by the ordinary cooling process. From the measurement of *P-E* hysteresis loops, both the remanent polarization P_r and the coercive field E_c of BNT samples prepared by ordinary firing were almost the same as those quenched from 1100 °C. Additionally, from the measurements by a resonance-antiresonance method, the electromechanical coupling factor k_{33} of ordinarily fired BNT was 0.45, and that of the quenched BNT was 0.46. From these results, it is clarified that the quenching procedure is an effective way to increase the T_d of BNT ceramics without deteriorating ferroelectric and piezoelectric properties. In the presentation, we will focus on the quenching effects for electrical properties of not only pure BNT ceramic but also BNT-based solid solution ceramics with MPB composition.

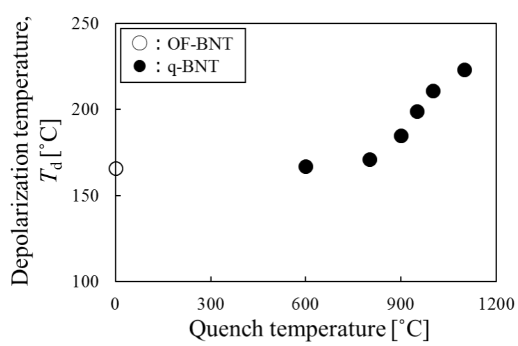

Fig. 1 Depolarization temperature T_d as a function of quenching temperature of OF- and q-BNT ceramics