Thermodynamic Studies of Relaxor Ferroelectrics $BaTi_{1-x}Zr_xO_3$
- Giauque Travel Award Presentation -

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Because of large dielectric, piezoelectric, and electromechanical properties, much attention has been paid to so-called relaxor ferroelectrics, which have large value of dielectric constant in a wide temperature range with a broad maximum, which shows frequency-dependent glass-like behavior. Unfortunately, the typical relaxors are mainly lead-based complex oxides with perovskite structure; $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN), and $Pb(Mg_{1/3}Ta_{2/3})O_3$ (PMT), and lead-free relaxors are highly desired in the view point of environmental. We have studied thermodynamic properties of PMN and PMT by heat capacity and dielectric measurements, and found a broad anomaly in the heat capacity curve, which should be attributed to formation of FNR (ferroelectric nano-region) [1]. Although the mechanism has been widely studied for such lead-based compounds, few studies have been published for lead-free compounds. However, it has been recently reported the broad heat capacity anomaly in $BaTi_{0.65}Zr_{0.35}O_3$ [2].

In the present study, barium titanate zirconate, $BaTi_{1-x}Zr_xO_3$ (BTZ, $0.15 < x < 0.90$) were prepared by a conventional solid state reaction method. The heat capacity measurements were carried out using an adiabatic calorimeter in the temperature range from 13 K to 420 K and a relaxational calorimeter between 2 K and 300 K. The dielectric constants were measured using an HP4284A LCR meter (HP Ltd.) from 1 kHz to 1 MHz in the temperature range from 20 K to 300 K.

A heat capacity anomaly was found at 330 K in the sample of $x = 0.15$, which should be attributed to a phase transition. With increasing the value of x, the anomaly decreased, and it vanished in the sample of $x = 0.35$, which showed typical relaxor behavior in the dielectric properties. Although the relaxor behaviors were observed in the samples of $x = 0.35$–0.70, the corresponding heat anomalies were undetectable. In the case of PMN, a broad anomaly was clearly observed in the heat capacity curve, and also typical relaxor behavior was observed in the dielectric properties [1]. It is considered that the relaxor mechanism of BTZ of $x = 0.35$–0.70 is different from that of PMN. Furthermore, in the sample of $x = 0.80$ the other dielectric dispersion was observed without remarkable heat capacity anomaly. The results of detailed studies will be presented.

References

[1] Y. Moriya, H. Kawaji, T. Tojo, and T. Atake, Phys. Rev. Lett., **90**, 205901 (2003)
[2] M. V. Gorev, V. S. Bondarev, I. N. Flerov, Ph. Sciau and J. –M. Savariault, Phys. Solid State, **47**, 2304 (2005)