Dielectric Properties of Amorphous PbTiO₃

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The dielectric constant of rapidly quenched amorphous PbTiO₃ was measured. As have already been reported,^{1,2)} the amorphous sample crystallizes when it is heated up to $500 \sim 550^{\circ}$ C, and by annealing it above this crystallization temperature ($T_{\rm crys}$), Raman spectra change continuously, owing to the structural relaxation toward the perfect crystalline state. The purpose of this report is to investigate the behavior of dielectric constant at $T_{\rm crys}$ and in this annealing process. A part of this study has already been reported in refs. 1 and 2.

Figure. 1 shows the 10 kHz result. The crystallization temperature of the specimen used was confirmed to be 530°C by DTA. Curve 1 is the result of the first heating run. Dielectric constant shows increase on heating. Just below $T_{\rm crys}$, a shoulder is seen. At $T_{\rm crys}$, it shows a jump and afterward further increase is seen. These two anomalies in the first heating process depend on the frequency. As the frequency becomes higher, the temperature of the shoulder shifts to a higher temperature and jump height at $T_{\rm crvs}$ decreases. The shoulder may be owing to a relaxation of ionic motion and the structural softening as a precursor of crystallization. High value in dielectric constant after crystallization is likely owing to the high ionic conductivity in the crystalline phase.

Curve 2 is the second heating run: after the first heating up to the maximum arrival temperature 540°C the sample was cooled down to the room temperature. Broad maximum appears around 400°C. Curve 3, 4 and 5 are third, fourth and fifth heating runs which follow the cooling from 600, 700 and 850°C to room temperature, respectively. The maximum becomes less broade and shifts to a higher temperature. The peak temperature in curve 5 is about 480°C which almost agrees with the crystalline Curie point.



Fig. 1. Dielectric constants and Raman spectra as functions of maximum arrival temperature.

Thus it is confirmed that the ferroelectric anomaly in dielectric constant develops by suitable annealing. This is in accord with the result of Raman spectroscopy. In Fig. 1 the Raman spectra from same samples used in dielectric measurements are also shown on right. The soft mode which does not exist in the amorphous state appears after crystallization and it show hardening with increasing the maximum arrival temperature. The shifts of dielectric constant peak above observed do not contradict the hardening of the soft mode. These seem to reflect the development of the ferroelectric ordering owing to the strutural relaxation from amorphous state toward the perfect crystalline one.

References

- M. Takashige, T. Nakamura, H. Ozawa, R. Uno, N. Tsuya and K. I. Arai: Jpn. J. Appl. Phys. 19 (1980) L225, L555.
- 2) T. Nakamura and M. Takashige: this issue.