Dielectric behavior of poled complex perovskite relaxor ferroelectrics

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RELAXOR ferroelectric ceramics with complex perovskite structure is considered as the first material chosen for multilayer capacitors (MLC) in technology and economy, because of their high permittivity, lower sintering temperature and lower capacitors changing rate with temperature (temperature coefficient) for diffuse phase transition (DPT). Thus, preparations and properties about the materials received more attention. There are many reports about their dielectric properties[1, 2] but there are few about their poled dielectric behavior[3]. In the present work, the dielectric behavior was carefully measured for poled and unpoled complex perovskite

relaxor ferroelectric ceramics PZN-PT-BT(80/10/10). We compare the results with their pyroelectric behavior under the same poling condition, and give the character of microdomain-macrodomain transition for poled relaxor ferroelectrics.

1 Experimental

1.1 Sample preparation

The PZN-PT-BT(80/10/10) relaxor ferroelectric ceramics was prepared with a conventional mixed oxide method[4], and sintering was performed at 1 100℃ for 2 h. The PZN-based relaxor ferroelectric ceramics powder was characterized by powder X-ray diffraction (XRD) patterns using a diffractometer. The result shows that the sample has a pure perovskite phase. Sample was polished carefully and measured with the thickness $d = 1.220$ mm and the diameter $D = 9.095$ mm. Then it was electroded with silver paint in the faces, and fired at 550℃ for 5 min.

1.2 Coercive field E_c measurements

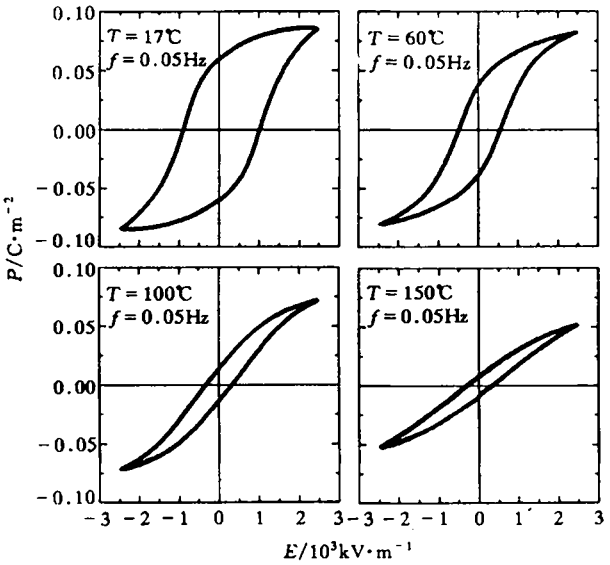

Fig. 1. Hysteresis of PZN-PT-BT(80/10/10) at several temperatures.

Using the automatic Sawyer-Tower circuit hysteresis loop measuring system, we m easured the hysteresis of the sample in the temperature range from 20 to 150℃. From the hysteresis loop, we can get the coercive field E_c and the remanent polarization P_r. Fig. 1 gives the hysteresis of the sample at different temperatures. Figs. 2 and 3 give E_c and P_r versus temperatures of the PZN-BT-PT (80/10/10) sample, respectively.

1.3 Dielectric properties measurements

The sample was placed in a test chamber, which could be operated between −196 and 300℃. The temperature was measured using a Hewlett-

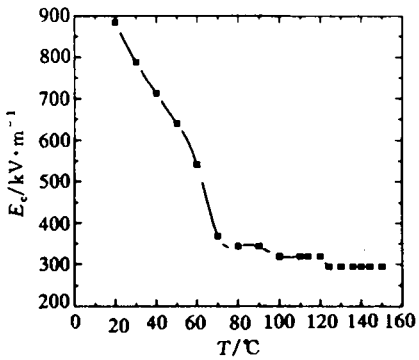

Fig. 2

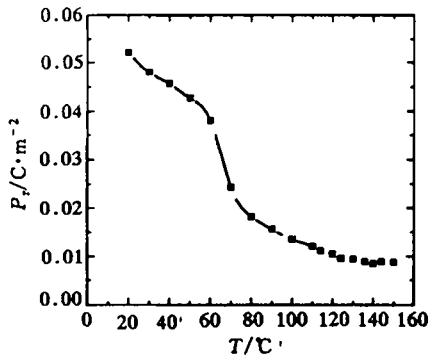

Fig. 3

Packard 3455A digital voltage meter via a platinum resistance thermometer mounted directly on the ground electrode of the sample fixture. Based on the above measurement results, the sample was heated to 200℃, in an applied DC field of 20 kV/cm for 30 min, and then quenched to −100℃ and shorted 15 min to release the surface charge. The dielectric response was measured as a function of frequency between −100 and 180℃ at a heating rate of 2℃/min, using a Hewlett-Packard 4274A LCR meter which could cover a frequency range from 100 to 100 kHz.

The poled sample was heated to 300℃ and stored for 60 min to depole, and then directly quenched to −100℃. The dielectric response was measured again as a function of frequency between −100 and 180℃ at the same heating rate of 2℃/min. Fig.4(a) gives the dielectric response results for the unpoled sample. Fig.4(b) gives the dielectric response dependence on temperature under poling condition.

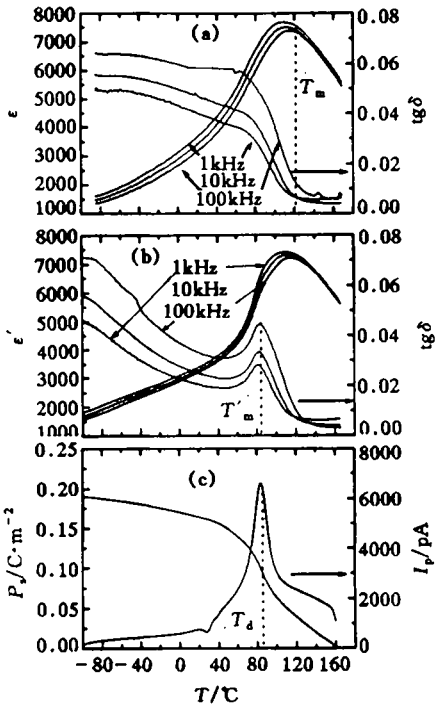

Fig. 4. Dielectric spectroscopy of unpoled sample (a), poled sample (b) and the pyroelectric spectroscopy of sample (c).

1.4 Pyroelectric properties measurements

According to Roundy-Byer pyroelectric measurement method[7, 8], we poled the sample as above, heated it from −100 to 180℃ at a rate of 2℃/min, and got the pyroelectric current using a Hewlett-Packard 4140B PA meter. Fig.4(c) shows the pyroelectric current and the calculated saturation polarization P_s.

2 Results and discussion

Comparing fig. 4(a), (b) and (c), we found that the temperature of dielectric loss angle tangent maximum (T'_m) does not change with frequency any more, which coincides with the temperature of pyroelectric current maximum (T_d) under the same poled condition, and the frequency dependence of dielectric permittivity becomes weak in the $T < T'_m$ region.

Based on the microdomain-macrodomain transition model[5, 6], when the poled sample was cooled under the DC field (field cooling), in microscopy the microdomains which caused the diffused phase transition in relaxor ferroelectrics turned in the DC field direction. Most of them coupled and transformed into macrodomains showing the normal ferroelectrics characteristics. Thus, when the heating measurement process was under zero DC bias (zero field heating), the frequency dependence of dielectric permittivity became weak in DPT region. Up to T'_m the thermal fluctuation was able to make the macrodomains transform into microdomains. In small-signal dielectric measurement, this transition shows a large sharply-dielectric loss angle tangent peak, and the temperature of dielectric loss angle tangent peak (T'_m) does not change with frequency because of the sudden change of the microscopy structure transition. In the pyroelectric measurement process, the pyroelectric current peak for normal ferroelectrics

shows the transition from ferroelectric phase to paraelectric phase, but the peak for relaxor ferroelectrics shows the microscopy structure transition from macrodomain to microdomain because there is no clear phase transition near ferroelectric-paraelectric phase transition temperature in relaxor ferroelectrics. This is the reason why T_d does not coincide with T_m but with T'_m and the peak is not so sharp as that for normal ferroelectrics. In ref. [7], more clear explanations and analyses about the results were given.

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References

1 Bell, A. J., Calculation of dielectric properties from the superparaelectric model of relaxors, *J. Phys. Condens. Matter*, 1993, 5(46): 8773.
2 Choi, S. W., Shrout, T. R., Jang, S. J. *et al.*, Dielectric and pyroelectric properties in the PMN-PT system, *Ferroelectrics*, 1989, 100(1): 29.
3 Yao, X., Chen, Z. L., Cross, L. E., Polarization and depolarization behavior of hot pressed PLZT ceramics, *J. Appl. Phys.*, 1983, 54(6): 3399.
4 Halliyal, C., Kumar, U., Newnham, R. E. *et al.*, Dielectric and ferroelectric properties in the PZN-PT-BT system, *J. Am. Ceramic Soc.*, 1987, 70(2): 119.
5 Yao, X., Chen, Z. L., Relaxor ferroelectrics, *Piezoelectrics and Acoustooptics* (in Chinese), 1984, 5(6): 1.
6 Cross, L. E., Relaxor ferroelectrics, *Ferroelectrics*, 1987, 76(1): 241.
7 Fan, H. Q., Xu, Z., Zhang, L. Y. *et al.*, A study on the dielectric behavior of poled relaxor ferroelectrics, in *Proceedings of the 9th International Symposium on Electrets* (in Chinese), Shanghai, 1996, 1019—1023.

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