International Symposium of Research Students on Materials Science and Engineering
December 20-22, 2004, Chennai, India
Department of Metallurgical and Materials Engineering, Indian Institute of Technology Madras

DOMAIN ALIGNMENT AND MICROCRACKING AS SOURCES OF ACOUSTIC EMISSION IN PZT CERAMICS DURING PHASE TRANSITION

K. Prabakar and S.P. Mallikarjun Rao
Acoustic Emission Lab., Physics Department, University College of Science, Osmania University, Hyderabad – 500 007, India

ABSTRACT

Acoustic emission (AE) has been monitored during the Phase Transition (PT) in soft and hard PZT ceramics. Both poled and unpoled samples were experimented between 30ºC to about 350ºC, and the results are presented. High and Low frequency AE signals were observed and their possible source mechanisms seem to be piezoelectric strain / domain alignments / fallbacks and debonding of counter electrode / microcrack initiation and propagation due to thermal inhomoginities – respectively. During temperature decrease cycle, with a dc bias being present, the PT seems to be indicated by the appearance of a low frequency AE signal.

Keywords: Acoustic Emission, PZT ceramics, Phase Transition

1. INTRODUCTION

Acoustic emission (AE) is a term used to describe the transient elastic waves generated from the deformation process of materials. All the moving defects and rapid structural changes are accompanied by AE activity. In the case of ferroelectric materials, which are of present interest, the domain alignment and fallbacks / propagation of micro / macro crack, stress induced twinning, structural phase transitions, electrical discharges, etc., are some of the possible source mechanisms of AE. Several workers[1-4] have reported AE in ferroelectric single crystals around their Phase Transition (PT) temperature and attributed it to the sudden lattice parameter changes and distortions at Tc. However, in the case of polycrystalline materials, Aparna[5] and Aparna et al[6] while studying AE during PT in soft and hard poled PZT ceramics did not observe any significant AE activity at PT. The authors in the present work give reason for this absence of AE and suggest an alternate method to observe measurable AE signals at PT and substantiate it experimentally. Apart from this, in the present work, AE was monitored from PZT samples subjected to different dc bias to verify the inferences made by Tsurumi et al[7] who opined that the observed strains in PZT are composed of both piezoelectric strain and strain due to 90º domain reorientations.

2. EXPERIMENTAL

Poled and unpoled PZT circular disk samples of - PZT-5A, PZT 5J, PZT 5H and PZT 4 (20 mm dia and 1 mm thick), obtained from Sparkler Ceramics, Pune, India, were used in the present studies. A simplified experimental setup for PT studies using AE is shown in Fig.1. The complete description of the setup is discussed else where[8]. The sample under study was heated through its curie point from room temperature to about 350 ºC, at an approximate rate of 60ºC / Hr using an oven and later air cooled.

AE was continuously monitored using a broad band transducer (PAC – micro 80D). For applying the field, a high voltage (HV) function generator cum amplifier (Model No. 609E-6-FG, M/s Trek Inc., USA) was used in the amplifier mode with an appropriate dc input from a lab power supply. Temperature of the sample was monitored using a chromel-alumel thermocouple. The output of the thermocouple and a fraction of the applied field were given as parametric inputs to DiSP card for monitoring.

3. RESULTS AND DISCUSIIONS

In the present work mainly two types of AE signals were observed (fig. 2) namely Low Frequency AE (LF-AE) and High Frequency AE (HF-AE), whose principal frequency components were, centered about 100 KHz and 300 KHz respectively. Aparna et al[6] during their studies on soft and hard PZT ceramics concluded that domain alignment / fall backs and microcrack signals give rise to typical HF-AE and LF-AE signals respectively. The authors in the present work used the above information to interpret the results and the signals observed at different temperatures are discussed in detail as follows.

3.1. LF-AE signal at PT under an applied field condition

As mentioned in the introduction, no AE was observed around PT in PZT ceramics by earlier workers. This absence of AE signals especially during the Temperature decrease cycle (T_{DEC}) was surprising and can possibly be explained as follows. During the PT from cubic to tetragonal lattice structure, the stresses induced in the grains are in all possible directions due to the randomly oriented effective polarisation vectors (Ps) of the grains (tetragonal phase). Thus there is no "co-operative" deformation of grains taking place in the direction of observation i.e. perpendicular to the thickness of sample, to yield any detectable AE. It was felt that, this co-operative deformation of grains can be achieved by applying a dc field much before PT in the paraelectric phase and retaining it even after PT. Thus, immediately at PT, the effective Ps vectors of grains will try to align in the direction of the applied field and generate a large stress, which will get released by an initiation of microcrack. This, inturn should appear as a LF-AE signal. This indeed was found to be the case and a LF-AE signal was observed indicating the PT in all the PZT ceramics experimented. The source mechanism of this LF-AE was confirmed by a SEM (not shown here) photograph of the body of the experimented sample, which revealed a microcrack. However, it may be noted that the PT temperature determined by this method is under the presence of an electric field and thus will be higher than what it would be without field[9].

Thus, to determine the PT temperature by AE method, a suitable dc field was applied between T_L and T_H (Table – 1 above shows these values for different varieties), where T_H - $T_L \approx 100$ºC encompassing the PT temperature. Threshold value for capturing AE was increased to compensate the background noise generated by switching on the high voltage power supply[10]. Several poled samples were experimented and the PT temperature was determined as the temperature at which the first LF-AE occurs during T_{DEC} cycle under the applied field condition. This LF-AE was attributed to the PT, as it was felt that at these temperatures no other source mechanism other than PT could be responsible for the initiation of a microcrack, since the sample will be in cubic phase during T_H to Tc. Further, a comparison of PT temperature determined by AE method with and without field (columns 6 & 3 respectively) was found to be in agreement with Le Chatelier's principle, which suggests an increase of PT temperature with increasing field. It may be noted that the PT temperature without field (column No. 3 of Table-1) was obtained experimentally from the dielectric constant Vs Temperature graph.
In the event of wishing to determine the PT temperature of an unknown composition of PZT sample, the electric field must be applied through out the experiment during T_{DEC} and the

temperature at which first LF-AE appears will indicate the PT. Further studies are underway to simultaneously monitor AE and obtain PT temperature by an independent method.

3.2. HF-AE signals due to poling

Poling is a process by which the randomly oriented domains inside the ceramic are aligned in a given direction by applying strong electric field while cooling from a suitably high temperature. In the present work, the dc bias applied to observe the AE at PT has resulted in the observation of HF-AE signals due to poling in the temperature range of T_L ~ Tc during both increasing and decreasing temperature cycles. It was decided to study the variation of these HF-AE signals with the applied field in order to understand its source mechanism(s). Several unpoled samples of all the soft ceramic varieties were experimented and the field was applied in steps of 0.5 kV/cm initially till 2 kV/cm and in steps of 2 kV/cm till 10 kV/cm. For every rise in the field new sample was used. Since the results obtained in PZT 5A, PZT 5J & PZT 5H were similar, representative results obtained in PZT 5A alone are discussed here. The representative AE distribution plots obtained at high and low fields are shown in Fig. 3 in the case of PZT 5A. From the AE distribution plots at different fields, it was observed that at higher fields (>2 kV/cm), multiple peaks were occurring, and in general these peaks shift towards lower temperature as the field increases. The observed multiple peaks can be explained based on the fact that domain switching occurs in bunches[11] and similar discontinuous nature of the AE counts was also observed by Jiyang et al[12] in PLZT ceramics. The shifting of peaks to lower temperatures at higher fields supports the known fact that the higher fields are required for poling at lower temperatures.

Representative HF-AE signals observed in all the varieties, obtained at lower fields (< 2 kV/cm) and higher fields (> 2 kV/cm) were subjected to unsupervised pattern recognition under Noesis environment[13]. This yielded two clusters (Fig.4) – signals arising due to the lower fields falling into one cluster and the rest of the signals obtained at higher fields falling in to another cluster. This indicates that the source mechanisms responsible are different for the observed AE at lower and higher fields. The authors feel that the results obtained in this work support the inference of Tsurumi et al[7] that total strain in a PZT comprises of both "piezoelectric" and "domain alignment" strain components. Additionally it was also found that the amplitude of HF-AE signals occurring due to piezoelectric strain (36 mV) are always much smaller than the signals due to domain alignment (80 mV). This is to be expected as it is well known that domain alignments lead to larger deformation of grains when compared with the deformation generated by piezoelectric strain.

3.3. HF-AE signals due to depoling

HF-AE signals were observed in the present work below T_L (i.e. under no filed condition) in the case of poled samples during T_{INC} cycle. These HF-AE signals were essentially same as that of poling signals but had lower amplitudes. These are attributed to the thermal depoling of the sample due to domain fallbacks facilitated by temperature. M.W. Hooker[14] while studying the properties of PZT 5A, PZT 5H ceramics between -150 ºC and 250 ºC found that PZT 5H gets depoled fully by the time it reaches 170ºC and PZT 5A does not get depoled till it reaches its Tc. In the present study this was confirmed by AE method i.e. in the case of PZT 5A and PZT 5J these HF-AE signals were observed till their Tc, where as in the case of PZT 5H these were observed only till about 190 ºC which indicates that PZT 5H gets totally depoled when it reaches this temperature. The higher value of 190ºC obtained by us may be due to the microstructural differences in the samples used by us and Hooker. It was also observed that these HF-AE signals normally cluster around two temperatures, around 50ºC and 150ºC in PZT 5A and PZT 5J. However, in PZT 5H these signals were observed continuously over the temperature range of ambient to about 150ºC. This probably is an indication that PZT 5H depoles more linearly while PZT 5A & 5J depoles nonlinearly.

3.4. LF-AE signals observed below T_L

The LF-AE signals observed below T_L were mainly grouped in to two groups – one below 100ºC and the other between 100ºC and T_L ºC. The SEM photographs of the experimented samples till T_L revealed large number of black spots on the periphery of the counter electrode and many microcracks in the body of the ceramic. Representative microphotographs in PZT 5A are shown in fig. 5. To understand the source mechanism of the signals observed below 100ºC - one of the samples was experimented only till 100 ºC and was taken for SEM observation at a magnification of 100. It revealed a number of black spots at the periphery of the sample, indicating the debonding of counter electrode at these places. It is a known fact that the periphery of a sample will be weak and may contain trapped gaseous matter during uniaxial compaction used for pelletisation. The release of these gasses could have given rise to AE. Further, an AE experiment conducted on an unelectroded sample did not give any LF-AE signals below 100ºC, indicating that the surface electrode alone was responsible for these signals.

The LF-AE signals observed between 100ºC to 200ºC were attributed to the microcracks generated due to the thermal inhomoginities at the weak boundary of the unpoled samples. However in the case of poled samples, apart from the thermal inhomogeneity; thermal anisotropic expansion could also have been a source. These results point to the fact that the origin of LF-AE signals could be either debonding of counter electrode or microcracking within the specimen. Pattern recognition studies are being conducted on these LF-AE signals to discriminate between these different source mechanisms.

4. CONCLUSIONS

1) In the present work for the first time the authors suggest that the observation AE at PT in PZT ceramics is possible if a suitable dc bias is applied around PT.

2) Pattern recognition was used to discriminate between the two source mechanisms responsible for the observed HF-AE under an applied field – namely “piezoelectric strain” and “strain due to domain alignments”.

ACKNOWLEDGEMENTS

One of the authors KPR is thankful to the DRDO for the award of JRF. Authors thank Osmania University for providing the facilities. Some of the results of the above work have been undertaken in a DRDO sponsored project and the authors thank the concerned authorities.

REFERENCES

1. Buchman,P., *Solid State Electron.*, **15**, (1972) 142.
2. Lambson,E.F., Linz,a., Madhava,M.R., Mohamad,I.J., Underhill,C. and Saunders,G.A., *Proc.Inst.Acoustics*, **4(8)**, (1977) 1.
3. Dulkin,E.A., Gavrilyachenko,V.G. and Semenchev,A.F., *Sov.Phys. Solid State*, **34(5)**, (1992) 863-864.
4. Zammit-Mangion, L.J. and Saunders. G.A., J.Phys.C:Solid State Phys., **17**, (1984) 2825-2830.
5. Aparna Dode, *Ph.D. Thesis: “ PR of AE signals from piezoelectric ceramics”,* The Osmania University, October, 1999.

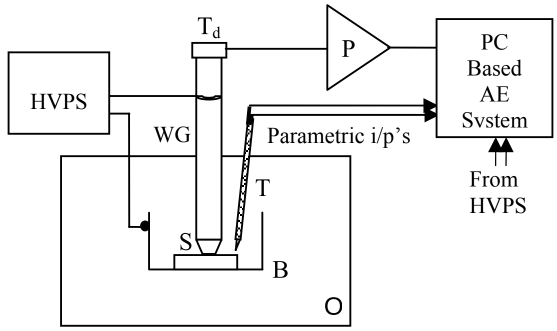
6. Aparna D., and Mallikarjun Rao S.P., *"Pattern Recognition of Acoustic Emission Signals from PZT ceramics", www.ndt.net*, **Vol.7, No.9**, September, 2002.
7. Tsurumi, T., Kumano, Y., Ohashi, N., Takenaka, T., and Fukunaga, O., *Jpn. J. Appl. Phys.*, **36**, (1997) 5970 -5975.
8. Prabakar K., Aparna Dode, Manjunath Rao S. and Mallikarjun Rao S.P.," *Phase transition studies on PZT ceramics using Acoustic Emission*", - Proceedings of 26th European Conference on Acoustic Emission Testing, Sep 15-17, 2004, Berlin Germany, *PP* 267-82.
9. Jaffe,B., Cook Jr.,W.R., and Jaffe,H., *"Piezoelectric Ceramics"* , 1971, Academic Press, London, Page Nos 57 & 76.
10. Aburthani, H., Uchino, K., *Jpn.J.Appl.Phys.*, **35**, L516-L518 (1996).
11. Subbarao, E.C., Srikanth, V., Cao, W., and Cross, L.E., *Ferroelectrics,* **145**, (1993) 271-281.
12. Jiyang, Q.Y., Subbarao. E.C., and Cross, L.E., *Ferroelectrics,* **154**, (1994) 113-118.
13. Pattern recognition software – NOESIS Pro Ver. 3.3, for windows developed by Envirocousitcs, Greece (2002).
14. Hooker M.W., NASA report – NASA / CR – 1998 – 208708, USA (1998).

TABLE

Table 1. PT determined by AE method (experimental conditions and results)

Sl.No.	Sample type	PT temp. from Dielectric Constant Graph during T_{DEC}(No field case) ºC	Field applied Kv/cm	T_L - T_H ºC	Temp. at which LF-AE signal is obtained while T_{DEC} ºC
1	PZT 5A	314	10	250-350	329
2	PZT 5J	239	10	150-300	254
3	PZT 5H	240	6	150-275	259
4	PZT 4	331	1	250-350	294

FIGURES



HVPS – High Voltage Power Supply, O – Oven, P – Pre-Amplifier (60 dB), S – Sample, B – Brass cup, T – Thermo couple, T_d – Transducer, WG – Waveguide

Fig. 1. Acoustic Emission Experimental Setup for PT studies

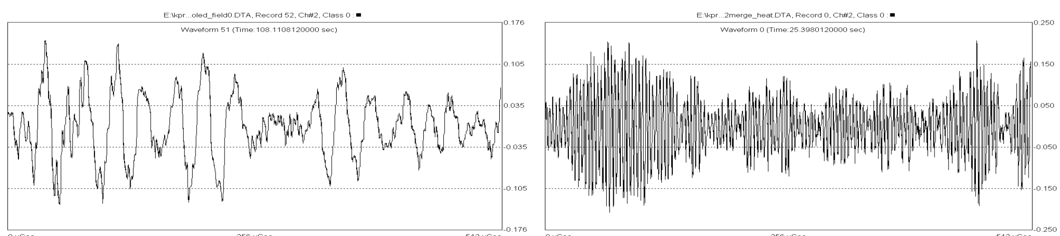


Fig. 2. Typical (a) LF-AE signal (b) HF-AE signal

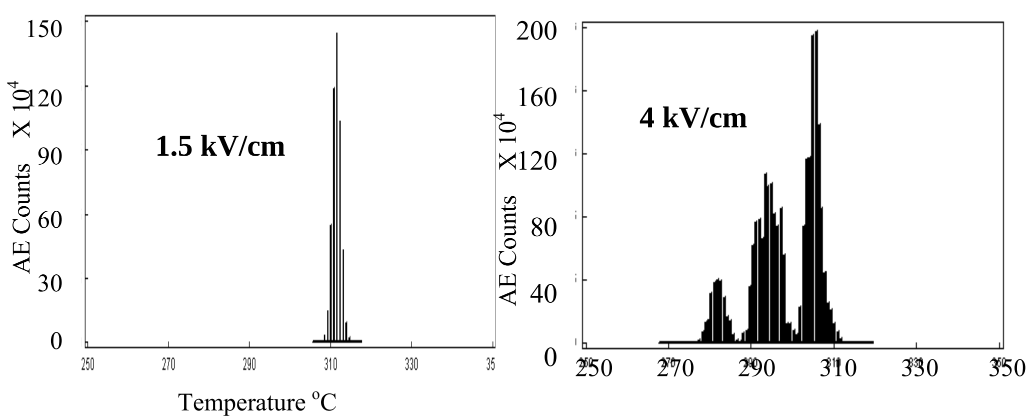


Fig. 3 Distribution plots showing the AE counts rate during T_{DEC} in PZT 5A. Plots are drawn between T_L and T_H.

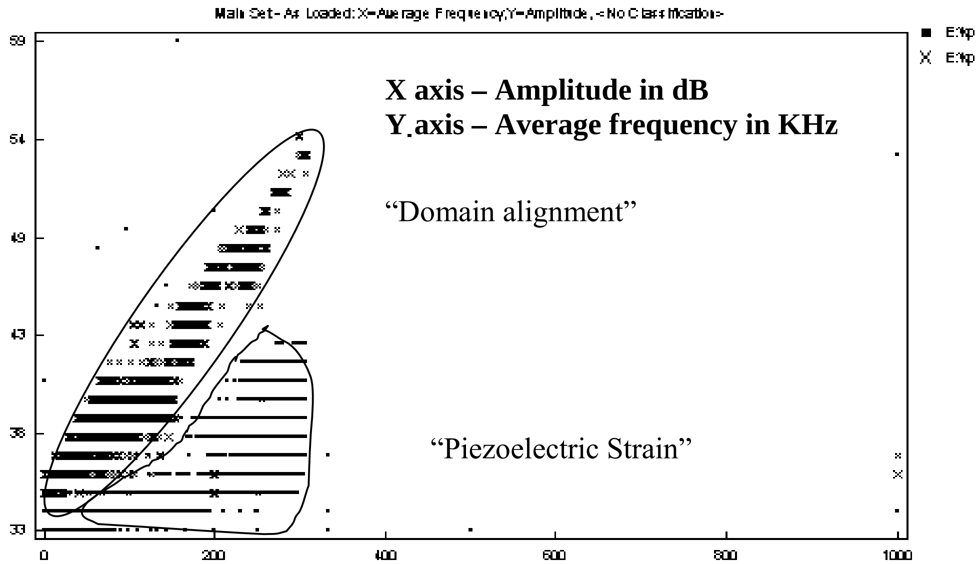


Fig. 4 PR chart showing two clusters indicating two possible source mechanisms for the observed HF-AE

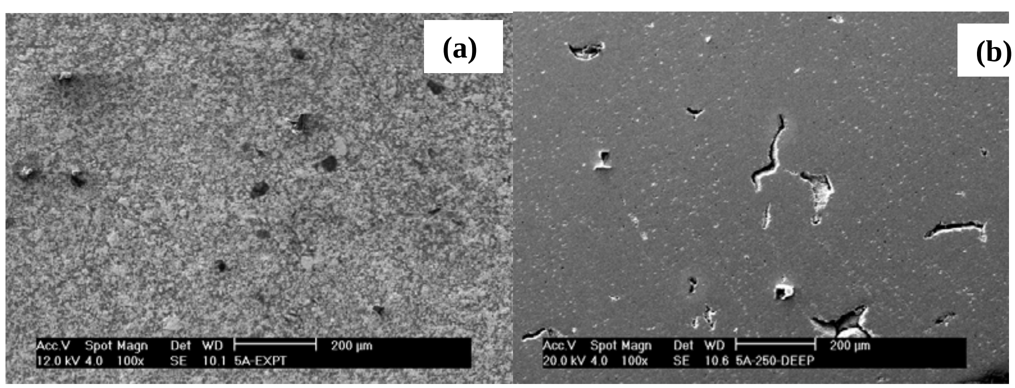


Fig. 5 SEM Photo graph of PZT 5A (a) experimented till 100 ºC – showing the black spots on the electrode, (b) experimented till 250 ºC - several microcracks can be seen on the surface beneath the electrode