Stress-mediated tuning of ferroelectric properties in $0.68Pb(Mn_{1/3}Nb_{2/3})O_3$-$0.32PbTiO_3$ single crystals

A Thesis

Submitted

By

ADITYA CHAUHAN (S12021)

For the award of degree of

Master of Science (*by research*)

SCHOOL OF ENGINEERING

INDIAN INSTITUTE OF TECHNOLOGY MANDI

Mandi, Himachal Pradesh -175001

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Dedicated to

my teachers

and

family

Declaration by the Research Scholar

This is to certify that the thesis titled "**Stress-mediated tuning of ferroelectric properties in $0.68Pb(Mn_{1/3}Nb_{2/3})O_3$-$0.32PbTiO_3$ single crystals**" submitted by me, to the Indian Institute of Technology Mandi for the award of the degree of **Master of Science (by research)**, is a bona fide record of the research work carried out by me in the School of Engineering, Indian Institute of Technology Mandi, under the supervision of Dr. Rahul Vaish. The contents of this thesis, in full or in parts, have not been submitted to any other Institute or University for the award of any degree or diploma.

In keeping with the general practice of reporting scientific observation, due acknowledgements have been made wherever the work described is based on the findings of other investigators.

School of Engineering
Indian Institute of Technology Mandi
Mandi (H.P.)

Aditya Chauhan

Date:

Thesis Certificate

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Dr. Rahul Vaish
Assistant Professor
School of Engineering
Indian Institute of Technology Mandi
Mandi, (H.P.)

Date:

Preamble

Ferroelectrics form an important class of materials and are employed for a variety of applications. However, specific systems and devices dictate the need of tailored ferroelectric response. This creates a requirement to obtain ferroelectric materials with tunable properties. It is an established fact the ferroelectric behaviour is a function of the domain response/switching behaviour when subjected to external impetus. Thus, domain engineering is the key for inception of tuneable material attributes. Generally, chemical modifications are employed to this effect in the form of doping or solid-solutions. However, this step complicates the material system unnecessarily and leads to long term predicaments including functional fatigue, electrical hardening and costly fabrication processes. Therefore, need has been felt for a physical alternative leading to desired domain engineering. In this regards, this study attempts to shed light on the use of compressive pre-stresses for tuning and enhancing the ferroelectric properties.

Chapter 1 forms the beginning of thesis with detailed explanation of the functional hierarchy in dielectric materials including the piezoelectric, pyroelectric, ferroelectric and relaxor ferroelectric materials.

Chapter 2 discusses the phenomenon of ferroelectric and ferroelastic switching behavior under combined electromechanical loading in PMN-32PT. Subsequently a brief description has been provided how this phenomenon can be employed to tune the pyroelectric and piezoelectric coefficients of PMN-32PT.

Chapter 3 provides a detailed discussion regarding the effect of directional confinement on the electrical energy storage characteristics of PMN-32PT. Indirect measurements have been made employing unipolar *P-E* loops obtained under various stress levels and operating temperatures.

In chapter 4 a discussion regarding the mechanism of various caloric effects in ferroelectric materials and their indirect measurement by using modified Maxwell's relations has been provided. Emphasis has been laid on differentiating the various modes of measurement, their inferences and the underlying mechanism of a particular caloric effect.

Chapter 5 presents the effects of stress-mediation on the electrocaloric behavior of PMN-32PT single crystals. Furthermore, a novel cycle has been proposed to successfully combine the two different phenomenon of electrocaloric and elastocaloric effect to enhance the cooling capacity in PMN-32PT.

The thesis concludes by summarizing the key findings of the investigation and highlighting where maximum benefits, enhancements and improvements can be obtained. This research provides a direct experimental verification of how application of premeditated multi-physical stimulus can be used as an effective route for augmenting desirable characteristics of relaxor based ferroelectric PMN-32PT single crystals. The following publications are largely based on the studies conducted as a part of the research work reported over here.

1. **Chauhan, A.**, Patel, S. and Vaish, R. Mechanical confinement for tuning ferroelectric response in PMN-PT single crystal. ***J. Appl. Phys.*** **117 (8)**: 084102, **2015.**

2. **Chauhan, A.**, Patel, S. and Vaish, R. Effect of directional mechanical confinement on the electrical energy storage density in $68Pb(Mn_{1/3}Nb_{2/3})O_3$-$32PbTiO_3$ single crystals. ***Ferroelectrics*** **478**: 40-53, **2015**.

3. **Chauhan, A.**, Patel, S. and Vaish, R. Multicaloric effect in $Pb(Mn_{1/3}Nb_{2/3})O_3$-$32PbTiO_3$ single crystals. ***Acta Mater.*** **89**: 384-395, **2015**.

4. **Chauhan, A.**, Patel, S. and Vaish, R. Multicaloric effect in $Pb(Mn_{1/3}Nb_{2/3})O_3$-$32PbTiO_3$ single crystals: modes of measurement. ***Acta Mater.*** (doi: 10.1016/j.actamat.2015.06.027), **2015**.

Other publications include:

1. **Chauhan, A.** and Vaish, R. Magnetic material selection using multiple attribute decision making approach. ***Mater. Des.*** **36**: 1-5, **2012**.
2. **Chauhan, A.** and Vaish, R. A comparative study on material selection for micro-electromechanical systems. ***Mater. Des.*** **41**: 177-181, **2012**.
3. **Chauhan, A.** and Vaish, R. Hard coating material selection using multi-criteria decision making. ***Mater. Des.*** **44**: 240-245, **2013**.

4. **Chauhan, A.** and Vaish, R. An assessment of bulk metallic glasses for microelectromechanical system devices. ***Int. J. Appl. Glass Sci*.** **4(3)**: 231-241, **2013**.
5. **Chauhan, A.**, Vaish, R. and Bowen, C. Piezoelectric material selection for ultrasonic transducer and actuator applications. ***J. Mater. Des. Appl.*** **229(1)**: 3-12, **2015**.
6. Patel, S., **Chauhan, A.** and Vaish, R. A technique for giant mechanical energy harvesting using ferroelectric/antiferroelectric materials. ***J. Appl. Phys*.** **115(8)**: 084908, **2014**.
7. **Chauhan, A.**, Patel, S., Vats, G. and Vaish, R. Enhanced thermal energy harvesting using Li, K doped $Bi_{0.5}Na_{0.5}TiO_3$ lead-free ferroelectric ceramics. ***Energ. Technol.*** **2(2)**: 205-209, **2014**.
8. Patel, S., **Chauhan, A.** and Vaish, R. Analysis of high field energy harvesting using ferroelectric materials. ***Energ. Technol.*** **2(5)**: 480-485, **2014**.
9. **Chauhan, A.**, Patel, S. and Vaish, R. Mechanical confinement for improved energy storage density in BNT-BT-KNN lead-free ceramic capacitors. ***AIP Adv.*** **4(8)**: 087106, **2014**.
10. Vats, G., **Chauhan, A.** and Vaish, R. Thermal energy harvesting using bulk lead-free ferroelectric ceramics. ***Int. J. Appl. Ceram. Technol.*** **12**: E49-54, **2015**.
11. Patel, S., **Chauhan, A.** and Vaish, R. Enhanced energy harvesting in commercial ferroelectric materials. ***Mater. Res. Expr.*** **1(2)**: 025504, **2014**.
12. Bowen, CR., Taylor, J., LeBoulbar, E., Zabek, D., **Chauhan, A.** and Vaish R. Pyroelectric materials and devices for energy harvesting applications. ***Energ. Environ. Sci.*** **7(12)**: 3836-3856, **2014**.
13. Rastogi, M., **Chauhan, A.**, Vaish, R. and Kishan, A. Selection and performance assessment of phase change materials heating, ventilation and air-conditioning applications. ***Energ. Conver. Manage.*** **89**: 260-269, **2015**.
14. Patel, S., Chauhan, A., **Chauhan, A.** and Vaish, R. Temperature dependent scaling behaviour of the dynamic hysteresis in $0.715Bi_{0.5}Na_{0.5}TiO_3$-$0.065BaTiO_3$-$0.22SrTiO_3$ ferroelectric ceramics. ***Mater. Res. Expr.*** **2(3)**: 035501, **2015**.
15. Sharma, M., **Chauhan, A.**, Vaish, R. and Chauhan VS. Finite element analysis on solar energy harvesting using ferroelectric polymer. ***Solar Energ.*** **115:** 722-732, **2015**.

16. **Chauhan, A.**, Patel, S. and Vaish, R. Giant electro-mechanical energy conversion in lead-free ferroelectric materials. ***Ferroelectrics Lett. Sec.*** **42(1-3)**: 35-42, **2015**.
17. **Chauhan, A.**, Patel, S. and Vaish, R. Elastocaloric effects in ferroelectric ceramics. ***Appl. Phys. Lett.*** **106(17)**: 172901, **2015**.
18. **Chauhan, A.**, Patel, S. and Vaish, R. Enhanced electrocaloric effect in pre-stressed ferroelectric materials. ***Energ. Technol.*** **3(2)**: 177-186, **2015**.
19. Patel, S., **Chauhan, A.** and Vaish, R. Improved electrical energy storage density in Vanadium-doped $BaTiO_3$ bulk ceramics by addition of $3BaO$-$3TiO_2$-B_2O_3 glass. ***Energ. Technol.*** **3(1)**: 70-76, **2015**.
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ADITYA CHAUHAN

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Table of contents