

TABLE OF CONTENTS

	Page
ACKNOWLEDGEMENT	iii
ABSTRACT (THAI)	iv
ABSTRACT (ENGLISH)	vi
LIST OF TABLES	xi
LIST OF FIGURES	xiii
ABBREVIATIONS AND SYMBOLS	xix
 CHAPTER 1 INTRODUCTION	 1
CHAPTER 2 LITERATURE REVIEW	
2.1. Basic Definitions and Characteristics of Ferroelectric Materials	3
2.1.1. Piezoelectricity	3
2.1.2. Pyroelectricity	4
2.1.3. Ferroelectricity	5
2.1.3.1. Ferroelectric phase and domains	5
2.1.3.2. Curie temperature and phase transition in ferroelectrics	7
2.1.3.3. Polarization switching process in ferroelectrics	8
2.1.3.4. The hysteresis loop of ferroelectric	9
2.1.3.5. Poling of ferroelectrics	11
2.1.4. Perovskite structure	12
2.2. Normal and Relaxor Ferroelectrics	13
2.2.1. PZT ceramic	16
2.2.2. PMN ceramic	19
2.3. Prior Studies of Hysteresis Properties on PMN-PZT ceramics	21

CHAPTER 3 EXPERIMENTAL PROCEDURE

3.1. Sample preparation	23
3.1.1. Powder preparation	23
3.1.1.1. Preparation of lead zirconate titanate powders	26
3.1.1.2. Preparation of lead magnesium niobate powders	26
3.1.1.3. Preparation of lead magnesium niobate - lead zirconate titanate powders	27
3.1.2. Ceramic preparation	27
3.1.3. Poling procedure	29
3.2. Sample characterizations	29
3.2.1. The X-ray diffraction (XRD) technique	30
3.2.2. Scanning electron microscopy (SEM)	31
3.2.3. Densification analysis	31
3.3. Experiment setup for hysteresis properties measurement	32
3.4. Measurements of hysteresis properties of lead magnesium niobate - lead zirconate titanate ceramics	36

CHAPTER 4 RESULTS AND DISCUSSION

4.1. Phase formation analysis	37
4.2. Microstructural analysis	40
4.3. Densification analysis	50
4.4. Hysteresis properties	51
4.4.1. Hysteresis loop evolution	52
4.4.2. Hysteresis properties dependence on poling field strength	59
4.4.2.1. PZT	59
4.4.2.2. 0.1PMN-0.9PZT	61
4.4.2.3. 0.3PMN-0.7PZT	63
4.4.2.4. 0.5PMN-0.5PZT	65
4.4.2.5. 0.7PMN-0.3PZT	67
4.4.2.6. 0.9PMN-0.1PZT	69

4.4.2.7. PMN	71
4.4.3. Hysteresis properties dependence on composition ratio	73
4.4.3.1. The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of (x)PMN-(1-x)PZT ceramics poled at 10 kV/cm	73
4.4.3.2. The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of (x)PMN-(1-x)PZT ceramics poled at 20 kV/cm	75
4.4.3.3. The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of (x)PMN-(1-x)PZT ceramics poled at 30 kV/cm	77
4.4.3.4. The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of (x)PMN-(1-x)PZT ceramics poled at 40 kV/cm	78
4.4.4. The maximum hysteresis parameters	80
CHAPTER 5 CONCLUSIONS AND SUGGESTIONS	
5.1. Conclusions	81
5.2. Suggestions	82
REFERENCE	84
APPENDIX	90
VITA	119

LIST OF TABLES

Table	Page
2.1 Differences between normal and relaxor ferroelectric	16
3.1 Sintering temperature, dwell time, and heating/cooling rates for (x)PMN-(1-x)PZT ceramics	28
4.1 Grain size range and average grain size of (x)PMN-(1-x)PZT ceramics	41
4.2 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of PZT ceramic	60
4.3 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.1PMN-0.9PZT ceramic	62
4.4 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.3PMN-0.7PZT ceramic	64
4.5 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.5PMN-0.5PZT ceramic	66
4.6 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.7PMN-0.3PZT ceramic	68
4.7 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.9PMN-0.1PZT ceramic	69
4.8 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of PMN ceramic	71
4.9 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of PMN-PZT ceramics poled at 10 kV/cm	74
4.10 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of PMN-PZT ceramics poled at 20 kV/cm	76
4.11 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of PMN-PZT ceramics poled at 30 kV/cm	77
4.12 Remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of PMN-PZT ceramics poled at 40 kV/cm	79

- 4.13 The maximum value of remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of PMN-PZT ceramics 80



ลิขสิทธิ์มหาวิทยาลัยเชียงใหม่
Copyright © by Chiang Mai University
All rights reserved

LIST OF FIGURES

Figure	Page
2.1 Interrelationship of piezoelectric and subgroups on the basis of symmetry	4
2.2 Unit cells of the ferroelectric phases of BaTiO ₃	6
2.3 Schematic illustration of 180° and 90° domain walls	7
2.4 (a) Surface charge associated with spontaneous polarization (b) formation of 180° domains to minimize electrostatic energy	7
2.5 Schematic of 90° and 180° domain switching induced by an electric field above the coercive strength ($E \geq E_c$)	9
2.6 Schematic circuit of Sawyer-Tower for the observation of P-E characteristics of ferroelectric	10
2.7 A typical P-E hysteresis loop in ferroelectric	11
2.8 Schematic illustration of the domain structure and poling process	12
2.9 A cubic ABO ₃ perovskite-type unit cell	13
2.10 Schematic temperature dependence of the dielectric permittivity (ϵ) and spontaneous polarization (P_s) for (a) a first-order and (b) a second-order ferroelectric	14
2.11 Temperature dependence of dielectric permittivity and loss factor for relaxor ferroelectric of PMN ceramic	15
2.12 Cubic perovskite-type structure of PZT	17
2.13 Phase diagram of the PbTiO ₃ -PbZrO ₃ solid solution (a) by Jaffe <i>et al.</i> , (b) by Noheda <i>et al.</i>	18
2.14 Dielectric and piezoelectric properties of the PbTiO ₃ -PbZrO ₃ solid solution	19
2.15 Two unit cells of the Perovskite structure of PMN showing the chemical-ordering model. The B-site marked Mg is referred to as the B' site while Nb ions are on the B'' sites	20
3.1 Experimental procedure diagram	24
3.2 Preparation route for powder	25
3.3 Sintering process diagram (when T _s is sintering temperature)	28

Figure

	Page
3.4 Poling setup used to pole samples in this study	29
3.5 X-ray diffractometer used in this study	30
3.6 Scanning electron microscopy (SEM) used in this study	32
3.7 Schematic of the standard Sawyer-Tower circuit ($R_1 = 6.8 \text{ M}\Omega$, $R_2 = 10 \text{ k}\Omega$, $C_0 = 0.1 \text{ }\mu\text{F}$, $C_s = \text{Sample}$, $C_0 \gg C_s$)	33
3.8 The experiment setup for hysteresis loop measurements	33
3.9 Example of a hysteresis loop obtained from the experimental set-up	34
4.1 XRD pattern of PZT ceramic which with JCPDS file no. 50-0346	38
4.2 XRD pattern of PMN ceramic (with JCPDS file no. 27-1199) including small amount of pyrochlore (with JCPDS file no. 33-0769)	39
4.3 XRD pattern of (x)PMN-(1-x)PZT ceramics	40
4.4 SEM micrograph of (a) free and (b) fracture surfaces of PZT ceramic.	42
4.5 SEM micrograph of (a) free and (b) fracture surfaces (0.1)PMN-(0.9)PZT ceramic	43
4.6 SEM micrograph of (a) free and (b) fracture surfaces (0.3)PMN-(0.7)PZT ceramic	44
4.7 SEM micrograph of (a) free and (b) fracture surface (0.5)PMN-(0.5)PZT ceramic	45
4.8 SEM micrograph of (a) free and (b) fracture surface (0.7)PMN-(0.3)PZT ceramic	46
4.9 SEM micrograph of (a) free and (b) fracture surface (0.9)PMN-(0.1)PZT ceramic	47
4.10 SEM micrograph of (a) free and (b) fracture surface PMN ceramic	48
4.11 Representative EDX spectra obtained from submicron size grains on the surface of (x)PMN-(1-x)PZT ceramics	49
4.12 Representative EDX spectra obtained from large grain on the surface of (x)PMN-(1-x)PZT ceramics	49
4.13 Densities of (x)PMN-(1-x)PZT ceramics	50
4.14 Hysteresis loops evolution of commercial sample (PKI-552 or soft PZT) taken at AC drive amplitudes of: (a) 5.13, (b) 10.14, (c) 12.17 and (d) 19.20 kV/cm	51
4.15 Hysteresis loops evolution of PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : (a) 6.04, (b) 8.03, (c) 10.09 and (d) 14.06 kV/cm	52
4.16 Hysteresis loops of 0.1PMN-0.9PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : (a) 4.57, (b) 6.03, (c) 9.04 and (d) 11.14 kV/cm	54

Figure

	Page
4.17 Hysteresis loops of 0.3PMN-0.7PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : (a) 4.04, (b) 5.04, (c) 6.54 and (d) 8.04 kV/cm	54
4.18 Hysteresis loops of 0.5PMN-0.5PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : (a) 8.11, (b) 9.12, (c) 9.56 and (d) 10.13 kV/cm	55
4.19 Hysteresis loops of 0.7PMN-0.3PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : (a) 6.05, (b) 11.03, (c) 14.00 and (d) 18.03 kV/cm	56
4.20 Hysteresis loops of 0.9PMN-0.1PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : (a) 5.59, (b) 11.08, (c) 15.03 and (d) 20.03 kV/cm	57
4.21 Hysteresis loops of PMN ceramics poled at 30 kV/cm taken at AC drive amplitudes of : (a) 6.04, (b) 7.05, (c) 15.03 and (d) 20.14 kV/cm	57
4.22 The saturated hysteresis loop of PZT ceramic poled at poling field strength of : (a) 10, (b) 20 and (c) 30 kV/cm	59
4.23 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of PZT ceramic as function of poling field strength	61
4.24 The saturated hysteresis loop of 0.1PMN-0.9PZT ceramic poled at poling field strength of: (a) 10, (b) 20, (c) 30 and (d) 40 kV/cm	62
4.25 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.1PMN-0.9PZT ceramic as function of poling field strength	63
4.26 The saturated hysteresis loop of 0.3PMN-0.7PZT ceramic poled at poling field strength of: (a) 10, (b) 20, (c) 30 and (d) 40 kV/cm	64
4.27 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.3PMN-0.7PZT ceramic as function of poling field strength	65
4.28 The saturated hysteresis loop of 0.5PMN-0.5PZT ceramic poled at poling field strength of: (a) 10, (b) 20, (c) 30 and (d) 40 kV/cm	66
4.29 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.5PMN-0.5PZT ceramic as function of poling field strength	67
4.30 The saturated hysteresis loop of 0.7PMN-0.3PZT ceramic poled at poling field strength of: (a) 10, (b) 20, (c) 30 and (d) 40 kV/cm	68

Figure

	Page
4.31 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.7PMN-0.3PZT ceramic as function of poling field strength	69
4.32 The saturated hysteresis loop of 0.9PMN-0.1PZT ceramic poled at poling field strength of: (a) 10, (b) 20, (c) 30 and (d) 40 kV/cm	70
4.33 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.7PMN-0.3PZT ceramic as function of poling field strength	70
4.34 The saturated hysteresis loop of PMN ceramic poled at poling field strength of: (a) 10, (b) 20, (c) 30 and (d) 40 kV/cm	71
4.35 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of 0.7PMN-0.3PZT ceramic as function of poling field strength	72
4.36 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of (x)PMN-(1-x)PZT ceramics poled at 10 kV/cm	75
4.37 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of (x)PMN-(1-x)PZT ceramics poled at 20 kV/cm	76
4.38 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of (x)PMN-(1-x)PZT ceramics poled at 30 kV/cm	78
4.39 The remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) of (x)PMN-(1-x)PZT ceramics poled at 40 kV/cm	79
A.1 Hysteresis loops evolution of commercial sample (PKI-552 or soft PZT) taken at AC drive amplitudes of: 3.11 to 19.20 kV/cm	93
A.2 Hysteresis loops evolution of PZT ceramics poled at 10 kV/cm taken at AC drive amplitudes of : 3.02 to 8.03 kV/cm	94
A.3 Hysteresis loops evolution of PZT ceramics poled at 20 kV/cm taken at AC drive amplitudes of : 4.05 to 17.08 kV/cm	95
A.4 Hysteresis loops evolution of PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : 3.02 to 18.04 kV/cm	96
A.5 Hysteresis loops evolution of 0.1PMN-0.9PZT ceramics poled at 10 kV/cm taken at AC drive amplitudes of : 4.57 to 11.05 kV/cm	97

Figure

	Page
A.6 Hysteresis loops evolution of 0.1PMN-0.9PZT ceramics poled at 20 kV/cm taken at AC drive amplitudes of : 4.57 to 11.51 kV/cm	98
A.7 Hysteresis loops evolution of 0.1PMN-0.9PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : 3.03 to 11.14 kV/cm	99
A.8 Hysteresis loops evolution of 0.1PMN-0.9PZT ceramics poled at 40 kV/cm taken at AC drive amplitudes of : 4.57 to 14.52 kV/cm	100
A.9 Hysteresis loops evolution of 0.3PMN-0.7PZT ceramics poled at 10 kV/cm taken at AC drive amplitudes of : 4.04 to 8.04 kV/cm	101
A.10 Hysteresis loops evolution of 0.3PMN-0.7PZT ceramics poled at 20 kV/cm taken at AC drive amplitudes of : 4.04 to 8.04 kV/cm	102
A.11 Hysteresis loops evolution of 0.3PMN-0.7PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : 4.04 to 8.57 kV/cm	103
A.12 Hysteresis loops evolution of 0.3PMN-0.7PZT ceramics poled at 40 kV/cm taken at AC drive amplitudes of : 4.04 to 8.57 kV/cm	104
A.13 Hysteresis loops evolution of 0.5PMN-0.5PZT ceramics poled at 10 kV/cm taken at AC drive amplitudes of : 8.11 to 10.13 kV/cm	105
A.14 Hysteresis loops evolution of 0.5PMN-0.5PZT ceramics poled at 20 kV/cm taken at AC drive amplitudes of : 8.11 to 10.13 kV/cm	106
A.15 Hysteresis loops evolution of 0.5PMN-0.5PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : 8.11 to 10.13 kV/cm	107
A.16 Hysteresis loops evolution of 0.5PMN-0.5PZT ceramics poled at 40 kV/cm taken at AC drive amplitudes of : 8.11 to 10.13 kV/cm	108
A.17 Hysteresis loops evolution of 0.7PMN-0.3PZT ceramics poled at 10 kV/cm taken at AC drive amplitudes of : 5.10 to 15.07 kV/cm	109
A.18 Hysteresis loops evolution of 0.7PMN-0.3PZT ceramics poled at 20 kV/cm taken at AC drive amplitudes of : 5.10 to 16.49 kV/cm	110
A.19 Hysteresis loops evolution of 0.7PMN-0.3PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : 5.10 to 17.08 kV/cm	111

Figure

	Page
A.20 Hysteresis loops evolution of 0.7PMN-0.3PZT ceramics poled at 40 kV/cm taken at AC drive amplitudes of : 6.05 to 18.03 kV/cm	112
A.21 Hysteresis loops evolution of 0.9PMN-0.1PZT ceramics poled at 10 kV/cm taken at AC drive amplitudes of : 4.05 to 18.01 kV/cm	113
A.22 Hysteresis loops evolution of 0.9PMN-0.1PZT ceramics poled at 20 kV/cm taken at AC drive amplitudes of : 4.05 to 20.03 kV/cm	114
A.23 Hysteresis loops evolution of 0.9PMN-0.1PZT ceramics poled at 30 kV/cm taken at AC drive amplitudes of : 5.59 to 20.03 kV/cm	115
A.24 Hysteresis loops evolution of 0.9PMN-0.1PZT ceramics poled at 40 kV/cm taken at AC drive amplitudes of : 5.59 to 20.03 kV/cm	116
A.25 Hysteresis loops evolution of PMN ceramics poled at 10 kV/cm taken at AC drive amplitudes of : 2.09 to 11.00 kV/cm	117
A.26 Hysteresis loops evolution of PMN ceramics poled at 20 kV/cm taken at AC drive amplitudes of : 3.02 to 11.00 kV/cm	118
A.27 Hysteresis loops evolution of PMN ceramics poled at 30 kV/cm taken at AC drive amplitudes of : 4.03 to 20.14 kV/cm	119
A.28 Hysteresis loops evolution of PMN ceramics poled at 40 kV/cm taken at AC drive amplitudes of : 5.04 to 25.03 kV/cm	120

ABBREVIATIONS AND SYMBOLS

PZT	lead zirconate titanate
PZ	lead zirconate
PT	lead titanate
PMN	lead magnesium niobate
BT	barium titanate
MPB	morphotropic phase boundary
T_c	Curie temperature
XRD	X-ray diffraction
JCPDS	joint committee for powder diffraction standard
SEM	scanning electron microscopy
EDX	energy dispersive X-ray spectrophotometry
W	weight
a	lattice parameter of a
D	diameter
ρ	density
V	voltage
C	capacitance
P_r	remanent polarization
P_s	spontaneous polarization
E_c	coercive field
ϵ_r	relative permittivity

ลิขสิทธิ์มหาวิทยาลัยเชียงใหม่

Copyright © by Chiang Mai University

All rights reserved