

## LIST OF CONTENT

<b>Chapter</b>		<b>Page</b>
<b>I      INTRODUCTION.....</b>		<b>1</b>
Overview.....		1
Objectives of this research.....		3
<b>II     THEORY AND LITERATURE</b>		<b>4</b>
Perovskite Structure.....		4
Piezoelectricity.....		5
Ferroelectrics.....		7
Normal ferroelectric.....		9
Relaxor ferroelectric.....		10
Antiferroelectric.....		12
Paraelectric.....		13
Morphotropic phase boundary (MPB).....		14
Typical methods to synthesize ceramics.....		15
One component systems.....		21
Binary systems.....		27
Ternary Systems.....		32
<b>III    MATERIALS AND METHOD.....</b>		<b>37</b>
Sample preparation.....		37
Sample Characterization.....		38
Physical properties.....		39
<b>IV    RESULTS AND DISCUSION: <math>(1-x-y)\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3 - x\text{Bi}_{0.5}\text{K}_{0.5}</math></b>		
$\text{TiO}_3 - y\text{BiFeO}_3$ .....		42
Introduction.....		45
Experimental.....		43

## LIST OF CONTENT (CONT.)

<b>Chapter</b>		<b>Page</b>
	Results and Discussion.....	44
	Conclusions.....	67
<b>V</b>	<b>RESULTS AND DISCUSION: <math>(1-x-y)Bi_{0.5}Na_{0.5}TiO_3</math></b>	
	$-xBi_{0.5}K_{0.5}TiO_3 -yBi_{0.5}Li_{0.5}TiO_3$ .....	68
	Introduction.....	68
	Experimental.....	69
	Results and Discusstion.....	71
	Effect of $x$ and $y$ on electrical properties of BNKLT	
	Ceramics.....	84
	Conclusions.....	94
<b>VI</b>	<b>RESULTS AND DISCUSION: <math>(1-x-y)Bi_{0.5}Na_{0.5}TiO_3</math></b>	
	$-xBi_{0.5}K_{0.5}TiO_3 -yK_{0.5}Na_{0.5}NbO_3$ .....	95
	Introduction.....	95
	Experimental.....	97
	Results and Discusstion.....	97
	Conclusions.....	117
<b>VII</b>	<b>SUMMARY.....</b>	<b>118</b>
	<b>REFERENCE.....</b>	<b>121</b>
	<b>BIOGRAPHY.....</b>	<b>133</b>

## LIST OF TABLES

Table	Page
1 Different properties of normal and relaxor ferroelectrics.....	13
2 Some properties of organic compounds.....	20
3 Effect of various fuels of the Ni-Zn ferrites prepared by the combustion technique.....	21
4 The optimal piezoelectric constant ( $d_{33}$ ) and electromechanical coupling factor ( $k_p$ ) of the composition in various BNT-based piezoelectric ceramics.....	32
5 Specifications of starting materials used in this study.....	38
6 Perovskite phase, lattice parameter and average particle size of BNKFT- $x/0.03$ powders calcined at various temperatures.....	52
7 Perovskite phase, lattice parameter and average particle size of BNKFT- $0.18/y$ powders calcined at various temperatures.....	53
8 Average grain size, density and shrinkage of BNKFT ceramics at various temperatures.....	57
9 Average grain size, density, dielectric constant ( $\epsilon_r$ ), dielectric loss ( $\tan\delta$ ), remnant polarizations ( $P_r$ ) and coercive fields ( $E_c$ ) of BNKLT- $x/y$ ceramics.....	66
10 Perovskite phase, lattice parameter and average particle size of BNKLT- $x/0.10$ powders calcined at various temperatures.....	74
11 Perovskite phase, lattice parameter and average particle size of BNKLT- $x/0.10$ powders calcined at various temperatures.....	75
12 Perovskite phase, lattice parameter and average particle size of BNKLT- $0.20/y$ powders calcined at various temperatures.....	76
13 Perovskite phase, lattice parameter and average particle size of BNKLT- $0.20/y$ powders calcined at various temperatures.....	76

## LIST OF TABLES (CONT.)

<b>Table</b>		<b>Page</b>
14	Average grain size, $\rho$ , $\varepsilon_r$ , $\tan\delta$ , $T_d$ , $T_c$ and shrinkage of BNKLT-0.22/0.10 ceramics at various temperatures.....	80
15	Average grain size, $T_d$ , $T_m$ , $P_r$ , $E_c$ , $S_{Max}$ , $S_{Neg}$ , $S_{Max}/E_{Max}$ and $d_{33}$ of BNKLT- $x/y$ ceramics.....	93
16	Percent perovskite phase, lattice parameter $a$ , average particle size, average grain size, density and shrinkage of BNNKT.....	101
17	Average grain size, $T_d$ , $T_m$ , $P_r$ , $E_c$ , $\varepsilon_r$ , $S_{Max}$ , $S_{Neg}$ , $S_{Max}/E_{Max}$ and $d_{33}$ of BNNKT- $x/y$ ceramic.....	116

## LIST OF FIGURES

<b>Figures</b>		<b>Page</b>
1	The perovskite structure as an $\text{ABO}_3$ perovskite-type unit cell.....	5
2	Piezoelectric effects in ferroelectric ceramics (a) direct effect (b) converse effect.....	6
3	Interrelationship of piezoelectric and subgroups on the basis of symmetry.....	8
4	A ferroelectric hysteresis loop.....	9
5	Phase transition in a ferroelectric (a) first order and (b) second order.....	10
6	Antiferroelectric hysteresis loop.....	12
7	Paraelectric hysteresis loop.....	14
8	Phase diagram and piezoelectric properties for $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ taken from.....	15
9	Thermogravimetric of fuel.....	18
10	XRD patterns of the Ni-Zn ferrites with various organic fuels: (a) alanine (b) glycine (c) carbohydrazide (d) urea and (d) citric acid.....	19
11	TEM micrographs of (a) original magnification x120,000 of $\text{Pb}_{0.60}\text{Ba}_{0.40}\text{TiO}_3$ powder calcined at 1050 °C and (b) original magnification x100,000 of $\text{Pb}_{0.20}\text{Ba}_{0.80}\text{TiO}_3$ powder calcined at 1150 °C.....	20
12	FESEM micrograph of the BNT powders.....	22
13	Temperature dependences of dielectric constant ( $\epsilon_r$ ) and loss tangent ( $\tan\delta$ ) in temperature range from RT to 500 °C of BNT ceramics .....	23
14	FESEM micrograph of the BKT powders was studied By Hou et al.....	25

## LIST OF FIGURES (CONT.)

<b>Figures</b>		<b>Page</b>
15	FESEM micrograph of the BKT powders was studied by Lencka et al.....	25
16	Temperature dependence of dielectric constant $\varepsilon_r$ and loss $\tan\delta$ at various frequencies for $(K_{0.5}Bi_{0.5})TiO_3$ ceramics.....	26
17	Polarization of KBT measured at temperatures of (a) 100 °C, (b) 200 °C, (c) 240 °C and (d) 260 °C.....	26
18	Phase diagram of BNT-BT showing the MPB between the ferroelectric rhombohedral phase and the ferroelectric tetragonal phase, reproduced from Takenaka et al.....	27
19	Temperature dependence of dielectric constant ( $\varepsilon_r$ ) and dielectric loss ( $\tan\delta$ ) for $(1-x)NBT-xNN$ ceramics at 1, 10, 100 kHz with (a) $x = 0.02$ , (b) $x = 0.04$ , (c) $x = 0.06$ , and (d) $x = 0.08$ ...	28
20	The dielectric constant and $\tan\delta$ of BNT-ST (a) $x = 0.04$ , (b) $x = 0.08$ , (c) $x = 0.14$ , (d) $x = 0.20$ and (e) $x = 0.24$ .....	29
21	$T_d$ , $T_{R-T}$ and $T_m$ of BNT-ST ceramics.....	30
22	Density of BNT-BKT ceramics.....	30
23	The dielectric constant and dielectric loss of BNT-BKT ceramics: (a) 12 mol% of BKT (b) 16 mol% of BKT (c) 18 mol% of BKT (d) 20 mol% of BKT.....	31
24	Piezoelectric constant $d_{33}$ and planar electromechanical coupling factor $k_p$ of $(1-x-y)BNT-xBKT-yBF$ ceramics as a function of the amount of BKT and BF: (a) $x = 0.18$ and (b) $y = 0.03$ ...	33
25	The electromechanical coupling factor $k_p$ of $(1-x-y)BNT-$ $xBKT-yBF$ ceramics with $x = 0.18$ , $y = 0$ ; $x = 0.18$ , $y = 0.03$ and $x = 0.15$ , $y = 0.03$ .....	34

## LIST OF FIGURES (CONT.)

<b>Figures</b>		<b>Page</b>
26	Piezoelectric coefficient $d_{33}$ and planar coupling factor $k_p$ of (1-x-y)BNT-xBKT-0.30KNN at $x = 0.100.40$ .....	35
27	Bipolar strain vs electric field of 0.80BNT-0.20BKT0.01KNN ceramic at room temperature at 1 Hz.....	35
28	Variation of piezoelectric coefficient $d_{33}$ and planar electromechanical coupling factor $k_p$ with $x$ for the (1-x-y)BNT-xBKT-yBLT ceramics ( $y=0.05$ and $0.10$ ).....	36
29	Diagram of experimental procedure on sample characterization...	38
30	XRD patterns of (a) BNKFT-0.12/0.03 and (b) BNKFT- 0.18/0.01 powders calcined at various temperatures for 2 h: ( $\nabla K_4Ti_3O_8$ ).....	47
31	SEM images of BNKFT-0.12/0.03 calcined at (a) 600 °C, (b) 700 °C, (c) 800 °C and BNKFT-0.18/0.01 calcined at (d) 600 °C, (e) 700 °C, (f) 800 °.....	48
32	32X-ray diffraction patterns of (a) BNKFT-x/0.03 and (b) BNKFT-0.18/y calcined powders.....	49
33	SEM images of BNKFT-x/0.03 calcined powder with (a) $x=0.12$ , (b) $x=0.18$ , (c) $x=0.24$ and BNKFT-0.18/y calcined powder with (d) $y=0$ , (e) $y=0.03$ , (f) $y=0.07$ .....	50
34	TEM images of BNKFT-x/0.03 calcined powder with (a) $x=0.12$ , (b) $x=0.18$ , (c) $x=0.24$ and BNKFT-0.18/y calcined powder with (d) $y=0$ , (e) $y=0.03$ , (f) $y=0.07$ .....	51
35	X-ray diffraction patterns of BNKFT0.18/0.03 sintered ceramics at various temperatures in the $2\theta$ range of (a) $10^\circ$ - $60^\circ$ , (b) $39^\circ$ - $41^\circ$ and (c) $45^\circ$ - $48^\circ$ .....	55

## LIST OF FIGURES (CONT.)

<b>Figures</b>		<b>Page</b>
36	Surface morphologies and cross-sectional micrographs of the BNKFT0.18/0.03 sintered ceramics at various temperatures: (a) and (b) sintered at 950 °C, (c) and (d) sintered at 1000 °C, (e) and (f) sintered at 1050 °C, (g) and (h) sintered at 1075 °C.....	56
37	X-ray diffraction patterns of BNKFT-x/0.03 sintered ceramics in the 2θ rang of (a) 10° to 60°, (b) 39° to 41° and (c) 45° to 48°..	59
38	X-ray diffraction patterns of BNKFT-x/0.03 sintered ceramics in the 2θ rang of (a) 10° to 60°, (b) 39° to 41° and (c) 45° to 48°..	59
39	SEM images of BNKFT-x/y sintered ceramics with (a) $x=0.12$ , (b) $x=0.18$ , (c) $x=0.21$ , (d) $y=0$ , (e) $y=0.03$ and (f) $y=0.07$ .....	61
40	The temperature dependences of dielectric constant ( $\varepsilon_r$ ) of the (a) BNKFT-x/0.03 and (b) BNKFT-0.18/y ceramics.....	63
41	Ferroelectric hysteresis loops of (a) BNKFT-x/0.03 and (b) BNKFT-0.18/y ceramics.....	64
42	Piezoelectric properties of (a) BNKFT-x/0.03 and (b) BNKFT-0.18/y ceramics.....	65
43	XRD patterns of (a) BNKLT-0.18/0.10 and (b) BNKLT-0.20/0.05 powders calcined at various temperatures for 2 h: ( $\nabla$ $K_4Ti_3O_8$ ) and ( $\square$ $K_2Ti_6O_3$ ).....	72
44	SEM images of BNKLT-0.18/0.10 calcined at (a) 600 °C, (b) 700 °C, (c) 800 °C and BNKLT-0.20/0.05 calcined at (d) 600 °C, (e) 700 °C, (f) 800 °C.....	73
45	X-ray diffraction patterns of (a) BNKLT-x/0.10 and (b) BNKLT-0.20/ycalcined powders.....	74

## LIST OF FIGURES (CONT.)

Figures		Page
46	X-ray diffraction patterns of BNKLT-0.22/0.10 sintered ceramics in the $2\theta$ rang of (a) $10^\circ$ to $60^\circ$ , (b) $39^\circ$ to $41^\circ$ and (c) $45^\circ$ to $48^\circ$ .....	78
47	Surface morphologies micrographs of the BNKLT-0.22/0.10 sintered ceramics at various temperatures: (a) sintered at $950\text{ }^\circ\text{C}$ , (b) sintered at $1000\text{ }^\circ\text{C}$ , (c) sintered at $1025\text{ }^\circ\text{C}$ , and (d) sintered at $1050\text{ }^\circ\text{C}$ .....	81
48	The temperature dependences of dielectric constant ( $\varepsilon_r$ ) of the BNKLT-0.22/0.10 and ceramics at differences temperature...	81
49	X-ray diffraction patterns of BNKLT- $x$ /0.10 sintered ceramics at different $x$ content.....	82
50	X-ray diffraction patterns of BNKLT-0.20/ $y$ sintered ceramics at different $y$ content.....	82
51	SEM images of BNKLT- $x$ / $y$ sintered ceramics with (a) $x=0.18$ , (b) $x=0.22$ , (c) $x=0.26$ , (d) $y=0$ , (e) $y=0.07$ and (f) $y=0.12$ ....	83
52	SEM images of BNKLT- $x$ / $y$ thermally etched surfaces with (a) $x=0.18$ , (b) $x=0.22$ , (c) $x=0.26$ , (d) $y=0$ , (e) $y=0.07$ and (f) $y=0.10$ .....	83
53	Temperature dependence of dielectric properties in the BNKLT system: (a) BNKLT-0.18/0.10, (b) BNKLT-0.20/0.10, (c) BNKLT-0.22/0.10, (d) BNKLT-0.24/0.10 and (e) BNKLT-0.26/0.10.....	86
54	Temperature dependence of dielectric properties in the BNKLT system: (a) BNKLT-0.20/0, (b) BNKLT-0.20/0.03, (c) BNKLT-0.20/0.05, (d) BNKLT-0.20/0.07, (e) BNKLT-0.20/0.10 and (e) BNKLT-0.20/0.12.....	87

## LIST OF FIGURES (CONT.)

Figures		Page
55	Logarithm of $(1/\varepsilon - 1/\varepsilon_m)$ against logarithm of $(T - T_m)$ for the BNKLT system measured at 1 kHz (a) BNKLT-x/0.10 and (b) BNKLT-0.20/y.....	88
56	P-E hysteresis loop of (a) BNKLT-x/0.10 and (b) BNKLT-0.20/y ceramics measured at 0.1 Hz.....	90
57	Strain loop of (a) BNKLT-x/0.10 and (b) BNKLT-0.20/y ceramics measured at 0.1 Hz.....	91
58	XRD patterns of BNNKT0.20/0.03 powders calcined at various temperatures for 2 h: ( $\nabla$ $K_4Ti_3O_8$ ).....	99
59	X-ray diffraction patterns of BNKLT-0.20/0.03 sintered ceramics in the $2\theta$ range of (a) $10^\circ$ to $60^\circ$ , (b) $39^\circ$ to $41^\circ$ and (c) $45^\circ$ to $48^\circ$ .....	99
60	SEM images of BNNKT-0.20/0.03 calcined at (a) $600^\circ C$ , (b) $700^\circ C$ , (c) $800^\circ C$ and (d) $850^\circ C$ .....	100
61	Cross-sectional and surface morphologies micrographs of the BNKLT-0.20/0.10 sintered ceramics at various temperatures: (a) and (b) sintered at $950^\circ C$ , (c) and (d) sintered at $1000^\circ C$ , (e) and (f) sintered at $1025^\circ C$ , (g) and (h) sintered at $1050^\circ C$ .....	102
62	X-ray diffraction patterns of BNKKTx/0.03 sintered ceramics in the $2\theta$ range of (a) $10^\circ$ to $60^\circ$ , (b) $39^\circ$ to $41^\circ$ and (c) $45^\circ$ to $48^\circ$ .....	104
63	X-ray diffraction patterns of BNKKT0.20/y sintered ceramics in the $2\theta$ range of (a) $10^\circ$ to $60^\circ$ , (b) $39^\circ$ to $41^\circ$ and (c) $45^\circ$ to $48^\circ$ .....	105

## LIST OF FIGURES (CONT.)

<b>Figures</b>		<b>Page</b>
64	SEM images of BNKKT- $x$ /0.03 sintered ceramics with (a) $x=0.18$ , (b) $x=0.20$ , (c) $x=0.22$ , (d) $x=0.24$ , (e) $x=0.26$ and (f) $x=0.28$ .....	105
65	SEM images of BNKKT-0.20/ $y$ sintered ceramics with (a) $y=0$ , (b) $y=0.01$ , (c) $y=0.03$ , (d) $y=0.05$ and (e) $y=0.07$ .....	106
66	Temperature dependence of dielectric properties in the BNNKT system: (a) BNNKT-0.18/0.03, (b) BNNKT-0.20/0.03, (c) BNNKT-0.22/0.03, (d) BNNKT-0.24/0.03, (e) BNNKT- 0.26/0.03 and BNNKT-0.28/0.03.....	109
67	Temperature dependence of dielectric properties in the BNNKT system: (a) BNNKT-0.20/0, (b) BNNKT-0.20/0.01, (c) BNNKT-0.20/0.03, (d) BNNKT-0.20/0.05 and (e) BNNKT-0.20/0.07.....	110
68	Logarithm of $(1/\varepsilon - 1/\varepsilon_m)$ against logarithm of $(T - T_m)$ for the BNNKT system measured at 1 kHz (a) BNNKT- $x$ /0.01 and (b) BNKLT-0.20/ $y$ .....	111
69	$S_{max}$ (%) and $d_{33}^*$ (pm/V) values plotted on the BNNKT phase diagram.....	113
70	$P$ - $E$ hysteresis loop of (a) BNKKT- $x$ /0.10 and (b) BNKKT- 0.20/ $y$ ceramics measured at 0.1 Hz.....	114
71	Strain loop of (a) BNKLT- $x$ /0.10 and (b) BNKLT-0.20/ $y$ ceramics measured at 0.1 Hz.....	115