CHAPTER I

INTRODUCTION

Overview

As is well known, piezoelectric materials have wide applications in electronic and microelectric devices. The most widely used piezoelectric materials are $PbZrO_3$-$PbTiO_3$ (PZT) based ceramics. However, in the process of obtaining this material, it is necessary to maintain the atmosphere with enriched vapor of lead oxide (PbO), which is opposite to the needs of environmental protection because of the toxicity. Furthermore, many countries have restricted the use of lead oxide by drafting legislation. Consequently, it is necessary and urgent to search for lead-free piezoelectric ceramics with excellent piezoelectric properties [1].

$Bi_{0.5}Na_{0.5}TiO_3$ (abbreviate to BNT), discovered by Smolenskii et al. in 1960 [1, 2], is an ABO_3 type ferroelectric with a perovskite phase. BNT is considered to be a promising candidate for lead-free piezoelectric ceramics with a relatively large remnant polarization (P_r = 38 $\mu C/cm^2$) and a high Curie temperature (T_c = 320 °C) [3, 4]. However, pure BNT ceramics have a high coercive field (E_c = 7.3 kV/mm), making the poling of the ceramics extremely difficult. In addition, pure BNT ceramic usually exhibits very poor piezoelectricity (d_{33} = 58 pC/N). To decrease the coercive field and improve the piezoelectric properties, a number of solid solutions of BNT with ABO_3-type ferroelectrics or non-ferroelectrics, such as BNT-$BaTiO_3$ [5], BNT-$NaNbO_3$ [6], BNT-$CaTiO_3$ [7], BNT-$SrTiO_3$ [7], BNT-$Bi_{0.5}K_{0.5}TiO_3$ [1] have been studied extensively. Xinyou et al. [5] reported that the dielectric constant (ε_r), the loss tangent (tanδ) and the piezoelectric constant (d_{33}) of 0.94 BNT-0.06 $BaTiO_3$ ceramics are 1650, 0.048 and 120 pC/N. In the case of 0.98 BNT-0.02 $CaTiO_3$, 0.80 BNT-0.20 $SrTiO_3$ and 0.98 BNT-0.02 $NaNbO_3$, the d_{33} values were 75 [7], 133 [7] and 88 pC/N [6]. 0.82 BNT-0.18 $Bi_{0.5}K_{0.5}TiO_3$ (BNT-BKT) ceramics demonstrated the ε_r, tanδ, d_{33}, k_p and E_c of 893, 0.037, 144 pC/N, 29.0 % and 4 kV/mm [1]. Among these compositions, BNT-BKT ceramics have exhibited excellent piezoelectric properties, but are still far from satisfactory in terms of practical applications.

The ternary system has demonstrated thata favorable design can improve piezoelectric and ferroelectric properties [8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19]. The ternary system of BNT-BKT with $BaTiO_3$ (BT) [10, 11, 12], $LiNbO_3$ [13], $(K_{0.5}Na_{0.5})NbO_3$ (KNN) [14], $(Bi_{0.5}Li_{0.5})TiO_3$ (BLT) [15, 16], and $BiFeO_3$ (BF) [17, 18, 19] were intensively investigated. Among the solid solutions that have been developed so far, $(1-x-y)Bi_{0.5}Na_{0.5}TiO_3$–$xBi_{0.5}K_{0.5}TiO_3$–$yBiFeO_3$ (BNKFT), $(1-x-y)Bi_{0.5}Na_{0.5}TiO_3$–$xBi_{0.5}K_{0.5}TiO_3$–$y(Na_{0.5}K_{0.5})NbO_3$ (BNKNT) and $(1-x-y)Bi_{0.5}Na_{0.5}TiO_3$–$xBi_{0.5}K_{0.5}TiO_3$–$yBi_{0.5}Li_{0.5}TiO_3$ (BNKLT) system has attracted considerable attention[10, 11, 12, 13, 14, 15, 16, 17, 18]. Zhou et al. [18] reported that the optimum values of d_{33} and k_p are 170 pC/N and 36.6 % obtained from $0.79Bi_{0.5}Na_{0.5}TiO_3$-$0.18Bi_{0.5}K_{0.5}TiO_3$-$0.03BiFeO_3$ ceramics. Yang et al. [15] found that doping BLT could reduce the coercive field and improve the piezoelectric properties of BNT-BKT ceramics as follows: E_c=3.24 kV/mm, d_{33}=203 pC/N and k_p=31.0% with a composition of $0.68Bi_{0.5}Na_{0.5}TiO_3$-$0.22Bi_{0.5}K_{0.5}TiO_3$-$0.10Bi_{0.5}Li_{0.5}TiO_3$ ceramic. Singh et al. [14] revealed that the $0.79Bi_{0.5}Na_{0.5}TiO_3$-$0.20Bi_{0.5}K_{0.5}TiO_3$-$0.01K_{0.5}Na_{0.5}NbO_3$ ceramic has the largest strain ever reported for a polycrystalline lead-free ceramics (~0.80%), which is even higher than the strains obtained in Pb-based antiferroelectrics.

BNKFT, BNKNT and BNKLT have been prepared by the solid state reaction method, at a calcination stage above 900 °C and a sintering stage above 1150 °C for 3-5 h [14, 15, 18]. Although the solid-state reaction method is relatively simple, it is time consuming, energy intensive and a poor quality ceramic is obtained. Recently, our previous works have successfully fabricated high quality different oxides ceramics such as: $(Ba_{1-x}Sr_x)(Zr_xTi_{1-x})O_3$ [20], $Ba(Ti_{1-x}Zr_x)O_3$ [21], $(Pb_{1-x}Ba_xTiO_3)$ [22], $BaZrO_3$ [23] using the combustion technique. The advantages of this technique include inexpensive precursors, simple preparation process, and resulting good electrical properties with lower firing temperatures and a shorter dwell time [24, 25, 26]. Furthermore, from a survey of the literature, BNKFT, BNKNT and BNKLT ceramics, prepared by the combustion method, have not been studied. Thus, in this work, $(1-x-y)Bi_{0.5}Na_{0.5}TiO_3$–$xBi_{0.5}K_{0.5}TiO_3$–$yBiFeO_3$ (x=0.12-0.24, y=0-0.07),

$(1\text{-}x\text{-}y)Bi_{0.5}Na_{0.5}TiO_3$–$xBi_{0.5}K_{0.5}TiO_3$–$y(Na_{0.5}K_{0.5})NbO_3$ (x=0.18-0.28, y=0-0.07) and $(1\text{-}x\text{-}y)Bi_{0.5}Na_{0.5}TiO_3$–$xBi_{0.5}K_{0.5}TiO_3$–$yBi_{0.5}Li_{0.5}TiO_3$ (x=0.18-0.26, y=0-0.12) ceramics were prepared by the combustion method. The effects of firing temperatures and the change of the x and y content on the phase formation, microstructure, and electrical properties of ceramics were investigated.

Objectives of this research

The objectives of this research are as follow:

1. To prepare BNKFT, BNKNT and BNKLT ceramics ternary system using the combustion technique.

2. To investigated the optimum calcination and sintering conditions of BNKFT, BNKNT and BNKLT ceramics for combustion preparation.

3. To study the effect of calcination and sintering temperatures on the crystal structure, microstructure, electrical properties of BNKFT, BNKNT and BNKLT ceramics prepared via the combustion technique.

4. To study the effect of $BiFeO_3$, $(Na_{0.5}K_{0.5})NbO_3$ and $(Bi_{0.5}Li_{0.5}Ti)O_3$ co-doped on the properties of $Bi_{0.5}Na_{0.5}TiO_3$–$Bi_{0.5}K_{0.5}TiO_3$ based ceramics and special significance was focused on seeking the MPB composition.