



Effect of Li_2CO_3 addition in $\text{BiFeO}_3\text{-BaTiO}_3$ ceramics on the sintering temperature, electrical properties and phase transition



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ABSTRACT

In this work, we reported that a $\text{BiFeO}_3\text{-BaTiO}_3$ (BF-BT) ceramics with Li_2CO_3 as a sintering aid was fabricated in order to develop a new materials sintered at low-temperature for piezoelectric applications. The phase transition, microstructure, dielectric, piezoelectric and ferroelectric properties, and temperature stability of the ceramics were investigated. The results show that the addition of Li_2CO_3 improves the electrical properties of BF-BT ceramics and decreases the sintering temperature from 990 °C to 930 °C. The obtained Li_2CO_3 -doped ceramics sintered at 930 °C when $x = 0.3$ mol% have an optimized electrical properties, which were listed as follows: ($k_p = 0.294$, $Q_m = 38.33$, $\tan\delta = 0.043$ and $d_{33} = 163$ pC/N), high Curie temperatures ($T_c = 508$ °C) and high depolarization temperature ($T_d = 480$ °C). These results indicate that this material is a promising candidate for high-temperature lead-free piezoelectric applications.

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1. Introduction

High Curie temperatures (T_c) piezoelectric ceramics were widely used in petrochemical, automotive, and aerospace industries. However, the serious depoling and aging problems of commercial Lead zirconate-titanate ceramics (PZT) hindered its application at temperature above 200 °C. In general, their operation temperature was limited to one half of the T_c value due to thermal depolarization and increased conductivity at high temperatures [1]. The T_c of the commercial PZT piezoelectric ceramics varies between 180 °C and 386 °C, which depends on the composition of the ceramics. Furthermore, the PZT ceramics contains more than 70% toxic lead and could cause serious damage to people's health. Legislation governing the use of lead containing compounds has been enforced in many countries. Therefore, it is necessary to develop lead-free piezoelectric materials with high Curie temperature, high piezoelectric strain coefficients and thermal stability to replace the lead-based ceramics. Recently, $\text{BiFeO}_3\text{-BaTiO}_3$ (BF-BT) ceramics has received considerable attention due to its high Curie temperature and depoling temperature ($T_c > 500$ °C and $T_d > 400$ °C) [2]. In our previous research, we also found that Mn- and Cu-modified BF-BT ceramics showed excellent piezoelectric and dielectric properties

[3]. However, Bi is a volatile constituent at elevated temperature due to its high saturation vapor pressure. The BF-BT based ceramics have a high sintering temperature that is above 980 °C, which could induce the evaporation of Bi element during the sintering process [4], and lead to the deviation from the optimally designed composition [5,6]. At the same time, high temperature sintering process leads to high dielectric losses and high conductivity. Excess Bi element were added to compensate the volatilization during the high temperature sintering process, and ceramics with improved piezoelectric and dielectric properties were obtained [4]. However, the researches on reducing the volatilization of Bi element by decreasing the sintering temperature has not been reported systematically to this day. Therefore, low temperature sintering of materials with excellent piezoelectric properties were desired. Furthermore, low-temperature sintering has many other advantages such as the compatibility with low temperature co-fired ceramics (LTCCs) and less energy consumption. Therefore, a BF-BT based piezoelectric ceramics that can be successfully sintered at a lower sintering temperature is attractive for many applications. Previously we found that the addition CuO , B_2O_3 and MgO can reduce the sintering temperature [7–9]. It was reported by Yang [10] that the sintering temperature had a significant effect on the piezoelectric properties of $(1-x)\text{BiFeO}_3\text{-xBaTiO}_3$ ceramics. The Hee-Wook You reported that they reduced the sintering temperature from 1350 °C to around 900 °C by add 1~5 wt % Li_2CO_3 into BaSrTiO_3 materials [11]. The Li_2CO_3 and MgO was also added into $(\text{Ba,Sr})\text{TiO}_3$

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material to reduce the sintering temperature and improve dielectric properties such as loss tangent, and frequency dispersion [12]. In this work, we reported that a $\text{BiFeO}_3\text{-BaTiO}_3$ (BF-BT) ceramic with Li_2CO_3 as a sintering aid was fabricated in order to develop a new materials sintered at low-temperature for piezoelectric applications. The effect of the Li_2CO_3 addition on the phase transition, microstructure, dielectric, piezoelectric and ferroelectric properties, and temperature stability of the ceramics were also reported.

2. Experimental procedures

$0.725\text{BiFeO}_3\text{-}0.275\text{BaTiO}_3 + 0.35\text{molMnO}_2 + x\text{Li}_2\text{CO}_3$ (BF-BT-xLC) ceramics, where x is the fraction of Li_2CO_3 , ranging from 0 to 0.5 mol%, were prepared by solid state reaction method. Starting materials of commercial reagent grade metal oxide and carbonate powders of Bi_2O_3 (99.99%), TiO_2 (99.99%), Fe_2O_3 (99.99%), BaCO_3 (99.99%), MnO_2 (99.99%) and Li_2CO_3 (99.99%) were weighed according to the compositions. The oxide powders were batched in stoichiometric proportions and mixed by ball milling for 24 h in absolute ethyl alcohol (99.7%). The mixtures were calcined at 780°C for 4 h and then different amount of Li_2CO_3 were added into the calcined powders and milled again in alcohol suspensions. After that, the powders were pressed into pellets with a diameter of 11 mm and thickness of 1.5 mm under 150 MPa. Following removal of binder at 600°C for 4 h, green compacts were sintered in air at $890^\circ\text{C}\text{-}990^\circ\text{C}$ for 2 h. The crystalline phase of the sintered pellets was identified by X-ray diffraction (XRD) technique using $\text{Cu K}\alpha$ radiation (XRD, D8-2-Advanced, Bruker AXS, Germany). Surface microstructure was examined by scanning electron microscopy (SEM, JSM-5610LV, JEOL, Japan). The sintered pellets were polished down to 1.2 mm, and were electroded with a post fire silver paste. For electric measurements, BF-BT-xLC ceramics were poled in an oil bath under a field of 5 kV/mm at 100°C for 15 min. Poled samples were aged for 24 h before conducting any electrical measurement. Dielectric properties as a function of temperature were measured using a computer controlled precision LCR meter (4294A, Agilent). The $P\text{-}E$ hysteresis loops and piezoelectric strains were measured by a ferroelectric tester (Radiant P-PMF.USA). The piezoelectric constant d_{33} was measured using a quasi static piezoelectric meter (ZJ-3A, CAS Shanghai China). The electromechanical coupling factor k_p and the mechanical quality factor Q_m was calculated based on the resonate method using an impedance analyzer (4294A, Agilent). The temperature dependent electromechanical coupling factor k_p were determined by attaching the poled specimens to a conductive jig placed in a furnace. The poled pellets were supported only by the silver wires fired on both surfaces.

3. Result and discussion

Fig. 1 shows the XRD pattern of all specimens sintered at 930°C . As shown in Fig. 1(a), a single perovskite phase without extra diffraction peaks can be observed for the systems, indicating that a solid solution is formed. However, a small amount of miscellaneous peak (represents the heterogeneous $\text{Bi}_2\text{Fe}_4\text{O}_9$) can be also observed. With increasing the Li_2CO_3 content, the hybrid peaks disappear gradually. According to the XRD patterns in Fig. 1(b), the number of the XRD between 38° and 40° changed from one to two peaks and that between 44° and 46° remains a single peak, with increasing the x value. With increasing of Li_2CO_3 content, the single (111) peak splits to (003) and (021) peaks. The lattice parameters for BF-BT-xLC are listed in Table 1. The lattice parameter (a , b , c) decreases firstly and then increases with the increasing of x value. The crystal structure changed from tetragonal to rhombohedral phase. When $x = 0, 0.001, 0.002$, the crystal cell parameters are $a = b = 3.98740$, $c = 3.97389$, $\alpha = \beta = \gamma = 90^\circ$ and $a = b = 3.98060$, $c = 4.00270$,

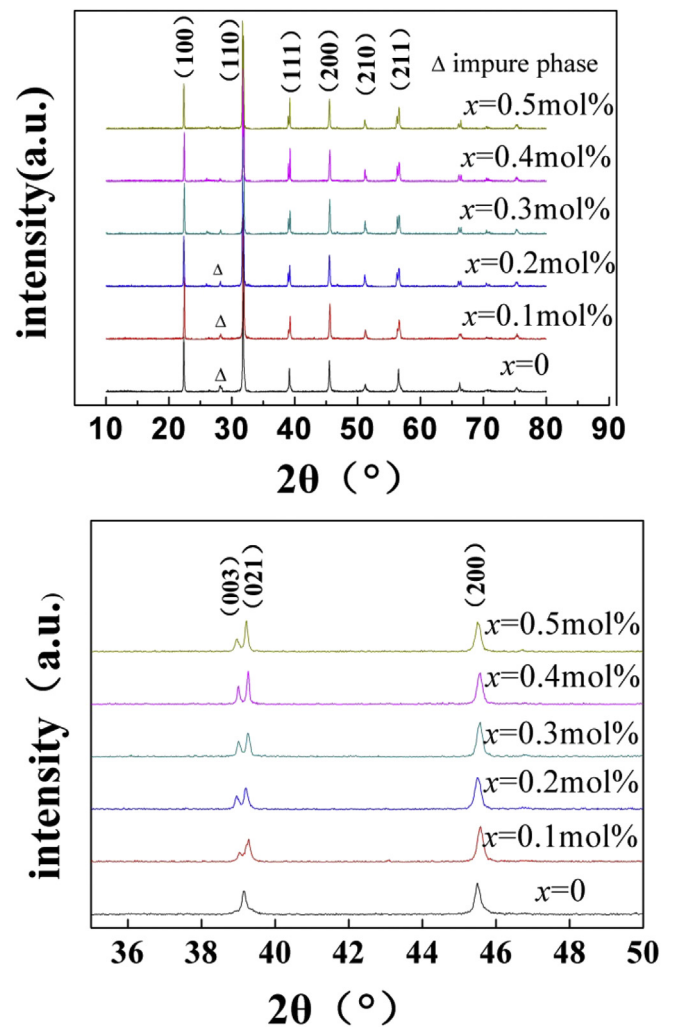


Fig. 1. XRD pattern of specimens sintered at 930°C as a function of Li_2CO_3 content.

Table 1
The lattice parameter for BF-BT-xLC.

x	$a(\text{\AA})$	$b(\text{\AA})$	$c(\text{\AA})$	$\alpha(^{\circ})$	$\beta(^{\circ})$	$\gamma(^{\circ})$
0	3.9874(1)	3.9874(1)	3.9738(9)	90	90	90
0.001	3.9806(1)	3.9806(1)	4.0024(7)	90	90	90
0.002	3.9860(4)	3.9860(4)	3.9860(4)	89.7234	89.7234	89.7234
0.003	3.9878(7)	3.9878(7)	3.9878(7)	89.8018	89.8018	89.8018
0.004	3.9974(3)	3.9974(3)	3.9974(3)	89.7582	89.7582	89.7582
0.005	4.0039(1)	4.0039(1)	4.0039(1)	89.6597	89.6597	89.6597

$\alpha = \beta = \gamma = 90^\circ$ and $a = b = c = 3.98604$, $\alpha = \beta = \gamma = 89.7234^\circ$, respectively. It is showed Li^+ may incorporated into the BF-BT, resulting in lattice distortion which changes lattice parameters. On the other hand, the results show that the lattice constant increases first and then decreases with the increase of Sc doping content in $0.725\text{BiFe}_{1-x}\text{Sc}_x\text{O}_3\text{-}0.275\text{BaTiO}_3$, and maintains a small fluctuation [13]. The sample has still a crystalline structure with a tetragonal phase at $x = 0.1$ mol%. The tetragonal phase structure had a tendency to transform into a rhombohedral structure ($R3c$) when $x > 0.1$ mol%. The rhombohedral phase can be obtained when Li_2CO_3 content is above 0.2 mol%. The polymorphic transition appears at room temperature for the composition around 0.1 mol% $< x < 0.2$ mol%, in which the tetragonal and rhombohedral phases coexist. It was reported by Chandarak et al. [14] that the

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