ARTICLE IN PRESS

Available online at www.sciencedirect.com

ScienceDirect

Ceramics International ▮ (▮▮▮▮) ▮▮▮–▮▮▮

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

A lead-reduced ferrolectric solid solution with high curie temperature: $BiScO_3$–$Pb(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$

Dongfang Pang[a,b], Xifa Long[a,*], Hamel Tailor[c]

[a]*Key Laboratory of Optoelectronic Materials Chemistry and Physics, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, Fujian 350002, China*

[b]*University of Chinese Academy of Sciences, Beijing 100049, China*

[c]*Department of Chemistry and 4D LABS, Simon Fraser University, Burnaby, British Columbia, Canada V5A 1S6*

Received 25 March 2014; received in revised form 28 April 2014; accepted 28 April 2014

Abstract

A lead-reduced ternary ferroelectric solid solution system of $BiScO_3$–$Pb(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (BS–PZN–PT) in the form of the ceramic was investigated for its structure and properties in order to develop novel ferroelectric materials with high Curie temperature and good piezoelectric properties. A morphotropic phase boundary region was delimited in this system between rhombohedral and tetragonal phases. The Curie temperature T_C of this ternary system varied from 321 °C to 131 °C and the coercive fields E_C from 24.6 kV/cm to 8.62 kV/cm. The piezoelectric coefficient d_{33} varies in the range of 100–320 pC/N with increasing PZN content. Those compositions within the MPB region possess good comprehensive piezoelectric properties, for example, d_{33} and T_C of compositions with 0.32BS–0.15PZN–0.53PT, 0.34BS–0.15PZN–0.51PT and 0.28BS–0.30PZN–0.42PT are 313 pC/N and 319 °C, 300 pC/N and 304 °C, 320 pC/N, and 236 °C, respectively. The high T_C and d_{33} make it a promising material for high-powder ultrasound transducers for use over a wide temperature range.

Keywords: C. Ferroelectric properties; C. Piezoelectric properties; D. Perovskites; D. Traditional ceramics

1. Introduction

Lead-based perovskite-type oxide ferroelectric materials are used in a wide range of application that are of great importance in various industrial and scientific areas such as in sensors, actuators, and ultrasonic transducers [1,2]. However issues such as the environmental restrictions on the usage of lead and the depoling and aging of $PbZrO_3$–$PbTiO_3$ (PZT) piezoelectric materials at temperatures above 200 °C have attracted considerable attention to search for alternative materials [3,4]. $BiMeO_3$–$PbTiO_3$ (Me: Fe^{3+}, Sc^{3+}, In^{3+}, Y^{3+}, Yb^{3+}, Ga^{3+}, etc.) ceramics have been found to exhibit a significantly higher Curie temperature than those of Pb $(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (PZNT, T_C ~70 °C), PZT(T_C ~360 °C), and $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (PMNT, T_C ~165 °C) systems [3,5–8]. Therefore, $BiMeO_3$ addition into lead-based perovskite solid solutions can not only reduce the content of lead to lower pollution, but also increase the number of possible application by increasing the maximum operating temperature [3,9–12]. Many $BiMeO_3$–$PbTiO_3$ systems have been studied extensively but the majority have shown either limited solid solubility, such as $BiInO_3$–$PbTiO_3$, $BiGaO_3$–$PbTiO_3$ or display inferior properties such as $Bi(Mg_{1/2}Ti_{1/2})O_3$–$PbTiO_3$, $Bi(Mg_{1/2}Zr_{1/2})O_3$–$PbTiO_3$ and $BiFeO_3$–$PbTiO_3$ [11–16]. Among them, $BiScO_3$–$PbTiO_3$ (BS–PT) was discovered to possess high Curie temperature of 460 °C [7], which possesses better properties than other members, for example, the piezoelectric coefficient of BS–PT reaches values up to 200 pC/N [17]. If $BiScO_3$ is introduced into the binary systems such as PMNT or PZNT, it is believed that these systems will possess high Curie temperature and good piezoelectric properties with the less lead content. In actuality, the investigations on the ternary systems of $BiMeO_3$–$PbTiO_3$ and a classic relaxor as an end-member have been reported and it is found that these systems show relaxor ferroelectricity with high Curie temperature [18]. Craig et al. reported that the BS–PMN–PT system exhibits relaxor properties of T_{max} ~250–350 °C and ε_{max} ~10,000–24,000 at

*Corresponding author. Tel./fax: 86 591 83710369.
E-mail address: lxf@fjirsm.ac.cn (X. Long).

http://dx.doi.org/10.1016/j.ceramint.2014.04.156
0272-8842/

Please cite this article as: D. Pang, et al., A lead-reduced ferrolectric solid solution with high curie temperature: $BiScO_3$–$Pb(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$, Ceramics International (2014), http://dx.doi.org/10.1016/j.ceramint.2014.04.156

1 KHz [3,19]. Moreover, some ternary systems containing BS–PT such as BS–PZ–PT [20], and $BiScO_3$–$PbTiO_3$–$Pb(Cd_{1/3}Nb_{2/3})O_3$ [7] has also been reported. Although PZNT exhibits excellent piezoelectric and electromechanical properties, the reports about BS–PZN–PT system are quite few due to the difficulty of the preparing samples. Up to now, only few compositions of the ternary system BS–PZN–PT have been investigated showing both high Curie temperature and good piezoelectric properties near MPB region [21,22]. In the paper, we report the preparation, structure, phase diagram and electric properties of the ternary BS-PZN-PT ceramics.

2. Experimental procedure

The ceramic samples of $(1-x-y)BiScO_3$–$xPb(Zn_{1/3}Nb_{2/3})O_3$–$yPbTiO_3$ with compositions of x=0.15, y=0.47, 0.49, 0.51, 0.53, 0.55; x=0.3, y=0.48, 0.46, 0.44, 0.42, 0.40 and x=0.38, y=0.35, 0.37, 0.39, 0.41, 0.43 were prepared by conventional mixed oxide ceramic processing. Ceramic samples were fabricated using metal oxide powders with 99.9% purity. PbO, Bi_2O_3, Sc_2O_3, TiO_2, ZnO, Nb_2O_5. PbO and Bi_2O_3 were weighed with 2 mol% excess of PbO to compensate for evaporation during sintering. At first, ZnO and Nb_2O_5 powders were mixed and milled with 5% excess of ZnO for 2 h. Then these metal oxide powders and the milled powder were mixed in the stoichiometric amount and milled using ethanol for 1 h. After milling, the powders were pressed into pellets at 4 MPa. The pressed powders were calcined at 900 °C for 6 h in air. After calcination, the pieces were remilled using ethanol for 1 h and mixed with 2 wt% polyvinyl alcohol (PVA) as a binder. The powders were pressed again at 7 MPa, and then heated up to 500 °C for 2 h to eliminate the binder. Finally, the

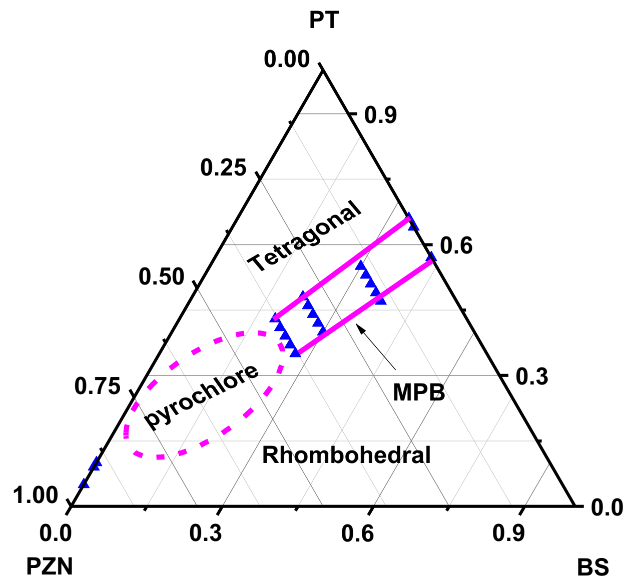

Fig. 2. Phase region of $(1-x-y)$BS–xPZN–yPT ternary system and MPB region.

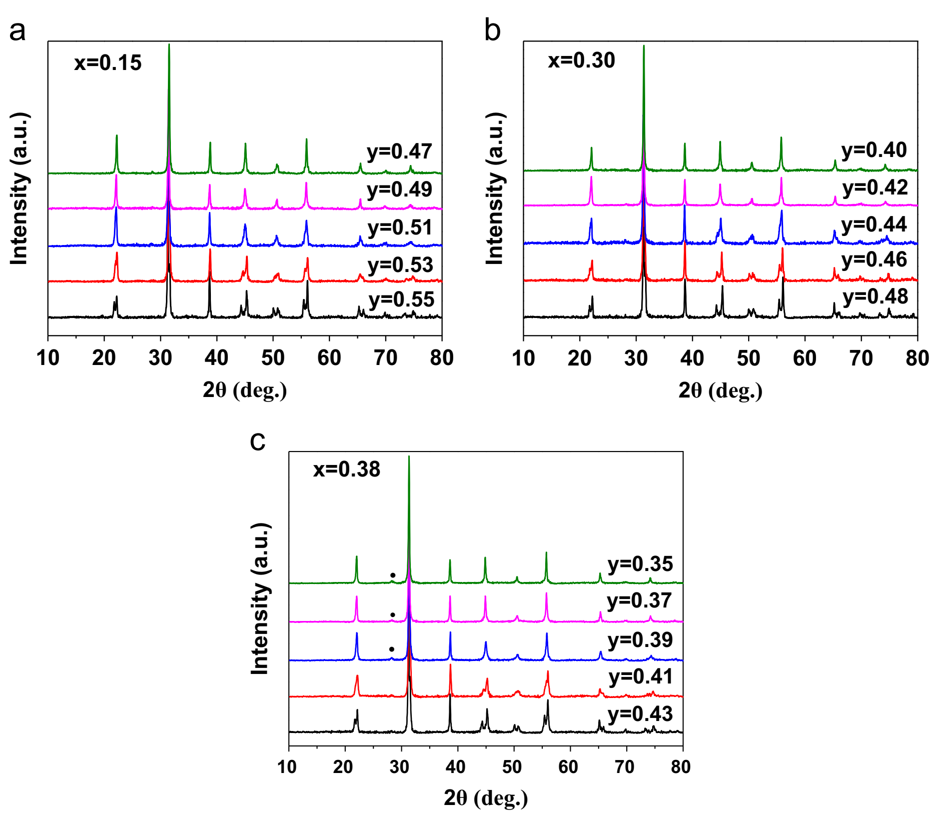

Fig. 1. X-ray powder diffraction patterns of the $(1-x-y)$BS–xPZN–yPT ceramics: $x=0.15$, $x=0.30$, and $x=0.38$, indicating a phase transition from the rhombohedral to tetragonal phase.

Please cite this article as: D. Pang, et al., A lead-reduced ferrolectric solid solution with high curie temperature: $BiScO_3$–$Pb(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$, Ceramics International (2014), http://dx.doi.org/10.1016/j.ceramint.2014.04.156