# Preparation and Electrical Properties of (1-x)$LiNbO_3$-x$BiYbO_3$ Ceramics

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**Abstract.** A series of (1-$x$)$LiNbO_3$-$x$$BiYbO_3$ ($x$ = 0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.06) lead-free piezoelectric ceramics were prepared by conventional solid-state reaction. The effect of $BiYbO_3$ content in $LiNbO_3$ system on crystal structure, microstructure, dielectric and piezoelectric properties have been investigated. The results indicate that the ceramics of $x$ = 0.01 and $x$ = 0.02 can form perovskite single phase. When the $x$ is more than 0.03, a little impure phases, such as $Nb_2O_5$ and $Bi_2O_3$, begin to appear. All the studied ceramics show compact microstructures with the relatively density of above 97%. With the trace of $BiYbO_3$ addition, the grain size decreases slightly, and the resonance peak of permittivity moves toward high frequency. The amount of $BiYbO_3$ seems to have little influence on the piezoelectric constant $d_{33}$, but affect the real part of permittivity ε′ of the ceramics, which attain maximum values at $x$ = 0.06.

## Introduction

Since the vast majority of the piezoelectric ceramics used are lead at the moment, which seriously damage both environment and human health during the processes of preparation, use and disposal treatment, scholars in different countries have paid great attention to the research and development of lead-free piezoelectric ceramics [1-4]. Lithium niobate is an important piezo-ferroelectric material, with hexagonal structures and point groups of R3C at room temperature, and the ferroelectric-paraelectric phase transition occurs at 1,200 ℃. Now, the lithium niobate is widely used in photoconduction, light modulation, nonvolatile memory and second harmonic generator, becoming one of preferred materials of electro-optical devices and surface acoustic wave devices, because of high Curie temperature, great spontaneous polarization, high electromechanical coupling coefficient, high mechanical quality factor and excellent characteristics of electro-optics, acousto-optics, photo refraction and nonlinear optical property [5]. However, lithium niobate piezoelectric ceramic is a single-component crystal system with a very narrow sintering temperature range, and it is hard to sinter; meanwhile, since alkali metal ions are partially vaporized during the sintering process, it is hard to obtain homogeneous solid solution. With regard to this, adding other components to form dual- or multi-component solid solution ceramics systems has been a research highlight [6, 7]. This article uses $LiNbO_3$ as a basic research component and adds $BiYbO_3$ to form a new perovskite-type (1-$x$) $LiNbO_3$-$x$ $BiYbO_3$ ($x$ = 0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.06) lead-free ceramic system by conventional solid-state reaction method, with the aim of investigating the influences of $BiYbO_3$ on crystal structure, microstructures, dielectric as well as piezoelectric properties of the ceramics.

## Experimental Procedures

All chemicals are of analytical grade and used as received without further purification. Ceramics with compositions (1-$x$)$LiNbO_3$-$x$$BiYbO_3$ were prepared via a conventional solid-state reaction method. First, stoichimetric $Nb_2O_5$ and $Li_2CO_3$ were mixed according to $LiNbO_3$ and then ball-milled in alcohol for 24 h. After drying at 70 ℃, the mixed powders were calcined at 800 ℃ for 3 h in air to get $LiNbO_3$ powders. Then, materials $LiNbO_3$, $Bi_2O_3$ and $Al_2O_3$ were weighted according to (1-*x*)$LiNbO_3$-*x*$BiYbO_3$ with the *x* of 0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.06, mixed in alcohol and milled for 24 h, followed by drying at 70 ℃ and calcining at 800 ℃ for 3 h in air. The powders were finally ground and mixed with 5 wt% polyvinyl alcohol (PVA) binders, then pressed into cylindrical pellets with a diameter of 10 mm and thickness of about 2 mm under a pressure of 4 MPa. After burning off the PVA at 500 ℃ for 2 h, the disk pellets were sintered at 1000 ℃ for 2 h in air. The surfaces of the sintered ceramics were polished and coated with silver paste to act as the electrodes. To measure the relevant piezoelectric properties, the prepared samples were poled in silicone oil at room temperature under 2 KV/mm for 10 min.

The phase composition of samples were determined using an X-ray diffractometer (XRD, DX-2500, Dandongfangyuan Co. Ltd., China) with Cu Kα radiation operated at 30 kV and 25 mA in a 2θ range of 20-70$^o$. A scanning electron microscope (SEM, JSM-6460, JEOL, Tokyo, Japan) was used to observe the microstructures of (1-*x*)$LiNbO_3$-*x*$BiYbO_3$ ceramics. The samples were aged at room temperature for 24 h after poling to measure the piezoelectric constant $d_{33}$,using a quasi-static $d_{33}$ meter (ZJ-6 A, Beijing, China). The dielectric properties were measured using an impedance analyzer (E4991, Agilent, USA) in the frequency range from 1 MHz to 1 GHz at room temperature.

## Results and Discussion

### XRD Characterization

XRD patterns of (1-*x*)$LiNbO_3$-*x*$BiYbO_3$ (*x* = 0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.06) ceramics prepared by solid-state reaction are shown in Fig. 1. No impurity phase was found in samples *x* = 0.01 and *x* = 0.02, which means pure (1-*x*)$LiNbO_3$-*x*$BiYbO_3$ ceramics can be obtained by solid-state reaction method. However, with the increasing of doping levels, the second phase such as $Nb_2O_5$ or $Bi_2O_3$, is gradually increased. The results demonstrate that the addition of $BiYbO_3$ did not cause a remarkable change in $LiNbO_3$ crystal structure.

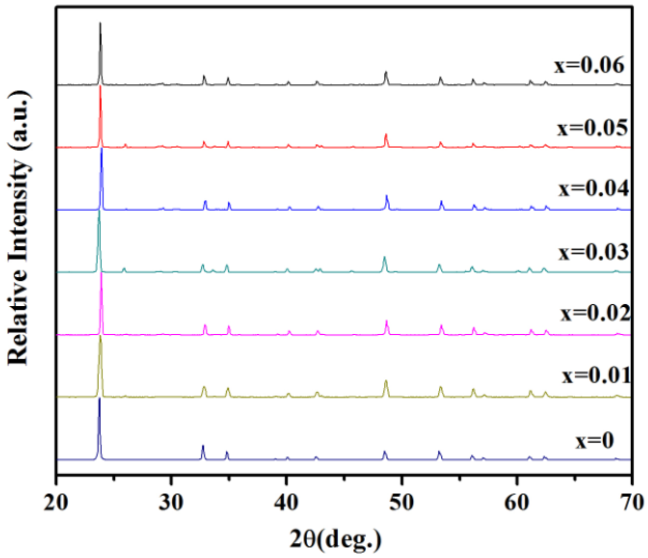


Fig. 1 XRD patterns of (1-*x*)$LiNbO_3$-*x*$BiYbO_3$ (*x* = 0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.06) ceramics prepared by solid-state reaction

### Microstructures

Fig.2 shows the typical SEM photographs of (1-*x*)$LiNbO_3$-*x*$BiYbO_3$ (*x* = 0, 0.01, 0.02, 0.03, 0.04, 0.05) ceramics. It can be observed that all the ceramic samples show compact and uniform microstructures, and the grain boundary is very clear. Moreover, with the addition of $BiYbO_3$, the grain size of the ceramics decrease slightly. So the additon of $BiYbO_3$ acts as a grain growth inhibitor.

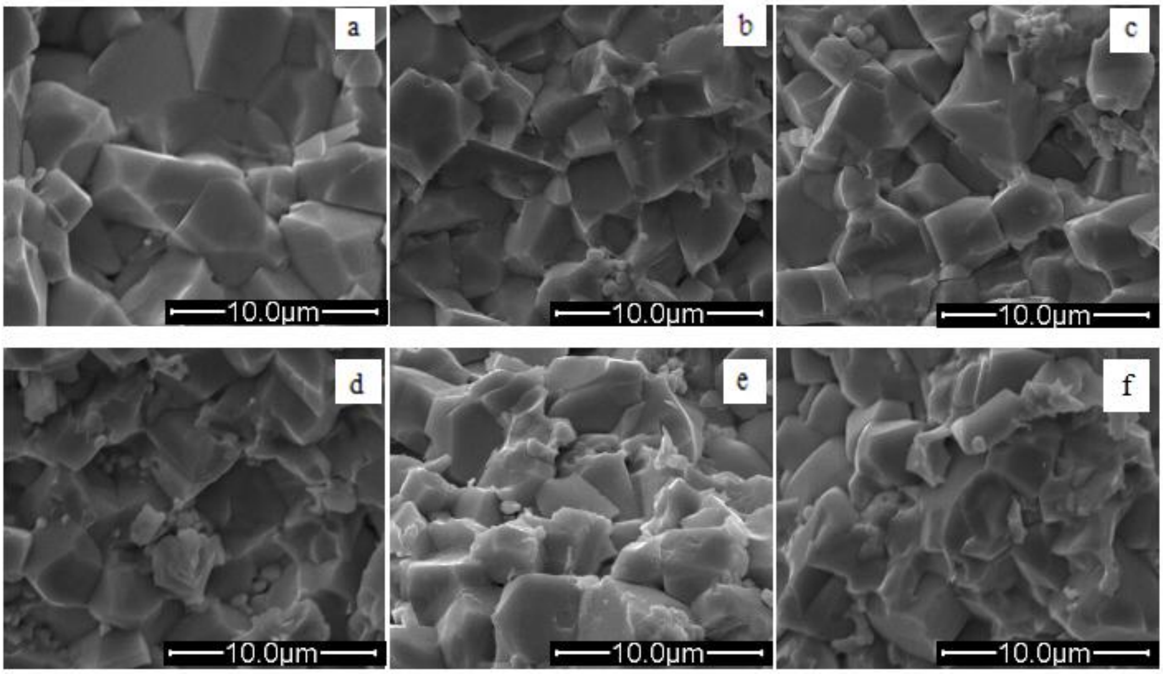


Fig. 2 SEM images of different (1-$x$)$LiNbO_3$-$x$$BiYbO_3$ ceramics: (a) $x$ = 0 (b) $x$ = 0.01 (c) $x$ = 0.02 (d) $x$ = 0.03 (e) $x$ = 0.04 (f) $x$ = 0.05

**Piezoelectric Properties**

Fig.3 shows the piezoelectric constant $d_{33}$ of samples at room temperature as a function of $x$. The hightest piezoelectric constant $d_{33}$ appears at $x$ = 0.04 or $x$ = 0.05, which maybe caused by the impure phase. The results revealed that the addition of $BiYbO_3$ for $LiNbO_3$ ceramics did not cause an obvious change of piezoelectric constant $d_{33}$.

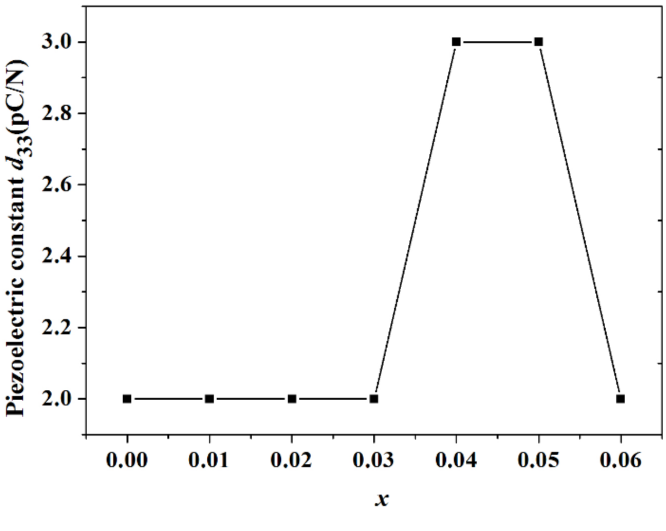


Fig.3 Piezoelectric constant $d_{33}$ of samples at room temperature as a function of $x$

**Dielectric Properties**

Fig. 4 shows the permittivity of (1-$x$)$LiNbO_3$-$x$$BiYbO_3$ ceramics as a function of frequency for sintered samples at room temperature. The frequency range is measured from 1 MHz to 1 GHz. It is found that the dielectric resonance peak of permittivity moves toward high frequency with an increase the doping amount of $BiYbO_3$. The value of real part of permittivity is strongly dependent on the $BiYbO_3$ content. For instance, the sample with $x$ = 0 has a permittivity ε' of about 45 at 100 MHz; however, in the case of $x$ = 0.06, the permittivity ε' attains the maximum values of 55 at 100 MHz.

Similar to real part of permittivity, imaginary part of permittivity ε'' also dependent on the $BiYbO_3$ amount.

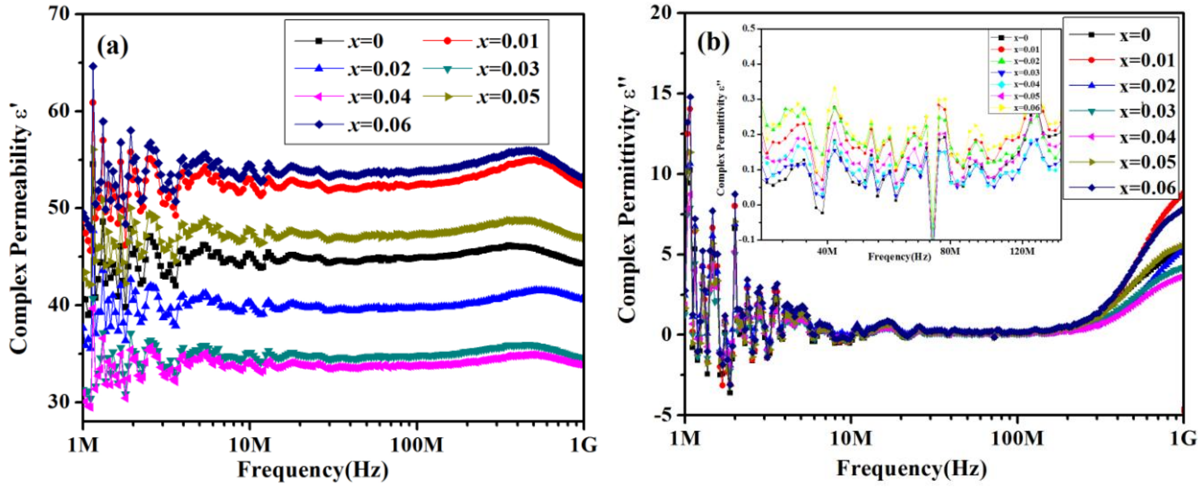


Fig. 4 Frequency dependence of permittivity for all samples: (a) real part of permittivity, and (b) imaginary part of permittivity. Inset shows a zoom around 25 MHz to 150 MHz of (b)

## Summary

A series of $(1\text{-}x)LiNbO_3\text{-}xBiYbO_3$ ceramics have been successfully prepared using conventional solid-state reaction method. No impurity phase was found in samples $x = 0.01$ and $x = 0.02$. With the increase of the content of $BiYbO_3$, the second phase is gradually increased. The microstructures of all ceramics are dense and uniform. The mean grain size tends to reduce when $BiYbO_3$ content increase. The addition of $BiYbO_3$ did not cause an obvious change of piezoelectric constant $d_{33}$ for $LiNbO_3$ system.Both permittivity of ε' and ε'' significantly dependent on the increase doping level of $BiYbO_3$.

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