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Letter

A rational designed multi-layered structure to improve the temperature stability of Li modified (K,Na)NbO_3 piezoceramics

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A B S T R A C T

The temperature dependent properties of (K,Na)NbO_3-based piezoelectric materials enlightened us that the combination of compositions gradient could remedy the fluctuation of the electrical properties induced by the temperature variation. Then Li modified (K,Na)NbO_3 ceramics was chosen as target materials and a multi-layered structure containing a series of composition gradient layers was prepared via a conventional solid state reaction. The results of relative permittivity against temperature demonstrated that the as-called relaxor feature was observed in this proposed multi-layered structure and the change rate of the dielectric anomaly peak got evidently reduced. Besides, the field-induced strain varying less than 10% from the room temperature to 100 °C was also achieved in this layered-structure ceramics. Moreover, the investigation here could render us an important reference for designing ferroelectric materials to satisfy the diversified requirements in the functional device.

1. Introduction

A wide range of lead-free piezoelectric composition systems have been investigated for the sake of replacing (Pb,Zr)TiO_3-based piezoelectric materials (abbreviated as PZT) owing to the ever-increasing concerns of environmental sustainable development. Among these reported lead-free materials systems, (K,Na)NbO_3 materials (abbreviated as KNN) have emerged as one of the most promising candidates owing to its outstanding piezoelectric properties and high Curie temperature point [1,2]. However, KNN-based materials system exhibits the inferior temperature stability of piezoelectric property owing to the limited temperature region for the coexistence of orthorhombic and tetragonal phase [3–5]. In order to enhance the temperature stability of KNN-based materials system, an effective approach has been universally adopted so far: shifting the transition temperature of orthorhombic to tetragonal phase ($T_{O\text{-}T}$) below the room temperature by the chemical substitution [2,6–8]. However, the essence of this approach is actually to avoid polymorphic phase transition (abbreviated as PPT) problem by creating a single tetragonal phase between the room temperature and Curie temperature. Meanwhile this approach would ineluctably sacrifice the piezoelectric activity of KNN-based materials system to some extent. On account of the absence of great advance in the enhancement of temperature stability, the further development and wide application of KNN-based piezoelectric materials is still hampered.

As for KNN-based materials with the composition lying in the region of PPT, the electrical properties (such as the temperature dependent the relative permittivity and the piezoelectric coefficient) would achieve the maximum value at a certain temperature and exhibit an approximate normal distribution around this temperature point. And the above mentioned circumstances have been already reported in previous literature and generally accepted by the material researchers [9–11]. As for Li element modified (K,Na)NbO_3, the critical temperature point (possessing the maximum value for these above-mentioned electrical properties) accordingly varied with the concentration of Li element. Taking $T_{O\text{-}T}$ (the temperature point for the transition from the orthorhombic to tetragonal phase) for example, with the concentration of Li element changing from 0 to 8% mole percent, $T_{O\text{-}T}$ got reduced from more than 200 °C to far below the room temperature. However, the change trend (the normal distribution around the critical temperature point) for these temperature dependent electrical properties, such as relative permittivity and piezoelectric constant would not show obvious change. With the consideration of an approximate normal distribution of these electrical properties, we put forward a sketch to theoretically illustrate this feature in Fig. 1, which indicated the variation of electrical properties (such as relative permittivity, etc.) as the function of temperature for Li element modified KNN ceramics. The peak of relative permittivity (ε_{max}) was located at the phase transition temperature point ($T_{O\text{-}T}$) for all of these Li modified KNN ceramics. However, by noticing the change trend of each curve (with different Li concentration) illustrated in Fig. 1, if we combine the compositions with the different Li concentration together, it would arrive at the possibility of weakening the normal distribution of these temperature depended electrical properties. This consideration inspired us in this current research to put up with a multi-layered structure with improved temperature stability as illustrated in Fig. 1. The gradient color in these layers indicated the different Li concentration in KNN-based ceramics.

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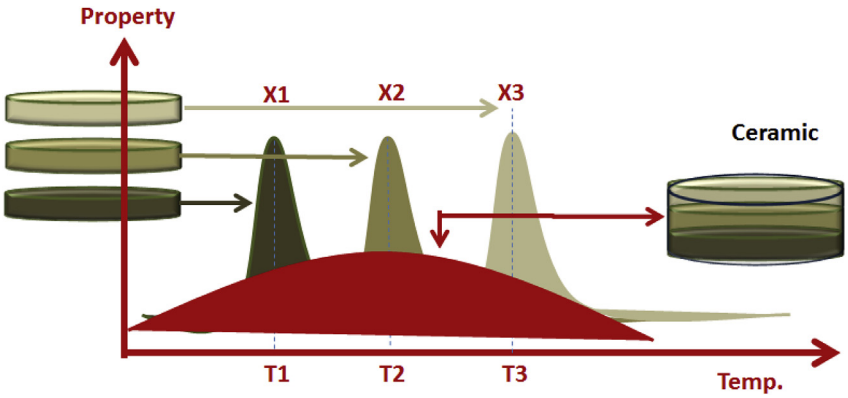

Fig. 1. The illustration of the relationship between PPT temperature and different composition.

2. Experimental procedure

Different Li concentration modified $[Li_x(K_{0.48}Na_{0.52})_{1-x}]NbO_3$ powders (x = 2%, 4%, 6%), which were named as L2 (Li: 2%), L4 (Li: 4%) and L6 (Li: 6%), were separately prepared by a conventional solid state reaction method. High purity powders of Na_2CO_3 (99.5%), K_2CO_3 (99.9%), Li_2CO_3 (99%) and Nb_2O_5 (99.99%) were used as starting raw materials. The powders in the stoichiometric ratio were mixed in the alcohol using ball milling for 24 h. After ball milling, the raw materials were dried and calcinated at the temperature range of 750–850 °C for 5 h according to Li concentration. The calcinated powders with different Li concentration were pressed into disks with the diameter of 10 mm using polyvinyl butyral as the binder. While the multi-layered structure were synthesized as S1 (L2+L4), S2 (L4+L6), S3 (L2+L4+L6) and the weight of each layer (corresponding to the different Li element concentration) was kept at 0.1 g. After the binder was burn out, the sintering procedure was carried out in the air for 2 h without protection and the sintering temperature varied within the temperature region of 1050–1100 °C. After being polished, silver paste was fired on the both sides of as-obtained ceramics at 550 °C for 20 min to form electrodes for the electrical measurements. The

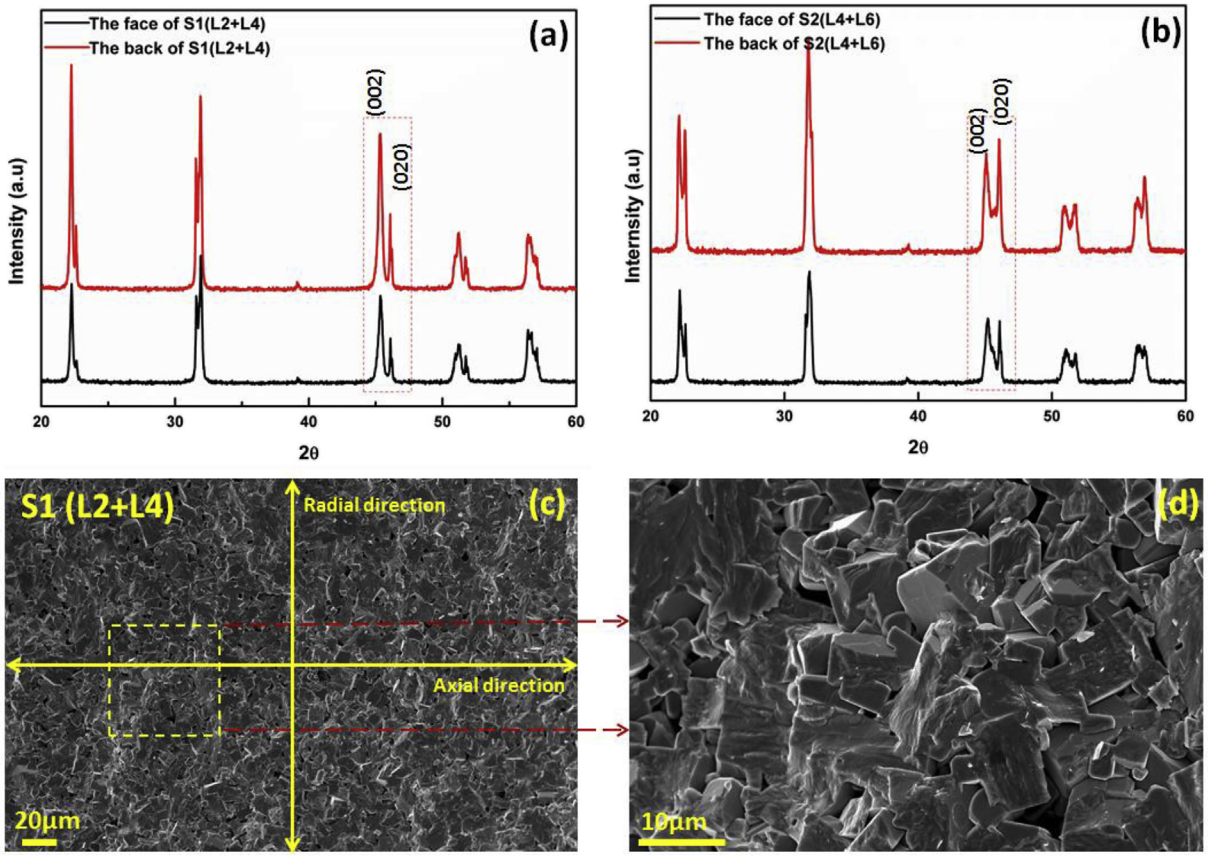

Fig. 2. (a) The XRD patterns for the face and back of the sample S1, (b) The XRD patterns for the face and back of the sample S2, (c) and (d) The cross-sectional SEM images for the sample S1.