Synthesis and characterization of two-dimensional lead-free (K, Na)NbO₃ micro/nano piezoelectric structures

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Received: February 14, 2019; Revised: May 21, 2019; Accepted: July 20, 2019

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Abstract: Two-dimensional (2D) lead-free (K, Na)NbO₃ (KNN) micro/nano structures with controllable K/Na ratio were successfully fabricated via a two-step molten salt synthesis (MSS). In this work, the reaction factors, including the proportion of molten salts, the types of carbonates, the sintering temperature, and the sintering time, were discussed in detail and the optimized condition was identified. The microstructure of KNN was confirmed by confocal Raman spectroscopy, while piezoresponse force microscopy (PFM) was applied to measure three-dimensional (3D) morphology and piezoelectric properties of KNN particles. The as-synthesized KNN platelets apparently possess anisotropic morphology and uniform structure, the size of which reaches 5–20 μm in length/width and 0.5–1 μm in thickness. It should be noted that the K/Na ratios of the KNN crystals are basically consistent while the proportion of salts changes within a certain range. The enrichment of Na element in the products is also observed, which owes to the smaller ionic radius of Na⁺ comparing to that of K⁺. This result provides a reference for the further preparation of textured ceramics and flexible piezoelectric generators.

Keywords: (K, Na)NbO₃ (KNN); lead-free; two-dimensional (2D) structure; piezoelectric property

1 Introduction

Piezoelectric materials are functional materials which can interconvert mechanical and electrical energies. Such smart materials can be fabricated as sensors, actuators, transducers, etc., which are widely applied in the fields of electronic communications, smart homes, and medical imaging [1–8]. Although lead zirconate titanate (Pb(Zr,Ti)O₃, abbreviated as PZT) has excellent

electromechanical performance, lead-based materials have been gradually replaced by lead-free materials due to the toxic nature of lead. Among all the lead-free material candidates, alkali niobate ceramics based materials have made a breakthrough in 2004. Saito *et al.* [9] prepared textured (K, Na)NbO₃ (KNN) based ceramics with a high piezoelectric coefficient d_{33} of 416 pC/N, which is comparable to that of PZT. This work has ignited worldwide interest in exploring lead-free KNN based piezomaterials. More and more studies on KNN materials concentrate on the improvement of piezoelectricity [10–15].

Driven by an increasing demand for the textured

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ceramics and the flexible piezoelectric generators, the research on low dimensional lead-free niobate based micro/nano-structures have made great progress [16–18]. For instance, the well-textured KNN ceramics are considered to possess high piezoelectric response [9,19]. Yan *et al.* [20] fabricated grain-oriented PbTiO_3 ceramic with 95% $\langle 001 \rangle$ texture, which has an extremely large g_{33} . Thus, the textured lead-free ceramics with high piezoelectric performance are also expected according to the work reposted by Saito *et al.* [9]. In order to obtain the textured KNN materials with high piezoelectric response, one of the challenges is the fabrication of plate-like particles as templates [21]. As is known, the reactive template grain growth (RTGG) is a texturing technique, which uses plate-like or needle-like particles as templates for oriented growth of crystals. The resultant samples would obtain the anisotropic structures with improved physical and mechanical properties [9,22]. During the process, the required characteristics of the templates are crucial, such as uniform structure, complete crystal morphology, and stable chemical properties [23–27]. Furthermore, the plate-like KNN particles are expected to inherit the morphology and structure of the precursor [28–30].

There are many studies on the preparation of single-crystal NaNbO_3 structures through the plate-like precursors [9,31–33], while the reports on KNN structures are relatively few. Ishii and Tashiro [34] reported the orientation control of KNN ceramics using NaNbO_3 templates, and the sinterability of the textured ceramics, together with the diffusivity of K atoms into the templates was investigated. Li *et al.* [35] studied the synthesis of mechanism of KNN plate-like particles. Lee *et al.* [36] found that the salt contributed to the conversion reaction from plate-like precursor to KNN or NaNbO_3 platelets. Zhu *et al.* [37] fabricated Li-doped KNN thick film by employing NaNbO_3 as template. Compared to NaNbO_3 two-dimensional (2D) templates, KNN plate-like crystals are expected to possess high piezoelectric properties, and might be much similar to the ceramic matrix [35,36]. Moreover, the enhanced performance is reasonably expected when the K/Na ratios of KNN materials are tailored at an optimal composition. Thus, the preparation of the KNN micro/nano-plates would be beneficial to the piezoelectric composite and ceramics. However, it would be a challenge for synthesizing KNN materials with controlled compositions at the micro/nano-scale. For instance, the type of molten salt, together with the ratio of $\text{K}_2\text{CO}_3/$

BNN_5 , would lead to the deviated structures results [35,36]. Besides, it should be stressed that the cationic sizes of K^+ and Na^+ differ from that of Bi^{3+} , thus it is difficult to predict the practical amount of cations entering the lattice.

In this work, the plate-like KNN platelets were fabricated via topochemical micro-crystal conversion (TMC). During the preparation procedure, the bismuth layered-perovskite $\text{Bi}_{2.5}\text{Na}_{3.5}\text{Nb}_5\text{O}_{18}$ (BNN5) precursors were initially prepared by molten salt synthesis. Then, the KNN crystals were obtained by cations substitution of the BNN5 structures. The reaction conditions including molten salts, the carbonates, the sintering temperature, and the sintering time were also investigated. Furthermore, the microstructure and piezoresponses of the KNN micro/nano-structures were addressed by Raman and piezoresponse force microscopy (PFM), respectively.

2 Experimental

2.1 Synthesis of the plate-like KNN crystals via TMC

The BNN5 precursors were synthesized by the molten salt synthesis (MSS). Reagent-grade Bi_2O_3 (99.99%), Nb_2O_5 (99.99%), and Na_2CO_3 (99.8%) were weighed according to the stoichiometric ratio and mixed together with the same weight of NaCl (99.5%). The mixtures were fully milled in the mortar for 2 h. Then, the mixture of reactants was calcined at 1100 °C for 2 h in the Al_2O_3 crucible. The as-prepared products were washed several times with hot deionized water to remove the residual salts. Finally, the BNN5 particles were obtained as precursors.

The plate-like KNN structures were fabricated via TMC. The BNN5 precursors reacted with Na_2CO_3 or K_2CO_3 (99.0%) in a molten salt flux. The carbonate and KCl/NaCl (99.5%) salts were milled for ~1 h, and then the BNN5 particles were added and milled for another 1 h. The mixtures were kept at 970–1020 °C for different hours. The reaction products were washed several times with hot deionized water and HCl solution alternately to remove the NaCl salts and the Bi_2O_3 , respectively.

2.2 Characterization of KNN structures

The crystalline structures of the platelets were characterized by X-ray diffraction (XRD Rigaku, D/Max2500, Japan) using $\text{Cu K}\alpha$ radiation. The morphologies of the particles were observed by scanning

electron microscope (SEM, JEOL JSM-7001F, Japan). The elemental analysis was carried out by energy dispersive spectroscopy (EDS, Oxford, Japan). The phase structure of the products was examined by Raman spectroscopy (LabRAM HR, HORIBA Jobin Yvon, France) with an excitation wavelength of 488 nm. The microscopies of 2D KNN structures were observed by atomic force microscope (AFM, Asylum Research MPF-3D, USA) with a Cr/Pt coated tip at tapping mode, while the piezoelectric responses were measured using switching spectroscopy piezoresponse force microscopy (SS-PFM).

3 Results and discussion

The XRD patterns of the BNN5 particles synthesized by molten salt synthesis at 1100 °C for 2 h with a heating rate of 10 °C/min are shown in Fig. 1(a). Almost all of the diffraction peaks could be assigned to the BNN5 phase. The intensities of (0012), (0014),

(0018), and (0024) peaks in the diffraction pattern are stronger than those in the PDF#42-0399, indicating that the BNN5 structures tend to orientate along (001) crystallographic plane direction. Although there are a few of BNN2 and BNN4 phases as marked in Fig. 1(a), it has no significant effect on the TMC due to their similar structures to that of the BNN5 crystals. As shown in Fig. 1(b), the BNN5 products are plate-like with relatively smooth surface and quite complete rectangle section. A typical particle of the BNN5 crystal is presented in the inset of Fig. 1(b), revealing that the BNN5 particles are high-aspect-ratio. According to the statistics of sample sizes, the BNN5 samples possess an average size of 5–20 µm in length/width and 0.5–1 µm in thickness. Hence, the BNN5 platelets with anisotropic morphology would be suitable precursors to fabricate the KNN crystals.

In order to fully investigate the KNN crystals, the NaNbO_3 particles were first prepared as comparison. Figure 2(a) shows the XRD patterns of NaNbO_3 crystals produced in NaCl salt at 950 °C for 4 h. All

Fig. 1 (a) XRD patterns and (b) SEM images of BNN5 products.

Fig. 2 (a) XRD patterns and (b) SEM images of NaNbO_3 platelets.

the peaks can be indexed to the perovskite phase well, identified by PDF#33-1270. Moreover, the surfaces of the NaNbO_3 crystals are parallel to (001) since the intensities of the (001) and (002) are much stronger than those in the PDF#33-1270. Figure 2(b) shows the SEM images of the plate-like NaNbO_3 platelets. The morphology of the NaNbO_3 crystals has inherited that of BNN5 precursors according to the similarity between Figs. 1(b) and 2(b). Moreover, a representative NaNbO_3 particle is shown in the inset of Fig. 2(b), the morphology of which is also in accordance with that of BNN5 particle. Whereas, there are a few of fragments perhaps due to the structure reconstruction during the reaction.

The XRD patterns of the KNN particles prepared at 1020 °C for 10 h with K_2CO_3 and different ratios of KCl/NaCl are shown in Fig. 3(a). Although there are a tiny amount of BNN5 structures, the KNN particles mainly possess perovskite phases when the ratios of KCl/NaCl are set at 2/1 and 4/1. However, a certain amount of BNN5 phase emerges with the increasing KCl/NaCl ratio, and the XRD results of the resultant product with the salts ratio of 8/1 is also presented. As is known, the cation radius of Na^+ (1.02 Å) is much close to that of Bi^{3+} (1.03 Å). Moreover, the cation radius of K^+ is 1.38 Å, thus Na^+ is considered easier to incorporate into the BNN5 structures. However, the concentration of Na^+ is considerably low in the reaction solution when the salts ratio of KCl/NaCl is 8/1 with K_2CO_3 as carbonate. Hence, the Na^+ cations would be insufficient to replace Bi^{3+} , and the BNN5 precursor was retained. In addition, there is a small shift of diffraction peaks to a lower angle $\sim 46^\circ$ with the increase of the salts ratio, illustrating that a certain amount of K^+ incorporate into the structures. It could be inferred that the KNN particles with high K

concentration would be rather difficult to be obtained only with the high ratio of KCl in the salts due to the remnant BNN5 and big radius of K^+ . In order to confirm the effect of carbonate on the KNN particles, further comparative experiments were carried out. Figure 3(b) shows the XRD patterns of the KNN particles synthesized with K_2CO_3 and Na_2CO_3 when the salts ratio of KCl/NaCl is 4/1, respectively. The samples with perovskite structure can be identified by PDF#33-1270. The types of carbonates have no significant influence on the final products, which is in good agreement with Ref. [35]. During the TMC process, the CO_3^{2-} could mainly contribute to removing the weak network $[\text{Bi}_2\text{O}_2]^{2+}$. In addition, some peaks at 22° and 45° indexed by PDF#32-0822 reveal the existence of KNN phase.

Moreover, the synthesis factors such as reaction temperature, together with the sintering time, should also be emphasized, and the results are shown in Fig. 4. Figure 4(a) shows the KNN structures prepared at 970 °C for various sintering time. The BNN5 structures are clearly observed in the KNN particles synthesized for 6 h. It was decided to extend the sintering time from 6 to 10 h for the purpose of promoting more complete reaction. As shown in Fig. 4(a), the BNN5 phases obviously decrease, and there are mainly perovskite phases in the KNN particles assigned by PDF#33-1270. Thus, it is revealed that extending the sintering time helps K^+/Na^+ to replace the Bi^{3+} completely. In order to further control the KNN structures, the reaction temperatures varied from 970 to 1020 °C. It can be seen from Fig. 4(b) that the intensities of BNN5 phase are weaker at 1020 °C comparing to that at 970 °C, indicating that increasing sintering temperature helps the fabrication of KNN structures with less impurity.

Fig. 3 XRD patterns of KNN platelets synthesized with (a) different ratios of KCl/NaCl and (b) carbonates.

Fig. 4 XRD patterns of KNN platelets fabricated with different (a) sintering time and (b) sintering temperatures.

The morphology and EDS images of the KNN particles are shown in Fig. 5. The morphology of the KNN platelets synthesized with the KCl/NaCl ratio of 2/1 and 4/1 is basically consistent with that of BNN5 structures. The length/width of the KNN samples reaches 5–20 μm , while the thickness is close to 1 μm . The morphology of the KNN particles prepared with the salts ratio of 2/1 is quite complete in edge as shown in Fig. 5(a); when the samples are broken, more obvious sample fragments can be seen in Fig. 5(c). The reason might be that the perovskite structures would be crashed to some extent when the considerable K^+ incorporates into the structures during the TMC procedure. The EDS spectra of the KNN particles with different molten salt ratios are shown in Figs. 5(b) and 5(d). There are Na^+ ,

K^+ , Nb^{5+} , and O^{2-} detected in the KNN particles, and the atomic ratios are also listed. The enrichment of Na^+ cations is also observed in the EDS results, and the quantity of K^+ cations entered the crystal structures rises to some extent with the increase of the KCl/NaCl ratio. Thus, the chemical formulas of the KNN structures synthesized with the KCl/NaCl ratio of 2/1 and 4/1 are roughly considered as $(\text{K}_{0.08}\text{Na}_{0.92})\text{NbO}_3$ and $(\text{K}_{0.15}\text{Na}_{0.85})\text{NbO}_3$, respectively. According to the experimental results, the K/Na ratio of the KNN particles could be mainly attributed to the salt ratio and the sintering condition, while the types of carbonates seem do not play a critical role on the KNN template synthesis. As a result, the content of Na in the product is much higher than that of K element, and the change

Fig. 5 SEM images of KNN platelets synthesized with KCl/NaCl ratios of (a) 2/1 and (c) 4/1; (b, d) EDS results corresponding to (a, c), respectively.

of K atomic ratio is not so obvious while the proportion of molten salts varies in the range from 2/1 to 4/1.

Raman spectroscopy is very sensitive to the structure [38]. In order to figure out the effect of the salts proportion, Raman spectra of 2D NaNbO_3 and KNN templates measured at $\sim 25^\circ\text{C}$ are shown in Fig. 6. The structures were mechanically pressed into thin bulks as the initial step. Then, the spot of incident light was set at $1\ \mu\text{m}$ and focused on the top of the thin bulk. It should be noted that the light would not raise the

Fig. 6 Raman spectra of NaNbO_3 structures and KNN structures with KCl/NaCl ratios of 4/1 and 2/1.

temperature of NaNbO_3 and KNN micro/nano structures due to its low power which is small than 8 mW. As is known, there are kinds of internal vibrational modes of NbO_6 octahedra. ν_1 and ν_5 modes are relatively strong and could be easily identified in the Raman spectra as shown in Fig. 6. It can be seen that some bands in the region of $180\text{--}200\ \text{cm}^{-1}$ are observed as ν_6 modes in NaNbO_3 structures, while the intensity of these bands significantly decreases in the KNN ones. Moreover, the $\nu_1+\nu_5$ mode locates at $880\ \text{cm}^{-1}$ in the NaNbO_3 sample and shifts to the lower frequency in the KNN sample, which is related to the change of vibration mode causing by the addition of K^+ cations. In addition, the Raman results for the KNN templates synthesized with different salts proportions, i.e., KCl/NaCl of 2/1 and 4/1, are basically consistent. It could be concluded that there is little difference between the two components of the KNN samples, which is in accordance with the EDS results in Fig. 5. It should also be noted that random regions of the samples were selected, and the Raman scattering results remain essentially unchanged, indicating that the samples are homogeneous in composition.

Figure 7 shows the topography and the piezoelectric

Fig. 7 (a) 3D and (b) 2D AFM images of KNN structures with the KCl/NaCl ratio of 2/1; (c) height variation of the line across the KNN structure as marked in (b); (d) piezoresponse and phase information gathered from points 1–3, as marked in (b).

responses of the plate-like KNN structures with the KCl/NaCl ratio of 2/1. AFM at tapping mode was employed to map the topography of the KNN particles, and the 3D and 2D images of the KNN particles are shown in Figs. 7(a) and 7(b), respectively. A straight line was drawn across the KNN particle as marked in Fig. 7(b), and the height information of the line was gathered, indicating that the thickness of the plate is ~500 nm as shown in Fig. 7(c). Whereas, the topography of the KNN plate in Fig. 7(c) seems to be rough, and it could be attributed to the fragment of KNN samples adhered to the surface of the plate. The particle size corresponds well with the SEM analysis results (Fig. 5(a)). Furthermore, the SS-PFM was applied to measure the piezoelectric response of the KNN plates, and three points are arranged on the surface of the particle as marked in Fig. 7(b). A sequence of DC signal with “on” and “off” states in a periodic triangle wave ranging from –30 to +30 V was applied, while an AC excitation signal of 3 V was added at intervals of increasing DC voltage to minimize electrostatic effects [39]. The amplitude butterfly curves and the phase hysteresis loops of the three points obtained during the “off” state are shown in Fig. 7(d), which represent the typical amplitude–voltage and phase–voltage hysteresis loops, respectively. The phase contrast of the three regions on the KNN structure is approximately 180°, which is a signature of domain reversal in typical ferroelectrics [40,41]. Moreover, the piezoresponses from different districts are not extensively different, indicating the uniformity of the compositions and crystal structures of the KNN plates.

4 Conclusions

In conclusion, 2D plate-like lead-free KNN structures were successfully synthesized via a two-step MSS. The layered-perovskite BNN5 structures were synthesized as precursors in the initial step, and then the high-aspect-ratio KNN crystals were prepared using the BNN5 precursors by TMC. The reaction factors were discussed in detail, and the optimized condition was confirmed. It should be emphasized that confocal Raman spectroscopy and PFM were used to characterize the microstructure and piezoelectric property of the KNN plates. Resultantly, the KNN crystals with uniform structure and anisotropic morphology were obtained, which possessed the size of 5–20 μm in length/width and 0.5–1 μm in thickness. Additionally, the enrichment

of Na element in the products is also observed when compared to K element, indicating the different cation substitution in the BNN5 precursors. The 2D KNN micro/nano piezoelectric structures are expected to play an important role on the fabrication of high-performance KNN based textured ceramics.

Acknowledgements

The authors acknowledge the financial supports from the National Natural Science Foundation of China (Grant No. 51602345), the State Key Laboratory of New Ceramics and Fine Processing Tsinghua University (Grant No. KF201512), Open Fund of State Key Laboratory of Coal Resources and Safe Mining (Grant No. SKLCRSM19KFA13), and Fundamental Research Funds for the Central Universities (Grant No. 2016QJ01).

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