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Preparation and Characterization of New $Pb(Yb_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ Ternary Piezo-/Ferroelectric Crystals

Chao He,[†,‡] Xiuzhi Li,[†] Zujian Wang,[†] Xifa Long,*[,†] Shaoyu Mao,[§] and Zuo-Guang Ye*[,∥]

[†]*Key Laboratory of Optoelectronic Materials Chemistry and Physics, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fujian, Fuzhou 350002, China,* [‡]*Graduate University of Chinese Academy of Sciences, Beijing 100049, China,* [§]*College of Chemistry & Chemical Engineering, Xiamen University, China, and* [∥]*Department of Chemistry and 4D LABORATORIES, Simon Fraser University, 8888 University Drive, Burnaby, British Columbia, V5A 1S6, Canada*

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Piezo-/ferroelectric crystals of a new $Pb(Yb_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ ternary solid solution system with the dimensions of $30 \times 30 \times 12\ mm^3$ have been grown by the top-seeded solution growth method for the first time. The crystal structure is analyzed by means of X-ray diffraction at various temperature. The composition of the grown crystals is found to be near the ternary morphotropic phase boundary. The Curie temperature T_C reaches 205 °C. The piezoelectric coefficient d_{33} and the longitudinal electromechanical coupling factor k_{33} are found to be 1800 pC/N and 90%, respectively. A remanent polarization Pr of $27.8\ \mu C/cm^2$ is displayed with a coercive field E_c of about 7 kV/cm. These results show that the PYMNT crystals exhibit a higher T_C and a larger coercive field than the binary PMN-PT piezocrystals, making this material a promising candidate for high-power electromechanical transducers that can operate in a wider temperature range.

Introduction

Recently, there is considerable interest in relaxor-based piezo-/ferroelectric crystals of perovskite-type with compositions close to or within the morphotropic phase boundary (MPB) region, due to their high piezoelectric properties which make them promising candidates for the applications as high-performance solid state actuators and electromechanical transducers.[1−5] The most typical of these kinds of crystals are $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMNT)[6−8] and $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PZNT),[9−11] which exhibit very high piezoelectric coefficients, large longitudinal electromechanical coupling factors, high dielectric constants, low dielectric losses, and exceptionally high strain levels. However, the PMNT and PZNT crystals near the MPB region show a low Curie temperature T_C (about 150 °C for PMNT and 170 °C for PZNT), and an even lower depoling temperature T_{rt} (75−95 °C), which arises from the rhombohedral-tetragonal phase transition in the MPB region. This drawback seriously limits their applicability in the high temperature range. Another drawback of the PMNT and PZNT crystals is the low coercive field ($E_c = 2-3$ kV/cm), which causes the crystals very easily to depole and thereby their piezoelectric performance to degrade.[2] Therefore, new piezo- and ferroelectric single crystals with high performance and high T_C are desired. In the past several years, some other relaxor-based ferroelectric crystal systems have been investigated, such as $Pb(Yb_{1/2}Nb_{1/2})O_3$-$PbTiO_3$ (PYNT),[12,13] $Pb(In_{1/2}Nb_{1/2})O_3$-$PbTiO_3$ (PINT),[14,15] and $Pb(Sc_{1/2}Nb_{1/2})O_3$-$PbTiO_3$ (PSNT),[16] which seem to exhibit a higher Curie temperature. However, progress in the growth of these crystals has been slow up to now, mostly due to the difficulties in controlling the proper composition. In order to solve this issue, researchers have tried to modify the PMNT and PZNT crystals by doping a small amount of other ions with the intention to increase

*To whom correspondence should be addressed. X. Long: phone, (86)-591-83710369; fax, (86)-591-83714946; e-mail, lxf@fjirsm.ac.cn. Z.-G. Ye: phone, 778-782-8064; fax, 778-782-3765; e-mail, zye@sfu.ca.

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the morphotropic phase transition temperature T_{rt} and also the E_c.[17−19] However, the results are not particularly promising as the effects of chemical modification may vary depending upon the systems and dopants. For instance, Mn-doping increases the E_c, T_C, and T_{rt} of PMN-29PT crystals but the piezoelectric properties are reduced,[18] while Fe-doped PZN-4.5PT crystals show reduced T_C and T_{rt} by 20 °C, even though the E_c and d_{33} are increased.[17]

In recent years, great attention has been paid to some ternary solid solution systems, for example, $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PIMNT)[20,21] and $Pb(Sc_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PSMNT),[22] which show promising piezoelectric properties with a higher Curie temperature. On the other hand, the binary PYNT crystals were reported to possess high piezoelectric properties and high T_C, but it has been very difficult to grow large and high-quality crystals.[12,13,23] We believe that the $Pb(Yb_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PYMNT) ternary system should present an MPB and the PYMNT single crystals of the MPB compositions could exhibit excellent piezoelectric properties, a higher T_C, and a larger E_c, compared with the PMNT and PZNT crystals. Therefore, in this work, we take on the challenge of growing the PYMNT ternary crystals. In this paper, we report the growth, phase analysis, and dielectric, piezo-, and ferroelectric characterizations of the PYMNT ternary crystals. The characterization of the dielectric, piezo-, and ferroelectric properties of the grown crystals confirms the advantages anticipated which make the PYMNT crystals the most promising candidate for high piezoelectric applications. The properties of the PYMNT crystals are also compared with those of some typical ferroelectric single crystals with compositions near the MPB.

Top-seeded solution growth (TSSG) method has been developed to grow large size and high quality relaxor-based ferroelectric crystals.[2,8,19] Compared with other methods, the TSSG technique offers some advantages in growing single crystals of good quality, high compositional homogeneity, and controlled morphology thanks to its unique temperature field design and slow growth process.

Experimental Section

In this work, the PYMNT single crystals were grown by the TSSG method. The starting chemicals, PbO (99.9%), TiO_2 (99.9%), MgO (99.9%), Yb_2O_3 (99.99%), Nb_2O_5 (99.9%), were mixed according to the (nominal) formula of 0.20PYN-0.45PMN-0.35PT [PYMNT (20/45/35)] (solute). The mixture of PbO and H_3BO_3 (99.9%) was used as flux (with a molar ratio of PbO/H_3BO_3 = 4:1) and the flux to solute molar ratio was 3:1. The mixture was placed in a Pt crucible, which was then palced into a vertical tubular furnace to melt. The PYMNT seed crystals used were obtained from preliminary flux growth by spontaneous nucleation. The saturation temperature T_s of the solution was determined to be about 1100 °C by repeated seeding trials. At the stage of growth, the slow cooling rate was set between 0.2 and 0.5 °C/h. The growing crystal was rotated at a speed of 0−30 rpm. At the end of growth process, the grown crystal was pulled out of the melt surface and then cooled down to room temperature at a rate of 20 °C/h. Large size and high-quality PYMNT single crystals were obtained at the end of the growths. For structural analysis, X-ray diffraction was performed at room temperature (25 °C) and 100 °C on a Rigaku diffractometer (DMAX2500) equipped with Cu Kα radiation and a graphite monochromator. The actual chemical composition of the grown crystals is analyzed by energy dispersive spectrometry (EDS, JSM 6700 field emission scanning electron microscope). The single crystals were sliced into platelets (wafers) and bars (with an aspect ratio of 3.7 mm height/1 mm base) with (001) orientation. Then, the samples were polished and sputtered with gold layers as electrodes for electric measurement. The dielectric constant (ε') and dielectric loss tangent (tan δ) were measured using a Solartron 1260 impedance analyzer in conjunction with a 1296 dielectric interface, with an ac signal of 0.3 V (peak-to-peak) applied. The same setup was used to measure the resonance frequency (f_r) and antiresonance frequency (f_a) of a bar sample. The piezoelectric coefficient was measured using a quasistatic d_{33} meter (Institute of Acoustics, Chinese Academy of Sciences, model ZJ-4AN) and calculated using resonance and antiresonance frequencies. The ferroelectric hysteresis loops were displayed using an aixACCT TF2000 analyzer (at f = 2 Hz).

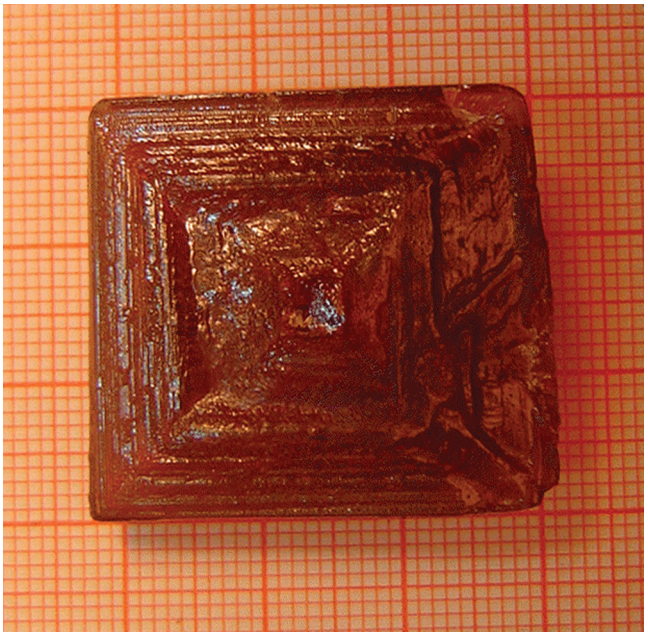

Figure 1. An as-grown PYMNT(15/53/32) single crystal by the TSSG method (scale in millimeters).

Results and Discussion

Figure 1 shows a PYMNT single crystal with dimensions of 30 × 30 × 12 mm^3. It can be seen that the PYMNT crystal grown by the TSSG method exhibits a

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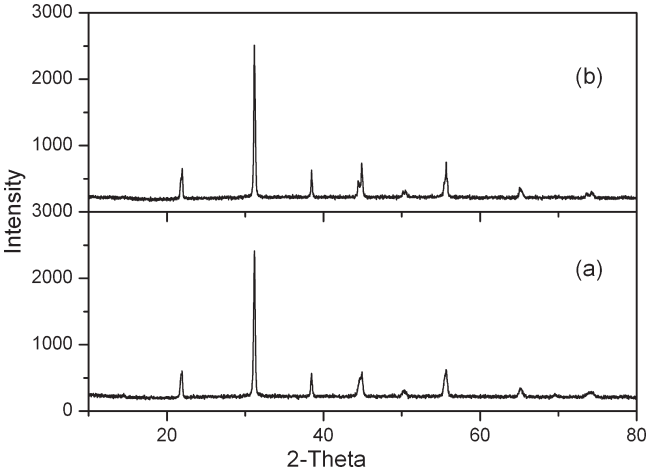

Figure 2. X-ray powder diffraction patterns of the crashed PYMNT(15/53/32) crystals: (a) at 25 °C and (b) at 100 °C, indicating a pure complex perovskite phase and a phase transition from the rhombohedral to tetragonal phase upon heating.

quadrate morphology with naturally developed (100) faces, suggesting that the growth speed of the (100) faces is the slowest. The actual chemical composition of the grown crystals is determined to be PYMNT(15/53/32) based on the data from energy dispersive spectrometry. This slight deviation from the nominal composition resulted from the inevitable phase segregation in this ternary system, as observed in many other solid solution crystals.

Figure 2 presents the X-ray diffraction (XRD) patterns measured at 25 and 100 °C, respectively. It can be seen that the as-grown PYMNT single crystals were of pure perovskite phase. In order to accurately study the possible change of phase symmetry as a function of temperatures, special attention was focused on (200) reflections around $2\theta = 45°$. It is well-known that the XRD profiles of (200) reflections will show only a single peak R(200) in the rhombohedral structure because all the planes of (200) share the same lattice parameters, whereas in the tetragonal phase the (200) reflections should be split into two peaks, the T(200)/(020) and T(002) profiles.[24,25] As shown in Figure 3, the experimental XRD data (○) of (200) reflection peaks were deconvoluted with the Gaussian functions. The (200) reflection is composed of three peaks at room temperature (Figure 3a). The dominant peak 1 corresponds to the rhombohedral phase, and peaks 2 and 3 represent the tetragonal phase. The ratio of rhombohedral to tetragonal phase in the perovskite structure was determined by eq 1 as follows:[26]

$$\mathrm{R/T} = I_{\mathrm{R}(200)}/[I_{\mathrm{T}(200)} + I_{\mathrm{T}(002)}] \qquad (1)$$

where $I_{\mathrm{R}(200)}$ is the intensity of the rhombohedral (200) reflections, and $I_{\mathrm{T}(200)}$ and $I_{\mathrm{T}(002)}$ are the intensities of tetragonal (200) and (002) reflections, respectively. According

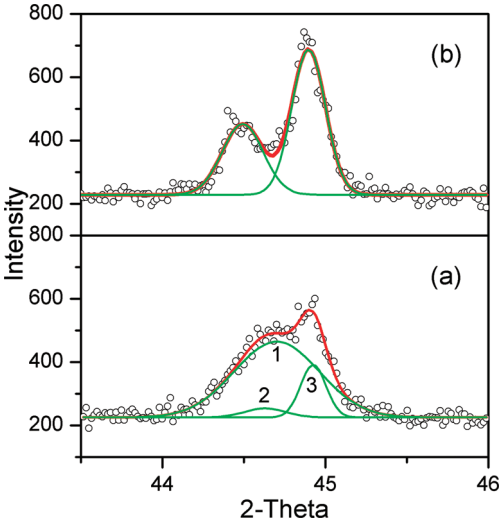

Figure 3. Pseudocubic (200) reflections (○) of the PYMNT(15/53/32) crystals: (a) at 25 °C, with a broad profile, indicating the coexistence of rhombohedral and tetragonal phases; (b) at 100 °C, indicating a pure tetragonal phase.

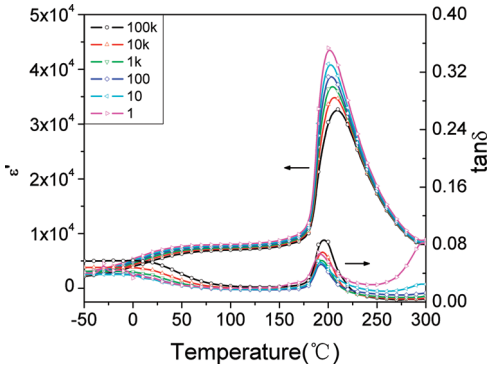

Figure 4. Variations of dielectric constant (ε') and loss tangent (tan δ) of an unpoled (001)-oriented PYMNT(15/53/32) single crystal platelet (thickness = 0.8 mm) measured upon heating at various frequencies.

to eq 1, the value of R/T is 3.8 at room temperature, indicating that the rhombohedral phase is the main phase at room temperature. This result confirms that the composition of the grown crystal within the MPB region. At 100 °C, the tetragonal symmetry can be clearly identified by two distinct peaks for the $(200)_{\mathrm{cub}}$ reflection, while the rhombohedral peak has completely disappeared (as shown in Figure 3b). In the tetragonal phase, the area ratio of the split right peak over left peak is 2:1, which arises from the intensity ratio of the T(200)/(020) (right) reflections over the T(002) (left) reflection. These results confirm that a phase transition from the rhombohedral to tetragonal structure takes place in the temperature range between 25 and 100 °C, corresponding to the morphotropic phase transition.

The variations of the dielectric constant (ε') and dielectric loss tangent (tan δ) of an unpoled (001)-oriented PYMNT(15/53/32) crystal platelet (thickness = 0.8 mm) as a function of temperature measured upon heating at various frequencies of 1 Hz, 10 Hz, 100 Hz, 1 kHz, 10 kHz, and 100 kHz are shown in Figure 4. The sharp peak of the dielectric constant indicates the Curie temperature T_{C} of the

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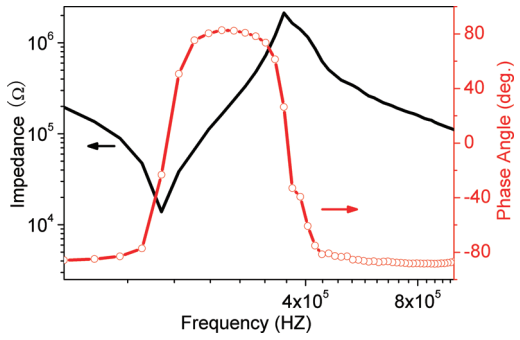

Figure 5. Impedance and phase angle as a function of frequency for the PYMNT(15/53/32) single crystal at 20 °C, showing the resonance (f_r = 164 285.4 Hz) and antiresonance (f_a = 349 999.7 Hz) frequencies.

crystal at 205 °C (at 1 kHz), which is much higher than that of the PMNT binary crystals and also slightly higher than that of the PIMNT ternary crystals.[20,21] In addition, a rather broad weak dielectric maximum was observed around 50 °C, which corresponds to the temperature (T_{rt}) of the morphotropic transition from the rhombohedral to the tetragonal phase, as revealed in the above-mentioned XRD analysis. It can also be seen that the temperature of maximum dielectric constant (T_{max}) is dependent on frequency, i.e., it shifts to higher temperatures with increasing frequencies (e.g., from T_{max} = 200 °C at 1 Hz to 210 °C at 100 kHz). This variation suggests some degrees of relaxational behavior, which is reminiscent of the typical relaxor properties of PMN and PYN, resulting from the dynamics of polar nanoregions (PNRs) that inherently exist in relaxors.[27,28] The relative permittivity ε' shows a maximum value of over 37 000 (at 1 kHz) at T_{max}, attesting to the good quality of the grown crystal. The room temperature values of the dielectric constant and loss tangent at 1 kHz are 6270 and 0.03, respectively. The high dielectric constant can be attributed to the relatively low T_{rt}.

The resonance (f_r) and antiresonance (f_a) frequencies for the PYMNT(15/53/32) crystal bar with dimensions of 1.0 × 1.0 × 3.7 mm^3 were measured at various temperatures by impedance spectroscopy. Figure 5 shows the impedance and phase angle as a function of frequency at 20 °C. The impedance spectrum shows two peaks at 164 285.4 and 349 999.7 Hz, indicating resonance frequencies f_r and f_a, respectively. From these values, the longitudinal electromechanical coupling coefficient k_{33} was calculated according to the IEEE Standards, as follows:[29,30]

$$k_{33}^2 = \frac{\Pi}{2}\frac{f_r}{f_a}\cot\left(\frac{\Pi}{2}\frac{f_r}{f_a}\right) \qquad (2)$$

Figure 6 shows the variation of the longitudinal electromechanical coupling factor k_{33} as a function of temperature. It can be seen that the value of k_{33} reaches 90% around room temperature, which is comparable to that of PIMNT crystals of MPB compositions. Upon heating,

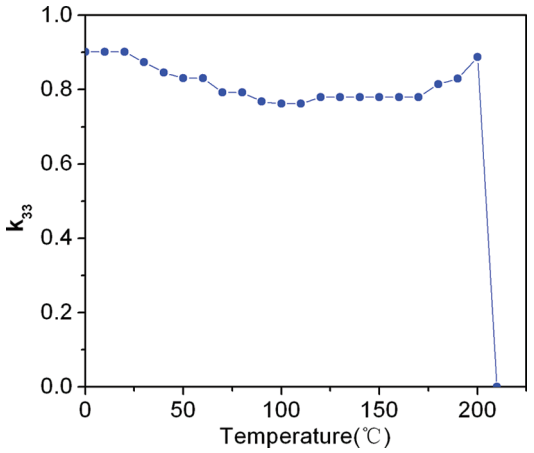

Figure 6. Temperature dependence of the longitudinal electromechanical coupling factor k_{33} of a (001)-oriented PYMNT(15/53/32) bar crystal.

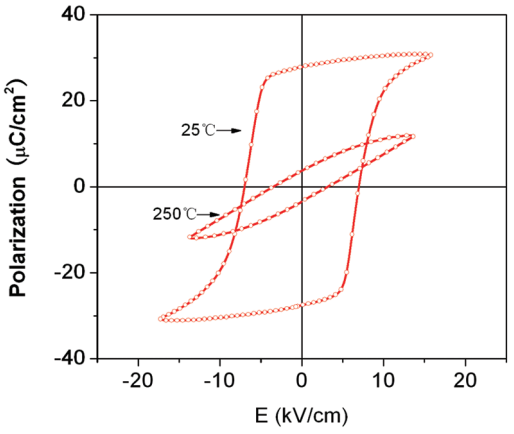

Figure 7. Ferroelectric hysteresis loops of a (001)-oriented PYMNT(15/53/32) single crystal measured at 25 and 250 °C, respectively (f = 2 Hz).

the value of k_{33} decreases gradually to its minimum value of 76% at about 100 °C, which is believed to result from partial depoling caused by the rhombohedral−tetragonal phase transition, as in the case of the PMNT crystals.[19] It then increases again upon further heating and reaches 90% at 200 °C, i.e., near the T_C, above which it steeply drops toward zero upon entering into the paraelectric phase. This result shows that in a wide temperature range between room temperature and 200 °C, the PYMNT(15/53/32) crystal exhibits a relatively high electromechanical coupling factor of 76−90%. This temperature range is much wider compared with the range of 20−80 °C for PMNT crystals or 20−95 °C for PZNT crystals, providing a much better temperature stability for the piezoelectric properties of the PYMNT crystals and a wider operating range. The piezoelectric coefficient d_{33} was also calculated to be 1980 pC/N based on the resonance and antiresonance frequencies, following the IEEE Standards,[29,30] which is close to the value of 1800 pC/N measured by the quasistatic method at room temperature. It is shown that the piezoelectric performance of PYMNT crystals is comparable to that reported for the PMNT and PIMNT crystals.

The polarization−electric field hysteresis loops of a (001)-oriented PYMNT(15/53/32) crystal platelet are displayed in

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Table 1. Comparison of the Properties of the (001)-Oriented PMNT, PIMNT, PMN-PZT, and PYMNT Crystals

crystals	ε'	T_C (°C)	T_{rt} (°C)	k_{33} (%)	d_{33} (pC/N)	E_c (kV/cm)	ref
PMNT(67/33)	4470	150	54	90	2000−2300		31
PMNT(70/30)		125		85	1240	2	8
PMN-PZT	3100	205	165	90	1050	5	32
PIMNT(24/44/32)	4290	150	110−117	83−92	900−1900	6	21
PIMNT(24/42/34)	4900	184	89	80	2200	6.8	33
PIMNT(28/40/32)	5200	192	119	92	1700−2200	10.88	34
PYMNT(15/53/32)	6270	205	50	90	1800	7	this work

Figure 7, which indicates the ferroelectricity with a coercive field E_c of 7 kV/cm and a remanent polarization Pr of 27.8 μC/cm^2 at room temperature. The polarization is saturated at an electric field of ±18 kV/cm at room temperature. It is worth noting that the strength of the coercive field E_c of PYMNT(15/53/32) crystals is about two times as high as that of PMNT crystals and comparable to that of PIMNT crystals, indicating a high stability for the poled state. Therefore, the PYMNT crystals is useful for high-power piezoelectric applications. Above T_C, P_r and E_c do not become zero. Instead, a residual remanent polarization of ~3.6 μC/cm^2 is to subsist even up to 250 °C. This kind of behavior is quite different from normal ferroelectrics whose remnant polarization and coercive field fall to zero in the vicinity of T_C but rather is typical of relaxor ferroelectric materials in which a macroscopic polarization can be induced.[27] Note that the electromechanical coupling factor k_{33} becomes zero at T_C, which is in support of the induced ferroelectric nature above T_C.

The comparison of the dielectric, piezoelectric, and ferroelectric properties of PMNT, PIMNT PMN-PZT, and PYMNT ferroelectric crystals is presented in Table 1. It can be seen that the PYMNT crystals exhibit excellent overall piezoelectric properties, with a high T_C, a high dielectric constant, a large coupling factor k_{33}, a high piezoelectric coefficient d_{33}, and a strong coercive field E_c.

Conclusions

Complex perovskite single crystals of ternary solid solution PYN-PMN-PT(15/53/32) have been successfully grown by the top-seeded solution growth method, with dimensions of 30 × 30 × 12 mm^3. The TSSG method produces ternary single crystals of quadrate morphology with large (001) faces, resulting from stable (100) two-dimensional growth. The composition of grown crystals is located near the MPB region, as revealed by the analysis of the XRD data. The characterization of the dielectric, piezoelectric, and ferroelectric properties of the PYMNT crystals shows that the Curie temperature T_C is 205 °C, which is higher than that of PMNT and PIMNT crystals so far reported. The piezoelectric coefficient d_{33} is found to be about 1800 pC/N, and the longitudinal electromechanical coupling factor k_{33} reaches 90% at room temperature. The remanent polarization and coercive field are found to be 27.8 μC/cm^2 and 7 kV/cm, respectively. Compared with the PMNT and PZNT so far developed, the PYMNT(15/53/32) ternary crystals show some advantages: a higher Curie temperature (205 °C) and a much larger coercive field (7 kV/cm). However, the T_{rt} of the grown crystals is relatively low, which limits their applicability. In order to increase the T_{rt}, the chemical composition of the PYN-PMN-PT ternary system needs to be tuned and optimized carefully and appropriate chemical doping may also be needed. In this way, the PYMNT crystals can become a promising candidate for advanced electromechanical transducer applications that require a wider operation temperature range and higher acoustic power.

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