Fabrication and ferroelectric/dielectric properties of La-doped PMN-PT ceramics with high optical transmittance

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Abstract

Relaxor ferroelectric $0.75(\text{Pb}_{1-3x/2}\text{La}_x)(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.25(\text{Pb}_{1-3x/2}\text{La}_x)\text{TiO}_3$ ($\text{La}^{3+}:\text{PMN-PT } x/75/25$, where $x = 2.8, 3.0, 3.5$, and 4.0 mol% of La^{3+}) transparent ceramics were fabricated by the combination of oxygen atmosphere pressureless sintering and hot-pressing sintering process. The optical transmittances of above four ceramics are higher than 60% at the wavelength of 500 - 900 nm. $\text{La}^{3+}:\text{PMN-PT } 3.0/75/25$ exhibits the highest transparency around 70% at 900 nm which is very close to the theoretical transmittance 71%. Each of the four ceramics exhibits the pure perovskite phases. They show fully dense microstructures and their relative densities are higher than 99.8%. The ferroelectric and dielectric measurements indicate that these four ceramics exhibit relaxation characteristics. With increasing La^{3+} content, (200) peak in XRD patterns shifts to higher angles and the average grain size increases, while the temperature $T_{\varepsilon_{\max}}$ corresponding to the maximum ε_r , the remanent polarizations P_r and coercive fields E_c decrease gradually.

Keywords: PMN-PT; Transparent ceramics; La doping; Relaxor ferroelectric

1. Introduction

Transparent ceramics have drawn greater attention due to their advantages compared to transparent single crystals, such as low cost, easy fabricating, good compositional control and so on [1]. Transparent ferroelectric ceramics possess great application prospects in the fields of electro-optical switches and electro-optical modulators owing to their notable electro-optic (EO) effect. La-doped PbZrTiO_3 (PLZT) transparent ferroelectric ceramics have been systemically investigated for many years because of high EO effect compared to LiNO_3 transparent single crystals [2–5]. However, PLZT transparent ferroelectric ceramics have large polarization-dependent scattering loss and significant field-induced detention, which restrain their application in high-frequency dynamic devices. The newly developed rare earth elements doped $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - PbTiO_3 (RE^{3+} :PMN-PT) transparent ceramics have stood out from the rest due to higher EO effect and weaker field-induced detention [6,7]. That means, RE^{3+} :PMN-PT transparent ceramics are more suitable for being applied in electro-optical modulators and high speed electro-optical switches. These outstanding properties make RE^{3+} :PMN-PT become a promising alternative to PLZT in some applications.

High optical transmittance is the key for transparent ceramics to be applied in optical devices. Up to now, Boston Applied Technologies Incorporated (BATI) is only the one that successfully fabricated transparent ceramics based on PMN-PT, with high optical transmittance 70%, to applying in such devices as variables optical attenuator, Q-switches and so on [6]. However, some details, like the high-optical-transmittance

composition, have not been mentioned. Owing to the complexity and poor repeatability of preparation technology, most of the PMN-PT transparent ceramics, fabricated by scientists from different countries, are still translucent and imperfect. Table 1 shows the research progress of PMN-PT transparent ceramics. One can see that the values of optical transmittance are not generally high [6-13]. However, low values can't meet the requirements of application. For this reason it is requisite to solve this problem to improve the optical transmittance of PMN-PT transparent ceramics now.

A pore-free microstructure (or porosity < 0.1%) needs to be developed in the process of fabricating transparent ceramics, because the probability of light scattering would increase and the value of optical transparency would drop when pores occur in the transparent ceramics [1,14]. It has been reported that oxygen has great solubility and diffusivity in PMN-PT system, and pores in ceramics can be easily removed by diffusion through the vacancies because of oxygen [5,15]. Moreover, the hot-pressing sintering process is beneficial to eliminate pores. In addition, it has been reported that the doping of rare-earth element is advantageous to optical and electrical properties of lead-based relaxors [16-18]. However, few works have been carried out to investigate the effect of La-doping on the optical, ferroelectric and dielectric properties of PMN-PT transparent ceramics, or the influence mechanism of La-doping on the performance is still not clear.

In this article, highly transparent La^{3+} :PMN-PT ceramics were fabricated successfully using combination of oxygen atmosphere pressureless sintering and hot-pressing sintering process. The influences of La-doping on microstructure, phase transition,

ferroelectric and dielectric properties of PMN-PT transparent ceramics were studied systematically.

2. Experimental procedure

The $0.75(\text{Pb}_{1-3x/2}\text{La}_x)(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.25(\text{Pb}_{1-3x/2}\text{La}_x)\text{TiO}_3$ transparent ceramics with different La-doping contents ($x = 2.8, 3.0, 3.5$, and 4.0 mol%) were prepared by the columbite precursor method to avoid pyrochlore phase [19]. They are denoted as $\text{La}^{3+}:\text{PMN-PT } x/75/25$ ($x = 2.8, 3.0, 3.5, 4.0$) ceramics in this paper. First, high-purity powders of Nb_2O_5 (99.99%; Aladdin) and excess MgO (99.99%; Aladdin) were mixed by using high energy ball-milled technique, then they were calcined at 1100°C for 3 h to form MgNb_2O_6 powders. This step effectively prevented the reaction of Nb_2O_5 and PbO , which is the key to avoid the formation of pyrochlore phase. Second, MgNb_2O_6 powders were mixed with high-purity powders of PbO (99.99%; Aladdin), TiO_2 (99.99%; Aladdin) and La_2O_3 (99.99%; Aladdin), then they were calcined at 800°C for 3 h, and finally PMN-PT powders can be obtained. Excess PbO was added to compensate the loss of PbO resulted from evaporation during high temperature sintering process. The PMN-PT powders were pressed into pellets with 13 mm in diameter. The pellets were sintered at 1200°C - 1260°C in the oxygen atmosphere firstly, and then sintered at 1200°C - 1240°C for 3 h with the uniaxial pressure of 100 MPa. Any temperature within this range fits for fabricating transparent ceramics. It is worth noting that these four transparent ceramics obtained in this paper were sintered at the same temperature of 1230°C .

The phase structures of $\text{La}^{3+}:\text{PMN-PT } x/75/25$ transparent ceramics were

characterized by X-ray diffraction (XRD, SmartLab, Rigaku, Tokyo, Japan). The microstructures were investigated by scanning electron microscopy (SEM, JSM-6390LV, JEOL, Tokyo, Japan). For electrical properties measurements, the gold electrodes were sputter-coated on a sample surface, using vacuum ion sputtering system. The room-temperature dielectric frequency spectra and dielectric temperature spectra were measured using precision impedance analyzer (4294A, Agilent, USA). The ferroelectric hysteresis P - E loops were characterized at room temperature on a ferroelectric test system (RT Premier II, Radiant, USA). All four ceramics were cut and polished (0.5 mm in thickness) to measure the optical transmittance using a UV-VIS spectrophotometer (UV-2550, Shimadzu, Tokyo, Japan).

3. Results and discussion

3.1. Optical transmittance

Fig. 1 shows the transmission spectra as a function of wavelength (300 - 900 nm) of La^{3+} :PMN-PT $x/75/25$ ($x = 2.8, 3.0, 3.5, 4.0$) transparent ceramics with thickness of 0.5 mm. The inset on the same figure shows the photography of obtained La^{3+} :PMN-PT ceramics. One can see that possibility of clearly seeing and legibility of the text underneath samples confirms their high transparency. From the transmission spectra, it is obviously found that all four ceramics have considerable optical transmittance at visible light wavelength and near-infrared wavelength. Especially La^{3+} :PMN-PT 3.0/75/25 exhibits the highest transparency around 70% in near-infrared wavelength and 67% at 633 nm wavelength, which is better than the data obtained by most research institutes [7-13]. The comparison of detailed data is shown in Table 1. Moreover, the

high optical transparency of $\text{La}^{3+}\text{:PMN-PT}$ 3.0/75/25 is comparable to that fabricated by BATI [6]. Detailed information, like the high-optical-transmittance composition, has not been mentioned by BATI. In addition, the transparency of other three ceramics obtained in this work is higher than 63% at 633 nm and 66% in near-infrared wavelength, which is also similar to the data reported in the published literature.

For an ideal transparent sample with a thickness t , the theoretical optical transmittance T can be described as the following equations [20]:

$$T = (1 - R)^2 \exp(-\beta t) \quad (1)$$

$$R = (n - 1)^2 / (n + 1)^2 \quad (2)$$

where n is the refractive index, R is the reflectivity, and β is the absorption and scattering index. It has been reported that the value of n can be considered to be a constant when the ratio of PMN/PT is fixed [13]. That means the theoretical optical transmittance T of all ceramics in this study can be regarded as approximately equal. It has been reported that n of PMN-PT ceramics is about 2.3 in near-infrared wavelength of 800 - 2500 nm, while it is 2.44 at visible light wavelength of 633 nm [7]. According to Eq. (1)-(2), the theoretical optical transmittance T is 71% in near-infrared wavelength and 68% at 633 nm wavelength. The $\text{La}^{3+}\text{:PMN-PT}$ 3.0/75/25 transparent ceramic obtained in this work has considerable transparency around 99% when neglecting the influence of twice air/ceramic interface reflection by appropriate antireflection coating, and such high transparency can fulfill the basic requirements of electro-optical devices.

3.2. Phase structure

Fig. 2 shows the XRD patterns of $\text{La}^{3+}\text{:PMN-PT}$ $x/75/25$ transparent ceramics. No

peak other than perovskite phase peaks can be observed in Fig. 2(a), implying that all La^{3+} :PMN-PT $x/75/25$ ceramics are pure and homogeneous without pyrochlore phase. The formation of pyrochlore phase was effectively suppressed by using the columbite precursor method. Since pyrochlore phase has different optical properties to the perovskite phase, light scattering is more likely to occur at the phase interface. Therefore, pure phase is the key point for obtaining the La^{3+} :PMN-PT ceramics with high optical transmittance. Fig. 2(b) shows the fine scanning XRD patterns corresponding to (200) peaks at $2\theta = 44.8 - 45.3^\circ$, in a function of La^{3+} dopant in PMN-PT ceramics (step - 0.002° , duration time - 2 seconds). It is clear that there is no obvious peak splitting for all La^{3+} :PMN-PT transparent ceramics. In addition, with increasing La^{3+} content, it is obviously found that (200) peaks shift to higher angles. That's because La^{3+} occupies A-site by replacing Pb^{2+} in PMN-PT ceramics. However, the ionic radius of La^{3+} is 0.1032 nm which is smaller than 0.1190 nm of Pb^{2+} , so this occupation leads to lattice distortion and reduces the interplanar spacing d . According to Bragg equation $2d\sin\theta=\lambda$, when interplanar spacing d decreases, θ will increase, which means diffraction peaks shift to higher angles.

3.3. Density and microstructure

Cell parameters and relative density of La^{3+} :PMN-PT $x/75/25$ transparent ceramics are shown in Table 2. Cell parameters were calculated from the XRD results and the bulk density was measured using Archimedes method. The relative densities of all four ceramics are greater than 99.8% of the theoretical density, as shown in Table 2. It's well-known that possessing high density is the critical step for fabricating transparent

ceramics, which is well verified by La^{3+} :PMN-PT 3.0/75/25 transparent ceramic in this study. La^{3+} :PMN-PT 3.0/75/25 shows the highest optical transmittance and has the highest value of relative density. Nonporous structure can improve the optical transmittance because pores scatter incident light as scattering centers in ceramics. In this work, the combination of oxygen atmosphere pressureless sintering and hot-pressing sintering process were implemented to fabricate La^{3+} :PMN-PT transparent ceramics. It has been reported that oxygen has high solubility and diffusivity in PMN-PT system, and the pores in ceramics filled with oxygen are easily removed through diffusion of vacancies formed by PbO evaporation [5,15]. Moreover, the hot-pressing sintering process is beneficial to eliminate pores, so the La^{3+} :PMN-PT transparent ceramics become denser and more transparent.

The high densification of La^{3+} :PMN-PT x /75/25 transparent ceramics is further confirmed by SEM observation as shown in Fig. 3. The fractured surfaces in case of all ceramics exhibit similar microstructure. The grain boundaries are clear and pores are hardly observed in each of the obtained ceramics, not only in the region as shown in the images, but also in other regions not shown here. All ceramics composition mostly show intergranular fracture character, which could be attributed to the presence of La^{3+} . According to Winter's research [18], PMN-PT ceramic shows transgranular fracture character without La^{3+} doping, while intergranular fracture character occurs gradually with the increase of La^{3+} content. Lanthanum ions influences on the increasing of the nonuniformity in the compositions between grain boundaries and grains, which leads to increased amount of weak grain-boundary phase, so the intergranular fracture is more

likely to occur. Fig. 4 shows the distribution of grain sizes of $\text{La}^{3+}\text{:PMN-PT } x/75/25$ transparent ceramics, which were obtained from their SEM images and the statistics were obtained for the population of 100 grains. The grain sizes of all four ceramics distribute between 1.5 μm to 11.4 μm . From Fig. 4, it is obviously found that the grain sizes of four ceramics largely concentrated in 3-7 μm . With the increase of La^{3+} content, the percentage in 3-5 μm decreases gradually, while the percentage in 7-9 μm increases. After calculating, it's found that the average grain sizes of $\text{La}^{3+}\text{:PMN-PT } x/75/25$ ($x = 2.8, 3.0, 3.5, 4.0$) transparent ceramics have a slight increasing trend, and they are approximately 4.58, 4.61, 4.98, and 5.18 μm , respectively.

3.4. Ferroelectric and dielectric properties

Fig. 5 shows the ferroelectric hysteresis (P - E) loops of $\text{La}^{3+}\text{:PMN-PT } x/75/25$ ($x = 2.8, 3.0, 3.5, 4.0$) transparent ceramics, recorded at the room temperature. Rectangular hysteresis loops are observed for $\text{La}^{3+}\text{:PMN-PT } 2.8/75/25$ transparent ceramic with large values of coercive field (E_c) and remnant polarization (P_r). That means the behavior of transparent $\text{La}^{3+}\text{:PMN-PT } 2.8/75/25$ ceramic is close to normal ferroelectric. With the increase of La^{3+} content, the P - E loops of $\text{La}^{3+}\text{:PMN-PT } x/75/25$ transparent ceramics become more inclined and slimmer, especially for $x = 4.0$ % of dopant. In this case it possesses the slimmest P - E loops and exhibits the typical characteristic behavior of a relaxor ferroelectric. The descending trends of P_r and E_c are shown in Fig. 6. It's obviously found that ferroelectric property of $\text{La}^{3+}\text{:PMN-PT } x/75/25$ ceramics becomes weaker with increasing La^{3+} doping content. Undoubtedly, the variation of ferroelectric property has an inseparable relation with La^{3+} doping. Because La^{3+} can weak the

long-range coupling interaction between the ferroelectric BO_6 octahedrons in ABO_3 perovskite-type structure [21,22]. PMN-PT can be regarded as a network of BO_6 octahedrons, where Mg^{2+} , Nb^{5+} , and Ti^{4+} are located inside octahedrons as B-site cations and A-site cations Pb^{2+} are positioned in the interstitial site of octahedrons. According to Thomas' study [21], in La-undoped PMN-PT ceramics, NbO_6 and TiO_6 octahedrons have ferroelectric activity and couple with each other through A-site cations Pb^{2+} , and the coupling is sufficiently strong to offer a normal ferroelectric response. In other words, the presence of Pb^{2+} is a requirement for ferroelectric property besides ferroelectric BO_6 octahedrons in PMN-PT system. Due to the doping of La^{3+} in PMN-PT ceramics, La^{3+} occupies A-site by replacing Pb^{2+} . That indicates the amount of the medium which connects ferroelectric BO_6 octahedrons decreases. Therefore, the long-range coupling interaction among ferroelectric BO_6 octahedrons becomes weaker with increasing La^{3+} doping content. Thus the P - E loops become more inclined and slimmer, the values of P_r and E_c decrease, and the transformation from normal ferroelectric to relaxor ferroelectric occurs.

Fig. 7 shows the dielectric properties of La^{3+} :PMN-PT $x/75/25$ transparent ceramics. At the room temperature, the relative dielectric constants ϵ_r as a function of frequency are shown in Fig. 7(a). It is obviously observed that the relative dielectric constants ϵ_r of La^{3+} :PMN-PT $x/75/25$ ($x = 2.8, 3.0, 3.5, 4.0$) transparent ceramics increase with the increasing of La^{3+} content, which are 15792、18117、21383 and 29339 at 1 kHz, respectively. Fig. 7(b)-(e) show the relative dielectric constants ϵ_r as a function of temperature with frequencies ranging from 0.1 kHz to 100 kHz. All ceramics show the

typical relaxor characteristic: broad peaks and frequency dispersion. As said above, La^{3+} doping can break the coupling among ferroelectric BO_6 octahedrons, and this decoupling effect results in the formation of polar nanodomains or clusters, which is considered to be responsible for the relaxor characteristics [23]. As shown in Fig. 7(f), when the La^{3+} amount increases from 2.8 mol% to 4.0 mol%, the temperatures $T_{\varepsilon_{\max}}$ corresponding to the maximum ε_r have a downward trend, from 60°C、55°C、46°C to 32°C respectively. According to Kim's research [16], when doping content of La^{3+} increases by 1 mol%, $T_{\varepsilon_{\max}}$ decreases by about 25°C. Therefore, the results in this study match well with Kim's. In addition, similar decreasing tendency of $T_{\varepsilon_{\max}}$ by La^{3+} doping was also observed previously in our experiment and other researches [16-18,24]. Therefore, the downtrend of $T_{\varepsilon_{\max}}$ verifies that La^{3+} doping can weaken the long-range coupling interaction among the BO_6 octahedrons and suppress the ferroelectric and dielectric properties once again.

4. Conclusions

Highly transparent $\text{La}^{3+}\text{:PMN-PT } x/75/25$ ($x = 2.8, 3.0, 3.5$, and 4.0 mol%) ceramics with high density and pure perovskite phase were fabricated using combination of oxygen atmosphere pressureless sintering and hot-pressing sintering process. $\text{La}^{3+}\text{:PMN-PT } 3.0/75/25$ exhibits the highest transparency around 70% in near-infrared wavelength and 67% at 633 nm wavelength which are very close to its theoretical transmittance. Due to the increase of the amount of weak grain-boundary phase caused by La^{3+} doping, all ceramics mostly show intergranular fracture in SEM micrographs. Moreover, pores are hardly found and grains are closely packed, and the relative

densities of all ceramics are above 99.8%. The ionic radius of La^{3+} is smaller than the value of Pb^{2+} , which results in the interplanar spacing d reducing and diffraction peaks shifting to higher angles with the increase of La^{3+} content. La^{3+} doping weakens the long-range coupling interaction among ferroelectric BO_6 octahedrons, so the temperature $T_{\varepsilon_{\max}}$ corresponding to the maximum ε_r shifts from 60°C to 32°C, the values of remanent polarizations P_r and coercive fields E_c decrease gradually, and transforms from normal ferroelectric to relaxor ferroelectric with La^{3+} content increasing gradually.

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References

- [1] G. H. Haertling, C. E. Land, Hot-Pressed (Pb, La)(Zr, Ti)O₃ Ferroelectric Ceramics for Electro-optic Applications, J. Am. Ceram. Soc. 54 (1971) 1-11.
- [2] X. N. Zhang, B. Xia, X. Zeng, P. S. Qiu, W. X. Cheng, X. Y. He, Temperature dependence of electric-induced light scattering performance for PLZT ceramics, J. Am. Ceram. Soc. 97 (2014) 1389-1392.
- [3] M. Nakada, K. Ohashi, J. Akedo, Optical and electro-optical properties of Pb(Zr,Ti)O₃ and (Pb,Lu)(Zr,Ti)O₃ films prepared by aerosol deposition method, J. Cryst. Growth. 275 (2005) 1275-1280.
- [4] G. S. Snow, Fabrication of transparent electro-optic PLZT ceramics by atmosphere sintering, J. Am. Ceram. Soc. 56 (1973) 91-96.
- [5] Y. Yoshikawa, K. Tsuzuki, Fabrication of Transparent Lead Lanthanum Zirconate Titanate Ceramics from Fine Powders by Two-Stage Sintering, J. Am. Ceram. Soc. 75 (1992) 2520-2528.
- [6] H. Jiang, Y. K. Zou, Q. Chen, K. K. Li, R. Zhang, Y. Wang, H. Ming, Z. Q. Zheng, Transparent electro-optic ceramics and devices, Proc. SPIE 5644 (2005) 380-394.
- [7] W. Ruan, G. R. Li, J. T. Zeng, J. J. Bian, L. S. Kamzina, H. R. Zeng, L. Y. Zheng, A. L. Ding, Large electro-optic effect in La-doped 0.75Pb (Mg_{1/3}Nb_{2/3})O₃-0.25PbTiO₃ transparent ceramic by two-stage sintering, J. Am. Ceram. Soc. 93 (2010) 2128-2131.
- [8] K. Uchino, Electro-optic ceramics and their display applications, Ceram. Int. 21 (1995) 309-315.
- [9] D. A. McHenry, J. Giniewicz, S. J. Jang, A. Bhalla, T. R. Shroud, Optical properties of hot pressed relaxor ferroelectrics, Ferroelectrics 93 (1989) 351-359.
- [10] F. A. L. Badillo, J. A. Eiras, F. P. Milton, D. Garcia, Preparation and Microstructural, Structural,

Optical and Electro-Optical Properties of La Doped PMN-PT Transparent Ceramics, *Opt. Photon. J.* 2 (2012) 157-162.

[11] I. Fujii, R. Yoshida, T. Imai, S. Yamazoe, T. Wada, Fabrication of Transparent $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$ Based Ceramics by Conventional Sintering, *J. Am. Ceram. Soc.* 96 (2013) 3782-3787.

[12] S. Y. Si, C. Z. Zhang, X. H. Li, W. M. Lv, Y. L. Wang, Y. C. Zhang, W. N. Ye, C. J. Lu, Microstructure and analysis on the high transparent 0.88PMN-0.12PT ferroelectric ceramics prepared by pressureless sintering, *J. Chin. Electr. Mic. Soc.* 33 (2014) 495-498.

[13] W. L. Ji, X. Y. He, X. Zeng, W. X. Cheng, P. S. Qiu, B. Xia, D. Wang, Effects of PMN/PT ratio on optical and electro-optic properties of PLMNT transparent ceramics, *Ceram. Int.* 41 (2015) 10387-10393.

[14] Z. Z. Yang, Y. C. Zhang, W. N. Ye, C. J. Lu, Light Scattering by Residual Pores and Grain Boundaries in Transparent 0.65PMN-0.35PT Ceramic, *J. QDU. (Natural Science Edition)* 25 (2012) 42-46.

[15] J. J. Choi, J. Ryu, H. E. Kim. Microstructural evolution of transparent PLZT ceramics sintered in air and oxygen atmospheres, *J. Am. Ceram. Soc.* 84 (2001) 1465-1469.

[16] N. Kim, W. Huebner, S. J. Jang, T. R. Shrout, Dielectric and piezoelectric properties of lanthanum-modified lead magnesium niobium-lead titanate ceramics, *Ferroelectrics* 93 (1989) 341-349.

[17] W. Ruan, G. Li, J. Zeng, L. S. Kamzina, H. R. Zeng, K. Y. Zhao, L. Y. Zheng, A. L. Ding, Origin of the giant electro-optic Kerr effect in La-doped 75PMN-25PT transparent ceramics, *J. Appl. Phys.* 110 (2011) 074109 7pp.

- [18] M. R. Winter, S. M. Pilgrim, M. Lejeune. Study on the Effects of Lanthanum Doping on the Microstructure and Dielectric Properties of $0.9\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - 0.1PbTiO_3 , J. Am. Ceram. Soc. 84 (2001) 314-320.
- [19] S. L. Swartz, T. R. Shrout, Fabrication of Perovskite Lead Magnesium Niobate, Mater. Res. Bull. 17 (1982) 1245–1250.
- [20] J. G. J. Peelen, R. Metselaar, Light scattering by pores in polycrystalline materials: Transmission properties of alumina, J. Appl. Phys. 45 (1974) 216-220.
- [21] N. W. Thomas, A new framework for understanding relaxor ferroelectrics, J. Phys. Chem. Solids. 51 (1990) 1419-1431.
- [22] X. Dai, D. Viehland. Effects of lanthanum modification on the antiferroelectric - ferroelectric stability of high zirconium-content lead zirconate titanate, J. Appl. Phys. 76 (1994) 3701-3709.
- [23] S. M. Gupta, D. Viehland. Compositional Studies of Lanthanum-Modified Morphotropic Phase Boundary $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - PbTiO_3 Ceramics, J. Am. Ceram. Soc. 80 (1997) 477-485.
- [24] J. R. Zhang, Y. C. Zhang, C. J. Lu, W. N. Ye, J. Su, Effect of La-doping content on the dielectric and ferroelectric properties of $0.88\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - 0.12PbTiO_3 ceramics, J. Mater. Sci: Mater. Electron. 25 (2014) 653-658.

Figure Captions

Fig. 1. Optical transmission spectra of La^{3+} :PMN-PT $x/75/25$, with a thickness of samples 0.5 mm.

The inset is the photography of the obtained transparent ceramics.

Fig. 2. (a) XRD patterns of La^{3+} :PMN-PT $x/75/25$ transparent ceramics, (b) the fine-scanning patterns showing the shift of (200) peaks in a function of lanthanum-dopant.

Fig. 3. SEM micrographs of the fractured surfaces of the La^{3+} :PMN-PT $x/75/25$ transparent ceramics.

Fig. 4. (a-d) Grain size distribution for the La^{3+} :PMN-PT $x/75/25$ transparent ceramics.

Fig. 5. Ferroelectric hysteresis loops (P - E) recorded for La^{3+} :PMN-PT $x/75/25$, measured at the room temperature and the frequency $f = 10$ Hz.

Fig. 6. La-content dependences of remanent polarizations P_r and coercive fields E_c .

Fig. 7. Dielectric properties of La^{3+} :PMN-PT $x/75/25$ transparent ceramics. (a) Frequency dependence of relative dielectric constant measured at the room temperature, (b-e) temperature dependence of relative dielectric constant, (f) La-content dependences of ε_r (measured at the room temperature) and $T\varepsilon_{max}$.

Table 1

Comparison of Transmittance of Various PMN-PT Ceramics

Time	Component	Transmittance		Thickness	Reference
		@ 633 nm	@ NIR ^{††}		
1989	0.9PMN-0.1PT	52% [†]	61% [†]	1.03 mm	[9]
1995	0.88PMN-0.12PT	49%	—	0.5 mm	[8]
2005	PMN-PT	67% [†]	70% [†]	1.45 mm	[6]
2010	0.75PMN-0.25PT	58% [†]	65%	0.5 mm	[7]
2012	<i>x</i> PMN-(1- <i>x</i>)PT	43% [†]	50% [†]	0.63 mm	[10]
2013	0.9PMN-0.1PT	58% [†]	68%	0.5 mm	[11]
2014	0.88PMN-0.12PT	—	67%	0.8 mm	[12]
2015	0.78PMN-0.22PT	63% [†]	66% [†]	0.35 mm	[13]
2016	0.75PMN-0.25PT	67%	70%	0.5 mm	This work

[†]The optical transmittances were estimated from their optical transmission spectrum. ^{††}Near-infrared waveband.

Table 2 Cell Parameters and Densities of La^{3+} :PMN-PT Transparent Ceramics

La content	Cell parameters (Å)		Cell volume (Å ³)	Theoretical density (g/cm ³)	Bulk density (g/cm ³)	Relative density
	<i>a</i> = <i>b</i>	<i>c</i>				
2.8	4.0227	4.0238	65.1136	8.1072	8.1023	99.94%
3.0	4.0222	4.0231	65.0861	8.1071	8.1039	99.96%
3.5	4.0215	4.0221	65.0473	8.1032	8.0902	99.84%
4.0	4.0207	4.0208	65.0004	8.1003	8.0849	99.81%