

Dielectric, ferroelectric and ferromagnetic properties of 0.8PbFe_{0.5}Ta_{0.5}O₃-0.2PbTiO₃ ceramics

Sung Yung Cho · Jin Soo Kim · M. S. Jang

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Abstract Lead based complex compounds, 0.8PbFe_{0.5}–Ta_{0.5}O₃-0.2PbTiO₃ (0.8PFT-0.2PT) ceramics, were prepared by the solid state reaction method, and the corresponding dielectric, ferroelectric and ferromagnetic properties were investigated. As the PT phase was added to the PFT phase, the Curie temperature of 0.8PFT-0.2PT ceramics increased. The ferroelectric P-E and ferromagnetic M-H hysteresis loops were observed at the same time. The ferroelectric properties depend on the formation of the perovskite 0.8PFT-0.2PT phase; however, the ferromagnetic properties depend on the formation of the pyrochlore Pb₃FeTaO₇ phase.

Keywords Ferroelectric · Dielectric · Relaxor · Perovskite · Lead based complex compound · PMN · PFT

1 Introduction

Lead based compounds A(B', B'')O₃ show ferroelectric relaxor behaviors [1, 2]. The relaxor behaviors were explained by the partially ordered B' and B in the perovskite structure. The ferroelectrics relaxor behaviors were well known for various solid solutions such as PbMn_{1/3}Nb_{2/3}O₃-PbTiO₃ (PMN-PT), PbMn_{1/3}Nb_{2/3}O₃-PbMg_{2/3}W_{1/3}O₃-PbTiO₃ (PMN-PMW-PT) and PbZn_{1/3}Nb_{2/3}O₃-PbTiO₃ (PZN-PT) [2]. However, such lead based compounds are difficult to

fabricate without the formation of a pyrochlore phase that can be determinate to the dielectric properties [3]. One of them, PbFe_{0.5}Ta_{0.5}O₃ (PFT), is a compound belonging to the lead based complex perovskites with a general formula Pb(B_{1/2}³⁺, B_{1/2}⁵⁺)O₃ [4]. Next, Fe and Ta ions are believed to randomly occupy the B sites of ABO₃ perovskite structure, which caused the relaxor behaviors. On the other hand, PbTiO₃ (PT) is a normal ferroelectrics with a sharp dielectric peak at $T_c \approx 500^\circ\text{C}$. In this study, PbFe_{0.5}Ta_{0.5}O₃-PbTiO₃ solid solutions systems [5, 6] were prepared, and these ceramics exhibit ferroelectric P-E and ferromagnetic M-H hysteresis loops at the same time. Referring to the formation of the pyrochlore Pb₃FeTaO₇, the dielectric, ferroelectric and ferromagnetic properties of the 0.8PbFe_{0.5}Ta_{0.5}O₃-0.2PbTiO₃ were investigated.

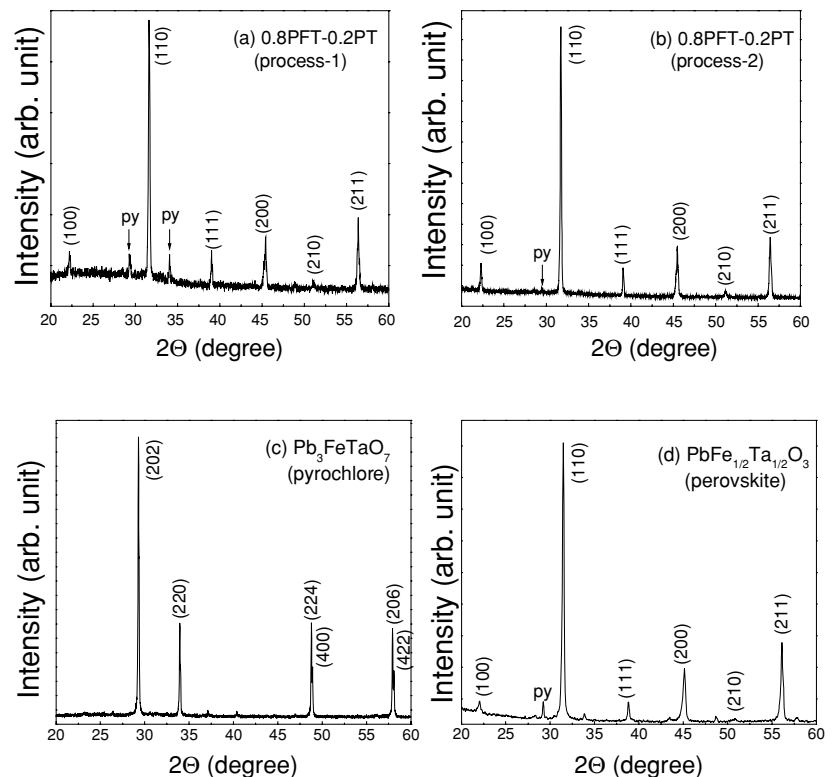
2 Experimental work

PbFe_{0.5}Ta_{0.5}O₃ (PFT), 0.8PbFe_{0.5}Ta_{0.5}O₃-0.2PbTiO₃ (0.8PFT-0.2PT) and Pb₃FeTaO₇ ceramics were prepared by the solid state reaction method. The raw materials for the PFT and 0.8PFT-0.2PT and Pb₃FeTaO₇ ceramics were lead monoxide (99.9%), α -iron Oxide (99.9%), titanium oxide (99.95 %) and tantalum oxide (99.99%). The mixed raw materials firstly calcined at 950°C for 5 h, and secondly calcined at 1100°C for 5 hours. The calcined powders were ground and mixed in a jar. In addition to a dry mixed compound (process-1) of the 0.8PFT-0.2PT compositions, an alcohol mixed compounds (process-2) was also prepared. The powders were pressed into pellets with 15 mm diam and 2.5 mm thick at a pressure of about 50 kg/cm². The ceramic pellets were sintered at 1170°C for 5 h in air. A Pt electrode was coated on the ceramic samples by DC sputtering. The capacitance and loss tangent were measured

S. Y. Cho · J. S. Kim · M. S. Jang
Department of Physics, and Research Center for Dielectric and Advanced Matter Physics, Pusan National University, Busan 609-735, Korea

J. S. Kim (✉)
School of Electrical Engineering, University of Ulsan, Ulsan 680-749, Korea
e-mail: kimjjin@pusan.ac.kr

Fig. 1 XRD patterns of the PFT and 0.8PFT-0.2PT (process-1 and Process-2) and $\text{Pb}_3\text{FeTaO}_7$



by an impedance analyzer (HP4194A). Ceramic samples were heated with a ratio of $1^\circ\text{C}/\text{min}$ and measured over the frequency range of 100 Hz–1 MHz. Ferroelectric P-E and ferromagnetic M-H hysteresis loops were measured by using a Sawyer-Tower circuit and a VSM (Vibrating Sample Magnetometer), respectively.

3 Results and discussion

The phase formation and crystal structure were examined by X-ray diffraction (XRD) [7, 8]. Figure 1 shows the XRD patterns of the PFT, 0.8PFT-0.2PT and $\text{Pb}_3\text{FeTaO}_7$ ceramics. The PFT-PT phase is accompanied by the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase and PbO phase. Previously, it was understood that the lead-based ferroelectric relaxors inevitably involved the formation of the pyrochlore phase [9]. In this study, the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ and PbO phases formed as well as the perovskite 0.8PFT-0.2PT phase, and the formation of mixed phases depended on various processing factors.

In addition to the dry mixed compound (process-1), the alcohol mixed compound (process-2) was also prepared. Compared to process-1, the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase decreased when utilizing process-2. The formation of the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase depends on such factors as the sintering temperature, raw material composition and processing procedure. Previously, it was reported that the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase decreased at sintering temperatures above

950°C [9]. However, the mechanism of the phase formation is different when examined in detail. In addition to the preparation of the 0.8PFT-0.2PT ceramics, under the same process, the $\text{Pb}_3\text{FeTaO}_7$ ceramics with only pyrochlore phases were also prepared to explain the ferromagnetic properties. Under these conditions, the sintering temperatures of the $\text{Pb}_3\text{FeTaO}_7$ ceramics were 700°C , 950°C , 1170°C and 1200°C at 6 h. Below the sintering temperature of 1170°C , the prepared ceramics show mixed phases of PbO, $\text{Pb}_3\text{FeTaO}_7$ and PFT. After being sintered at 1200°C , only the well-defined $\text{Pb}_3\text{FeTaO}_7$ pyrochlore phase remained.

Figure 2 shows the temperature dependence of the dielectric constant of the PFT and 0.8PFT-0.2PT ceramics

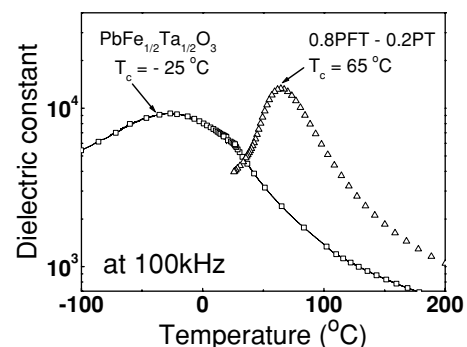
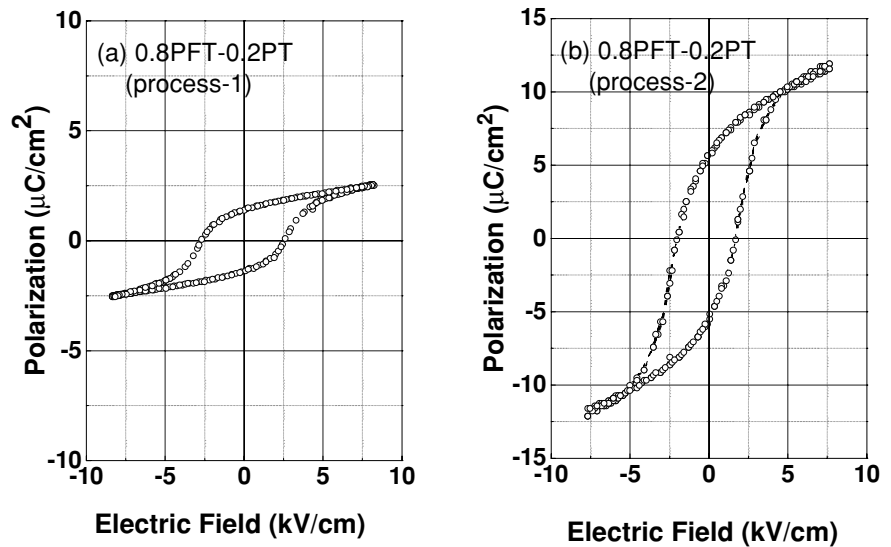


Fig. 2 Temperature dependent dielectric constant of the PFT and 0.8PFT-0.2PT measured at 100 kHz

Fig. 3 Ferroelectric P-E hysteresis loops of the 0.8PFT-0.2PT ceramics measured at room temperature



measured at a frequency of 100 kHz. The dielectric anomaly of the PFT ceramic was observed at about -25°C [6, 10]. The phase transition shows a broad dielectric constant maximum peak, which agrees with the previous results [10]. Previously, the PFT was known to the relaxor ferroelectrics [11], which was related to the disordered state of Fe and Ta ions in the B-site cation.

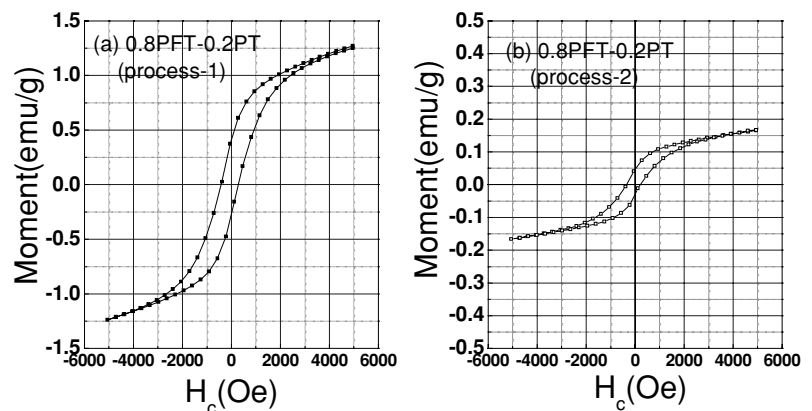
As the PT phase ($T_c = 500^{\circ}\text{C}$) added to the the PFT phase ($T_c = -25^{\circ}\text{C}$), the T_c of 0.8PFT-0.2PT ceramics increased, and the corresponding phase transition showed a sharp dielectric constant maximum peak. The phase transition behaviors of PFT-PT ceramics are similar to that of PMN-PT ferroelectrics relaxors [12, 13] in the view of a solid solution system of a relaxor (PMN)—a normal ferroelectrics (PbTiO_3).

In addition to the dielectric properties, the ferroelectric and ferromagnetic properties of 0.8PFT:0.2PT ceramics remarkably depend on the formation of the perovskite phase and pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase. Figure 3 shows the ferroelectric

P-E hysteresis loops of 0.8PFT-0.2PT ceramics prepared by process-1 and process-2. Well-saturated ferroelectric P-E hysteresis loops were observed in both. At an applied electrical field of about 7 kV/cm, the remnant polarization P_r of the 0.8PFT:0.2PT ceramics prepared by process-1 and process-2 is $5 \mu\text{C}/\text{cm}^2$ and $2 \mu\text{C}/\text{cm}^2$, respectively. Thus, the remnant polarization of 0.8PFT:0.2PT ceramics with in a low pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase was remarkably enhanced [14].

Figure 4 shows the ferromagnetic M-H hysteresis loops of 0.8PFT:0.2PT ceramics prepared by process-1 and process-2. Well-saturated ferromagnetic M-H hysteresis loops were observed in both. The magnetic moment M_r of 0.8PFT:0.2PT ceramics prepared by process-1 was higher than that of 0.8PFT:0.2PT ceramics prepared by process-2. At the applied magnetic field of 5000 Oe, the M_r of 0.8PFT:0.2PT ceramics by process-1 was 0.5 emu/g, which is higher than that of 0.8PFT:0.2PT ceramics prepared by process-2 with $M_r = 0.05 \text{ emu/g}$.

Fig. 4 Ferromagnetic M-H hysteresis loops of the 0.8PFT-0.2PT ceramics measured at room temperature



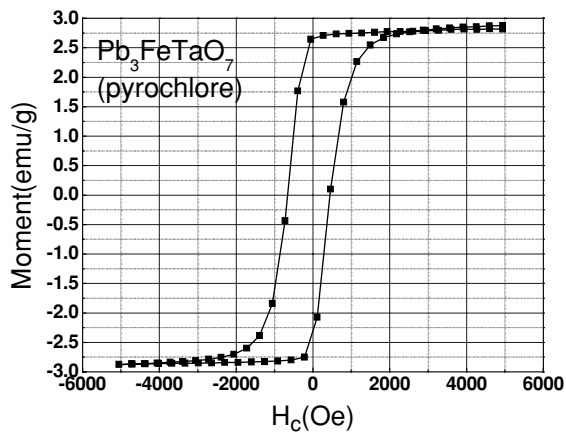


Fig. 5 Ferromagnetic M-H hysteresis loops of the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase

Regarding the results of the XRD, P-E and M-H hysteresis loops, the ferroelectric and ferromagnetic properties depend on the formation of the perovskite 0.8PFT:0.2PT and the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phases.

Figure 5 shows the ferromagnetic M-H hysteresis loops of the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase. Well-saturated ferromagnetic M-H hysteresis loops were observed, and the magnetic moment M_r was 2.7 emu/g. Thus, the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase contributed to the ferromagnetic properties.

For the preparation of the lead complex compound 0.8PFT:0.2PT ceramics, the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase was formed as well as the perovskite 0.8PFT:0.2PT phase. The lead-based ferroelectrics inevitably involved the pyrochlore phase, and the formation of pyrochlore phases depended on processing factors. The degree of the perovskite 0.8PFT:0.2PT and the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phases contributed to the ferroelectric and ferromagnetic properties, respectively.

4 Conclusions

The dielectric, ferroelectric and ferromagnetic properties of the lead complex compound 0.8PFT:0.2PT ceramics were investigated. The Curie temperature of 0.8PFT-0.2PT ceramic was 65°C , which is higher than that of PFT ceramics. For the preparation of 0.8PFT:0.2PT ceramics, the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase was also formed. Then, the ferroelectric P-E and ferromagnetic M-H hysteresis loops were observed. The formation of the perovskite phase contributed to the ferroelectric properties. However, the formation of the pyrochlore $\text{Pb}_3\text{FeTaO}_7$ phase contributed to the ferromagnetic properties.

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