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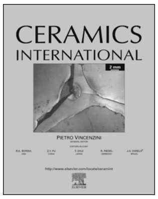

Large strain in poled and aged Mn-doped $Pb(Mn_{1/3}Sb_{2/3})O_3$–Pb(Zr,Ti)O_3 ferroelectric ceramics under a low electric field

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ABSTRACT

Reversible domain switching in aged acceptor-doped ferroelectric ceramics is a powerful method to achieve electrostrain due to the exchange of nonequal crystalline axes of domains. Nevertheless, previous work suggested that the strain obtained via reversible domain switching in unpoled ceramics was much lower than theoretical value. In this paper, we demonstrate that the strain in poled and aged acceptor-doped ferroelectric ceramics can be significantly enhanced when the driving electric field is perpendicular to the poling direction, originating from the exchange of nonequel crystalline axes of more domains. Consequently, the electrostrain of poled and aged 0.1 wt% MnO_2-doped $0.05Pb(Mn_{1/3}Sb_{2/3})O_3-0.50PbZrO_3-0.45PbTiO_3$ could reach 0.225% which was 4.17 times larger than that of unpoled sample under a low electric field of 15 kV cm^{-1}. Moreover, the corresponding normalized strain d_{33}^{*} reached 1500 pm V^{-1} which significantly exceeded the values reported in ferroelectric ceramics. Our work provide a general effective method to enhance the electrostrain behavior via reversible domain switching in aged acceptor-doped ferroelectric ceramics.

1. Introduction

Actuators which are based on electric field-induced-strain behavior of ferroelectric materials are widely used in ultrasound motors, fuel injectors and ink cartridges [1,2]. Therefore, the actuator materials, especially ferroelectric ceramics which can generate large electrostrain, have attracted considerable research attention in recent years [3–9]. Generally, converse piezoelectric effect, electrostrictive effect and phase transition from antiferroelectric to ferroelectric are the conventional mechanisms to realize electrostrain in ferroelectric ceramics [5,8,9]. Recently, a novel mechanism based on reversible domain switching in aged acceptor-doped ferroelectric ceramics was proposed to achieve electrostrain originating from the exchange of nonequal crystalline axes of domains [10]. However, previous studies about this novel mechanism were mainly focused on unpoled ceramics [10–13], which could only utilize part of the domains to exchange nonequal crystalline axes to generate strain due to the random orientations of domains in polycrystal ceramics, as demonstrated in Fig. 1(a). The domains whose polarization orientations are parallel to the external electric field cannot exchange their nonequal crystalline axes to contribute to the strain.

Here we proposed an effective approach to enhance the electrostrin performance: (1) Firstly, the acceptor-doped ceramics should be poled; (2) Secondly, the ceramics needed to be aged for enough time; (3) Finally, the applied electric field should be perpendicular to the poling direction, as illustrated in Fig. 1(b). According to this approach, more domains of the ceramics can switch to exchange nonequal crystalline axes, which will result in a larger electrostrain.

Mn-doped $Pb(Mn_{1/3}Sb_{2/3})O_3$–Pb(Zr, Ti)O_3 (abbreviated as Mn-doped PMS-PZT) ceramics are typical acceptor-doped ferroelectric ceramics. In previous studies, it was found that they exhibited large strain memory effect and linear temperature scaling behavior of ferroelectric hysteresis [2,14]. Additionally, the Mn-doped PMS-PZT ceramics possessed a low coercive field of about 11 kV cm^{-1}, indicating that the domains could be driven to switch under a low electric field. As a consequence, it is expected that a large strain under a low electric field can be obtained in the Mn-doped PMS-PZT ceramics through our approach proposed above.

In this paper, we investigated the polarization–electric field (*P–E*) hysteresis loops and unipolar strain behavior of the poled and aged Mn-doped PMS-PZT ceramics when the electric field was perpendicular to the poling direction according to our method proposed above. Typical double *P–E* hysteresis loops were observed at various

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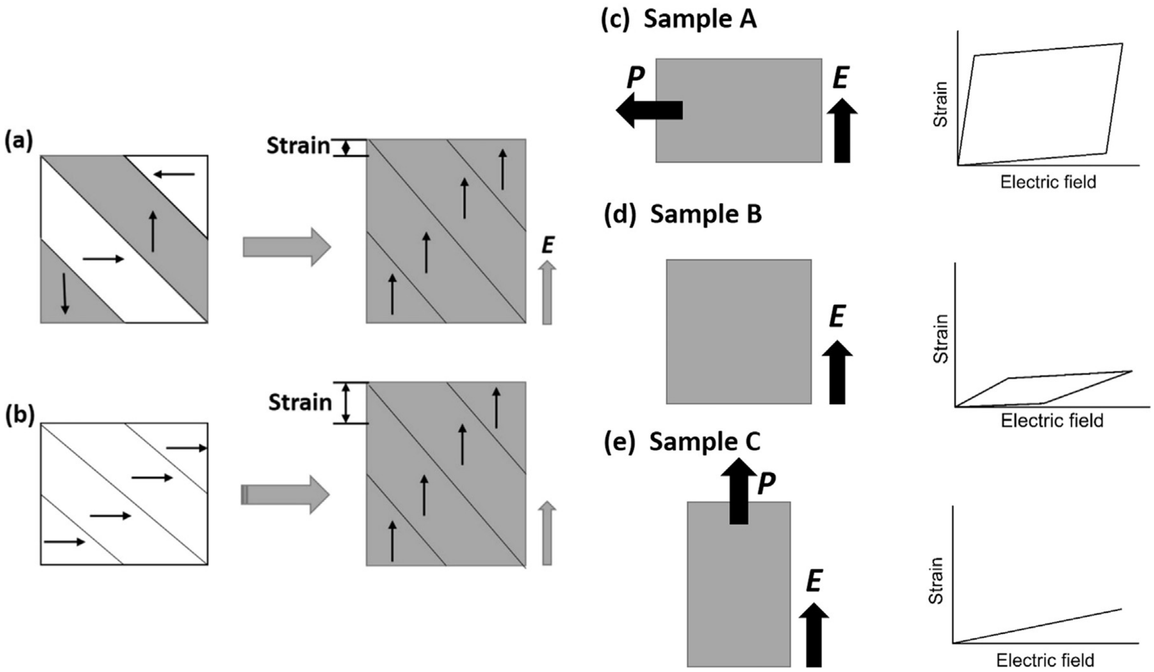

Fig. 1. (a) The schematic of electrostrain achieved via reversible domain switching in unpoled ceramics, which can be found in previous literature [17]; (b) The schematic of electrostrain achieved via reversible domain switching in poled ceramics when the external electric field is perpendicular to the external electric field; (c–e) The illustrations of sample A, sample B and sample C with respective unipolar strain curves.

electric fields. It was found that the unipolar strain could reach 0.225% which was 4.17 times larger than that of unpoled sample at a low electric field of 15 kV cm^{-1}. Thus the corresponding normalized strain d_{33}^* reached 1500 pm V^{-1} which significantly exceeded the values reported in ferroelectric ceramics. Our work provide a general effective method to enhance the electrostrain behavior via reversible domain switching in aged acceptor-doped ferroelectric ceramics.

2. Experimental procedures

The 0.1 wt% MnO_2-doped $0.05Pb(Mn_{1/3}Sb_{2/3})O_3-0.50PbZrO_3-0.45PbTiO_3$ ceramics used in this paper were fabricated by a conventional solid-state reaction method. The detailed fabrication process can be found elsewhere [14]. It needs to be pointed out that three samples with different poling states were utilized in our experiment for comparison. Sample A was poled under a dc field of 50 kV cm^{-1} for 15 min at 160 °C, but the external driving electric field was perpendicular to the poling direction according to our method, as shown in Fig. 1(c). Sample B was not poled, which was consistent with previous reports, as revealed in Fig. 1 (d). Sample C was also poled under a dc field of 50 kV cm^{-1} for 15 min at 160 °C, but the driving field was parallel to the poling direction, which demonstrated converse piezoelectric effect, as displayed in Fig. 1(e). The schematic illustrations of unipolar strain curves of sample A, sample B and sample C are also shown in Fig. 1 (c–e). It needs to be noted that all the samples were aged at room temperature for more than one month before the measurement of polarization–electric field (*P*–*E*) loops and strain-electric field (*S*–*E*) curves. The *P*–*E* loops and *S*–*E* curves were detected by an aixACT TF 2000 analyzer (aixACT Systems GmbH, Aachen, German) equipped with a laser interferometer.

3. Results and discussion

Fig. 2(a) shows the *P*–*E* hysteresis loops of sample A under various electric fields at 10 Hz. Typical double - hysteresis-loop can be observed at 10 kV cm^{-1} and 15 kV cm^{-1}. It needs to be pointed out that low external electric field cannot drive the domains to switch. As a result, the *P*–*E* loop is nearly linear with extremely low hysteresis under a relatively low electric field of 5 kV cm^{-1}. Moreover, both the polarization and the hysteresis become larger accompanied with more remarkable double-loop when the applied electric field becomes higher, originating from that more domains are forced to switch reversibly by the higher external field. It is also interesting to note that the double *P*–*E* loop can be predicted to occur according to the reversible domain switching model. It is well known that no polarization will be demonstrated in ferroelectric ceramics due to the random orientations of domains and grains before the application of electric field. Nevertheless, the applied electric field (sufficiently high) will force the domains to align along the same direction with demonstration of large polarization. Additionally, the domains will switch back to their initial orientations resulting in double *P*–*E* loop and large hysteresis due to the reversible domain switching mechanism after removal of the external field. The insert of Fig. 2 (a) demonstrates the comparison of *P*–*E* loops of sample A, sample B and sample C at 15 kV cm^{-1}. In comparison with sample A, sample B shows non-saturated *P*–*E* loop with lower maximum polarization and higher remnant polarization, which results from the reversible switching of part of the domains [15]. In addition, sample C exhibits asymmetric *P*–*E* loop which is attributed to the internal bias field associated with defect dipoles [2,14]. According to Fig. 2(b), the *P*–*E* hysteresis loop of sample A is sensitive to the measurement frequency. As the frequency declines, the maximum polarization tends to increase, indicating that the domain switching process is restricted by the high measurement frequency. In fact, low measurement frequency is favorable to make domains switch sufficiently, leading to high polarization.

Fig. 3(a) exhibits unipolar *S*–*E* curves of sample A at various electric fields (10 Hz). It can be seen that the strain increases from 0.038% at 7 kV cm^{-1} to 0.225% at 15 kV cm^{-1} accompanied with augment of strain hysteresis. At low field, few domains can be drove to switch to