

Regular article

Nanodomains in metal/ferroelectric 0–3 type composites: On the origin of the strong piezoelectric effect



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ABSTRACT

Outstanding piezoelectric properties were obtained in $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})_{0.20}(\text{Zr}_{0.50}\text{Ti}_{0.50})_{0.80}\text{O}_3/6 \text{ vol.}\% \text{ Ag}$ (PZN–PZT/Ag) metal/ferroelectric 0–3 type composites. A combination of X-ray diffractometer (XRD) and high resolution transmission electron microscopy (HRTEM) revealed that the PZN–PZT/Ag ferroelectric composite presents a novel intragranular structure. It is suggested that the uniform distribution of nano silver particles in a PZN–PZT matrix was the origination of nanodomains in PZN–PZT/Ag composites, further leading to the strong piezoelectric effect. We believe that our discovery here will be helpful in the development of new piezoelectric composites with outstanding properties.

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Nowadays, it is increasingly being recognized that new applications for materials require functions and properties that are not achievable by monolithic materials. Therefore, composite materials, which were combined by dissimilar materials, were used for these new applications [1–3]. However, the properties and processing of the composite materials still need to be understood to bridge the gap between the composite material microstructure and the end-product. Among the many composite materials, ferroelectric–metal composites formed by metallic particles dispersed within an insulating ferroelectric matrix have received extensive attention.

Over the past 40 years, ferroelectric–metal composites have attracted much attention due to a singular combination of properties (including mechanical, optical, electrical and magnetic properties), which make them excellent candidates to fabricate multifunctional devices with unique features. For example, the improvement of the dielectric behavior near their percolation threshold by incorporating dispersed metal particles (e.g., Ni, Ag, Pd, etc.) to a ferroelectric ceramic matrix (BaTiO_3 , $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$, etc.) is well reported nowadays and this approach has been successfully studied in numerous systems [4–7]. In previous studies, the ferroelectric and piezoelectric properties of the PZT/Ag composites were also investigated aiming at potential applications in functionally graded piezoelectric actuators with enhanced mechanical reliabilities [8]. Though, the enhanced mechanical properties make the PZT/Ag composites a promising candidate for piezoelectric actuator applications, however, at the same time, the ferroelectric and piezoelectric properties of PZT/Ag composites greatly deteriorated due to

the difference in the polarization characteristics of PZT/Ag composites associated with Ag particle dispersion in the PZT matrix [9].

As a result, it is highly desired to explore the possibility whether high ferroelectric and piezoelectric properties can be obtained in ferroelectric–metal composites. Here, in this letter, we reported an excellent electrical property in $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})_{0.20}(\text{Zr}_{0.50}\text{Ti}_{0.50})_{0.80}\text{O}_3/6 \text{ vol.}\% \text{ Ag}$ (PZN–PZT/Ag) nanocomposites. Fig. 1(a and b) gives the comparison of ferroelectric and piezoelectric behavior between PZN–PZT and PZN–PZT/Ag specimens, where the data is derived from the electrical properties versus Ag volume concentration (f_{Ag}), shown in Fig. S1. As can be seen, both ferroelectric behavior and piezoelectric behavior of PZN–PZT/Ag are much superior to that of pure PZN–PZT ceramic. We discovered that the origin of the strong piezoelectric effect is the existence of nanodomains in PZN–PZT/Ag composites.

The compositions used in this study are as follows: $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})_{0.20}(\text{Zr}_{0.50}\text{Ti}_{0.50})_{0.80}\text{O}_3 + x \text{ Ag}$ (PZN–PZT/xAg), where $x = 0 \text{ vol.}\%$ to 10 vol.%. The specimens were prepared using a simple blending procedure and low temperature sintering route. Reagent-grade oxide powders, i.e. Pb_3O_4 , ZrO_2 , TiO_2 , ZnO , Nb_2O_5 and Ag_2O , were used as raw materials. Here, it is worth noting that the Ag_2O used in our work shows good dispensability, which was pre-prepared by AgNO_3 and KOH , following reactions (1) and (2):



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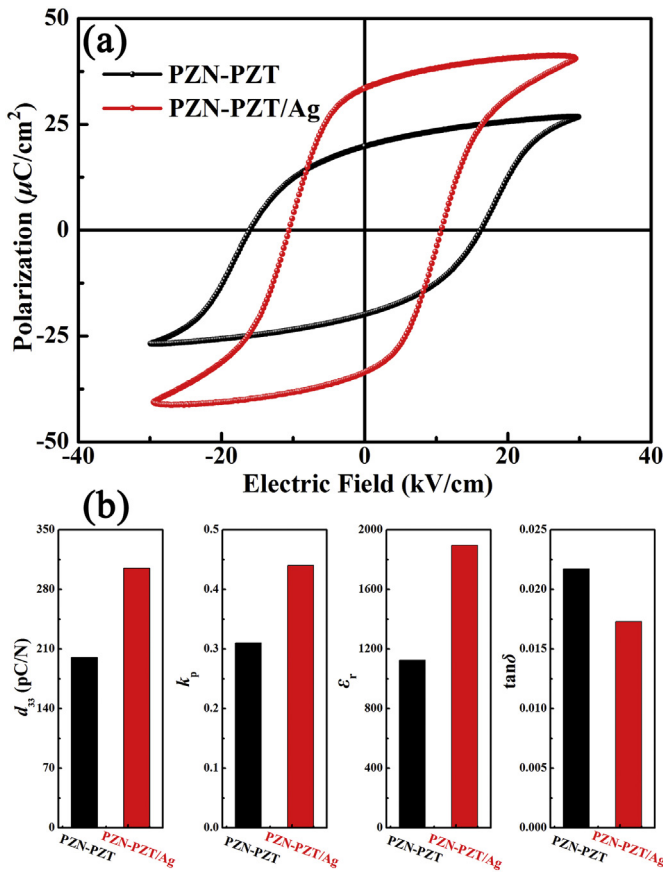


Fig. 1. (a) The polarization–electric field (P - E) hysteresis loops and (b) d_{33} , k_p , ϵ_r , and $\tan\delta$ values for PZN-PZT and PZN-PZT/Ag specimens.

The powders of Ag_2O together with other raw materials were weighed according to stoichiometric ratio and mixed through ball milling, with partially stabilized zirconia balls as media, in alcohol for 12 h. After drying, the mixture was calcined in a covered alumina crucible at 850°C for 2 h. At elevating temperature region, Ag_2O decomposed to Ag and O_2 at 250 – 300°C , following reaction (3):

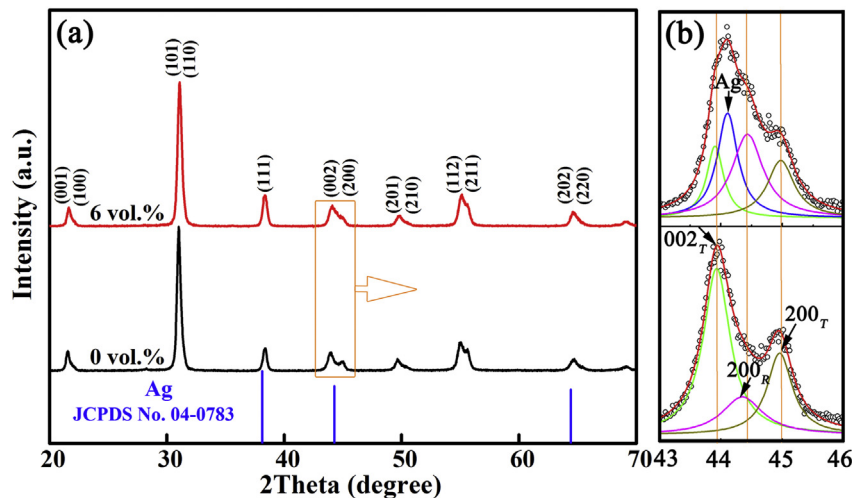


Fig. 2. (a) XRD patterns and (b) comparison of (002) and (200) reflections (from left to right) for 0 vol.% and 6 vol.% Ag specimens.

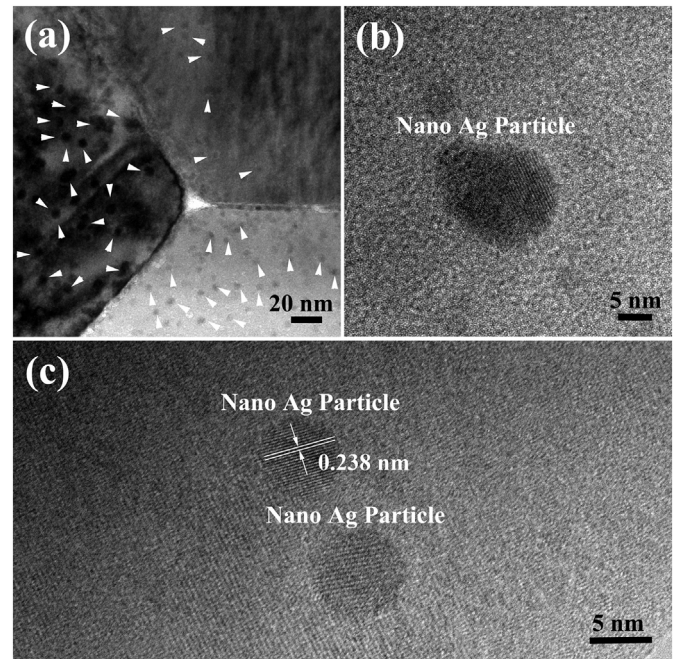


Fig. 3. (a) Intragranular structure in 6 vol.% Ag sample; (b)–(c) HRTEM imaging of several intragranular structures.

The calcined powders were remilled for 12 h and then pressed into disks of 11.5 mm in diameter at around 100 MPa. The green disks were sintered at 1050°C for 2 h in a sealed alumina crucible. And the detailed measurement methods have been described elsewhere [7,10].

Fig. 2(a) presents the XRD patterns of the 0 vol.% and 6 vol.% Ag specimens. All of the peaks in the profiles were assigned to ferroelectric PZN-PZT and metallic Ag phase, and no unwanted or reaction phases between the PZN-PZT matrix and Ag were found in the composites. Analysis on the fitting of $2\theta = 45^\circ$ peak and the relative intensities of (002) and (200) peaks reveal the coexistence of rhombohedral and tetragonal (R and T) perovskite phase for the PZN-PZT ceramic, while further observation on the 45° peak of 6 vol.% Ag specimen reveals obvious changes. Referring to the standard data of Ag (JCPDS No. 04-0783), it is deduced that the location of (200) (44.28°) diffraction peak of Ag was overlapped with those of PZN-PZT. According to the peak fitting results shown in Fig. 2(b), it is clear that the evolution of 45° peak patterns

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