

Structures and properties of novel antiferro-/ferroelectric materials with high energy-storage performance

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Energy resources are the driving force of social development and electrical energy-storage technology has received extensive attention. With their excellent positive DC bias feature, accompanied by high density, ultra-fast discharge and expansion of cell volume in the field-induced antiferroelectric (AFE) to ferroelectric (FE) phase transition process, antiferroelectrics have been considered as promising materials for high energy storage capacitors, large strain actuators, explosive transducers, electric-caloric refrigeration and microelectromechanical systems. To meet the increasing demands for high-performance energy storage capability, new AFE materials need to be developed.

In this paper, we present a novel concept for designing AFE materials based on the formation of solid solutions between two AFE systems of different anti-dipolar structures, namely between the conventional AFE PbZrO_3 or PbHfO_3 and the complex perovskite AFE $\text{Pb}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3$. A series of new AFE solid solutions of $(1-x)\text{PbZrO}_3 - x\text{Pb}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3$ (PZ-PMW) and $(1-x)\text{PbHfO}_3 - x\text{Pb}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3$ (PHf-PMW) have been synthesized, which has allowed us to reduce the critical field, and to realize the electric-induced AFE-FE phase transition at room temperature. In addition, the intermediate phase and structure-property relationship are studied in connection with the features of Pb^{2+} ion on the A-site and the evolution of chemically ordered structures on the B-site, leading to the establishment of comprehensive phase diagrams.

In particular, the crystal structure and energy storage properties of the PHf-PMW solid solution are systematically studied. The mismatch of B-site cations is believed to disrupt the long-range order of AFE dipoles. The competition between two different types of Pb ion displacement on A-site causes dipole frustration, reduces the critical field E_{cr} and increases the maximum polarization P_{max} , thus improving the recoverable energy density W_{rec} . With the PMW content of 10%, the best energy storage performance is obtained, which is superior to the so-far reported perovskite ceramics under similar electric field, and the W_{rec} is the highest in AFE-AFE system, making the PHf-PMW AFE solid solution ceramics a new class promising material for room-temperature energy storage applications.

These results indicate that the novel solid solutions form an interesting family of soft antiferroelectric materials, promising for high-density energy storage applications. The structural characterization and theoretical modeling shed light on the mechanisms underlying the AFE properties. This work not only gives the design method and synthesis strategy, but also provides ideas and guiding significance for developing other new AFE-AFE solid solution materials for high energy storage applications.