11th International Conference on Advanced Materials
Rio de Janeiro Brazil
September 20 - 25
ICAM2009

First principle based phase stability in PMN-*x*PT

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Abstract – We have applied a shell model potential developed by fitting first principle results to describe the behavior of the relaxor-ferroelectric PMN-*x*PT as function of concentration. The solid solution exhibits three regions with different characteristics according to Ti content in agreement with the experiments. At low Ti the polar and structural disorder dominates, and the symmetry is rhombohedral. The intermediate region is ferroelectric with easy rotation of the polarization direction, and the symmetry is monoclinic. Finally, in the high Ti content region, the solid solution adopts a ferroelectric behavior similar to PT, with tetragonal symmetry.

Relaxor ferroelectric solid solutions like *(1-x)*$PbMg_{1/3}Nb_{2/3}O_3$-*x*$PbTiO_3$ (PMN-*x*PT) or *(1-x)*$PbZn_{1/3}Nb_{2/3}O_3$-*x*$PbTiO_{1/3}$ (PZN-PT) are technological important compounds due to their extraordinary dielectric and piezoelectric properties. As a consequence, the basic understanding of the origin of their behavior has becoming an intensive area of research. Many progresses have been made from theoretical and experimental studies, but other many aspects of such rich behavior still not clearly understood due to the complexity of these materials.

We apply a shell model potential to determine equilibrated zero-temperature structures of PMN-*x*PT as function of the concentration. The model was developed by the simultaneous fitting of the parameter to LDA first principles results of the end members PMN and PT [1]. The simulations were performed at intervals of 10% in concentration in system sizes of 12*x*12*x*12 unit cells (8640 atoms) with periodic boundary conditions. All atoms and cell parameters were allowed to relax in each case. The atomic distribution of Nb and Mg are according to random site model while Ti atoms are randomly distributed. We perform simulations for three different arrange of ions or 'layouts'.

Figure 1 and 2 shows the dependence of lattice parameters and components of polarization with concentration respectively, where we can distinguish a series of transitions as function of the concentration. Starting from a cubic non polar system at PMN limit, a first transition to a rhombohedral phase takes place when small amount of Ti is added. In this low Ti content region, a relative small polarization shows up approximately along [111] direction while the cubic structure is slightly distorted. The polarization *P* is growing with *x*, and there is a second transition to an intermediate region at *x*=0.3. In this region, the solid solution develops a clear ferroelectric state, although different phases as function of the composition and layout are present. Finally, there is a last transition at x=0.6 to an ending region with tetragonal structure. The zero temperature predicted sequence of transitions as function of the concentration is in excellent agreement with the experimental evidence, and it allows us to investigate the temperature evolution of the solid solution.

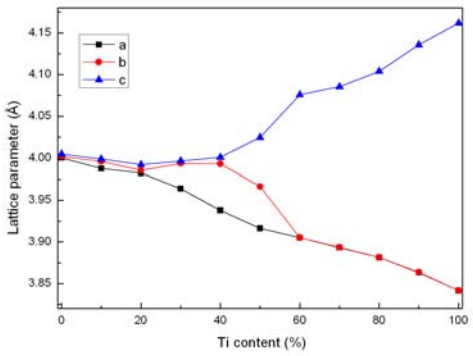

Figure 1: Lattice parameters as function of Ti-content in PMN-*x*PT at zero temperature obtained from model simulations.

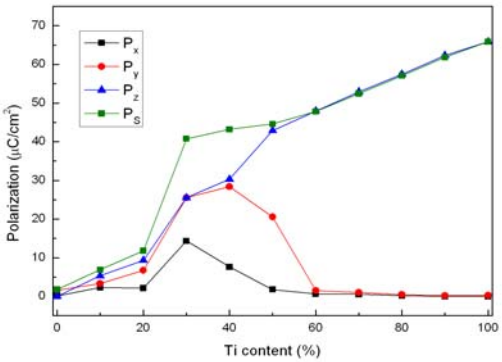

Figure 2: Polarization as function of Ti-content in PMN-*x*PT at zero temperature obtained from model simulations.

[1] A. Asthagiri, Z. Wu, N. Choundhry, and R. E. Cohen, Ferroelectrics **333**, 69 (2006).