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Synthesis, dielectric properties and structural characterization of $Ba_{0,9}Ca_{0,1}Zr0_{0,9X}Ti_{1-0,9X}O_3$ ferroelectric ceramic system

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Abstract – The $Ba_{0,9}Ca_{0,1}Zr_{0,9x}Ti_{1-0,9x}O_3$ ceramic system have been studied by dielectric experiments, high-resolution synchrotron x-ray diffraction and x-ray absorption spectroscopy. Temperature dependence of dielectric permittivity and the degree of diffuseness of relaxor ferroelectric ceramics became more pronounced for higher zirconium concentration. Phase transition over the temperature was observed for non-relaxor ceramics while cubic symmetry was maintained for relaxor ferroelectric system.

Ferroelectric materials have been applied to a large field of applications because of their excellent dielectric, piezoelectric and ferroelectric properties. Lead-free ferroelectric materials have attracted the attention of different researches due to the protection environment restriction and thus, many investigations were concentrated on the BaTiO3-based materials. The substitution of barium by calcium and zirconium atoms in the $BaTiO_3$ structure ($Ba_{0,9}Ca_{0,1}Zr_{0,9X}Ti_{1-0,9X}O_3$, BCZT) lead to significant changes in the electrical and structural properties which can varies from a normal to a relaxor ferroelectric behavior depending of the Zr/Ti ratio as well as the amount of calcium. The ceramic properties are also deeply affected by the powder characteristics such as the particle size and chemical composition. Recently, it was proposed the existence of a spontaneous phase transition at temperatures below the maximum of dielectric permittivity; however, the existence of this transition is controversial and until now under discussion.

In this work the effect of composition variation of the $Ba_{0,9}Ca_{0,1}Zr_{0,9x}Ti_{1-0,9x}O_3$ ($0<x<30$) system on the electrical and structural properties was revised. The ceramics were prepared by a sol-gel method and the dielectric permittivity, the short and long range order structures were characterized. Dielectric measurements show that samples with $x > 20$ presents a typical relaxor behavior and that the degree of diffuseness became more pronounced as the amount of Zr increases. HRXRD results showed changes in the symmetry for samples with a lower amount of zirconium and the nonexistence of a spontaneous phase transition for a relaxor ferroelectric ceramics as proposed in the literature. XAS experiments collect at Ti K edge shows no significant changes on the TiO6 octaedra structure.