

Origin of enhanced dielectric and piezoelectric properties in Li and Ta co-substituted $K_{0.5}Na_{0.5}NbO_3$ ceramics

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Ferroelectric ceramic oxides have been widely used in various devices such as non-volatile random access memory, sensors, actuators, transducers and high energy density capacitors, due to its excellent dielectric, ferroelectric and piezoelectric properties

Potassium sodium niobate ($K_{0.5}Na_{0.5}NbO_3$, KNN) based ceramics and its solid solutions are considered as one of the promising lead free ferroelectric materials. Unfortunately, due to the evaporation of the volatile potassium (K) and sodium (Na), the synthesis of stoichiometric KNN is difficult [1,2]. At the same time the piezoelectric properties of KNN based ferroelectrics need to be enhanced for the device applications. It has been reported that addition of Li^+ at the K/Na site in KNN improves densification, lowers the T_{O-T} (orthorhombic to tetragonal phase transition temperature), and raises the T_C (Curie temperature) [1, 3]. On the other hand, Ta^{5+} substitution at the Nb^{5+} site in KNN lowers both T_{O-T} and T_C and enhances the piezoelectric properties [1, 3]. So in the present study, Li and Ta co-substituted KNN ceramics are considered. In view of this, we have prepared

$(K_{0.48}Na_{0.48}Li_{0.04})(Nb_{1-x}Ta_x)O_3$ ($x=0$ to 0.40) ceramic using conventional solid state reaction route and investigated the compositional driven structural phase transitions using X-ray diffraction and Raman spectroscopic techniques. Rietveld

refinement analysis of X-ray diffraction patterns indicate a compositional induced structural phase transition from orthorhombic (Amm2) phase ($x<0.10$) to tetragonal (P4mm) phase ($x>0.20$) through the coexistence of both orthorhombic and tetragonal (Amm2+P4mm) phases for $0.10\leq x\leq 0.20$. This compositional driven phase transitions is well correlated with the Raman spectroscopic analysis. FESEM micrograph shows a highly dense microstructure with inhomogeneous distribution of grains and the grain size decreases with increase in Ta content. The temperature-dependent dielectric properties of the ceramic were measured to further study the ferroelectric phase transitions behaviour. Based on the RT XRD, Raman spectroscopic analysis and temperature dependent dielectric properties, a phase diagram has been constructed. For the ceramic with $x=0.20$, we have obtained maximum piezoelectric coefficient ($d_{33}=159pC/N$) and dielectric constant ($\epsilon_r=556$) at RT which can be explained due to the coexistence of O+T phases around the MPB (morphotropic phase boundary). The in-depth analysis of the crystal structure and its relationship to the improvement of physical properties will be discussed.

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