World Academy of Science, Engineering and Technology
International Journal of Materials and Metallurgical Engineering
Vol:19, No:03, 2025

Theoretical and Experimental Investigation of Structural, Electrical and Photocatalytic Properties of $K_{0.5}Na_{0.5}NbO_3$ Lead- Free Ceramics Prepared via Different Synthesis Routes

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Abstract : The $K_{0.5}Na_{0.5}NbO_3$ (KNN) system has emerged as one of the most promising lead-free piezoelectric over the years. In this work, we perform a comprehensive investigation of electronic structure, lattice dynamics and dielectric/ferroelectric properties of the room temperature phase of KNN by combining ab-initio DFT-based theoretical analysis and experimental characterization. We assign the symmetry labels to KNN vibrational modes and obtain ab-initio polarized Raman spectra, Infrared (IR) reflectivity, Born-effective charge tensors, oscillator strengths etc. The computed Raman spectrum is found to agree well with the experimental spectrum. In particular, the results suggest that the mode in the range ~840-870 cm^{-1} reported in the experimental studies is longitudinal optical (LO) with A_1 symmetry. The Raman mode intensities are calculated for different light polarization set-ups, which suggests the observation of different symmetry modes in different polarization set-ups. The electronic structure of KNN is investigated, and an optical absorption spectrum is obtained. Further, the performances of DFT semi-local, metal-GGA and hybrid exchange-correlations (XC) functionals, in the estimation of KNN band gaps are investigated. The KNN bandgap computed using GGA-1/2 and HSE06 hybrid functional schemes are found to be in excellant agreement with the experimental value. The COHP, electron localization function and Bader charge analysis is also performed to deduce the nature of chemical bonding in the KNN. The solid-state reaction and hydrothermal methods are used to prepare the KNN ceramics, and the effects of grain size on the physical characteristics these ceramics are examined. A comprehensive study on the impact of different synthesis techniques on the structural, electrical, and photocatalytic properties of ferroelectric ceramics KNN. The KNN-S prepared by solid-state method have significantly larger grain size as compared to that for KNN-H prepared by hydrothermal method. Furthermore, the KNN-S is found to exhibit higher dielectric, piezoelectric and ferroelectric properties as compared to KNN-H. On the other hand, the increased photocatalytic activity is observed in KNN-H as compared to KNN-S. As compared to the hydrothermal synthesis, the solid-state synthesis causes an increase in the relative dielectric permittivity (ε^') from 2394 to 3286, remnant polarization (P_r) from 15.38 to 20.41 μC/cm^², planer electromechanical coupling factor (k_p) from 0.19 to 0.28 and piezoelectric coefficient (d_33) from 88 to 125 pC/N. The KNN-S ceramics are also found to have a lower leakage current density, and higher grain resistance than KNN-H ceramic. The enhanced photocatalytic activity of KNN-H is attributed to relatively smaller particle sizes. The KNN-S and KNN-H samples are found to have degradation efficiencies of RhB solution of 20% and 65%, respectively. The experimental study highlights the importance of synthesis methods and how these can be exploited to tailor the dielectric, piezoelectric and photocatalytic properties of KNN. Overall, our study provides several bench-mark important results on KNN that have not been reported so far.

Keywords : lead-free piezoelectric, Raman intensity spectrum, electronic structure, first-principles calculations, solid state synthesis, photocatalysis, hydrothermal synthesis
Conference Title : ICMMSE 2025 : International Conference on Metallurgy, Materials Science and Engineering
Conference Location : Miami, United States
Conference Dates : March 11-12, 2025