INDIAN INSTITUTE OF TECHNOLOGY GUWAHATI
SHORT ABSTRACT OF THESIS

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Thesis Title: Studies on Bismuth Sodium Titanate-based Lead-Free Piezoelectric Bulk Ceramics and Thin Films

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Thesis Submitted to the Department/ Center	:	Physics
Date of completion of Thesis Viva-Voce Exam	:	17/06/2020
Key words for description of Thesis Work	:	Lead-free Ceramics and Composites, $Bi_{0.5}Na_{0.5}TiO_3$ (BNT), VRH-Mechanism, Dielectric, Ferroelectric, Piezoelectric, Microwave Dielectric & Nonlinear Optical Properties, PLD, Oxygen Partial Pressure, SPDR and Z-Scan Method

SHORT ABSTRACT

The present thesis is mainly focused on the lead-free piezoelectric bulk ceramics and thin films, which are highly interesting from the point of view of scientific, research, and industrial applications. In this work, successful efforts have made to improve the structural, dielectric, piezoelectric, and other physical properties of bismuth sodium titanate (BNT) lead-free piezoelectric ceramics with suitable dopants/compositions and to understand the origin of dielectric, relaxor/ferroelectric, piezoelectric response on their structure, polarization, strain, stability, temperature, defects, etc. The electrical properties are mostly controlled by the relative sizes of the ions and are mainly dependent on the electronic configuration of the ions. Dopants and substituents have a profound influence on the tailoring properties of the ABO_3 oxides.

In order to enhance dielectric and piezoelectric properties, suitable dopants (Ce^{+3}, Gd^{+3}, and K^{+}) at A- and/B-sites and compositions ($K_{0.5}N_{0.5}NbO_3$ + Gd; KNNG) of BNT ceramics were chosen, which have been prepared by a conventional solid-state reaction method. The Rietveld refinement XRD and Raman spectroscopy of all samples revealed the rhombohedral crystal symmetry with the R3c space group, whereas K doped BNT ceramics revealed a morphotropic phase boundary between rhombohedral and tetragonal phases with improved properties. The average grain size was found to decrease with the substitution of Ce, Gd, K and KNNG, which inhibits grain growth and enhanced density. The dielectric studies of pure BNT ceramics as a function of temperature exhibited two phase transitions correspond to the ferroelectric to antiferroelectric (T_d ~198ºC) and anti-ferroelectric to paraelectric phase

(T_C~330ºC). All the samples have exhibited two phase transitions, the T_d and T_C values shifted towards lower temperatures with the addition of Gd, K and KNNG composition. The frequency dispersion with a diffuse transition in T_d and T_C exhibits relaxor behavior, which is confirmed by the conclusion drawn from modified Curie Weiss and Vogel-Fulcher laws. Temperature dependence of resistivity analysis provides the evidence of the variable range hopping mechanism between charge carriers of BNT and doped systems. The average distance between the two successive hops, associated with hopping energy decreased, which are signifying the formation of additional localized states due to the incorporation of these dopants into the BNT system. The leakage current density effectively reduced with the substitution of K and KNNG in the BNT system and improved the dielectric and piezoelectric properties. The leakage current conduction mechanism of K and KNNG based compositions followed a space charge limited conduction which is related to deep acceptor traps. The dielectric and piezoelectric response of the Ce, Gd, K and KNNG doped BNT ceramics improved significantly and high dielectric constant, low loss tangent (ε_r = 1074 and tanδ = 0.059 at 1 kHz), best piezoelectric constants (d_{33} = 108 pC/N and g_{33} = 1.14×10^{-2} Vm/N) large d_{33} (108 pC/N) and electromechanical coupling factors (k_{33}, k_{31}, k_t ~ 48% and k_p = 53%), were obtained for the BNT-KNNG composition of x = 0.01.

Further, the pure BNT and best-modified composition (BNT-KNNG) chosen to deposit thin films by pulsed laser deposition technique to study their optical, dielectric, and ferroelectric properties. The larger nonlinear refractive index (n_2 = 5.775 ×10^{-6} cm^2/W) and strong absorption coefficient (β = 0.973 cm/W) and improve dielectric (ε_r = 411, *tanδ* = 0.156 @ 1 kHz), microwave dielectric (ε_r = 317 and *tanδ* =0.0074 @ 10 GHz), and ferroelectric (P_r = 25.31 μC/cm^2, E_C = 42.62 kV/cm @ 1 kHz) properties with low leakage current density are observed for the BNT-KNNG thin films (10 Pa), which is followed a space charge limited conduction behavior. The obtained best dielectric and piezoelectric properties of bulk BNKT and BNT-KNNG ceramics are good candidates for the high performance of electromechanical (energy harvesting) applications. The optimized best linear, nonlinear optical properties and microwave dielectric properties with low leakage current density properties of the BNT-KNNG composite thin films are making the material suitable for nonlinear photonics, optoelectronic, integrated electronic and microwave tunable device applications.