Study of the Magnetoelectric Coupling Behavior of Divalent Cations Substituted Barium-Zircon-Titanate and Gadolinium Substituted Nickel-Copper-Zinc Ferrite Multiferroic Composites

by

Bablu Chandra Das
Roll No: 1014144008
Session: October 2014

DOCTOR OF PHILOSOPHY

Supervisor

Prof. Dr. A. K. M. Akther Hossain

Department of Physics
BANGLADESH UNIVERSITY OF ENGINEERING AND TECHNOLOGY
April 2021

Abstract

The room temperature magnetoeletric coupling was investigated thoroughly in various $(1\text{-}x)Ba_{0.985}D_{0.015}Zr_{0.10}Ti_{0.90}O_3+xNi0_{.48}Cu_{0.12}Zn_{0.40}Gd_{0.04}Fe_{1.96}O_4$ [(1-*x*)BDZT+*x*NCZGF] (where D = Ca, Sr) multiferroic composites. Ferroelectric and ferrite phase of the composites have been prepared by the standard solid state reaction technique. Structural study was conducted by the X-ray diffraction (XRD) and the Rietveld refinement was adopted for further analysis of the structure. The refined of XRD patterns confirmed all composites having a biphasic perovskite-spinel structure with $P4mm$, and $Fd\bar{3}m$ space group, respectively. Microstructural characteristics of all samples have been analyzed using Field Emission Scanning Electron Microscopy images. The magnetoeletricaly coupled composites is illustrated by an obvious modification in lattice, dielectric, ferroelectric and magnetic properties as a function of ferrite content. Frequency dependence of the dielectric constant exhibits dispersion, which can be modeled using a modified Debye function that considers the probability of several ions contributing to the relaxation. The dielectric constant of the composites was found to decrease with the addition of ferrite content in line with the theoretical estimations of the Bruggeman, Maxwell-Garnett, Lichtencker and Looyenga models, respectively. The dielectric constant and initial permeability were calculated theoretically using the different models and compared with the experimental values. The discrepancy between the calculated and experimental values of dielectric constant and initial permeability might be attributed to the diffusion of ions and interaction between two phases of the composites. The ac conductivity analysis revealed that the conduction mechanism is attributed to the small polaron hopping and also obeyed the Jonscher's power law. Complex impedance was evaluated using the Nyquist plot which ensured the dominance of both grain and grain boundary resistance. The hysteresis loops (*M-H* and *P-E*) ensured the typical ferromagnetic and ferroelectric nature for all composites at room temperature. Magnetic studies show an improvement of initial permeability, relative quality factor and magnetization, whereas magnetic loss gradually reduces with the increment of ferrite content. The maximum magnetoelectric response ~173 mV/(cm Oe) and ~194 mV/(cm Oe) were found for 0.9BCZT+0.1NCZGF and 0.9BSZT+ 0.1NCZGF composite that could be utilized in the future multifunctional devices.

List of peer-reviewed journal articles

[1] **B. C. Das,** A. K. M. Akther Hossain, "Rietveld refined structure, ferroelectric, magnetic and magnetoelectric response of Gd- substituted Ni-Cu-Zn ferrite and Ca, Zr co-doped $BaTiO_3$ multiferroic composites", *J. Alloy. Compd.*, vol. 867, 159068, 2021.

[2] **B. C. Das,** M. A. Matin, A. K. M. Akther Hossain, "Rietveld refinement structure, electric, dielectric and ferroelectric properties of lead free $Ba_{0.985}Sr_{0.015}Zr_{0.10}Ti_{0.90}O_3$ ceramics", *J. Mater. Sci.: Mater. Electron*, vol. 32, 4916-4936, 2021.

[3] **B. C. Das,** F. Alam, A. K. M. Akther Hossain, "The crystallographic, magnetic, and electrical properties of Gd^{3+}-substituted Ni–Cu–Zn mixed ferrites," *J. Phys. Chem. Solid.*, vol. 142, 109433, 2020.

[4] **B. C. Das,** H. Das**,** M. A. Matin, A. K. M. Akther Hossain, "Rietveld refinement analysis and influence of individual phases on the magnetoelectrically coupled (1-*x*)BSZT+*x*NCZGF multiferroic composites" *J. Alloys Compd*., (Review completed).

[5] F. Alam, M. L. Rahman, **B. C. Das,** A. K. M. Akther. Hossain, "Effect of Cu on structural, elastic and magnetic properties of nanostructured Mn–Zn ferrite prepared by a sol-gel auto-combustion method," *Physica B: Condens. Matter*, vol. 594, 412329, 2020.

[6] M. M. Rhaman, M. A. Matin, M. A. Al Mamun, A. Hussain, M. N. Hossain, **B. C. Das,** M. A. Hakim, M. F. Islam, "Enhanced electrical conductivity and multiferroic property of cobalt-doped bismuth ferrite nanoparticles," *J. Mater. Sci.: Mater. Electron.*, vol. 31, pp. 8727–8736, 2020.