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Room temperature magnetoelectric properties of lead-free alkaline niobate based particulate composites

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ABSTRACT

The room temperature magnetoelectric properties of lead-free $K_{0.5}Na_{0.5}NbO_3$ (KNN) and $CoFe_2O_4$ (CFO) particulate composites having general formula (1-*x*) KNN/ *x* CFO (where $x = 0.1$, 0.2, 0.3, 0.4 and 0.5)are presented here. Structural studies confirm the presence of phase pure KNN and CFO in the composites. Microstructural properties and grain size evaluations are carried out by using FESEM and TEM micrographs. The well-defined P-E and M-H hysteresis loops obtained for all the composites confirmed their multiferroic properties. The highest magnetocapacitance of 30% at a low frequency of 4 kHz and 5% at a high frequency of 100 kHz are noted for the composite with 40% CFO. Also, the composite with 40% CFO showed a maximum ME coupling coefficient of 28 mV/cm.Oe. The room temperature coupling properties and high magnetocapacitance at low frequencies make this non-lead-based ME nanocomposites as a promising candidate in the field of MEMS.

1. Introduction

The concept of magnetoelectric effect in composites was predicted by Van Suchtelen in 1972 [1]. Followed by this, several combinations such as BTO/ferrites, PZT/ferrites, PMN-PT/ferrites etc. were studied experimentally and theoretically [2–6]. Magnetoelectric composites are generally described according to their structures or connectivity schemes. Particulate bulk composites are the first explored ME composite geometry. In this structure, magnetic nanoparticles are embedded in a piezoelectric matrix. The original works on insitu formation of particulate composites were done at Philip's Laboratories [7–9]. They prepared magnetoelectric composites of $BaTiO_3$ and $CoFe_2O_4$ by unidirectional solidification and reported a high ME coupling coefficient of 130 mV/cm Oe. But sintering techniques were found to be easier and cheaper for the fabrication of particulate composites especially having piezoelectric and ferrite phases. Most of the particulate composites prepared by sintering techniques exhibited ME coefficients of about 1–100 mV/cm Oe in magnitude at lower frequencies. Ryu *et al.* [10] reported an ME voltage coefficient of 115 mV/cm Oe for 3–0 PZT/NFO composites. Due to the hazardous effect of lead, the lead-free ME systems are increasing interest in recent years. $BaTiO_3$ and derivatives, alkaline niobates, Bismuth layered composites etc. are some of the lead-free piezoelectric material alternatives to PZT.

Mainly in bulk composites, the magnetic/piezo particles are randomly distributed in a matrix of piezoelectric/magnetic materials. The magnetic to piezoelectric volume ratio is an important parameter to determine the geometry of this type of bulk composites. Generally, in this type of particulate composites, the magnetoelectric voltage reaches the maximum at the middle concentration region of the ferrite/piezoelectric phases i.e., $f \sim 0.4$–0.6. The excess amount of ferrite phase (more than 60 wt%) can result in poor piezoelectric and ferroelectric properties due to the low resistivity of ferrite grains. It can affect the electric poling at high voltages and resulted in high leakage current which in turn reduces the magnetoelectric voltage output.

Here, magnetoelectric bulk composites were fabricated with piezoelectric and magnetic materials as constituent phases. A promising lead-free piezoelectric candidate such as $KNaNbO_3$ (KNN) with high piezoelectric properties is used as the piezoelectric matrix material. There are only rare reports about this combination of $KNaNbO_3$ and $CoFe_2O_4$ which is used as constituent materials in ME bulk composites. Here, magnetoelectric coupling properties of KNN-CFO composites with a different loading percentage of CFO are analyzed.

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