Ceramics International 42 (2016) 18238–18246

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

Reduced leakage current and improved multiferroic properties of 0.5((1−x)BLPFO-xPZT)-0.5PVDF composite films

Nidhi Adhlakha [a], K.L. Yadav [b,*], Marco Truccato [a], Manjusha [b]

[a] Department of Physics, University of Torino, via Giuria 1, Torino, I-10125 Italy
[b] Smart Materials Research Laboratory, Department of Physics, Indian Institute of Technology Roorkee, Roorkee, 247667 India

ARTICLE INFO

Article history:
Received 9 June 2016
Received in revised form
27 July 2016
Accepted 23 August 2016
Available online 24 August 2016

Keywords:
Ceramic-polymer composite films
Magneto-dielectric coupling
Leakage current
Magnetization
X-ray diffraction

ABSTRACT

$0.5((1-x)Bi_{0.8}La_{0.1}Pr_{0.2}FeO_3$ (BLPFO)-$xPb(Zr_{0.52}Ti_{0.48})O_3$ (PZT))-0.5Polyvinylidene difluoride (PVDF) composite films with x variations 0.25, 0.40 and 0.50 were synthesized using two step mixing, followed by hot pressing. The structural, microstructural, dielectric, magnetic, ferroelectric and magnetodielectric properties of composite films have been systematically investigated. The measurement of the dielectric properties at 1 kHz shows that the dielectric loss (tan δ) decreases with increasing the volume fraction of PZT. The value of maximum room temperature $\varepsilon_r \sim 78$ and low tan δ ~0.061 for 0.5((1−x)BLPFO-xPZT)-0.5PVDF composite film with $x=0.50$ suggests its usefulness for capacitor applications. For predictions of effective dielectric constant of composite films experimental data were fitted with Lichtenecker model. Among all the composite films, the film with $x=0.50$ was found to exhibit smallest leakage current density $\sim 7\times10^{-8}$ A/cm^2 and hence improved electrical resistivity. The variation of magnetization with temperature indicates the presence of spin glass behavior along with the ferromagnetic component at 5 K. The value of remnant polarization ($2P_r$) is found to increase with increase of PZT content in composite films. In the present composite films a significant dependence of dielectric constant on magnetic field has been observed, and highest value of magnetodielectric response of 2.85% is observed for composite film with $x=0.50$.

1. Introduction

In the past several years, a significant amount of work has been carried out on various ceramic-polymer composites, especially on high dielectric constant ceramic-polymer composites for their applications in embedded capacitors [1,2]. During study of these high dielectric constant materials, relatively less importance has been given to dielectric loss. However in many applications such as integral thin film capacitors or electric stress control devices, a low dielectric loss along with high dielectric constant is required [3]. In spite of high dielectric constant exhibited by conductive fillers embedded in insulating polymer, a high dielectric loss and low breakdown strength has been observed which limits their applications [3]. In recent years ferroelectric ceramics with high dielectric constant and low dielectric loss such as Pb(Zr, Ti)O_3 [4] and $BaTiO_3$ [5] have been used as fillers in order to overcome the aforementioned limitations. On the other hand, multiferroic materials is a class of interesting materials which simultaneously show electric and magnetic orderings, and are considered to be promising materials both scientifically and technologically due to their potential for design of multifunctional devices such as sensors, MEMS devices, ferroelectric storage and spintronic devices etc [6,7]. The existence of cross-coupling between magnetic and electric order parameters, called magnetoelectric (ME) coupling, has drawn great interest, as this effect opens the possibility to manipulate electric properties magnetically, and vice versa [8]. $BiFeO_3$ (BFO) is one of the most widely studied room temperature multiferroic materials with high Curie temperature ($T_c=1103$ K) and high Neel temperature ($T_N=643$ K). Despite of exhibiting certain interesting features it has limitations from the application point of view as a multiferroic system due to its high conductivity, antiferromagnetic nature and weak magnetoelectric (ME) coupling. In bulk BFO the presence of defects, low resistivity and nonstoichiometry gives rise to leakage problems which further hinders the measurement of ferroelectric and transport properties. Apart from this, the macroscopic magnetization is canceled due to the presence of inhomogeneous cycloidal spin structure which further hinders the linear magnetoelectric effect [9]. Therefore, it is essential to improve its multiferroic properties for practical applications. For instance, in BFO nanostructures an unexpectedly large spontaneous polarization (~60 μC/cm^2) was observed and considered to be due to strain effect [10]. Similarly in

* Corresponding author.
E-mail address: klyadav35@yahoo.com (K.L. Yadav).

http://dx.doi.org/10.1016/j.ceramint.2016.08.147
0272-8842/