International Journal of Scientific Research in Science and Technology
Print ISSN: 2395-6011 | Online ISSN: 2395-602X (www.ijsrst.com)
© 2021 | IJSRST | Volume 8 - Issue 1

Magnetoelectric Effects in LSMO - BCZT Multiferroic Composites

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ABSTRACT

$(Ba_{(1-x)}Ca_{(x)})(Zr_{(y)}Ti_{(1-y)})O_3$ (BCZT), for x= 0.05 and 0.075 & y = 0.075 and 0.10 is a ferroelectric material known to possess ferroelectric transition temperature Tc in the vicinity of room temperature is chosen to investigate their possible magnetoelectric (ME) applications. The $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) is selected to be ferromagnetic phase. Hydroxide co-precipitation route is adopted so as to synthesis of LSMO phase and ceramic synthesis route is adopted for the synthesis of BCZT phase. Starting with the LSMO and BCZT powders, the composites yLSMO + (1- y) BCZTx = yLBCZTx with y = 0.10, 0.15 & 0.20 and x = 1, 2 & 3. are synthesized for ME properties. The parent composition of LSMO as well as the BCZT are characterized for dielectric and magnetic properties to confirm the formation of desire ferroelectric and magneto strictive phases. The composites are investigated for the structural and microstructural analysis, dielectric, magnetoelectric properties. The results show that the composite yLBCZTx exhibit excellent ME properties.

Keywords - BCZT, LSMO, Ferroelectric, Magnetoelectric.

I. INTRODUCTION

Multiferroic materials have gained momentum in recent years for their coexistence of ferroelectricity, ferromagnetism and ferroelasticity [1-3]. The coupling between ferroelectric and ferromagnetic orders can produce magnetoelectric (ME) effects. The ME composites are extensively studied over last 20 years. In recent years, ME materials prepared by combining the ferrite and ferroelectric as a constituent phases have drawn significant interest due to their multifunctionality [4].These materials provide opportunities for potential applications such as transducers, actuators and sensors [5] .The selection of ferrite and ferroelectric phases are mainly depend on the various factors viz., high magnetostriction coefficient of ferrite phase and piezoelectric coefficient of ferroelectric phase, high dielectric constant and poling strength [6]. Thus one may select a ferroelectric material with useful values of polarization P, ε and ferrite materials with sufficient CMR systems possessing Tc in the vicinity of room temperature. Therefore, considering the discussion above, it appears that after proper selection of the ferroelectric and ferromagnetic phases the composites may produce sufficiently large values of both ME effect.

BCZT is a good candidate for a variety of applications. Its properties can be controlled by varying the Ba/Ca and Zr/Ti compositions. The (Ca, Zr) co-doping plays

a critical role in maintaining the electrical properties of $BaTiO_3$ ceramics. Regarding CMR materials $La_{(1-x)}Ba_xMnO_3$ perovskite manganites have attracted considerable research interest in recent years because of the observation of a huge magneto-resistance (MR) called the colossal magneto-resistance (CMR) effect.

Considering the earlier reports, $(Ba_{(1-x)}\ Ca_{(x)})(Zr_{(y)}Ti_{(1-y)})O3$ (BCZT), for x= 0.05 and 0.075 & y = 0.075 and 0.10 was selected as the ferroelectric phase for the formation of composites. Similarly, $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) was selected as for ferromagnetic phase. Using these compositions, the ME composites are formed using the formula yLSMO + (1- y) BCZTx = yLBCZTx with y = 0.10, 0.15 & 0.20 and x = 1, 2 & 3.

The composites are investigated for their structural, microstructural and magnetoelectric properties. The present paper report details of synthesis and also presents a qualitative analysis of the observed ME properties.

II. METHODS AND MATERIAL

2.1 Synthesis of BCZT composition

The BCZT solid solutions have been synthesized using standard ceramic route of synthesis. High purity (>99.9%) $BaCO_3$, CaO, ZrO_2 and TiO_2 of AR grade are used as precursors. The stoichiometric amount of the precursors was well mixed together and grounded thoroughly for 2 h in an agate mortar with pestle. Considering the earlier reports, the pre sintering was carried out at 1150^0C.The pre sintered powder was mixed with a polyvinyl acetate (PVA) binder solution and compacted into disk shaped samples with a diameter of 1.0 cm and a thickness of nearly 1.0 mm. The final sintering process was carried out at 1200^0C for 24 h in two steps. The product of final sintering is formed as a powder. The calcinated powder of BCZT was pelletized for the investigation of physical and dielectric properties of BCZT. It is observed that the dielectric properties of BCZT are in concurrence with the earlier reports. The powder has been used for the formation of ME composites.

2.2 Synthesis of LSMO composition

To achieve near atomic level uniformity of the constituents, the hydroxide co-precipitation route has been adopted for synthesis of $La_{0.67}Sr_{0.33}MnO_3$ (LSMO). The $La(NO_3)_3.6H_2O$, $Sr(No_3)_2$, $KMnO_4$, and $(CH_3COO)_2.Mn.4H_2O$ of AR grade are used as precursors for the hydroxide co-precipitation route of synthesis. The precursors are dissolved in distilled water to form nearly 40 mM solution of the constituents and NH_4OH is used as precipitant. The precipitates are thoroughly washed in distilled water keeping alkaline medium using NH_4OH solution with pH between 9 to 10 [7]. The dried precipitates are calcinated at 1000 ^{0}C for 12 h, and final sintering is carried out at 1100 ^{0}C for 12 h with intermediate grinding. The product of final sintering is formed as a powder and also in pellets of 1.2cm diameter. The powder has been used for the formation ME composites, while the pellets were used for determination of their physical and magnetic properties.

2.3 Formation of composites

The powder of LSMO and BCZT composition are used to form the required ME composites using the following formula

yLSMO + (1- y) BCZTx = yLBCZTx with y = 0.10, 0.15 & 0.20 and x = 1, 2 & 3.

These sintered yLBCZTx composites with y = 0.10, 0.15 & 0.20 and x = 1, 2 & 3 were studied by X-ray diffraction technique using Cu Kα (0.154nm) radiation. The microstructure of sintered composites was studied by using JEOL JSM – 6360A Analytical Scanning Electron Microscope. The linear and quadratic magnetoelectric coefficients α and β were determined using a custom designed instrument as reported earlier [8].

III. RESULTS AND DISCUSSION

Fig. 1 show the XRD pattern of 0.10LBCZT1 composite sintered at 1100°C. The peaks corresponding to BCZT and LSMO phase are separately identified in the XRD pattern of composites. This indicates that pure bi phase composites are formed in this process. The peaks of plane *(012), *(104) are corresponds to LSMO ferromagnetic phase, whereas peaks of plane (100), (110) are corresponds to BCZT ferroelectric phase.

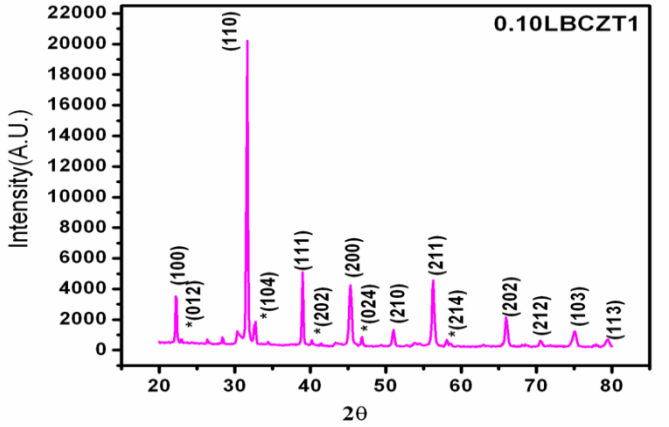

Fig.1 XRD pattern of composite 0.10LBCZT1 composite

Fig. 2 show SEM image of 0.10LBCZT1 composite. The SEM image clearly show that the sintered sample have dense structure with non-uniform grain size distribution and it is seen to be spongy. The average uneven grain size of 0.10LBCZT1composite is observed to be 0.7935 μm, 0.6135 μm for 0.15LBCZT1 composite and 0.8411 μm for 0.20BCZT1 composite. The average uneven grain size of 0.10LBCZT2 composite is observed to be 1.5628 μm, 1.1321 μm for 0.15LBCZT2 composite and 1.4653 μm for 0.20BCZT2 composite. The average uneven grain size of 0.10LBCZT3 composite is observed to be 1.5577 μm, 1.6156 μm for 0.15LBCZT3 composite and 1.1682 μm for 0.20BCZT3 composite.

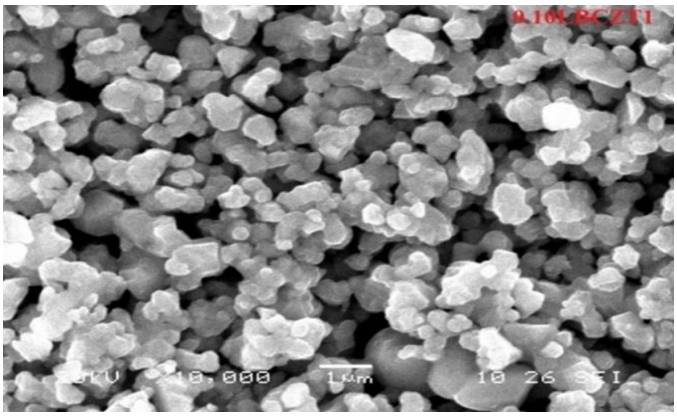

Fig.2 SEM image of 0.10LBCZT1 composite.

3.1 Magnetoelectric properties

The linear ME coefficient α and quadratic ME coefficients β of the composites has been measured at 4 KHz using a custom designed measurement unit [8].

Linear ME coefficients α is calculated using the following formula. $\alpha = \frac{dv}{dh \times d \times G}$

Where dv is r. m. s output voltage developed across the sample in mv, h is r. m. s value of A.C field at frequency 4KHz, G is Gain of the amplifier =19 and d are the thickness of the sample.

Quadratic ME coefficients β is calculated using the following formula. $\beta = \frac{dv}{dH \times 2h \times d}$

Where dv is r. m. s output voltage developed across the sample in mv , dH is imposed d. c magnetic field and h - r. m. s value of A.C field at frequency 4KHz.

Tables 1, 2 and 3 shows the variation of linear and quadratic ME coefficients α and β as a function of y for yBCZTx with y = 0.10, 0.15, 0.20 and x =1,2 and 3 composites sintered at 1100°C. Further, from these tables, it can be seen that the magnitude of linear ME coefficients α is substantially large for all studied composites and thus these composites are suitable for ME applications.

In all studied composites, linear ME coefficient α is observed to be maximum for y = 0.20 composite than y = 0.15 and 0.10 composites. This feature is attributed to the y × (1-y) type proportionality of α [9]. As expected on the basis of the magnitude of λ, linear ME coefficient α is maximum for 0.20LBCZT as compared with 0.10LBCZT & 0.15LBCZT composites, but both

of these values are large and useful. In the present case, the quadratic ME coefficient β is also determined. Here, β occurs mainly because of the variation of λ with the applied magnetic field. The greater dλ/dH, the greater β will be also. As a device requirement, quadratic ME coefficient β is required to be as low as possible. Table 1,2 and 3 presents the values of quadratic ME coefficient β for yLBCZTx composites with y = 0.10, 0.15, 0.20 and x =1,2 and 3 composites sintered at 1100 °C. It is observed that quadratic ME coefficient β is fairly low as required for device applications.

Tables 1: Linear and Quadratic Magnetoelectric coefficients α and β for yLBCZT1composites with y = 0.10, 0.15 and 0.20.

Composition	α (mV/Oe.cm)	β × 10^{-4} (mV/Oe^2.cm)
0.10LBCZT1	2.419	3.20
0.15LBCZT1	2.762	4.502
0.20LBCZT1	3.480	4.958

Tables 2: Linear and Quadratic Magnetoelectric coefficients α and β for yLBCZT2 composites with y = 0.10, 0.15 and 0.20.

Composition	α (mV/Oe.cm)	β × 10^{-4} (mV/Oe^2.cm)
0.10LBCZT2	11.05	2.534
0.15LBCZT2	15.98	5.068
0.20LBCZT2	20.12	7.10

Tables 3: Linear and Quadratic Magnetoelectric coefficients α and β for yLBCZT3 composites with y = 0.10, 0.15 and 0.20.

Composition	α (mV/Oe.cm)	β × 10^{-4} (mV/Oe^2.cm)
0.10LBCZT3	8.718	4.520
0.15LBCZT3	9.540	6.721
0.20LBCZT3	12.54	7.820

IV.CONCLUSION

The multiferroic composites are observed to show useful magnitude of linear ME coefficient α and quadratic ME coefficient β .It is observed that the variation of ME coefficient α with y could be correctly correlated through the basic feature of ME properties of composites and observed morphological features.

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