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Abstract

The thesis discusses the synthesis and characterisation of Composite Multiferroics prepared by solid state route. The prepared samples were analysed by XRD, SEM, FTIR, VSM, Impedance analyser and PE loop tracer. We prepared the composites of various materials to study their structural, electrical and magnetic properties.

The $BiFeO_3$-$SrTiO_3$ composites were successfully synthesised utilising the solid-state route. The temperature of 900ºC was utilised in making the composites. The Rietveld refinement confirmed the rhombohedral structure with R3c space group bearing lattice parameters a=b=5.56812Å, c= 13.77369Å $\alpha=\beta=90$, $\gamma=120$. The stabilisation in the structure was observed due to the substitution of STO in BFO. The composites did not show any impurity peak in the XRD plot. Among the four prepared samples, in the 6B4S sample much decline was detected in the value of leakage current. The decreased value of current density was $4.205\times10^{-7}Acm^{-2}$ at an applied field of 7.41kV/cm. In addition, we observed fine PE loop of 6B4S sample with values of P_{max}(=0.5472μC/cm^2) and P_r(=0.1422μC/cm^2), which are fairly high as compared to lower concentration composites. The dielectric measurements also presented improvement in the dielectric properties with increase in the STO content in BFO. When we paid attention towards the magnetic properties of the composite system, we found 8B2S sample giving best values. An increase in the value of both remanent and saturation magnetisation was observed. The values recorded are Mr = 1.4emu/g, Ms = 4.210emu/g and $\mathbf{n}_B$=0.210. The bond length of Fe-O bond was calculated from the observed FTIR spectrum (lower wavelength region) and matching value of bond length was observed.

Secondly, the composites of $NdFeO_3$-$SrTiO_3$, were prepared using the sintering temperature of 1200ºC. XRD was performed to know the crystal structure of the prepared system and Rietveld refinement was done to find the lattice parameters and volume of the unit cell. The value of lattice parameters found was a=5.55Å, b= 7.75Å and c= 5.47Å and $\alpha=\beta=\gamma=90^o$ for NFO phase and a=3.89Å, b= 3.89Å and c= 3.89Å and $\alpha=\beta=\gamma=90^o$ for STO phase. On performing the electric measurements, we observed enhancement in both PE and dielectric properties which is credited to STO content in NFO. In addition, VSM performed also showed enhancement in the magnetic properties of the composite system. Among the prepared samples, 6N4S was found to have the highest value of saturation magnetisation (3.2emu/g) whereas the 7N3S had the largest value of saturation polarisation equal to 0.7μC/cm^2 and P_r =2.91 μC/cm^2. The variation of the relative dielectric constant of NFO-STO composite system at room temperature as a function of frequency (up to 2 MHz) and at variable temperature (100–400ºK) has also put forth promising values.

The third composite was prepared from $CuFe_2O_4$, an antiferromagnetic substance and $BaTiO_3$ a ferroelectric substance. The prepared samples were investigated by many instruments to know the crystal structure, surface morphology, electric and magnetic properties. Structure study and surface morphology were was carried out using x-rays and SEM. ^{57}Fe Mossbauer spectroscopy of x=0 sample was used to know the magnetic moment. Magnetization and ferroelectric loop tracer were used to know the nature of the hysteresis loops. The XRD results fitted with Reitveld, confirmed the single-phase formation of both $BaTiO_3$ and $CuFe_2O_4$ samples and their composite. The SEM micrographs showed the homogeneity and uniformity of the samples. The observed M-H and P-E loop of the composite $0.1CuFe_2O_4$-$0.9BaTiO_3$ sample showed the presence of spontaneous electric polarization and spontaneous magnetization. The MH loop of $0.9BaTiO_3$–$0.1CuFe_2O_4$ composite gave Ms = 3.09 emu/g, Mr = 0.3 emu/g and Hc = 200Oe. Further, The PE loop gave the value of maximum polarisation P_{max} = 5.86μC/cm^2, the value of remanent polarisation P_r =2.23μC/cm^2 and Vc= 2.24kV/cm. In conclusion, we observed that the prepared composite exhibited both ferroelectric and magnetic ordering arising due to the respective ferroelectric and ferrite phases present in the sample.

In addition to the preparation of the composites, we worked on the theme of substituting BFO at A and B sites separately and sintering temperature used was 900^{o}C. In $Bi_{1-x}Nd_xFeO_3$ ceramic samples, change in the structure has been detected with doping of Nd. The structure was observed to change from the distorted rhombohedral structure bearing space group R3c to triclinic structure whose space group is P1. The change in the structure is believed to be due to the change in the atomic radius of the substituent element. The experimentally observed ferroelectricity of BNFO system has got improved with doping Nd content. The value of leakage current density was observed to decrease with Nd doping. The value of leakage current density observed for B9N1 is 4.67×10^{-7} Acm^{-2} at an applied electric field of 13.62kV/cm. The remnant polarisation increases with increasing Nd content from 0.1 to 0.30 and shows a decrease when x becomes greater than 0.3. In addition, the doped $BiFeO_3$ samples showed ferromagnetic behaviour with higher values of Mr and Ms than BFO which is believed to exhibit antiferromagnetic behaviour. The substitution at B site in BFO has also shown improvement in its multiferroic properties. The XRD of pure BFO sample confirmed the formation of $BiFeO_3$ phase with a small amount of $Bi_2Fe_4O_9$/Bi_2O_3 phase also present in the sample. There was significant structural change observed in doped compound as observed from the XRD pattern. The XRD pattern was made to compare with the Pcpdfwin software and was found to match with PDF No. 450397. The formation of $Bi_3FeMo_2O_{12}$ phase was observed possessing monoclinic structure and space group C2/c. The nature of bonds and the value of Fe-O bond length was determined by Fourier Transform Infrared spectroscopy (FTIR) found equal to 1.88Å. Mo doping is helpful in reducing the leakage current and also enhancing the PE loop. We observed an increase in the value of P_r and P_{max} in BF4M6 sample. The Field Emission Scanning Electron Microscope showed an increase in the grain size of BF4M6 sample as compared to pure BFO.