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Effect of Ba5RTi3V7O30 (R=Ce, Pr) in the magnetic properties of BiFeO3.

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Abstract— Polycrystalline Samples of the composite $(BiFeO_3)_{1-x}(Ba_5HoTi_3V_2O_{30})_x$ (BFO-BHTV) is prepared by high temperature Solid State Reaction Technique. The formation of the material has been confirmed through X-Ray Diffractometer (XRD) to ensure the single phase formation. Dielectric characterisation has been done by Impedance Analyser (HIOKI-IM3536) for variation of dielectric constant with frequency and with temperature has been studied in a wide range to see the dielectric anomaly, magnetisation study is also done by Vibration Magnetometer (VBM) for different composition i.e. x in the composite (BFO- BHTV). The aim of the study is to determine the nature of composite material by changing the Holmium (Ho) in the composite with other similar types of Rare Earth elementthen to see at what x value the composite will have a balanced magnetic and electric properties.

Keywords—Dielectric, Solid State Reaction, Impedance Analyser, Magnetisation, XRD

1.INTRODUCTION

Ferroelectricity was first discovered in 1920 in Rochelle salt by Valasek. He found the non-linear relationship between the dielectric polarization and the electric field of Rochelle salt, and thus a hysteresis loop was observed similar to B-H loop obtained in ferromagnetic materials. Thehysteresis loop, showing remanent polarization is the main deciding factor for the data storing. In 1950ceramicferroelectric $BaTiO_3$ (Barium

Titanate) was discovered. The compound of this family exhibit/possess many interesting properties such as electro-optic, pyroelectric, piezoelectric, photorefractive properties etc.

In 1957 Royen and Swars synthesized a multiferroic material $BiFeO_3$. It is one of the few single phase material that exhibits both ferroelectric and antiferromagnetic properties in the room temperature. Now research is going on to synthesis and to produce many material of multiferroic and their new applications are being discovered day by day.

In multiferroic material they show both magnetic hysteresis and electric hysteresis. Now research is going on to find out a particular composite material made by combining multiferroic with some

ferroelectrics. By balancing the structure,the composite material can be perturbed by both electric and magnetic field. We have to investigate whether by suppressing some properties of electric property if magnetic property regenerate or further suppressed or remain invariant.

By detail literature survey we have found that when some multiferroic materials are mixed with some ferroelectric material by solid state reaction then a particular balanced condition between electrical and magnetic property is achieved.

Some of our Collaborator investigated the effect of adding someferroelectric material like $Ba_5RTi_3V_7O_{30}$ (R=Rare earth element such as Ho, Sm, La, Nd, etc.) withmultiferroic material BiFeO3.

To see the change in the magnetic property of Bismuth Ferrite pure ferromagnetic materials are added forming the composition $(BiFeO_3)_{1-x}(Ba_5RTi_3V_2O_{30})_x$,then the value of x is changed to get balanced magnetic and electric properties.

In our proposed work we have gone through some literature where we have taken a case study where ferroelectric materials$Ba_5HoTi_3V_2O_{30}$(BHTV) of certain composition is mixed with multiferroic$BiFeO_3$ (BFO)in proper balancing, then there is a better electric and magnetic ordering, showing both in proportionate form. Our aim is to see the effect of addition in the magnetic hysteresis of BFO at the same time to see how it is suppressing the electric property at which proportion of x it will show the best performance for both magnetic and electric response

The outline of this paper is as follows. In sec. 2 we present the Methodology employed. First we prepare $Ba_5HoTi_3V_2O_{30}$. Then we prepare$BiFeO_3$.Both the sample is mixed to form a composit$(BHTV)_{1-x}(BFO)_x$. In sec. 3 we discuss the Characterisation i.e. Electrical, Magnetic and Structural Characterisation. In sec. 4 we study the Results and Discussions, focussing on how the dielectric constant ($\mathcal{E}_r$) varies with frequency and temperature. In the same section we study Dielectric of $(BHTV)_{1-x}(BFO)_x$ and also study Magnetization. In sec.5, we conclude with some remarks.

2.METHODOLOGY

a.Preparation of $Ba_5HoTi_3V_2O_{30}$(BHTV)

For the synthesis of ceramics sample for our investigation solid-state reaction technique could be employed

For preparation of polycrystalline specimens of $Ba_5RTi_3V_7O_{30}$, (R=Ho, Ce, Nd, etc.). The stoichiometryamount are calculated by the reaction:

$5BaCO_3 + (1/2)R_2O_3+TiO_3+ (7/2)V_2O_5=Ba_5RTi_3V_7O_{30}$
$+5CO_2$

These compounds are stoichiometry ground in agate mortar by mixing with methanol then the mixture is calcined in muffle furnace at temperature of ~ 750^oC

The calcined powder is further ground to fine form so that it will be in nanoscale size.

The powder isdie pressed under a pressure of ~ 7 tons by hydraulic pressure to a pallets form of size about 1 cm. The pallets are sintered at ~800^oC.

b.Preparation of $BiFeO_3$ (BFO)

Fe_2O_3 and Bi_2O_3 are mixed in stoichiometric ratio and calcined at 700^oC for 4 hr to form $BiFeO_3$.

c.Preparation of $(BHTV)_{1-x}(BFO)_x$

The calcined powder of $BiFeO_3$ and $Ba_5HoTi_3V_2O_{30}$ with different proportions are mixed with a small amount of PVB (polyvinyl butyral) as a binder and the powder is die pressed under a pressure of ~ 7 tons by hydraulic pressure to a pallets form of size about 1 cm. The pallets are sintered at 800^oC.For 6 hrs in environs air followed by cooling at 2^oC/min.

Finally the pallet are painted with silver paste to make it conducting on both sides then the electrical measurement is done.

3.CHARACTERISATION

a.Electrical Characterisation

Using a HIOKI model dielectric constants and the loss factor (tanδ) could be measured for different frequency at different temperature. The Transition temperature (T_c) is marked for different variations of x and see how T_c is changing. Rare earth are also changed and observation is taken for tanδ, Ɛ, maximum Ɛ, T_c response.

b.Magnetic Characterisation

Magnetic hysteresis is obtained by the Vibration magnetometer, the remnant magnetisation is measured for further harnesses.

c.Structural Characterisation

Then structural characterisation of the calcined powder has been put into X-Ray Diffractometer (XRD) from XRD the structure of the material could be found whether the compound form is a single phase or not

4.RESULTS AND DISCUSSIONS

Variation of (ε_r) with frequency

From fig.1 it can be seen that the value of relative dielectric constant (ε_r) decreases from ~64 to ~18 for frequencies increasing from 1 kHz to 1MHz, which is a general features of polar dielectric materials.

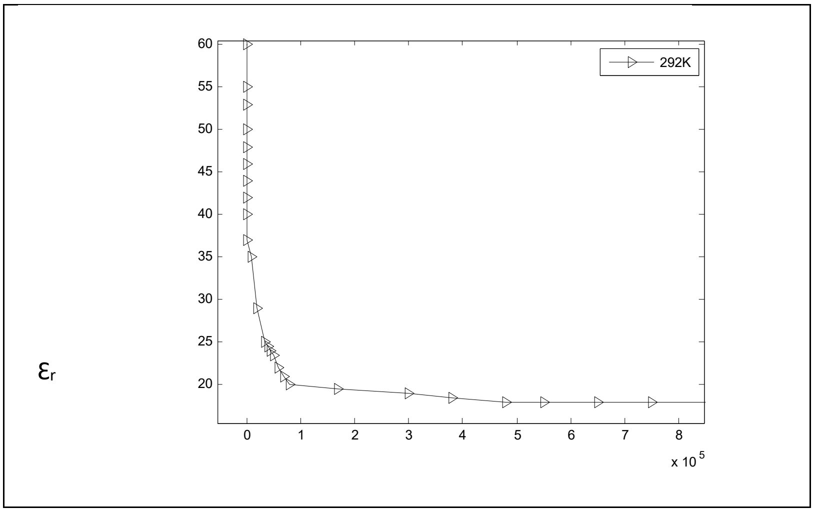

Fig. 1Frequency variation of ε_r of BHTV (at room temp)

Variation of (ε_r) with temperature

From fig.2 it can be seen that with the rise of temperature the value of ε_r first increases reaches its peak value which is its transition temperature (T_c), and after slight decrease it further increases for the mentioned frequencies.

From Table II, it can be seen that for 1 kHz, dielectric constant, ε_{rmax} is 2316which is relatively very high in comparison to values at 10 kHz and 100 kHz, it may be because it represent the grain boundary region. This dielectric anomaly at $T_c = 240^oC$ (513K) corresponds the ferroelectric-paraelectric phase transition. The presence of peaks in ε_r- T graph is an indicator for ferroelectric nature of the compound. There is another peak at 378^{o}C for 10 kHz.

Hence, the dielectric constant may be due to the contribution from both dipole orientation and long-range migration of charge species. The second peak may be considered because of an ionic transition.

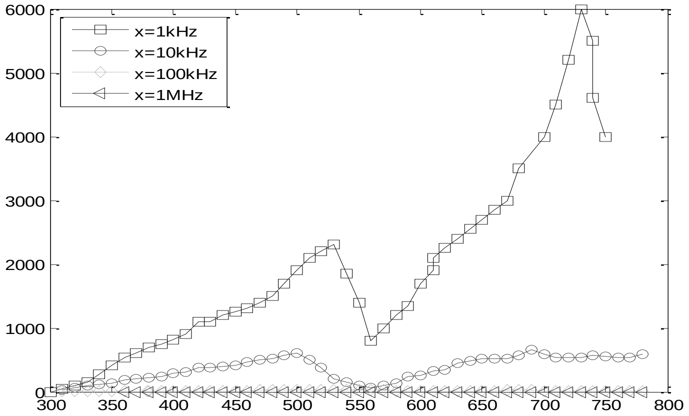

Fig. 2 Variation of Ɛr with temperature (K) for BHTV at various frequencies

TABLE VII

Compounds	**$Ba_5HoTi_3V_7O_{30}$**			
f(kHz)	1	10	100	1000
$Ɛ_{RT}$	64	41	23	18
$Ɛ_{max}$	2316	651	163	41
T_c(K)	513	501	500	498

Comparison of dielectric properties and T_c(K) of BHTV compounds at different frequencies

Dielectric study of $(BHTV)_{1-x}(BFO)_x$

Fig.3 shows dependency of dielectric constant ($Ɛ_r$) with temperature at frequency 100 kHz for $(BHTV)_{1-x}(BFO)_x$ with x ranging from 0 to 0.5 and temperatures ranging 300K-800K.

Table II shows the values of T_c and $Ɛ_r$ for $(BHTV)_{1-x}(BFO)_x$ (x=0, 0.1, 0.2, 0.3, 0.4, 0.5). It is clear that T_c significantly increases when x changes from 0 to 0.3 and then decreases for 0.3 to 0.5.

The change in T_c is due to the addition of BFO into BHTV in different proportions which might have caused mismatch in the ionic radii of Fe^{3+} and V^{5+} as the V^{5+} ionic radii is smaller than Fe^{3+}. It was found that even with small amount of BFO, i.e., x=0.1, there is considerably change in the ferroelectric property of BHTV.

The decrease of T_c for x=0.4 and 0.5 indicates that there is a modification in ferroelectric property of BHTV by addition of BFO. Upto x=0.3, the compound is predominately ferroelectric in nature.

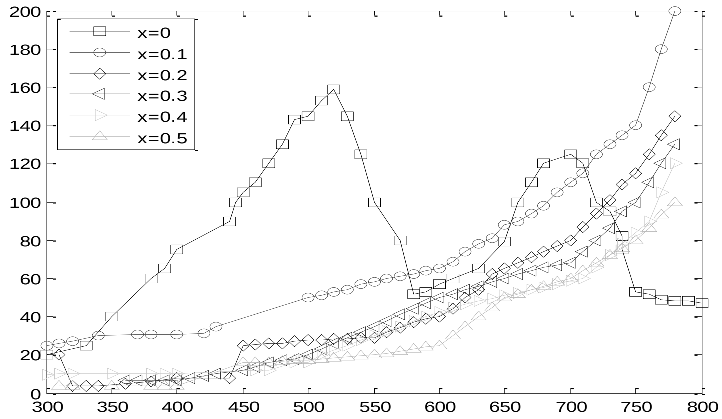

Fig. 3 Variation of ε_r with temperature (K) for

$(BHTV)_{1-x}(BFO)_x$ at 100kHz frequency

TABLE VIII

Frequency	**100 kHz**	
$(BHTV)_{1-x}(BFO)_x$	**T_c(K)**	**ε_r**
X=0	500K	163
X=0.1	603K	48
X=0.2	625K	41
X=0.3	631K	42
X=0.4	613K	43
X=0.5	538K	48

There is an increase Curie temperature till x=0.3 and beyond x=0.3 the solid solution is dominated by BFO (i.e. multiferroics) where Neel temperature (T_N) decreases because of the presence of BHTV. This mismatch may be caused by the presence of different concentration of BHTV and BFO which introduces distortion in the oxygen octahedron of BHTV showing significant variation in T_c and ε_r. The oxygen octahedron undergoes distortion due to mechanical stresses caused by the increase in the percentage of BFO, resulting in the decrease of transition temperature since T_c depends upon the structural changes. Here the little amount of BFO can make a complete solid solution which indicates that until x=0.3 in composite falls in the morphotropic phase boundary (MPB) region. Even with the low concentration of BFO in solid solution, BFO dominates the behaviour of compound and it is substantiated by the magnetization behaviour.

Magnetization study

The Magnetization vs. magnetic field has been studied by VSM and fig. 4 show that for x = 0.1, it is perfectly paramagnetic. But at x=0.4 it starts showing ferromagnetic properties (fig. 5). These results are in agreement with the dielectric data i.e. x>0.3 onwards it is predominately multiferroics.

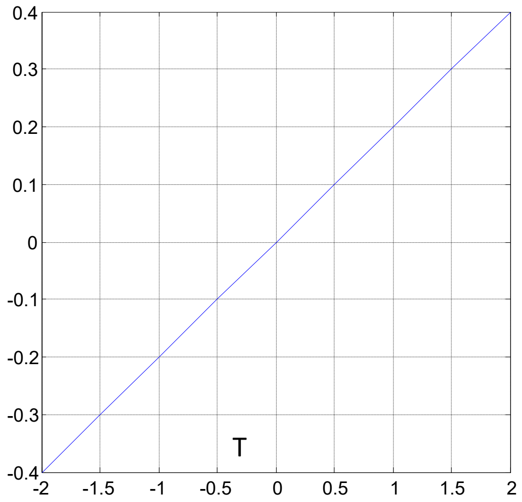

Fig.4 Coercive field (T) vs Magnetization (M) for $(BHTV)_{1-x}(BFO)_x$ at x= 0.1

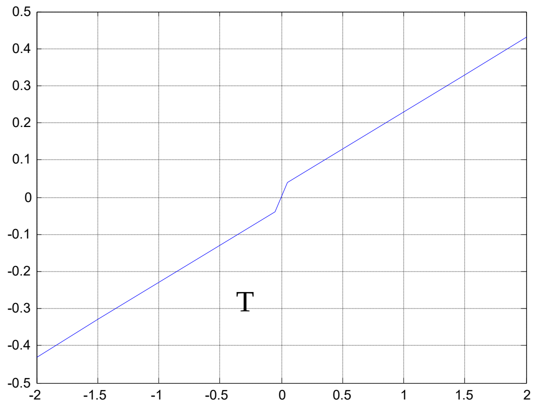

Fig. 5 Coercive field (T) vs. Magnetization (M) for $(BHTV)_{1-x}(BFO)_x$ at x=0.4

5.CONCLUSIONS

After detail literature survey we have seen that when multiferroic $BiFeO_3$ is mixed with ferroelectric materials $Ba_5HoTi_3V_2O_{30}$to form a composite material $(BiFeO_3)_{1-x}(Ba_5HoTi_3V_2O_{30})_x$, the magnetic property of Bismuth Ferrite is modified.

We are planning to study the nature of composite material by changing the Holmium (Ho) in the composite by other Rare Earth element (R: Ce, Pr, Tb, etc.) then to see at what x value the composite will have a balanced magnetic and electric properties.

Further plan is to investigate by doping Ti element in $Ba_2RTi_3V_7O_{30}$ with different proportion of similar group element such as Zr and to see how the properties of multiferroic compound is being changed overall.

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