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Magnetic Behaviors of $BaTiO_3$-$BaFe_{12}O_{19}$ Nanocomposite Prepared by Sol-Gel Process Based on Differences in Volume Fraction

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Abstract. Barium titanate $BaTiO_3$ (BTO) - barium hexaferrite $BaFe_{12}O_{19}$ (BHF) nanocomposite could be as a raw material of multiferroic. Multiferroic is a class of materials with coupled electric, magnetic and structural order parameters that yield simultaneous effects of ferroelectric, ferromagnetism and ferroelasticity in the same material. This material has potential applications in such as spintronic devices and sensors. This work was an earlier research towards formation of multiferroic material. Knowing magnetic properties that will lead to a better understanding of magnetoelectric coupling in multiferroic material is the objective of this research.The samples were BTO and BHF prepared by sol-gel and then were mixed in bulk system by a conventional techniques in various of volume fraction between BTO : BHF = 1:1 ; 1:2 and 2:1, then samples were sintered at 925^{o}C for 5, 10 and 15 hours. Composite phase study was carried out using X-Ray Diffraction (XRD). MPS Magnet – Physik EP3 – Permagraph L was used to characterize magnetic properties. XRD results confirm that composite with volume fraction of BTO : BHF = 1:1 with sintering at 925^{o}C for 5 hours consists only of 2 phases BTO and BHF. There is impurity phase $BaFe_2O_4$ beside BTO and BHF phases at samples with volume fraction BTO:BHF = 1:2 and 2:1 for longer sintering. Composite with volume fraction of BTO:BHF = 1:1 for 5 hours sintering has a high value of remanent magnetization 0.081 T and the lowest value of intrinsic coersive 333.6 kA/m leading to good characteristics of multiferroic materials.

Introduction

Over the last few years, magnetoelectric (ME) nanocomposites have received a lot of interest due to their potential applications on many multifunctional devices such as sensor, non-volatile memory etc. Due to the reason that single phase multiferroic materials are rare and their ME coupling response is weak, the magnetic field in barium hexaferrite, $BaFe_{12}O_{19}$ (BHF) induced electric polarization and electric field in barium titanate, $BaTiO_3$ (BTO) induced magnetization in ferromagnetic/ferroelectric nanocomposite. It is being extensively studied. Traditional ME effect in composite system is originated from the strain coupling around the interface of ferroelectric and ferromagnetic components, therefore there has been a great effort to optimize the growth and study of the coupling mechanism of ferroelectric and magnetic nanocomposite in bulk system.

Such materials are also known as multiferroic which have a wide range of potential applications in information storage such as spintronic devices and sensors. They are $BiFeO_3$ (BFO) produced by sol-gel hydrothermal method [1], $BaTiO_3$/$CoFe_2O_4$ superlattice films deposited by laser molecular beam epitaxy [2], BFO thin films on Pt-coated silicon wafers by pulsed laser deposition [3], YMn_2O_5 fine powder derived from hydrothermal process [4], BTO-BHF prepared by conventional solid state sintering technique [5], BFO produced by spark plasma process [6] and gel combustion [7] etc.

It is not an easy task to produce nanomaterials by conventional techniques. Coupled with high reaction temperatures (>2000K), makes it difficult to synthesize nanosize structures with high surface area [8]. The ferroelectric-ferromagnetic ceramics of BTO-BHF in this research were prepared by sol-gel process which could produce nanosize powder. This research was being an earlier research to form multiferroic materials. The objective of this research was to know magnetic properties first leading to a better understanding of the origin of ME coupling in composites.

Experimental Procedure

The ceramic nanocomposites of BTO-BHF were prepared by sol-gel method for each ceramic material. The starting materials for BTO were $Ba(NO_3)_2$, TiO_2 and for BHF were $Ba(NO_3)_2$, $Fe(NO_3)_3.9H_2O$, all of analytic purity (99.0%) from Merck KGaA Chemicals, Damstadt, Germany. It was used citric acid as a fuel. First, stoichiometric amounts of high purity nitrates were processed to be dried gel after heating at 300^{o}C, 24 hours for BTO and 450^{o}C, 24 hours for BHF. Afer grinding in the mortar, the dried gel was sintered at 700^{o}C, 2 hours for BTO and 850^{o}C, 10 hours for BHF to have nanopowder ceramics. The ceramics were then mixed by conventional technique with volume fraction of BTO : BHF = 1:1, 1:2 and 2:1. Then the mixed ceramics were uniaxially pressed into pellets (bulk system) and finally sintered again at 925^{o}C for 5, 10 and 15 hours for each ratios. Composite phase study was carried out using X-Ray Diffraction (XRD) Phillips, PW 1835 Type. MPS Magnet – Physik EP3 – Permagraph L was used to characterize magnetic properties.

Results and Discussion

The calculation of Crystallite and particle size of BTO and BHF ceramics from research before shows that all samples are in nanoceramics with size of <100 nm [9]. The effecst of volume fraction of BTO : BHF in composite and time of sintering on XRD pattern could be shown in Fig. 1,2 and 3.

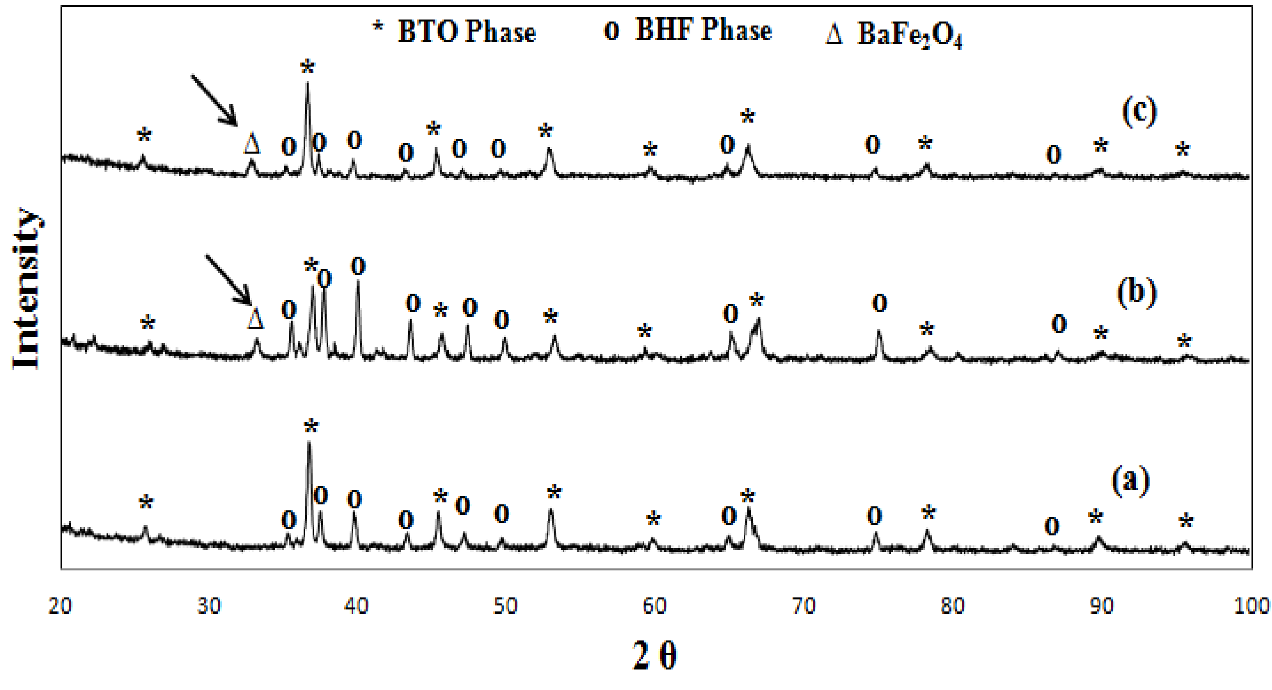


Figure 1. XRD Pattern for Samples at 925^{o}C for 5 Hours (a) Volume Fraction of BTO : BHF = 1:1 (b) Volume Fraction of BTO:BHF = 1:2 (c) Volume Fraction of BTO:BHF = 2:1

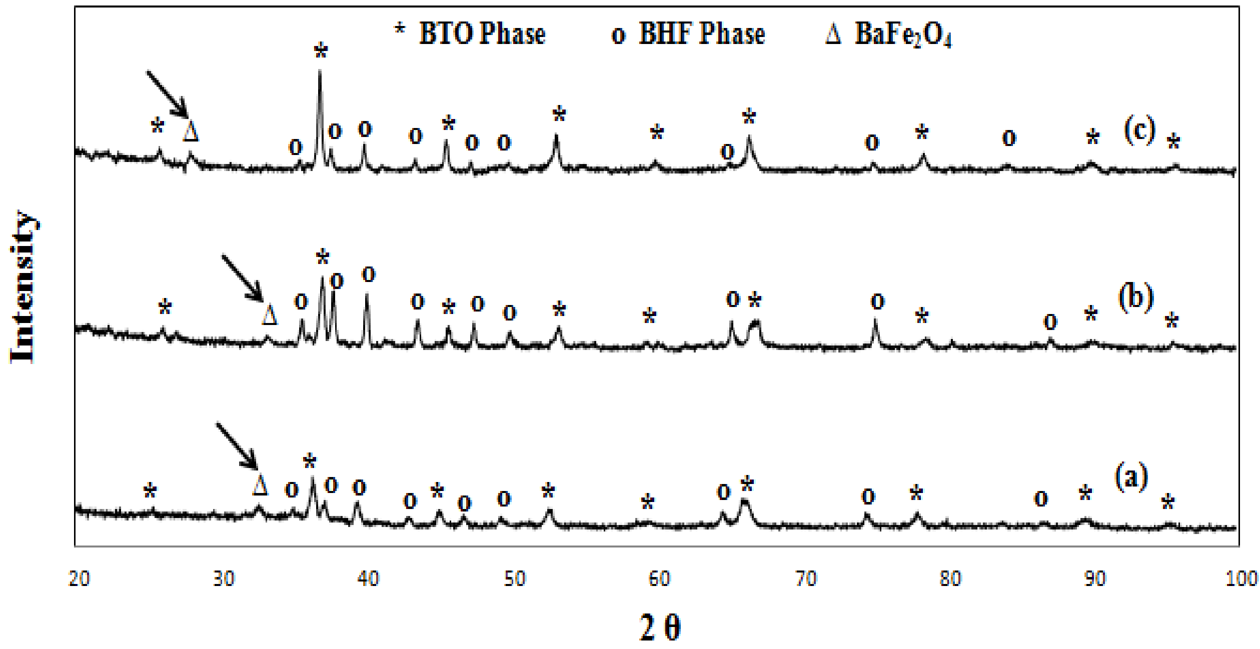


Figure 2. XRD Pattern for Samples at 925°C for 10 Hours (a)Volume Fraction of BTO:BHF=1:1 (b) Volume Fraction of BTO:BHF = 1:2 (c) Volume Fraction of BTO:BHF = 2:1

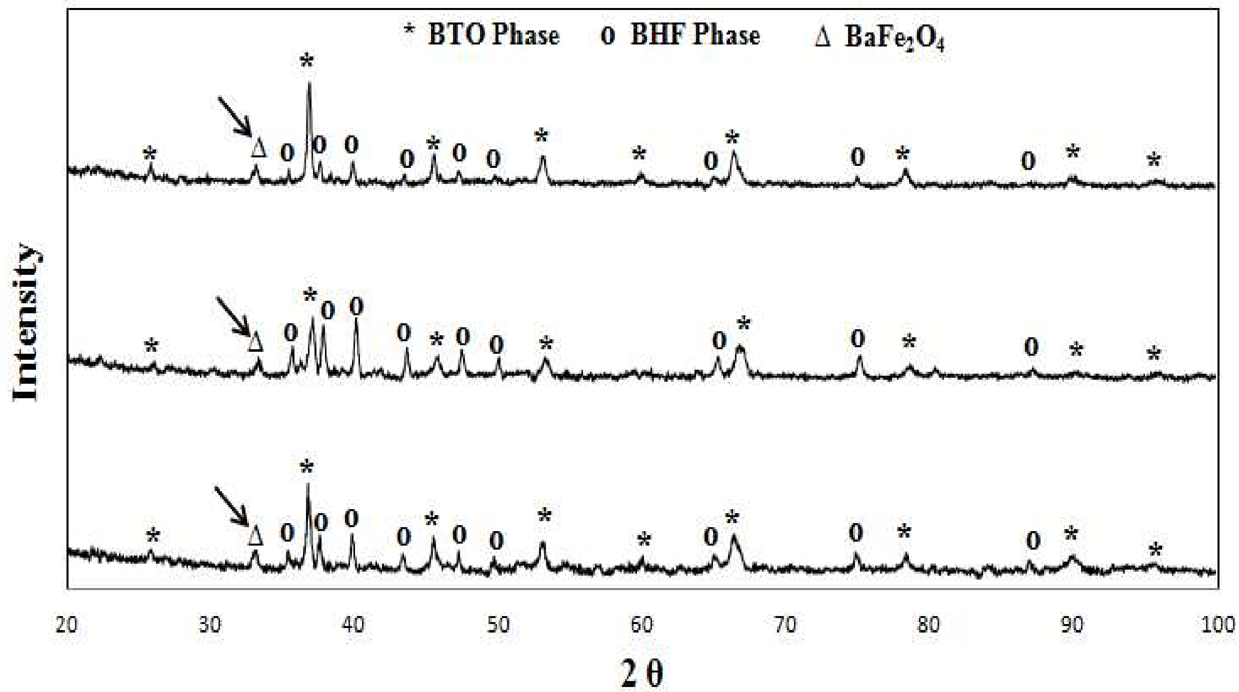


Figure 3. XRD Pattern for Samples at 925°C for 15 Hours (a) Volume Fraction of BTO:BHF=1:1 (b) Volume Fraction of BTO : BHF = 1:2 (c) Volume Fraction of BTO : BHF = 2:1

XRD results from Fig. 1,2 and 3 confirm that composite which has only BTO and BHF phases is sample with volume fraction of BTO:BHF = 1:1 with sintering at 925°C for 5 hours. Composites with volume fraction of BTO : BHF = 1:2 and 2:1 for 5,10 and 15 hours sintering and volume fraction of BTO:BHF=1:1 for 10 and 15 hours sintering, have some impurity phase $BaFe_2O_4$ beside BTO and BHF phases. That is due to reaction between excess of Ba^{2+} and Fe^{3+}in composites which have volume fraction of BTO:BHF = 1:2 and 2:1 during longer sintering than 5 hours. At the same time no sign of α Fe_2O_3 appeared. Magnetic characteristics of composites could be seen in Fig. 4, 5 and 6 as a hysteresis loops.

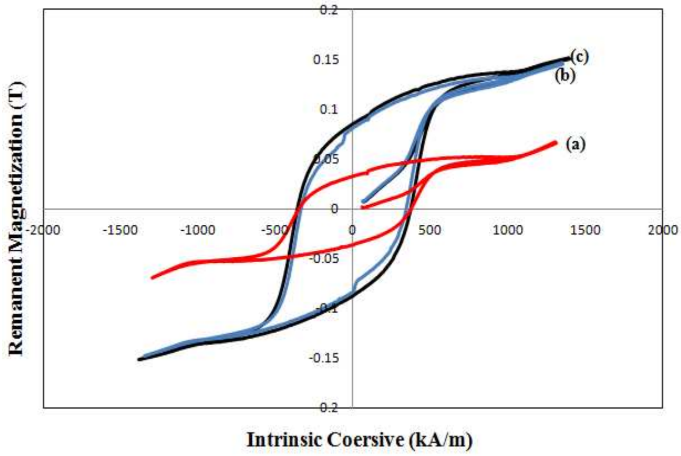


Figure 4. Magnetic Hysteresis Curve for Composite with sintering at 925^{o}C for 5 Hours
(a) Volume Fraction of BTO:BHF = 2:1 (b) Volume Fraction of BTO:BHF = 1:1
(c) Volume Fraction of BTO: BHF = 1:2

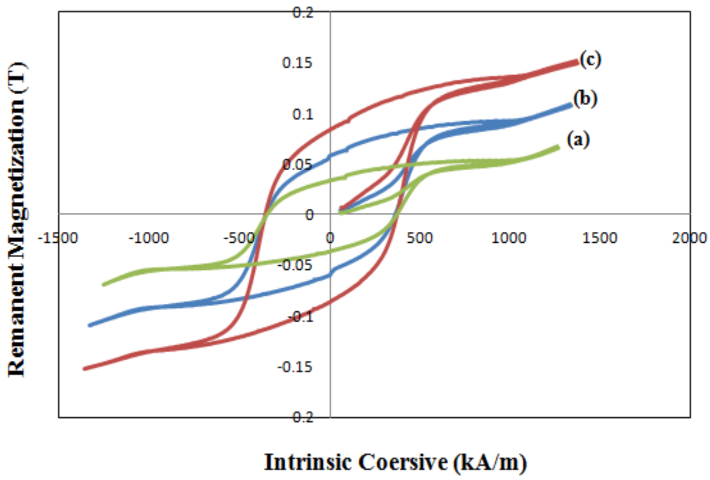


Figure 5. Magnetic Hysteresis Curve for Composite with sintering at 925^{o}C for 10 Hours
(a) Volume Fraction of BTO:BHF = 2:1 (b) Volume Fraction of BTO:BHF = 1:1
(c) Volume Fraction of BTO: BHF = 1:2

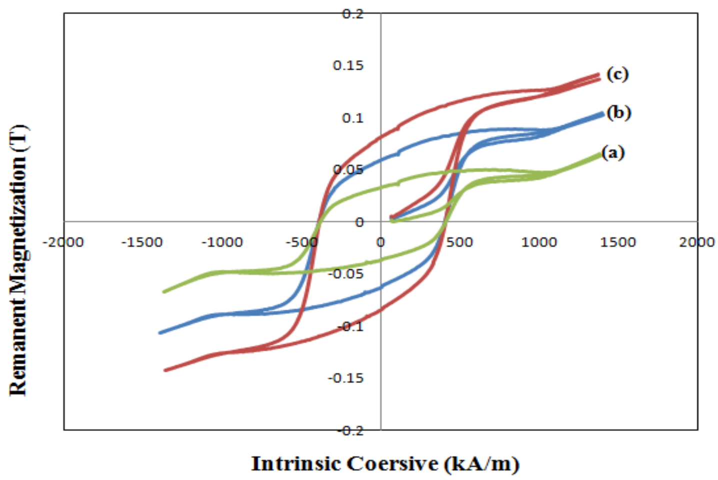
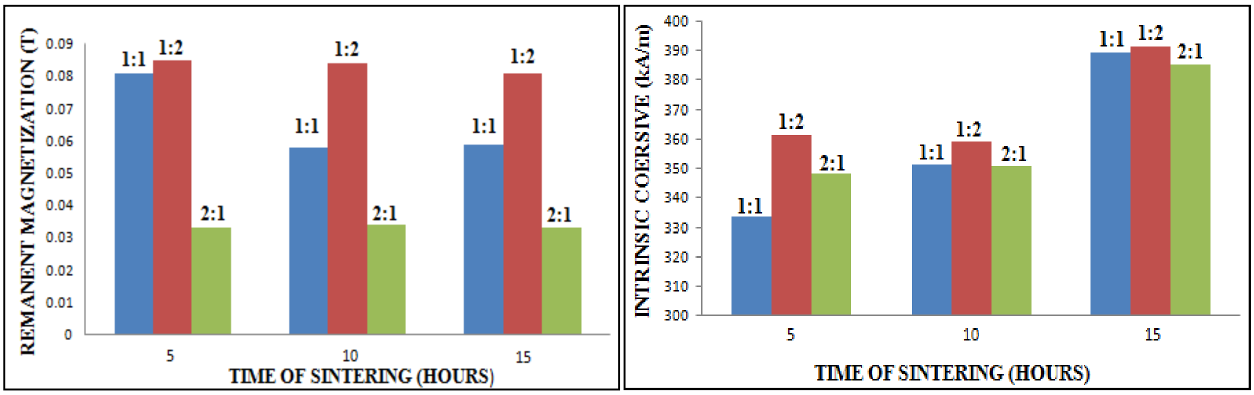


Figure 6. Magnetic Hysteresis Curve for Composite with sintering at 925^{o}C for 15 Hours
(a) Volume Fraction of BTO:BHF = 2:1 (b) Volume Fraction of BTO:BHF = 1:1
(c) Volume Fraction of BTO: BHF = 1:2

Magnetic behaviors of as synthesized composites are presented in Fig. 4,5 and 6. All of the samples show hard magentic properties. Commonly composites with volume fraction of BTO:BHF= 1:2 and 2:1 for 10 and 15 hours sintering have lower value of remanent magnetization due to the presence of impurity phase $BaFe_2O_4$ showed in XRD pattern in Fig 1,2 and 3. It could contribute to the reduction of magnetization value. The magnetic behaviors from Fig. 4,5 and 6 could be converted to numeric value presented in Table 1 and bar chart shown in Fig 7 and 8.

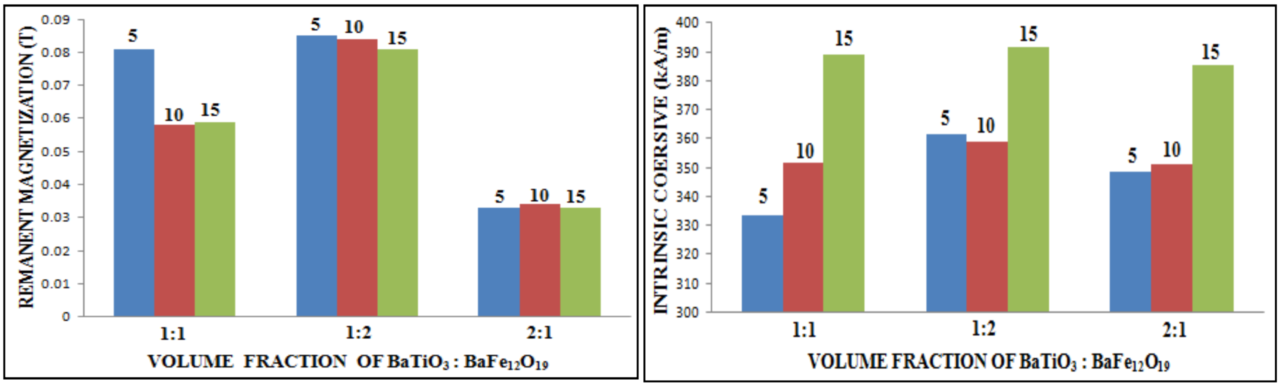
Table 1. Magnetic Properties of BTO-BHF Nanocomposite

No	Sintering Treatment	Volume Fraction of BTO:BHF	Remanent Magnetization (T)	Intrinsic Coersive (kA/m)
1	925^{o}C 5 Hours	1:1	0.081	333.6
		1:2	0.085	361.3
		2:1	0.033	348.4
2	925^{o}C 10 Hours	1:1	0.058	351.5
		1:2	0.084	359.0
		2:1	0.034	350.0
3	925^{o}C 15 Hours	1:1	0.059	389.3
		1:2	0.081	391.6
		2:1	0.033	385.3



(a) (b)

Figure 7. Remanent Magnetization vs Time of Sintering (a) and Intrinsic Coersive vs Time of Sintering Based on Differences in Volume Fraction of BTO-BHF Nanocomposite



(a) (b)

Figure 8. Remanent Magnetization vs Volume Fraction of BTO-BHF (a) and Intrinsic Coersive vs Volume Fraction of BTO-BHF Based on Differences in Time of Sintering (Hours)

From bar chart of magnetic properties in Fig. 7, it could be seen that there is a decreasing of remanent magnetization and increasing of intrinsic coersive with increasing of sintering time for each volume fraction of BTO : BHF except for volume fraction of BTO:BHF= 1:2 at 10 hours sintering, there is little decreasing of intrinsic coersive. Sample with volume fraction of BTO:BHF = 1:2 has the highest value of remanent magnetization for all time of sintering and sample with volume fraction of BTO:BHF = 1:1 has the lowest value of intrinsic coersive for all time of sintering except for 15 hours sintering, volume fraction of BTO:BHF = 2:1 has the lowest value of intrinsic coersive. From bar chart of magnetic properties in Figure 8, it could be seen that sample for 5 hours sintering has the highest value of remanent magnetization and the lowest value of intrinsic coersive for each volume fraction of BTO:BHF.

CONCLUSIONS

It could be concluded that XRD study confirms that sol-gel method could produce ceramic nanocomposite of BTO-BHF without impurity phase at volume fraction of BTO:BHF = 1:1 for 5 hours sintering at 925^{o}C . There is a trace of $BaFe_2O_4$ phase on XRD pattern at composites with volume fraction of BTO:BHF = 1:2 and 2:1. It is due to effect of excess of Ba^{2+} in volume fraction mentioned. There is a tendency of decreasing of remanent magnetization with increasing sintering time and there is a tendency of increasing of intrinsic coersive with increasing sintering time for each volume fraction of BTO:BHF. Comparing study results among all parameters, the well composite is at volume fraction of BTO : BHF = 1:1 at sintering 925^{o}C for 5 hours since it has the best perfomances due to high remanent magnetization, low intrinsic coersive and there is no impurity phase. It confirms material lead to form good characteristics of multiferroic materials. Work is ongoing to improve magnetic and ferroelectric phase distribution in order to achieve high ME coupling while maintaining good ferroelectric and magnetic properties.

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