Microstructural Studies of (Barium Strontium Hexaferrite-Barium Strontium Titanat) Composite System by Mechanical Alloying Process

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Abstract: ***In this paper, we report our investigation on material structure analysis of $(Ba_{(0.7)}Sr_{(0.3)}Fe_{12}O_{19})_{1-x}$-$(Ba_{(0.7)}Sr_{(0.3)}TiO_3)_x$ with x = 0.2, x = 0.5 and x = 0.8 composite system prepared by a mechanical alloying process to promote feroic properties. It is shown that the x-ray diffraction patterns of each composition for the composite materials are the same. It was consisted of the mixture for the two phases. The average of particle size for each respective phase in the composite materials was found initially increased,up to 18-20 μm after mechanically milled for 40 hours, then start to decreased to a smaller size ~ 8-10 μm after 80 hrs milling time. However, a plot of particle size against the milling time for each composite phase shown a trend of further reduction in the mean particle sizes. In addition, the x-ray traces ofdense pellet samples after sintering the milled powders at a temperature of 1100 ^oCshowed broadened diffracted peaks pattern due to fine crystallites in the samples. Results of mean crystallite size determination of respective phases in the composite samples showed the same trend, a decrease with milling time toward values about 10 nm at 80 hrs milling time.Hence, sintering to the milled particles has promoted the formation of nanocrystal containing particles. When compared between the mean particle size and mean crystallite size of respective phase in the composite sample at 80 hrs milling time, it is found that the mean crystallite size for magnetic phase (B_7S_3F) was more than 100 times smaller than the mean particle size of composite particles. However, finer mean crystallite sizes were found in the ferroelectric phase (B_7S_3T) in which the mean was about 200 times smaller than the mean particle size.***

Keywords: Mechanical alloying; feroic; composite; permanent magnets

1. Introduction

Barium-strontium hexaferrite and barium-strontium titanate are both well established materials which widely used respectively for permanent magnets and piezoelectric applications. As the properties are a structure sensitive, materials structure must be properly designed to meet a specific application. Both types of materials have excellent chemical stability and relatively cheap to produce. Barium Strontium Titanate (B_7S_3T) is known as ferroelectric material and considered as a good candidate for high performance piezoelectric applications [1], [2]. Ferrite magnets such as Barium Strontium Hexaferrite (B_7S_3F) is widely used to a differenceapplications such as magnetic, microwave and recording divice [3]-[5]. However, in new type of materials the so-called multiferoic [6] the magnetization can be induced by electric fields and electric polarization by magnetic fields. Thus, it is not surprise to find that researchs on coupling between magnetic and electric fields effects such as magneto electric (ME) in a material system have attracted great attentions of many researcher over the last few years [7]. The coupling effects between magnetization and electric polarization were shown also occured in a multi-layer of two phase multiferroic material [8]. The effects raised due to intimate contact between the layer of magnetic phase and that of feroelectric phase which further facilitating the cyrtallite exchange interaction in interfacial areas of the multi-layer system.

In this paper, we report some results of our recent investigation on the study of crystallites exchange interacting effects in a two-phase bulk system. The system is a composite between magnetic phase of $Ba_{0.7}Sr_{0.3}Fe_{12}O_{19}$($B_7S_3F$) and feroelectric phase of $Ba_{0.7}Sr_{0.3}TiO_3$(B_7S_3T) prepared by conventioanal mechanical milling technique. We present results of investigation on synthesis and material structure analysis of $(Ba_{(0.7)}Sr_{(0.3)}Fe_{12}O_{19})_{1-x}$-$(Ba_{(0.7)}Sr_{(0.3)}TiO_3)$x with x = 0.2, x = 0.5 and x = 0.8composite system prepared by a mechanical alloying process.The magnetic properties of the composites are also discussed in this report.

2. Experiment

The materials respectively $Ba_{0.7}Sr_{0.3}TiO_3$ (coded B_7S_3T) and $Ba_{0.7}Sr_{0.3}$ $Fe_{12}O_{19}$ (coded B_7S_3F) were prepared through mechanical alloying route employing a planetary ball mill with ball to powder ratio 10:1 for 80 hrs. Stoichiometric quantities of the analytical-graded precursors $BaCO_3$, $SrCO_3$, Fe_2O_3 and TiO_2 with purity better than 99 % were mixed in a planetary ball mill. Milled powders of various milling time for respective composition were taken and successively sintered at temperature 1100 ^0C to produce fully crystalline materials. The sintered powders were analysed by Particle Size Analyser for the average particle size determination.

Additional analysis by XRD was performed on un overlapping diffraction peak employing step scanning and calculation for crystallite size determination using *Debye Scherer* formula[9] :

$$d = 0.9 \times \lambda / \beta \times \cos \Theta$$

Where d is the mean crystallite size, λ is the X-ray wavelength, β is the full width at half maximum (FWHM) of a diffraction peak after correcting for instrumental peak broadening (β expressed in radians), θ is the Bragg angle. Peak broadening due to lattice strain was neglected.

Preparation steps for$(Ba_{(0.7)}Sr_{(0.3)}Fe_{12}O_{19})_{1-x}$-$(Ba_{(0.7)}Sr_{(0.3)}TiO_3)_x$ with x = 0.2, x = 0.5 and x = 0.8composite system were carried out by a mixing the respective component materials with the right mass fraction and successively co-milled in the planetary ball milling apparatue leading to a homogeneous mixture in the composite network. The milled mixture was then pressed into a cylindrical die of 25 mm diameter to form a green pellet. The pellet was sintered at 1050 ^{0}C for 5 hours towards dense composites. Powders morphology was observed under Scanning Electron Microscope. The magnetic properties of composite samples were evaluated by a permagraph equipped with an external magnetic field up to 2.3 T.

3. Result and Discussion

Plots of diffraction traces for $Ba_{(0.7)}Sr_{(0.3)}Fe_{12}O_{19})_{1-x}$-$(Ba_{(0.7)}Sr_{(0.3)}TiO_3)_x$ composite samples with x = 0.2, x = 0.5 and x = 0.8 is shown in Figure 1. All diffraction peaks for each x-ray diffraction trace were carefully identified and found that the trace consisted of a mixture between diffraction trace of $BaTiO_3$and that of $BaO.6(Fe_2O_3)$. No other phases were found in the diffraction trace. Thus,a sintering treatment at 1050 ^{0}C for 5 hours to the composite samples has resulted in a two-phase mixture material which consisted of $(Ba,Sr)TiO_3$ and $(Ba,Sr)O.6(Fe_2O_3)$ phases. The intensity of all diffracted peaks for $Ba_{0.7}Sr_{0.3}TiO_3$ increased proportionally as the fraction x increased. This is in contrary with those of $Ba_{0.7}Sr_{0.3}Fe_{12}O_{19}$ and thus confirm the composition of composite samples qualitatively. We have carried out quantitative analysis to the diffraction traces from which results of analysis is presented in Table 1.Composite samples with x = 0.2, 0.5 and 0.8 must have mass fraction rasioB_7SF/B_7ST respectively 4.0, 1.0 and 0.25. Thus, results of quantitative analaysis for the mass fraction of phases as listed in Table 1 for the three samples have confirmed the designated value for the mass fraction of phases present in the composite samples.

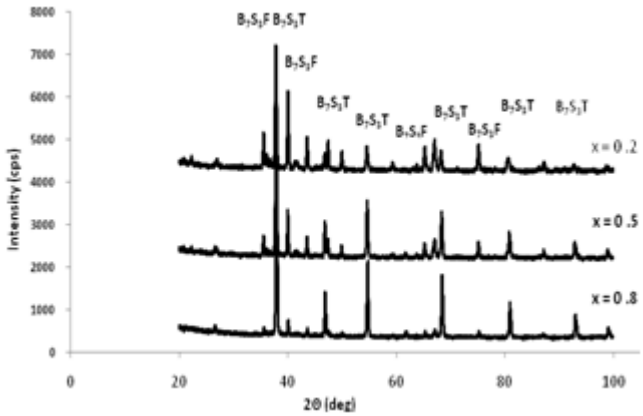


Figure 1.XRD traces of B_7S_3F- B_7S_3T composites with x = 0.2, x = 0.5, and x = 0.8

Table 1: Results of quantitative analysis of composite samples

No	$(B_7S_3F)_{(1-x)}$-$(B_7S_3T)_x$			
		Mass fraction		Wt fraction
	x	B_7S_3F (%)	B_7S_3T (%)	rasioB_7S_3F/ B_7S_3T
1	0.2	73.6	26.3	2.8
2	0.5	52.5	48.5	1.1
3	0.8	22.7	78.3	0.28

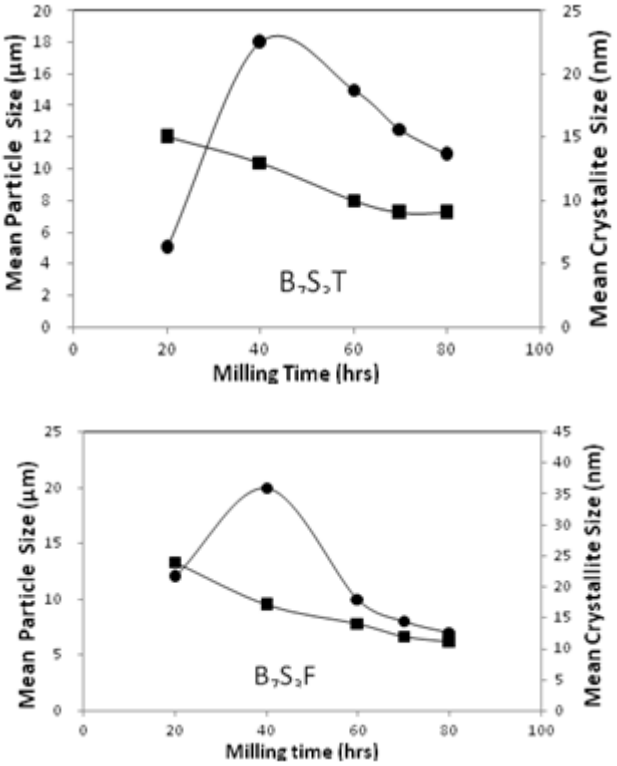


Figure 2: XRD traces of B_7S_3F- B_7S_3T composites with x = 0.2, x = 0.5, and x = 0.8

Figure 3 compares the average of particle size and crystallite size obtained from $Ba_{0.7}Sr_{0.3}TiO_3$ based samples. The average particle size was found initially increasedup to ~ 18 µm after mechanically milled for 40 hours, then start to decreased to a smaller size ~ 10 µm after 80 hrs milling time.

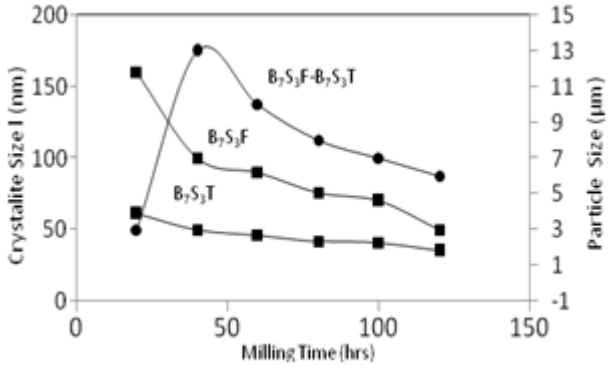


Figure 3: Crystallite Size (■) and ParticleSize (•) of B_7S_3T andB_7S_3F

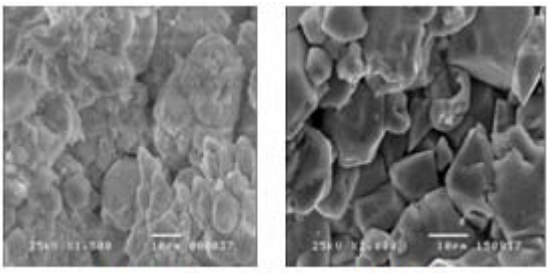
Plot of particle size against the milling time showing a trend of further reduction in the mean particle sizes.It was also observed, the x-ray traces ofdense pellet samples after sintering at a temperature of 1100 ^{o}C showed broadened diffracted peaks pattern due to fine crystallites in the samples in which values of mean crystallite size are compared in Figure 3. A similar trend was also found in the $Ba_{0.7}Sr_{0.3}Fe_{12}O_{19}$ based samples.

Table 2: Crystallite Size (■) and Particle Size (•) of composite B_7S_3F-B_7S_3T mixed during 120 hours

Milling Time	*Particle Size (μm)*	*Crystal Size (nm) B_7S_3F*	*Crystal Size (nm) B_7S_3T*
20	3	160	61
40	13	100	50
60	10	90	46
80	8	76	42
100	7	70	41
120	6	50	35

When the mean particle size and mean crystallite size of respective phase in the composite samples are compared, it was found that the mean crystallite size for magnetic phase (B_7S_3F) was more than 100 times smaller than the mean particle size of composite particles. However, finer mean crystallite sizes were found in the ferroelectric phase (B_7S_3T) in which the mean was about 200 times smaller than the mean particle size.

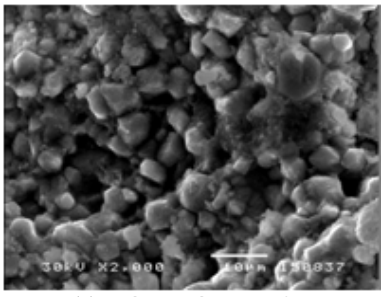
Fig 4 and Fig 5 shows the morphology of based materials and composite B_7S_3F-B_7S_3Tcrystallite observed by the scanning electron microscope. Moreover, of a fracture of the tested composite in presented in the figure 7 for each combined composite material for composition x = 0.2, x = 0.5, x = 0.8



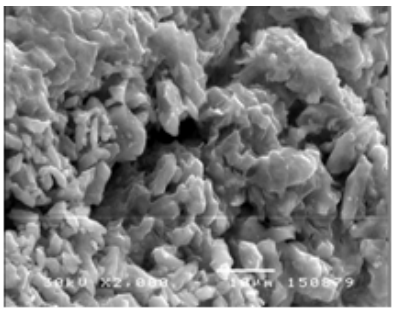
(a). B_7S_3T (b).B_7S_3F

Figure 4.SEM Result for materials (a) B_7S_3T, and (b). B_7S_3F

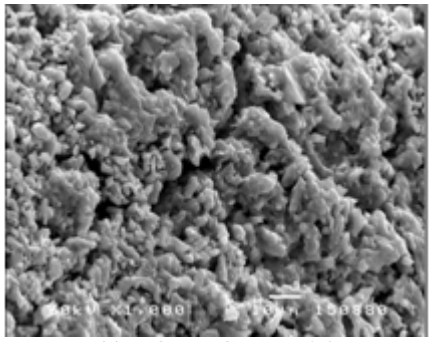
During observedof morphology of crystallite andfractures of composite's samples it was observed that the distribution of crystal in matrix in irregular. Crystallite hasirregular shape and size.



(a).B_7S_3F-B_7S_3T, x = 0.2



(a).B_7S_3F-B_7S_3T, x = 0.5



(a).B_7S_3F-B_7S_3T, x = 0.8

Figure 5: Morphology of composite(a). B_7S_3F-B_7S_3T, x = 0.2, (b). B_7S_3F-B_7S_3T, x = 0.5, (c). B_7S_3F-B_7S_3T, x = 0.8

4. Conclusion

On the light of the discussion as described above, some conclusions can be drawn. The $(Ba_{0.7}Sr_{0.3}Fe_{12}O_{19})_{1-x}$-$(Ba_{0.7}Sr_{0.3}TiO_3)_x$ with x = 0.2, x = 0.5 and x = 0.8 composites were succesfully prepared by mechanically milling the crystalline materials of the composite components. All composites consisted of the mixture of two phases. The mean crystallite size for magnetic phase (B_7S_3F) in the compoistes was more than 100 times smaller than the mean particle size of composite particles. However, finer mean crystallite sizes were found in the ferroelectric phase (B_7S_3T) in which the mean was about 200 times smaller than the mean particle size. This research will be continu to multiferoic materials.

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Author Profile

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