

EXPLORING THE EFFECT OF PARTICLE CONCENTRATION AND IRRADIATION TIME IN THE SYNTHESIS OF BARIUM STRONTIUM TITANATE (BST) $\text{Ba}_{(1-x)}\text{Sr}_x\text{TiO}_3$ (X:0-1) NANOPARTICLES BY HIGH POWER ULTRASONIC IRRADIATION

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ABSTRACT

Barium strontium titanate (BST) or $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ with $x=0-1$ possesses superior dielectric properties, which are widely used in many applications like in communication technology, electronic instrumentations, and various electrical devices. In this paper, the characterization of the particle and crystallite size of $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ (x : 0; 0.3; 0.7) is described. A two-step refinement commenced: first by mechanical milling, and then a further refinement under ultrasonic irradiation in a high power sonicator was applied to $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ (x : 0; 0.3; 0.7) particles. The crystalline powders were obtained through mechanically alloyed standard research grade BaCO_3 , TiO_2 , and SrCO_3 precursors in a planetary ball mill. The powders were first found heavily deformed after 60 hours of milling and then went through a sintering process at 1200°C for 4 hours to form multicrystallite particles. The presence of a single phase in the three samples was solidly confirmed in their respective X-ray diffraction (XRD) patterns. The changes of multicrystallite particles into monocrystallite particles were obtained only after crystalline powders were irradiated ultrasonically in a high power sonicator. The processing variable during ultrasonic irradiation was limited to the duration time of irradiation and particle concentration in the exposed media. It is shown that the average sizes of BST particles at $x=0$; 0.3; 0.7 before ultrasonic irradiation were 353, 348, and 385 nm, respectively. These respective sizes decreased drastically to 52, 35, and 49 nm, respectively, after 12 hours of ultrasonic irradiation. These particle sizes are almost identical with that of their crystallite size. Hence, the synthesis of monocrystallite particles has been achieved. As the particle concentration of media takes effect, it is shown that an exposed media with a higher particle concentration tends to form multicrystallite particles.

Keywords: Barium strontium titanate; Mechanical alloying; Nanoparticle; Particle size; Ultrasonic irradiation

1. INTRODUCTION

The dielectric property of materials is one of the most popular and intensive branches of science and technology that is constantly being updated. It is not surprising to note that dielectric materials like barium strontium titanate (BST) are found commonly in a wide range of communication technology products, electronic instrumentations, sensor technologies, and

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various electrical devices. Also, the applications of dielectric materials continue to grow rapidly. The need for cost-effective dielectric materials with superior properties that can meet volume demands will be the most important factor in product competition in the future. It has been reported that BST with $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ ($x:0-1$) compositions possesses attractive dielectric properties and fulfills the requirements of high permittivity capacitors, electro-optic devices, pyroelectric sensors, and piezoelectric transducers (Manavalan, 2005). BST has a unique perovskite crystal structure with piezoelectric, pyroelectric, and ferroelectric properties (Motaleb et al., 2013).

BST may be considered as a modified BaTiO_3 (BTO) that was intentionally made to derive new, improved properties by partial ionic substitution of Sr to Ba in BaTiO_3 . It was reported that the dielectric constant of Sr-substituted BTO of $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ ($x:0-1$) increased continuously with the increase of Sr and achieved the highest value of 1195 (1 kHz) at $x=0.3$ (Patil et al., 2007) and a value of 13,960 (10 kHz) at $x=0.3$ (Yu et al., 2013). The report demonstrates that partial ionic substitution in BTO has changed the electronic responses of modified materials towards a specific value. In addition to the composition, the microstructure of materials should also affect the electronic properties due to various factors like crystal defects, scattering sites, and crystallite size, representing the internal structure of materials. It was shown in many reports that materials with nanostructure would be an appropriate alternative for producing superior materials. In nanostructured materials, the physical phenomenon on the surface plays an important role in the enhancement of properties because the surface-to-volume ratio in materials is extremely large. Thus, the interactions between grain surfaces in nanostructured materials are expected to give additional properties and, hence, to increase their respective extrinsic properties' value when compared with that of conventional materials.

Various synthetic methods to produce nanocrystalline materials have been developed by previous researchers. These include mechanical alloying, co-precipitation (Hessien, 2008; Matutes-Aquino et al., 2000), sol-gel (Czekaj, 2010), hydrothermal (Liu et al., 1999), and sonochemical materials (Shafi & Gedanken, 1999). Selection of a synthesis method is critical since various effectiveness factors must be considered to achieve the objective; for example, it should be waste-free and not exaggerate the process. However, it should produce high yields, and, more importantly, the method should be simple. The mechanical alloying method, in particular, is considered a simple method that can also be applied to BST. The method is based on the delivery of mechanical energy through ball impacts to the elemental powder mixtures and allows the production of homogeneous materials (Suryanarayana, 2003).

The mechanical energy delivered to elemental powder mixtures would be stored in the form of stresses trapped in laminated materials obtained during the initial deformation. The mechanical treatment applied to the laminated materials with a longer milling time would eventually fragment the materials into fine powders. Crystalline powder may be obtained through a solid state reaction at a particular temperature to grow the crystal embryo (Lu & Lai, 1998; Suryanarayana, 2003). Once the crystalline powders are already obtained, an ultrasonic wave at a frequency of 20 kHz was applied to the particles containing liquid, causing wave propagation in the form of compression and rarefaction cycles. By giving an acoustic effect, it will form destruction forces to the particles and inter-particle collisions, which eventually decrease the average size of particles further (Shafi & Gedanken, 1999; Nguyen et al., 2011).

For the current research, we have explored the effects of particle concentration and irradiation time in the synthesis of BST nanoparticles with $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ ($x:0-1$) compositions. The technical parameters applied during nanoparticles preparation through high power ultrasonic irradiation was the variation of BST concentration in the reactor and the variety of irradiation

time. In this study, non-ionic surfactant was introduced to the media containing particles as the last treatment after ultrasonic irradiation.

2. EXPERIMENTAL METHODS

The BST synthesis was performed through stoichiometry mixing the standard research grade BaCO_3 , SrCO_3 , and TiO_2 Sigma Aldrich of 99% purity precursors in 97% alcohol media. All precursors were ball-milled in a planetary ball mill apparatus for 60 hours at a 160 RPM. The ratio of ball mills to precursors was 10:1. The mechanical milling process produced heavily deformed powder materials. A 2 wt% of polyvinyl acetate (PVAc) emulsion was then added to the powder materials as a binder agent. A compacting force of 10 tons was employed to produce the green compact samples that were 25 mm in diameter and 3 mm thick. Sintering the green compact samples at a temperature of 1200°C for 4 hours resulted in polycrystalline samples. The characterization of polycrystalline samples was performed by XRD PANalytical Empyrean and refined using HighScore Plus software (Speakman, 2012), which then showed the formation of a single-phase BST. The ultrasonic treatments were then applied to the crystalline powders obtained through a two-step grinding; the first refinement, through a hand grinding process, was followed by the second refinement, achieved through mechanical milling for 20 hours under a similar planetary ball mill. The preparation of liquid containing the particles was made in the ultrasonic reactor with variations of 1, 3, and 5 grams of BST in 100 ml of demineralized water. All ultrasonic treatments were performed with a Qsonica sonicator Q700, operated at 20 kHz with a transducer of 60 μm amplitude for 3 hours. To prevent agglomeration before the ultrasonic treatment finish, two drops (0.54 μL) of non-ionic surfactant as polyoxyethylene (20) sorbitan monolaurate of 0.01 by volume were added into the liquid in the reactor. Then, ultrasonic irradiation was re-applied for 5 minutes. To observe the influence of ultrasonic treatment to the particles further, ultrasonic irradiation was applied to 1 g of BST in 100 ml of demineralized water for 0, 3, 6, 9, and 12 hours. The fine powders that resulted from the ultrasonic treatment were characterized by XRD and analysed by Whole Powder Pattern Modeling (WPPM) software (Leoni, et al., 2016; Leoni & Confente, 2012) to derive the mean size of crystallites and the crystallite size distribution. The distribution of particle sizes was obtained by a Malvern Zetasizer Nano ZS Particle Size Analyzer (PSA). Field emission scanning electron microscopy (FE-SEM, FEI Inspect F50) and transmission electron microscopy (TEM, FEI Tecnai G2 SuperTwin) were employed in microstructure studies.

3. RESULTS AND DISCUSSION

Figure 1 compares the diffraction pattern of a polycrystalline sample of $x=0$; 0.3 and 0.7 compositions in the range of 2-theta 20° – 90° . It shows that diffraction patterns of the three samples remain the same, despite a small shift in the peak positions due to the presence of a partial Sr^{+2} ion substitution for Ba^{+2} in BST. The shifting in diffraction peaks was confirmed in the enlargement of (002) peak at 2-theta 45.0° – 46.5° (inset plot of Figure 1).

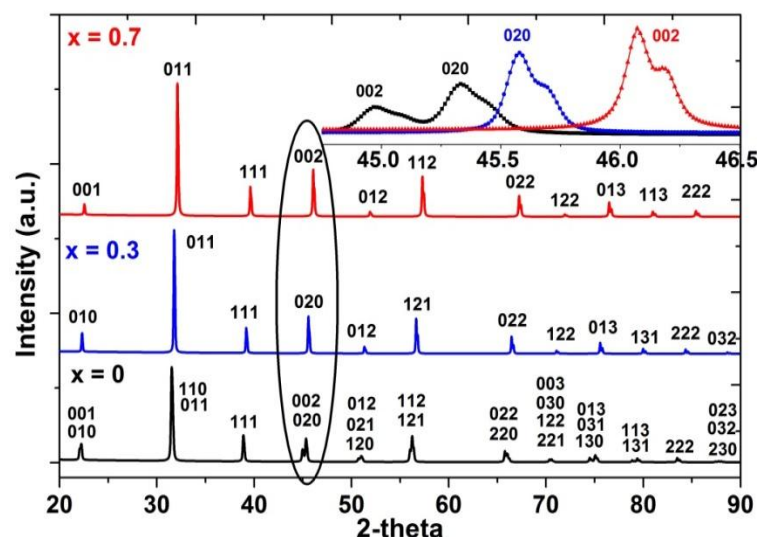


Figure 1 The comparison of a single-phase of BST diffraction patterns $x=0$; $x=0.3$ and $x=0.7$

The XRD data processing using HighScore Plus software (Speakman, 2012) for the phase identification and the determination of crystal parameters referred to the convergent fitting process parameter after an acceptable goodness of fit value was achieved. The results of the XRD data refinement showed the formation of a single-phase BST with 12 peaks, which is in line with the Inorganic Crystal Structure Database (ICSD) (www.crystallography.net, 2016) no. 98-010-0800, no. 98-005-4150 and no. 98-008-8532 respectively for $x=0$, 0.3 and 0.7 . In addition, all XRD traces consistently exhibit diffraction line broadening. All parameters resulting from data refinement for these samples are provided in Table 1.

Table1 Results of XRD data refinement by HighScore Plus software for the 3 samples

Description	X=0	X=0.3	X=0.7
Goodness of Fit	1.03845	1.43415	1.55156
Chemical Formula	BaTiO ₃	Ba _{0.7} Sr _{0.3} TiO ₃	Ba _{0.3} Sr _{0.7} TiO ₃
Mass Formula(g/mol)	233.23	216.82	196.24
Calculated Density (g/cm ³)	6.02	5.74	5.36
Crystal system	Tetragonal	Tetragonal	Cubic
Space Group	P4mm	P4mm	P m-3m
<i>LatticeParameters (Å):</i>			
a	3.99593	3.97378	3.93249
b	3.99593	3.97378	3.93249
c	4.02621	3.9716	3.93249
alpha, bbeta, gamma (deg.)	90	90	90
Volume of cell (×10 ⁻²³ cc)	6.42884	6.27154	6.08140

As shown in Table 1, there were differences in the calculated mass density of the three BST materials because of differences in the molecular weight and the unit cell volume. Obviously, this occurred due to the presence of a partial Sr²⁺ ion substitution for Ba²⁺ in BST. It was also found that a phase transformation occurred as the result of a large fraction of Sr²⁺ substitution to Ba²⁺ in a BST of $x=0.7$ composition. It is then assured that the BST crystal structure of $x=0$ and $x=0.3$ compositions was tetragonal while that of $x=0.7$ was a cubic structure. Previous research has reported (Ioachim et al., 2007) that the substitution of Sr²⁺ into Ba²⁺ in a large fraction resulted in a distortion in the tetragonal system. Hence, the phase transformation from

tetragonal to cubic takes place followed by a decrease in a tetragonal lattice. The decrease of the lattice parameter value was due to a smaller radius of Sr^{2+} (0.125 Å) than that of Ba^{2+} (0.142 Å) (Thongtem et al., 2010).

We explored further the effects of powder refinement by hand grinding, remilling and ultrasonic irradiation on a pre-ground, sintered bulk BST sample, with a deeper focus given to samples under ultrasonic treatment.

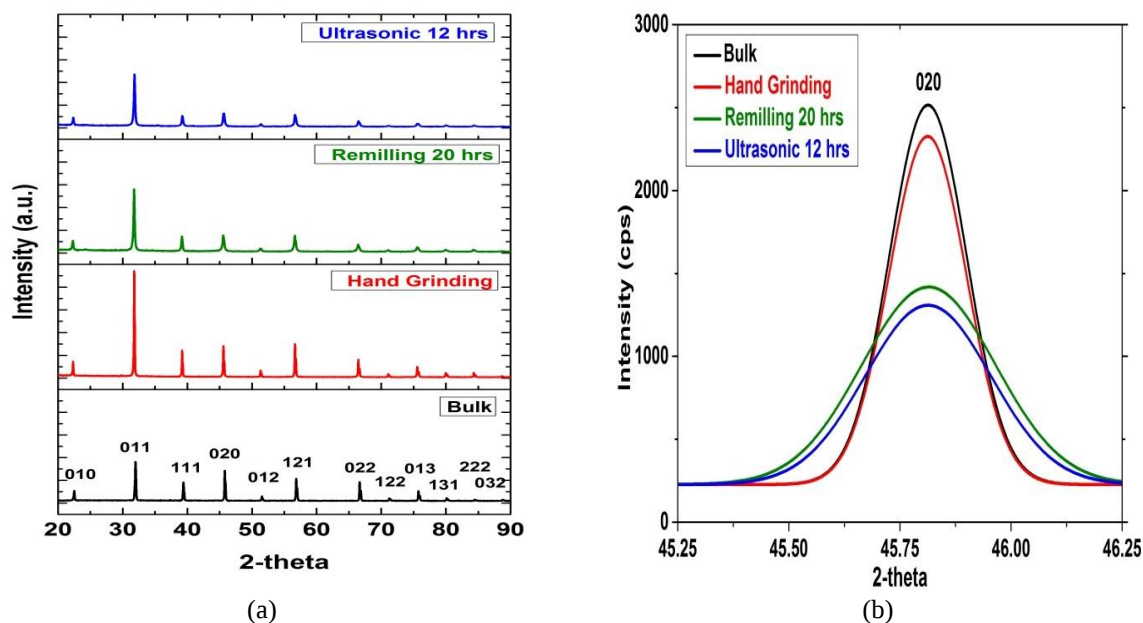


Figure 2 The diffraction patterns at: (a) different stages of treatment; and (b) the plot of a (020) diffracted peak

Figure 2a compares the XRD pattern of the samples in question, taken from a BST sample of $x=0.3$ composition as a representative. It shows that all samples have a similar diffraction pattern at various stages of refinement, which consisted of hand grinding, remilling, and 12 successive hours of ultrasonic irradiation. Such treatments did not change the XRD pattern, indicating that no phase changing has taken place so far. However, once again, all XRD traces consistently exhibit diffraction line broadening as shown in Figure 2b, which then also ensured no shifting in diffraction peaks. As to our current understanding, it is still well-accepted that the line broadening of diffraction peaks is caused by fine crystallite sizes, crystal defects, and induced microstrains, in addition to instrumental broadening.

The measured full width half maximum (FWHM) values of the diffraction peaks can be used to estimate the mean crystallite size for samples by using the Scherrer formula (Monshi, 2012). Figure 3 compares the results of the mean crystallite size evaluation of samples after receiving various refining treatments. It was found that the mean crystallite size for treated samples progressively reduced. The smallest mean particle size was obtained in the ultrasonically treated sample in comparison to the first three.

The provision of ultrasonic waves at 20 kHz created millions of bubbles, and the bubbles continuously enlarged with the increase in ultrasonic irradiation time, so that, at any time, the bubble could burst (Gedanken, 2004). This cycle created vacuum waves moving at high speed (~ 400 km/h) when the waves were destroyed, resulting in a high local temperature (~ 5000 K), high pressures (~ 1000 atm), and a fast cooling rates ($>10^9$ K/sec) (Heielscher, 2005). The particles around the bubbles strongly collided, causing damage in the particles and breaking

them into smaller particle sizes. The local pressure of about 1000 atm that resulted from the bubbles certainly was responsible for breaking the particles into fine crystals.

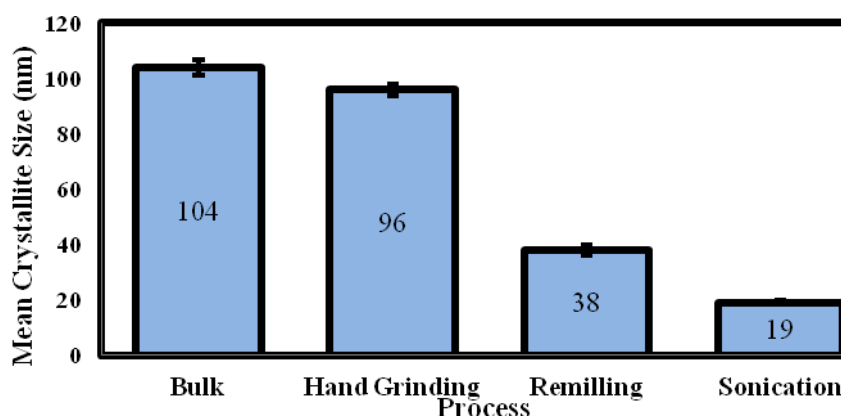


Figure 3 The mean crystallite size of a BST sample with $x=0.3$ obtained after different stages of treatment

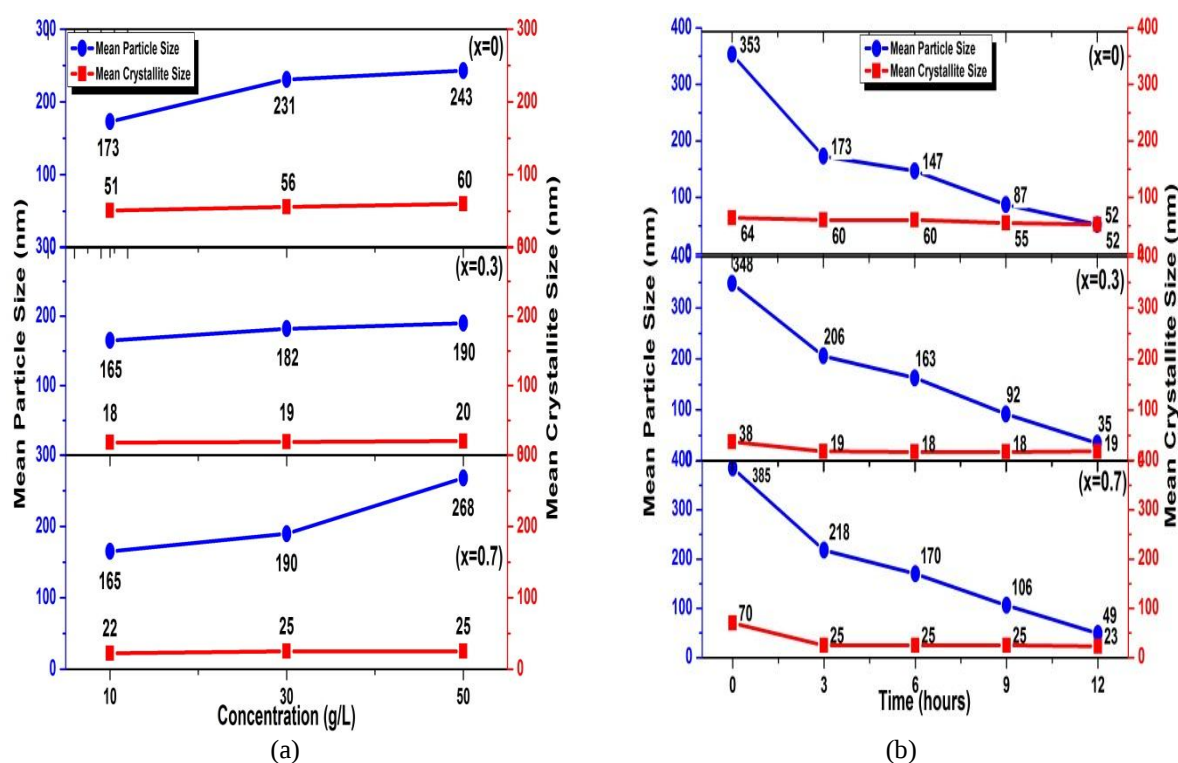


Figure 4 Comparison between mean particle size and crystallite size of BST after Ultrasonic treatments: (a) in an ultrasonic medium containing particles with different concentrations; and (b) with various ultrasonic treatment times

The results of the mean particle and crystallite size evaluation of BST samples were summarized in Figures 4a and 4b. Particle concentration in the ultrasonic reactor during the 3 hours of irradiation time seems to have had almost no effect on the crystallite size. However, the average sizes of particles tend to slightly increase with the increase in the particle concentration of irradiated media. These apply to the three samples (see Figure 4a). In a high particle concentration, the space between particles becomes more limited compared to that of low concentrations. Consequently, particles of high concentration that contain in the medium

would not collide maximally due to bubbles produced by ultrasonic waves. A slight increase in mean particle size was observed in all irradiated media containing BST particles with a concentration higher than 10 g/L. This indicated that the reduction of mean particle sizes by ultrasonic irradiation is less effective in irradiated media with high particle concentrations. Similar results were also reported, showing that higher particle concentrations enhanced the viscosity of irradiated media and hindered the cavitation effect (Ali et al., 2014).

Contrary to the effect of particle concentration on the mean particle size, the length of irradiation time applied to the exposed media seems much more effective at decreasing particle sizes. However, the mean crystallite size was not affected significantly by the increased irradiation time. As is clearly shown in Figure 4b, the mean particle size decreased progressively as irradiation time increased while the mean crystallite size was almost unchanged. A significant drop in the mean particle size occurred in the first 3 hours of irradiation time. The mean size of particles continued to decrease linearly with longer irradiation times, approaching their respective mean crystallite size value. At 12 hours of duration, both the mean particle and the crystallite sizes have almost the same value.

Table 2 summarizes the numerical value of the mean size of particles and crystallites from Figure 4a, which was used to calculate the number of crystallites in the particle. It was assumed that both particles and crystallites have solid spherical physical forms with the diameter being their respective mean particle and crystallite sizes.

Table 2 A summary of the number of crystallites in the polycrystalline particles obtained in 3 different particle concentrations after 3 hours irradiation time

Concentration g/L	X								
	0			0.3			0.7		
	PS (nm)	CS (nm)	N	PS (nm)	CS (nm)	N	PS (nm)	CS (nm)	N
10	173	51	39	165	18	770	165	22	422
30	231	56	70	182	19	879	190	25	439
50	243	60	66	190	20	857	268	25	1232

PS: particle size; CS: crystallite size; and N: number of crystallites in the particle

Table 3 A summary of the number of crystallites in the polycrystalline particles obtained in a 10 g/L particle concentration after being irradiated for up to 12 hours

Irradiation time (hrs)	X								
	0			0.3			0.7		
	PS (nm)	CS (nm)	N	PS (nm)	CS (nm)	N	PS (nm)	CS (nm)	N
0	353	64	168	348	38	768	385	70	166
3	173	60	24	206	19	1275	218	25	663
6	147	60	15	163	18	743	170	25	314
9	87	55	4	92	18	134	106	25	76
12	52	52	1	35	19	6	49	23	10

Similarly, Table 3 listed mean values of particle and crystallite sizes, which are evaluated after different amounts of irradiation time along with a calculated number of crystallites in the particles. The data listed in Table 2 show that the particles of BST contained a cluster of crystallites of a few tens in number in un-doped BTO to a few hundred in doped BTO (BST of

$x=0.3$ and 0.7 compositions). A relatively large number of crystallites in the particles of doped BTO, obviously due to their mean crystallite size, are finer than that of un-doped BTO. Hence, the nucleation of crystallites in BST must be different in that of BTO.

Contrary to this, the data in Table 3 show that the duration of irradiation time is critical to produce monocrystallite particles. It shows that, basically, monocrystallite particles could be achieved after being ultrasonically irradiated for at least 12 hours in BTO and BST.

In general, it was proved that the method of mechanical alloying followed by a high power ultrasonic irradiation was effective at producing monocrystallite particles. In the current results, monocrystallite particles were found in BTO. However, the results also showed that particles containing 6–10 crystallites were obtained in the BST. In our previous report, it was also concluded that the use of a high power sonicator proved effective at reducing the size of particles prepared through mechanical alloying, leading to nanoparticles (Manaf & Hafizah, 2012).

Figure 5a shows high-resolution imaging of BST for $x=0.3$ before being ultrasonically irradiated under a high power sonicator. The SEM image shows particles of a few hundred nanometers. Hence, it shows clusters of nanocrystallites that formed due to a strong attractive force among fine crystallites. Obviously, the particles have a solid spherical morphology with a particle size of a few hundred nanometers.

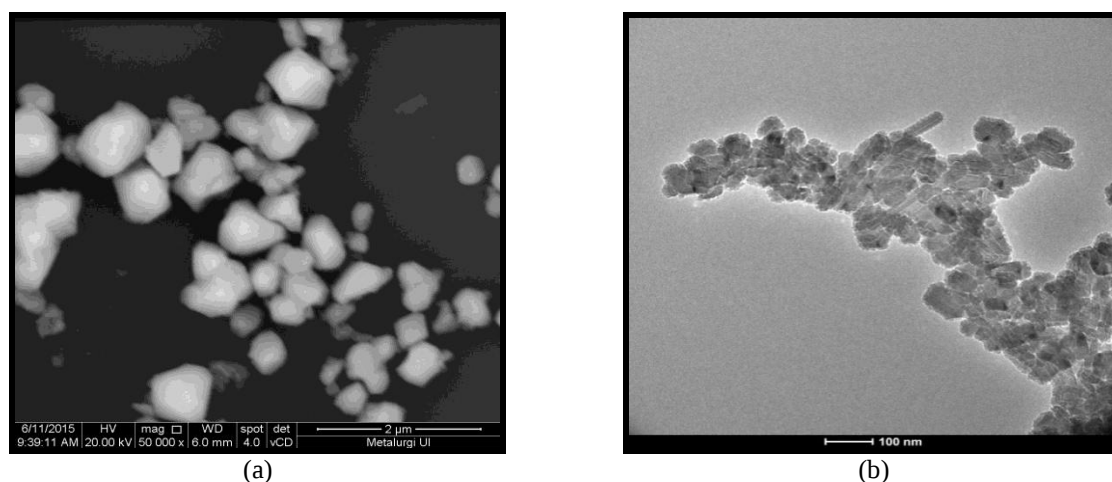


Figure 5 The morphology of BST $x=0.3$ nanoparticles: (a) before ultrasonic irradiation by SEM-FEI; and (b) after 12 hours of ultrasonic irradiation by TEM-FEI

The individual crystallites themselves can be seen in the TEM micrograph in Figure 5b, which shows fine crystallites of a few tens of nanometers in size that apparently have an elongated crystal morphology. The micrographs in both Figures 5a and 5b confirmed the results of the mean particle size evaluation by PSA and the mean crystallite size evaluation by WPPM.

4. CONCLUSION

This research has successfully synthesized a single phase of nanoparticles $\text{Ba}_{(1-x)}\text{Sr}_x\text{TiO}_3$ at $x=0$; 0.3 ; 0.7 through a mechanical alloying method coupled with ultrasonic irradiation treatments using a high power sonicator. The duration of ultrasonic irradiation time is much more effective, taking at least 12 hours to obtain monocrystallite particles at $x=0$; and at $x=0.3$; 0.7 containing 6–10 crystallites. The particle concentration of liquid media has no significant effects on the reduction of particle sizes. The mean crystallite size of $x=0$ particles is in the range 51–60 nm and even finer sizes in the range 18–25 nm have been obtained in the particles

5. ACKNOWLEDGEMENT

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