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Novel hot milling approach for the synthesis of fine $PbZr_{0.95}Ti_{0.05}O_3$ powders

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ABSTRACT

A new and effective hot milling system was designed and applied to the synthesis of $PbZr_{0.95}Ti_{0.05}O_3$ (PZT) powder. PbO, ZrO_2 and TiO_2 powders were used as starting materials and hot milled at various temperatures ranging from 10° °C to 800° °C. The obtained powders were characterized by X-ray diffraction analysis (XRD) and scanning electron microscopy (SEM). Results indicated that pure, crystalline, and fine PZT powders with the average particle size of 170 nm can be successfully synthesized directly after a relatively short synthesis time of 12 h at lower temperature of 800° °C by the hot milling technique. In contrast to this method, this study demonstrates that conventional solid-state synthesis route, which includes milling and subsequent thermal annealing, would not give rise to the synthesis of pure PZT powder, indicating the simultaneous heating and milling effectively promotes formation of the perovskite PZT in the hot milling process. Finally, the results show that the hot milling method is a simple and effective approach for the large-scale production of PZT powders.

1. Introduction

$PbZr_{0.95}Ti_{0.05}O_3$ (PZT) is a member of perovskite ceramic materials with ABO_3 general formula and a Zr:Ti ratio of 95:5. PZT has ability to release charge for generating megawatts of electrical power in a short period of time when subjected to a shock-wave compression-induced phase transition from rhombohedral ferroelectric to orthorhombic anti ferroelectric phase [1–3]. This fascinating characteristic has captured the attention of many researchers for several therapeutic applications, such as high intensity ultrasound therapy [4].

Commercial PZT powders are usually prepared by the conventional solid-state reaction process. In this method, starting materials, oxide and carbonate powders, are blended and then the obtained mixture is exposed to high temperature heat treatment to promote reactions for the synthesis of final product. Although the synthesis method is simple, the produced PZT powders with this route suffer from some important drawbacks such as non-stoichiometry in composition due to loss of Pb element, compositional fluctuation and agglomeration of the obtained PZT powders because of the high temperatures used in this process [5,6]. According to the sintering theories, the agglomerated powders have serious disadvantages to the fabrication of ceramic bodies when highly densified samples with homogenous microstructures are needed [7]. To remove agglomeration effect, a grinding step is almost always required to break large agglomerates and produce powders with better features. However, this additional milling step is time and energy-consuming and often leads to the contamination of the synthesized powders with impurities which in turn can degrade functional performance of PZT ceramics. A variety of methods have been developed to prepare fine, agglomerated-free PZT powders, including chemical co-precipitation, sol–gel process, hydrothermal reaction, reactive calcination, gel-combustion, spray pyrolysis, shock synthesis, and other related methods which are based on organic precursors [8–21]. However, there are some key disadvantages of these organic-based processes among which cost of raw organic chemicals, lack of standardization, and complicated terminology are considerable [22].

It is well-known that chemical reactivity of starting materials could be significantly improved after an appropriate milling treatment, so that the subsequent calcination temperature for the synthesis of ceramic powders can be reduced in the conventional solid-state technique [23]. Moreover, the produced powder through ball milling was reported to possess better characteristics from densification point of view. For example, it was observed that sintering temperature of the high-energy

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ball milled PZT samples was declined by at least 100 °C [24]. This process has been successfully employed to synthesize nano-sized compounds [25–27]. In addition to the appropriate milling, the reactivity of the raw materials can be influenced by temperature to a large extent for the synthesis of PZT product, increasing the purity of PZT phase by rising calcination temperature in the solid-state method [28]. Therefore, ball milling and high temperature are two effective factors in producing high purity PZT powders with small average particle size.

In this work, ball milling and high temperature (called hot-milling) were simultaneously used to synthesize high purity and fine PZT powders. It is demonstrated that, in comparison with the conventional solid-state technique, the combination of milling and heat treatment, hot milling, makes a profound impact on the production of pure and fine PZT. To the best of the authors' knowledge, to date there has been no published report on the synthesis of PZT powder based on the milling at elevated temperatures. Just a few studies have been devoted to milling at high temperatures for some other compounds such as $Li_4Ti_5O_{12}$, $(Y,Gd)BO_3:Eu^{3+}$, Nd-Fe-B, $Nd_{12}Fe_{82}B_6$, $Nd_{14}Fe_{80}B_6$, and $LiFePO_4/C$ [29–34]. Effects of milling time and temperature on hot milling of elemental powders including PbO, ZrO_2, and TiO_2 for PZT synthesis were studied in the present work. Our results demonstrate that high-temperature ball milling method is a simple and effective route for the synthesis of the pure PZT powder with high percentage of crystallinity and particle size close to nanoscale. It was observed that PZT phase is formed in a very short time compared to common room-temperature milling methods.

2. Experimental procedure

At the beginning, high-temperature ball milling system with zirconia media was designed. To synthesize PZT powders, commercially available PbO, ZrO_2 and TiO_2, all of > 99% purity were employed as starting materials. These oxide powders exhibited an average particle size distribution from submicron to tens of microns. The appropriate amounts of the constituent oxides according to the stoichiometric $PbZr_{0.95}Ti_{0.05}O_3$ composition were weighed and stirred for 20 min continuously in acetone at room temperature, and then the obtained slurry was completely dried at 50 °C. In the next step, the obtained powder was milled for 6, 12 and 36 h at room temperature (10 °C), 200 °C, 400 °C, 600 °C, and 800 °C. The milling was carried out within cylindrical Yttria-stabilized zirconia (YSZ) grinding jar (diameter of 40 mm and length of 60 mm) and balls. The hot milling setup designed and used in this work is depicted in Fig. 1. The zirconia grinding jar was affixed to a steel mold (Fig. 1b) and the mold was inserted into an electric furnace for the high temperature experiments (Fig. 1c). An electric motor was used to provide rotational force and the rotational speed could be adjusted by a digital power inverter, as shown in Fig. 1a. The YSZ grinding spheres were consisted of 30 balls with diameter of 5 mm and 3 balls with diameter of 15 mm. Ball-to-powder weight ratio was fixed to be 4:1, the rotation speed was 120 rpm. It is worth mentioning that the media distance from horizontal axis of rotation was designed to be 6 cm so that impact strength of the milling system was improved as a greater moment of inertia was provided. The milling was performed in air atmosphere with heating rate to the desired temperature of 5 °C/min.

The synthesized powders were characterized by X-ray powder diffraction analysis (XRD) and scanning electron microscopy (SEM). The X-ray diffraction studies were carried out using a Philips X Pert-Pro instrument operated at 40 kV and 30 mA with CuKα radiation (λ = 1.5406 Å). The particle size and morphological characteristics of the powders were studied by high magnifications via Field Emission Scanning Electron Microscopy (FE-SEM, Hitachi S-4160 Microscopy). Particle size distribution and average particle size of the synthesized powders were measured through ImageJ software.

In order to evaluate electrical properties of the synthesized PZT powder, green cylindrical ceramic pellets of 1 cm diameter were pressed and formed under 120 MPa uniaxial pressure. Sintering was performed at 1280 °C for 3 h in a sealed alumina crucible under PbO-rich atmosphere to minimize lead loss during high temperature sintering. Heating and cooling rate were set to be 300 °C/h and 180 °C/h, respectively. Both sides of the sintered pellets were ground to remove surface layers and were coated with silver electrodes. Piezoelectric charge coefficient (d_{33}) was measured by a direct method based on a Quasi-static d_{33}-meter and dielectric constant and dielectric loss factor were measured using an LCR meter (Model HP 4284A) at 1 kHz at room temperature.

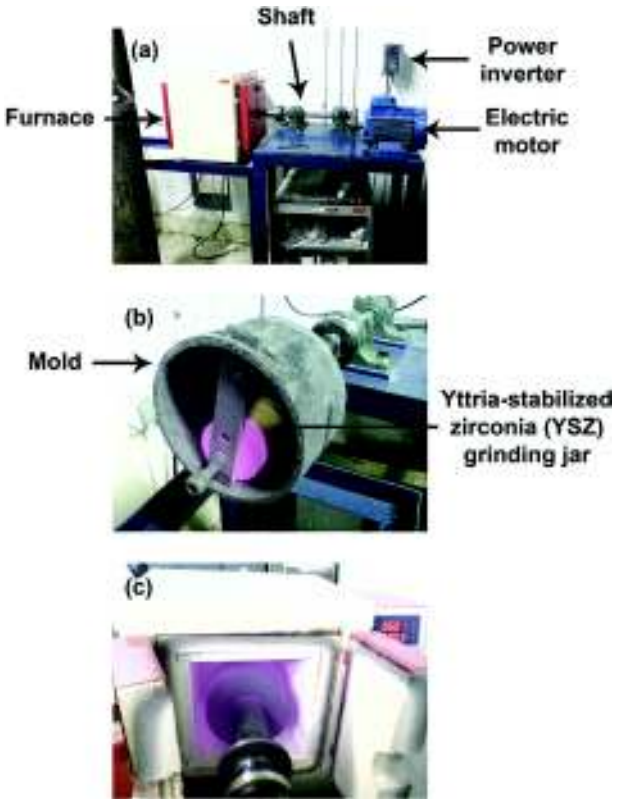

Fig. 1. Pictures of the designed hot milling setup; (a) the whole setup consisted of furnace, electric motor, shaft, digital inverter, etc. (b) the zirconia jar affixed to the steel mold, and (c) the grinding media inserted into the high temperature furnace.

3. Results and discussion

Fig. 2 shows XRD patterns for mixture of PbO, ZrO_2 and TiO_2 commercial powders milled for 6 h at different temperatures ranging from 10 °C to 800 °C. All of the diffraction peaks were successfully assigned to the oxides and compounds as marked in the figure. It is seen that milling at 10 °C did not result in any chemical interaction between the starting materials and all the diffracted peaks exhibited crystalline PbO, ZrO_2, and TiO_2. Besides, the XRD results demonstrate that when milling temperature was increased to 600 °C, the mixture of oxides was still present and no desirable PZT phase was triggered. The XRD pattern of the powder milled at 200 °C is very similar to that recorded from the powders milled at 10 °C. However, it was found that by milling at 400 °C, a new oxide phase of Pb_3O_4 was appeared. The formation of this phase can be attributed to reaction between PbO and O_2, since milling process was carried out in air atmosphere, leading to the formation of Pb_3O_4. It is also worth mentioning that the color of the powder was changed from yellow to orange after milling at 400 °C, which can be attributed to the appearance of Pb_3O_4 product, as it possesses a bright red or orange. The color of powder after milling at 600 °C has gone

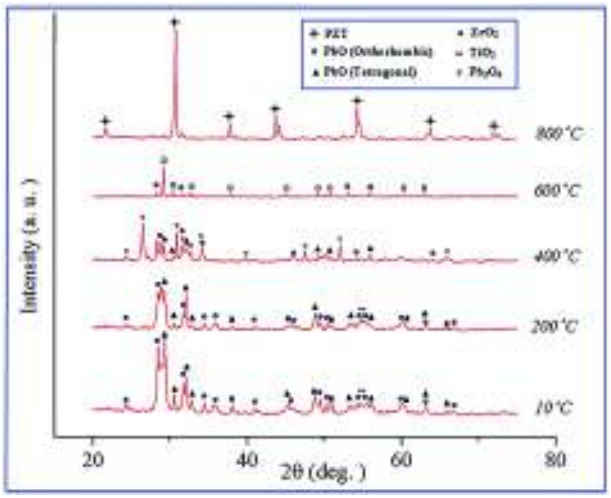

Fig. 2. X-ray diffraction patterns of the obtained powders after hot milling of PbO, ZrO_2 and TiO_2 at temperatures of 10 °C to 800 °C for 6 h.

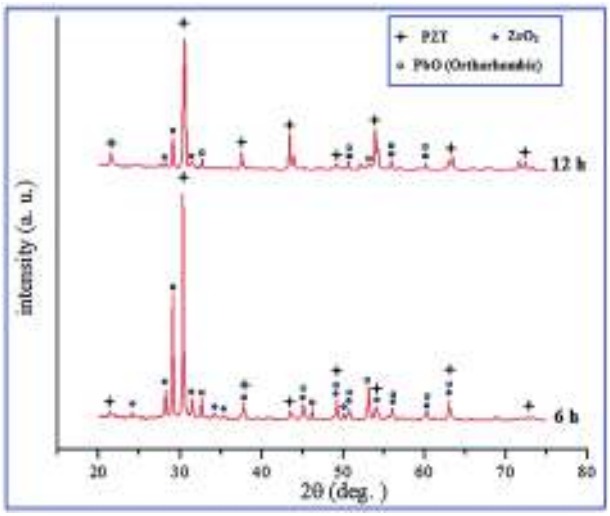

Fig. 3. X-ray diffraction patterns of the synthesized powders after hot milling at temperature of 700 °C for 6 and 12 h.

yellow again because of Pb_3O_4 decomposition and formation of PbO [35]. Nevertheless, it was found from the XRD patterns that crystal structure of the newly formed PbO after milling at 600° °C was orthorhombic, which is different from the initial PbO with tetragonal structure used as starting material. Transformation of $\alpha - PbO$ with tetragonal lattice (a = 3.9729 Å, c = 5.0192 Å) to the high temperature modification of $\beta - PbO$ with orthorhombic (a = 5.489 Å, b = 4.755 Å, c = 5.891 Å) was also reported by other researchers [36]. According to the XRD analysis, single phase PZT powder with full crystallinity was achieved when hot milling was performed at 800° °C. Considering the relevant literature indicates that, in contrast to the present study, relatively longer milling times are needed to synthesize single phase PZT. For example, Lee et al. [37] employed high-energy shaker mill (SPEX 8000) and found that milling times more than 25 h is required to convert the mixture of PbO, ZrO_2, and TiO_2 to perovskite PZT phase. Kong et al. [26] also reported ultra-fine $Pb(Zr_{0.52}Ti_{0.48})O_3$ could be produced using the high-energy ball milling technique in air at room temperature only when 80 h milling was carried out. Similar to these studies, Parashar et al. [38] observed that the reaction of oxides appears to be incomplete even after 35 h of high-energy milling and hence, subsequent post-annealing is required for the completion of the reaction. Table 1 gives synthesis parameters of pure PZT powder prepared with various methods. It is seen that even expensive organic-based processes need calcination at high temperatures of 600–900° °C, and high energy milling route at room temperature also suffers from the main disadvantage of significantly long milling time required to synthesize pure PZT powder. By contrast, pure PZT can be obtained by simple and almost inexpensive route of hot milling for short time of 6 h at 800° °C.

It is clear that the synthesis of pure PZT phase using a low-energy milling system (similar to the system designed in this work) at low temperatures would take even much more time. However, in the present work, a modified milling system was utilized, in which ball milling and heating was used simultaneously to synthesize PZT phase. In this system, the mechanical milling plays an important role of obtaining fine and homogenous particles and on the other hand the thermal energy provided by a controllable heating device promotes reaction between the oxide constituents. Additionally, the mechanical milling generates extra heat by transforming mechanical energy into thermal energy, facilitating the associated reactions. In the following section, it is demonstrated that only simultaneous heating and milling (hot milling) would be effective in the formation of single phase PZT, since milling step and subsequent high-temperature calcination in a furnace did not give rise to the desirable results.

In order to evaluate the possibility of PZT formation at lower temperatures, hot milling was carried out for 6, 12, and 36 h at 700 °C. The X-ray diffraction patterns of the obtained powders are presented in Fig. 3. The patterns clearly show that PZT ceramic powder can be synthesized at lower temperatures, but small amount of unreacted PbO (orthorhombic) and ZrO_2 have been still detected. In addition, Fig. 3 indicates that with increasing milling time from 6 to 12 h, the content of the unreacted powders was declined, as intensities of the diffraction peaks related to PbO and ZrO_2 phases dropped. However, it is obvious that pure PZT could not be synthesized even after 12 h hot milling at 700 °C. Another experiment was performed at 700 °C and prolonged milling time of 36 h; however, as indicated in Fig. 4, the unreacted compounds still existed. Therefore, it can be realized that the effect of milling time is less evident than that of temperature and hot milling at temperatures ≥ 800 °C is required to obtain pure PZT phase.

Fig. 5 indicates the FE-SEM micrographs of the PZT powders hot milled at 800 °C. In addition to the successful synthesis of PZT phase, the SEM micrographs of the PZT powders hot milled at 800 °C for 12, shown in Fig. 5c and d, demonstrate that very fine powder with an average particle size of 170 nm can be obtained by the hot milling technique. Fig. 5e and f depict particle size distribution of the PZT

Table 1
Synthesis conditions of pure PZT powder prepared by hot milling (this work) and some other methods reported in the literature.

Synthesis route	Composition	Temperature (°C)	Time (h)	Ref.
Coprecipitation	$Pb(Zr_{0.52}Ti_{0.48})O_3$	900	2	Rao et al. [8]
Evaporative decomposition	$PbZr_{0.53}Ti_{0.47}O_3 + 0.5$ wt% Nb_2O_5	600	2	Lee et al. [9]
Sol–gel process	(PZT) (52/48)	600	1	Wu et al. [11]
Reactive calcination	$Pb(Zr_{0.52}Ti_{0.48})O_3$	750	4	Shrout et al. [14]
High-energy ball milling	$Pb(Zr_{0.52}Ti_{0.48})O_3$	RT	80	Kong et al. [26]
High-energy shaker mill	$Pb(Zr_{0.52}Ti_{0.48})O_3$	RT	25	Lee et al. [37]
Hot milling	$Pb(Zr_{0.95}Ti_{0.05})O_3$	800	6	This work

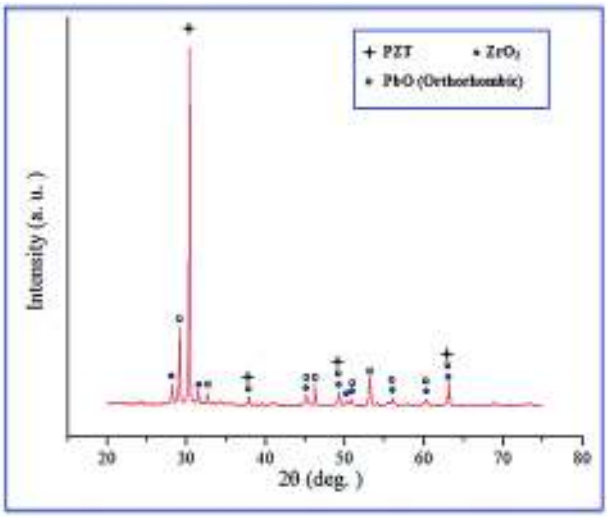

Fig. 4. X-ray diffraction pattern of the obtained powder after hot milling at 700 °C for 36 h.

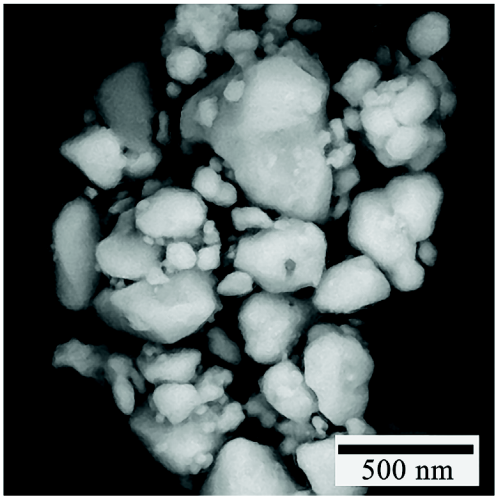

Fig. 6. Backscattered image of the PZT powders hot-milled for 6 h at 800 °C.

powders hot-milled for 6 h and 12 h, respectively. Average particle size was reduced from 410 nm to 170 nm by prolonging milling time. The produced fine PZT powder can be attributed to the decrease of hardness and impact strength of the particles at higher temperatures [39]. Additionally, variation in unit cells volume and a rise in internal stress because of the occurred transition from PbO (tetragonal) to Pb_3O_4 and then, Pb_3O_4 to PbO (orthorhombic), thermal expansion in oxides, and change in unit cells volume after formation of PZT were reported to contribute effectively in the high-temperature particle size reduction [40–42]. Backscattered electron image of the PZT powders prepared at 800 °C for 6 h was given in Fig. 6. It can be seen that there is not any clear image contrast between powder particles, indicating almost pure PZT powder was synthesized by hot milling at 800 °C. This observation is in agreement with the XRD findings shown in Fig. 2. The synthesized PZT powder by hot milling at 800 °C for 6 h was used to prepare dense ceramic pellets through sintering at 1280 °C for 3 h. Physical properties of the sintered PZT pellets were measured and the samples demonstrated piezoelectric charge coefficient (d_{33}) of 36 pC/N, dielectric constant (ε_r) of 264 and dielectric loss (tan δ) of 2%.

To understand impact of simultaneous ball mill and heating (hot milling) on the synthesis of PZT phase, an experiment was carried out in which first the oxide materials (PbO, ZrO_2, and TiO_2) was milled for 6 h at room temperature and then the obtained powder mixture was heated at 800 °C for 6 h. In contrast to the hot milling results, it was found that the separated steps of milling and heating, which is called conventional synthesis method, could not be effective in the complete formation of perovskite PZT.Fig. 7a demonstrates that although the conventional route would fairly result in the synthesis of PZT, a large amount of unreacted oxides, PbO and ZrO_2, are still present in the final product.Fig. 7c and Fig. 7d give the SEM observation of the conventionally synthesized powders. Generally large ceramic particles with average size of 1 μm are seen and are covered by a large number of fine powders as shown in Fig. 7c. A more magnified image clearly indicated morphology of the fine particles (Fig. 7d). Nanoparticles with hexagonal platelet-like morphology and a thickness of about 20 nm and lateral dimensions of 100 nm were clearly observed that were stuck on the surface of the larger particles. Efforts were taken to identify the nature of these nanoparticles. According to our EDS analysis, shown in Fig. 7b, chemical composition of the platelet particles is very close to the composition of PZT ($PbZr_{0.95}Ti_{0.05}O_3$) phase, characterized by $\frac{Pb}{Zr}$~1.

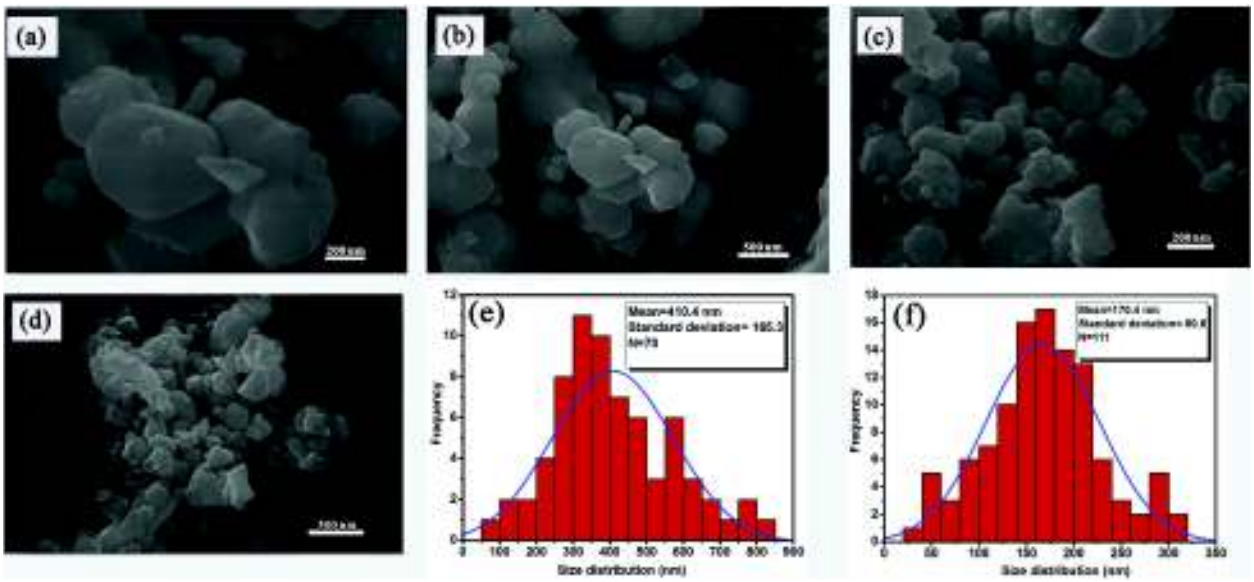

Fig. 5. FE-SEM micrographs of the obtained PZT powder after milling at 800 °C for (a, b) 6 h and (c, d) 12 h at two different magnifications. Particle size distribution of the PZT powders synthesized for (e) 6 h and (f) 12 h.

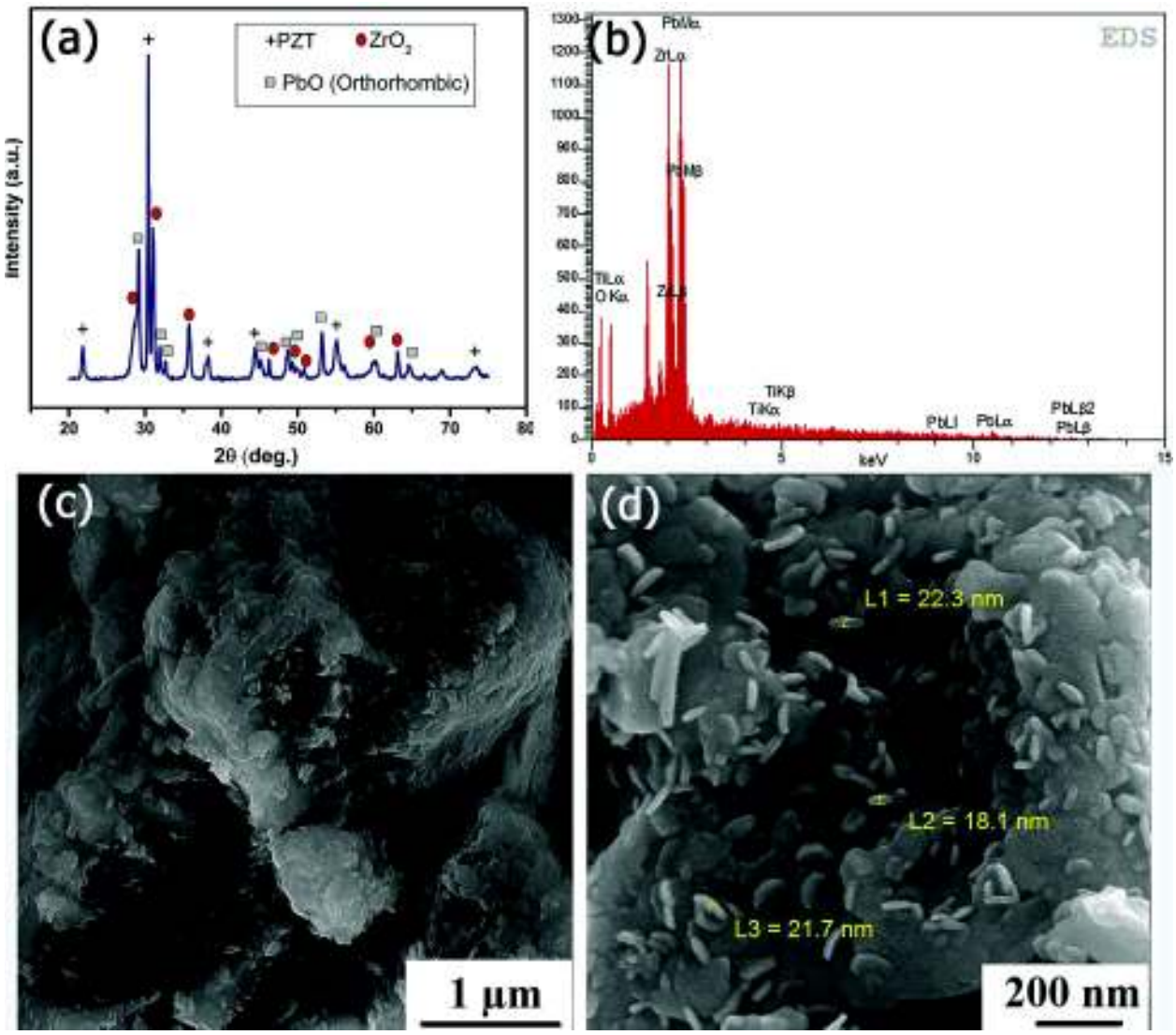

Fig. 7. (a) XRD pattern of the obtained powders by conventional solid-state technique, (b) EDS analysis and (c, d) FESEM micrographs of the synthesized powders.

Therefore, the nanoparticle might be the newly formed PZT phase identified in the XRD results. However, it can be seen from the SEM images that beside the PZT particles, powders with irregular and regular morphologies are also observed that can be attributed to the unreacted raw oxides. Therefore, it is found that the conventional synthesis route is less effective in the synthesis of pure PZT in comparison to the hot milling technique. The enhanced performance of the hot milling might be explained on the ground of the increased number of "fresh contact points" in this method. During the hot milling the fresh contact points between PbO, ZrO_2, and TiO_2 components are continuously created and at the same time chemical reactions of PZT formation are occurred at these points. By contrast, in the conventional route, once milling is done at the first step, there are not any rearrangement to the oxide particles during the post heat treatment step and hence no fresh contact points would be established, which in turn would lengthen diffusion route for the formation of PZT product.

4. Conclusion

High-temperature ball milling is a simple and effective technique to synthesize very fine and crystalline PZT powder. Hot milling of PbO, ZrO_2, and TiO_2 raw materials at 800 °C for 6 h can successfully convert all the oxide materials to PZT product, which in contrast to the conventional synthesis route, i.e. milling for 6 h and then post heating at 800 °C, demonstrated much more efficiency in the synthesis of pure PZT phase. The average particle size of the obtained PZT powder after 12 h milling at 800 °C was measured to be very close to nanoscale (~170 nm). The hot milling method has several advantages over the common milling methods such as simplicity in production as calcination process is not required. Additionally, uniform and fine powder can be obtained because of milling at high temperatures. Moreover, according to the designed milling system (a low energy type), the results demonstrate that hot milling system, which is relatively cheap and simple in use, can be an attractive candidate for the industrial production of various ceramic powder materials.

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