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Synthesis, Structural and Ferroelectric Properties of Lead Titanate ($PbTiO_3$) Nanoparticles by Sol-Gel Auto Combustion Method

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Abstract: $PbTiO_3$ nanoparticles were prepared at low temperatures using the sol-gel auto combustion method. XRD analysis confirmed the formation of single phase tetragonal structure without any impurity phases. Using XRD data we have calculated lattice constant (a and c), unit cell volume (V), X-ray density (ρ_x), bulk density (ρ_B), porosity (P%) and average crystallite size (t) was calculated. The average crystallite size (D) was found to be ~42 nm. The polarization versus electric field (P-E) loops showed a well saturated hysteresis loop at room temperature confirming the ferroelectric nature of the prepared sample.

Keywords: Nanoparticles, sol-gel, $PbTiO_3$, ferroelectric.

I. INTRODUCTION

Ferroelectric materials have been applied to a large field of applications because of their excellent dielectric, piezoelectric and ferroelectric properties. $BaTiO_3$ was the first perovskite type ferroelectric material developed and intensively studied ever since its discovery about 70 years ago [1, 2]. This fact is due to its high dielectric constant and low dielectric loss, good piezoelectric, pyroelectric, and ferroelectric properties, positive temperature coefficient [3].

The lead titanium oxide ($BaTiO_3$) is a ferroelectric material with perovskite-type structure, with a simple cubic structure (ABO_3) which consists of a small cation B (Ti^{4+} ions) in the center of an octahedron of oxygen anions, and a large cation A (Pb^{2+} ions) in the corners of the unit cell. $PbTiO_3$ has found extensive use in the ceramic capacitor industry [4].

Conventionally, lead titanate has been produced by conventional solid state reaction techniques. However, in recent years there has been a trend towards the use of wet chemical methods [5].

In the present study, lead titanate ceramics of the compositional formula $PbTiO_3$were synthesized using sol-gel auto combustion method. An extensive literature survey shows that ball milling, solid state methods have been used to synthesize several ferroelectric materials [6]. However, very few reports are available on the wet chemical synthesis of $PbTiO_3$. This encouraged us to synthesize the pure $PbTiO_3$ using sol-gel. The structural, microstructural and ferroelectric properties of $PbTiO_3$ were investigated.

II. EXPERIMENTAL

Analytical grade lead nitrate hexahydrate ($Pb(NO_3)_2{\cdot}6H2O$), tetra butyl titanate ($Ti(OC_4H_9)_4$), citric acid ($C_6H_8O_7$), ethanol (C_2H_5-OH) and ammonium hydroxide (NH_4OH) provided by Merck with ~99 % purity were used as a starting materials without further purification for the synthesis. Firstly, tetra butyl titanate solution diluted with ethanol was added into the citric acid aqueous solution with pH = 8 which is adjusted by adding the appropriate amount of ammonia. Ethanol was used to chelate tetra butyl titanate.

A yellowish transparent liquid was obtained which is marked as solution 'A' after being stirred at 80 °C for 1 h. At the same time, barium nitrate were dissolved into distilled water, accompanying continuous stirring until all salts were absolutely solved which is marked as solution 'B′. Subsequently, solutions 'A' and 'B′ were poured together. Followed by a continuous stirring for 3 h, the viscosity of solution increased gradually and then a stable transparent sol formed. Uninterrupted heating of 100 °C initiates the gel formation. Under constant stirring and heating, viscous gel transforms into dry gel. The dried gel formed from metal nitrates and citric acid exhibited self-propagating combustion behavior. The obtained powders dried, crushed and were annealed at 900 °C for 5 h in a muffle furnace in order to get the nanocrystalline powders.

Characterizations

The X-ray diffraction (XRD) pattern of the prepared sample was taken by using Phillips X-ray diffractometer (Model PW-1710) using Cu-Kα radiation (λ = 1.5418 Ǻ). The surface morphological studies were carried out using scanning electron microscopy using JEOL JSM-6360

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scanning electron micrograph (SEM). The P-E hysteresis loop was measured under an alternative electric field using a ferroelectric test system (RT6000HVS, Radiant Technology Incorporation) at 50 Hz.

III. RESULTS AND DISCUSSIONS

Fig.1 shows the X-ray diffraction pattern of $PbTiO_3$ ceramics. It exhibits all the characteristic peaks of perovskite structured material without any impurity peak and the most intense peak was observed at (110). The other planes observed are (100), (110), (111), (200), (210), (211), (220), (221), (300) and (310).

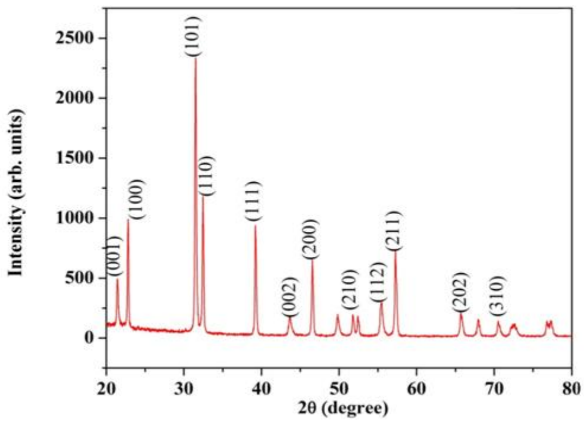

Fig.1 XRD pattern of $PbTiO_3$

The XRD pattern confirms the formation of perovskite tetragonal structure without any impurity peaks. The pattern shows well defined peaks and there is no any intermediate phase is formed, confirming the single phase formation of PbTiO3 sample. The lattice parameters are found to be a = b=3.8405 Å and c = 4.0496 Å with c/a = 1.0544 for pure lead titanate nanopowders which are in good agreement with the reported values [7]. This suggests that the crystal structure is tetragonal at room temperature.
The average crystallite size was calculated by using Scherrer formula and is found to be 42 nm. The unit cell volume (V), bulk density (ρm) and porosity (P) values were also calculated for the present sample and their values are tabulated in Table 1.

TABLE I Lattice constant (a and c), c/a ratio, unit cell volume (V), average crystallite size (D), X-ray density (ρx), bulk density (ρm) and porosity (P) of PbTiO3 nanoparticles

Parameters	Values
a (Å)	3.8405
c (Å)	4.0496
c/a	1.0544
V(Å)3	59.7272
D (nm)	42.28
ρx (g cm-3)	8.4250
ρm (g cm-3)	6.5572
P (%)	22.17

The scanning electron micrograph (SEM) of the lead titanate is shown in Fig. 2. It is observed from SEM micrograph that the lead titanate particles obtained by sol-gel auto combustion method used in the present investigation are mostly uniform in morphology with having agglomeration to some extent. The grain size obtained from SEM micrograph by using the linear intercept method is of the order of 210 nm.

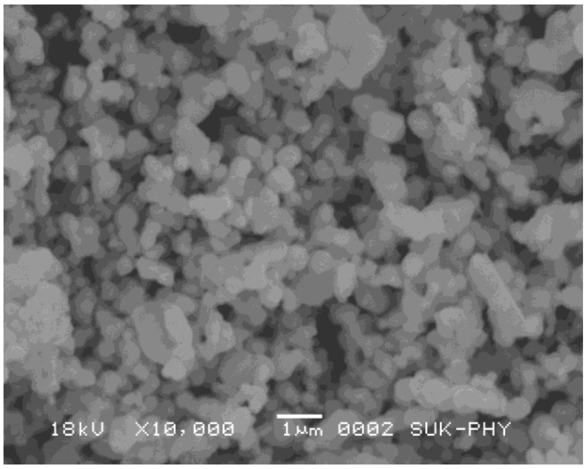

Fig. 2 SEM image of $PbTiO_3$

It is known fact that composition, microstructure, crystalline phase, and lattice defects like oxygen vacancies can significantly affect the ferroelectric properties of ceramics [4]. The strength of the ferroelectric phase can also be determined by the structural transition with additional aspects like the incorporation of foreign species. It is obvious that the perseverance of ferroelectricity results from the long-range polar orders of dipoles; and any disruption in the polar order would affect the ferroelectricity [8].
Fig. 3 shows the P–E hysteresis loop at room temperature of $PbTiO_3$ nanoceramics. Well saturated hysteresis shape typical of ferroelectric materials was evident for the prepared nanoceramics. . The values of saturation polarization (Ps), remanent polarization (Pr) and coercive field (Ec), determined from the hysteresis loop.

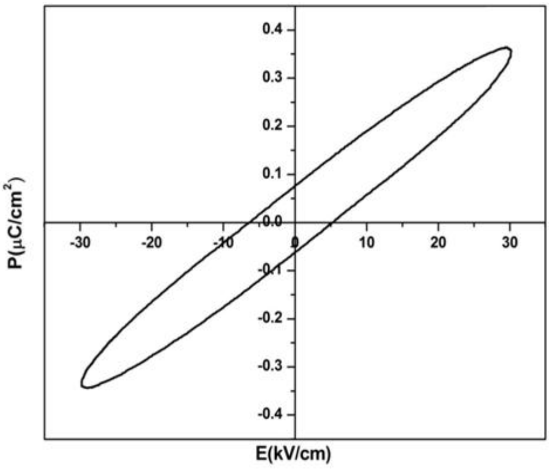

Fig.3 P-E loop for $PbTiO_3$ nanoparticles

IV. CONCLUSION

$PbTiO_3$ nanoparticles wear successfully synthesized by sol-gel auto combustion method. XRD pattern confirmed the formation of tetragonal perovskite structure. Using XRD data, unit cell volume (V), X-ray density (ρ_x), bulk density (ρ_B) and porosity (P%) values were calculated. Average crystallite obtained by Debye-Scherrer's formula shows the nanocrystalline nature of the prepared sample. The lattice constants were found to be in reported range. Well saturated P-E hysteresis shape curve typical of ferroelectric materials was evident for the prepared nanoparticles.

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