PEROVSKITE MICROSTRUCTURE STUDIED OF BARIUM TITANATE - POLYVINYL ALCOHOL WERE SINTERED AND SYNTHESIZED BY SOL-GEL

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Nanoparticle barium titanate powder has been made by sol-gel preparation and by sintered in several temperatures. Polyvinyl alcohol (PVA) as a filler had uses for made some samples of barium titanate-PVA in its various with differences of temperature at 700[°]C, 800[°]C, and 900[°]C. It was found that structural were appropriated with tetragonal and occurs for all samples. Optimally occurs at 800[°]C and tends to binding of atoms between Ba, Ti, and O, in the perovskite form of BaTiO₃. Their grain sizes, volume fractions, densities, and dielectric constants has been studied. It was both effectively indicates of PVA and temperature affected to form its appropriate microstructure of barium titanate perovskite.

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1. Introduction

Perovskite lattice of barium titanate is compound with formula BaTiO₃, where their atom Ba and Ti are two cations in different size, a atom O is an anion that bonds formed [1]. The stable form of barium titanate into the perovskite structure is greatly determined for its uses [2], particularly as a raw material ferroelectric or piezoceramic. There is example for another perovskite form of lead zirconium titanate (PZT), where it was uses to electronic devices [3-4]. However, a PZT is not good for environment and also tends to toxic for living organism, due to had a atom Pb in their compound. Meanwhile, a compound of $BaTiO_3$ is better for living organism and environment. Therefore, in this work is concerned study to nanoparticle barium titanate with polyvinyl alcohol added to enhances the perovskite lattice throughout in the tetragonal structure formed by of sol-gel preparation and sintered too. Sol-gel preparation were preffered to synthesis their compound in the form of BaTiO₃ from its sources in this works due to that product always in nanoparticle size. Subsequently, its temperature of sinter were preffered to tetragonal phase in their structural could be optimizing becames a perovskite lattice of barium titanate [5-7]. By means of both were effectively optimize their microstructure as well as a crystallite grain size, number of volume fraction, number of density, and dielectric constants [8]. In the other words, these ways effectively controlled to enhance that atoms binding were optimum.

2. Experimental

Pro Analysis of materials with 99.0% of the purity was uses in this work. Those materials such as a barium carbonate powder, a titanium dioxide powder, an ammonium hydroxide, nitric acid, polyvinyl alcohol, and aqua bidestilate. By sol-gel preparation, it was results a few of samples in gel formation with different contents of PVA. Subsequently by sinter in various of

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temperature, eaxctly for 700°C, 800°C, and 900°C had been carried out to obtained dry powder sample. The phase formation and number of volume fraction was traced by X-ray diffraction tools, with a type PHILIPS-binary (scan.RD) and by *Match Program!* for quantitative refinement analysis [9-10]. The distribution of crystallite grain size was traced by particle size analysis (PSA-DLS-size distribution by intesity with dry dispersion type). Density of sample was calculated by wet bulk density methods [11]. These methods was conducted to appropriate technique for bulk density material like a nanocomposite BaTiO₃-PVA. Finally, that ferroelectrical characteristics was tested by electrical voltage response uses a analog oscilloscope and by measurement that capacitance to frequency versus dielectrical constants count uses a LRC digital meters.

3. Results and discussion

3.1. Crysttalite phase analysis by XRD results

Fig. 1. has been shown the XRD patterns for every samples of each PVA contents like in the notation of BI0PVA, BI1PVA, BI2PVA, and conducted by various temperature of sinter from 700° C to 900° C. Their crystallite phase structure instead of perovskite lattice or tetragonal phase structure were indicated by calculating that is crystallite lattice. In which it was shown that peak to peak of the patterns is crystallite phase structure for perovskite lattice and were dominantly occurs. Therefore, it was indicates to effects of PVA as a filler in the sol-gel preparation could be made their atoms Ba, Ti, O or by their ions of Ba²⁺ and TiO₃²⁻ tends to tightly binding. In the other words, this attempts indicates was successfully made for a barium titanate in the perovskite lattice most dominant by means of sol-gel preparation and by sintered.



Fig. 1. XRD patterns of : BIOPVA (a) at $700^{\circ}C$, (b) at $800^{\circ}C$, (c) at $900^{\circ}C$; BI1PVA (d) at $700^{\circ}C$, (e) at $800^{\circ}C$, (f) at $900^{\circ}C$; BI2PVA (g) at $700^{\circ}C$, (h) at $800^{\circ}C$, (i) at $900^{\circ}C$.

Fig. 2 was shown the peak shifting of XRD patterns in interval angle 2theta = 30° up to 2theta = 35° and were indicated to effects of temperature enhance for fixed three samples BI0PVA, BI1PVA, and BI2PVA in the curves (a), (b), (c), (g), (h), (i), (m), (n), (o), and by PVA enhanced for fixed temperature of sinter at 700° C, 800° C, and 900° C in the curves (d), (e), (f), (j), (k), (l), (p), (q), (r).



Fig. 2. Peak shifting of XRD patterns were reflected by third different samples, (a), (b),and (c) for BIOPVA; (g),(h), and (i) for BI1PVA; (m), (n), and (o) for BI2PVA; (d), (e), and (f) for 700^oC; (j), (k), and (l) for 800^oC; (p), (q), and (r) for 900^oC

Certainly, perovskite lattice (110) it was found by calculation at 2theta = 31.69° and for lattice (101) at 2theta = 31.77° . These lattices had been calculated by appropriate formula of tetragonal phases [12-13], insted of perovskite lattice of barium titanate was found in dominantly formed. It was indicated strongly peak shifting occurs by effects of sintered and by effects of PVA.

3.2. Particle size analysis by PSA distributions

Particle size distributions for all samples was tracked by PSA and then obtained to its distributions less than 100.0 nm with almost uniform. The best results was found for samples B11PVA and B12PVA, it shown in the Figure 3. Where it was really sintered into the temperatures 700°C, 800°C, and 900°C. In the other words, the quantities of particle size have been incressed or decreased its number of particle size less than 100.0 nm when they were conducted by sintered and synthesized by sol-gel. Even that, it was really effectively for PVA enhanced if that was compared by a sample without of PVA such as in the BI0PVA.



Fig. 3. Particle size distributions of third samples BIOPVA, BI1PVA, and BI2PVA was tracked by PSA in differences of sintered temperature; (a), (b), and (c) for 700^oC; (d), (e), and (f) for 800^oC; (g), (h), and (i) for 900^oC.

3.3 Volume fraction of the crystallite lattice

The volume fraction of crystallite lattice for all contents was shown in the Fig. 4., where a volume fraction of tetragonal of $BaTiO_3$ is frequently appears if that compared by others contents like $BaCO_3$ and TiO_2 . As well as in the Fig 4(b), 4(e), and 4(h), particularly for all samples when sintered at 800^oC seems dominant or optimum occurred in about hundreds percent for tetragonal of $BaTiO_3$ with measly other content like $BaCO_3$ in about 8.0%. Instead of the perovskite lattice of $BaTiO_3$ is successfully synthesized and by sinter at 800^oC for all samples tends to be stable microstructure formed by much more quantities.



Fig. 4. Volume fraction of sample; (a) BIOPVA, (b) BI1PVA, and (c) BI2PVA for 700^oC; (d) BI0PVA, (e) BI1PVA, and (f) BI2PVA for 800^oC; (g) BI0PVA, (h) BI1PVA, and (i) BI2PVA for 900^oC.

3.4. Number of density and constant dielectrics of the sample

The number of density were calculated by wet bulk method for all samples and was shown in the Fig. 5(a), 5(b), and 5(c). Its results was closely with the number of density of barium titanate pure in about 6.02 g/cm³ [14]. Therefore, this quantizations was indicates for all samples is a porous materials.

Meanwhile, Fig 5(d), 5(e), and 5(f) has been shown the characteristics of dielecctric constant for all samples as a function of frequency. When it was measured by frequencies from 100 Hz to 100 kHz at room temperature.



Fig. 5. Density distribution of all samples; (a) BIOPVA, (b) BI1PVA, and (c) BI2PVA. The characteristics of dielectric constant of all samples; (f) BIOPVA, (e) BI1PVA, and (d)BI2PVA

The results was exponentially and decreased by the increased of the frequencies such as their purely characteristics of barium titanate. However this result is smaller than dielectric constant of purely of barium titanate in near curie temperature [15]. In the other words, their effects of temperature and polyvinyl alcohol was effectively to increased its dielectric constant.

4. Conclusion

The powders of barium titanate in its quantities of nanoparticle size have been obtained by sol-gel preparation and sintered. This works was shown the perovskite microstructure of barium titanate is proper affected by effects of temperature and polyvinyl alcohol, throughout this studied by their crystallite lattice, volume fraction of crystallite lattice, particle size distributions, number of density, and dielectric constant.

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References

- [1] G. H. Jonker, E. E, Havinga, Mat. Res. Bull., **17**, 346 (1982).
- [2] U. Al-Amani Azlan, A Fauzi Mohd Noor., Sci. Sinter., 49, 108 (2017).
- [3] A. Kareira, S. Tautkvs, R. Rapalaviciute., Journal of Material Science., 34, 4853 (1999).
- [4] M. C. Cheung, H. L. W. Chan, C. L. Choy., Nanostruct Matter., 11, 837 (2001).
- [5] M. Reza, C. Hu, W. H. Shepherd., IEEE Transactions on Electron Devices. 39, 2044 (1992).
- [6] Suastiyanti, Dwita., Advanced Materials Research., 789, 118 (2014).
- [7] Irinela Chilibon, Jose N. Marat-Mendes., J. Sol-Gel Sci Technol., 64, 571 (2012),
- [8] M. Cernea., Journal of Optoelectronics and Advanced Materials., 7, 3015 (2005).
- [9] V. V. Zyryanov, V.A. Sadykov, M.I. Ivanovskaya, J.M. Criado, S. Neophytides., Sci. Sinter., 37, 45 (2005).
- [10] J. F. Scott, A. Schilling, S. E. Rowley, J. M. Gregg., Sci. Technol Adv Mater., 3, 16 (2015).
- [11] J. Rouquerol., et al., Pure Appl. Chem., 84, 107 (2012).
- [12] A. S. Bhalla, R. Guo, R. Roy., Mat. Res. Innovat., 4, 3 (2000).
- [13] M. T. Buscaglia, V. Buscaglia, M. Viviani, P. Nanni, M. Hanuskova., Journal of the European Ceramic Society., 20, 1997 (2000).
- [14] Ma, Chi; Rossman, R. Geoge., American Mineralogist., 93, 154 (2008).
- [15] Ken-ichi Sakayori, Y. Matsui, H. Abe., et al., Japanese Jounal of Applied Physics., 34, 9 (1995).