Electric polarization properties of BaTiO$_3$-BiFeO$_3$ as nanomultiferroic material produced by sol-gel method

Dwita Suastiyanti$^{1,*}$, Maykel T.E. Manawan$^2$ and Marlin Wijaya$^3$

$^1$Mechanical Engineering Study Program, Institut Teknologi Indonesia, Puspiptek-Rayastreet, 15320 Serpong, Indonesia
$^2$Politeknik Negeri Jakarta, 16424 Depok, West Java, Indonesia
$^3$Badan Pengkajian dan Penerapan Teknologi (BPPT), 10340 Serpong, Indonesia

Abstract. The nanomultiferroic material which is synthesized in this research used sol-gel method. The research used weight ratio of BaTiO$_3$:BiFeO$_3$ of 2:1. Gel formed after heating at 80-90°C was calcined at 350°C for 4 hours and then sintered at 700, 750 and 800°C for 2, 4 and 6 hours respectively. Powder produced after sintering was characterized by X-Ray Diffraction (XRD) test using XRD Phillips PW 1835 type, 20°-100° diffraction angle and CuKα, electric polarization properties test and particle size measurement using Particle Size Analyzer of Beckman Coulter DelsaTM Nano instrument. From the characterization results, it is obtained that the dominant phase is Barium Bismuth Iron (III) Oxide (BaBiFe$_2$O$_5$). Electrical polarization properties such as remanent, coercivity and saturation reach maximum value at sinter temperature of 750°C and sinter time of 6 hours. This result is supported by the smallest particle size of powder (54-57 nm) and also supported by the largest number of dominant phase (98.79%) at same condition.

1 Introduction

The nanomultiferroic material is a material with nano particle size (<100 nm) having 2 or more physical properties within the material. If the material is given external effects in the form of magnetic field then the material will show a response in the form of electrical voltage values. Similarly if material is given external effects of electrical field then the material will show a response of magnetic properties. There will occasionally arise a mechanical response in the form of a strain effect. Barium titanate (BaTiO$_3$ / BTO) is a very popular ferroelectric material and is a good example to explain the nature of ferroelectricity and piezoelectricity in perovskite structures. Barium titanate is the first developed piezoelectric ceramics and even today it is still widely used. This popular material is also used as a capacitor. The crystallographic dimension of the BTO lattice changes with

* Corresponding author: dwita.suastiyanti@iti.ac.id
temperature changes. Given these distortions there will be enormous spontaneous polarization that can provide an increase in the dielectric constant and temperature dependence on the dielectric constant. The perfect perovskite structure has a general formula ABO₃ which shows the presence of divalent and trivalent cations and B is a tetravalent or trivalent cation. The A ions occupy the corners of the cube while the B ions are in a concentration position of space in the octahedron oxygen, which is the side-centered position. The B ions can move relatively freely inside oxygen octahedrons with a relatively small force.

The multiferroic material that has been successfully synthesized and shows better value of the MagnetoElectric (ME) coupling constant is based on ferrite material [1]. The material shows a large voltage value of 130 volts when given the effect of external magnetic field only 150 Gauss. The constant of ME is a constant showing the multiferroic properties of a material. The ferrite-based material mentioned is BiFeO₃ (bismuth ferrite / BFO). Other researchers have already done a lot of research on multiferroic BFO materials either as a single compound [2,3] and as a substitution compound [4,5,6,7]. From the results of the study, it was found that single-compound multiferroic BFO materials had better physical properties compared to the doped multiferroic of BFO compounds. This research has the objective of obtaining nanomultiferroic materials with good electric polarization properties with the addition of ferroelectric materials (BTO) into multiferroic materials (BFO). Good electric polarization properties could increase the opposite response when given external effects such as magnetic fields, electric fields or stresses / strain [8]. It means could improve multiferroic properties (increase ME coupling constant).

Applications of this material could be used such as ultimate memory devices for the electronics industry and machine tools (due to have high hardness of the material) that could be fabricated, among others, by the process of Taguchi and Fuzzy Logic Taguchi wire processes [9].

2 Methods

This research uses sol-gel synthesis method which is a method that could produce nano-sized powder, homogeneous, no agglomeration and requires low process temperature. The basic compound used is of pro analysis Merck product with a purity of 99.99% such as Bi₅O(OH)₉(NO₃)₄, Fe(NO₃)₃, 9H₂O, HNO₃, H₂O, Zn(NO₃)₂, Ba(NO₃)₂, TiO₂ and citric acid C₆H₈O₇ as fuel. Flow diagram of the research is shown in Figure 1.

![Flowchart of nanomultiferroic powder synthesis.](https://example.com/flowchart.png)

Fig. 1. Flowchart of nanomultiferroic powder synthesis.
Figure 1 shows flow diagram of nanomultiferroic powder synthesized which used low calcination temperature of 350°C and low sinter temperatures of 700-800°C (< 1000°C). The variation of parameter used is sintering time of 2, 4 and 6 hours. It will produces powder with nanosize particle evidenced by test of particle size analyzer. The other characterizations are X-Ray Diffraction (XRD) test using XRD Phillips PW 1835 type, 20°-100° diffraction angle and CuKα to know types of phases formed, electrical test to know electric polarization properties of remanent, coersivity and saturation.

3 Results and Discussion

3.1. Result of XRD Test

From XRD test results, there are obtained graphs of XRD patterns for all samples, one example shown in Figure 2 for sinter temperature of 750°C and sinter time of 6 hours.

![XRD pattern of powder with sintering temperature of 750°C for 6 hours sintering.](image)

Figure 2 shows that powder has some peaks at certain positions belonged to BaBiFe_2O_5 phase. After refining the pattern by HighScrePlus system, it obtains percentage of dominant phase of BaBiFe_2O_5 shown in Table 1.

| Number | Sintering Treatment | BaBiFe_2O_5 (%) |
|--------|---------------------|----------------|
| 1      | 700°C, 2 hours      | 88.50          |
| 2      | 700°C, 4 hours      | 91.49          |
| 3      | 700°C, 6 hours      | 94.44          |
| 4      | 750°C, 2 hours      | 95.98          |
| 5      | 750°C, 4 hours      | 97.37          |
| 6      | 750°C, 6 hours      | 98.79          |
| 7      | 800°C, 2 hours      | 95.07          |
| 8      | 800°C, 4 hours      | 93.12          |
| 9      | 800°C, 6 hours      | 90.55          |

Table 1 shows that it produces dominant phase of BaBiFe_2O_5 where as the largest number of as much as 98.79% at sinter temperature of 750°C and sinter time of 6 hours. At the temperature of 700°C there is an increase in the number of dominant phases with increasing
sinter time and at temperature of 800°C there is a decrease in the number of dominant phases with increasing sinter time. This is because at higher sinter temperatures, the oxide phases occur due to the reaction of Ba, Bi, and Fe elements with oxygen still present in the powder formed.

### 3.2 Electric Test

From the result of electric test, it is obtained graphs of sinter time vs electric polarization properties such as remanent, coersivity and saturation shown in Figure 3,4 and 5.

**Fig. 3.** Graph of electric properties (remanent) for all samples.

**Fig. 4.** Graph of electric properties (coersivity) for all samples.
Figures 3, 4 and 5 show that remanent, coercivity and polarization values increase with increasing sinter time for all sinter temperatures. The greatest value is owned by sintered samples at 750°C for 6 hours. While at highest sinter temperatures of 800°C, the value of electric polarization will decrease again, this is because at highest temperature, it occurs reaction among metal elements in nanomultiferroic compounds with oxygen gas so it will form impurities phases which are not expected. While at lower temperature (700°C) the amount of impurities phases is still more than of 750°C, so it causes value of electric polarization also lower than of 750°C [10]. This is because at that temperature, it is not enough heat is given to form a single dominant phase (BaBiFe₂O₅). The presence of impurities phases cause dipoles of electric charge is undirectional which could decrease electric polarization properties of the sample.

3.3 Particle Size Analyzer

From results of particle size analyzer, it is obtained graph of sinter time vs particle size (nm) shown in Figure 6.
Figure 6 shows that the powder produced from the research is a nanoparticle because it shows the particle size <100 nm. The longer of sintering time there will be an increase in particle size because the particles will grow with increasing sintering time. The smallest particles (54-57 nm) belongs to powder sintered at 750°C for all sintering time.

4 Conclusion

Powder synthesized by so-gel method for all variables (sintering temperatures of 700, 750 and 800°C for 2, 4 and 6 hours respectively) is nanoparticle because the size < 100 nm. So it could be claimed that powder produced by the research is as nanoparticle powder where as the smallest particle belongs to powder sintered at 750°C for 6 hours. The synthesized produces powder with dominant phase of BaBiFe₂O₅ (Barium Bismuth Iron (III) Oxides) more than 85% and the most amount of BaBiFe₂O₅ phase belongs to powder sintered at 750°C for 6 hours. The Values of electric polarization properties such as remanent, coersivity and saturation increase with longer time of sintering for all sintering temperatures. The highest values of electric polarization properties belong to powder sintered at 750°C for 6 hours as linear with the most amount of BaBiFe₂O₅ phase (98.79%) at the same parameter. So there is a relation among the amount of dominant phase and particle size with electrical polarization properties.

This research is supported financially by Grant of Research based on Competency 2018 through letter of number decree of 044/KM/PNT/2018, March 6th, 2018, Ministry for Research and Technology DIKTI, The Republic of Indonesia and letter of number decree of 32/KP/LPKT-ITI/III/2018, March 29th, 2018, Institut Teknologi Indonesia, the Republic of Indonesia.

References

1. D. Suastiyanti, M. Wijaya, *ARPN Journal of Engineering and Applied Sciences*, **11** (2016)
2. D. Suastiyanti, Ismojo, *IOP Conference Series : Material Science and Engineering*, **214** (2017)
3. D. Suastiyanti, Ismojo, *The International Journal of Engineering and Sciences (IJES)*, **5** (2016)
4. D. Suastiyanti, Ismojo, *ARPN Journal of Engineering and Applied Sciences*, **12** (2017)
5. V. James, P.P. Rao, S. Sameera, S. Divya, *Ceram Int.,* **40** (2014)
6. A. Chaudhuri, K. Mandal. J. Magn. Magn, Mater. **353** (2014)
7. D. Suastiyanti, Ismojo, M. Wijaya, *International Journal of Advanced Research (IJAR)*, **5** (2017)
8. Y. Cai, S. Luo, Z. Zhu, H. Gu, *The Journal of Chemical Physics*, **139** (2013)
9. P. Rupajati, B.O.P. Soepangkat, B. Pramujati, H.C.K. Agustin, *Applied Mechanics and Materials, Trans Tech Publication*, **493** (2014)
10. L. Michael, W. Gerald, L. Vera, S. Peter, M. Hiwa, J.V.B. Margriet, V. Andre, T. Kristiaan, O. Oliver, G. Marius, *Applied Physics Letters*, **106** (2015)