Improving Magnetic Properties of $BiFeO_3$-$BaFe_{12}O_{19}$ Solid Solution by Different Sintering Time and Temperatures of Sol-Gel Method

Dwita Suastiyanti[1,*], Yuli Nurul Maulida[2] and Marlin Wijaya[3]

[1] Mechanical Engineering Study Program-Institut Teknologi Indonesia
Puspiptek Raya Street, Serpong, Tangerang Selatan, Banten, Indonesia

[2] Chemical Engineering Study Program-Institut Teknologi Indonesia
Puspiptek Raya Street, Serpong, Tangerang Selatan, Banten, Indonesia

[3]Badan Pengkajian dan Penerapan Teknologi, Puspiptek - Indonesia
Puspiptek Raya Street, Serpong, Tangerang Selatan, Banten, Indonesia

[*]*Corresponding author's email: dwita_suastiyanti [AT] iti.ac.id*

ABSTRACT— ***Synthesis of $BiFeO_3$-$BaFe_{12}O_{19}$ solid solution is aimed to enhanced magnetic properties of the material which can improve the quality of multiferroic properties of material. As we know that $BiFeO_3$ is a multiferroic material if it is in single phase but unfortunately it is difficult to produce $BiFeO_3$ in single phase, which can cause a large current leakage arising from non stoichiometric. It used sol gel method to produce $BiFeO_3$-$BaFe_{12}O_{19}$ solid solution with weight ratio of 1;1. To know magnetic properties, it was used permagraph test which is type of MPS magnet – Physic EP3 – Permagraph L . The sintering temperature was 750, 800 and 850^oC for 8, 10 and 12 hours respectively. There is no regularity in increasing and decreasing of remanent and coersivity properties with increasing sinter temperatures and time of sintering but there is an increasing magnetic energy with increasing sinter temperatures and time of sintering. The highest value of magnetic energy, 10.716 GkA/m belongs to powder sintered at 850^oC for 12 hours.***

Keywords— magnetic properties, magnetic energy, sol-gel method

---

1. INTRODUCTION

Multiferroic materials is an attractive material due to their potential application in electronic device (i.e. information storage, packaging materials, sensors, ultimate memory devices etc) [1]. The material will give electric voltage response if given an external magnetic field or will give magnetic response if given an external electric field or will give a plasticity response if given an external magnetic or electric field. Bismuth ferrite ($BiFeO_3$) is a potential multiferroic material with ferroelectric and ferromagnetic properties at room temperature if it is in single phase. Unfortunately it is difficult to produce $BiFeO_3$ in single phase, which can cause a large current leakage arising from non stoichiometric. $BiFe0_3$ has high Curie (Tc~830 °C) and Neel temperatures (TN~367 0c), has G type antiferromagnetic spiral spin perovskite structure (space group R3c) with aperiod of 62 nm [2,3]. The other problems of $BiFe0_3$ are dielectric loss, low dielectric constant and weak magnetic properties. H.Y. Dai et al. reported effect of $BaTi0_3$ doping on the structural, electrical and magnetic properties of $BiFe0_3$ ceramlcs [4]. Ren et al. reported comparative study of Mn^{3+} and Mn^{2+} doping effects on structure and electrical properties of $BiFe0_3$ thin film [5]. Liu et al. reported dielectric Characteristics in $BiFe0_3$-Modified $SrTi0_3$ incipient ferroelectric ceramics [6]. $BaFe_{12}0_{19}$ is a ferromagnetic material and also permanent magnetic material. It is known as hexagonal M-type barium ferrite, M from the magnetite stone crystal structure name and the crystal structure belongs to the cubic hexahedral structure.

The excellent properties of $BaFe_{12}0_{19}$ among other are high frequency field, large saturation magnetization and high coercive force [7]. In this research, it is alloyed ceramics of $BiFe0_3$ and $BaFe_{12}0_{19}$ together hoping to improve magnetic properties which could improve value of magnetoelectric coupling (related to higher quality of multiferroic materials). Increasing value of magnetic properties (magnetic energy) can also increase magnetoelectric coupling. So the problem solution to avoid large current leakage is to form ceramics of $BiFeO_3$ – $BaFe_{12}O_{19}$ solid solution.

2. METHOD

It was used sol-gel method to produce ceramic of $BiFeO_3$-$BaFe_{12}O_{19}$ solid solutions. By using this method, it can produce nanoparticle powder (useful to have high quality of multiferroic material), minimal agglomeration, lower process temperatures and higher homogenity. The research used basic material which are chemical compound of $Bi_5O(OH)_9(NO_3)_4$, $Fe(NO_3)_3.9H_2O$, $C_6H_8O_7$, and aquabidestilate for stoichiometric calculation. The compounds are pro analysis Merck product with a purity of 99.99%. The stoichiometry was based on parameter varied of weight ratio of $BiFeO_3$: $BaFe_{12}O_{19}$ = 1 : 1. All basic materials were then dissolved using aquabidestilate. The solution was heated on a hot plate at 80-90 ºC for 4-5 hours until the gel formed. During the heating process, it used magnetic stirrer for stirring it to be homogeneous. Stirring was continued manually when the solution was very thick. It was then produced gel finally. The gel was then heated in the furnace at 150ºC for 2 hours to remove the remaining moisture from the process before. Then the gel was heated again in the furnace at 450ºC for 24 hours (calcination process) to remove unexpected elements e.g. C, H, and N from the basic materials. The powder was then crushed to avoid agglomerated and to have fine powder finally. After having finer powder than before, it was then sintered at 750, 800, 850ºC fo 8,10 and 12 hours respectively. The aim of sinter processing is occurring crystallization of powder. After sintering the powder was cruched again finally. To know the characteristics of ceramic powder which has multiferroic properties, it was used permagraph test which is type of MPS magnet – Physic EP3 – Permagraph L. to know magnetic properties. For permagraph test, powder was pressed first into pellet in diameter of 100 mm and thickness of 50 mm. Data obtained from the test results were then processed to obtain the graphs for analysis. It was used particle size analyzer with Beckman Coulter DelsaTM Nano instrument to know particle size of powder produced the research.

3. RESULTS AND DISCUSSIONS

The results of permagraph test are in hysteresis curve shown in Figure. 1, 2 and 3 for ceramic powder at sinter temperatures of 750ºC for 8 hours, 750ºC for 10 hours, and 850ºC for 2 hours sintering (random sample taken).

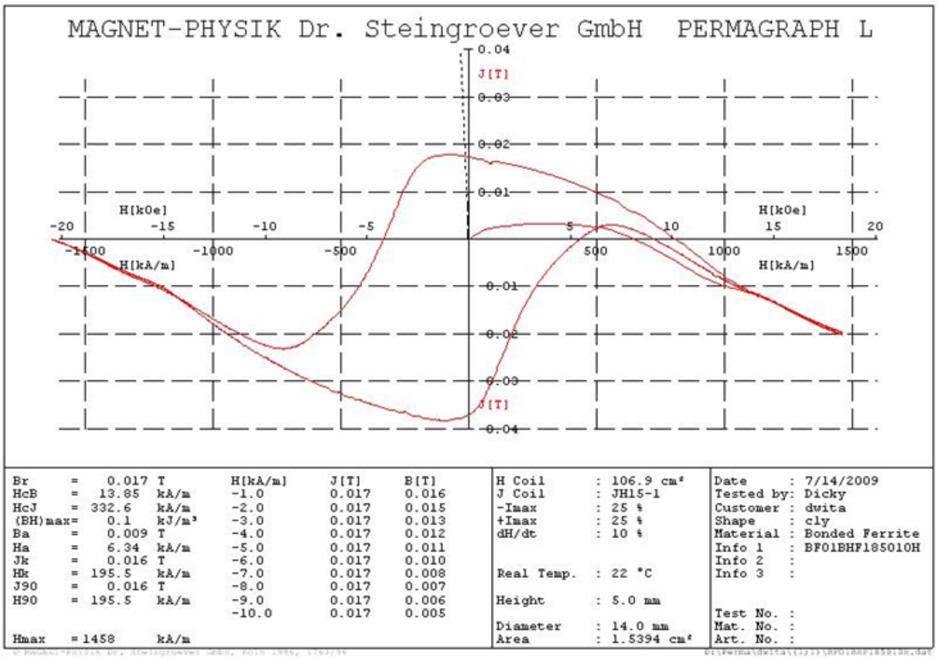

Figure 1. Hysteresis Curve for Powder at Sinter Temperature of 750ºC for 8 Hours Sintering

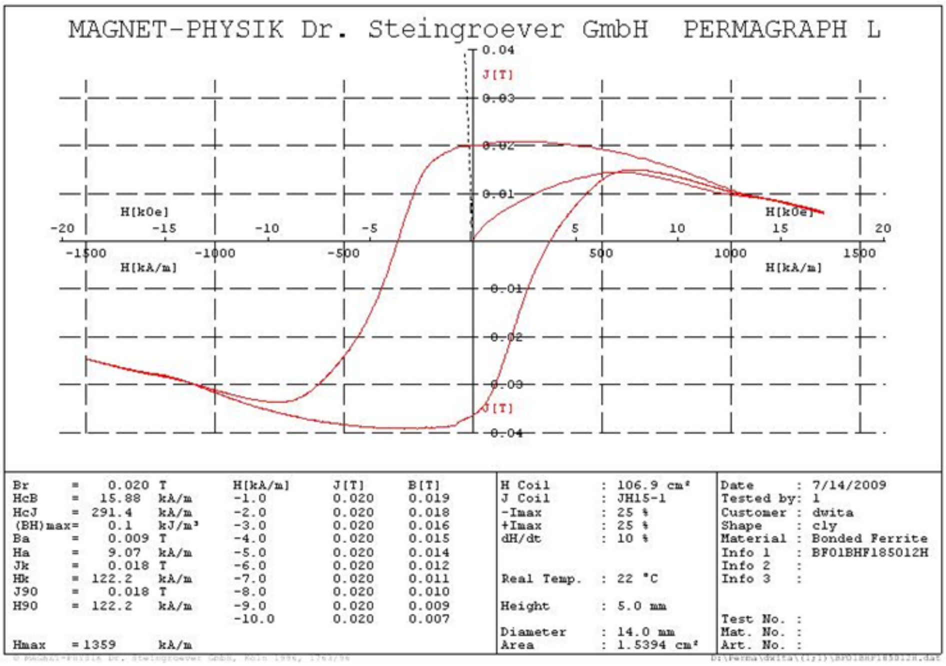

Figure 2. Hysteresis Curve for Powder at Temperature Sinter of 750^{o}C for 10 Hours Sintering

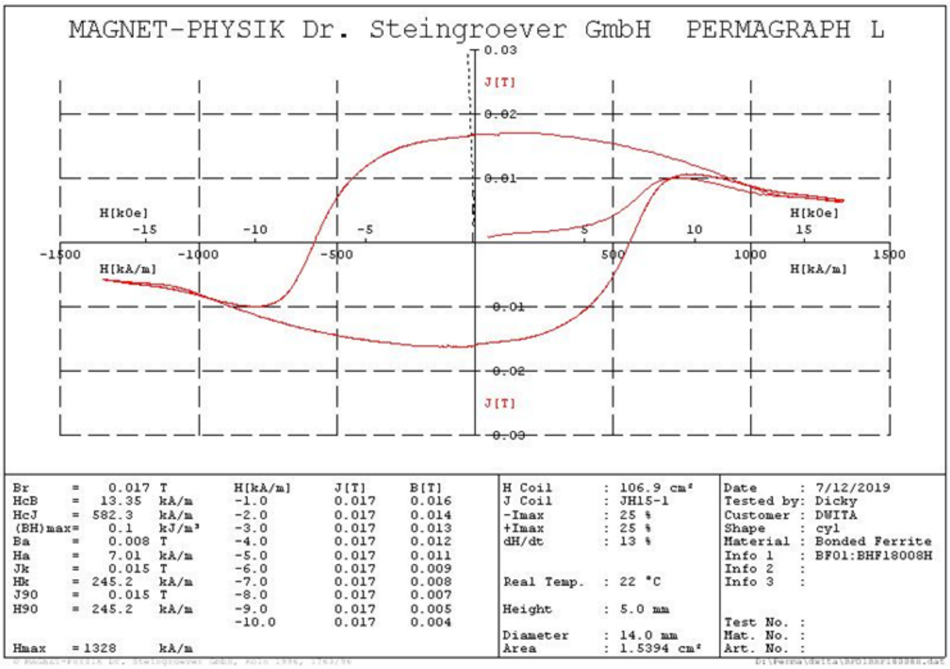

Figure 3. Hysteresis Curve for Powder at Temperature Sinter of 850^{o}C for 10 Hours Sintering

All samples have the same curve (hysteresis curve) showing properties of remanent, coersivity and saturation polarization. The curves are magnetization hysteresis (M-H) loops at room temperature measured with magnetic field of 1500 kA/m. The curves show that there is no regularity in increasing and decreasing of magnetic properties (remanent and coersivity) with increasing temperature and sintering time. But in this case it will calculated magnetic energy described the overall magnetic properties as shows in Table 1. as follows :

Table 1. Quantitative Data of Magnetic Properties From Samples of All Conditions

Sinter Temperature (°C)	Time of Sintering (Hours)	Remanent ($x10^4$ G)	Coersivity (kA/m)	Magnetic Energy (GkA/m)
750	8	0.017	332.6	5.654
750	10	0.020	291.4	5.828
750	12	0.024	310.5	7.452
800	8	0.013	652.1	8,4773
800	10	0.018	530.3	9.5454
800	12	0.015	644.1	9.6615
850	8	0.015	652.3	9.7845
850	10	0.017	582.3	9.8991
850	12	0.020	535.8	10.716

Table 1 shows that there is an increase of magnetic energy with increasing of temperatures and sintering times. Value of magnetic energy is the result of multiplication of remanent and coersivity value. This phenomenon shows that longer and higher of sintering times and sintering temperatures can decrease residual moment from canted Fe^{3+} spin structure which results in the disappearance of weak ferromagnetism [8]. Doping of $BaFe_{12}O_{19}$ in $BiFeO_3$ – $BaFe_{12}O_{19}$ solid solution should result in variety of powder particle resulting in enhanced magnetic energy of the samples. Since the result of the smaller particle can increase in grain boundaries, and the increase in grain boundaries contributes to the increase in magnetic energy, so the increase in magnetic energy may imply that the particle size of the sample becomes smaller [9.10,11,12] as shown in Table 2.

Table 2. Particle Size of Powder for All Conditions

Sinter Temperatures (°C)	Time of Sintering (Hours)	Particle Size (nm)	Magnetic Energy (GkA/m)
750	8	110	5.654
750	10	108	5.828
750	12	100	7.452
800	8	98	8,4773
800	10	95	9.5454
800	12	90	9.6615
850	8	86	9.7845
850	10	82	9.8991
850	12	79	10.716

Table 2 shows that powder with particle size less than 100 nm belongs to powder sintered at 800 and 850°C for 8, 10 and 12 hours sintering. Higher and longer of sinter temperatures and sinter time can reduce particle size and the smallest particle size belongs to powder at sinter temperature of 850°C for 12 hours sintering. There is a correlation between particle size and magnetic energy as discussed before (Table 2) due to presence of a large particle surface with increasing particle refinement, which can cause increasing interactions between magnetic atomic.

4. CONCLUSIONS

The conclusions of the research among others :

1. Sol-gel method with sinter temperatures of 800 and 850°C for 8, 10 and 12 hours sintering produced powder in nanoparticle size (less than 100 nm).
2. There is no regularity in increasing and decreasing of remanent and coersivity properties with increasing sinter temperatures and time of sintering but there is a regularity in increasing magnetic energy with increasing sinter temperatures and time of sintering.
3. There is a correlation between particle size and magnetic energy that is an increasing mangetic energy with decreasing of particle size.

5. ACKNOWLEDGEMENT

This research was supported financially by the Ministry of Research and Technology DIKTI, The Republic of Indonesia under Research Grant of Research based on Fundamental Reaserch 2019, letter of number decree of 43/AKM/MONOPNT/2019, March 27th, 2019, Ministry for Research and Technology DIKTI, The Republic of Indonesia. This research is also under contract by letter of number decree of 057/KP/LPKT-ITI/IV/2019, March 28th, 2019, Institut Teknologi Indonesia, the Republic of Indonesia

6. REFERENCES

[1] M.Zhong N, P, Kumar, E. Sagar, Z. Jian, H. Yemin, P>V. Reddy, "Structural Magnetic and Dielectric Properties of Y Doped $BiFeO_3$", Materials Chemistry and Physics, vol. 173, pp.126-131, 2016.

[2] Y.Chen, W.Y. Xing, Q. Yun, W. Gao, C.H. Nie, S.F. Zhao, "Eflects of Ho, Mn, Co-Doping on Ferroelectric Fatigue of BiFeO3 Thin Films", Electronic Materials Letters, vol. 11, pp. 601-608, 2015

[3] Y.X. Sun, X. Y. Liu, Y. Zeng, W. Yan, D.Q. Shi, H.R. Liu, "Photovoltaic Eflects in Polarized Polycrystalline $BiFeO_3$ Films", Journal of Electronic Materials, vol. 44, pp. 4207-4212, 2015

[4] H. Y. Dai, I. Chen, T.Li, D.W. Liu, R.Z. Xue, H.W. Xing, et al, "Eflects of $BaTiO_3$ Doping on The Structural , Electrical and Magnetic Properties of BiFeO3 Ceramics", Journal of Materials Science-Materials in Electronics, vol. 26, pp. 3717-3721, 2015

[5] Y.I. Ren, Z.H. Zhu, I.L. Zhu, I.G. Zhu, D.Q. Xiao, "Comparative Study of Mn^{3+} and Mn^{2+} Doping Effects on Structure and Electrical Properties of $BiFeO_3$, Thin Films", Ceramics International, vol. 41, pp. S234-S239, 2015

[6] J. Liu, Y.Qin, X.Q. Liu, X.M. Chen, "Dielectric Characteristics in BiFeO-Modified SrTiO, Incipient Ferroelectric Ceramics", Chinese Physics Letters, vol. 32, 2015

[7] J. Li. H. Zhang, Y. Liu, G. Ma, Q.Li, "Low-Temperature co-fired Ni-Ti co-Substituted Barium Ferrites" Journal of Composite Materials, vol. 50, pp. 173-178, 2015

[8] R. Dahiya, A. Agarwal, S. Sanghi, A. Hooda, P. Godara, "Structural Magnetic and Dielectric Properties of Sr and V Doped BiFeO Multiferroics, "Journal of Magnetism and Magnetic Materials, vol. 385, pp. 175-181, 2015

[9] F.Esat, T.P. Comyn, A.J. Bell, "Microstructure Development of BiFeO-PbTiO, Films Deposited by Pulsed Laser Deposition on Platinum Substrates", Acta Materialia, vol. 66, pp 44-53, 2014

[10] S.M.H. Khalkhali, M.M. Tehranchi, S.M. Hamidi, "Photo-magnetic Assisted Ferroelectric Polarization in Magneto-Electric BiFeO-BaTiO Thin Film", Journal of Magnetism and Magnetic Materials, vol. 355, pp.188-191, 2014

[11] D. Suastiyanti, Ismojo, M. Wijaya, "Magnetic Properties of Magnesium Doped Bismuth Ferrite as Multiferroic Material Produced by Sol-Gel Method at Low Temperature", International Journal of Advanced Research, 5(5), pp. 1954-1963, 2017.

[12] D. Suastiyanti, S. Yatmani, M. Wijaya, "Magnetic Properties of Nanomagnetic Material Based on $BaTiO_3$ and $BiFeO_3$ with Variation of Temperatures and Times of Sintering", IOP Conf. Series: Journal of Physics: Conf. Series 1153 (012052), 2019