Magnetic properties of nanomagnetic material based on BaTiO₃ and BiFeO₃ with variation of temperatures and times of sintering

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Abstract. This research uses basic compound of BaTiO₃ (BTO) and single phase multiferroic material BiFeO₃ (BFO) with weight ratio of BTO: BFO = 2: 1 to produce multiferroic ceramic. The purpose of this research is to know magnetic properties of the ceramic which consists of BFO multiferroic material combined with BTO electrical material. Changes in magnetic properties will affect the quality of multiferroic materials. The process used sol-gel method using temperature and time of calcination of 350°C and 4 hours, then sintered at 700, 750 and 800°C for 2, 4 and 6 hours respectively. The sample was characterized by XRD test with diffraction angle of 20°-100° and the machine was made in Phillips type of PW 1835. This test is to know types of phases. To know magnetic properties of sample, it was used Permagraph test. To know particle size, it was used Particle Size Measurement from Nano Instrument of Beckman Coulter DelsaTM. The results show that new phase is formed which is the dominant phase, Barium Bismuth Iron (III) Oxide (BaBiFe₂O₅) with the largest percentage (98.79%) obtained at sintering process with temperature of 750°C for 6 hours. Increasing the percentage of dominant phases is linear by increasing of magnetic properties of the sample. The highest value of magnetic properties among other coercivity of 247.1 kA / m, remanent of 0.275 Tesla and Saturation of 0.41 Tesla belongs to ceramic with condition of sintering temperature and time of 750°C and 6 hours. The smallest particle size is also obtained under the same conditions equal to the particle size of 54-57 nm.

1. Introduction
Permanent magnets require high coercivity values to show strong and stable of net magnetization in external fields requiring high coercivity. In hard magnetic material, uniaxial magnetic anisotropy is required and some of the following characteristics are high coercivity, large magnetization and rectangular hysteresis loop [1,2,3,4,5,6,7]. Coercivity is also called the coercive field is the intensity of the magnetic field required to reduce the magnetization of the material to zero after the magnetization of the sample reaches saturation. The intrinsic properties of magnetics are grouped into primary and secondary. Primary properties such as saturation magnetization Js and magneto crystalline anisotropy K1 constants are directly related to magnetic structures, whereas secondary intrinsic magnetic forces such as HA anisotropic field strength and energy of specific domain wall γw, are derived from the primary properties. The primary and secondary magnetic properties characterize the actual magnetic state.
One multi-function material is a multiferroic material that has 2 or more properties such as ferromagnetism, ferroelectric, ferroelasticity and ferrotoroidicity that appear simultaneously in a material. The average particle size of material phase in the nanometer scale causes the increasing of interacting surface fraction as the grain size increases. Therefore, material preparation will be directed to a nanoceramic system instead of a conventional ceramic system. To obtain this nanoceramic material it is necessary to prepare the basic material in a nanoparticle size which is carried out by sol-gel process which is a simple and easy method. The multiferroic material successfully synthesized in the form of single-phase ceramics is BiFeO$_3$ [8,9,10,11] which is produced by sol-gel method. The purpose of this research is to know the influence of temperatures and times of sintering on the magnetic properties of ceramic consist of BiFeO$_3$ multiferroic material combined with BaTiO$_3$ electrical material with weight ratio of BTO: BFO = 2: 1. Given that magnetic properties are one of the properties that exist in multiferroic properties so the change of magnetic properties will also affect the multiferroic properties of the material. Material characterization was done by using XRD test to know the phase type formed by XRD machine which was made in Philips of PW 1835 type, used diffraction angle of 20°-100°, permagraph test to know magnetic properties using MPS Magnet-Physik EP3-Permagraph L. To know particle size, it was used Particle Size Measurement which was made in Beckman Coulter DelsaTM Nano instrument.

The application of the material was as ultimate memory devices for mechatronics industry and manufacture tools (it must have high hardness). The tools were manufactured by wire processes of Taguchi and Fuzzy Logic Taguchi [12].

2. Methods
It uses sol-gel method which could produce nanoparticle, homogen powder, little agglomeration and lower temperature. The basic compound used is of pro analysis Merck product with a purity of 99.99% such as Bi$_5$O(OH)$_9$(NO$_3$)$_4$, Fe(NO$_3$)$_3$.9H$_2$O, HNO$_3$, H$_2$O, Ba(NO$_3$)$_2$, TiO$_2$ and citric acid C$_6$H$_8$O$_7$. Figure 1 shows the flowchart of research.

![Figure 1. The Process Step of Nanomagnetic Ceramic Synthesized by Sol-Gel Method](image-url)
3 Results and Discussion

3.1. X-Ray Diffraction Test Result
The pattern of XRD test results show that for all conditions of sinter temperatures and times, synthesis process produces new phase dominantly, Barium Bismuth Iron (III) Oxide (BaBiFe$_2$O$_5$) more than 80%. Some diffraction patterns of the ceramic is shown in Figure 2.

![Figure 2: Diffraction Pattern of Ceramic at Sinter Temp of 700, 750 and 800°C for 2 and 6 Hours](image)

Figure 2 shows that the diagram shows new phase (dominant phase), BaBiFe$_2$O$_5$ in largest percentage is condition with temperature of sinter of 750°C and time of sinter of 6 hours. Difference of dominant phase percentage is determined by position of 2 Theta shown in Figure 2, where as at 2 Theta of 25-30° it shows different peak. Generally all process conditions produce the same dominant phase with a percentage greater than 80% [13]. The recent research shows that the largest percentage of dominant phase is owned by ceramic sintered at 750°C for 6 hours. At lower temperature (700°C), elements of Ba$^{2+}$, Bi$^{3+}$ and Fe$^{3+}$ are still form individual oxide compounds [14,15,16]. While at higher temperature (800°C) the dominant phase will decompose into barium oxide, bismuth oxide and ferri oxide compounds.

3.2. Permagraph Test Result
From permagraph test result, it is obtained magnetic properties of ceramic, among other magnetic field remanent and coersivity shown in figure 3.
Figure 3. Remanent of Magnetic Field (a) and Coercivity of Magnetic Field (b) for Ceramic at Differences of Sintering Temperatures and Times

Figure 3 (a) shows that remanent value goes up with increasing sinter time. The highest remanent value is obtained by condition at 750°C sinter and 6 hours of sinter time. Figure 3 (b) shows that the coercivity value increases with going up sinter time for sinter temperature of 750°C but decreases with increasing sinter time for sinter of 700 and 800°C. The highest coercivity value obtained at the same condition that is at temperature of sinter of 750°C and 6 hours sinter time. As we know from table 1, at this condition it is also obtained the largest percentage of the dominant phase, so it shows that increasing of dominant phase percentage also causes increasing value of remanent and coercivity. Saturation of magnetic field value obtained from permagraph test is shown in Figure 4.

Figure 4. Saturation of Magnetic Field for Powder at Differences of Sintering Temperatures dan Times
Figure 4 shows that the saturation value increases with increasing sinter temperature and time. The highest saturation value is obtained at sinter temperature of 750°C and sinter time of 6 hours condition. This phenomenon is linear with the increasing percentage of dominant phase at sinter temperature of 750°C for sinter time of 2, 4 and 6 hours.

3.3. Particle Size Measurement
The result of nanoparticle size is shown in Figure 5.

![Figure 5. Particle Size for Ceramic at Differences of Sinter Temperatures and Times](image)

Figure 5 shows that all process condition produces nanopowder with particle size less than 100 nm. The powder which is sintered at 750°C has the smallest particle size (54-57 nm). At higher temperature (800°C), particle will grow causes it is getting bigger.

4. Conclusion
Sol-gel method at this research produces powder with nanoparticle at all process condition (sinter temperatures of 700, 750 and 800°C and sinter time of 2, 4 and 6 hours). Percentage of new phase which is dominant phase, BaBiFe₂O₅ of 95-98% is obtained by condition at sinter temperature of 750°C. Percentage of dominant phase affects value of remanent, coersivity and saturation of magnetic field which increase with increasing percentage of dominant phase.

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