Synthesis and characterization of iron (III) oxide from natural iron sand of the south coastal area, Purworejo Central Java

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Abstract. The purpose of this study is to synthesize and characterize of iron sand from the south coastal area of Purworejo, Central Java. Iron sand was mechanically filtered using permanent magnets 35 times. The filtered iron sand was characterized using x-ray diffraction (XRD) to determine the compound contained therein and his physical characteristics. The filtered iron sand was heated at temperatures of 800 ° C and 900 ° C for 5 hours in the furnace and after it was cold and followed by XRD testing. Based on the x-ray diffraction pattern, a single phase Fe_2O_3 as a permanent magnet base can be formed after a 900 ° C heating process for 5 hours.

1. Introduction

The presence of iron sand in Indonesia is abundant, especially those located on the southern coast of Java. In Purworejo Regency, Central Java province, especially in the southern coast, the iron sand has not been optimally used for electronic devices [1, 2] such as permanent magnets on electric speakers or electric motors. Iron sand in nature itself generally is composed of magnetite (Fe_3O_4), maghemite (γ-Fe_2O_3) and hematite (α-Fe_2O_3) [3-6]. These compounds have different characteristics and uses and are usually mixed with other impurities.

Magnetite (Fe_3O_4) has been widely used especially as nanoparticles as high-density information storage, ferrofluid, catalysis, electronic devices, biomedical and pigments [7-10]. There have been various methods or techniques used to synthesize magnetite, especially nanoparticles, including co-precipitation, solvothermal processing, and high temperature organic phase decomposition [11-17]. The method or technique in synthesizing magnetite affects the physical properties of magnetite itself.

Hematite is the mineral form of iron (III) oxide (α-Fe_2O_3) are antiferromagnetic. Hematite, especially nanoparticles, has several applications such as pigments for paints, drug targeting cancer cells and for labeling and tracking target cells using magnetic resonance imaging (MRI) [18]. Iron (III)

oxide can be synthesized using several methods including co-precipitation, thermal decomposition, hydrothermal synthesis, microemulsion, and sonochemical synthesis [18, 19-26].

This research reported the process of synthesis and characterization of Iron (III) oxide (α-Fe_2O_3) from iron sand at south coastal area of Purworejo, Central Java, which has been mechanically filtered using a permanent magnet for 35 times and followed by a thermal decomposition process at a temperature of 800 ° C and 900 ° C for 5 hours during which the characterization process at each stage was carried out using x-ray diffraction (XRD).

2. Experiments

Iron sand powder comes from the coastal area of Purworejo, Central Java. It was then characterized using XRD to determine the content of the compound. The X- ray diffraction patterns were recorded by "step-scan" method in 2 θ range from 20 ° to 80 ° with Cu $K\ \alpha$ radiation. Iron sand powder was filtered using a permanent magnet for 35 times, then XRD testing was carried out to determine the compound and its physical properties. The Rietveld analysis was performed applying the High Score Plus program that is an updated version for Rietveld refinement with PC and mainframe computers. A sample of the filtered product was put into the furnace for the oxidation process with a heating temperature of 800° C and 900° C for 5 hours then was cooled in the furnace (Nabertherm N31/H) until room temperature was achieved. Oxidized samples were characterized using XRD to determine the compounds therein and their physical properties.

3. Result and Discussion

3.1 Testing of X-ray Diffraction (XRD) Raw Material

Iron sand taken from the coast of Purworejo district was used as raw material in this study by XRD testing to ascertain the compounds in the sand. The test results are shown in figure 1.

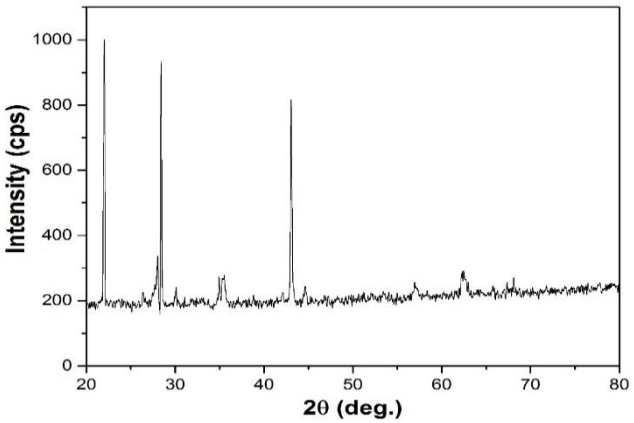

Figure 1. X-ray diffraction pattern from iron sand.

The diffraction pattern in figure 1 was analyzed using the High Score Plus software from PANanalytic where the results of the initial analysis were shown in figure 2. Figure 2 shows that the x-ray diffraction pattern of the iron sand matches with the diffraction pattern of Magnetite (Fe_3O_4) and Cristobalite low (SiO_2) compounds in the Inorganic Crystal Structure Database (ICSD) number 98-015-8740 and 98-005-2371. Based on figure 2, it can also be concluded that iron sand as raw material in this study is dominantly composed of Fe_3O_4 and SiO_2. There may still be other compounds contained in iron sand with a composition below 5%. Because the characterization using XRD can only identify the compounds in a material with the percentage above 5%.

The next analysis process was to quantitatively calculate the content of two main compounds in the iron sand. The results of further analysis of x-ray diffraction pattern are shown figure 3. The

quantitative analysis shows the content of the two main compounds in iron sand as shown in figure 3 is 61.1% SiO_2 and 38.9% Fe_3O_4. Characteristics of the physical properties of SiO_2 in iron sand are shown in figure 4 and figure 5 for Fe_3O_4 compounds .

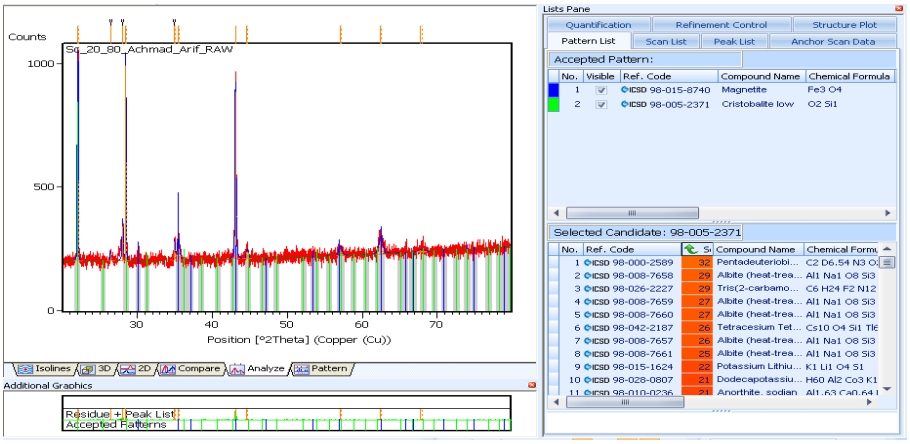

Figure 2. Results of initial analysis of x-ray diffraction patterns from iron sand.

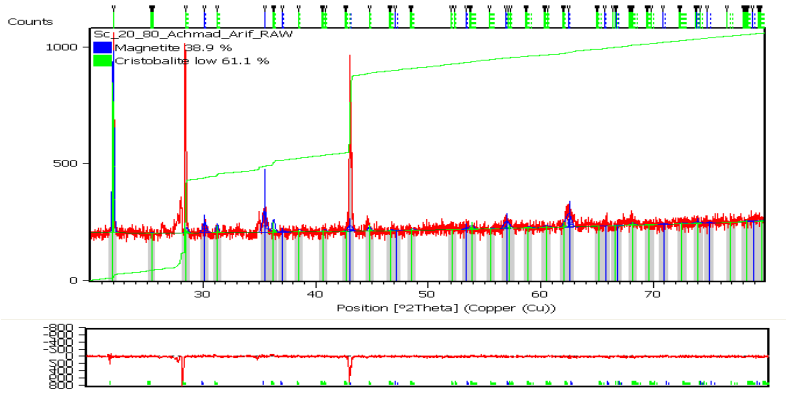

Figure 3. The results of quantitative analysis of two main compounds in iron sand.

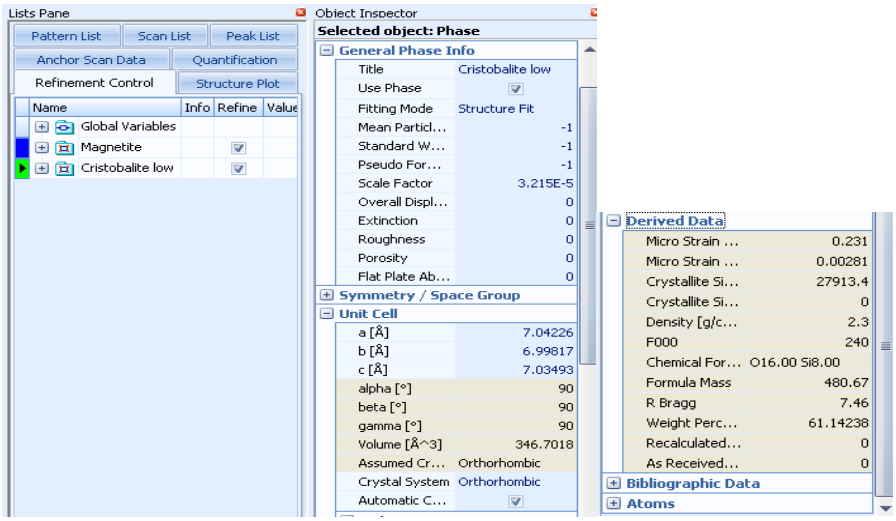

Figure 4. Characteristics of physical properties of Cristobalite low (SiO_2) in iron sand.

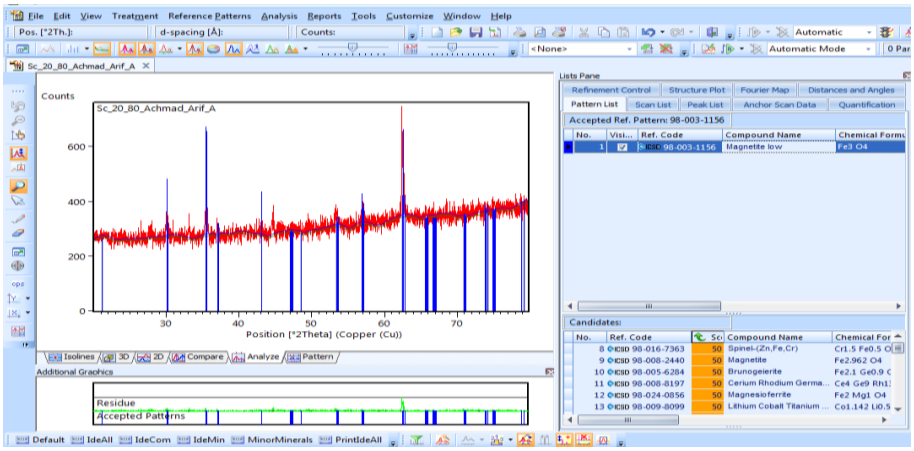

Figure 5. Characteristics of the physical properties of magnetite (Fe_3O_4) in iron sand.

3.2 Mechanical Sand Filtering Process

Iron sand has been taken from the coast of Purworejo district and has been characterized for its chemical and physical properties using High Score Plus software analysis, then the next step was the process of filtering or separating other particles from iron sand particles. The filtering process used two methods or techniques: mechanical and chemical. The mechanical filtering process is the process of separating other particles from iron sand using permanent magnets for 35 times. This process was performed to obtain iron sand that is free of other particles that do not contain the Fe element (iron). The filtering results was then characterized for its chemical and physical properties using XRD. The test results using XRD produce diffraction patterns as shown in figure 6.

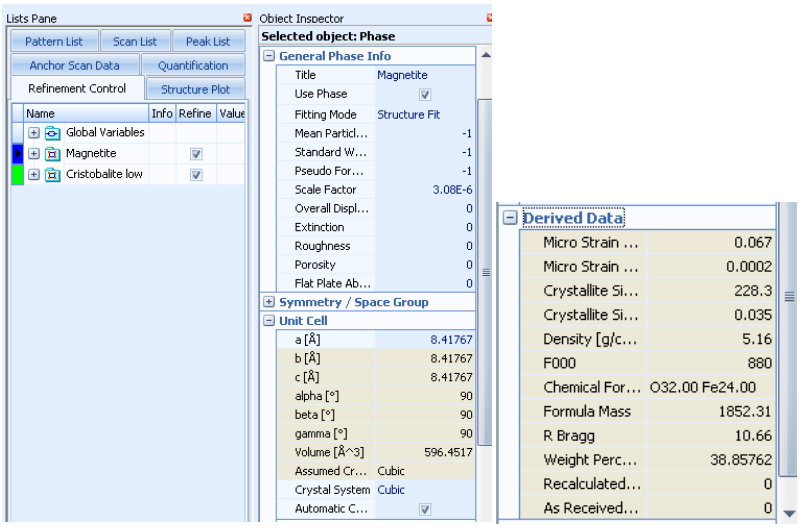

Figure 6. Iron sand x-ray diffraction pattern after mechanical filtering

Figure 6 shows that the x-ray diffraction pattern of iron sand after mechanical filtering for 35 times matches with the diffraction pattern of the magnetite low (Fe_3O_4). Then the characteristics of the physical properties of Fe_3O_4 were analyzed, the results are shown in figure 7 and figure 8. Figure 7 shows that the unit cell of magnetite low (Fe_3O_4) is orthorombic. The crystallite size of the magnetite low (Fe_3O_4) based on Figure 8 is 100 nm.

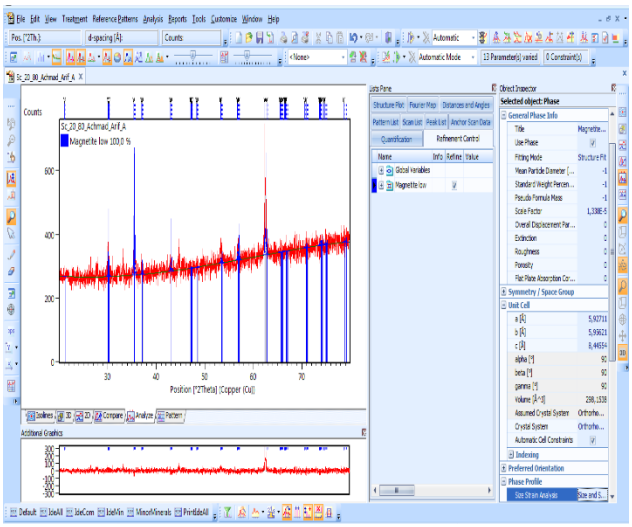

Figure 7. The cellular unit of magnetite low (Fe_3O_4)

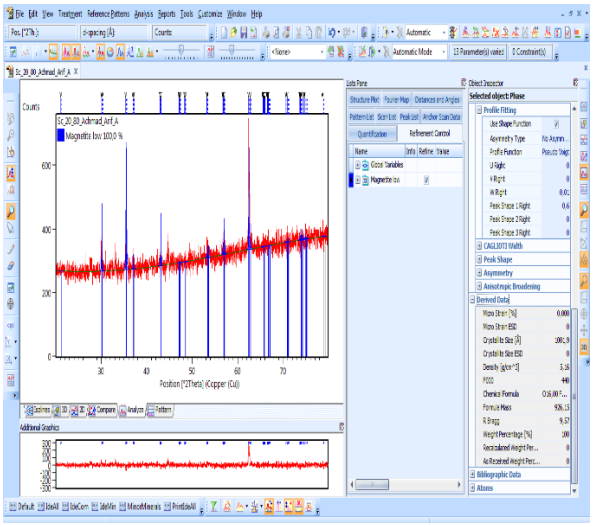

Figure 8. Sizes of magnetite low (Fe_3O_4) crystalline compounds

3.3 Enrichment Process or Oxidation of Magnetite low (Fe3 O4)

The magnetite low (Fe_3O_4) as the result of the mechanic filtering must be transformed into Fe_2O_3 compounds. This transformation process is called the enrichment process. The enrichment process was carried out by heating the powder of a magnetite low compound (Fe_3O_4) to a temperature of 900°C or this process is called the magnetite low oxidation process (Fe_3O_4). The magnetite low (Fe_3O_4) oxidation process was carried out at 800°C for 5 hours then cooled to achieve room temperature and the characterization process was carried out using XRD. The results of the characterization are shown on figure 9.

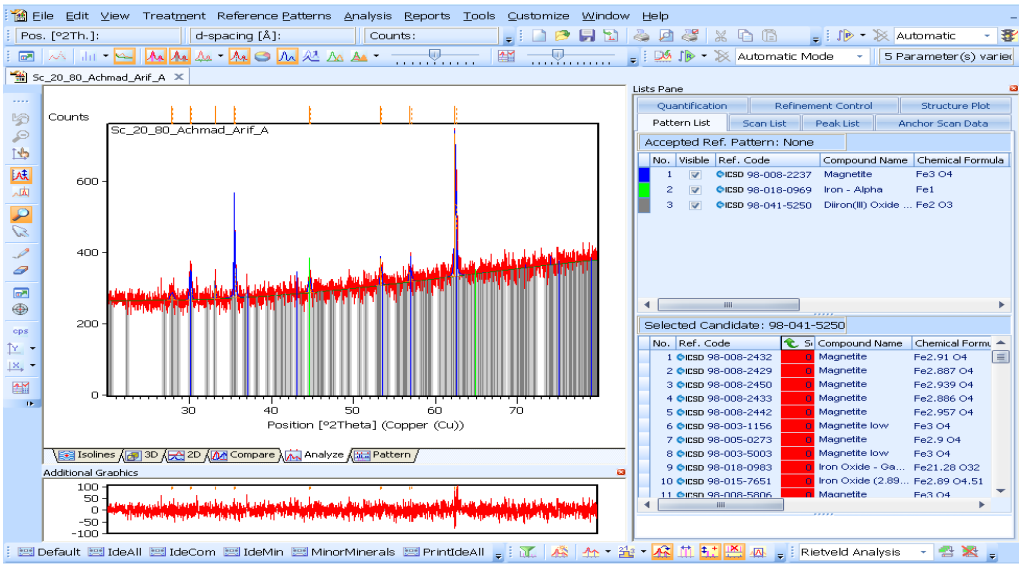

Figure 9. X-ray diffraction pattern of iron sand which has been oxidized at a temperature of 800°C

Figure 9 shows that after magnetite low (Fe3O4) is oxidized at 800 ° C, the present phases are magnetite (Fe3O4), Fe and Fe2O3. Fe and Fe2O3 are present as the results of decomposition of magnetite low (Fe3O4). The quantitative analysis of the three phases on the magnetite low

(Fe3O4) oxidation process was carried out at 800 ° C for 5 hours shown in Figure 5.10. The percentage of magnetite (Fe3O4) phase is 37.9%, Fe is 13.9% and Diiron (III) Oxide (Fe2O3) is 48.3%. In the oxidation process of 800 ° C for 5 hours, the Fe2O3 phase is present and has the highest content, although it has not achieved 100%. The transformation process of magnetite low (Fe3O4) phase into fully Fe2O3 phase is the expected result from the enrichment process or oxidation process, where the phase of the Fe2O3 compound is a base material of permanent magnet.

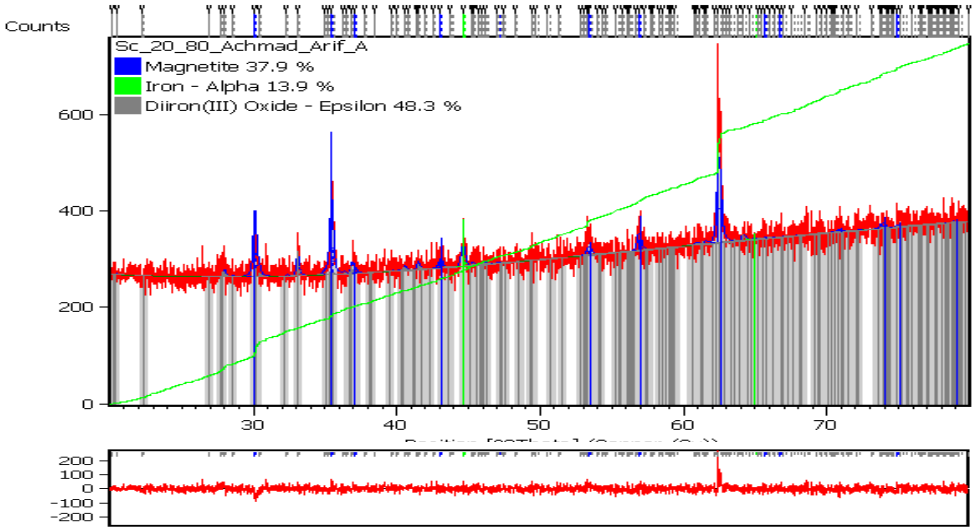

Figure 10. Results of quantitative analysis of magnetite low (Fe_3O_4) oxidation process at 800 ° C

The process of analyzing physical properties based on diffraction patterns in Figure 10 using High Score Plus software found that the shapes of the magnetite (Fe_3O_4) and Iron-Alpha (Fe) phase units are cubic, and Diiron (III) Oxide (Fe_2O_3) is orthorombic. The crystallite size of the magnetite (Fe_3O_4) phases formed after oxidation of 800 ° C for 5 hours is 35 nm, Iron-Alpha (Fe) is 34 nm and the Diiron (III) Oxide (Fe_2O_3) phase is 69 nm.

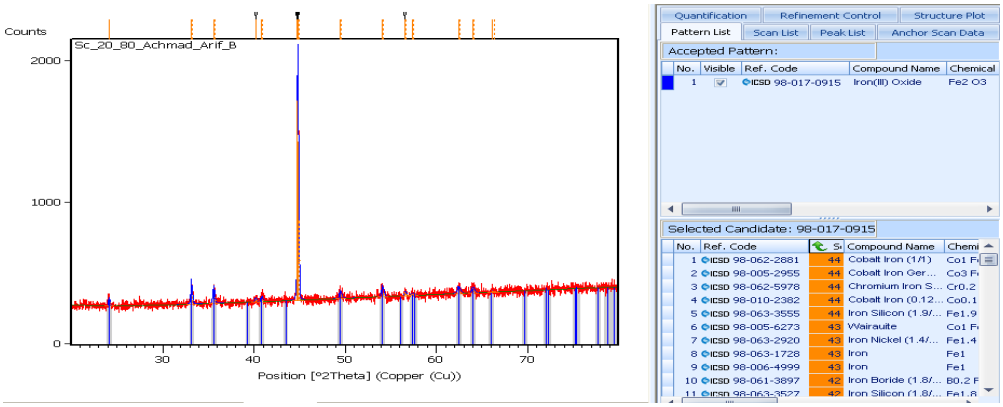

Figure 11. X-ray diffraction pattern as a result of the oxidation process to a temperature of 900 ° C

Because the composition of the Fe_2O_3 compound as the main material of permanent magnets has not reached 100%, it is necessary to do the next oxidation process at a temperature of 900 ° C for 5 hours. The XRD pattern formed after oxidation to 900 ° C for 5 hours is shown in figure 11. Based on figure 11, the XRD pattern shows that the diffraction pattern of the Iron (III) Oxide (Fe_2O_3) compound, which indicates the formation of a single phase Fe_2O_3 (100%). This indicates that the oxidation process or enrichment of iron sand to a temperature of 900° C for 5 hours has succeeded in

transforming the low magnetite (Fe_3O_4) phase into a single phase Fe_2O_3 which is the main material of permanent magnet.

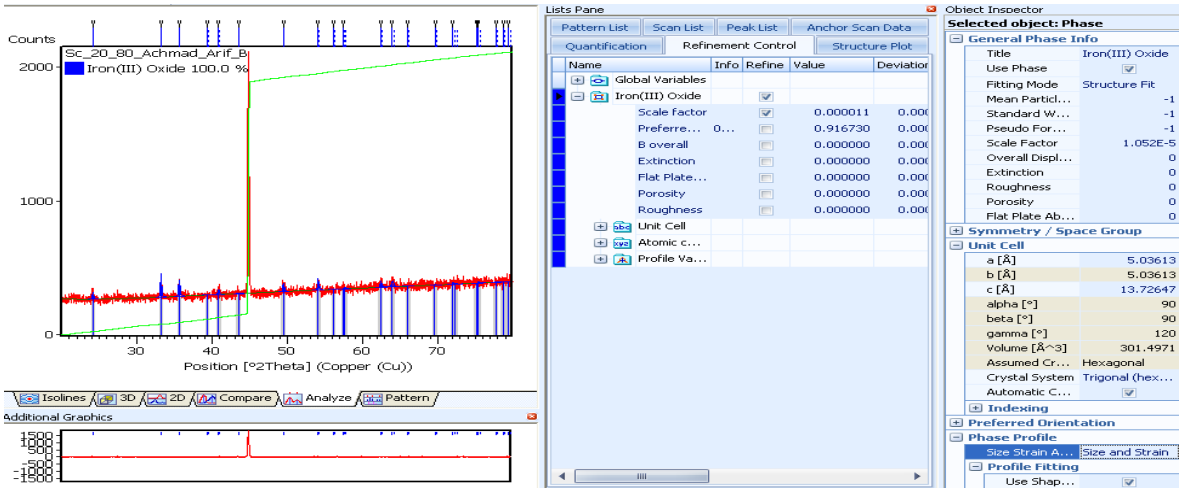

Figure 12. The Results of physical properties analysis based on XRD patterns from oxidation process up to 900 ° C

The process of analyzing physical properties based on diffraction patterns in figure 11 using High Score Plus software shows that the Fe_2O_3 phase has the form of cell unit is trigonal (hexagonal) as shown in figure 12. Based on XRD pattern in figure 11, the crystal size of single phase Fe_2O_3 after the analysis process equals to 71 nm, as shown in figure 13.

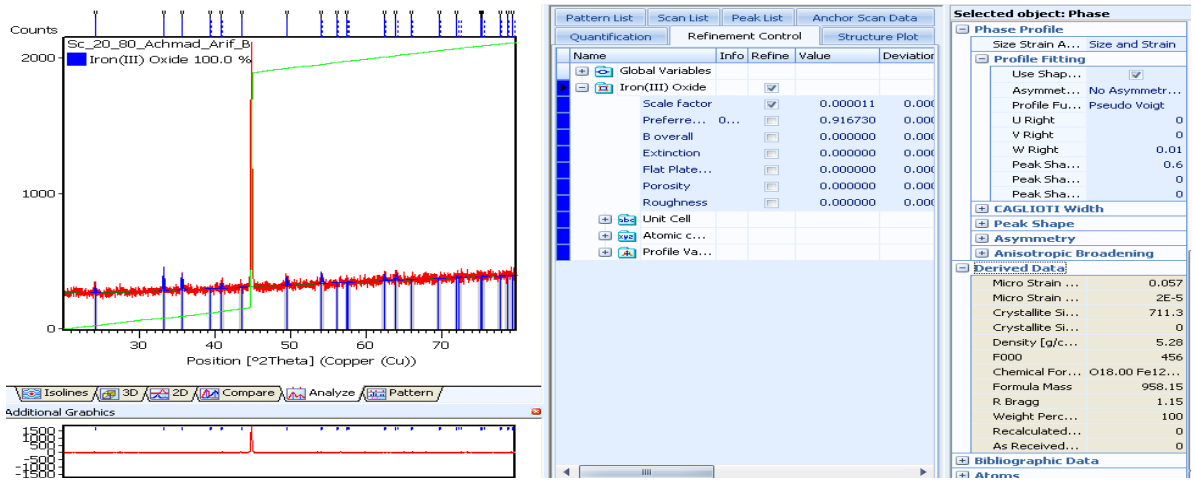

Figure 13. Results of analysis of other physical properties based on XRD patterns resulting from oxidation process up to 900 ° C

4. Conclusion

The process of filtering or refining iron sand which is performed mechanically for 35 times succeeded in getting iron sand which has a low magnetite low compound (Fe_3O_4). The process of enrichment or oxidation of low magnetite compounds (Fe_3O_4) at temperatures up to 800 ° C for 5 hours has not been successful in producing a single phase of Fe_2O_3. Single phase Fe_2O_3 as the main compound of the magnetic material was successfully obtained after the oxidation process of the low magnetite compound (Fe_3O_4) at a temperature of up to 900 ° C for 5 hours.

References

[1] Andimutiafitri 2018 Synthesis of Magnetite Based Iron Sand by Using Coprecipitation method *J. of Applied Physics (IOSR-JAP)* **10(3)** 40-42.
[2] Rianna M, Sembiring T, Situmorang M, Kurniawan C, Setiadi E A, Tetuko A P, Simbolon S, Ginting M, Sebayang P 2018 Preparation and Characterization of Natural Iron Sand from Kata Beach, Sumatera Barat Indonesia with High Energy Milling (HEM) *J. Natural* **18(2)** 97-100.
[3] Manjunatha M, Kumar R, Anupama A V, Khopkar V B, Damle R, Ramesh K P, Sahoo B 2019 XRD, Internal Field-NMR and Össbauer Spectroscopy Study of Composition, Structure and Magnetic Properties of Iron Oxide Phases in Iron Ores *J. of Materials Research and Technology* (Article in Press).
[4] Muflikhah, Rusdiarso B, Putra E G R, Nuryono 2017 Modification of Silica Coated on Iron Sand Magnetic Material with Chitosan for Adsorption of Au(III) *Indonesian J. of Chemistry* **17(2)** 264-273.
[5] Sehah, Raharjo S A, Kurniawan M A 2016 Distribution of Iron Sand in the Widarapayung Coast Area at Regency of Cilacap Based on Magnetic Anomaly Data *Indonesian J. of Applied Phys.* **6(02)** 97-106.
[6] Fahlepy M R, Wahyuni Y, Andhika M, Tiwow V A, Subaer 2019 Synthesis and Characterization of Nanopraticle Hematite (α-Fe_2O_3) Minerals from Natural Iron Sand Using Co-Precipitation Method and its Potential Applications as Extrinsic Semiconductor Materials Type-N *Materials Science Forum* **967** 259-266.
[7] Mamani J B, Gamarra L F, Brito G E S 2014 Synthesis and Characterization of Fe_3O_4 Nanoparticles with Perspectives in Biomedical Applications *Materials Research* **17(3)** 542-549.
[8] Rusianto T, Wildan M W, Abraha K, Kusmono 2012 The Potential of Iron Sand from The Coast South of Bantul Yogyakarta as Raw Ceramic Magnet Material *J. Teknologi* **5(1)** 62-69.
[9] Fahlepy M R, Tiwow V A, Subaer 2018 Characterization of Magnetite (Fe_3O_4) Minerals from Natural Iron Sand of Bonto Kanang Village Takalar for Ink Powder (Toner) Application *J. of Phys.: Conf. Series* **997** 012036.
[10] Morel M, Martínez F, Mosquera E 2013 Synthesis And Characterization Of Magnetite Nanoparticles From Mineral Magnetite *J. of Magnetism and Magnetic Materials* **343** 76–81.
[11] Harvey D T and Linton R W 1981 Chemical Characterization of Hydrous Ferric Oxides by X-ray Photoelectron Spectroscopy *Analytical Chemistry* **53(11)** 1684.
[12] Rockenberger J, Scher E C, Alivisato A P 1999 A New Nonhydrolytic Single-Precursor Approach to Surfactant-Capped Nanocrystals of Transition Metal Oxides *J. of American Chemical Society* **121** 11595-11596.
[13] Chin S F, Pang S C, Tan C H 2011 Green Synthesis of Magnetite Nanoparticles (via Thermal Decomposition Method) via Controllable Size and Shape *J. of Materials and Environmental Science* **2** 299–302.
[14] Fajaroh F, Setyawan H, Widiyastuti W, Winardi S 2012 Synthesis of Magnetite Nanoparticles by Surfactant-free Electrochemical Method in an Aqueous System *Advanced Power Technologies* **23** 328–333.
[15] Han C, Zhao D, Dung C, Hu K 2012 A Facile Hydrothermal Synthesis of Porous Magnetite Microspheres *Materials Letters* **70** 70–72.
[16] Xu J, Yang H, Fu W, Du K, Sui Y, Chen J, Zenh Y, Li M and Zou G 2007 Preparation and Magnetic Properties of Magnetite Nanoparticles by Sol–gel Method *J. of Magnetism and Magnetic Materials* **309** 307–311.
[17] Vergés M A, Costo R, Roca A G, Marco J F, Goya G F, Serna C J and Morales M P 2007 Uniform and Water Stable Magnetite Nanoparticles with Diameters Around The Monodomain–Multidomain Limit *J. of Phys. D: Applied Physics* **41** 134003.
[18] Wu W, He Q, Jiang C 2008 Magnetic Iron Oxide Nanoparticles: Synthesis and Surface

Functionalization Strategies *Nanoscale Research Letter* **3** 397–415.
[19] S.A. Abd El Aal, Abdelhady A M, Mansour N A, Hassan N M, Elbaz F, Elsayed, Elmaghraby K 2019 Physical and Chemical Characteristics of Hematite Nanoparticles Prepared Using Microwave-assisted Synthesis and its Application as Adsorbent for Cu, Ni, Co, Cd and Pb from Aqueous Solution *Materials Chemistry and Physics* **235** 121771.
[20] Tadic M, Trpkov D, Kopanja L, Vojnovic S, Panjan M 2019 Hydrothermal synthesis of hematite (α-Fe_2O_3) nanoparticle forms: Synthesis Conditions, Structure, Particle Shape Analysis, Cytotoxicity and Magnetic Properties *J. of Alloys and Compounds* **792** 599-609.
[21] Trpkov D, Panjan M, Kopanja L, Tadic M 2018 Hydrothermal Synthesis, Morphology, Magnetic Properties and Self-assembly of Hierarchical α-Fe_2O_3 (Hematite) Mushroom-, Cube- and Sphere-like Superstructures *Applied Surface Science* **457** 427–438.
[22] Lassoued A, Lassoued M S, Dkhil B, Gadri A, Ammar A 2017 Synthesis, Structural, Optical and Morphological Characterization of Hematite Through The Precipitation Method: Effect of Varying The Nature of The Base *J. of Molecular Structure* **1141** 99-106.
[23] Darezereshki E, Bakhtiari F, Alizadeh M, Vakylabad A B, Ranjbar M 2012 Direct thermal decomposition synthesis and characterization of hematite (α-Fe_2O_3) nanoparticles *Materials Science in Semiconductor Processing* **15** 91-97.
[24] Housaindokht M R and Pour A N 2011 Precipitation of Hematite Nanoparticles via Reverse Microemulsion Process *J. of Natural Gas Chemistry* **20** 687–692.
[25] Han L H, Liu H, Wei Y 2011 In Situ Synthesis of Hematite Nanoparticles Using a Low-temperature Microemulsion Method *Powder Technology* **207** 42-46.
[26] Roshan A H, Vaezi M R, Shokuhfar A, Rajabali Z 2011 Synthesis of Iron Oxide Nanoparticles via Sonochemical Method and Their Characterization *Particuology* **9** 95-99.

Acknowledgments
We thank Engineering Faculty, Universitas Negeri Semarang for the support of laboratory facilities and funding. This research was funded from the DIPA Number: 042.01.2.400899/2019 through the collaborative research scheme. We also thank the Department of Physics, FMIPA Diponegoro University for supporting the laboratory facilities.