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EFFECT OF SINTERING TEMPERATURE ON CRYSTALLOGRAPHY AND MICROSTRUCTURE OF YTTRIUM IRON GARNET VIA MECHANICAL ALLOYING TECHNIQUE

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ABSTRACT

This work focused on the preparation of yttrium iron garnet ($Y_3Fe_5O_{12}$, YIG) via mechanical alloying technique derived by steel waste product. The steel waste was purified by using magnetic and non-magnetic particles (MNM) and Curie temperature separation (CTS) technique. The powder from the CTS technique was oxidized at 500°C for 9 hours in air to form the iron oxide (Fe_2O_3). The Fe_2O_3 was mixed with Y_2O_3 using high energy ball milling for 9 hours. The obtained mixed powder was pressed and sintered at varied temperature 500/600/700/800/900/1000/1100/1200 °C. X-ray diffraction (XRD) showed the YIG was completely formed at 1100 °C. The crystallite size and grain size of YIG powder were observed. The results show the grain size and crystallite size increased as a function of sintering temperatures.

During processing of steel, mill scale is generated during hot rolling and considered as a waste. Mill scale is formed when steel surface is in contact with humid air at high temperatures in the hot strip mill process. This condition is formed an oxide layer called commonly mill scales. Mill scales generated in steel plants is represents about 2% of the steel produced. Almost 5 million tons of the steel wastes are manufactured from steel mill factories annually. Steel waste contains highest percentage of iron (≈ 72 % Fe) [1], consists of iron oxides; wustite (FeO), hematite (Fe_2O_3), and magnetite (Fe_3O_4) [2]. YIG ferrite is a soft ferrimagnetic material with garnet structure, and the most representative and a well-known compound among of rare-earth iron garnets [3]. YIG is formed by heating a combination of a mixture of Y_2O_3 and Fe_2O_3 giving Fe and Y to the ratio of 5:3 (Fe:Y) [4]. The ferrimagnetic behavior possessed by YIG garnet ferrite is used for many applications in magnetic and magneto-optical devices. The investigations of YIG gives much attention to researcher due to its low energy loss, used in the microwave frequency range, having the smallest magnetic-resonance linewidth among the magnetic materials, and high saturation magnetization [5]. Various techniques have been used to prepare YIG. The solid state method of preparing YIG ferrites from the corresponding oxides Y_2O_3 and Fe_2O_3 is a very tedious process, requiring prolonged grinding operation, and yielding large particles with broad size distribution and poor chemical homogeneity [6]. In order to overcome the shortcomings of the solid-state reaction method, besides many wet-chemical approaches, high-energy ball milling, also known as mechanical alloying is the most suitable technique due to its simple, controllable process, and can be operated on a large scale. It is well-known that crystal structure and microstructure play an important role in determining the magnetic properties of YIG [5]. This however, in previous literature has been much concerned on these effects to the magnetic properties of pure YIG, fabricates from a commercially product of Fe_2O_3 raw materials [4, 5]. Therefore, this research aims to study the crystallographic and morphology of YIG derived by steel waste product as the Fe_2O_3 source. It is believe that this motivation will be significantly contributes to the low-cost production of YIG without reducing its superior properties.

The mill scales were crushed for several hours to obtain the precise sized of powder. The scale was mixed in a steel pot ball milling using distilled water and steel balls as a medium. The slurries were dried in an oven before carried out to separation technique. The process was continued with magnetic technique as done by Azis et.al (2015) [7]. Fe_2O_3 (2.70 g) derived from mill scale and Y_2O_3 (2.30 g, Alfa Aesar 99.99 %) were weighed and mixed in a stoichiometric ratio according to the equation:

$$5Fe_2O_3 + 3Y_2O_3 \rightarrow 2Y_3Fe_5O_{12} \quad \text{Eq. (1)}$$

The powders were milling in a SPEX 8000D mechanical alloying machines at room temperature for 9 hours. The balls to powder weight ratio is 10:1. To prevent excessive heating of the milling system, the milling was carried out for 90 min followed by 20 min of pause. The as-milled powders were pressed uniaxially into pellets and toroid form in a die with the diameter of 10 mm and 18 mm respectively at about 3 MPa. The pressed samples were sintered at elevated temperatures from 500/600/700/800/900/1000/1100/1200 °C for 9 hours in air with the heating rate of 3 °C/min. The phase of the YIG samples were characterized using an X-ray diffractometer (PANalytical X'Pert PRO PW3040) with Cu Kα radiation (λ = 0.1542 nm). The diffraction patterns were compared to the patterns in the JCPDS index. The microstructure of the bulk samples was characterized by field emission scanning electron microscopy (FeSEM) using an FEI NOVA NanoSEM 230 machine. The real permeability values were measured by using a HP4291B Materials Impedance Analyzer.

The particle size distribution was measured by TEM observation. Figure 1 shows an image of YIG nanoparticles after HEBM. The average particle size estimated from the image is 57.80 nm. The particle size is considered to be small because of the enough time of impact between the powder that gives sufficient energy to complete the reaction between yttrium oxide and iron oxide.

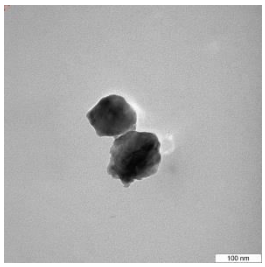

Figure 1: TEM image of YIG nanoparticles after mechanical alloying.

Figure 2 shows the XRD patterns powder sinter at different temperature from 500°C to 1200°C. The XRD pattern of the powder sinter at 500-800°C shows the presence of orthorhombic yttrium ferrate (III) Y(FeO_3) phase. By increasing the heat treatment temperature to 900°C the YIG pattern start to appear along with Y(FeO_3) phase. For the powder heat treated at 1100°C, the XRD pattern exhibited diffraction peaks corresponding to the garnet phase, indicating that the formation of garnet phase was completed in this temperature range. The XRD results showed that a cubic garnet phase was formed in the samples sintered at 900-1200^{o}C. This indicates that the energy provided from the high-energy ball milling with subsequent sintering was enough to produce a desired single cubic garnet phase. The changes observed in the samples with the increase of intensity of the samples with increasing the temperatures. The width of XRD peaks also decreases with increase the temperatures. This indicates the crystallite growth with increase in the sintering temperature. The crystallite size measurements were determined from the full-width at half maximum (FWHM) of the strongest reflection of the (024) peak by using the Scherrer equation (Eq. 2).

$$D = \frac{K\lambda}{\beta cos\,\theta} \quad \text{Eq. (2)}$$

where D is the crystallite size, k is a shape function for which a value of 0.9 is used, λ is the wavelength of the radiation, β is the fullwidth at half maximum (FWHM) in radians in the 2θ scale, and θ is the Bragg angle. By applying Scherrer's formula and considering FWHM of the diffraction peaks, average crystallite size of the YIG ferrite samples sintered at various temperatures was calculated. Table 1 showed mean crystallite sizes of sintering temperature from 500°C to 1200°C. The FWHM peaks widths show a decrease that indicate the grain growth and increase the crystallite size as a function of sintering temperatures. Higher sintering temperature increase atomic mobility and this cause grain growth, which results improve the crystallinity. The YIG ferrites samples show a variation of size, ranging from 13.1 to 197.0 nm.

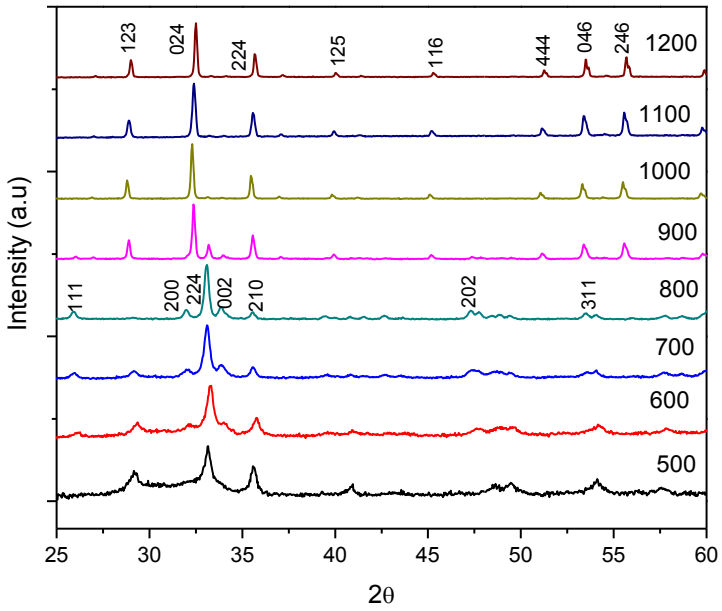

Figure 2: XRD pattern of YIG nanoparticles sintered at 500°C to 1200°C.

Table 1: The hkl, peak position, peak width, space group and crystallite sizes of YIG ferrites at 500°C to 1200°C

T (°C)	(*hkl*)	Peak position	Peak width (°)	Space group	Crystallite sizes (nm)
500	224	33.1071	0.7036	P n m a	13.1
600	224	33.2293	0.5336	P n m a	17.9
700	224	33.0776	0.3389	P n m a	30.9
800	224	33.0537	0.2499	P n m a	46.3
900	024	32.3633	0.1345	P n m a	131.3
1000	024	32.2846	0.1166	I a -3 d	179.8
1100	024	32.3737	0.1641	I a -3 d	88.9
1200	024	32.4851	0.1134	I a -3 d	197.0

Representatives FeSEM micrographs of the YIG sintered at different temperature are shown in Figure 3. The micrographs of the YIG sintered at 500 °C and 600 °C showed that the particles had 36-43 nm in diameter with some agglomeration. The grain size was fairly homogeneous and the crystal started to grow when the temperature increased up to 700 °C. At 700 °C, the formation of necks between particles has formed which lead to the

development of grains. At 800 °C, a small irregular and rounded morphology is observed. The grain becomes rounder and bigger as sintering temperature increased. The grain became finally close to spherical with a smooth surface at 1100 °C. It is believed that a mass transport mechanism started with atomic surface diffusion at relatively low temperatures continued to occur by the grain boundary diffusion, resulting in formation of necking, contact growth, pores elimination and particles grain growth. The micrographs of FESEM for samples sinter at 1200 °C exhibit the final stage of sintering where the micrographs show the well formation of polyhyedral grains with diminished pores.

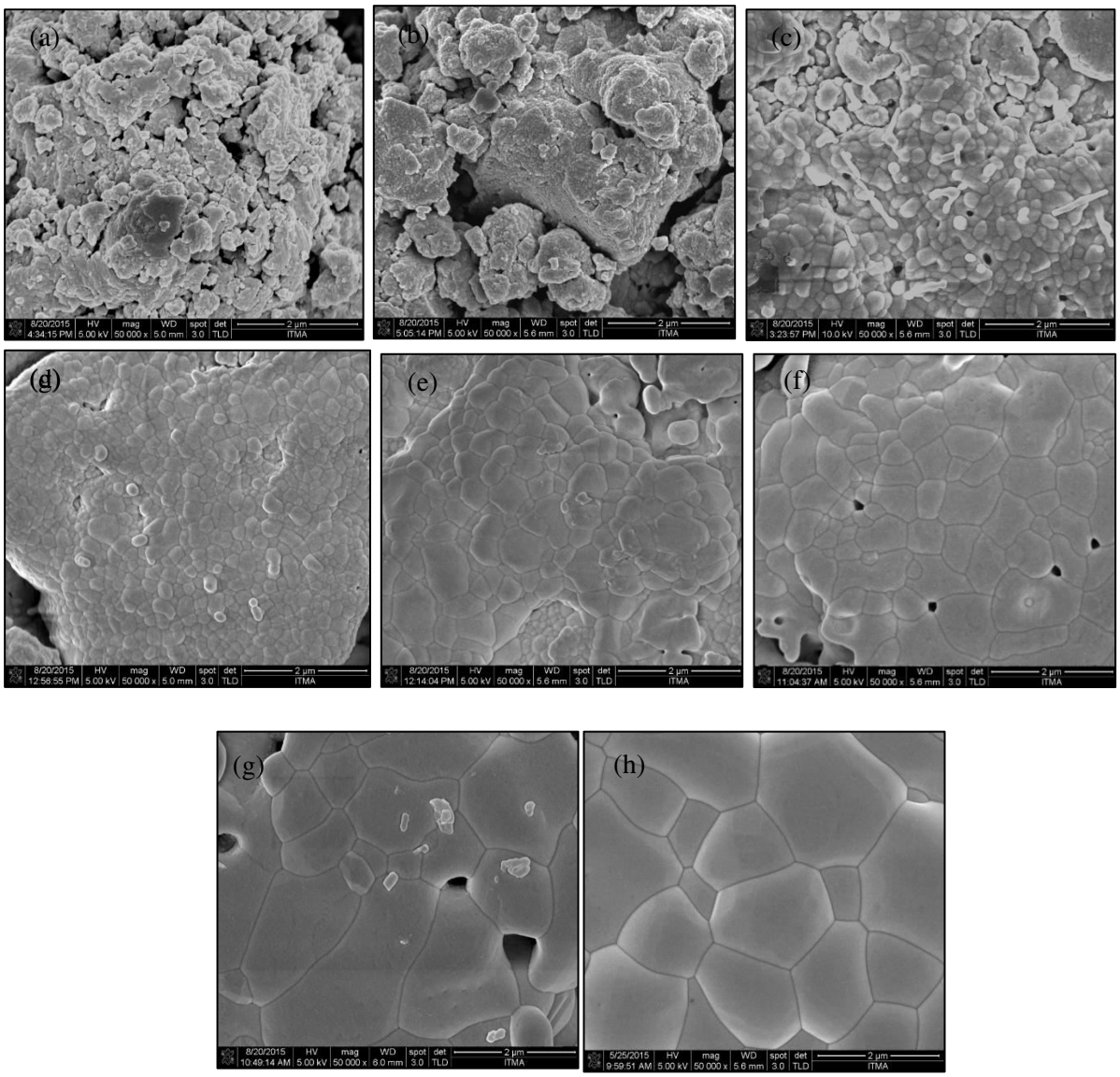

Figure 3: FeSEM images of the samples sintered at (a) 500 °C, (b) 600 °C, (c) 700 °C, (d) 800 °C, (e) 900 °C, (f) 1000°C, (g) 1100°C, and (h) 1200°C.

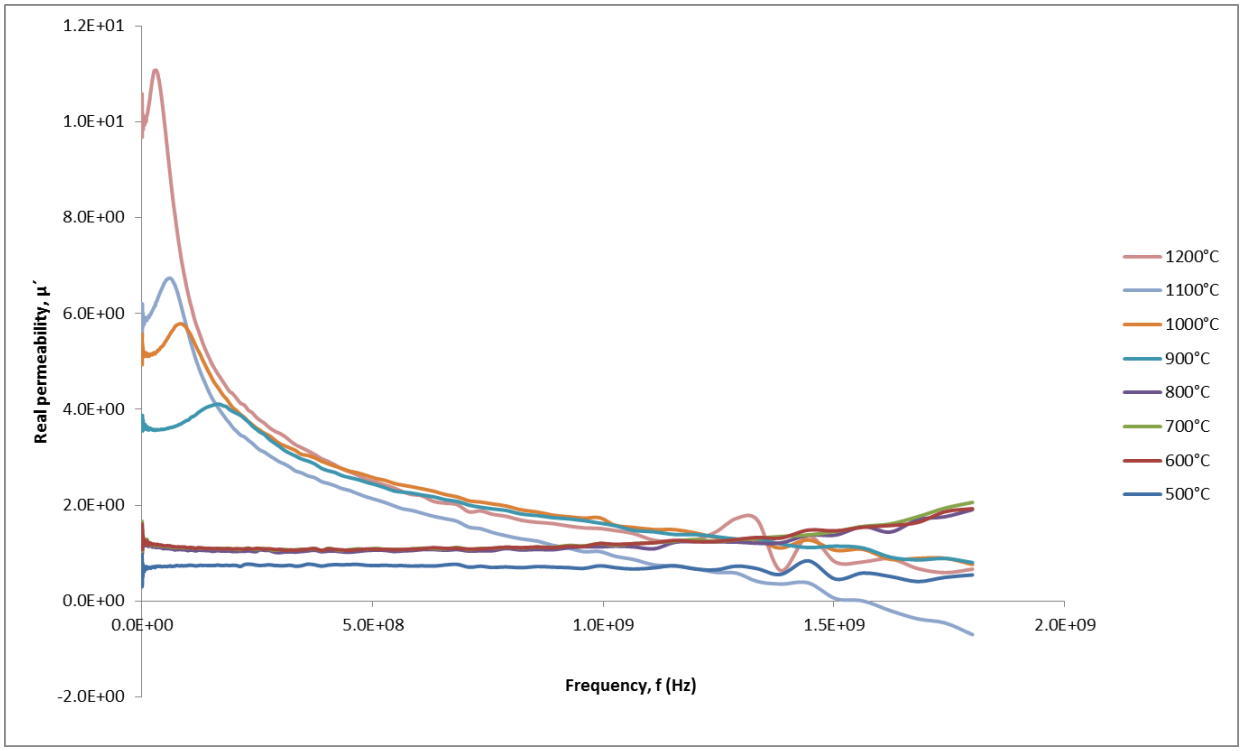

Figure 4: Real permeability, μ´ of YIG at 500°C to 1200 °C sintering temperature

Figure 4 showed the real permeability, μ´, for YIG measured from 10 MHz to 1 GHz. The μ´ value was increased to a maximum value and decreased rapidly to a very low value. The μ´ at 10 MHz showed the highest value for the sample sintered at 1200 °C, which is 57. The increased of μ´ value with increased in of sintering temperature was attributed to the increased of grain sizes, where larger grains diminished the number of grain boundaries. This leads to a reduction of magnetic anisotropy, which contributes by the ease of the domain walls movement.

YIG powders have been prepared from the steel waste product via the mechanical alloying technique. YIG ferrites was successfully persists a complete garnet structure starting at 1100 °C. As the sintering temperature increases, the average grain sizes and mean crystallite size also increases. The XRD spectra indicated the presence of orthorhombic yttrium ferrate (III) Y(FeO_3) phase for samples sintered at 500 to 800°C. The YIG garnet structure start to appear for sample sintered at 900°C, and completely fomed at 1100°C. The FWHM showed an increase in the mean crystallite size from 13.1 to 197 nm when the sintering temperature was increased, as agreed with enlargement of the grains size observed by FeSEM micrographs. The real permeability also increases as a function of sintering temperature, gives maximum μ´ is 57 for samples sintered at 1200°C.

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