J. Solid St. Sci. & Technol. Letters, 2015, Vol. 16 No. 1-2, pp. 62-66
http://letters.masshp.net/ | ISSN 0128-8393

COMPOSITION AND MAGNETIC PROPERTIES OF ALUMINIUM SUBSTITUTED YTTRIUM IRON GARNET WASTE MILL SCALES DERIVED VIA MECHANICAL ALLOYING TECHNIQUE

Makiyyu Abdullahi Musa[1], Rabaah Syahidah Azis[1,2], Mansor Hashim[2], Nurul Huda Osman[1], Siti Irryani Adnan[1], Noruzaman Daud[1], Nuraine Mariana Mohd Shahrani[2], Sakinah Sulaiman[2], Nor Nadhirah Che Muda[2] and Abddul Halim Azizan[1]

[1]Department of Physics, Faculty of Science, University Putra Malaysia, 43400 UPM, Serdang, Selangor, Malaysia
[2]Institute of Advanced Materials, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

ABSTRACT

This paper presents the effects of aluminium substitution on sample composition, density and magnetic properties of yttrium iron garnet, $Y_3Fe_5O_{12}$ (YIG). Mill scales, in a form of flakes was obtained from the steel industry in Malaysia. The mill scales was purified to produce high purity hematite, Fe_2O_3. The mill scales derived Fe_2O_3 were used as raw material to prepare the aluminum substituted yttrium iron garnet $Y_3Fe_{(5-x)}Al_{(x)}O_{12}$ (Al-YIG) with variation compositional x = 0.0, 0.5, 1.0, 1.5 and 2.0 using mechanical alloying technique. X-ray fluorescence (XRF) was used to investigate the percentage compositions of the raw mill scales. The magnetic hysteresis of sample were investigate by using B-H tracer (MATS). Density of the Al-YIG bulk samples was found to decrease with increasing xAl_2O_3 content. The saturation magnetization M_s also shows a decrease with increase xAl_2O_3 content. The maximum saturation magnetization M_s and coercivity H_c was found at 310 G and 14.98 Oe, respectively. With increase xAl_2O_3 to 2.0%, the M_s and H_c were reduced to 29.35 G and 3.15 Oe, respectively.

Yttrium iron garnet (YIG), also called microwave ferrites, belongs to the iron-containing oxides with general formula $A_3B_5O_{12}$. It has specific magnetic and magneto-optical properties that make it used in many technological applications. YIG has attracted much scientific researches and investigations since it was discovered [1]. It is best applied in microwave devices including isolators, circulators and phase shifters. This is due to its narrowest line width and the smallest losses of all materials ($\Delta H \approx 0.1$ G at 10 GHz) [2]. YIG is also applied in permanent magnets, magnetic recording media and telecommunications systems [3][4]. YIG has cubic crystal structure with oxygen ions surrounding the nonmagnetic Yttrium (Y^{3+}) and magnetic iron (Fe^{3+}) ions. The nonmagnetic Y^{3+} ions occupy the dodecahedral (c) sites, three Fe^{3+} ions occupy the tetrahedral (d) sites and the remaining two Fe^{3+} ions occupy the octahedral (*a*) site. Due to the antiparallel alignment of the magnetic moments of (*a*) and (*d*) sites, a resultant magnetism emerges, reflecting the magnetic moment of the surplus Fe^{3+} ion in (*d*) site. Thus, YIG suited well for magnetism studies because it has well defined cation distribution. The magnetic properties of YIG are highly flexible due to the three crystallographic sites available for different cation distribution and substitution. It is interested to determine the nature of substitution of iron ions by rare-earth materials like aluminium and study the cation distribution pattern in the crystals. This is because aluminium-substituted YIG have led to a new phenomenon that favors wider range of technological applications [1], [5]. The substituted Al ions prepare to replace the tetrahedral iron ions, especially at low Al concentrations. [2] also reported that Al or Ga substitution at the tetrahedral site lowers the magnetization to $4\pi M_s$ = 300 G. This improves the microwave properties of the materials, as low magnetization is required for microwave applications. In this work, we used mill scale to prepare Al-YIG via mechanical alloying technique. Mill scale is a type of iron oxide that is formed during steel hot-rolling process. It is bluish grey in color because the steel surface has to be subjected to a very high temperature and rolling pressure. The mechanical alloying technique was proved to produce high quality ferrites and improved its physical and mechanical properties [6]. The effect of Al substitution in YIG will be investigate and discuss in this paper.

Preparation of hematite from mill scales. The mill scales were obtained from the steel factories in Malaysia. The steel waste flakes were ball milled for several hours to form a fine powder. The steel powder undergo the magnetic and nonmagnetic separation process by using impurity separation process (ISP) and Curie temperature technique (CTST) [7]. The purified magnetic powder was then oxidized at 500^oC for 6 hours to form the hematite, Fe_2O_3 according to Eq. 1.

$$2FeO\ (s) + {}^1/_2\, O_2 \quad \rightarrow \quad Fe_2\, O_3 \qquad (1)$$

Preparation of aluminium-substituted yttrium iron garnet ($Y_3Fe_{5-x}Al_xO_{12}$). The hematite, Fe_2O_3, formed was mixed with commercial Y_2O_3 and Al_2O_3 to prepare aluminium-substituted Yttrium Iron Garnet ($Y_3Fe_{5-x}Al_xO_{12}$) with x = 0.0, 0.5, 1.0, 1.5, and 2.0. Stoichiometric amount of the powders were mixed using mortar to form a homogeneous mixture. SPEX800D mill was used to mix the sample in a steel vial using different diameters of ball mill. The sample is grinded to form fine powder size with ball to powder weight ratio (BPR) of 10:1. The milled powder was calcine at 950^oC. The powder was then pressed using hydraulic press to form bulk toroid samples with diameter × thickness (11.4 × 3.1 mm), and sintered at 1200^oC.

Characterizations. The chemical composition of the Al-YIG powder was analyzed by using energy dispersive X-ray fluorescence (EDXRF). The B–H hysteresis curves of the Al-YIG toroidal samples were obtained by measuring the impedance of a coil wound around the toroid samples, with a soft magnetic materials automatic test system (MATS, MATS3010s, China) at room temperature under an applied magnetic field of 4000 A/m (~50 Oe). The Curie temperature was measured at 10 kHz using an Agilent 4284A LCR-meter with the sample placed in an oven as the temperature at which the sample's inductance, L, fell most sharply. The density of Al-YIG ferrites was measured using the Archimedes principle at room temperature and the density were estimated quantitatively.

The chemical percentage of the mill scales were investigate by using XRF analysis (Figure 1). The graph indicate the mill scales contains high content of iron Fe (98.59%) and the presence of small amount other compound such as Mn (0.55%), Ca (0.08%) and Tm (0.78%). With the high content of Fe, it encourages us to purify the mill scales powder to produce high purity of ferrites.

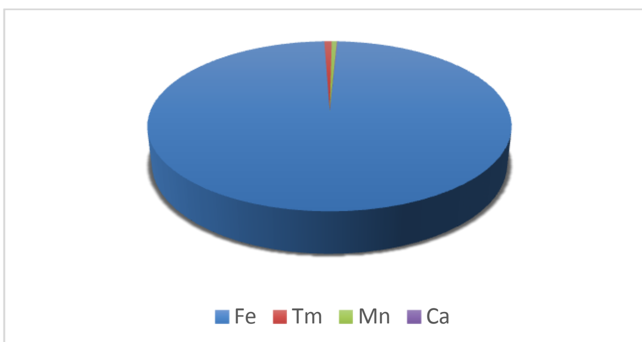

Figure 1: XRF analysis of raw mill scales powder.

Figure 2 shows the density of Al-YIG with xAl_2O_3 content. The density show a decrease with the addition of Al_2O_3 content. The decreasing in density of the Al-YIG ferrite is due to the heavier aluminium atomic mass as compared to the other element in the Al-YIG ferrites samples. The atomic mass of aluminium is 26.982 u which is lower compared to iron (55.845 u) and yttrium (88.906 u). The density show a decrease with increase Al content from 3.548 to 3.294 g/cm^3.

J. Solid St. Sci. & Technol. Letters, 2015, Vol. 16 No. 1-2, pp. 62-66
http://letters.masshp.net/ | ISSN 0128-8393

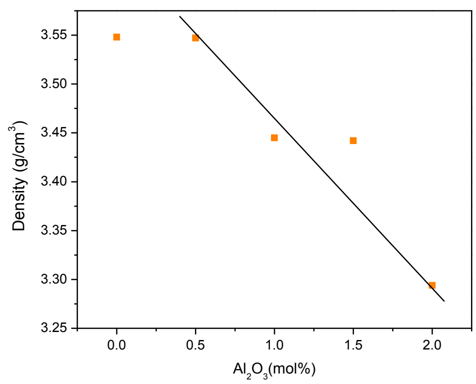

Figure 2: The variation of density of Al-YIG ferrites system.

In order to understand the effect of $x\mathrm{Al_2O_3}$ in the substituted garnet, a study of magnetic hysteresis was plotted. Figure 3 shows hysteresis loops of $Y_3Fe_{5-x}Al_xO_{12}$ samples, thermal treated at 1200 °C, at room temperature. As Fe^{3+} ions are substituted by the non-magnetic Al^{3+} ions in (*d*) and (*a*) sites the super exchange interactions of iron ions in the lattice is reduced, consequently reducing the magnetization, as it can be lower Fe/Al ratios [8]. Reduction of magnetization for higher Al^{3+} concentration is due to the substitution of Fe^{3+} with Al^{3+} in octahedral and tetrahedral sites in substituted YIG and the consequent reduction of super exchange interactions in the lattice [8]. The remanence, M_r and saturation magnetization, M_s decreases with increasing Al concentration (Figure 4). This is due to the decrease in the super exchange interaction between the (*a*) and (*d*) sites. The Al ions distribution between tetrahedral and octahedral sites also affects the domain and nature of the super exchange interaction. The density and magnetic parameters of Al-YIG ferrites is tabulated in Table 1. Curie temperature of Al-YIG also decreases as increase $x\mathrm{Al_2O_3}$ content.

Table 1: The density and magnetic parameters of Al-YIG ferrites.

$x\mathrm{Al_2O_3}$	Density (g/cm^3)	Saturation magnetization M_s (G)	Remanence M_r (G)	Coercivity H_c (Oe)	Curie temperature T_c (^oC)
0.0	3.548	310.00	165.90	8.79	225
0.5	3.547	172.00	100.00	12.58	230
1.0	3.445	100.00	56.10	14.98	240
1.5	3.442	39.32	8.80	8.80	270
2.0	3.294	29.35	0.34	3.15	-

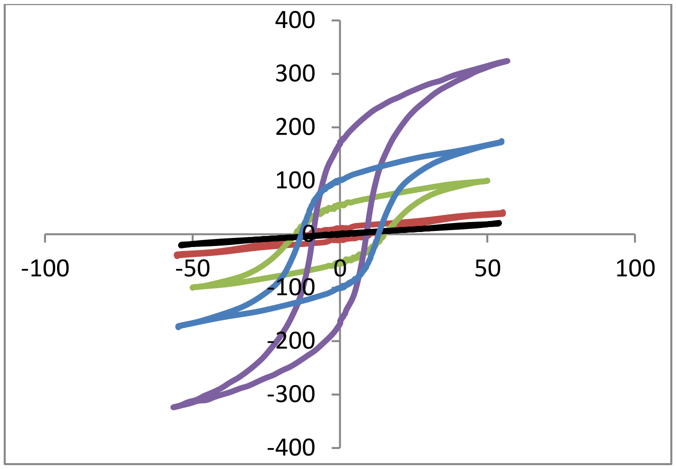

Figure 3: Hysteresis M-H loops of $Y_3Fe_{5-x}Al_xO_{12}$ powders.

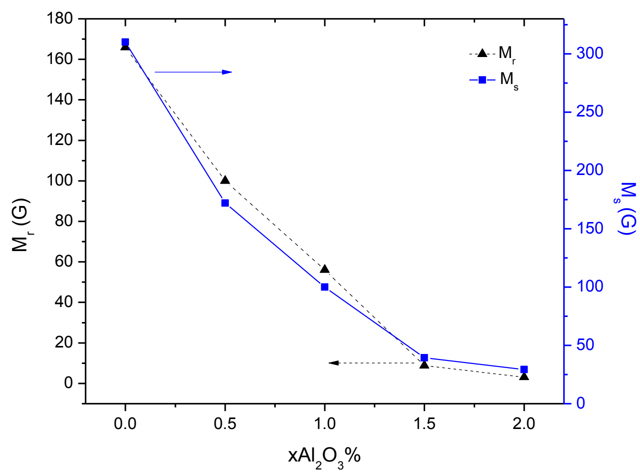

Figure 4: The effect of M_s and M_s values with variation in Al_2O_3 content.

Aluminium-substituted YIG, $Y_3Fe_{5-x}Al_xO_{12}$ (0.0 < x < 2.0), has been prepared from waste mill scale by using mechanical alloying technique. The density of the samples was measured using density meter and was observed to decrease as Al concentration increases. The magnetic properties such as remanence M_r, saturation magnetization M_s and coercivity H_c also show dependence on Al concentration. Remanence M_r and saturation magnetization M_s show a decreasing trend with increase in Al concentration. The reduction of magnetization for higher Al^{3+} concentration is due to the substitution of Fe^{3+} for Al^{3+} in octahedral and tetrahedral sites in substituted YIG and the consequent reduction of super exchange interactions in the lattice. Variation of T_c showed that Al substitution leads to the increase of the Curie temperature.

REFERENCES

[1] F. W. Aldbea, N. B. Ibrahim, and M. Yahya, (2014). "Effect of adding aluminum ion on the structural, optical, electrical and magnetic properties of terbium doped yttrium iron garnet nanoparticles films prepared by sol–gel method," *Appl. Surf. Sci.*, vol. 321, pp. 150–157.

[2] M. Pardavi-Horvath, (2000). "Microwave applications of soft ferrites," *J. Magn. Magn. Mater.*, vol. 215, pp. 171–183.

[3] R. D. Sánchez, J. Rivas, P. Vaqueiro, M. a. López-Quintela, and D. Caeiro, (2002). "Particle size effects on magnetic properties of yttrium iron garnets prepared by a sol–gel method," *J. Magn. Magn. Mater.*, vol. 247, no. 1, pp. 92–98.

[4] N. G. Imam and a. Hashhash, (2014). "Photoluminescence of γ-irradiation induced distortion on Ga based ferrospinel material to be used as γ-rays detector," *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 767, pp. 353–358.

[5] C. S. Kim, B. K. Min, S. J. Kim, and S. R. Yoon, (2003). "Crystallographic and magnetic properties of Y 3 Fe 5 À x Al x O 12," vol. 255, pp. 553–555.

[6] C. Suryanarayana, (2001). "Mechanical alloying and milling," vol. 46.

[7] R. S. Azis, M. Hashim, N. M. Saiden, N. Daud, and N. M. M. Shahrani, (2015). "Study the Iron Environments of the Steel Waste Product and its Possible Potential Applications in Ferrites," *Adv. Mater. Res.*, vol. 1109, pp. 295–299.

[8] R. D. Sánchez, C. a. Ramos, J. Rivas, P. Vaqueiro, and M. a. López-Quintela, (2004). "Ferromagnetic resonance and magnetic properties of single-domain particles of Y3Fe5O12 prepared by sol–gel method," *Phys. B Condens. Matter*, vol. 354, no. 1–4, pp. 104–107.