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STRUCTURAL AND MAGNETIC PROPERTIES OF ALUMINUM SUBSTITUTED YTTRIUM IRON GARNET VIA SOL-GEL SYNTHESIS

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ABSTRACT

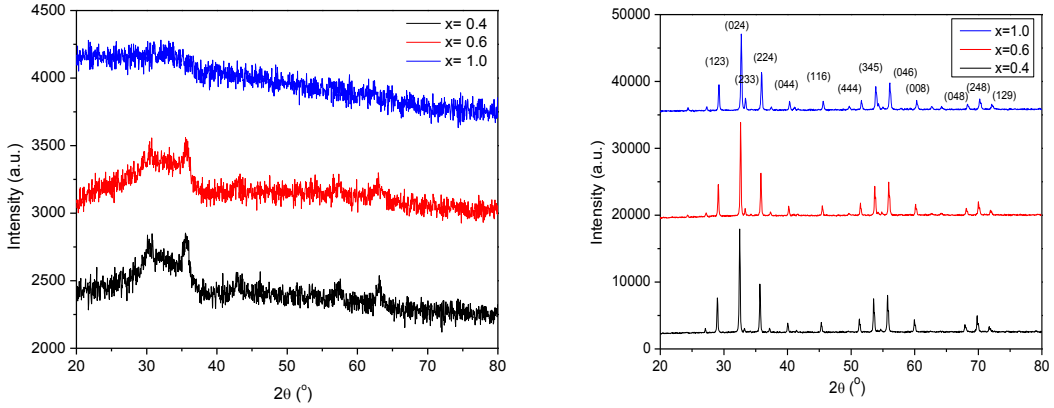
Aluminum-substituted yttrium iron garnet (Al-YIG) powders was synthesize by using sol-gel citrate nitrate combustion technique with different doping concentration (x = 0.4, 0.6 and 1.0). The $Y_{3-x}Al_xFe_5O_{12}$ samples were analyse of phase, structural and hysteresis by using X-ray diffraction (XRD), Fourier transform infra-red (FTIR) and Vibrating Sample Magnetometer (VSM).The powder resulted a single phase nanostructured garnet was formed. Room temperature saturation magnetization M_s and coercivity of Al-YIG powders decreased as a function of increasing Al content. The samples has a room temperature M_s of 9.2 emu/g and decreased to 1.5 emu/g. Coercivity H_c value decreases from 71.7 G to 51.4 G.

Yttrium iron garnet ($Y_3Fe_5O_{12}$, YIG) is an important materials due to its high resistivity and their application in microwave devices [1]. Its excellent microwave properties including relatively low magnetization, extremely narrow line width and low dielectric loss behavior make it suitable for microwave devices such as circulators, isolators, and phase shifters [2-8]. YIG also important for magneto-optic applications in telecommunications especially in high frequency devices [2]. The rare-earth doped YIG are very useful in microwave communication devices such as circulators, isolators, gyrators and phase shifters due to its narrow ferromagnetic resonance and low dielectric loss. Besides that, YIG has low hysteresis loss at room temperature that are useful in radio-electronics. YIG compounds either in the form of bulk, single crystal or thin film, have been used for many applications. YIG compound has cubic structure (space group *Ia3d*) and every cell will contain eight $Y_3^{3+}\ Fe_5^{3+}O_{12}$ molecules. Y^{3+} ions has large ion radius and exhibit in dodecahedral sites which have larger space [2]. There are several methods were introduced which can be employed for the preparation of YIG on large ceramic or metal surfaces such as co-precipitation method, sol-gel, spray pyrolysis, slurry coating and atmospheric plasma spraying. The citrate–nitrate gel combustion process is a good method to produce nanocrystalline oxide powders and has the advantage of using inexpensive precursors. In citrate–nitrate gel methods, the role of citric acid is to form a complex with metal ions which is expected to reduce segregation of cations. Citrate nitrate gel combustion offers excellent opportunities to synthesize large-scale ceramic nanopowders in an economically viable way. In this recent study, we report the mechanical alloying technique to prepared nanoparticles $Y_{3-x}Al_xFe_5O_{12}$. Phase study, microstructure and magnetic properties of Al substituted YIG powders were studied as a function of Al contents.

The starting raw materials used are yttrium nitrate hexahydrate ($Y(NO_3)_3.6H_2O$), aluminum nitrate nonahydrate ($Al(NO_3)_3.9H_2O$), and Iron (III) nitrate ($Fe(NO_3)_3{\cdot}9H_2O$). ($Al(NO_3)_3$ are used as substituted material into this formula $Y_{3-x}Al_xFe_5O_{12}$ with different concentration (x = 0.4,0.6 and 1.0). The nitrate salts are dissolved in iso-propanol, and the solution is placed on the magnetic hotplate. The solution was stirred using magnetic stirres at 40 °C for 20 to 40 minutes until it dissolved. The citric acid was added into the mixture with the ratio of citrate-nitrate is 0.75. The solution was continuously stirring at constant speed and increases up the temperature to 70 °C to form a brownish gels. The precursor solution became thick and viscous after evaporating to remove about 80 vol. % of its water. After approximately 30 min, the gel began to foam, swell and self-combusted to produce greenish-black powder. The hot plate temperature was then increases to 300 °C and the alumina crucible was covered with an alumina lid to allow hot gases to escape with dark brownish fume was formed. During combustion the temperature was recorded by a thermocouple and the maximum temperature reached was about 280 °C. The combustion-formed powder was then grind and crush using a mortar and the powders are calcined at different temperatures in air using a high temperature box furnace. Crystallography and composition were investigated by using X-ray diffraction (XRD). X-ray powder diffraction data were collected in a Philips Expert PW3040

diffractometer operating at 40 kV/30 mA using Cu Kα (0.154 nm) radiation to generate diffraction patterns from the crystalline powder samples at ambient temperature over the range from 10° to 80°. Infrared spectra (280–4000 cm^{-1}) is recorded using an FTIR spectrometer (Perkin Elmer model 1650), with the samples pressed onto diamond-coated CsI pellets. Both the XRD and the FTIR results were used to establish the structural study of the prepared Al-YIG ferrite powder. Magnetic characterization of the YIG ferrite nanoparticles was performed by using a vibrating sample magnetometer (VSM) (Lake Shore 4700) at room temperature with a maximum magnetic field of 15 kOe.

Figure 1 (a) and Figure 1 (b) shows the XRD spectra for the precursor and heated at 1200 °C powder, respectively. The XRD spectra for the precursor (Figure 1(a)), shows no reaction and a broad peak is revealed, which does not have sharp diffraction patterns and is still amorphous phase. Also, precursor powder shows the formation of intermetallic as indicated by the low intensity peaks. For powder heated at 1200 °C (Figure 1(b)), the XRD peaks show relatively intense diffraction peaks of Al-YIG phase and could be indexed to a garnet phase (JCPDS 44–0228). The heated powder confirms the presence of Al-YIG ferrite with a cubic structure. The main peak for Al-YIG (x = 0.4) is located at 2θ (32.4851°). It shifts slightly toward higher diffraction angles where 2θ (32.6293°) for (x = 0.6) then 2θ (32.7374°) for (x = 1.0).



(a) (b)

Figure 1: X-ray diffraction patterns of $Y_{3-x}Al_xFe_5O_{12}$ with different concentration of Al sol-gel as powder (a) precursors and (b) heated powder

Figure 2 shows the hysteresis M (H) loops of Al-YIG powders that were measured at room temperature in the range of approximately –15 to +15 kOe. Hysteresis loops were plotted and the coercive field H_c and saturation magnetization M_s were recorded. The saturation magnetization M_s for Al-YIG samples are 9.3 emu/g, 2.7 emu/g and reduced to 1.5 emu/g, respectively. The decrease in saturation magnetization of these samples, compared to that of bulk material, depends on different parameters. In the sol-gel method, the heating rate of calcination is one of the most important parameters that can effectively increase or decrease the saturation magnetization. The coercivity H_c of Al-YIG particles continuously decreased with increase Al content due to the increase in particle size. This behavior is in good agreement with that expected for small particles and is related to the presence of different magnetic processes in the particles [9]. The low magnetization value exhibited by YIG nanoparticles was attributed to a non-collinear arrangement of magnetic spins [10].

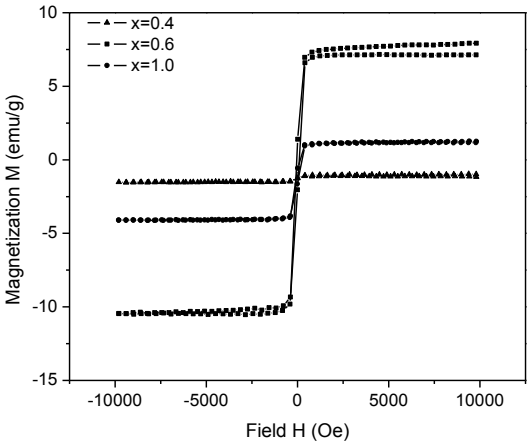


Figure 2: Room temperature M–H curves of combustion-formed of Al-YIG for x = 0.4,0.6 and 1.0

Figure 3 shows the FT-IR spectra analysis for Al-YIG with wave numbers between 280 and 4000 cm^{-1} for heated powders. Figure 3 shows all absorption peaks at 3427, 1633, and 602 cm^{-1}, corresponding to the stretching and bending vibrations of O–H, C=O and metal-oxygen vibrations, respectively. The FTIR spectrum shows very weak carbonate vibrations. The metal-oxygen vibrations at 602 cm^{-1} which are due to the lattice vibrational modes of the YIG unit cell [9].

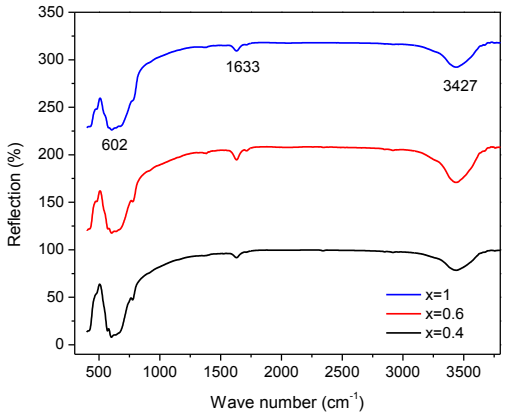


Figure 3: FTIR spectra of Al-YIG samples for x=0.4,0.6 and 0.10

In summary, the synthesis of sol-gel citrate nitrate combustion process produce high purity Al-YIG ferrites. Different ratio of aluminum substitution on YIG has been prepared for x = 0.04, 0.06 and 0.10 by using sol-gel technique. The XRD and FTIR results confirm the sol-gel process produce Al-YIG garnet structure and no trace other composition. The magnetic properties of YIG at different Al^{3+} substitution were recorded and show the room temperature saturation magnetization M_s and coercivity H_c are decreases as a function of increasing the Al content.

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