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Synthesis and Characterization of Nanocrystalline Yttrium Aluminum Garnet by Sol Gello Method

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Abstract—*The fabrication of transparent polycrystalline Yttrium Aluminum Garnet (YAG) is still a challenge, requiring the achievement of high purity, homogenous and nanosize material. In this work, Yttrium Aluminum Garnet (YAG) has been synthesized by sol- gello method using chloride precursor and agar-agar as gelling agent. The Influence of pH of solution, NH_4OH addition with stirring or putting solutions in closed container and calcination temperature on quality of YAG powder has been investigated. A variety of techniques, such as thermogravimetry (TG), X-ray diffraction (XRD) and scanning electron microscopy (SEM) were employed to characterize synthesized powders. These result show that hydrolysis in acid medium (pH = 6.0) show better crystalline size range (10-30 nm) and particle size (Less than 100 nm) than base medium (pH = 8.5) which show crystalline size (25-50 nm) and particle size (above 500 nm).Pure YAG phase obtained when calcined at 800°/2 hr in case of solution closed in container and 900°/2 hr with stirring.*

Keywords*: YAG, Agar, XRD, Agglomeration.*

1. INTRODUCTION

Yttrium aluminum garnet ($Y_3Al_5O_{12}$, YAG) exhibits high chemical stability and unique optical properties. Therefore, it has been extensively used as a host material for solid-state lasers, phosphors, and LED (light emitting diode) applications [1-4].Another important property that is being explored for YAG is its infrared transparency in the range of 3-5 μm. This makes it useful for such applications as missile domes [5] In addition, YAG ceramics provide a variety of potentially advanced structural materials owing to their high creep and oxidation resistance at high temperature and their low heat conductivity [6-8]. For the fabrication of high quality YAG-based materials, it is essential to prepare high purity powders, with fine particle size along with good dispersion and high sintering activity.

YAG powders are traditionally prepared with a solid-state reaction using yttria (Y_2O_3) and alumina (Al_2O_3) compounds. With this method, the precursors are mechanically milled followed by high temperature treatment typically higher than 1500 °C for prolonged time to eliminate intermediate phases [8-10]. Solid-state reaction method is a relatively simple way to prepare YAG powders. However, the incorporation of some impurities is unavoidable during ball milling. In addition, prolonged heat treatment at high temperature leads to hard agglomerates and extensive grain growth. In recent years, several wet chemical methods such as precipitation [11-14], spray pyrolysis [15,16], sol-gel [17,18], solvo-thermal synthesis [19,20] combustion synthesis [21,22] have been developed to produce YAG nanopowders.

The sol-gello method was developed to ensure that a more homogeneous gel can be made. In this work of synthesizing YAG, Yttrium chloride hexahydrate ($YCl_3.\ 6H_2O$) and Aluminum chloride hexahydrate ($AlCl_3.6H_2O$) as raw material and the gelling agent is agar. During hydrolysis, the agar "freezes" the metal ions in place preventing them from segregating during the hydrolysis process. The gelling agent also helps to control the particle size. Having nanosize precursors allow the low preparative temperatures and better sintering result.

2. EXPERIMENT PROCEDURE

Yttrium chloride hexahydrate ($YCl_3.\ 6H_2O$) and Aluminum chloride hexahydrate ($AlCl_3.6H_2O$) as raw material, the gelling agent is agar and Ammonium hydroxide to maintain final pH of solution.

First,concentrated solutions were obtained by dissolving $YCl_3.\ 6H_2O$ and $AlCl_3.6H_2O$ in distilled water, these solutions were mixed homogeneously with appropriate molar ration 3:5 according to the stoichio- metry of YAG(Y3Al5O12). Next, 10 w/w % agar solution were prepared by mixing agar powder in hot water (80°C) with continuous stirring. Concentrated solution mixes with agar solution and stirring is continuous. Next, Mixed solution was put in water bath maintained at 10 ° C for 6hr till transparent yellow gel was obtained.

Further NH_4OH addition to diffuse NH_3 Ions was approached by two methods. One was NH_4OH addition by dropwise in gel with stirring till proper pH obtains then. Another was the gel was placed inside an ammonium hydroxide chamber. The ammonium hydroxide chamber was a closed container with aqueous ammonium hydroxide inside. As the hydrolysis occurred, the gel transformed from a transparent light yellow color to white.

Once hydrolysis was completed, the white solid was removed from the chamber and dried at 80°C for one day. Next, dried powder grind in mortar pastel and calcined at temperature 700°C, 800°C, 850°C, 1000°C for 2 hr .Table 1 shows specification of various samples prepared for study. B refer to Base Medium, A refer to Acid

Table 1: Various Samples at different parameter

B1-700 (Sample 1)	pH = 8.3, first approach after water bath treatment, 700°C
B1-800 (Sample 2)	pH = 8.3, first approach after water bath treatment, 800°C
B1-900 (Sample 3)	pH = 8.3, first approach after water bath treatment, 900°C
B2-800 (Sample 4)	pH = 8.3, Second approach after water bath treatment, 800°C
A1-800 (Sample 5)	pH = 6.0, first approach after water bath treatment, 800°C
A1-900 (Sample 6)	pH = 6.0, first approach after water bath treatment, 900°C
A2-800 (Sample 7)	pH = 6.0, Second approach after water bath treatment, 800°C

3. CHARACTERIZATION METHOD AND INSTRUMENT

Thermal gravimetric analysis (TG) of the precursor was made on the Thermal Analysis (STA-800C, CSIO, Chandigarh) with a heating rate of 10 °C/min and upper temperature limit of 800°C. The phase analysis of the synthesized powders was accomplished with X-ray diffraction (XRD, D/Max2500, SAIF Lab Panjab University,Chandigarh) at 40 kV and 200 mA using Cu K_ radiation (_=0.15406 nm) in the scanning range of 5°–80°. Powder particle morphology analysis was carried out by scanning electron microscopy (FESEM,,ISSER Mohali).

4. RESULT AND DISCUSSION

4.1 TG Analysis

Fig. 1 shows the TGA profile for the pH = 8.3 uncalcined YAG sample. The first step was due to evaporation of water and was followed by sublimation of ammonium chloride in the second step. Next, the agar was burned out, a process that was completed at 530ºC Once the extraneous species were removed, the amorphous yttrium aluminum oxide/hydroxide began to crystallize into YAG from 575°C.Profile shows that complete YAG form at temperature 900°C.

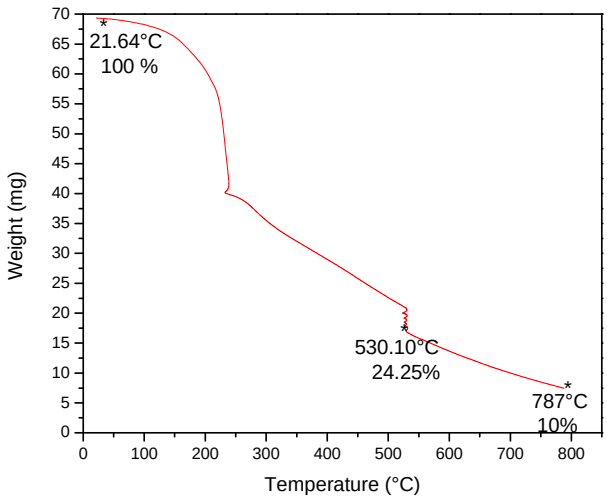


Fig. 1: TGA Analysis of Uncalcined Sample

4.2. XRD Analysis

The XRD was done to calculate crystalline size using Scherrer's equation, and to confirm the time and temperature of calcination for complete crystallization of YAG. The obtained peaks are compared with the standard JCPDS card no. of $Y_3Al_5O_{12}$ (YAG, JCPDS card no 79-1840), $YAlO_3$ (YAP, ICPDS card no. 74-1334) and $Y_4Al_2O_9$ (YAP, ICPDS card no. 34-0368).

Fig.2 show XRD peaks of various samples obtained. These result show that powder is amorphous in sample 1(at 700° C) in both approach, peaks obtained in sample2 (800°C) with first approach show YAG peaks with intermediate phase of YAP which was remove completely in Sample 3 (900°C). Peaks Obtained in sample 4 (800°C) with second approach show pure YAG peaks has crystalline size 30-50 nm.

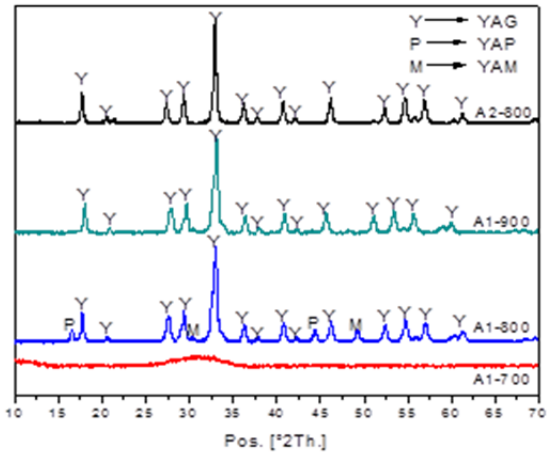


Fig. 2: XRD Pattern of calcined powder at various temperatures at pH = 6.0

Fig 3 show XRD peaks of sample prepared in acid medium showed better results. Sample 5 XRD peaks show that pure YAG phase not obtained at this temperature peaks of intermediate phase of YAP and YAM also shown in graph. Peaks Obtained in sample 6 (950°C) with second approach show pure YAG peaks has crystalline size 10-30 nm. Peaks Obtained in sample 7 (800°C) with second approach show YAG peaks with intermediate peaks of YAP but sample 8 show pure YAG Peaks.

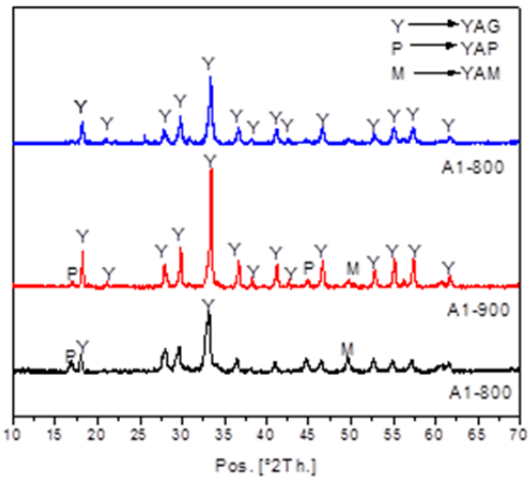
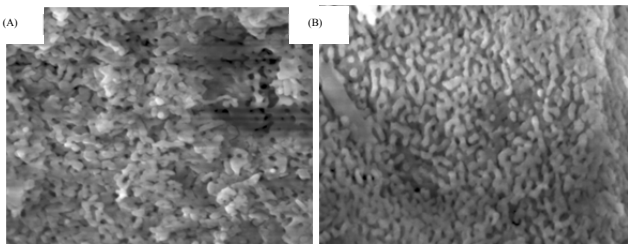


Fig. 3: XRD Pattern of calcined powder at various temperatures at pH = 8.3

Powder obtained by second approach showed pure YAG phase obtained at lower temperature but higher crystalline size.

4.3 FESEM Analysis

Fig. 4 shows FESEM image at 50000X of various sample synthesized at different pH, Calcination temperature and method. This result show that average particle size in in acid medium (pH = 6.0) was 225 nm as compare to base medium (pH = 8.3) which was above 500 nm.This shown that amount of agglomeration was less in acid medium than base medium.



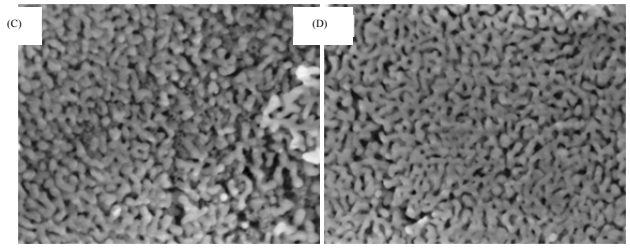


Fig. 4: FE-SEM Image of various calcined powder at 50000 X (a) Sample 3 (b) Sample 4 (c) Sample 6 (d) Sample 7

Lowest Particle size in sample 6 and sample 7 is less than 20 nm but because of agglomeration average particle size is above 100nm.This is because of small particle has higher tendency to agglomerated so if we able to prevent ions attraction or motion of sol by increases agar concentration in gel.

4.4 DLS Anaysis

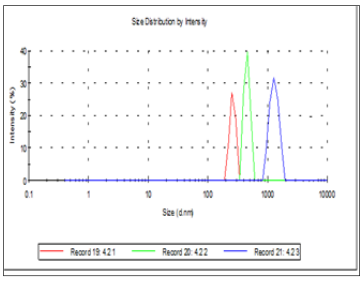
Fig. 6 shows particle size distribution of sample6 and sample 7. DLS confirm the results obtained from FESEM. This results shows that average particle size is less in acid medium than base medium. Processing through first approach show lesser particle size that second approach.

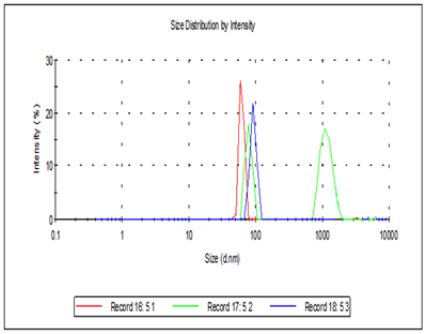
Table 2: Average crystalline size and average particle size of various samples

YAG Powder	Average crystalline size(By XRD)	Average Particle size (By DLS)
Sample 3	28 nm	1120 nm
Sample 4	35 nm	2235 nm
Sample 6	30 nm	545 nm
Sample 7	25 nm	220 nm

Table 2 also shows that powder obtained by first approach has lesser particle size than second approach but higher crystalline size. In each sample obtained YAG powder is nanocrystalline and nanoscale particle size in case of acid medium

Size (nm)	Intensity
220.2	3.8
255	9.1
295.3	6.3
342	0
396.1	9.2
458.7	13.2





Size (nm)	Intensity
50.75	0.3
58.77	8.7
68.06	9.3
78.82	8.5
91.28	10.5
105.7	3.4

Fig. 6: DLS result (a) Sample 6 (b) Sample 7

5. CONCLUSION

The fabrication of nanocrystalline YAG powder was obtained by sol Gello process. According to results of TGA, XRD pure YAG phase begin to form at lower temperature 900°C is case of first approach and 800° C in case of second approach which was lower than reported previously. FESEM and DLS result showed that in case of acid medium powder have low agglomeration and success to obtained nanosacle powder at 900°C through first approach and at 800°C through first approach.

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