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Cathodic Electrodeposition and Characterization of YAG Nanostructure: Effect Current Density on the Morphology

M. Hosseinifard [a,*], K. Ahmadi[a], A. Badiei[b]

[a] *Department of Semiconductors, Materials and Energy Research Center, P.O. Box 14155-4777 Tehran, Iran*
[b] *School of Chemistry, College of Science, University of Tehran, Tehran, Iran*

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ABSTRACT

YAG (Yttrium Aluminum Garnet) was successfully prepared through cathodic electrodeposition process by applying different current densities to the mixture of YCl_3 and $AlCl_3$ solution (water/ethanol 1:1 volume ratio). Hydroxide precursors were cathodically grown on the cathode surface (at different current densities) and then the obtained hydroxide powder was heat-treated at 1100 °C for 2 h. The oxide products were characterized by XRD, FTIR, DSC-TGA, SEM and EDAX techniques. The effect of applied current density on the morphology and particle size of YAG nanostructures were investigated through SEM images. Experimental results showed that the cathodic electrodeposition followed by heat-treatment can be used as a facile method for preparation of YAG nanostructures with different morphologies.

1. INTRODUCTION

Due to their excellent chemical and physical stability, creep resistance and optical transparency, yttrium aluminum garnets have been studied as an important functional material [1-2]. YAG has been widely utilized for solid-state lasers and in luminescence applications, especially when it is doped with the transition metals or lanthanide ions [3-5]. It is also applied in fiber optic telecommunication systems. The Y_2O_3-Al_2O_3 system contains three stable phases with various Al^{+3}-Y^{+3} ratios, i.e. monoclinic yttrium aluminum ($Y_4Al_2O_9$, YAM), perovskite yttrium aluminum ($YAlO_3$, YAP) and $Y_3Al_5O_{12}$ (YAG). YAG powder is conventionally produced via a solid-state reaction using individual component oxides. In order to achieve the desired composition, this method requires repeated mechanical mixing and extensive heat treatment at temperatures above 1600 °C [3-5]. Additionally, the homogeneity and purity of the YAG powder cannot be readily controlled. Furthermore, the conventional method comprises distinct disadvantages including large and/or inappropriate particle size, stoichiometry impurities and formation of undesirable phases. The high sintering temperature is necessary for crystallization, which may cause some defects and impurities [6- 7]. Therefore, various synthesis methods have been developed to fabricate the YAG-based material in order to overcome the drawbacks of the solid-state reactions. The final grain structure in dense polycrystalline YAG can be carefully controlled due to the potential and properties of YAG nanopowders. Furthermore, nanosized YAG particles provide higher brightness in phosphor applications [8-10]. Numerous methods can be employed for the preparation of YAG, for instance, solid-state [11], hydrothermal [12], solvothermal [13], sol-gel [14], co-precipitation [15] and sol-gel combustion [16]. The development of a simple and low-cost method has been proposed in order to synthesize YAG while eliminates the disadvantages of the aforementioned methods. To this end, we have proposed the electrochemical method as a novel and unique method. The main advantages of the electrochemical method are simplicity and flexibility. It is also a suitable method for the preparation of nanostructured metal oxides and hydroxides [17-19]. In the electrochemical method, a metal hydroxide is prepared as a precursor by the electrodeposition process. Then, it is converted into metal oxide via heat treatment. The rate of electrochemical reactions, nucleation and growths can be controlled through changing

* *Corresponding Author Email: m.hosseini@merc.ac.ir (M. Hosseini)*

experimental parameters such as current density, potential, bath temperature and reaction medium. These parameters are adjusted in order to design new products with the desired crystallinity and morphology [20]. According to the literature, a few limited numbers of research works have been reported the electrochemical synthesis of YAG [21-22]. In this work, YAG nano-powder was prepared via a cathodic deposition method followed by heat-treatment. Moreover, the effect of different current densities (0.5, 1, 2 and 5 $mAcm^{-2}$) at 25 ^oC on the morphology and particle size of YAG nanostructure is also examined.

2. EXPERIMENTAL PROCEDURE

2.1. Chemicals

Yttrium chloride hexahydrate (YCl_3 .$6H_2O$) and anhydrous aluminum chloride ($AlCl_3$) (Aldrich) were dissolved separately as raw materials without further purification in ethanol/ distilled water 1:1 solution.

2.2. Synthesis procedure

The cathodic electrodeposition was performed in an electrochemical cell including a cathodic steel substrate (316 L, 100×50×0.3 mm) centered between two parallel graphite anodes and the required amount of electrolyte (YCl_3 (0.005 M), $AlCl_3$ (0.0083 M) solution. The steel substrate was electropolished prior to each deposition [17-19]. Deposition experiments were carried out at a constant (differing at each experiment) current density for 2 h at a temperature of 25^oC. The obtained deposit was scraped from the steel electrode and dried at room temperature for 5 h. In order to prepare the oxide product, the hydroxide sample was heated at 1100 °C in the air for 2 h.

2.3. Characterization

Crystal structure of the prepared samples was investigated using X-ray diffraction (XRD) analysis. FTIR spectrum of the sample was obtained using a Bruker Vector 22 Fourier transformed infrared spectroscope with samples in a KBr wafer at ambient temperature, in the range of 400-4000 cm^{-1}. Morphology of the synthesized YAG nanoparticles were studied using a scanning electron microscope (SEM, LEO 1455 VP). Thermogravimetric analysis of deposited hydroxide was performed using a TGA/DSC1- METTLER TOLEDO-STARe in oxygen flow between 25°C -1100°C with a heating rate of 10 °C/min.

3. RESULTS AND DISCUSSION

Fig. 1 shows the XRD patterns of YAG powders obtained via electrodeposition method in different current densities and calcination at 1100 ^oC. All of the diffraction peaks are fully consistent with pure cubic phase (space group la3d) crystalline YAG (JCPDS card No.033-0040) [11-13]. Results showed a change in current density from 0.5 to 5 $mAcm^{-2}$ which does not affect the crystal structure. In fact, the applied current density has a considerable effect on the kinetic of base electrogeneration reactions, nucleation and growth of the deposit. These changes can be exhibited in the particle size and morphology of substance. In the cathodic electrodeposition process, controlling the nucleation, growth and the rate of electrochemical reactions through the experimental parameters like current density plays a key role in designing new morphologies along with the desired crystallinity.

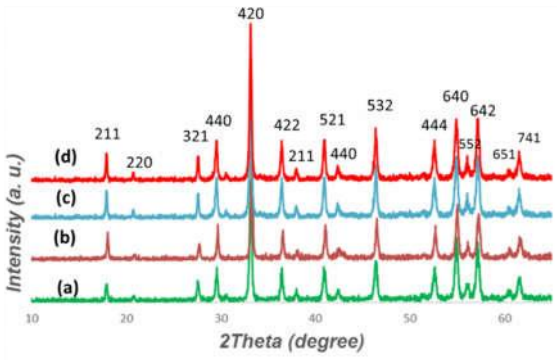

Figure 1. XRD patterns of the YAG powders prepared (a) i= 0.5 $mAcm^{-2}$, (b) i= 1 $mAcm^{-2}$, (c) i= 2 $mAcm^{-2}$ and (d) i= 5 $mAcm^{-2}$

The particle size and morphology of the synthesized YAG nanoparticles were characterized by SEM (Fig. 2 (a-d)). These different morphologies demonstrate the effect of applied current densities on the cathodic electrodeposition of YAG. It can be observed that the grain size is uniform and particle texture morphology is very fine (Fig. 2 (a)). It should be noted that it is not feasible to characterize the morphology and exact size of prepared particles from the SEM images. Crystals significantly grow when the current density is increased from 0.5 to 5 $mAcm^{-2}$ (see Fig. 2). Increasing current density leads to higher reaction kinetics and causes a raise in the production rate of OH^- ions on the surface of the cathode during the electrochemical reaction. In the cathodic electrodeposition process, H_2O plays a determinative role in the production of OH^-. Nucleation and growth of the nuclei can be developed with a faster velocity. In addition, releasing H_2 gas from the surface of the cathode in the double layer during the reaction results in porous products [23- 26]. Therefore, the average particle size increases with increasing the current density. Fig. 2(e) shows the representative EDAX pattern for YAG sample. The presence of Al, Y and O peaks determine YAG composition in the obtained sample [27].

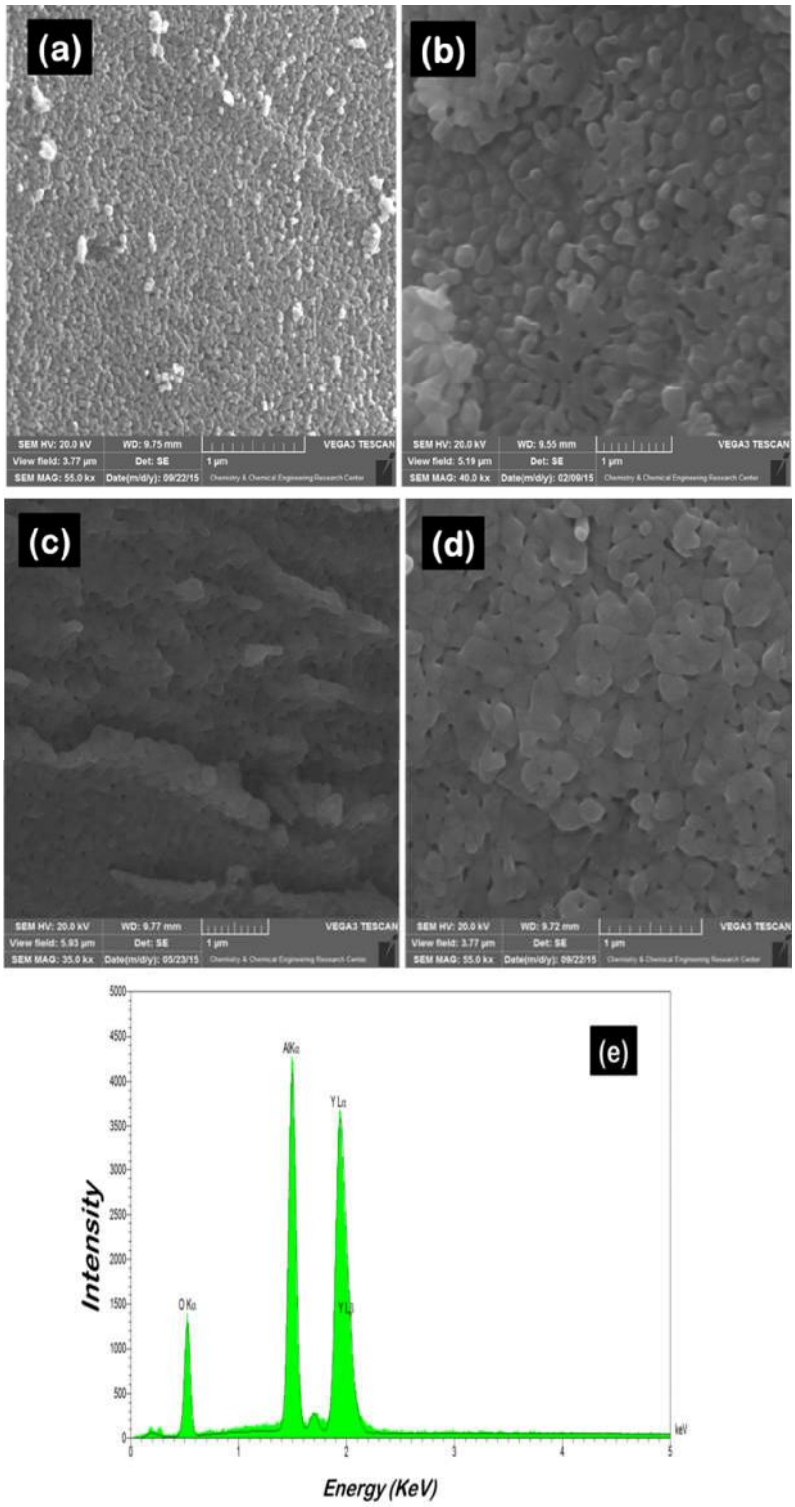

Figure 2.(a-d) SEM micrographs and (e) EDAX analysis of the prepared YAG samples (a) i= 0.5 $mAcm^{-2}$, (b) i= 1 $mAcm^{-2}$, (c) i= 2 $mAcm^{-2}$, (d) i= 5 $mAcm^{-2}$

The FTIR spectra of YAG powders which were prepared at different current densities and calcinated at 1100 °C, are shown in Fig. 3. The bands at 722 cm^{-1} and 790 cm^{-1} correspond to the stretching vibrations of Al-O and the other bands at 688 cm^{-1}, 570 cm^{-1} and 521 cm^{-1} associate with the Y-O metal–oxygen vibrations [12-15, 22]. These bands are characteristic peaks of the YAG structure. It can be concluded that the change in current density from 0.5 to 5 $mAcm^{-2}$ does not affect the crystal structure of YAG powder and FTIR analysis confirm the XRD results.

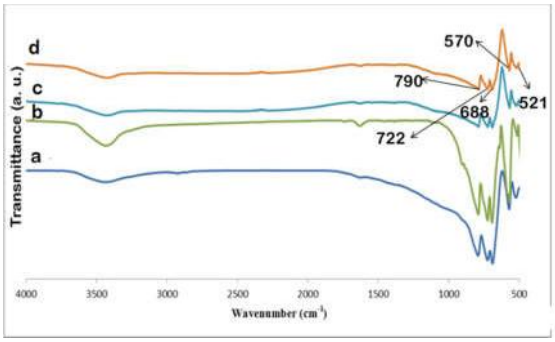

Figure 3. IR spectra of the prepared YAG powder calcinated at 1100 °C (a) i= 0.5 $mAcm^{-2}$, (b) i= 1 $mAcm^{-2}$, (c) i= 2 $mAcm^{-2}$, (d) i= 5 $mAcm^{-2}$

To investigate the thermal behavior of the deposited product, DSC-TG analysis was performed through a heat treatment at 25-1100 °C (Fig. 4). Two distinct endothermic peaks at 150 °C and 850 °C are observed in the DSC curve, which are related to the physical and chemical variations of the sample at these temperatures. Correspondingly, TG curve shows two stages of weight loss. The first region, including a 35%weight loss, is related to the dehydration of adsorbed and structural water and happened between 25°C -500 °C, [19, 22, 26]. The second region, which is happened at 500 °C - 920 °C and included a 39% weight loss during the heat treatment process, initiates from the formation of YAG powder.

4. CONCLUSION

YAG nanostructures were successfully prepared in two steps: First, the hydroxide precursors were cathodically deposited on the steel substrate from the solution of raw materials in the bath at different applied current densities; second, they were heat-treated. The powder was scraped off the substrate and subjected to further analyses by XRD, FTIR, DSC-TG, SEM and EDAX techniques. Results show that the obtained oxide product is composed of the crystalline YAG. Morphological characterization and structural analysis of the final product showed that the current density plays a vital role in the electrodeposition process. . It can be concluded that the change in current density from 0.5 to 5 $mAcm^{-2}$ does not affect the crystal structure of YAG powder. It can be stated that the current density is an effective and easy method for preparation of nanostructures of yttrium-aluminum garnet with different morphology and particle size in the cathodic electrodeposition method.

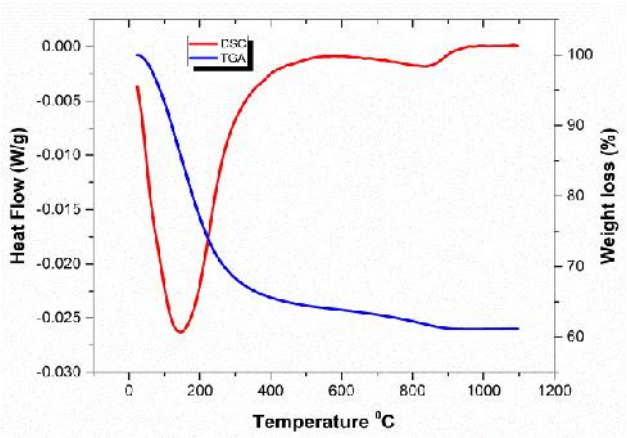

Figure 4. Thermogravimetric analysis (DSC-TGA) of the deposited hydroxide during heat treatment

5. ACKNOWLEDGEMENT

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