Synthesis of Nano-Sized Yag:Ce3+ by Sol- Gel Method

Loli Yusastri, I Made Joni[1], C. Panatarani[1]

Abstract—**We have been successfully synthesized YAG-:Ce3+ nanoparticle (60 nm in sized) having high crystalinity by using low temperature sol gel method. Due to additional of polyethylene glycol into the precursor source has been avoid inter particle hard-agglomeration. The influence of difference heating process on the crystallinity of YAG:Ce3+ was studied by means of XRD characterization. The highest crystallinity of YAG:Ce3+ was achieved via two step heating at 900 and 1100oC.**

Keywords—**Sol–gel process, YAG:Ce3+, Crystallinity**

I. Introduction

White light has been widely used in markets such as traffic lights, automobile brake lights, lightings for hazardous signs, backlights of colored liquid crystal displays and mobile phones[1]. Some effort have been done in order to be able to produce a white light LED which is one of the most expected feature of light. YAG:Ce3+ is the most excellent phosphor satisfactorily applied in white phosphor-based LED to enter into commercial market. Yttrium aluminum garnet activated with trivalent cerium (YAG:Ce3+) has been an efficient phosphor to convert the blue LED radiation into a broaden yellow emission band. The yellow emission from YAG:Ce3+ is intense enough to complement the residual blue light which escapes through the phosphor in order to produce a white light. It is well known that Y2O3–Al2O3 system exists in three different crystal phases: YAlO3 (YAP with a perovskite structure),Y4Al2O9 (YAM with a monoclinic structure) and Y3Al5O12 (YAG with a cubic garnet structure)[2]. YAG is much more stable than the other two intermediate phases in conventional solid-state reactions [3]. Therefore, high temperature annealing with repeated grinding and milling is required for preparing pure YAG by solid-state reaction. A number of investigations proved that the wet chemical methods could decrease the annealing temperature efficiently.[4], [8]

To obtain ultrafine YAG powders at relatively low temperature, we use the wet chemical methods where sol-gel is extensively applied to allow the starting materials (precursor source) mixed at molecular level. However, the crystalinity of product is relatively poor. To improve the crystalinity one need more annealing, but this often result particles in hard-agglomeration. The purpose of this research is to find methods of preparation in order to get good crystalinity without particles agglomeration during the processes. In order to solve that problem above, we propose the use of water soluble polymer to avoid particles agglomeration while still having a good cristalinity due to exothermic reaction of polymer during the annealing process. The water soluble polymer will be added to the precursor source before the annealing process and it is expected that water-soluble polymer behave as shield to prevent the particles from agglomeration. Besides, this method is relatively cheaper than other methods and also extremely applicable for practical large-scale production of phosphor assisted white LEDs.

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[1] I Made J. and C. Panatarani are with Department of Physics, Faculty of Mathematics and Natural Sciences, Universitay of Padjadjaran, Sumedang, West Java, INDONESIA.

II. Experiment

An alumunium triisopropoxide, (CH3)2CHO]3Al (99.99%, Kanto Chemical) has dissolved in ethanol (A solution). Yttrium nitrate hexahydrate, Y(NO3)3.6H2O and Ce(NO3)3.6H2O (99.99%, Kanto Chemical) have dissolved in aquades (B solution). B solution has been added into A solution to give a sol-gel reaction. The molecular ratio of Y:Al:Ce are fixed at 2.97:5:0.03. The water soluble PEG has added to the gel before the annealing process to prevent the particles from agglomeration. The heating process to intensify the particles crystallinity was varied as:

1. Sample I Heating at 900^{o}C for 2 hours.
2. Sample II Two steps heating for two hours at 900 ^{0}C and 1100 ^{0}C.
3. Sample III Heating at 1100 ^{o}C for two hours.
4. Sample IV Simultaneously heating at 900 ^{0}C (2 hours) followed by 1100 ^{0}C for 2 hours.

The phase purity, composition and morphology of YAG:Ce3+ particles will be studied by means of x-ray diffraction (XRD, Rigaku, Geiger Flex Difraksi Bragg) and field-emission electron microscopy (FE-SEM S5000, Hitachi, Tokyo) operated at 20 kV.

III. Result and Discussion

YAG:Ce3+ prepared particles has yellow in appearance due to activation of Ce3+ in YAG particles. The morphology of prepared particles is shown by Figure 1. Figure 1a is the particles prepared at 900 ^{0}C for 2 h and Figure 1b is the particles prepared at two step simultaneous heating at 900 ^{0}C 2h and 1100 ^{0}C for 2h. From the Fig. 1a, it is clearly shown that the morphology of particle having ~60 nm in sized is not spherical. Extended heating at 1100 ^{0}C for 2h caused the particles agglomeration (Figure 1b). PEG as a chelating agent completely evaporates at high temperature (>900 ^{0}C), therefore agglomeration was obvious. Figure 2 shows the XRD pattern of prepared particles with JCPDS No. 33-40 as reference. Figures 2a-2d are XRD pattern of particles prepared at

900 ^{0}C for 2h, two step heating (at 900^{0}C and 1100 ^{0}C), 1100 ^{0}C and simultaneously heating (900 ^{0}C, 2h and 1100 ^{0}C, 2h) respectively.

XRD pattern of Fig.2a show a broad pattern around 2è =30o indicated the formation of YAG structure. However, heating process at 900 ^{0}C for 2h is not adequate for fully formation of YAG crystal. The YAG crystal was increasingly formed by increasing the heating temperature (Figs. 2b-2d).

The utilization of higher temperature (>900 ^{0}C for 2h) and extended heating strongly improved the crystallinity of YAG (Figs 2b-2d). However, temperatures used in this research still did not adequate for create YAG in pure phase. YAP phase still remain in that of YAG.

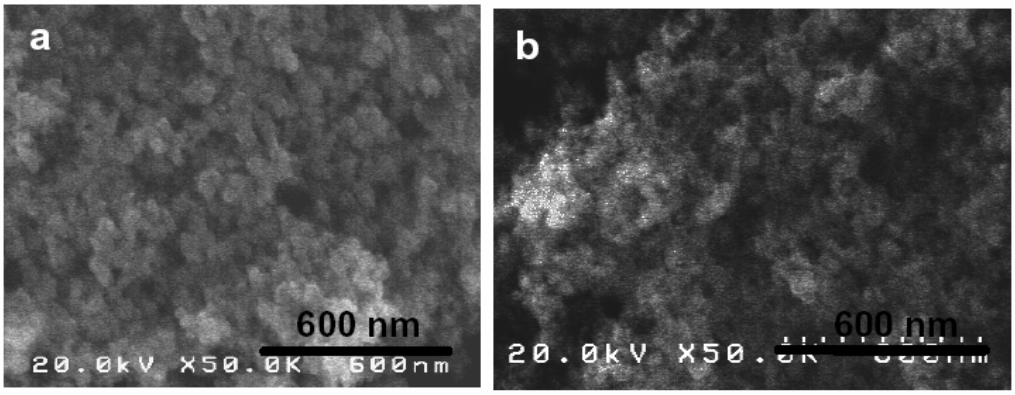

Fig.1. FE-SEM images of particles prepared at different heating process. (a) one step heating at 900 ^{0}C for 2 h and (b) simultaneously heating at 900^{0}C for 2h and 1100 ^{0}C for 2h

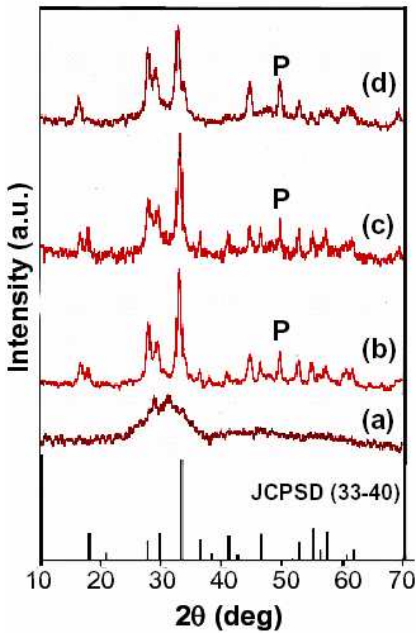

Fig. 2. XRD pattern of YAG:Ce3+ prepared at different heating process. (a) sample I, (b) sample II, (c) sample III and (d) sample IV. (P indicated YAP phase)

IV. Conclusions

Heating process which parameterizes by both duration and heating temperature is a crucial factor in YAG phase formation. Our investigation result shows that Additional polyethylene glycol could prevent hard agglomeration and two step heating process could produce good crystallinity and small size YAG particles.

V. Acknowledgment

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VI. References

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