

Glycothermal Synthesis and Characterization of YAG:Ce³⁺ Nanophosphors

Tetsuhiko Isobe*

Department of Applied Chemistry, Faculty of Science and Technology, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama 223-8522, Japan

isobe@aplc.keio.ac.jp

A glycothermal reaction is a thermal reaction in a sealed pressure reactor using glycol as a solvent. Inoue's group has developed glycothermal synthesis of several inorganic compounds such as rare earth aluminum garnets [1]. They reported that yttrium aluminum garnet (YAG) nanoparticles can be prepared from yttrium acetate and aluminum isopropoxide in 1,4-butanediol (BD), HO(CH₂)₄OH, at temperatures as low as 300 °C [2]. The mechanism on this glycothermal reaction is based on the following three steps: (i) alkoxy exchange between aluminum isopropoxide and 1,4-BD produces a glycoxide precursor, >Al-O-(CH₂)₄OH; (ii) the exchanged 1,4-BD cyclizes and the resulting protonated tetrahydrofuran detaches to form >Al-O⁻; (iii) >Al-O⁻ reacts with yttrium acetate to form Al-O-Y bond [1-3]. The cleavage of the O-C bond of >Al-O-(CH₂)_nOH is easiest at n=4 at the step (ii). The use of 1,4-BD therefore plays significant roles in the formation of crystalline YAG. Furthermore acidic and basic properties of aluminum and yttrium oxides, respectively, are associated with this mechanism.

We have intensively investigated glycothermal synthesis of YAG:Ce³⁺ nanoparticles and their structural and optical properties [4-9]. Glycothermal synthesis has several advantages with respect to the preparation of nanophosphors: (i) it is possible to crystallize by autoclave treatment at remarkably lower temperatures as compared to a conventional solid-state reaction; (ii) surface passivation by coordinating glycol solvents and acetate ions enables nanosizing in addition to suppression of Ostwald ripening due to lower solubility of YAG:Ce³⁺ in glycol, which is entirely different from hydrothermal synthesis in water; (iii) it is possible to well-disperse nanoparticles, which is important to produce transparent optical films.

One of characteristic optical properties of nanoparticles is transparency. Since the light scattering intensity depends on a particle diameter, d , to the sixth power, nanosizing decreases the light scattering loss. Nanoparticles with $d \leq 50$ nm can be dispersed in a matrix transparently to the eye because of the low scattering intensity of visible light. We therefore produced a color conversion film from YAG:Ce³⁺ nanoparticles and characterized its film. The back side of the film was excited by blue light and the intensity of photoluminescence (PL) at 530 nm corresponding to the 4f-5d transition of Ce³⁺ was measured in the front side of the film. The PL intensity of the film is proportional to the film thickness. Thus, we have demonstrated that YAG:Ce³⁺ nanoparticles dispersed in the film have low scattering loss of emission light. In this meeting we introduce a series of our works on YAG:Ce³⁺ nanoparticles synthesized by glycothermal method.

References

- [1] M. Inoue, H. Otsu, H. Kominami, and T. Inui, *J. Alloys Compd.*, 226, 146 (1995).
- [2] M. Inoue, H. Otsu, H. Kominami, and T. Inui, *J. Am. Ceram. Soc.*, 74, 1452 (1991).
- [3] M. Inoue, T. Nishikawa, H. Otsu, H. Kominami, and T. Inui, *J. Am. Ceram. Soc.*, 81, 1173 (1998).
- [4] R. Kasuya, T. Isobe, H. Kuma, and J. Katano, *J. Phys. Chem. B*, 109, 22126 (2005).
- [5] R. Kasuya, T. Isobe, and H. Kuma, *J. Alloys Compd.*, 408-412, 820 (2006).
- [6] R. Kasuya, A. Kawano, T. Isobe, H. Kuma, and J. Katano, *Appl. Phys. Lett.*, 91, 111916 (2007).
- [7] R. Asakura, T. Isobe, K. Kurokawa, T. Takagi, H. Aizawa, and M. Ohkubo, *J. Lumin.*, 127, 416 (2007).
- [8] Y. Kamiyama, T. Hiroshima, T. Isobe, T. Koizuka, and S. Takashima, *J. Electrochem. Soc.*, 157, J149 (2010).
- [9] T. Isobe, *ECS J. Solid State Sci. Technol.* 2, R3012 (2013).