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Preparation of Monodisperse Luminescent Particles of $Y_{2-x}Gd_xO_3$:Eu by Microwave-assisted Hydrolysis

A.S. Vanetsev[1,*], I.G. Chuvashova[1,2,†], O.M. Gaitko[1,2]

[1] *Kurnakov Institute of General and Inorganic Chemistry RAS, 31, Leninskii pr-kt, 119991 Moscow, Russia*
[2] *Lomonosov Moscow State University, 1, Leninskie Gory, 119992 Moscow, Russia*

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Development of energy-saving methods of synthesis of monodisperse particles of rare-earth oxides with improved luminescent properties is very vital task. In present work we've developed new method of microwave-assisted synthesis of $Y_{2-x}Gd_xO_3$:Eu solid solutions, performed a study of influence of Y:Gd ratio on morphology and luminescent properties of monodisperse oxide particles. We've established monotonous dependence of mean size of particles on Y:Gd ratio. On the other hand dependence of intensity of luminescence on Y:Gd ratio has a clear maximum.

Keywords: Microwave Synthesis, Monodisperse Particles, Luminescent Materials, Rare Earth Oxides

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1. INTRODUCTION

New methods of synthesis of monodisperse colloidal particles is one of the crucial steps on the way to production of various commercial materials including ink, cosmetics, different types of coatings and lubricants, paper products etc. One of the most important applications of monodisperse particles is luminescent coatings and highly efficient light sources. Among these luminescent materials, rare earth doped phosphors, for example, red luminescent phosphors doped with trivalent europium (Eu^{3+}) ions are of technological importance because they are widely used in color displays and fluorescent lamps [1]. Microcrystalline yttrium oxide doped with Eu^{3+} (Y_2O_3:Eu) represents a typical example and is considered to be one of the best red phosphors currently available [2]. These most promising luminescent materials at the same time require the largest amounts of energy during synthesis. Therefore, development of energy-saving methods of synthesis of monodisperse particles of rare-earth oxides with improved luminescent properties is very vital task.

Microwave treatment possesses many advantages in comparison with conventional heating methods, including higher velocity of heating, homogeneity of heat distribution and lower power inputs. One of the main merits of microwave treatment, especially in application to monodisperse particles synthesis, is volumetric and uniform heating which could be used to obtain materials with controlled morphology and functional properties. Earlier we've developed method of microwave-assisted synthesis of monodisperse RE-doped yttria powders consisting of spherical particles and possessing strong luminescence [3]. Comparing to conventional methods of synthesis microwave-assisted process allows increasing the yield of reaction dramatically.

In present work we've developed new method of microwave-assisted synthesis of $Y_{1.95-x}Gd_xEu_{0.05}O_3$ solid solutions and performed a study of influence of Y:Gd ratio on morphology and luminescent properties of monodisperse oxide particles.

2. EXPERIMENTAL

As starting compounds for preparation of $Y_{1.95-x}Gd_xEu_{0.05}O_3$ samples (x = 0, 0.4, 0.8, 1, 1.2, 1.6, 1.95) were used $Y(NO_3)_3 \cdot 6H_2O$ (Aldrich, 99.9 % purity), $Gd(NO_3)_3 \cdot 6H_2O$ (Aldrich, 99.9 % purity), $Eu(NO_3)_3 \cdot 6H_2O$ (Aldrich, 99.9 % purity), urea (KhimMed, 99 % purity). 0.5 L of solutions with 0.03 M total concentration of Y^{3+}, Gd^{3+} and Eu^{3+} (in various proportions depending on the composition of desired product) in deionized water were prepared.

Water solutions were transferred in 0.5 l flasks and exposed to microwave treatment for 3 hours using Panasonic NN-SD556M inverter microwave oven (2.45 GHz, 150 W minimum output power). During the microwave treatment process of precipitation of solid solution of rare-earth hydroxocarbonates proceeded as described in equation (1).

$$(Y^{3+} + Gd^{3+} + Eu^{3+}) + (NH_2)_2CO + (3+x)H_2O \rightarrow$$
$$\rightarrow (Y, Gd, Eu)(OH)CO_3 \cdot xH_2O\downarrow + 2NH_4^+ + H^+ \quad (1)$$

After treatment all samples were centrifuged, washed several times with deionized water and air-dried at 50 °C for 10 hours.

Resulting powders were annealed in laboratory furnace at 700 °C for 2 hours to form corresponding oxide solid solutions.

XRD analysis of synthesized samples was preformed using Rigaku D/MAX 2500 diffractometer (CuKα-emission). Identification of diffractions peaks was carried out using JCPDS PDF2 database. Morphology of samples were studied using Leo Supra 50 VP scanning electron microscope with acceleration voltage 1-10 kV. Photoluminescent properties were studied using Perkin Elmer LS-55 spectrometer with Xenon lamp as a source of excitation. Powdered samples were placed on copper carrier. Registration of emission was performed using photoelectron multiplier in the range of 400-900 nm.

* vanetsev@gmail.com
† irina.chuvashova@gmail.com

3. RESULTS AND DISCUSSION

XRD-analysis showed that all synthesized powders after annealing represent corresponding $Y_{1.95-x}Gd_xEu_{0.05}O_3$ solid solution with value of x very close to the one put during synthesis. Analysis of shift of (222) diffraction peak with increase of x (Fig. 1) allows one to see that synthesized solutions obey Vegard's law. As it is known that such solutions of yttrium and gadolinium oxides are disordered and radii of Y^{3+} and Gd^{3+} are not very different, this seems to be reasonable result.

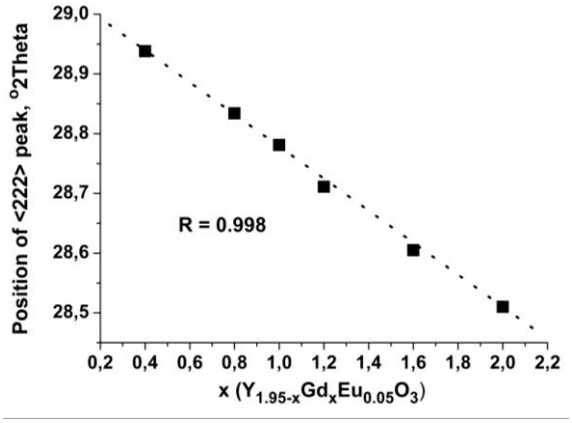

Fig. 1 – Dependence of position of (222) diffraction peak on composition of solid solution

Scanning electron microscopy (SEM) showed that all synthesized samples consist of spherical particles with very narrow size distribution. On Fig. 2 presented typical micromorphology for synthesized powders.

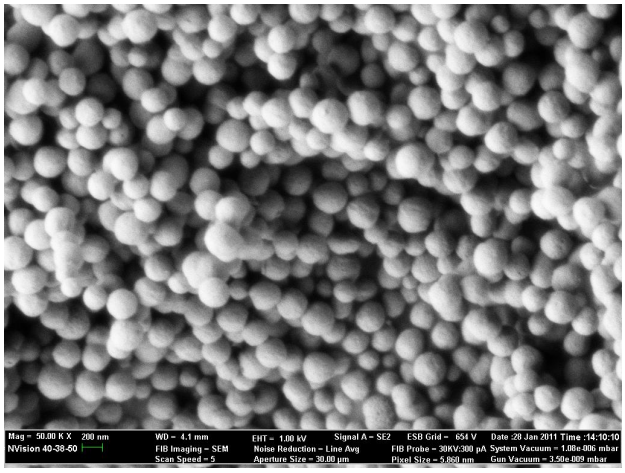

Fig. 2 – Morphology of $Y_{1.55}Gd_{0.4}Eu_{0.05}O_3$ particles

Dependence of mean size of spherical particles on composition of solid solution (Fig. 3) derived from SEM data shows that from pure yttrium oxide to pure gadolinium oxide mean size of particles decreases monotonously.

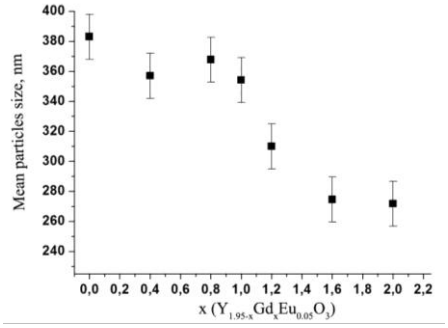

Fig. 3 – Dependence of mean size of spherical monodisperse particles on composition of solid solution

This most likely is related to the difference in solubility of corresponding hydroxocarbonates and, therefore, velocity of growth of precipitating particles.

As Eu^{3+} ion main luminescent emission band refers to symmetry forbidden transition its intensity is very sensitive to local symmetry. This most likely is the reason why relative intensity of $^5D_0 \rightarrow {}^7F_2$ to $^5D_0 \rightarrow {}^7F_1$ for these solid solutions has clear maximum near x = 0.5 (Fig. 4) as this composition corresponds to the most distorted local surrounding of Eu^{3+} ion in crystal lattice.

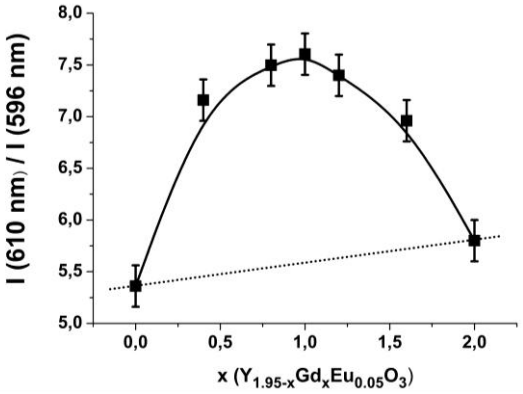

Fig. 4 – Dependence of mean size of spherical monodisperse particles on composition of solid solution

Therefore it is possible to state that developed method of synthesis of $Y_{1.95-x}Gd_xEu_{0.05}O_3$ solid solutions allows producing powders consisting of monodisperse spherical particles which size depends on composition of solid solution. Obtained results also show that use of solid solutions of yttrium and gadolinium instead of pure yttria as luminescent matrix will allow increasing the efficiency of luminescence significantly.

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