Synthesis and Luminescent Properties of Barium-Europium (III) Silicate Systems

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Abstract : Previous studies have shown that the involvement of silica hydrogel derived from serpentine minerals (Mq(Fe))₆ [Si₄O₁₀](OH)₈ as a source of silicon dioxide in SiO₂-NaOH-BaCl₂-H₂O system results in precipitating via one-hour stirring of boiling suspension such intermediates that on heating up to the temperature of 800 °C crystallize into the product composed of barium ortho- Ba₂SiO₄ and metasilicates BaSiO₃. Taking into account the fact that the suggested precipitation method based on the silica hydrogel mentioned allowed avoiding a number of drawbacks related with tetraethoxysilane Si(OC₂H₅)₄ frequently used in sol-gel routes, this approach has been decided to be adapted to inserting europium (III) Eu³+ ions into the structure of the synthesized compounds. A series of experiments was performed for the investigation of optical properties evolution observable in the final samples. Intermediates previously precipitated in SiO₂·H₂O (silica hydrogel)-NaOH-BaCl₂-Eu(NO₃)₃ system via stirring for 60 min at room temperature underwent one-hour heat-treatment at different temperatures (600]1200 °C). When the silica hydrogel was metered, SiO₂ content in the silica hydrogel that is 5.8 % was taken into consideration in order to guaranty the molar ratios of both SiO₂ to BaO and SiO₂ to Na₂O equal to 1:2. BaCl₂ and Eu(NO₃)₃ reagents were weighted so that the formation of appropriate compositions was guaranteed. A number of samples including various concentrations of Eu³+ ions (1.25, 2.5, 3.75, 5, 6.35, 8.65, 10, 17.5, 18.75 and 20 mol%) has been synthesized by the described method. Luminescence excitation, emission spectra of the final products were recorded on the Agilent Cary Eclipes fluorescence spectrophotometer (scanning rate = 30 nm/min, slit width = 5 nm, and Voltage = 800 V) as the excitation source. X-ray powder diffraction (XRPD) measurements were made on the SmartLab SE diffractometer. Emission spectra recorded for all the samples at an excitation wavelength of 394 nm exhibit peaks centered at around 536, 555, 587, 614, 653, 690 and 702.5 nm. The most intensive emission peak is observed at 614 nm due to 5D0 →7F2 of europium (III) ions transition. Luminescence intensity achieves its maximum for Eu³+17.5 mol% and heat-treatment at 1200 °C. The XRPD patterns revealed that the diffraction peaks recorded for this sample are identical to NaBa₆Nd(SiO₄)₄ reflections. As Nd-containing reagents were not involved into the synthesis, the maximum luminescent intensity is most likely to be conditioned by NaBa₆Eu(SiO₄)₄ formation whose reflections are not available in the ICDD-JCPDS database of crystallographic 2024. Up to Eu3+2.5 mol% the samples demonstrate the phases corresponding to Ba₂SiO₄ and BaSiO₃ standards. Subsequent increasing of europium (III) concentration in the system leads to NaBa₆Eu(SiO₄)₄ formation along with Ba₂SiO4 and BaSiO3. NaBa₆Eu(SiO₄)₄ share gradually increases and starting from 17.5 mol% and more NaBa₆Eu(SiO₄)₄ phase is only registered. Thus, the variation of europium (III) concentration in silica hydrogel-NaOH-BaCl₂-Eu(NO₃)₃ system allows producing by the precipitation method the products composed of europium (III)-doped Ba₂SiO₄ and BaSiO₃ and/or NaBa₆Eu(SiO₄)₄ distinguished by different luminescent properties. The work was supported by the Science Committee of RA, in the frames of the research project № 21T-1D131.

Keywords : europium (III)-doped barium ortho- Ba2SiO4 and metasilicates BaSiO₃, NaBa₆Eu(SiO₄)₄, luminescence, precipitation method

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