Versatile Phosphors $BaY_2Si_3O_{10}$: RE (RE = Ce^{3+} , Tb^{3+} , Eu^{3+}) for Light-emitting Diodes

W.-R. Liu (劉偉仁)¹, C.-C. Lin (林群哲)², Y.-T. Yeh (葉耀宗)¹, R.-S. Liu (劉如熹)², and B.-M. Cheng (鄭炳銘)³

¹Material and Chemical Research Laboratories, Industrial Technology Research Institute, Hsinchu, Taiwan ²Department of Chemistry, National Taiwan University, Taipei, Taiwan ³National Synchrotron Radiation Research Center, Hsinchu, Taiwan

Considerable interest in phosphors has led recently to the rapid development of promising display and illumination technologies. In particular, full-color photoluminescent materials are required for use in plasma display panels (PDPs) and ultraviolet lightemitting diodes (UVLEDs). Silicates are good candidates for use in stable host structures because they have high physical-chemical stability, waterresistant properties, stable crystal structures, and excellent optical properties. Blasse et al. reported on the fluorescent properties of Eu²⁺-activated binary and ternary silicates, including (Ca,Sr)₂MgSi₂O₇, MSiO₃ (M = Ba, Sr, Ca), BaSi₂O₅, BaMgSiO₄, CaMgSiO₄,and Sr₂LiSiO₄F. The systemization and luminescent properties of blue-excitable or UV-excitable Eu²⁺activated new silicates, such as Sr₃Al₁₀SiO₂₀, Ba₃SiO₅, Li₂CaSiO₄, and Li₂SrSiO₄ have also been elucidated. The results of these studies reveal these silicate phosphors are potential candidate in blue-pumping or UVpumping LED phosphors. Not only phosphors doped with divalent europium ions but also those doped trivalent rare-earth ions, such as Ce³⁺, Tb³⁺, and Eu³⁺, have been investigated: examples include $Na_3La_9O_3(BO_3)_8:RE^{3+}$ (RE = Eu, Tb), Ca- α -SiAlON:RE (RE = Eu^{2+} , Tb^{3+} , and Pr^{3+}), (Ca,Y)SiAlON:RE (RE = Eu^{2+} , Tb^{3+} , and Ce^{3+}), and $Y_2Si_4N_6C:Ce^{3+}$ or $Y_2Si_4N_4C:Tb^{3+}$.

To the best of our knowledge, the luminescent properties of rare-earth ions – activated $BaY_2Si_3O_{10}$ have not yet been reported upon. The aim of this study is to elucidate the synthesis, photoluminescence, thermal-stability, and color chromaticity of the new blue $(BaY_2Si_3O_{10}:Ce^{3^+}),$ green $(BaY_2Si_3O_{10}:Tb^{3^+}),$ and red phosphors $(BaY_2Si_3O_{10}:Eu^{3^+})$ and present their corresponding spectroscopic properties under VUV and UV excitation.

BaY₂Si₃O₁₀:xCe³⁺ (x = 1, 3, 5, 10, and 15 mol%), BaY₂Si₃O₁₀:yTb³⁺ (y = 5, 10, 20, 30, 40, and 60 mol%), and BaY₂Si₃O₁₀:zEu³⁺ (z = 5, 10, 20, 30, 40, 50, 60, and 70 mol%) were synthesized via a solid-state reaction. The starting materials that were used (to prepare OR in the preparation of) these phosphors were BaCO₃ (99.98%, Aldrich), Y₂O₃ (99.99%, Aldrich), SiO₂ (>99.9%, Strem Chemicals), Eu₂O₃ (99.9%, Aldrich), Tb₄O₇ (99.9%, Strem Chemicals), and CeO₂ (99.998%, Strem Chemicals). The raw materials were weighed out in stoichiometric proportions and the mixtures were then fired at 1350°C for 10 h in a 15%H₂/85%N₂ atmosphere (Ce³⁺,

Tb³⁺) or an air atmosphere (Eu³⁺). The resulting powder was cooled to room temperature in a furnace, ground, and pulverized for further measurements.

The VUV photoluminescence (PL) and photoluminescent excitation (PLE) spectra were obtained at the National Synchrotron Radiation Research Center (NSRRC) in Taiwan using the BL03A beam line. The PLE spectra were collected by scanning a 6 m in length cylindrical grating monochromator with a grating at 450 l/min, over a wavelength range of 100-350 nm.

The series of BYSO samples is determined to be suitable for VUV excitation ($\lambda = 172$ nm), as displayed in Fig. 1. The excitation bands of BYSO:Ce³⁺, BYSO:Tb³⁺, and BYSO:Eu³⁺ at ~172 nm were attributed to host absorption. A small hump observed at ~220 nm (Fig. 1a) resulted from the f-d excitation of Ce³⁺. The excitation band at around 230 nm (Fig. 1b) was caused by the ⁷D_J transition of Tb³⁺. The band between 200 and 280 nm was the CT band of Eu³⁺-O²⁻. The emission peak at ~460 nm may be caused by the occupation by Ce³⁺ of Ba²⁺ sites at the high-resolution synchrotron radiation beam line. The results indicate that these BYSO with the new compositions are good candidate luminescent materials for excitation under VUV and UV.

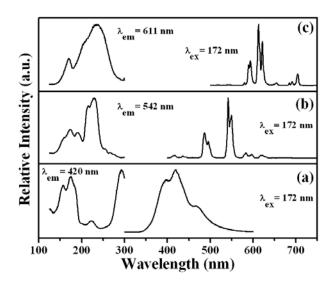


Fig. 1: VUV PL and PLE spectra of $BaY_2Si_3O_{10}$:RE involving in (a) RE = Ce^{3+} , (b) RE = Tb^{3+} , and (c) RE = Eu^{3+} measured at room temperature, respectively.