

Luminescence of the Green-Emitting $\text{Ca}(\text{La,Gd})_4(\text{SiO}_4)_3\text{O}:\text{Tb}^{3+}$ Phosphors with VUV Excitation

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We have investigated the synthesis, VUV photoluminescence spectra, optical properties, and chromaticity of hexagonal $\text{Ca}(\text{La}_{1-x-y}\text{Tb}_x\text{Gd}_y)_4\text{Si}_3\text{O}_{13}$ phosphors by using synchrotron radiation in the VUV spectral range. The VUV PLE and PL spectra and the correlation among VUV PL intensity, λ_{em} , and Tb^{3+} and Gd^{3+} -content have been established.

To investigate the VUV luminescence performance, we have measured the PLE spectra for $\text{CaLa}_4\text{Si}_3\text{O}_{13}$, $\text{CaGd}_4\text{Si}_3\text{O}_{13}$, and $\text{CaM}_4\text{Si}_3\text{O}_{13}$ ($M = \text{La, Gd, or both La and Gd}$) doped with 5% of Tb^{3+} , respectively. We compare the PLE spectra of $\text{CaLaSi}_3\text{O}_{13}$ with $\text{CaGdSi}_3\text{O}_{13}$ in the upper section in Figure 1. The broad band from 170-220 nm in curve a is host-related absorption. When La^{3+} is replaced by Gd^{3+} , shown in curve b, the band becomes broader, because the transitions ($^8\text{S}_{7/2} \rightarrow ^6\text{G}_J$, $^8\text{S}_{7/2} \rightarrow ^6\text{F}_J$) within Gd^{3+} ions overlap the host absorption. Besides, the narrow peaks at 273 and 276 nm are attributed to $^8\text{S}_{7/2} \rightarrow ^6\text{I}_J$ transition. When doping Tb^{3+} ions, other broad bands in curve c, d, and e which are resulting from the $f-d$ transitions of Tb^{3+} in the host lattices are observed. The electrons configuration of Tb^{3+} is $4f^8$, so the ground state is $^7\text{F}_6$ and there are two kinds of spin states, $^9\text{D}_J$ and $^7\text{D}_J$, within the $4f^75d$ excitation levels. For this reason, Tb^{3+} in a specific host exhibits two groups of $f-d$ transitions, spin-allowed with high-energy and spin-forbidden with lower energy. From 190 nm to 250 nm in curve c is a strong broad band and it is assignable to the spin-allowed transitions ($^7\text{F}_6 \rightarrow ^7\text{D}_J$) of Tb^{3+} while another weak broad, from 240 nm to 270 nm, is assigned to the spin-forbidden transitions. However, the intensity of the $f-d$ transitions within Tb^{3+} reduces in curve d. Here also saw the $^8\text{S}_{7/2} \rightarrow ^6\text{I}_J$ transition, at 274 nm, within Gd^{3+} ions, indicating the existence of the energy transfer from Gd^{3+} to Tb^{3+} in $\text{Ca}(\text{Gd}_{0.95}\text{Tb}_{0.05})_4\text{Si}_3\text{O}_{13}$. If Gd^{3+} ions only substitute half of La^{3+} ions, there is no significant change occurring, except for slight red shift of the peaks which belong to the $f-d$ transitions, as shown in curve e.

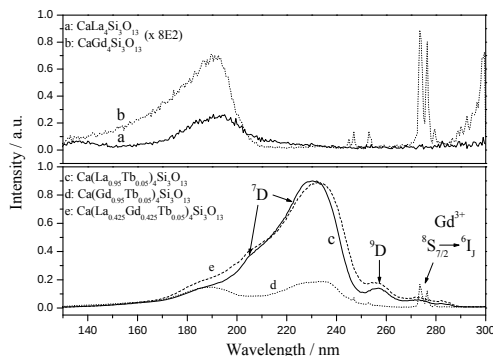


Figure 1. VUV PLE spectra of $\text{CaLa}_4\text{Si}_3\text{O}_{13}$ (a), $\text{CaGd}_4\text{Si}_3\text{O}_{13}$ (b), $\text{Ca}(\text{La}_{0.95}\text{Tb}_{0.05})_4\text{Si}_3\text{O}_{13}$ (c), $\text{Ca}(\text{Gd}_{0.95}\text{Tb}_{0.05})_4\text{Si}_3\text{O}_{13}$ (d), and

$\text{Ca}(\text{La}_{0.425}\text{Gd}_{0.425}\text{Tb}_{0.05})_4\text{Si}_3\text{O}_{13}$ (e).

To improve the absorption efficiency of phosphors in the VUV spectral region, we have also investigated the VUV PL spectra of $\text{Ca}(\text{La}_{0.9}\text{Gd}_y\text{Tb}_{0.1})_4\text{Si}_3\text{O}_{13}$ as a function of doped Gd^{3+} content under VUV excitation at 147 and 172 nm and the results are represented in Figures 2(a) and 2(b), respectively. The existence of the $^8\text{S}_{7/2} \rightarrow ^6\text{I}_J$ transition, at 274 nm, within Gd^{3+} ions reveals that the energy transition from Gd^{3+} to Tb^{3+} happening. Consequently, we found that the PL spectra for the $\text{Ca}(\text{La}_{0.9-y}\text{Gd}_y\text{Tb}_{0.1})_4\text{Si}_3\text{O}_{13}$ phosphors increases with increasing Gd^{3+} content from $y = 0$ to 0.90, reaching an optimal dopant value at 0.90 under both excitation conditions.

Furthermore, as indicated by a comparison of VUV PL spectra excited by 147 and 172 nm, we observed that under VUV excitation the PL intensity of our phosphor $\text{Ca}(\text{Gd}_{0.9}\text{Tb}_{0.1})_4\text{Si}_3\text{O}_{13}$ is 33 % (i.e., excited at 147 nm) or 67 % (i.e., excited at 172 nm) of that for commodity P1-G1S from Kasei Optonix. These observations hint that $\text{Ca}(\text{Gd}_{0.9}\text{Tb}_{0.1})_4\text{Si}_3\text{O}_{13}$ might serve as a promising substitute for P1-G1S as a green-emitting PDP phosphor.

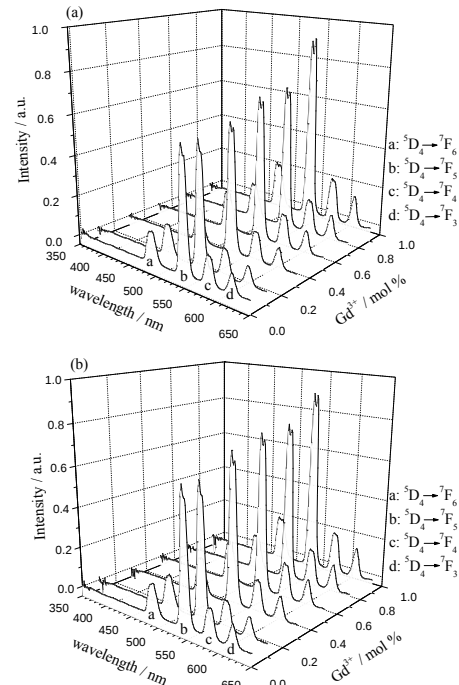


Figure 2. VUV PL intensity of $\text{Ca}(\text{La}_{0.9-y}\text{Gd}_y\text{Tb}_{0.1})_4\text{Si}_3\text{O}_{13}$ as a function of Gd^{3+} content: λ_{ex} = (a) 147 nm and (b) 172 nm.