Shaping scintillation and UV-VIS-NIR luminescence properties through synergistic lattice disordered engineering and exciton-mediated energy transfer in Pr^{3+}-doped $Lu_{1.5}Y_{1.5}Al_{5-x}Sc_xO_{12}$ (x = 0.0–2.0) garnets

Autorzy

Karol Bartosiewicz

Yevheniia Smortsova

Piotr Radmoski

Marcin E. Witkowski

Konrad J. Drozdowski

Masao Yoshino

Takahiko Horiai

Damian Szymański

Wioletta Dewo

Justyna Zeler

Paweł Socha

Maksym Buryi

Andrey Prokhorov

David John

Jakub Volf

Tomasz Runka

Tomasz Pędziński

Karol Hauza

Vitêzslav Jarý

Yasuhiro Shoji

Kei Kamada

Eugeniusz Zych

Winicjusz Drozdowski

Streszczenie

Akira Yoshikawa

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Artykuł

This study investigated the crystallization behavior, luminescence and scintillation properties of Pr^{3+}-doped Lu$_{1.5}$Y$_{1.5}$Al$_{5-x}$Sc$_x$O$_{12}$ (0.0, 0.5, 1.0, 1.5, 2.0) garnets, grown using the micro-pulling-down method, to address challenges associated with the substitution of Sc^{3+} for Al^{3+} ions due to their mismatched ionic radii in the same octahedral crystallographic site. A specially engineered crucible with five independent crystallization capillaries was used, which revealed that Sc^{3+} substitution caused localized melt heterogeneity, resulting in non-uniform melt ejection during crystallization. The threshold of Sc^{3+} ions concentration (x = 1.5) was identified, beyond which further substitution led to the formation of a garnet/bixbyite-like distorted perovskite hypoeutectic structure. This discovered a novel method for crystallization of hypoeutectic crystal growth by exploiting ionic radii mismatches. Vibrational spectroscopy confirmed that Sc^{3+} ions incorporation disrupted lattice symmetry, increasing structural disorder around Pr^{3+} ions. This structural modification significantly enhanced luminescence, particularly in the visible and near-infrared (NIR) ranges, achieving a sixteenfold increase in NIR luminescence intensity. Synchrotron radiation excitation spectra revealed that the band gap energy progressively decreased with increasing Sc^{3+} ions concentration. This finding provided crucial insights for designing materials based on band gap engineering strategies. A sixfold improvement in scintillation light yield, reaching 11 200 photons per MeV, was observed in the Lu$_{1.5}$Y$_{1.5}$Al$_{3.5}$Sc$_{1.5}$O$_{12}$ crystal (x = 1.5). The enhancement resulted from a Sc^{3+}-mediated energy transfer pathway , which optimized charge carrier dynamics by reducing deep trapping center density by an order of magnitude while preserving shallow traps. The EPR spectroscopy showed that Sc^{3+} incorporation reduced concentration of trace impurities, enhancing scintillation light yield. It also confirmed that F$^+$–Pr^{3+} interactions intensified Pr^{3+} emission at 370 nm and identified the 410–420 nm band as originating from F$^+$–O$^-$ defect pairs. These findings demonstrate that controlled lattice modification through Sc^{3+} incorporation allows for tuning structural and luminescent properties, offering a new approach for the design of advanced scintillators and luminescent materials with improved performance for targeted applications.

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