Composite Scintillators Based on Ce-doped Garnets as an Efficient and Inexpensive Alternative

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Single crystals of Ce-doped garnets, especially after appropriate band gap engineering and aliovalent codoping, exhibit an exceptional performance as scintillators. However, the growth of high-quality single crystals requires high temperatures and is costly. The current study was aimed at the development of inexpensive alternatives of scintillators based on Ce-doped garnets. We developed a two-step sol-gel procedure to fabricate composite scintillators containing Ce-doped yttrium aluminum garnet (YAG:Ce) grains embedded in a silica matrix. Ce-doped garnet powder was synthesized in the first step, followed by fabrication of the composites in the second step. Composites with partial or full substitution of Y by Lu to improve the stopping power for ionizing radiation were also fabricated.

High phase purity of the scintillator grains was confirmed by X-ray diffractometry, whereas scanning electron microscopy (SEM) and cathodoluminescence (CL) images were used to characterize the size and distribution of the grains, typically of ~1 μm in diameter. Agglomeration of the grains was observed at increasing garnet content in the composite. The nonequilibrium carrier dynamics and the emission decay characteristics were studied after short-pulse excitation with high-energy electrons and selective photoexcitation of activator ions Ce^{3+} in CL and photoluminescence (PL) experiments with time resolution on the order of tens of picoseconds.

The quantum efficiency of the emitting Ce^{3+} ions initially increases with increasing content of the scintillator material in the composite and saturates at a level close to that in bulk crystals, whereas the luminescence intensity increases superlinearly with garnet content (at least in the studied range of up to 2wt%). The increase in quantum efficiency is attributed to grain agglomeration into larger clusters. Statistical analysis of CL data from a large number of grains revealed the influence of surface recombination. The improvement of the normalized emission intensity with increasing garnet content is explained by an increased volume-to-surface ratio. A fast component in the luminescence decay was observed in the composites containing Lu and is interpreted by the formation of Ce^{3+} emission centers modified by nearby Lu and having smaller energy barrier for depopulation. Comparison of CL and PL decays revealed the influence of carrier trapping.

Additional emission peaked at 420 nm from the defects in silica matrix is observed but is weak, spectrally separated from the main emission of Ce^{3+} and decays faster (decay time of 14 ns instead of 60 ns for Ce^{3+}).

In summary, we report on the development of two-step sol-gel procedures enabling a low-cost fabrication of composites containing microcrystals of Ce-doped YAG, LuYAG, and LuAG in a silica matrix and show that their performance as scintillators is close to that of analogous single crystals and has potential for further improvement through optimization of microcrystal size and increased scintillator content in the composite.

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