Contribution ID: **187** Type: **Poster presentation**

Control of dopant segregation in colquiriite-type fluoride single crystal scintillators

Wednesday 20 September 2017 10:25 (1 minute)

Eu²⁺ doped LiCaAlF₆ and LiSrAlF₆ [Eu:LiCAF, Eu:LiSAF] single crystals have been investigated as a neutron scintillator for homeland security. The Eu:LiCAF and Eu:LiSAF single crystals indicated high light yield, ˜30,000 photons/neutron, compared to present neutron scintillators. However, the segregation coefficient, *k*_{eff}, of Eu²⁺ ion in the Eu:LiCAF and Eu:LiSAF single crystals is extremely small, *k*_{eff} = 0.02˜0.03, and the small segregation coefficient generated inhomogeneity in the bulk single crystals and decreased the yield rate of bulk single crystals. On these backgrounds, we grew Eu:LiCAF and Eu:LiSAF single crystals using Al metal as a starting material. Generally, EuF₃ powder was used for starting material to grow the Eu:LiCAF and Eu:LiSAF single crystals. However, Eu ion is doped in LiCAF as Eu²⁺ ion, and Eu³⁺ ion has to be reduced before doping to LiCAF and LiSAF. Therefore, we tried to reduce the valence of Eu³⁺ ion in EuF₃ by Al metal.

Eu:LiCAF and Eu:LiSAF single crystals were grown using Al metal powder as a starting material by the micro-pulling-down (μ-PD) method. Mixed powders with nominal compositions of Li(Ca_{1-x}Eu_x)(AlF_{1-y}Al^M_y)F₆ and Li(Sr_{1-x}Eu_x)(AlF_{1-y}Al^M_y)F₆ with x = 0.005˜0.03 and y = 0, 0.01 were prepared from LiF (7.5% ⁶Li), CaF₂, SrF₂, AlF₃, EuF₃ and Al metal powders (> 4N purity). In the chemical formula, Al^M and AlF are Al elements derived from AlF₃ and Al metal, respectively. Rectangular specimens with the thickness of 1 mm were obtained from the grown crystals and they were polished for measurements of optical and scintillation properties.

Eu:LiCAF and Eu:LiSAF scintillator single crystals were grown using Al metal as a starting material to improve the segregation coefficient of Eu ion. All Eu:LiSAF single crystals using Al metal [Eu,Al^M:LiSAF] indicated high transparency while Eu3%:LiSAF crystal without Al metal included milky parts in the crystal. The powder X-ray diffraction patterns indicated that all Eu,Al^M:LiSAF single crystals were a single phase of colquiriite-type structure without impurity phase. Effective segregation coefficient, *k*_{eff}, of Eu2%,Al^M1%:LiSAF single crystal was significantly improved to the value of 0.986 using the Al metal. In the transmittance spectra, all Eu,Al^M:LiSAF single crystals indicated more than 70% transmittance and absorption peaks were observed around 200 and 300 nm. Light yield of Eu,Al^M:LiSAF single crystals under thermal neutron irradiation increased with an increase of Eu concentration and the crystal with Eu2% indicated the maximum light yield, 10,000 photon/neutron. Details of crystal growth, optical and scintillation properties for Eu,Al^M:LiCAF and Eu,Al^M:LiSAF will be reported.“

Has accepted

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Session Classification: Poster Session 2

Track Classification: P5_characterization