Evaluation of Radiation Resistance for Apatite Solidification Materials by Optical Properties Analysis

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Huge amount of wastes containing radioactive Cs, Sr and minor actinide such as Am were generated due to Fukushima Daiichi Nuclear Power Plant Accident. Immobilization method of such radioactive nuclides is needed for volume reduction of the radioactive waste and safe disposal. Therefore, we focused on apatite materials due to some merits such as their long-term chemical durability, high thermal stability and radiation resistance. Furthermore, the apatite materials can sustain multivalent cations stably in the structure by chemical bond and hydrogen gas production can also be minimized because of the lack of hydrated waters if the sintering step is applied in synthetic scheme. Recently, we successfully synthesized the apatite-type materials containing Cs, Sr and lanthanide such as $La_2Sr_6Cs_2(PO_4)_6(OH)_2$ by solid-state reaction at below 700 °C.[1] However, site selectivity of multivalent cations and the structural change by gamma radiation were not clearly detected by powder XRD though the slight color change was observed. To investigate the radiation effect in detailed, we synthesized the Eu doped apatite material, $La_{1.9}Eu_{0.1}Sr_6Cs_2(PO_4)_6(OH)_2$. Eu^{3+} was used as surrogate elements of Am^{3+} and also as the fluorescent probe to detect the slight changes in the structure. Fig.1 showed emission spectra of $La_{1.9}Eu_{0.1}Sr_6Cs_2(PO_4)_6(OH)_2$ by increase radiation dose up to 1000 kGy. With increase in radiation dose, decrease in luminescence intensities via Eu^{3+} f-f transitions were observed, indicating an increase in structural defects such as anionic vacancies and nonradiative pathways to the excited state of Eu^{3+}. It was also noteworthy that the intensity ratio of $^5D_0 \rightarrow {}^7F_2$ transition to $^5D_0 \rightarrow {}^7F_1$ transition decreased gradually with increase in the radiation dose. Generally, Eu^{3+} f-f transitions are insignificantly affected by local environment, and an asymmetric ratio defined as a ratio of intensity of electric dipole $^5D_0 \rightarrow {}^7F_2$ and magnetic dipole $^5D_0 \rightarrow {}^7F_1$ transitions can be used to evaluate the environmental change.[2] As a result of gamma irradiation, the increase of structural defects and the decrease of asymmetric ratio depending on local site symmetry of Eu^{3+} were clearly detected by luminescence spectra and the result may suggests the transformation of hydroxyl group surrounding trivalent cation in hydroxyapatite materials.

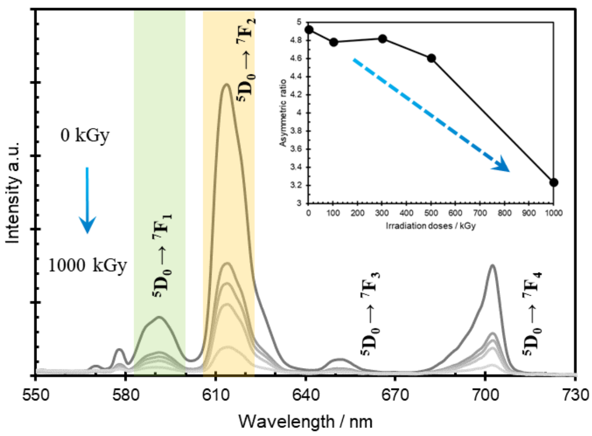

Fig.1 Emission spectra (λ_{ex}= 393.0 nm) of $La_{1.9}Eu_{0.1}Sr_6Cs_2(PO_4)_6(OH)_2$ by gamma irradiation, the inset show asymmetric ratio.

1) S. Kanagawa *et al*, *Chem. Lett.*, **2019**, **48**, 881.
2) I. E. Kolesnikov *et al*, *J. Rare Earths*, **2018**, **36**, 474.

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