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Power Generation Characteristics of Cr-doped Al_2O_3 Transparent Ceramics for Nuclear Battery Application

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Cr 0.05–1.0%-doped Al_2O_3 ceramics were synthesized by spark plasma and vacuum sintering to compare their power generation properties under α-, β- and X-ray irradiations for nuclear battery application. Moreover, Cr 0.6%-doped Al_2O_3 ceramics with different thicknesses fabricated by vacuum sintering were prepared to investigate the effects of sample size on power generation properties. The same as other phosphors, they showed the optimum Cr concentration in this specific application. The power generation decreased with increasing sample thickness under α- and β-ray irradiations, whereas the optimum sample size existed under X-ray irradiation.

1. Introduction

Many nuclear reactors are in operation around the world, and thus, nuclear wastes have become a problem since they emit ionizing radiation close to eternally.(1–3) Nuclear wastes are harmful when they are merely stored, but they become a useful resource when they contribute to electric power generation. Under this concept, the nuclear battery(4) has been studied for more than half a century. Mostly, three types of nuclear battery have been proposed, namely, thermoelectric,(5) semiconductor,(6) and phosphor.(7) The thermoelectric type has reached the practical application stage mainly in space applications,(8) and the semiconductor type has been demonstrated with a sample device in industrial fields.(9) Among these three types, the phosphor type is the most promising for large-scale power generation since the other two types are difficult to use in bulk form, which eventually leads to small-scale power generation.(8,9)

Despite such usefulness in a large scale, studies on the phosphor-type nuclear battery are limited. The concept of the phosphor type is as follows: bulk phosphor absorbs many quanta of ionizing radiation and converts them to UV–NIR emission, and the emitted photons are detected by Si solar cells to generate power.(10–13) Here, such a phosphor that converts ionizing radiation to low-energy photon emission is roughly classified into two types. One is a scintillator that

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immediately emits light after the absorption of ionizing radiation,[(14–17)] and the other is a storage phosphor that can temporally store the energy of ionizing radiation as a form of carrier trapping.[(18–21)] These phosphors have been traditionally applied to radiation detectors in some applications, including environmental monitoring,[(22)] optogenetics,[(23,24)] and astrophysics.[(25,26)] In most cases, they are used in bulk form, and the material forms are single crystal,[(27–48)] transparent ceramic,[(49–55)] glass,[(56–64)] and so on.[(65–68)] When such phosphors are used in nuclear battery applications, there remain several unclear points, such as which materials are preferable, the doping concentration dependence of emission centers in phosphors, the effect of sample size, and the effect of transparency. The aim of this work is to answer some of these questions such as the doping concentration, sample size, and transparency.

2. Materials and Methods

Samples are transparent ceramic $Cr:Al_2O_3$ produced by spark plasma sintering (SPS). The synthesis method was described in our previous paper where basic properties are also shown.[(69)] To compare the effects of transparency, vacuum-sintered $Cr:Al_2O_3$ ceramics were provided by Sumitomo Chemical. In both series, the same ultrafine, high-purity α-alumina, NXA-100[(70)] (purity > 99.99%, Sumitomo Chemical Co., Ltd., Japan), was used for sintering to fabricate the transparent ceramics and eliminate the effects of other impurities. Cr concentrations (Cr_2O_3 raw material was used) were 0.05–1.0% with respect to Al_2O_3, and here, the concentration was mass%. The sizes of SPS and vacuum-sintered samples were 10 mmφ × ~0.7 mmt and 10 × 10 × ~0.9 mm^3, respectively. Moreover, to compare the effects of sample size, vacuum-sintered samples with different thicknesses of 0.9, 2.2, 3.4, and 5.7 mm (the area was also 10 × 10 mm^2) were also provided by Sumitomo Chemical. In the thickness test, the Cr concentration was 0.6%, which had the highest scintillation intensity, as described later. The characterization method was the same as in our previous study,[(13)] and this time, to calculate the amount of power generated, the *I*–*V* property of the Si photodiode (Si-PD, Hamamatsu S12915-66R) was measured. Figure 1 shows samples with the same thickness. SPS samples were red transparent, and vacuum-sintered ones were red transparent-opaque. Under UV illumination, both samples exhibited a red emission. Moreover, in-line transmittance at 693 nm (emission wavelength of Cr^{3+}) was

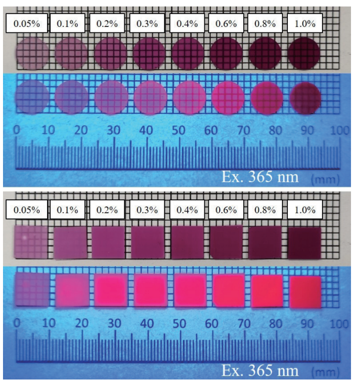

Fig. 1. (Color online) Picture of SPS (top) and vacuum-sintered (bottom) $Cr:Al_2O_3$ ceramics under room light and UV illumination.

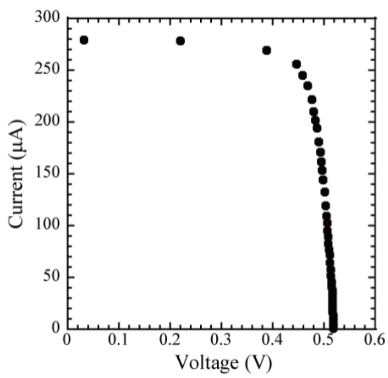

Fig. 2. *I*–*V* curve of Si-PD used in this experiment under white LED illumination.

measured using SolidSpec-3700 (Shimadzu). Figure 2 shows the *I–V* curve of Si-PD used in this experiment. The open-circuit voltage was estimated to be ~0.45 V. In these experiments, the X-ray source was the same as in our previous study,[65] and the delivered dose was calibrated using an ionization chamber (TN30013, PTW).

3. Results and Discussion

Figure 3 shows the output currents of all the $Cr:Al_2O_3$ samples with the same thickness under α- and β-ray irradiations. The uncertainty was around 5%, determined from several readouts under a fixed geometry, and only the readout error is plotted in Fig. 3. The output current ranged from 1 to 20 pA, and vacuum-sintered samples showed higher output. In this evaluation, the output current was corrected by the mass since the chemical composition was almost the same, and the active areas of the α- and β-ray-sealed sources were fully covered by the sample. The difference between SPS and vacuum-sintered samples is caused by the sample thickness. As described later, the sample thickness was insufficient to absorb all the secondary electrons. The same as the other phosphors having an optimum doping concentration, the SPS samples showed the optimum Cr concentration of 0.2–0.3%, whereas the outputs of the vacuum-sintered ones were diverse. Among the current samples, those with 0.05 and 0.6% Cr showed the highest output in vacuum sintering. This variation against Cr concentration is caused by several factors including the synthesis and polishing conditions and transparency. Figure 4 shows the output current under X-ray irradiation. The same as in the α- and β-ray cases, the vacuum-sintered samples showed higher output, and this tendency was easily understood by the difference in sample size. The optimum Cr concentration was 0.2–0.4% in the SPS samples. In vacuum-sintered ones, 0.3 and 1.0% Cr samples showed outstanding output, and except for them, the output was proportional to the Cr concentration. The vacuum-sintered samples were opaque, and in such cases, the X-ray absorption probability is a dominant factor.

The generated powers under α- and β-ray irradiations were roughly estimated as ~20 pA/g × 0.45 V / 0.3 s ~ 30 pW/g and ~5 pA/g × 0.45 V / 0.3 s ~ 3 pW/g for the brightest vacuum-sintered sample, respectively. Here, the accumulation time was 0.3 s. Nuclear wastes generally have a

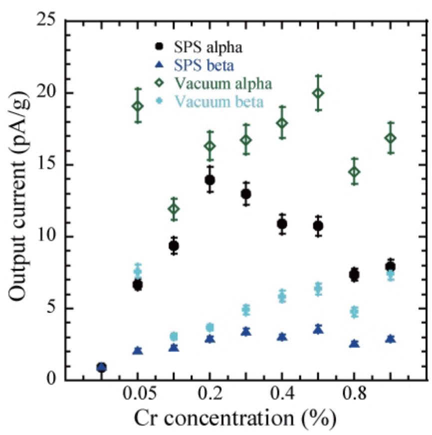

Fig. 3. (Color online) Output current of $Cr:Al_2O_3$ under α- and β-ray irradiations.

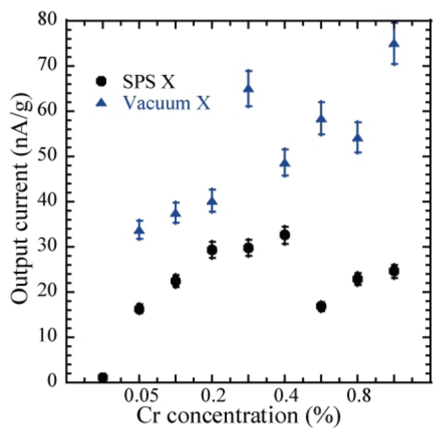

Fig. 4. (Color online) Output current of $Cr:Al_2O_3$ under X-ray irradiation.

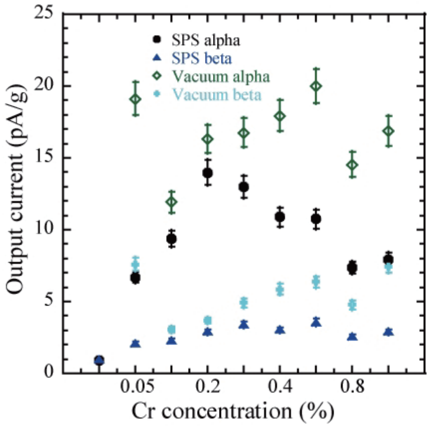

Fig. 5. (Color online) Output currents of vacuum-sintered Cr:Al_2O_3 under α- and β-ray irradiations with in-line transmittance at 693 nm.

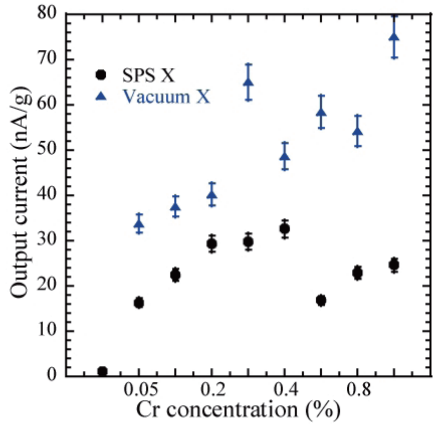

Fig. 6. (Color online) Output current of vacuum-sintered Cr:Al_2O_3 under X-ray irradiation with in-line transmittance at 693 nm.

bulk size of m^3 order, and instead of the mass, the volume is used for discussion here to easily calculate the amount of power generated by multiplying with the density (4 g/cm^3 in Al_2O_3). In the same sample, the power generation per volume was 30 pW/g × 4 g/cm^3 × 10^6 (from cm^3 to m^3) ~ 0.12 mW/m^3 with ~10^6 Bq radioisotopes (^{241}Am and ^{90}Sr). In the case of X-ray irradiation, the amount of power generated was ~60 nA/g × 0.45 V / 0.3 s = 81 nW/g, and it was 0.3 W/m^3 under the delivered X-ray dose of 85 mGy. In practical applications, the energy of radiation sources should be considered in addition to radioactivity.

To evaluate the effect of thickness on power generation, the output currents of samples with different thicknesses under α- and β-ray irradiations were compared, as shown in Fig. 5. In this experiment, the mass and volume were not corrected to compare the thickness (volume) effect. Since most interactions of α- and β-rays with matter occur at the surface of the sample, when the thickness increases, the output current decreases. Judging from this tendency, a 0.7–0.9 mm thickness (used in Fig. 3) was insufficient to cover the whole trajectory of secondary electrons since the effect of self-absorption appeared at 3.4 (α-ray) and 5.7 (β-ray) mm. The transmittance data also support the effect of self-absorption. Figure 6 shows the same plot under X-ray exposure. Because X-rays interact with matter not only at the surface but also inside, the graph shows a peak at the thickness of 2.2 mm. The graph shape was understood in terms of the trade-off between the volume and self-absorption. When the volume of the sample increases, the X-ray absorption probability increases, but at the same time, the effect of self-absorption becomes greater. If we can prepare transparent ceramics with different thicknesses, the effect of transmission will be further clarified.

4. Conclusions

The nuclear battery power generation properties of Al_2O_3 transparent and opaque ceramics doped with Cr at different concentrations were compared. In nuclear battery applications, the optimum dopant concentration appears the same as in other phosphor applications. Moreover, an optimum phosphor size also exists, and it depends on the target ionizing radiation. These results suggest that an adequate doping concentration and sample size must be selected for each power generation situation.

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