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Basic Research on Scintillator-based Nuclear Battery

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The power generation characteristics of many types of scintillator coupled with Si photodiodes under α-, β-, and X-ray irradiations were investigated. The current output of α-, β-, and X-ray irradiations showed a positive correlation. Throughout experiments, the required properties of scintillators for nuclear battery application turned out to be a long emission wavelength, high effective atomic number, and large volume as well as scintillation light yield.

1. Introduction

Ionizing radiation is composed of high-energy particles or electromagnetic waves and has been employed in many applications, including medical diagnostics,[1,2] radiation therapy,[3,4] individual dose monitoring,[5,6] optogenetics,[7,8] security,[9] resource exploration,[10] environmental monitoring,[11] and high-energy physics.[12,13] The monitoring of ionizing radiation is important to control them and avoid harmful damage to the human body, and to monitor them, phosphors have been used in many types of radiation sensor. Phosphors for radiation sensing are classified into two types, namely, scintillators and storage phosphors for dosimeters. Scintillators can convert the absorbed energy of a single quantum of ionizing radiation to numerous low-energy photons (scintillation photons) immediately.[14–17] Storage phosphors can store the absorbed energy of ionizing radiation as a form of carrier trapping, and after a certain period of time, the accumulated information (amount of absorbed energy) can be read as emission via the recombination of trapped carriers.[18–21] These two types of phosphor have different characteristics, but are useful in real-time and batch process measurements, respectively. In terms of emission efficiency, they generally show an inverse correlation with emission intensity.[18,19] Since their applications are wide, as mentioned above, and their required properties are different in each application, many types of phosphor have been developed so far with various material forms including single crystals,[22–41] ceramics (polycrystals[42–51]), and glasses.[52–65] Recently, in addition to the applications mentioned above, a nuclear battery has been considered as a new application in future power generation.

The nuclear battery is one of the power generation methods utilizing ionizing radiation, which is input energy, and power generation devices, which convert the energy of ionizing

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radiations to electric power.[66] Three main types of method, namely, thermoelectric,[67] semiconductor,[68] and scintillator types,[69] have been proposed. The thermoelectric type has reached the practical application stage mainly in space applications,[70] and the semiconductor type has been demonstrated as a sample device (not commercialized yet) in industrial fields.[71] Among these types, the scintillator type will be the most promising for large-scale power generation together with nuclear power plants since the use of the other two types is difficult in bulk power generation, which eventually leads to small-scale power generation such as that for space satellites and smartphones.[70,71] Despite such an ideal future application in a new battery, the scintillator type remains in the basic experimental stage, and only a few reports have been published.[69,72–74] Because the study of a scintillator-type nuclear battery is in a very early stage, the focus of most studies has been on simulation or basic experiments using commercial scintillators. Thus, the required properties of scintillators for this application are unknown, and this has large room for study. Furthermore, pioneering experiments have some problems. In past works, radiation sources with very high radioactivity/intensity were used,[69,75] but access to such powerful sources has been difficult for many researchers. In some investigations, since the intensity and penetration power of the ionizing radiation used were high, signals from scintillators and photodetectors (direct hit events) could not be distinguished. Unlike the thermoelectric and semiconductor types, the scintillator type must use photodetectors to convert scintillation photons to electric signals, and signals from the photodetectors should be distinguished in fundamental research studies.

To proceed with the research of a scintillation-type nuclear battery, in this study, we constructed a new measurement system for scintillator-type nuclear battery applications to solve the above problems. Since the ideal properties for this application are unclear, the scintillators shown in Table 1 have been examined.

2. Experimental Methods

A schematic drawing of the measurement system is shown in Fig. 1. A scintillator was coupled with an optical fiber, and the optical fiber was connected to a Si photodiode (Si-PD, Hamamatsu S12915-66R). Thus, scintillation photons were fed into Si-PD through the optical fiber (UD0065, Asahi spectra), and a sufficient distance could be achieved between Si-PD and an X-ray source to avoid a direct hit of X-rays to Si-PD. Although Si-PD was far from the ionizing radiation sources, it was also surrounded by 3 mm Pb seats to block scattering X-rays. The signals from Si-PD were transferred to a picoammeter (B2981A, Keithley) to observe an output signal current with 0.3 s integration time. The ionizing radiation sources were sealed ^{241}Am α-rays (4 MBq), sealed ^{90}Sr (1 MBq), and an X-ray generator supplied with a bias voltage of 40 kV and a current of 1 mA, which delivered a ~1 Gy/min dose to the sample. Since the energies of α- and β-rays were fully deposited on the surface of the scintillator, there was no concern for a direct hit to Si-PD. In addition, most parts of the sealed sources were covered by the sample, and the remaining part was also covered by black tape to block any signals from scintillations of air, which were excited by charged particles. In this system, the background electric noise level was suppressed to 0.1 pA so evaluations by weak sealed sources were possible, and all the experiments were carried out at room temperature.

Table 1
Samples used in this research. λ_{em} and V are the main emission wavelength and sample volume, respectively. V is calculated from density and mass due to undefined shapes of samples. In the remarks, abbreviations of the scintillator and supplier names are shown.

Sample	λ_{em} (nm)	V (cm^3)	Remarks
$Nd:Y_3Al_5O_{12}$	1064	0.89	Nd:YAG, Konoshima Chemical[76]
$Nd:Lu_3A_5O_{12}$	1064	0.09	Nd:LuAG, Self-made[77]
$Nd\text{-}Ce:Lu_3A_5O_{12}$	1064	0.29	Nd-Ce:LuAG, Self-made[77]
$Eu:Gd_3(Al,Ga)_5O_{12}$	1064	0.03	Eu:GAGG, Self-made[78]
$Eu:Y_3Al_5O_{12}$	610	0.03	Eu:YAG, Self-made[79]
$Sm:Y_3Al_5O_{12}$	625	0.03	Sm:YAG, Self-made[80]
$Cr:AlTaO_4$	900	0.04	Self-made
$0.1\%Cr:Al_2O_3$	693	0.05	Raw powder from Sumitomo Chemical
$0.4\%Cr:Al_2O_3$	693	0.07	Raw powder from Sumitomo Chemical
$Cr:Mg_4Ta_2O_9$	900	0.05	Self-made[81]
$Nd:LuVO_4$	1064	0.15	Self-made[82]
MgO	400	0.39	Self-made[83]
$Ce:Y_3Al_5O_{12}$	520	0.12	Ce:YAG, Konoshima Chemical[84]
$Ce:Gd_3(Al,Ga)_5O_{12}$	530	0.03	Ce:GAGG, Furukawa[85]
$Ce:Lu_3A_5O_{12}$	500	1.73	Ce:LuAG, Konoshima Chemical[86]
Lumilass-R	610	0.20	Sumita Optical Glass[87]
$Nd:30BaO\text{-}10PbO\text{-}60P_2O_5$	1064	1.30	Self-made
$CdWO_4$	480	0.64	NKK
$Bi_4Ge_3O_{12}$	480	0.88	BGO, Saint-Gobain
Tl:CsI	500	0.26	Saint-Gobain
$Ce:(Lu,Y)_2SiO_5$	420	0.13	Ce:LYSO, Saint-Gobain
$CsLiB_6O_{10}$	530	0.17	CLBO, Oxide[88]
ZnO	500	0.09	Self-made[89]
GaN	500	7×10^{-4}	Given by a company[90]
$Tb:Gd_2O_2S$	540	0.09	Tb:GOS, Hitachi Metals
$CaWO_4$	420	0.36	Mineral

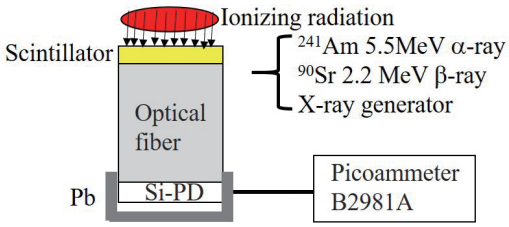

Fig. 1. (Color online) Schematic drawing of the experimental setup used.

3. Results and Discussion

Not all the samples have shown a detectable signal, and hereafter, only the results of samples with signal detection are discussed. Figure 2 shows the relationship between output signal currents under α- and β-ray irradiation. Some samples showed signal intensities of around 1– 30 pA. The output current under α-ray irradiation was higher than that under β-ray irradiation,

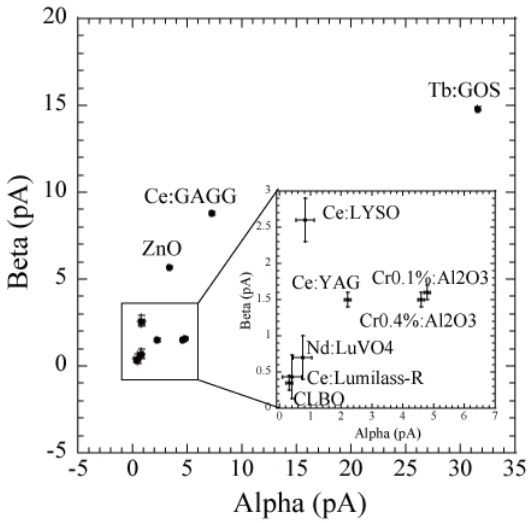

Fig. 2. Relationship between output signal currents under α- and β-ray irradiation.

which would be responsible for the energy and activity of ^{241}Am (5.5 MeV, 4 MBq) and ^{90}Sr (maximum energy of 2.2 MeV, 1 MBq) sources. Here, we demonstrate a rough calculation to consider the results. When the scintillation light yield (*LY*) is assumed to be typically 10^4 ph/MeV, the order of number output scintillation photons would be ~10^4/event for both α- and β-rays taking into consideration a typical α/β ratio of ~0.2. Under this condition, the output current would be estimated to be 10^4 (photons) × 0.8 (quantum efficiency of Si-PD) × ~10^6 (quanta)/10^{19} (conversion of number of electrons to current A) ≈ 270 pA/0.3 s. Therefore, the observed values of around 1–30 pA (few to ~10% efficiency) would be reasonable because no reflectors were used and sample shapes were not ideal for the optical fiber in this experiment.

The relationship between output currents under X- and α-ray irradiation is shown in Fig. 3. The positive correlation observed is the same as that in Fig. 2. Under X-ray irradiation, the irradiated dose rate was ~1 Gy/min, and a high signal intensity of around 1–800 nA was observed. Among the present samples, CLBO and Tb:GOS had the second and the third highest volume, respectively, and in X-ray measurement, the sample volume was considered important. Figure 4 shows the same relationship but for β-rays. The tendency observed was the same as that in Fig. 3. Throughout these experiments, α-, β-, and X-rays had the same positive correlation in current output, and all of these radiation sources could be applicable for this measurement. In practical applications, since there are various species of ionizing radiations for a nuclear battery, the selection of appropriate irradiation sources for each aim is required in basic experiments. If one would like to easily perform measurements (not considering the accessibility to radiation sources), X- or γ-ray (e.g., ^{60}Co) irradiation is simple to perform since the output current is very high and does not require a high technical ability in the measurements. The weak point of X- or γ-ray irradiation is the contamination of the signals from direct hit events of Si-PD, which can be avoided by setting a sufficient distance between the radiation source and Si-PD. In spite of the technical difficulty of detecting a very weak signal when using sealed α- and β-ray sources, their merit is the easy access to measurements since the radiation sources are very small (easy

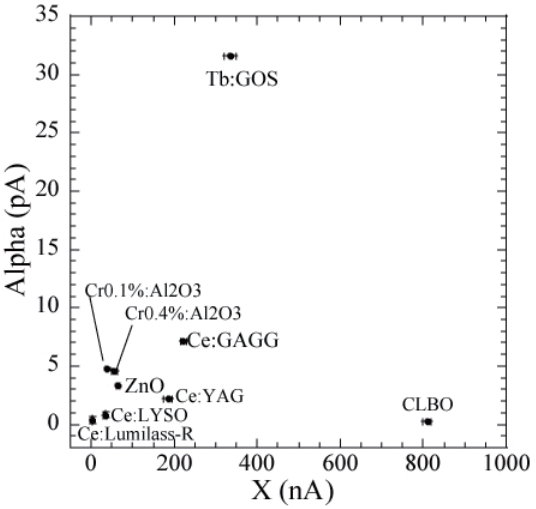

Fig. 3. Relationship between output currents under X- and α-ray irradiation.

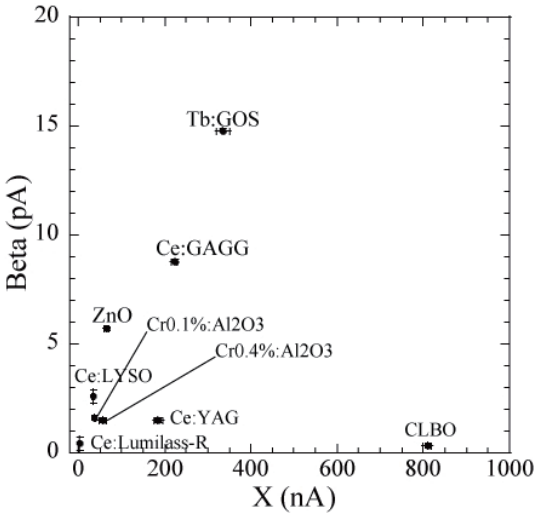

Fig. 4. Relationship between output currents under X- and β-ray irradiation.

handling) without any electric power input and can be easily used at the laboratory level. Furthermore, measurements using α- and β-ray sources offer a fair comparison since all the energy is fully deposited at the sample surface regardless of the chemical composition of the sample.

As a result of the above experiments, some required properties of scintillators for nuclear battery application become clear. Obviously, high scintillation *LY* is necessary, and the emission wavelength is also considered important. In this work, some scintillators with short (<500 nm) and long (>1 mm) wavelengths were examined, but most of them did not show a detectable signal. In terms of a short wavelength, only Ce:LYSO, which is a very common scintillator for positron emission tomography, exhibited a detectable signal, but its performance was inferior to that of Cr:Al_2O_3 even under X-ray irradiation. This was also true for long-emission-wavelength scintillators because Nd:YAG (a famous laser medium) and some other Nd-doped scintillators did not have a detectable signal. From these results, the ideal emission wavelength is 500–900 nm when Si-PD is used as a photodetector. Furthermore, the effective atomic number and sample volume are important when high-energy photons (X- and γ-rays) are used. In particular, the volume will be effective, as seen from the results of Lumilass-R glass. Although the scintillation *LY* of Lumilass-R glass is not very high,[87] it is the highest among the present samples. In practical applications for large-scale power generation, the cost of scintillators will be one of the important considerations since a large system is required, and cost-effective materials such as Al_2O_3,[91–93] ZnO,[94,95] and some glasses[96–98] may be interesting to study.

Apart from the experimental results, in terms of properties, afterglow should be reconsidered for this application. In conventional applications of scintillators, afterglow should be avoided to improve the signal-to-noise ratio, but in nuclear battery applications, afterglow, which is thermally stimulated luminescence (TSL) at room temperature, can contribute as a signal. Furthermore, in this application, the temperature of the sample will rise since the ionizing

radiation is irradiated continuously, and the final output signal is scintillation + afterglow. This is the biggest difference from conventional applications. In terms of TSL properties, high glow peak temperature and low fading rate are preferable for practical applications, but in this application, opposite properties will be ideal. For this application, a new concept of material design is required.

4. Conclusions

The required scintillation properties for nuclear battery application were experimentally determined. Long emission wavelength, high effective atomic number, and large volume as well as high scintillation *LY* are required for this application. Among the tested scintillators, Tb:GOS showed the best performance under α- and β-ray irradiation, and CLBO was the best for X-ray irradiation.

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