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Application of liquid scintillators as energy conversion materials in nuclear batteries

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ABSTRACT

The application of liquid scintillators as energy conversion materials in nuclear batteries was proposed. By introducing liquid scintillators, the output properties of radioluminescence (RL) nuclear batteries based on different liquid scintillators were investigated via low-energy X-ray. Results indicated that the values of short circuit current I_{sc} and maximum output power P_{max} increased as the X-ray intensity increased, and the output power of nuclear battery with Ultima Gold liquid scintillator was greater than those of batteries with Emulsifier-Safe or Bioscint liquid scintillator under excitation condition of 25 kV tube voltage. To analyze the RL effects of the liquid scintillators under X-ray excitation, we measured the RL spectra of the different liquid scintillators. Considering luminescence utilization in batteries, the surface of the quartz cuvette was covered with Al film. Al is a high-performance reflective material that can increase the scintillation light released by Ultima Gold, reaching the GaInP/GaAs/Ge device. After covering a thin Al film, the output power of the battery was significantly improved under X-ray excitation.

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1. Introduction

With the rapid development of micro-electromechanical systems (MEMSs) in military, aerospace, and biomedical applications, demand for power sources is increasing. Nuclear batteries have potential use as power supplies in MEMS because they exhibit high energy density, stability, and long life [1–5]. As a representative of nuclear batteries, low-energy X-ray nuclear batteries are attracting widespread interest because of their unique excellent properties [6–8]. Batteries based on low-energy X-rays can be easily shielded in contrast to γ-ray batteries and are safe because they only emit low-energy X-ray photons [9–12]. However, the semiconductor material of batteries suffers from radiation damage when exposed to intense fluxes of low-energy X-ray for a long time [13–16].

An alternative method to direct conversion of X-ray batteries is the addition of fluorescence materials, which directly absorb low-energy X-rays. Radiation energy is converted into luminescence energy, which is then converted into electricity [17–19]. Thus, the semiconductor material is shielded from X-ray radiation damage. In such nuclear batteries, fluorescence materials are exposed to X-ray irradiation, but radiation damage is slowed down because of these components. As a result, nuclear batteries can reduce radiation damage effectively.

Liquid scintillators are organic scintillators with high luminous efficiency; they are characterized by their low price, adequate attenuation length, long stability (with careful chemical treatment), and inherent radiation resistance [20–23]. Therefore, liquid scintillators can be used in nuclear batteries as intermediate energy conversion materials. The scintillation and fluorescence processes of liquid scintillators have been widely researched [24–28]. For a liquid system composed of one aromatic solvent and two fluorescent solutes, the scintillation process can be described in the following steps:

i The incident particle dissipates kinetic energy in the ionization and excitation of molecules along the track. Most of the ionized and excited molecules are solvent molecules, which comprise the majority of the solution. About 90% of the excited molecules, ions, and free radicals will recombine, de-excite, and dissipate their energy thermally. They do not contribute to the scintillation emission [25,26].
ii About 10% of the excited and ionized solvent molecules are populated to excited π-electronic singlet states. Depending on the

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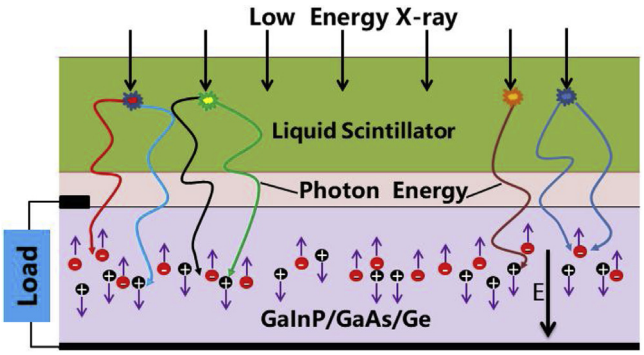

Fig. 1. Schematic of the GaInP/GaAs/Ge RL nuclear battery based on a scintillation cocktail.

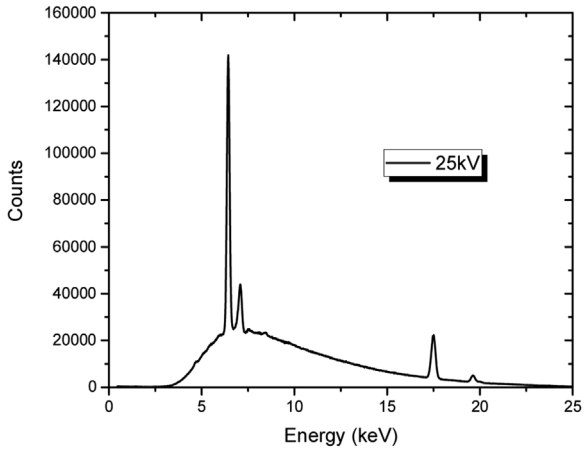

Fig. 2. X-ray energy spectrum for a tube voltage of 25 kV.

nature of the solvent, a fraction of these states undergo rapid internal conversion into the lowest excited singlet state [25,26].

iii The excitation energy from the singlet state is transferred to the primary solute and then to the secondary solute. Förster resonance energy transfer (FRET) is the dominant mechanism to transfer excitation energy between fluorescent molecules in a scintillator [27]. Given that FRET is driven by dipole–dipole coupling, it is radiationless and occurs over greater than interatomic distances (10–100 Å) between donor and acceptor molecules [27–29].

iv The de-excitation of the secondary solute leads to emission of photons in an appropriate wavelength range [26].

Nuclear batteries based on liquid scintillators are scarcely reported. This study aimed to study the characteristics of low-energy X-ray RL nuclear batteries based on different liquid scintillators under varying X-ray intensities. The optical properties of luminescent materials and spectral responsivity of a GaInP/GaAs/Ge device were analyzed. The output power of batteries was enhanced by increasing the luminescence intensity of liquid scintillators. Moreover, output performance improved after covering the surface of the quartz cuvette, which was filled with Ultima Gold liquid scintillator, with Al film. Our findings indicated that Al film could improve the luminescence utilization rate for batteries.

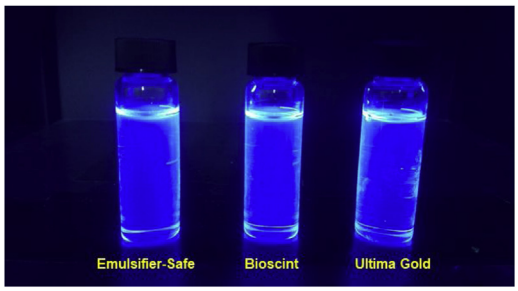

Fig. 3. Three different samples of liquid scintillators illuminated by 365 nm ultraviolet light. From left to right, samples are Emulsifier-Safe (PerkinElmer 6013381, USA), Bioscint (National Diagnostics LS-309, USA), and Ultima Gold (PerkinElmer 6013321, USA).

As illustrated in Fig. 1, the battery is composed of three parts: an X-ray emitter, liquid scintillator, and GaInP/GaAs/Ge device. In the battery, the liquid scintillator can absorb X-ray energy emitted from the X-ray emitter and produce radioluminescence (RL). The GaInP/GaAs/Ge photovoltaic material receives RL and creates many electron–hole pairs (EHPs). These EHPs are then separated by a built-in electrical field. Finally, the battery delivers an output current after being connected to an external circuit.

2. Materials and methods

2.1. Experimental materials

An X-ray tube (Shanghai KeyWay Electron Company Ltd., KYW900 A, China) was used as a suitable substitute for the low-energy X-ray source (such as ^{55}Fe, ^{241}Am, and ^{64}Cu) due to safety concerns and convenience. The X-ray tube provided easy adjustment of the intensity of emitted X-rays for this research. The basic specifications of the X-ray tube used in this study are listed in Table 1.

Under excitation condition of 25 kV tube voltage, the X-ray energy spectrum was obtained by a silicon drift detector (Amptek X-123, USA; Fig. 2). The high intensity of the X-ray was mainly concentrated in the middle section. The horizontal and vertical coordinates represent the photon energy and photon numbers, respectively.

Emulsifier-Safe liquid scintillator (product number: 6013381) and Ultima Gold liquid scintillator (product number: 6013321) were purchased from PerkinElmer, Inc. Bioscint liquid scintillator

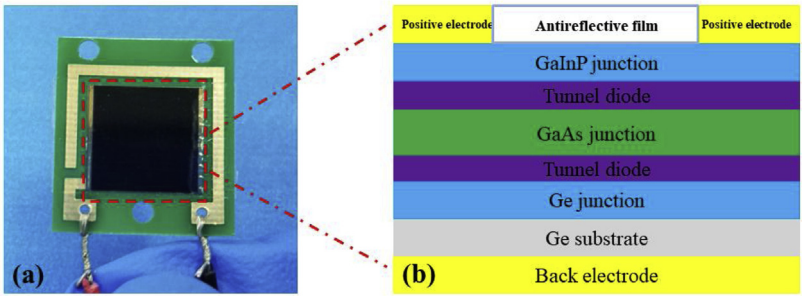

Fig. 4. (a) Prototype and (b) structure of the GaInP/GaAs/Ge device.

Table 1
Specification of the X-ray tube.

Anode voltage	Anode current	Maximum power	Filament voltage	Filament current
50 kV	0–1 mA	50 W	2.0 V	50 kV/1 mA, I_f = 1.7 A
Thickness of the beryllium window	Target angle	Focus spot size	Grounded mode	Target
200 μm	10°	0.1 mm × 0.1 mm	Grounded-cathode	Molybdenum

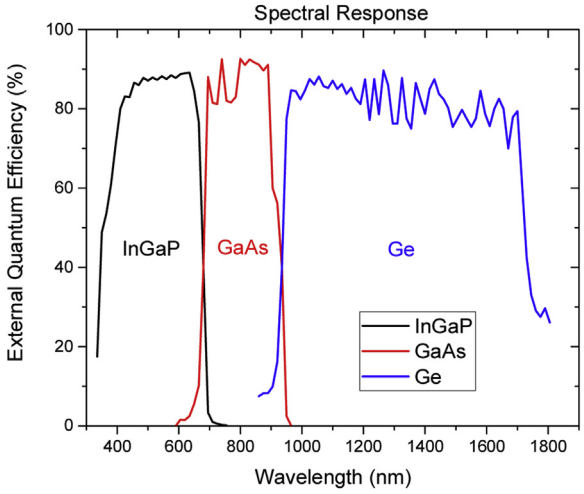

Fig. 5. Spectral response curves of the GaInP/GaAs/Ge device.

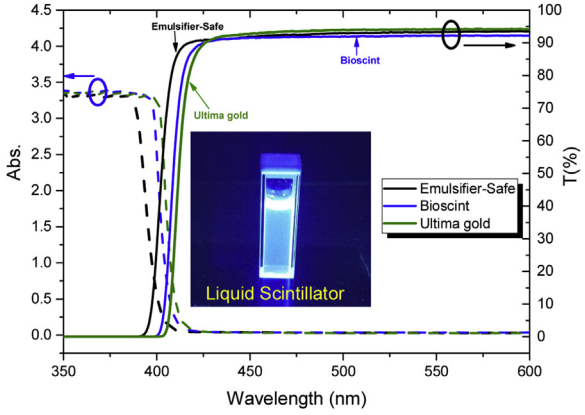

Fig. 7. Transmission and absorbance of Emulsifier-Safe, Bioscint, and Ultima Gold.

(product number: LS-309) was obtained from National Diagnostics, Inc. All these liquid scintillators have high flash points, good stability, and low toxicity, and they can be used as fluorescence materials for batteries. Fig. 3 shows some of these samples excited by a UV lamp at 365 nm. Each of these liquid scintillators stored in sample vials can emit blue light. A quartz cuvette was used to load the liquid scintillators because of its ultralight mass and good light transmittance. The inner dimension of the quartz cuvette was 10 mm × 20 mm × 45 mm.

The GaInP/GaAs/Ge device, which had a compact structure [30], large light absorption coefficient, wide band gap [31], and high crystal quality [32], was employed in the RL nuclear battery. The active area of the GaInP/GaAs/Ge device was 10 mm × 10 mm. The prototype and structure are illustrated in Fig. 4. Fig. 5 shows the spectral response curves of the GaInP/GaAs/Ge device. The spectral response curves of the GaInP/GaAs/Ge device were obtained at room temperature by a quantum efficiency spectrometer (Bentham PVE300, the UK).

2.2. Experimental measurements

2.2.1. Absorbance and transmittance tests

The absorbance (A) and transmittance (T) of the liquid scintillators were measured with a UV/Vis spectrophotometer (Shimadzu UV-2550, Japan) at 350–600 nm scanning range. A is expressed as follows:

$$A = lg(1/T) = lg(I_0/I_t) \quad (1)$$

where A is absorbance, T is transmittance, I_0 is the intensity of incident light, and I_t is the intensity of transmission light.

2.2.2. Excitation and emission spectra test

The excitation and emission spectra of the liquid scintillators were obtained by a Cary Eclipse fluorescence spectrophotometer (Agilent Technologies G9800a, Malaysia).

2.2.3. I–V test

The electrical properties of batteries, such as short circuit current (I_{sc}) and open circuit voltage (V_{oc}), were obtained with a dual-channel system source-meter instrument (Keithley Model 2636 A, USA) at room temperature. The maximum output power

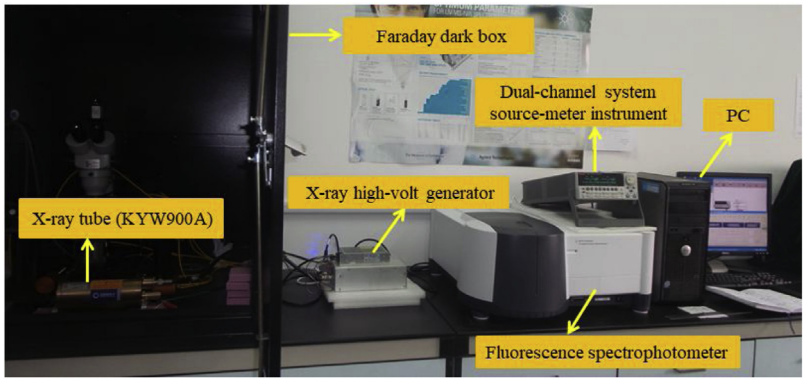

Fig. 6. Testing systems for electrical performance and RL spectra.

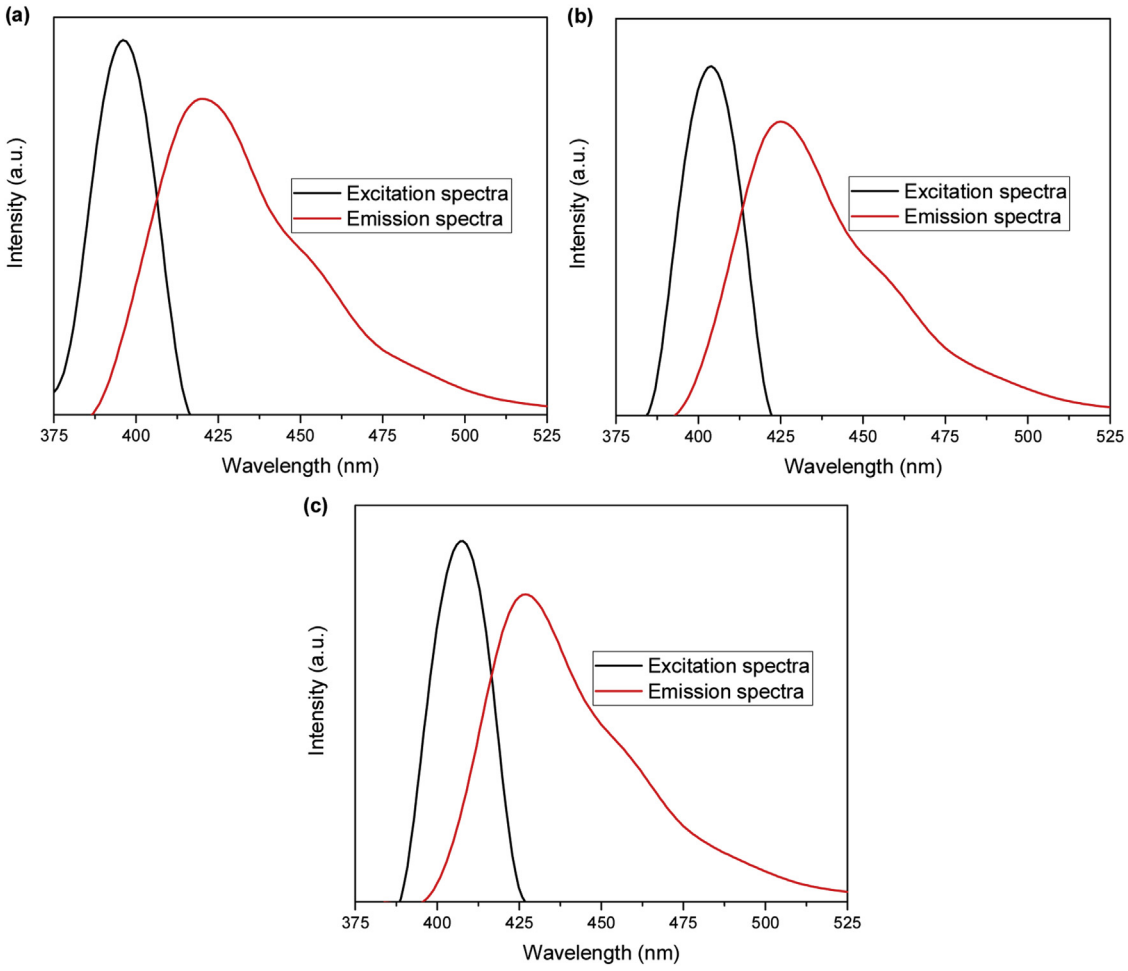

Fig. 8. Excitation and emission spectra of (a) Emulsifier-Safe, (b) Bioscint, and (c) Ultima Gold.

(P_{max}) and fill factor (*FF*) can be calculated by Eqs. (2) and (3) as follows:

$$P_{max} = V_{max} \times I_{max} \quad (2)$$

$$FF = \frac{V_{max} \times I_{max}}{V_{oc} \times I_{sc}} \quad (3)$$

where V_{max} and I_{max} are the voltage and current, respectively, at the maximum power point.

2.2.4. RL spectra test

The RL spectra of the liquid scintillators under different tube currents (100, 200, 300, 400, and 500 μA) were obtained with a Cary Eclipse fluorescence spectrophotometer (Agilent Technologies G9800a, Malaysia) at the tube voltage of 25 kV. In the process of RL spectra testing, X-ray tube was placed in fluorescence spectrophotometer and liquid scintillators were close to the X-ray emission window. The data mode of the Cary Eclipse fluorescence spectrophotometer was set as bio/chemiluminescence, and the emission slit and photomultiplier detector voltage were 20 nm and 700 V, respectively. Fig. 6 illustrates the corresponding measurement systems.

3. Results and discussion

3.1. Optical performances of the liquid scintillators

Emulsifier-Safe, Ultima Gold, and Bioscint liquid scintillators were used as luminescent materials in this experiment (Fig. 3).

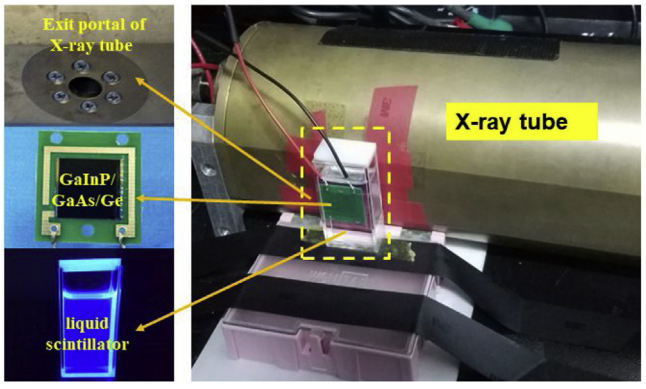

Fig. 9. Prototype of the experimental set up based on liquid scintillators.

By using a UV/Vis spectrophotometer (Shimadzu UV-2550, Japan), the absorbance (*A*) and transmittance (*T*) were measured over the scanning range of 350–600 nm. The optical path was 10 mm in the experimental measurements. As presented in Fig. 7, all these liquid scintillators exhibited high transmittance in the range of 425–600 nm, and the transmittance is higher than 91% in this wavelength range. This finding indicated that the RL at 425–600 nm was not easily absorbed by the liquid scintillators, thereby resulting in high transmission efficiency in this wavelength range. A sudden decrease in the transmission of these three liquid scintillators was

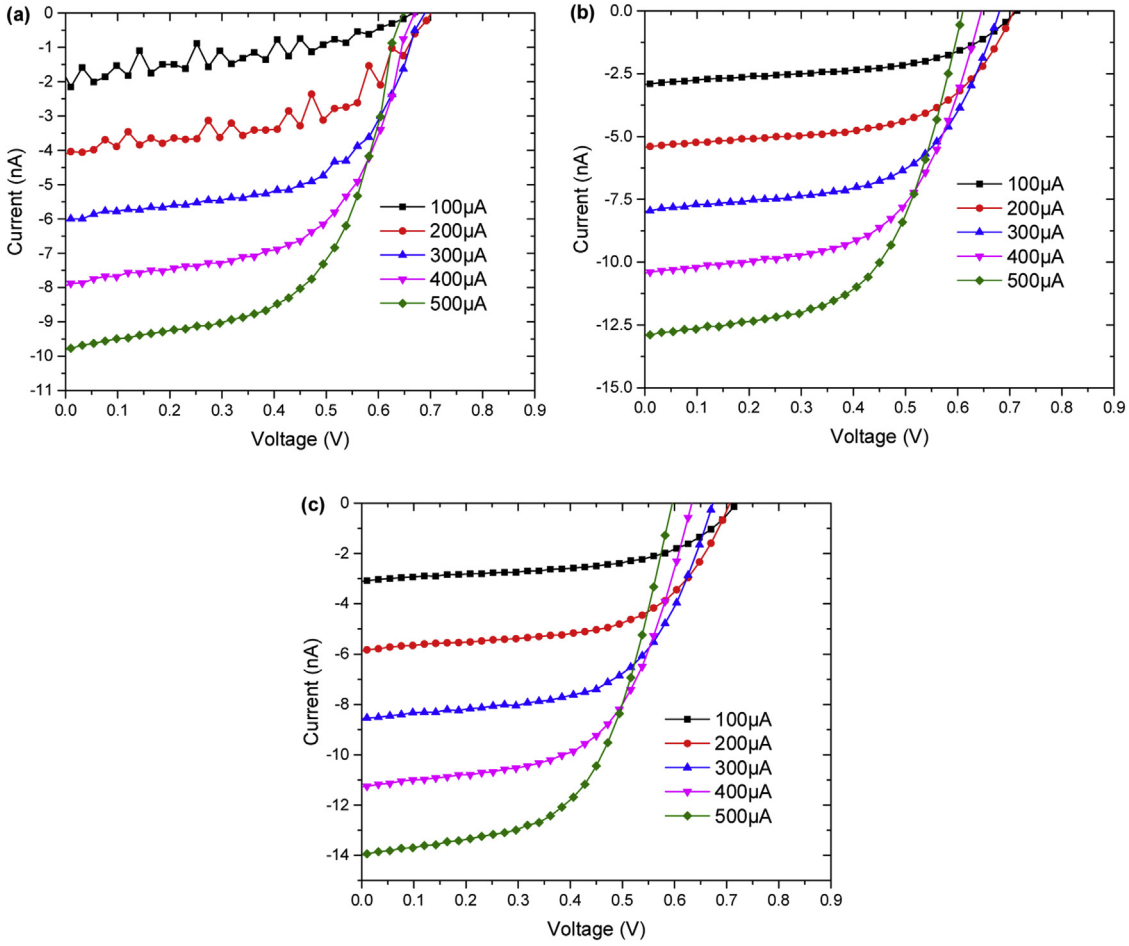

Fig. 10. *I–V* curves of the batteries with different liquid scintillators under different tube currents: (a) Emulsifier-Safe, (b) Bioscint, and (c) Ultima Gold.

observed when the wavelength was less than 425 nm, especially for Bioscint and Ultima Gold liquid scintillators.

The excitation and emission spectra of the liquid scintillators were obtained by fluorescence spectroscopy (Cary Eclipse fluorescence spectrophotometer, Agilent Technologies G9800a, Malaysia). As presented in Fig. 8, the emission peak wavelengths of Emulsifier-Safe, Bioscint, and Ultima Gold were approximately 418, 424, and 425 nm, respectively, and the excitation peak wavelengths of three liquids were 396, 404, and 408 nm, respectively. Self-absorption was noted at 388–418 nm for Emulsifier-Safe and at about 400–425 nm for Bioscint or Ultima Gold.

3.2. Electrical performances of the batteries with different liquid scintillators

The tube voltage was set to 25 kV, and the tube currents were 100, 200, 300, 400, and 500 μA. The electrical performances of the batteries with different liquid scintillators were measured during continuous and direct X-ray exposure. The experimental set up is shown in Fig. 9, and the corresponding *I–V* characteristics are illustrated in Fig. 10. I_{sc} remarkably increased with increasing tube current for the three liquid scintillators.

Fig. 11 shows the changes in I_{sc}, P_{max}, and *FF* for the batteries with different liquid scintillators as a function of tube current (100–500 μA). Changes in the electrical performances were observed after the tube current increased (Fig. 11). Obvious increases in I_{sc} and P_{max} were also noted. I_{sc} and P_{max} increased at a faster rate than *FF* with increasing tube current. The *FF* value stabilized at 300–500 μA. In Fig. 11**a** and **b**, I_{sc} and P_{max} of the battery with Ultima Gold were greater than those of the batteries with Emulsifier-Safe and Bioscint under the same tube current. In Fig. 11**c**, *FF* of the battery with Bioscint was extremely close to that of the battery with Ultima Gold but greater than that of the battery with Emulsifier-Safe under the tube current of 100–400 μA. However, the *FF* values in the batteries were nearly the same for Emulsifier-Safe, Bioscint, or Ultima Gold at the tube current of 500 μA.

3.3. Spectral matching between the liquid scintillators and GaInP/GaAs/Ge device

To clarify the differences in performance among Emulsifier-Safe, Bioscint, and Ultima Gold under X-ray excitation, experiments were carried out using a Cary Eclipse fluorescence spectrophotometer (Agilent Technologies G9800a, Malaysia). The RL spectra of Emulsifier-Safe, Bioscint, and Ultima Gold under different tube currents are provided in Fig. 12. The relative intensities of the RL spectra of the liquid scintillators were enhanced with increasing tube current. However, the emission peak wavelengths of the liquid scintillators remained steady and unchanged at different tube currents. The emission peak wavelengths of Emulsifier-Safe, Bioscint, and Ultima Gold were approximately 418, 424, and 425 nm, respectively.

The liquid scintillators exhibited different emission spectra because of the various luminescent materials used. The order of RL intensities for X-ray excitation was as follows: Ultima Gold > Bioscint > Emulsifier-Safe. Thus, Ultima Gold exhibited higher RL efficiency than Bioscint or Emulsifier-Safe under

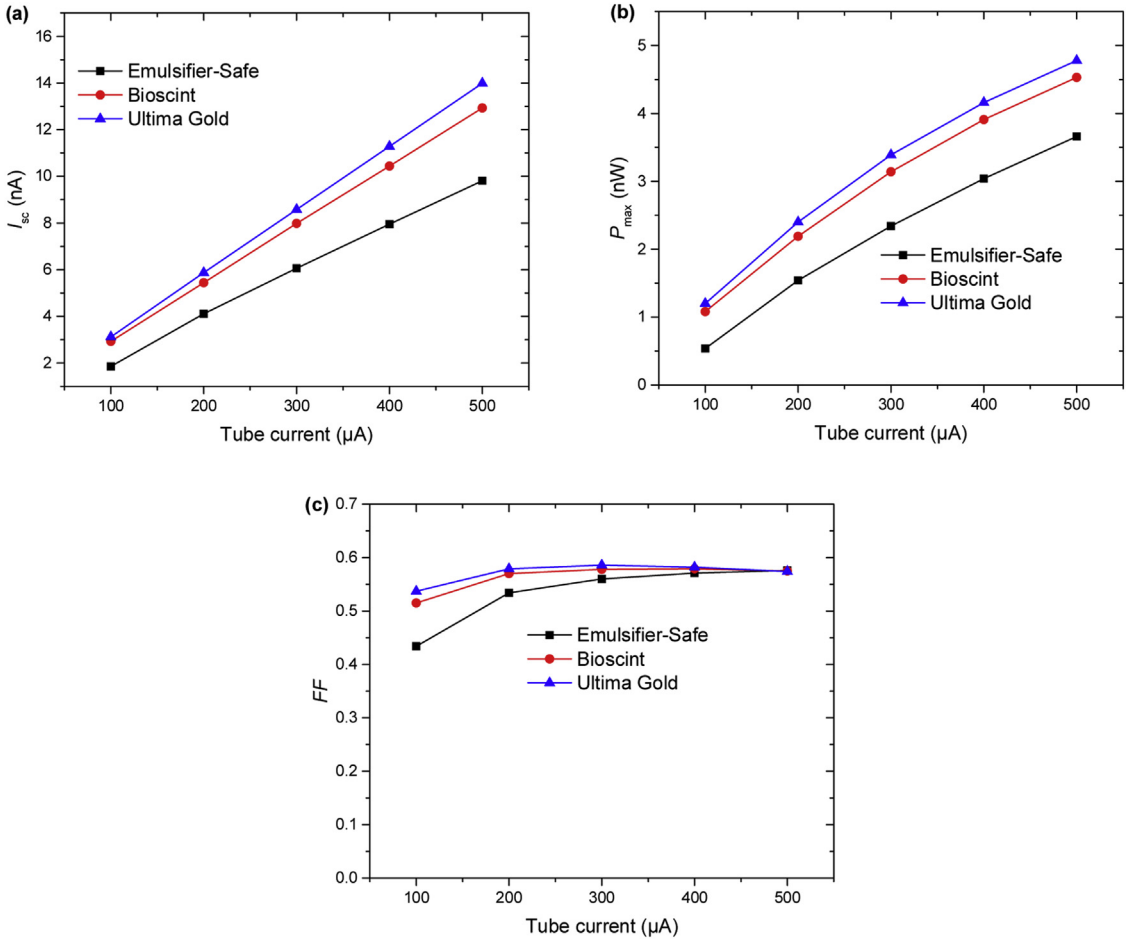

Fig. 11. Under excitation condition of 25 kV tube voltage, electrical properties of the batteries with different liquid scintillators as a function of X-ray tube current: (a) short circuit current I_{sc}, (b) maximum power P_{max}, and (c) fill factor *FF*.

X-ray excitation. The spectral response curves of GaInP and the RL spectra of the different liquid scintillators are given in Fig. 13. The external quantum efficiency (EQE) of the GaInP/GaAs/Ge device was 60%–88% at a wavelength range of 375–525 nm, where the light was absorbed efficiently by the device. Therefore, a good match was noted between the RL spectra of the three liquid scintillators and the spectral response of the GaInP/GaAs/Ge device. In addition, the EQE of the GaInP/GaAs/Ge device at the emission peak wavelength of Ultima Gold (425 nm) was higher than that at the emission peak wavelength of Emulsifier-Safe (418 nm) or Bioscint (424 nm). This result indicated that Ultima Gold was more suitable than Emulsifier-Safe or Bioscint to couple with the GaInP/GaAs/Ge device.

3.4. Increasing the utilization of scintillation photons

Ultima Gold exhibited higher RL efficiency than Emulsifier-Safe and Bioscint under X-ray excitation and was more suitable to couple with the GaInP/GaAs/Ge device than the other two scintillators. The energy attenuation plot of the X-ray photons in Ultima Gold simulated by GEANT4 and the screenshot of the GEANT4 model of the battery system are presented in Fig. 14**a** and **b**. The energy deposition of the X-ray photons increased with increasing thickness of Ultima Gold, and energy of the X-ray photons was almost completely deposited in Ultima Gold of 10 mm thickness (Fig. 14**a**). This phenomenon resulted in the increase in the generation of fluorescence. As can be seen from the Fig. 14**b**, X-ray tube produced a narrow beam of X-ray photons and some of the photons were scattered by solvent molecules when X-ray photons interacted with Ultima Gold. Al was introduced to improve utilization of the scintillation photons of Ultima Gold. Al film is a reflector that is thin enough so that it does not stop the low energy X-rays but thick enough so that it can reflect light from the liquid scintillator. The aim of this method is to reduce luminescent loss and increase luminescence availability. The structure and prototype of the battery are shown in Fig. 15**a** and **b**.

Table 2
Electrical parameters of the batteries with and without Al film.

Parameters	300 μA		400 μA		500 μA	
	UG	UG + Al	UG	UG + Al	UG	UG + Al
I_{sc} (nA)	8.58	9.77	11.28	12.83	13.99	15.88
V_{oc} (V)	0.67	0.65	0.63	0.61	0.60	0.57
P_{max} (nW)	3.39	3.62	4.16	4.43	4.78	5.04
FF	0.59	0.57	0.58	0.57	0.57	0.56

A thin Al film (10 μm thickness) was covered on the surface of a transparent cuvette, and the batteries were excited by an X-ray tube. The tube voltage was set to 25 kV, and the tube currents were 300, 400, and 500 μA. Fig. 16 shows the *I–V* and *P–V* characteristic curves of the batteries with and without Al film, and the corresponding electrical parameters are illustrated in Table 2. Ultima Gold was abbreviated as UG in the table. Fig. 16 reveals that I_{sc} and P_{max} of the batteries based on Ultima Gold increased significantly after covering the surface with Al film. The amount of luminescence that entered the PV material was affected by the

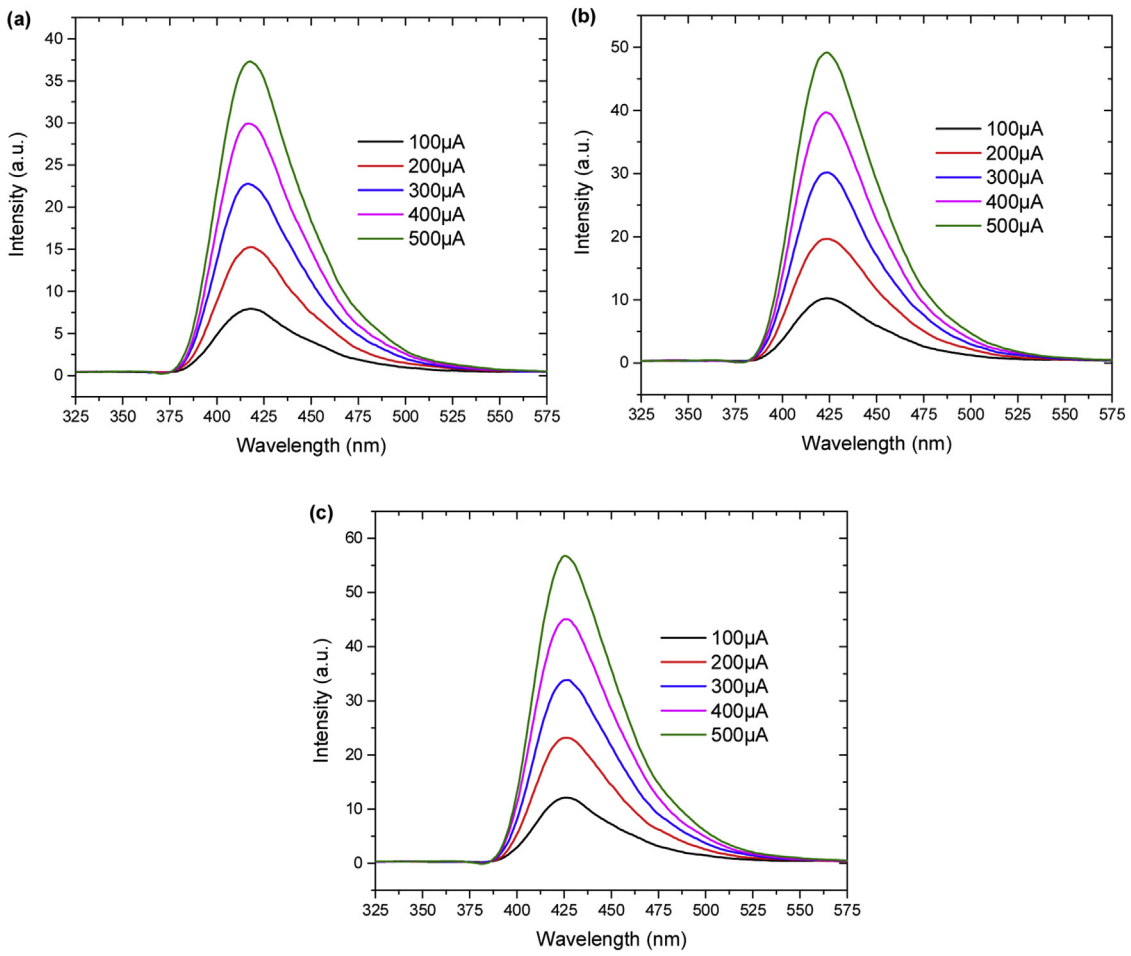

Fig. 12. RL spectra of liquid scintillators under different tube currents: (a) Emulsifier-Safe, (b) Bioscint, and (c) Ultima Gold.

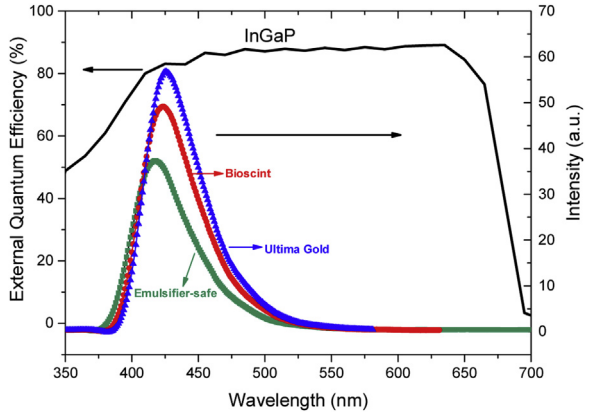

Fig. 13. Spectral response curves of the InGaP and RL spectra of different liquid scintillators under excitation of 25 kV tube voltage and 500 μA tube current.

amount of X-ray and luminescent utilization. Excited by X-rays, the liquid scintillator generated visible luminescence. The light emission of the liquid scintillator was isotropic, and the intensity of the light emitted toward the X-ray emitter was equal to that emitted toward the GaInP/GaAs/Ge device. The Al film was observed to be an excellent reflective material that could increase luminescent flux on the GaInP/GaAs/Ge device by reflecting the luminescence emitted toward the other directions. Table 2 shows that P_{max} of the batteries at 300, 400, and 500 μA increased by 6.78%, 6.49%, and 5.44%, respectively, after the Al film covered the surface of the cuvette.

4. Conclusions

GaInP/GaAs/Ge X-ray RL nuclear batteries based on liquid scintillators were investigated. In the experiment, I_{sc} and P_{max} of batteries increased with the increase in tube current under the tube voltage of 25 kV. The relative intensity of the RL spectra of the liquid scintillators also increased as the tube current increased, and Ultima Gold exhibited higher RL efficiency than Emulsifier-Safe and Bioscint. A good match was found between the RL spectra of the liquid scintillators and the spectral response of the GaInP/GaAs/Ge device, especially between Ultima Gold and the GaInP/GaAs/Ge

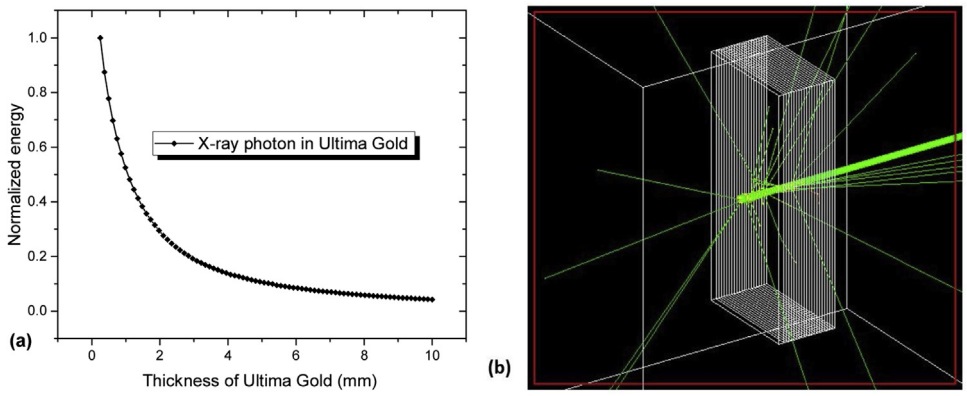

Fig. 14. For 25 kV X-ray tube voltage, (a) Energy attenuation of X-ray photons in Ultima Gold, and (b) screenshot of the GEANT4 model of nuclear battery based on Ultima Gold.

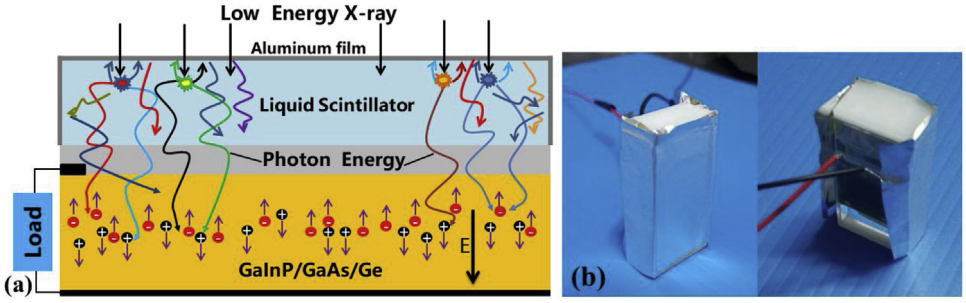

Fig. 15. (a) Schematic and (b) prototype of the battery enhanced by the Al film.

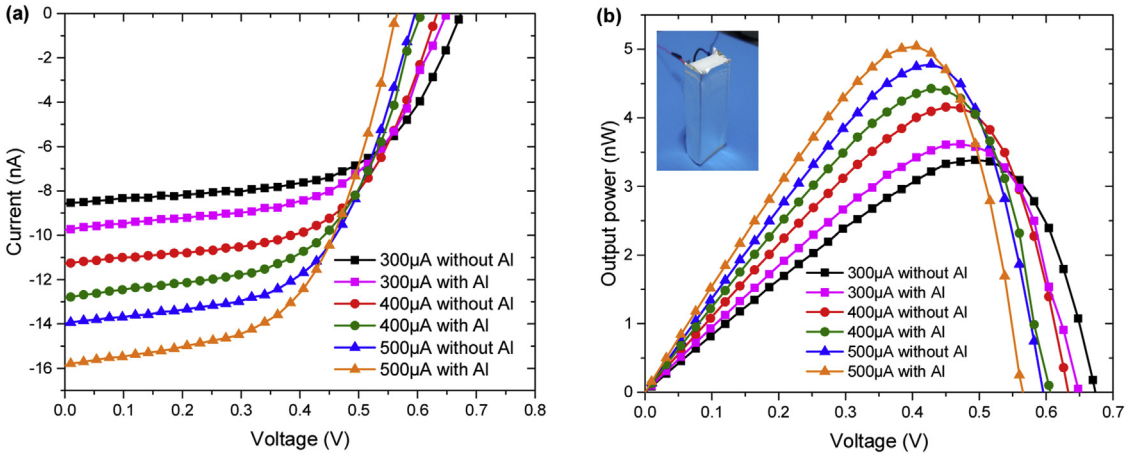

Fig. 16. *I–V* (a) and *P–V* (b) curves of the batteries with and without Al film.

device. Under the same excited condition, the battery coupled to Ultima Gold generated the best electric power compared with that coupled to Emulsifier-Safe or Bioscint. To improve the performances of the battery based on Ultima Gold, the surface of the quartz cuvette was covered with Al film to increase the availability of luminescence. Subsequently, the output power of the battery based on Ultima Gold improved. Thus, Al film could be used in X-ray RL nuclear batteries based on Ultima Gold to efficiently improve output performance.

Acknowledgements

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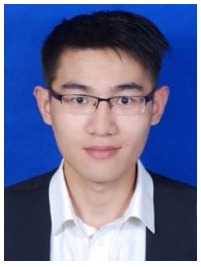

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