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Enhanced novel dual effect isotope batteries: Optimization of material and structure

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Summary

Materials and structures of radio-voltaic/radio-photovoltaic (RV/RPV) dual effect isotope batteries were optimized in this paper. The response relationship between different types of volt layer and the dual effect was studied. The GaAs volt layer with a high band gap can easily obtain a higher voltage output than the Si volt layer. Dual effect multilevel isotope batteries achieved better output performance by using the CsI scintillator due to its high photon yield. The CsI scintillator combined with the GaAs volt layer showed good spectral matching property, which is conducive to improve the RV/RPV dual effect isotope batteries. The overall scale of the multilevel structure was designed by performing Particle and Heavy Ion Transport code System (PHITS) calculation. When its overall scale reached 20 cm, the deposition rate of ^{60}Co gamma (γ) rays exceeded 90%. The response relationship between the thickness of each level of the CsI scintillator and the battery performance was analyzed by MCNP5. The total performance came up to the maximum when the CsI thickness in each level reached 2 cm. Finally, the multilevel isotope batteries were prepared. The output performance of the multilevel isotope batteries was characterized under ^{60}Co γ source at a dose rate of 0.103 kGy/hr. The multilevel isotope battery in series obtained open circuit voltage of 7.77 V, short circuit current of 1.69 μA, maximum output power of 7.98 μW, and energy conversion efficiency of 0.15%. These isotope batteries showed great potential for the power supply of electronic equipment in many special fields.

KEYWORDS
dual effect, gamma source, isotope battery, material and structure optimization, Monte Carlo simulation

1 | INTRODUCTION

Isotope batteries are good solutions for providing electrical energy to specific devices in deep space exploration, military, and other fields. With the rapid development of semiconductor research, the radio-voltaic (RV) effect isotope batteries have attracted the attention of many researchers in recent years. In the study of RV effect isotope batteries, the radioactive sources of these batteries are mostly beta (β) sources, such as Ni-63[1], Pm-147[2], and S-35[3]. The types of conversion units that match those sources include inorganic semiconductors (GaAs[4], Si[5], GaN[6], and SiC[7]), organic semiconductors (P3HT[8]), and nano-semiconductor materials[9,10]. The use of low-energy β sources can effectively avoid the problem of rapid decline of battery performance[11], but it also results in a low output performance. The radio-photovoltaic (RPV) isotope batteries effectively reduce the radiation damage effect of semiconductors. However, the RPV effect involves two energy conversions, leading to low conversion efficiency and weak output[12,13]. Therefore, due to the limitation of the choosing of radioactive source and the radiation damage of semiconductor, the current RV and RPV isotope batteries show low output performance and still have a large gap to meet the electrical energy requirements of actual electronic devices.

To solve the problems mentioned above and improve the performance of the radioisotope batteries in practical applications, we proposed a novel radio-voltaic/radio-photovoltaic (RV/RPV) dual effect multilevel isotope batteries based on a gamma (γ) radioactive source[14,15]. This isotope battery generates electricity by the RV/RPV dual effects through the interaction between γ rays and scintillator layers and volt layers. The γ source can effectively reduce the radiation damage on the semiconductor conversion materials. A multilevel structure was adopted to realize the gradual utilization of γ rays energy in consideration the strong penetration of γ rays. Previous work demonstrated that the energy conversion efficiency of the RV/RPV dual effect batteries is only 0.0235%[15] because of the low utilization of ^{60}Co γ rays in the two-level energy conversion structure.

To realize the further utilization of ^{60}Co γ ray, this work optimized the material and multilevel structure in the batteries. In accordance with the optimized results, multilevel isotope batteries were prepared and characterized under the 0.103 kGy/hr ^{60}Co γ source. The results show that the optimized multilevel isotope batteries obtain a considerable electrical output. The total energy conversion efficiency reaches up to 0.15%.

2 | MATERIALS OPTIMIZATION

In the isotope battery research, inorganic semiconductors have mature processing, good stability, and radiation resistance. Compared with other luminescent materials, scintillators show higher structural stability and have well-controlled thickness. Thus, inorganic semiconductors and scintillators are ideal energy conversion materials for multilevel isotope batteries. In this work, GaAs and Si semiconductor devices were selected as volt layers, and CsI and LYSO scintillators were selected as luminescent materials, respectively. The influence of the matching different volt layers and scintillators on the performance of RV/RPV isotope batteries was studied.

In this work, the X-ray tube was used as an equivalent γ source for irradiating the RV/RPV conversion module. The Si and GaAs volt layers were matched with CsI and LYSO scintillators, respectively. Four combinations, namely, Si + CsI, Si + LYSO, GaAs + CsI, and GaAs + LYSO, were formed. The thickness of the CsI and LYSO scintillators in each combination was 1, 5, 10, 20 mm, and the area was 1×1 cm^2. The performance of the RV/RPV effect isotope batteries was measured at 50 kV/800 μA.

Figure 1 shows that the battery performance was significantly improved by the dual effect after loading the scintillator. As the thickness of the scintillator increases, the performance of the dual effect batteries increases and then decreases. The decrease is mainly because of the self-absorption of the scintillator. For the same scintillator, the open circuit voltage (V_{oc}) and fill factor (FF) produced by using the GaAs volt layer were better than that by using the Si volt layer. At present, the reason that limits the practical applications of isotope batteries is that batteries cannot meet the voltage requirements of electronic equipment. To obtain a higher voltage output, the GaAs semiconductor was selected for the volt layer in the multilevel isotope batteries.

Under the condition of the same volt layer species and the consistent scintillator thickness, the performance by using CsI scintillator is superior to LYSO scintillator. To investigate the reason for this phenomenon, the fluorescence photon counting of the two kinds of scintillators was obtained by using a CCD camera (Andor-DU-888U3-CS0). As shown in Figure 2, the fluorescent photon relative count of CsI scintillator is significantly larger than that of the LYSO scintillator with the same thickness.

The scintillators have respective characteristic wavelengths. The volt layers show quantum efficiencies for different wavelengths. Thus, the suitability of different combinations has diversity. To characterize the suitability, the quantum efficiency tester (Newport IPCE/QE 200 tester) and the fluorescence spectrophotometer (Agilent Technologies, USA) were used to test the external quantum efficiency (EQE) curve of the volt layers and the emission spectrum of the scintillators, respectively. The EQE represents the ability of converting visible light to electric current, which positively correlates with the photoelectric conversion effect of the volt layer.

As shown in Figure 3A, the external quantum response peaks of the Si and the GaAs volt layer are ranged from approximately 500 to 850 nm. The peak wavelengths of the CsI scintillator and the LYSO scintillator are approximately 530 and 420 nm, respectively. This work used S to describe the matching between the volt layer and the scintillator quantitatively. The S can be expressed as follows:

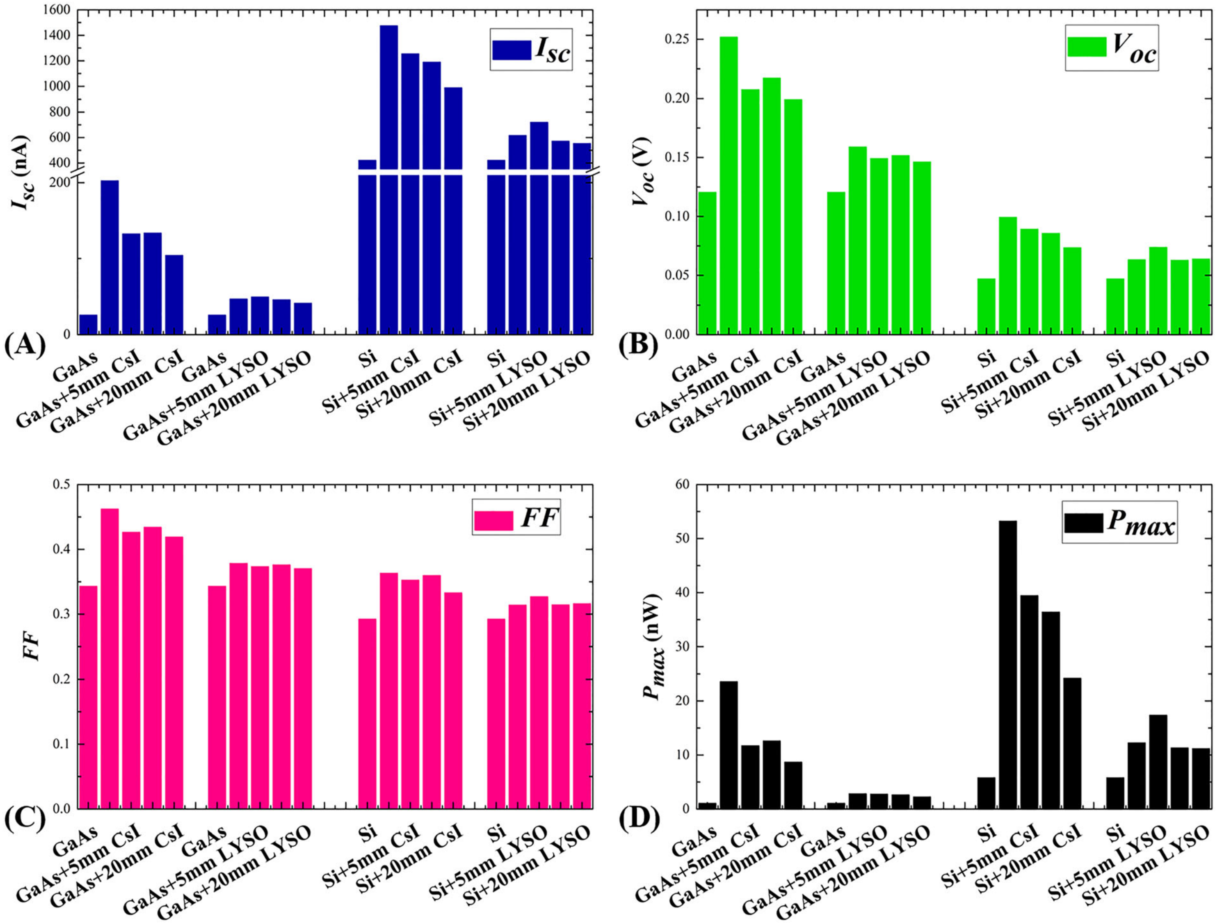

FIGURE 1 A, I_{sc}; B, V_{oc}; C, *FF*; D, P_{max} [Colour figure can be viewed at wileyonlinelibrary.com]

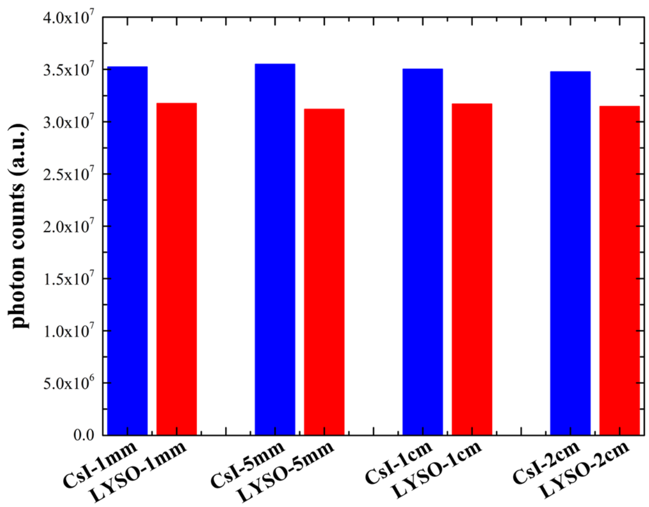

FIGURE 2 Comparison of the luminescence of the two scintillators under X-ray at 50 kV/800 μA [Colour figure can be viewed at wileyonlinelibrary.com]

$$S = \int_{\lambda-a}^{\lambda+a} \left(F_q \times F_e\right) d\lambda, \tag{1}$$

where F_q and F_e represent the polynomial fitting function of the EQE curve and the emission spectrum, respectively, λ represents the peak wavelength of the scintillator emission spectrum, and a represents the range of integration.

Figure 3B shows that the *S* between the CsI scintillator and the two kinds of volt layers is significantly better than that between the LYSO scintillator and the volt layers. Furthermore, the *S* between CsI and GaAs is the best. Hence, the effect of using CsI scintillator is better than that of LYSO scintillator.

3 | OPTIMIZATION DESIGN OF MULTILEVEL STRUCTURE

3.1 | Scale of multilevel structure

Scale length matching is an important concept in the design of isotope batteries[16]. Many nuclear batteries have been designed to consider the range of the ray and the scale of the conversion material for achieving a high-energy conversion efficiency. The γ rays released by ^{60}Co have a strong penetrating ability. Designing a suitable material scale is necessary to achieve an efficient utilization of γ energy.

The ability to absorb ^{60}Co γ rays of the four kinds of materials was calculated by Particle and Heavy Ion

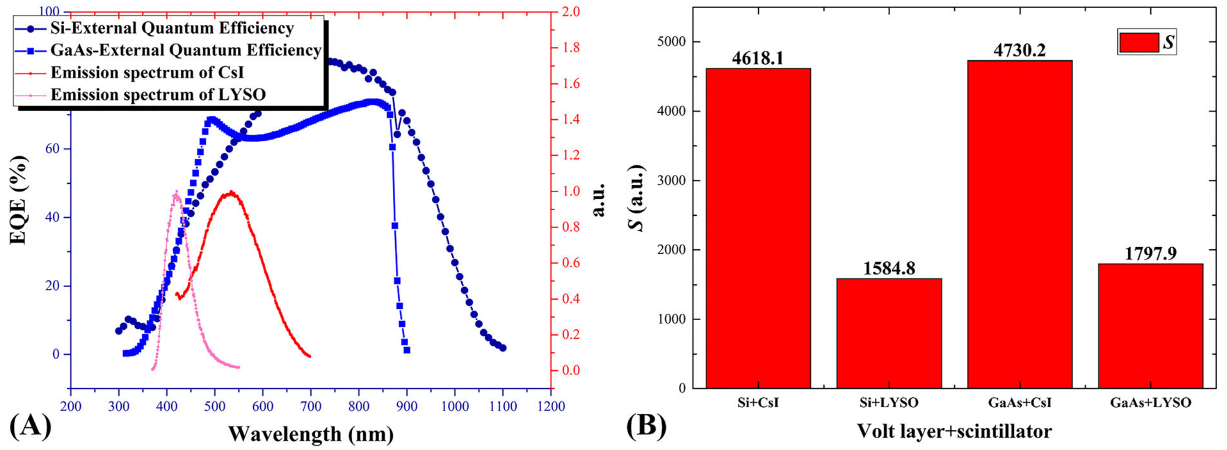

FIGURE 3 A, The emission spectrum of the scintillator and the external quantum efficiency curve of the volt layer; B, calculation result of suitability [Colour figure can be viewed at wileyonlinelibrary.com]

Transport code System (PHITS). PHITS is a Fortran program for particle transport simulation based on Monte Carlo method. It can simulate the transport of particles such as neutrons, protons, electrons, photons, and heavy ions in a three-dimensional magnetic field and gravity field. The PHITS calculation model was set to a sphere with varying diameter, which represents the energy conversion materials. A ^{60}Co point source was placed at the center of sphere to observe the ^{60}Co photon's deposition rate change with the thickness of the conversion material. Figure 4 reveals the calculation result.

As shown in Figure 4, the energy deposited in the conversion material generally increases with the increasing of the conversion material thickness. When the thickness reaches 20 cm, the deposition ratios of γ photons in Si, GaAs, CsI, and LYSO are 92.836%, 99.748%, 98.926%, and 99.952%, respectively. That means the use of the γ rays by the overall energy conversion structure tends to be saturated regardless of the combination type of those four materials. The structural design on this scale can maximize the use of γ energy.

3.2 | Effect of each level scintillator thickness on the performance

By determining the overall scales of the multilevel structure, this work discussed the response of the electrical properties to the thickness of each level scintillator. Six structures were created by using MCNP5. The parameters of all models are shown in Table 1. One CsI scintillator matched with two GaAs volt layers. The thickness one GaAs layer was set to 200 μm. Each CsI was equally divided by 100 μm per sublayer to calculate the emitted light

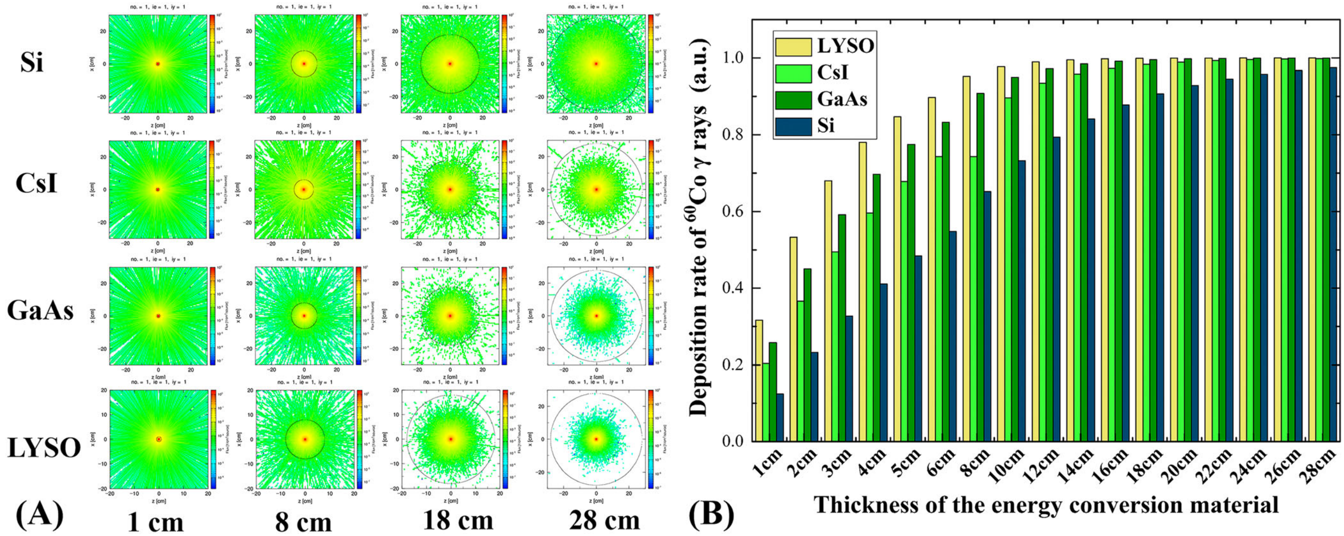

FIGURE 4 Relationship between γ photon distribution and material thickness: A, two-dimensional distribution; B, data statistics [Colour figure can be viewed at wileyonlinelibrary.com]

TABLE 1 Parameters of six multilevel structures

Serial number	1	2	3	4	5	6
Thickness of CsI in each level	1 cm	2 cm	3 cm	4 cm	5 cm	6 cm
Quantity of level	16	9	6	5	4	3
Total thickness	19.84 cm	20.16 cm	19.44 cm	21.2 cm	20.96 cm	18.72 cm

intensity. A PCB substrate with a thickness of 1 mm was set on the back of each GaAs volt layer. With a plane unidirectional ^{60}Co radioisotope source, the energy deposition in CsI scintillator and GaAs volt layer were calculated, respectively.

In our previous work, a calculation model for the limit output parameters of the dual effect isotope batteries have been established[15]. Self-absorption effects of scintillators need to be considered during the calculation of the RPV effect.

As shown in Figure 5, the UV-visible spectrophotometer (UV-2550, SHIMADZU) and fluorescence spectrophotometer (Agilent Technologies, USA) were used for testing the transmittance and characteristic wavelength of the CsI scintillator with different thicknesses, respectively. According to Lambert-Beer law, the self-absorption coefficient of scintillators can be obtained by fitting calculation[15].

According to the dual effect theoretical analysis model, the theoretical output parameters of I_{sc}, V_{oc}, P_{max}, internal efficiency and external efficiency of different structures under the 10 Ci ^{60}Co were obtained (Figure 6). The internal efficiency and external efficiency associate with the power of the γ rays deposited in the multilevel structure and the total output power of the ^{60}Co source, respectively.

As the CsI scintillator thickness of each level increases, the P_{max} of different models shows a trend of rising first and then decreasing. When the thickness of each layer scintillator reaches 2 cm, the P_{max} reaches the maximum value of 1.71 mW in series connection. Compared with Figure 6A with Figure 6B, it is easy to obtain a high V_{oc} in series and a high I_{sc} in parallel connection at the consistent thickness of the scintillator.

From Figure 6, as the scintillator thickness increases, the internal and external efficiencies of the isotope batteries first increases and then decreases. The maximum internal and external conversion efficiencies are 4.31% and 2.31% in series when the thickness of each level scintillator reached 2 cm, respectively.

4 | PREPARATION AND PERFORMANCE CHARACTERIZATION OF MULTILEVEL ISOTOPE BATTERIES

As a result of the optimized design, nine CsI scintillators combined with 18 GaAs volt layers formed a multilevel conversion module. Each level of the conversion module contains a CsI scintillator and two GaAs volt layers. The size of each scintillator is 1.5 cm × 1.5 cm × 2 cm, and the size of each GaAs volt layer is 1.1 cm × 1.1 cm × 200 μm. The prepared multilevel conversion module was placed in a custom enclosure to test and analyze its performance. The volt layers between each level were connected in

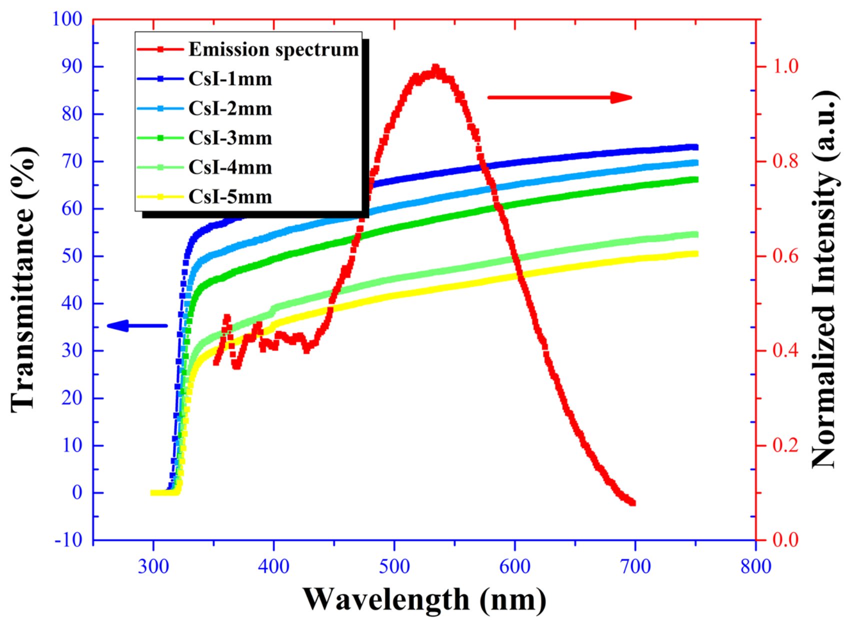

FIGURE 5 Transmittance of different thicknesses and emission spectra of CsI scintillators [Colour figure can be viewed at wileyonlinelibrary.com]

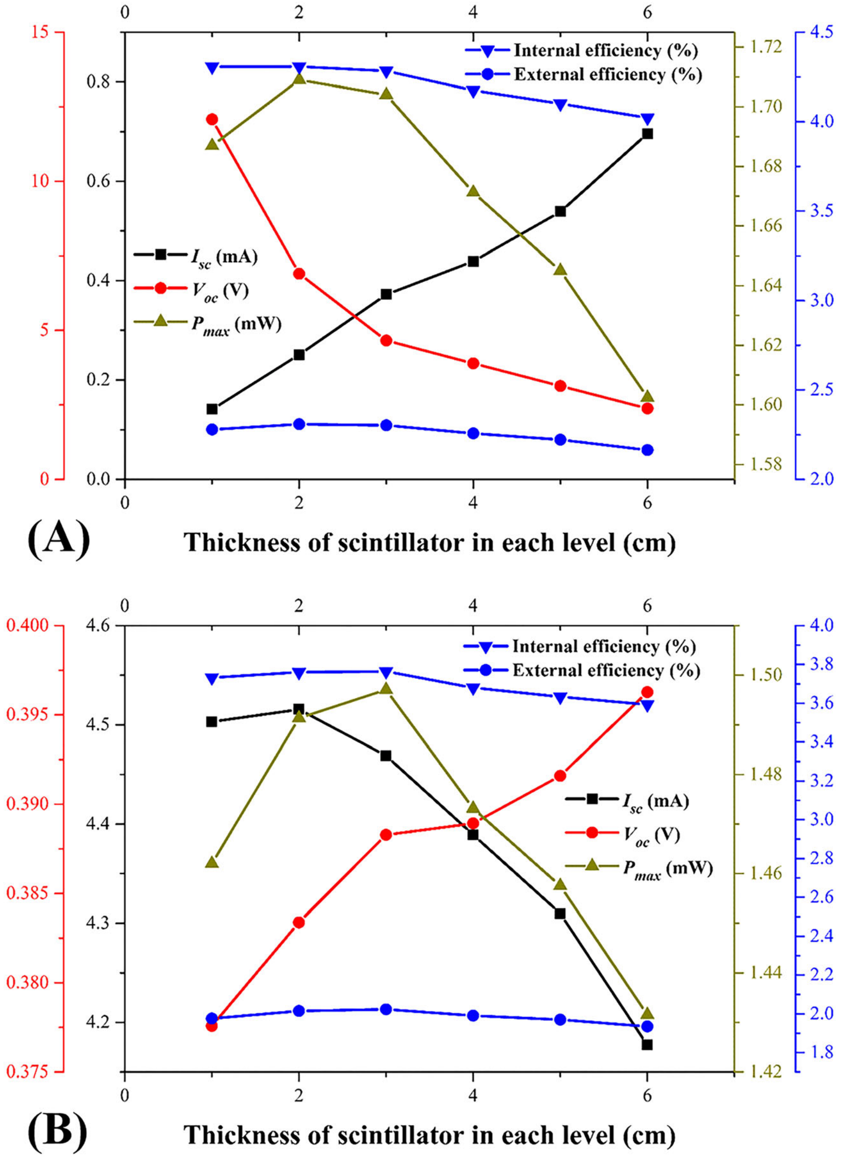

FIGURE 6 A, Calculation results for each structure in series connection; B, calculation results for each structure in parallel connection [Colour figure can be viewed at wileyonlinelibrary.com]

series, which makes it easier to obtained high V_{oc}, *FF*, and P_{max}. The electrical performance of the multilevel isotope batteries was measured at dose rate site of 0.103 kGy/hr. Figure 7 and Table 2 show the test results.

The measurement results show that the multilevel isotope batteries obtained considerable output performance after the optimized design. At the dose rate of 0.103 kGy/hr, the multilevel isotope batteries obtain the I_{sc} of 1.69 μA, V_{oc} of 7.77 V, P_{max} of 7.98 μW, and η of 0.15% when the volt layers are connected in series. The η increased an order of magnitude. The voltage output of these isotope batteries can meet the voltage requirements of many electronic devices. As such, these batteries have a strong ability to independently drive the electronic equipment in a specific field. The measured efficiency of the batteries is far less than that in the theoretical

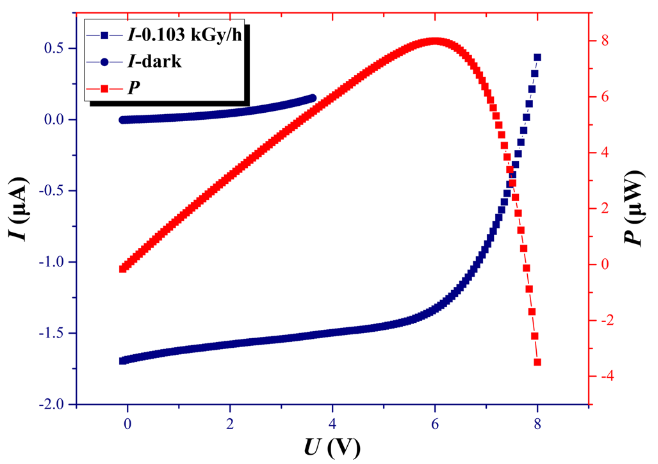

FIGURE 7 Performance test result at dose rate of 0.103 kGy/hr [Colour figure can be viewed at wileyonlinelibrary.com]

TABLE 2 Electrical parameters of multilevel isotope batteries at 0.103 kGy/hr

Parameter	I_{sc}, μA	V_{oc}, V	*FF*	P_{max}, μW	*η*
Value	1.69	7.77	0.61	7.98	0.15%

calculations, partly due to some idealized assumptions in the calculation process. Nonetheless, the battery performance has been greatly improved.

5 | CONCLUSIONS

In this work, the energy conversion materials and multilevel structures were optimized. Due to the higher photon yield, the CsI scintillator applied in dual effect multilevel isotope batteries achieved better electrical performance than the LYSO scintillator. The spectral matching of the scintillator and the volt layer is an important factor that affects the performance of the batteries. It was found that the CsI scintillator combined with the GaAs volt layer are suitable for the RV/RPV dual effect isotope batteries. The multilevel structure almost achieves full utilization of ^{60}Co γ rays when the scale reaches approximately 20 cm. With the increasing thickness of each level of the scintillator, the total P_{max} of the multilevel isotope battery increases first and then decreases. The output power came up to the peaked value when the thickness of each level scintillator reached 2 cm and the volt layer was connected in series.

Based on the optimized design of material and structure, the RV/RPV dual effect multilevel isotope batteries were prepared and finally obtained a huge improvement in battery performance with V_{oc} of 7.77 V, I_{sc} of 1.69 μA, P_{max} of 7.98 μW, and η of 0.15% under the 0.103 kGy/hr ^{60}Co source. The high open current voltage could meet the demands of many low-power electronic devices. This kind of isotope batteries is very promising as a new energy source for low-power devices in the military, deep space exploration, and other fields.

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