

Fabrication Study of Tritium-Integrated 4H-SiC Betavoltaic Cells

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Abstract

To improve the performance of radiovoltaic batteries, the effects of depletion region width, diffusion length, and electrode structure on output power were investigated, and through design optimization and process improvements in radioactive source thickness, transducer device structure, electrodes, and other aspects, an integrated tritiated titanium-SiC-based radiovoltaic battery with integrated radioactive source and transducer device was fabricated. Compared with the test results of non-integrated tritiated titanium-SiC-based batteries, the integration of tritiated titanium with the transducer device significantly improved the output performance of the radiovoltaic battery, with a maximum output power reaching 21.4 nW, which is at a relatively high level among similar types of radiovoltaic batteries reported in the literature. This work provides a valuable reference for the fabrication of high-performance radiovoltaic batteries.

Full Text

Fabrication of a 4H-SiC Betavoltaic Battery with Integrated Tritiated Titanium Source

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Abstract

To enhance the performance of betavoltaic batteries, we investigated the effects of depletion region width, diffusion length, and electrode structure on output power. By optimizing the design and improving the fabrication processes related to radioisotope source thickness, converter structure, and electrodes, we successfully developed an integrated tritiated titanium-SiC-based betavoltaic battery. Compared with test results from discrete tritiated titanium sources and converters, the integrated approach significantly improved the output performance of the betavoltaic battery, achieving a maximum output power of 21.4 nW, which is among the highest reported values for similar betavoltaic batteries. This work provides a valuable reference for developing high-performance betavoltaic batteries.

Keywords: betavoltaic batteries; 4H-SiC; tritiated titanium

Introduction

Betavoltaic batteries, which consist of a radioisotope source and a semiconductor converter, convert radioactive decay energy into electrical energy by generating and collecting electron-hole pairs. These batteries offer numerous advantages including long lifetime, high energy density, easy miniaturization, and strong anti-interference capability, making them promising for self-powered microdevices requiring low power, small volume, and long service life [1]. When selecting radioisotope sources, solid-state tritium (^3H) and ^{63}Ni sources that emit low-energy β particles are generally preferred to minimize lattice damage to the converter. Among common solid tritium sources, tritiated titanium (TiT_x , where x ranges from 1.5 to 1.8) is most widely used due to its high tritium content and strong thermal stability, while ^{63}Ni sources are typically electrodeposited nickel foils with ^{63}Ni isotopic abundance of 10%-20%. $\text{TiT}_{1.5}$ exhibits a maximum surface emission power density of approximately 1 W/cm², significantly higher than 0.4 W/cm² for ^{63}Ni [2], thus providing greater input power for betavoltaic batteries. Additionally, tritium costs about \$3.5 per curie internationally, nearly one-thousandth the price of ^{63}Ni (approximately \$4000 per curie), offering better economic viability [3]. Therefore, tritiated titanium represents a superior radioisotope source choice for betavoltaic batteries in terms of both power density and cost-effectiveness.

According to Olsen's theory that betavoltaic device efficiency is approximately positively correlated with the converter's bandgap [4], wide-bandgap semiconductor materials such as SiC and GaN have attracted significant attention for

betavoltaic applications [5, 6]. Among these, 4H-SiC benefits from relatively mature fabrication processes and demonstrates more stable device performance, making it the preferred converter material [7, 8].

Since Rappaport's first reported betavoltaic battery in 1954 [9], extensive theoretical and experimental research has been conducted. Theoretical studies have shown that PIN structures based on wide-bandgap semiconductors can achieve high device efficiency and output power, guiding the optimization of source and converter structures [10]. However, existing theoretical assumptions are often overly idealized and fail to adequately consider the actual material properties of radioisotope sources and semiconductor converters, causing theoretical predictions to significantly exceed experimental results [11-15]. For instance, theoretical predictions for Si-based [12] and GaN-based [13] betavoltaic batteries forecast short-circuit current densities and open-circuit voltages of 546.8 nA/cm², 0.23 V and 56 nA/cm², 0.48 V, respectively, while corresponding experimental values are only 77.5 nA/cm², 0.12 V and 2 nA/cm², 0.025 V—demonstrating substantial discrepancies.

Electron guns are widely used for simulated testing of betavoltaic batteries due to their flexible adjustable parameters including irradiation area, electron energy, and flux [16]. However, electron beams lack self-absorption effects and exhibit monoenergetic, unidirectional emission, unlike actual radioisotope sources with self-absorption, continuous energy spectra, and isotropic emission. Consequently, electron beams deposit more energy in the converter, leading to significantly overestimated performance measurements [10, 16]. For example, Zhao et al. found that using an electron gun with equivalent energy and beam intensity to simulate a ⁶³Ni source for testing 4H-SiC converters yielded an output power density of 15-20 nW/cm², substantially higher than the experimental value of 4-5 nW/cm² obtained with an actual ⁶³Ni source, with device efficiency also 20.6%-32% higher [16].

In experimental studies using actual radioisotope sources, short-circuit currents and output powers are generally low. For example, reported 4H-SiC-based betavoltaic batteries exhibit short-circuit currents and output powers of only 0.022-0.51 nA and 0.01-0.32 nW, respectively [17-19]. While devices reported in [15, 16] showed improved performance with short-circuit currents of 2.5-3.2 nA and output powers of 4.28-5.19 nW, the output power remained below 10 nW. The primary reason is that the ⁶³Ni sources used had low surface activity of only 0.28 mCi/cm², yielding a surface emission power density of 33.7 nW/cm²—significantly lower than the ⁶³Ni saturation value of 0.4 W/cm²—indicating substantial room for improvement. Replacing ⁶³Ni with tritiated titanium sources would provide greater source activity and surface emission power density. Thomas et al. [1] tested betavoltaic batteries using higher-activity tritium sources combined with 4H-SiC converters. In their work, a tritiated titanium source with surface activity of approximately 15 mCi/cm² achieved a surface emission power density of about 732.9 nW/cm². When the source and converter were tested separately, high performance metrics were obtained

with short-circuit current density and device efficiency reaching 75.47 nA/cm² and 18.6%, respectively. However, when the source and converter were integrated—by first fabricating the converter, then depositing a titanium film on its surface, and finally absorbing tritium—the device efficiency decreased to approximately 12%. Unfortunately, that work did not provide geometric dimensions of the betavoltaic device, preventing determination of the absolute maximum output power, and other similar experimental studies have failed to reproduce such high performance metrics. Notably, most experimental studies have used discrete tritium [20, 21] or nickel [17-19] source foils, highlighting the need for enhanced research on source-converter integration to meet practical application requirements for higher stability.

In summary, to improve betavoltaic battery output performance and meet practical application demands, research must focus on employing radioisotope sources with high surface emission power density, PIN-structured wide-bandgap converters, and integrated source-converter designs. Based on these considerations, this work employs tritiated titanium sources and 4H-SiC converters with optimized design and improved fabrication processes to prepare integrated betavoltaic batteries, followed by performance testing and analysis.

1. Theoretical Design of Betavoltaic Batteries

The working principle of betavoltaic batteries involves β particles incident on the converter generating electron-hole pairs through ionization; these pairs then drift, diffuse, and recombine under the built-in electric field, producing radiation-induced current ($J\beta$) and forward current (JF). These currents are collected by electrodes to drive loads, achieving conversion of decay energy to electrical energy, as illustrated in Figure 1 [Figure 1: see original paper]. The output performance of betavoltaic batteries is primarily determined by the radioisotope source's surface emission power density and device efficiency, making theoretical calculations of the correlation between structural parameters and output performance crucial for optimizing source and converter designs. In our theoretical calculations, we employ 4H-SiC as the converter material and $\text{TiT}_{1.5}$ as the radioisotope source, with device dimensions of 1 cm $\times$ 1 cm. By combining Monte Carlo and numerical simulation methods, we optimize structural parameters including source thickness, electrode structure, and epitaxial layer thickness and doping concentration to achieve optimal battery design and guide experimental fabrication.

1.1 Radioisotope Source and Electrode Design

Monte Carlo simulations were used to calculate the surface emission power and source efficiency of $\text{TiT}_{1.5}$ sources, as shown in Figure 2(a) [Figure 2: see original paper]. Due to self-absorption effects in $\text{TiT}_{1.5}$, source efficiency decreases with increasing source thickness, while surface emission power density initially

increases before saturating. When thickness increases from 0.5 μm to 1 μm , surface emission power density increases by only 7.2%. Therefore, considering both self-absorption and surface emission power density, the $\text{TiTi}_{1.5}$ thickness was set to 0.5 μm .

In electrode design for betavoltaic batteries, top electrode absorption of β particles should be minimized to reduce ineffective energy loss. This work employs a mesh structure with small area coverage for the top electrode. Based on fabrication processes for P-type ohmic contacts with low specific contact resistance ($\sim 10^{-5} \Omega \cdot \text{cm}^2$) [22], the electrode material stack of Ni/Ti/Al/W was selected with total thickness of 220 nm and single electrode line width of 20 μm . To analyze the effect of electrode structure on energy deposition loss in the converter, we define the energy deposition loss rate as $\eta = (E_0 - E_1)/E_0$, where E_0 and E_1 represent energy deposition in the converter without and with electrodes, respectively. Figure 2(b) [Figure 2: see original paper] shows the impact of mesh electrodes with different line counts and full-coverage electrodes on η . The loss rate increases with electrode line count, with full-coverage P-type electrodes causing approximately 88.4% loss, while mesh electrode structures with 4×4 , 8×8 , 12×12 , 16×16 , and 20×20 line distributions per cm^2 effectively reduce η . Therefore, electrode designs should employ mesh structures with line counts below $16 \times 16 \text{ per cm}^2$, yielding η below 4.3% and significantly reducing electrode-induced energy loss.

1.2 Converter Optimization Design

In this study, the 4H-SiC converter adopts a PIN structure. According to drift-diffusion theory, minority carriers located within the PN junction depletion region or within one diffusion length from it can be effectively collected. This region is defined as the effective charge collection region (ECR) with length $H_{\text{ECR}} = (W_d + L_n + L_p)$. To expand H_{ECR} , a “concentration-gradient I-layer” is inserted to form a PIN junction. Compared with PN junctions, the additional electric field region at the I/N interface in PIN structures facilitates electron-hole pair separation, increasing the effective charge collection region length to $H_{\text{ECR}} = (W_d + L_n + 2L_p + L_{p1})$, thereby effectively reducing recombination losses, as shown in Figure 1. Here, W_d , L_n , L_p , and L_{p1} represent the depletion region width, electron diffusion length, hole diffusion length in the I-region, and hole diffusion length in the N-region, respectively. Converter structural parameters include P-layer thickness, I-layer thickness, P-layer doping concentration (N_A), I-layer doping concentration (N_D), and N-layer doping concentration (N_d). Optimizing these parameters enables optimal betavoltaic battery design.

(1) Output Performance of Betavoltaic Batteries

The maximum output power (P_m) of a betavoltaic battery can be expressed as:

$$P_m = I_{SC} V_{OC} FF \quad (1)$$

where I_{SC} , V_{OC} , and FF are the short-circuit current, open-circuit voltage, and fill factor, respectively. The short-circuit current can be expressed as:

$$I_{SC} = q \int G(x) CE(x) \quad (2)$$

where q is the electron charge; H is the converter thickness; $G(x)$ is the electron-hole pair generation rate, expressed as $G(x) = A \cdot E_{dep}(x) /$, where A , $E_{dep}(x)$, and are the radioisotope source activity, energy deposition of a single β particle in the converter, and the average ionization energy of ${}^4\text{H-SiC}$ (6.88 eV [23]), respectively; $CE(x)$ is the carrier (electron and hole) collection efficiency. Within the depletion region, electron-hole pairs are considered to be effectively separated and collected by the electric field, so $CE(x)$ is taken as 100% in this region. Outside the depletion region, electron-hole pairs undergo diffusion and recombination, with collection probability dependent on their generation location and diffusion length. $CE(x)$ is given by:

$$CE(x) = 1 - \tanh \quad (3)$$

where $d(x)$ represents the distance between the electron-hole pair generation location and the depletion region boundary, and L is the minority carrier diffusion length determined by minority carrier mobility and lifetime (expressions given in reference [25]). Thus, $CE(x)$ is inversely correlated with the distance from the depletion region boundary and positively correlated with diffusion length. Larger depletion region widths and diffusion lengths enhance carrier collection and improve output performance.

Under ideal diode conditions, the open-circuit voltage can be expressed as:

$$V_{OC} = \frac{kT}{q} \ln \left(\frac{I_{SC}}{J_0} + 1 \right) \quad (4)$$

where k is the Boltzmann constant (1.38×10^{-23} J/K), T is the temperature (300 K in this work), and J_0 is the reverse saturation current (expression given in reference [24]).

In summary, increasing both short-circuit current and open-circuit voltage enhances maximum output power. Since short-circuit current correlates with $CE(x)$, increasing depletion region width and diffusion length raises $CE(x)$ and thus short-circuit current. Additionally, increased short-circuit current also improves open-circuit voltage to some extent. Therefore, enhancing $CE(x)$ is a key factor in improving betavoltaic battery performance.

(2) Epitaxial Layer Thickness Optimization

In optimizing epitaxial layer thickness, ineffective energy loss should be minimized while carrier collection efficiency is maximized. In PIN converters, the I-layer and P-layer are typically epitaxially grown on an N-type substrate, with the P-layer and N-layer heavily doped to facilitate ohmic contact formation, while the I-layer is lightly N-type doped [1, 16, 25].

Since the P-layer is heavily doped, the depletion region width is very narrow in this region, and generated electrons and holes readily recombine, making it an ineffective energy deposition region. The electron-hole pair generation rate decays quasi-exponentially with incident depth (see Figure 3(a) [Figure 3: see original paper]), and electron-hole pairs generated by tritiated titanium sources are predominantly distributed near the surface region. Therefore, recombination in this region severely impacts battery performance, and the P-layer should be as thin as possible. Based on current 4H-SiC epitaxial process capabilities, the P-layer thickness is typically set to 0.2 μm [1, 2].

The maximum β particle energy from $\text{TiT}_{1.5}$ is 18.7 keV, and Geant4 calculations show the range of such β particles in 4H-SiC is approximately 2.2 μm . In $\text{TiT}_{1.5}$ -4H-SiC betavoltaic batteries, setting the I-layer thickness greater than the β particle range can improve carrier collection efficiency. In 4H-SiC device fabrication, epitaxial processes for 10 μm -thick I-layers are well-established [16]; therefore, this work sets the converter I-layer thickness to 10 μm .

(3) Epitaxial Layer Doping Concentration Optimization

Epitaxial layer doping concentrations affect both depletion region width and minority carrier diffusion length. To form good ohmic contacts, the P and N regions require heavy doping, with P-layer doping concentration typically set to $N_A = 10^{19} \text{ cm}^{-3}$ [1, 15]. The N-layer doping concentration N_d is usually greater than $1 \times 10^{18} \text{ cm}^{-3}$, and is set to $N_d = 2 \times 10^{18} \text{ cm}^{-3}$ in this work. Therefore, optimization of epitaxial layer doping concentration primarily focuses on the I-layer doping concentration N_D .

Using formulas from reference [25], the correlation between depletion region width (W_d) and minority carrier diffusion lengths (L_n , L_p) with doping concentration can be calculated. Figure 3(b) [Figure 3: see original paper] shows the calculated relationship between depletion region width and I-layer doping concentration N_D , revealing that W_d increases with decreasing doping concentration. Figure 3(c) [Figure 3: see original paper] shows the correlation between minority carrier diffusion lengths and P-layer and N-layer doping concentrations, demonstrating that L_n and L_p increase with decreasing doping concentration. Therefore, lower doping concentrations enhance electron-hole pair collection efficiency and short-circuit current. Based on current low-doping process capabilities for 4H-SiC, the I-layer doping concentration is set to $N_D = 10^{14} \text{ cm}^{-3}$.

In summary, the optimized betavoltaic battery design in this work employs: a

0.5 μm -thick $\text{TiT}_{1.5}$ radioisotope source, mesh electrodes, and a PIN-structured 4H-SiC converter with heavily doped P and N layers and lightly doped I-layer. The P-region thickness is set to 0.2 μm , the I-region to 10 μm , and the doping concentrations of the P, I, and N regions are set to 10^{19} , 10^{14} , and $2 \times 10^{18} \text{ cm}^{-3}$, respectively.

2. Experimental Fabrication and Output Performance Testing

The fabrication process for betavoltaic battery converters primarily includes epitaxy, photolithography, etching, passivation, electrode preparation, and annealing. According to the optimization criteria in Section 1.1, this work employs a 350 μm -thick N-type 4H-SiC substrate with doping concentration of $2 \times 10^{18} \text{ cm}^{-3}$. Using chemical vapor deposition (CVD), a 10 μm -thick N-type lightly doped intrinsic layer (I-layer) with doping concentration of $1 \times 10^{14} \text{ cm}^{-3}$ is epitaxially grown, followed by a 0.2 μm -thick P-layer with doping concentration of $1 \times 10^{19} \text{ cm}^{-3}$. Inductively coupled plasma (ICP) etching is used to form 50 μm -wide trenches, and dry oxidation is performed on trench sidewalls to create passivation layers that isolate the active device region from its edges, with individual device dimensions of $1.75 \text{ cm} \times 0.44 \text{ cm}$. Passivation is performed using plasma-enhanced chemical vapor deposition followed by annealing in N_2 atmosphere to reduce surface leakage current.

Based on the electrode design in Section 1.1, Ni/Ti/Al/W is selected for the P-type ohmic contact electrode and Ti/Ni/Au for the N-type ohmic contact electrode [22]. To enhance current transport efficiency, the N-type electrode (back electrode) employs full coverage, while the P-type electrode uses a mesh structure to minimize β particle energy loss. The mesh P-type electrode Ni/Ti/Al/W (total thickness 220 nm) is fabricated via secondary photolithography and magnetron sputtering. On the backside electrode region, Ti/Ni/Au (total thickness 290 nm) is sequentially deposited by magnetron sputtering. Finally, stable ohmic contacts are formed by annealing at 950°C for 5 minutes in nitrogen atmosphere.

Based on the saturation thickness of 0.5 μm for tritiated titanium sources, a 0.5 μm -thick Ti layer is directly deposited on the P-layer of the 4H-SiC converter using magnetron sputtering, covering the mesh electrode. Using a low-pressure hydrogen storage system, ^3H is diffused into the Ti at 350°C to form the tritiated titanium source. The tritiated titanium layer thickness is measured to be approximately 0.5 μm using a step profiler, as shown in Figure 4(b) [Figure 4: see original paper]. Through these processes, an integrated betavoltaic battery with source and converter combined (hereafter “integrated battery”) was successfully fabricated.

I-V characteristics of the integrated battery were measured using a Keithley 2635B semiconductor analyzer, with results shown in Figure 5(a) [Figure 5: see

original paper]. The measured short-circuit current, open-circuit voltage, maximum output power, and fill factor are 20.7 nA, 1.26 V, 21.4 nW, and 82.5%, respectively. To fully utilize the energy from the tritiated titanium source, a non-integrated 4H-SiC converter was placed on top of the integrated battery, enabling a solid angle approaching 4π between the source and both converters. The measured I-V curve is shown in Figure 5(b) [Figure 5: see original paper], yielding a short-circuit current, open-circuit voltage, output power, and fill factor of 8.12 nA, 1.5 V, 8.76 nW, and 71.9%, respectively. These values are 154.9%, 144.3%, and 14.7% lower than those of the integrated battery for short-circuit current, output power, and fill factor, respectively. The primary reasons are: during tritium diffusion into Ti to form the tritiated titanium source, ^3H is predominantly distributed at the bottom of the Ti layer with relatively less at the top, resulting in lower surface activity from the top surface and consequently lower short-circuit current, output power, and fill factor compared to the integrated battery. Additionally, the tritiated titanium source on the integrated battery and the top converter are discrete with approximately 1 mm separation, resulting in a smaller emission solid angle than in the integrated configuration and potentially reduced output performance. Furthermore, β particles emitted from the tritiated titanium source undergo ionization interactions with air before entering the converter, causing additional power loss. These results demonstrate that integrating high surface-emission-power tritiated titanium sources with converters significantly enhances betavoltaic battery performance.

The maximum output power of the integrated 4H-SiC-based betavoltaic battery fabricated in this work is among the highest reported values (0.01-5.19 nW) for similar devices [15-19]. Notably, the fill factor of 82.5% achieved by our integrated battery is also among the highest reported values (50%-87.27%) for similar betavoltaic batteries, fully demonstrating that our device fabrication process and parameter selections are appropriate and that the source-converter integration technology has achieved excellent results. In summary, by employing a high surface-emission-power-density tritiated titanium source with a 4H-SiC converter, optimizing source thickness, electrode structure, and converter design, and improving fabrication processes, we have successfully fabricated high-performance betavoltaic batteries.

3. Conclusion

This study comprehensively considered the effects of radioisotope source surface emission power density, electrode structure, epitaxial layer thickness, and doping concentration on betavoltaic battery performance, designing and fabricating an integrated tritiated titanium-SiC betavoltaic battery. Through optimized integration design and improved fabrication processes, the fabricated battery demonstrated excellent performance with key metrics as follows: short-circuit current of 20.7 nA, maximum output power of 21.4 nW, and fill factor of 82.5%. Compared with non-integrated tritiated titanium-SiC batteries, the integrated

design improved short-circuit current, output power, and fill factor by 154.9%, 144.3%, and 14.7%, respectively. Moreover, the maximum output power represents a significant improvement over previously reported best results. This study demonstrates that employing integrated, high surface-emission-power-density radioisotope sources combined with optimized source thickness, electrode structure, and converter design is an effective approach for enhancing betavoltaic battery output power, providing valuable guidance for further performance improvements.

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Author Contributions

Hou-Jun He: Conceptualization, theoretical calculations, experimental fabrication, experimental testing, data analysis, manuscript drafting.

Yun-Cheng Han: Supervision, manuscript review and revision.

Xiao-Yu Wang: Manuscript review, editing, and revision.

Lei Ren: Manuscript editing and revision.

Xiang-Dong Meng: Manuscript revision.

Ming-Jie Zheng: Guidance, revision, and review.

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