

Exploratory study of betavoltaic battery using ZnO as the energy converting material

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Abstract Third-generation-semiconductor zinc oxide is utilized as an energy converting material in a betavoltaic battery, where 0.06 Ci ⁶³Ni and 8 Ci ¹⁴⁷Pm are used as the beta sources. Based on a Monte Carlo simulation, the full scales of the devices are derived as 17 and 118 µm, respectively, for both sources. The influences of semiconductor doping concentrations on the electrical properties of the devices are analyzed. For a typical doping concentration $N_{\rm A}$ = 10¹⁷ cm⁻³, $N_{\rm D}$ = 10¹⁶ cm⁻³, the conversion efficiencies are 7.177% and 1.658%, respectively, using ⁶³Ni and ¹⁴⁷Pm sources. The calculation results of energy deposition in materials for the two sources show that the doping concentrations drop to 1 × 10¹³–5×10¹⁴ cm⁻³ and 1 × 10¹²–5×10¹³, and accordingly, the energy conversion efficiencies rise to 14.212% and 18.359%, respectively.

Keywords Radioisotope · Beta voltaic effect · Zinc oxide · Nuclear battery · Monte Carlo simulation

1 Introduction

The nuclear battery is a device that uses energy from the decay of a radioactive isotope to generate electricity. It has a long life, high energy density, and is able to withstand

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extreme environmental conditions, and has thus been widely used in the aerospace industry, deep-sea and polar regions, heart pacemakers, micro-electromotors, and in other such applications [1].

Compared to other types of nuclear batteries, betavoltaic batteries are particularly well suited to low-power electrical applications due to their ultra-high energy density, ultralong lifetime and convenience of integration and miniaturization [2]. In order to enhance out power and reduce radiation damage, wide-bandgap semiconductors (SiC [3–5], GaN [6, 7]) and different energy conversion structures [8, 9] have been employed. However, the growth of SiC and GaN are relatively expensive and require high temperatures. In this paper, we attempt to apply the widebandgap semiconductor zinc oxide, which is more economical and easier to grow, as the energy conversion material. The most suitable p–n junctions and the corresponding electrical properties are acquired and reported here.

ZnO, with a direct band gap of 3.37 eV, along with SiC and GaN, is a third-generation semiconductor. ZnO films have high transparence and electrical conductivity, and are thus promising materials for optoelectronic applications, high-speed and space devices [10, 11]. Table 1 shows some parameters of ZnO and several other third-generation semiconductors, where the value E_g/E_{c-h} represents the most productive route for energy conversion, and is regarded as the theoretical efficiency limitation [12]. The comparison reveals that SiC has a narrower band gap, lower density and electron density. As shown in previous research [13, 14], lower density and electron density lead to a lower backscatter effect but longer electron range, and consequently enlarge the scales of devices and less energy deposition per unit, which is unfavorable for a practical

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Materials	4H-SiC	GaN	ZnO
Crystal structure	Wurtzite	Wurtzite	Wurtzite
Energy gap (eV)	3.26	3.39	3.37
Melting point (°C)	2810	> 2500	1975
Thermal conductivity $(W \cdot cm^{-1} \cdot K^{-1})$	4.9	1.5	1.16
Dielectric constant	9.6	8.9	8.1/9.0
Electron saturation velocity (cm·s ⁻¹)	2.7×10^{7}	2.5×10^{7}	3.2×10^{7}
$E_{\rm BR}$ (MV/cm)	3.0-5.0	2.6-3.3	3.5
Density $(g \cdot cm^{-3})$	3.2	6.09	5.605
Average atomic number	20	38	38
Relative molar mass	40.1	83.73	81.38
Electron density ($\times 10^{23}$)	9.61	16.64	15.76
Mean ionization energy E_{e-h} (eV)	8.4	10.3	About 9.9
E_{g}/E_{e-h}	0.388	0.329	0.340

Table 1 Parameters of thirdgeneration semiconductors

battery. ZnO and GaN have similar bandgaps, densities and electron densities. ZnO has a higher E_g/E_{e-h} value than GaN, which implies more electron-hole pairs produced by the betavoltaic effect and higher efficiency. Additionally, the much lower growth temperature and epitaxial growth of ZnO on native substrates [15] makes it easier to grow high-quality thin films than GaN. Along with its higher radiation resistance than Si, GaAs, CdS and GaN, its high break-down strength and low cost, ZnO is especially favorable for applications in radiation environment [16].

In order to fabricate a p-n junction diode nuclear battery, high performance and stable p-n junction diodes are necessary. However, fabrication of p-type ZnO has been a major challenge in the past because ZnO is normally an intrinsic n-type semiconductor [11, 15]. Recent successes in p-type doping of ZnO and considerable efforts devoted to the fabrication of ZnO homojunction diodes with positive evolvements have made it possible to fabricate ZnObased nuclear batteries by magnetron sputtering [17], spray pyrolysis [18] and pulsed laser deposition methods [19, 20]. Meanwhile, the technique of p- and n-ZnO using only one kind of dopant (Na element) in a single layer of NZO film has recently been developed [20]. In addition, the stability of the junction has been significantly improved, and extends up to 1 year without any obvious degradation [21]. Thus, ZnO is a potential energy converting material for nuclear batteries.

2 Choice of isotopes for nuclear batteries

2.1 Choice of radiation types

Some parameters of the most common beta sources are shown in Table 2. Pure β sources are typically utilized in micro-nuclear batteries because β particles have moderate

 Table 2
 Radioactive source parameters

Source	Radioactivity	Half-life (y)	$E_{\rm max}$ (keV)	$E_{\rm ave}~({\rm keV})$
³ H	β	12.33	18.6	5.7
⁶³ Ni	β	100.1	66	17.48
¹⁴⁷ Pm	β	2.62	224.5	59
¹³⁷ Cs	β, γ	30.17	1176	188.4
⁹⁰ Sr	β	28.8	546	195.8

mass, energy and transport length scale. Massive α particles with higher energy may cause severe radiation damage in materials, and gamma rays with long ranges in matter are difficult to shield. Considering the lifetime of the battery, the isotope should have a relatively long half-life without associated gamma rays, and its maximum energy should be below the lattice damage threshold of the energy converting material. Other factors such as state and price should also be taken into account. Among the radioactive sources listed below, ³H is in the gaseous state, which limits its scope of application. ¹³⁷Cs emits gamma rays, increasing the difficulty of shielding. With respect to ⁹⁰Sr, solid ⁶³Ni and ¹⁴⁷Pm have moderate energies and half-lives, and are therefore chosen as the radioactive sources.

2.2 Choice of source radioactivity

Beta particles can be absorbed by the sample material itself, a phenomenon called self-absorption [22]. Due to this effect, surface radioactivity initially increases with increasing sample thickness, but remains more or less unchanged beyond a particular thickness. A source with extra thickness is not only a waste of radiation energy but also leads to a rise in temperature and degradation of the nuclear battery. To reduce these effects and guarantee the efficiency of the battery, proper source thicknesses are required.

In this work, a $1 \text{ cm} \times 1 \text{ cm}$ rectangular source with a variable height is used in the simulation. The relationship between the height of the source and absolute activity is given by:

$$m = \rho \mathrm{SH} = \frac{\mathrm{MAT}_{1/2}}{N_{\mathrm{A}} \ln 2},\tag{1}$$

where *M* is the molecular mass (g/mol), *A* the absolute activity (Bq), $T_{1/2}$ is the half-life (s) and *S* is the superficial area (1 cm²). The self-absorption rate η can be defined as

$$\eta = \frac{P_{\rm in}}{P_{\rm total}},\tag{2}$$

in which $P_{\rm in}$ is the deposition power in the source and $P_{\rm total}$ is the total power. The surface radioactivity and self-absorption rate versus mass thickness are shown in Fig. 1. From the plots, a proper mass thickness (1.071507 mg·cm² for ⁶³Ni and 8.630406 mg·cm² for ¹⁴⁷Pm) can be selected according to the relatively high surface power and relatively low self-absorption rate. The surface powers are 1.37718 μ W and 826.08 μ W for ⁶³Ni and ¹⁴⁷Pm, respectively. Other calculation results are shown in Table 3.

The existence of self-absorption effect also leads to a shift in surface average energy, which is shown in Fig. 2. As the mass thickness increases, the increment in the surface average emitting energy gradually reduces, and the whole curve lies above the theoretical average energy line. This is because low-energy beta particles are absorbed by the source itself, and only particles with higher energies or those that are emitted near the surface can penetrate through the source body. When the source become thick enough, particles with the highest energy cannot be emitted from the source, so the surface average energy remains almost unchanged.

Table 3 Select parameters of sources

Parameters	⁶³ Ni	¹⁴⁷ Pm
Thickness (µm)	1.20394	13.31853
Mass thickness (mg·cm ²)	1.071507	8.630406
Absolute radioactivity (Ci)	0.06	8
Surface activity (single side) (Ci)	0.011155	1.674825
Surface power (single side) $(\mu W \cdot cm^2)$	1.37718	826.08
Surface average emission energy (keV)	20.83	83.21
Self-absorption rate (%)	55.93	54.10

3 Electron range and backscatter coefficient

The penetration depth and backscatter coefficient are simulated by CASINO v2.4.8.1. A monoenergetic beam of one thousand electrons is used to impinge vertically in the simulation, along with minimum electron energy of 0.01 keV. Electron ranges and backscatter coefficients are shown in Table 4. The maximum penetration depth restricts the full scale of the device, and accordingly, electrons with maximum energy should be completely shielded.

4 Energy deposition in ZnO

The simulation model is presented in Fig. 3. The thickness from the upper surface of the source to the stratification plane is variable, and energy deposition in this area is known. The energy deposition versus depth is shown in Fig. 4. For both sources, energy depositions initially increase as the depth of the sample increases. Beyond a certain depth, the energy deposition remains almost unchanged, where the energy of the source has been completely deposited. These depths are around 6 μ m and



Fig. 1 (Color online) Relation among the mass thickness, surface power and self-absorption rate. a ⁶³Ni source; b ¹⁴⁷Pm source



Fig. 2 Average energy shift. **a** 63 Ni source; **b** 147 Pm source

Table 4 Penetration depths andbackscatter coefficients	Source	⁶³ Ni	⁶³ Ni		¹⁴⁷ Pm	
		$E_{\rm average}$	$E_{\rm max}$	$E_{\rm average}$	E _{max}	
	Energy (keV)	17.48	66	59	224.5	
	Maximum penetration depth (µm)	1.40724	16.9041	12.2443	117.92	
	Backscatter coefficient	0.246	0.188	0.214	0.157	





 $40~\mu m,$ and the maximum deposited energies are about 2.96 and 13.88 keV for $^{63}\rm{Ni}$ and $^{147}\rm{Pm}$ sources, respectively.

However, not all deposited energy can be utilized to form electron-hole pairs, and the main effective coverage is the depletion layer. In a universal prototype of a nuclear battery, the source is set on the p-type doping layer. The effective deposition energy is the difference between the total deposition energy (i.e., the sum of the energy



Fig. 4 Energy deposition in ZnO. a 0.6 Ci ⁶³Ni source, b 8 Ci ¹⁴⁷Pm source

depositions in the depletion layer and p-type layer) and the p-type layer deposition energy. According to J.G. Lu et al. [23], the thickness of the p-type layer is selected as 300 nm, and then, the energies deposited in the p-type layer are 0.903 and 0.608 keV for ⁶³Ni and ¹⁴⁷Pm, respectively.

5 Build-in potential and depletion width

In a betavoltaic battery, most of the electron-hole pairs are produced in the depletion region. Electrons and holes are separated and collected by the build-in potential. In order to effectively utilize the beta source energy, the depletion width should be matched with the energy deposition range. In the equilibrium condition, the build-in potential for a p-n junction is given by the following relation [24]:

$$V_{\rm bi} = \frac{kT}{q} \cdot \ln\left(\frac{N_{\rm A}N_{\rm D}}{n_i^2}\right),\tag{3}$$

where $\frac{kT}{q}$ is about 0.0259 V at 300 k. N_A and N_D are doping concentrations of p and n (cm⁻³), respectively. At ambient temperature, they come close to carrier intensities. n_i is the intrinsic carrier concentration.

The depletion width is given by [24]:

$$W = \sqrt{\frac{2\varepsilon_{\rm s}\varepsilon_0}{q}} \cdot \left(\frac{N_{\rm A} + N_{\rm D}}{N_{\rm A}N_{\rm D}}\right) V_{\rm bi},\tag{4}$$

where ε_s is the relative permittivity of ZnO, which is 8.75, and ε_0 is the vacuum permittivity.

In several factual cases, the intrinsic carrier concentrations of ZnO vary by multiple orders of magnitude in the literature, which severely affect $V_{\rm bi}$ and W. In previous studies, we once regarded n_i as 10^6 cm⁻³.

Here, a theoretical intrinsic carrier concentration is used, which is given by:

$$n_{\rm i} = (N_{\rm c}N_{\rm v})^{1/2} \exp\left(-\frac{E_{\rm g}}{2k_0T}\right),\tag{5}$$

where N_c and N_v are the effective state densities of the conduction and valence bands, respectively, which can be calculated by $N_c = 2 \frac{\left(2\pi m_n^* k_0 T\right)^{3/2}}{h^3}$ and $N_v = 2 \frac{\left(2\pi m_p^* k_0 T\right)^{3/2}}{h^3}$, where m_n^* and m_p^* indicate effective masses of electrons and holes, respectively. In ZnO material, m_n^* and m_p^* are 0.318 m_0 and 0.64 m_0 , respectively [25]. Calculations indicate that $N_c = 4.49 \times 10^{18} \text{ cm}^{-3}$, $N_v = 1.28 \times 10^{19} \text{ cm}^{-3}$, and $n_i = 4.23 \times 10^{-10} \text{ cm}^{-3}$.

The formulas in this work are only suitable for nondegenerate semiconductors, i.e., $N_{\rm D} \ll N_{\rm c}$ and $N_{\rm A} \ll N_{\rm v}$. For ZnO material, $N_{\rm D}$ and $N_{\rm A}$ should be no more than



Fig. 5 (Color online) Relationship between build-in potential and doping concentrations



Fig. 6 (Color online) Relationship between depletion width and doping concentrations

 1×10^{18} and 5×10^{18} cm⁻³, respectively. Figures 5 and 6 give the relationships of $V_{\rm bi}$ and W versus doping concentrations when $N_{\rm D}$ and $N_{\rm A}$ vary from the order of magnitude of 12–18. The build-in potential increases, while the depletion width decreases with increasing doping concentrations. The build-in potential and depletion width lie from 2.55 to 3.31 V and 0.062 to 70.27 µm, respectively, corresponding to the scopes of variation of the doping concentration mentioned previously.

Considering the energy deposition range above, we assume that the most suitable depletion widths for ⁶³Ni and ¹⁴⁷Pm sources are around 6–7 and 40–50 µm and the doping concentrations are within the ranges of 1×10^{13} – 5×10^{14} and 1×10^{12} – 5×10^{13} cm⁻³, respectively. The build-in potential is between 2.588 and 2.826 V. Compared with the typical doping level of ZnO material ($N_A \ge 10^{17}$ cm⁻³, $N_D \ge 10^{16}$ cm⁻³), the selected doping

concentrations are considerably low, which make depletion widths much wider to deposit more energy. For a p-n junction with $N_{\rm A} = 10^{17}$ cm⁻³ and $N_{\rm D} = 10^{16}$ cm⁻³, the build-in potential and depletion width are 3.086 V and 0.573 µm, respectively. In this condition, the energy deposited in the depletion layer is reduced to 937.95 eV for ⁶³Ni and 974.54 eV for ¹⁴⁷Pm, respectively.

6 Electrical performance simulation

In the following calculations, it is assumed that the electron-hole pair collection rate in the depletion region is 1, while that outside the depletion is 0. In other words, the electron-hole pairs outside the depletion region are neglected.

The short-circuit current of the betavoltaic battery can be determined as [26]:

$$I_{\rm SC} = \frac{\rm Aq}{E_{\rm e-h}} \cdot \sum_{n=1}^{n} {\rm CE}(n) \times E(n), \tag{6}$$

where CE(n) is the collection rate of the electron-hole pair in layer n of the sample, and, according to the assumption above, CE(n) = 1. E(n) is the deposition energy in layer n, A is the absolute radioactivity, q is the electron charge and E_{e-h} is the average ionization threshold.

Because the E_{e-h} of ZnO is not found in the literature, an empirical formula is used to estimate E_{e-h} [27]:

$$E_{\rm e-h} = 2.8E_{\rm g} + 0.5,\tag{7}$$

where E_g is the forbidden band gap, the E_{e-h} is obtained as 9.9 eV.

Another important factor that evaluates the performance of a cell is the reverse saturation current. The density of the reverse saturation current (in A/cm²) can be expressed by [28]:

$$J_0 = 1.5 \times 10^5 \exp(-E_{\rm g}/kT),\tag{8}$$

where k indicates the Boltzmann constant. The J_0 is fairly low and is 4.65×10^{-52} A/cm², demonstrating that the quality of the device is excellent.

The build-in potential is the theoretical limit of the open-circuit voltage, which can be expressed as:

$$V_{\rm oc} = V_{\rm bi} = \frac{kT}{q} \cdot \ln\left(\frac{N_{\rm A}N_{\rm D}}{n_i^2}\right). \tag{9}$$

One of the determining factors in the overall behavior of a betavoltaic battery is the fill factor (FF). A higher fill factor results in greater efficiency and brings the cell's output power closer to its theoretical maximum. The fill factor is defined as the quotient of the maximum output power ($P_{\rm m}$)

divided by the product of the open-circuit voltage (V_{oc}) and the short-circuit current (I_{sc}) [28]:

$$FF = \frac{P_{\rm m}}{V_{\rm oc} \cdot I_{\rm sc}}.$$
(10)

An empirical formula [24] is used to calculate the fill factor:

$$FF = \frac{v_{oc} - \ln(v_{oc} + 0.72)}{v_{oc} + 1},$$
(11)

where $v_{\rm oc} = V_{\rm oc}/(kT/q)$.

The maximum output power is given by [24]:

$$P_{\rm m} = FF \times V_{\rm oc} \times I_{\rm sc}. \tag{12}$$

There are many definitions for the conversion efficiency of a betavoltaic battery. In this work, the power conversion efficiency is defined as:

$$\eta = \frac{P_{\rm m}}{P_{\rm source}},\tag{13}$$

which is the so-called device conversion efficiency, where P_{source} indicates the surface power of the source.

7 Results and discussions

The selected p-n junctions and electrical properties are shown in Table 5. For the same radiation source with a typical doping concentration, it has larger $V_{\rm bi}$ but much smaller W than the selected doping concentrations, which leads to lower energy deposition, and smaller $I_{\rm sc}$, $P_{\rm m}$ and η . This effect is more evident when a ¹⁴⁷Pm source is used, because a large amount of energy is wasted outside the depletion layer. Energy wastage is also inevitable for the ⁶³Ni source. As the doping concentration decreases, the $V_{\rm bi}$ decreases slightly, while W and energy deposition increase much more, and $I_{\rm sc}$, $P_{\rm m}$ and η are larger. It is noteworthy that, for ⁶³Ni and ¹⁴⁷Pm sources, to sufficiently deposit the energy and avoid exorbitant bulk resistances, doping concentrations within the ranges of 1×10^{13} – 5×10^{14} and 1×10^{12} – 5×10^{13} cm⁻³, respectively, are appropriate.

8 Conclusion

In summary, zinc oxide is applied as an energy conversion material in a betavoltaic battery, where 0.06 Ci ⁶³Ni and 8 Ci ¹⁴⁷Pm sources are used. The full scales of the 0.06 Ci ⁶³Ni and 8 Ci ¹⁴⁷Pm devices are 17 and 118 μ m, respectively. For typical doping concentrations $N_{\rm A} = 10^{17}$ cm⁻³, and $N_{\rm D} = 10^{16}$ cm⁻³, the conversion efficiencies drop to 7.177% and 1.658% because the depletion layer is too narrow for energy deposition. The most suitable doping

Properties	p-n junctions with doping concentrations derived from this work		Typical doping concentrations	
	For ⁶³ Ni	For ¹⁴⁷ Pm	For ⁶³ Ni	For ¹⁴⁷ Pm
Doping concentration (cm ⁻³)	$1 \times 10^{13} - 5 \times 10^{14}$	1×10^{12} -5×10 ¹³	$N_A = 10^{17}, N_D = 10^{16}$	$N_A = 10^{17}, N_D = 10^{16}$
$V_{\rm bi}$ (V)	2.794-2.826	2.588-2.662	3.086	3.086
W (µm)	6–7	40–50	0.573	0.573
Energy deposition (eV)	2060	12,971	937.95	974.54
$I_{\rm sc}$ (µA)	0.07391	62.0512	0.03365	4.6620
$J_0 (\mathrm{A} \cdot \mathrm{cm}^{-2})$	4.651×10^{-52}	4.651×10^{-52}	4.651×10^{-52}	4.651×10^{-52}
$P_{\rm m}$ (μW)	0.1957-0.1981	151.660–156.211	0.0988	13.694
FF (%)	94.78–94.82	94.44–94.57	95.18	95.18
η (%)	14.212–14.381%	18.359–18.910%	7.177%	1.658%

Table 5 p-n junctions and electrical properties

concentrations are within the range of 1×10^{13} – 5×10^{14} cm⁻³ for ⁶³Ni and 1×10^{12} – 5×10^{13} for ¹⁴⁷Pm, and accordingly, the conversion efficiencies rise to 14.212% and 18.359%, respectively.

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