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Temperature effects on gallium arsenide $^{63}$Ni betavoltaic cell

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ABSTRACT

A GaAs $^{63}$Ni radioisotope betavoltaic cell is reported over the temperature range 70 °C to −20 °C. The temperature effects on the key cell parameters were investigated. The saturation current decreased with decreased temperature; whilst the open circuit voltage, the short circuit current, the maximum power and the internal conversion efficiency values decreased with increased temperature. A maximum output power and an internal conversion efficiency of 1.8 μW (corresponding to 0.3 μW/μCi) and 7% were observed at −20 °C, respectively.

1. Introduction

A wide range of technological areas are undergoing miniaturisation putting increasing demands on their associated power supplies. Microelectromechanical systems (Judy, 2001) are ever decreasing in size requiring similarly shrinking power supplies; reducing power supply weight is vital in lowering the cost and thus increasing the accessibility of space science platforms (Landis et al., 2012). Size is not the sole consideration; replacing spent batteries in implantable medical devices (Lueke and Moussa, 2011), for example, is costly and inconvenient for both patient and healthcare service. In defense applications, FPGA encryption key battery-backed memory needs to be continuously powered since encryption keys are stored in memory systems only as long as energy is supplied to them. Therefore, longevity and durability are also paramount. In some circumstances, chemical or solar batteries are not ideal for these purposes due to their requirement for periodic recharge or replacement, their lack of environmental tolerance, or the need for solar illumination. In long life and harsh environment applications which only require small amounts of power, nuclear microbatteries are becoming increasingly attractive; they also offer useful properties such as high-energy density, and insensitivity to environment and temperature. In nuclear microbatteries, the high-energy particles, released by radioactive atoms during nuclear decay, are absorbed by the microbattery conversion material generating electrical energy (Bower et al., 2002). Recently, a number of alpha, beta and X-ray microbatteries have been reported (Landis et al., 2012; Revankar and Adams, 2014; Butera et al., 2016a, 2016b). Beta emitters, in particular, have received research interest because of their ability to produce relatively high output powers with minimal lattice damage to the conversion device. The use of wide bandgap semiconductors for the conversion material is attractive because of the lower thermally generated leakage currents with respect to narrower badgap semiconductors; moreover, analysis has shown that the efficiency of conversion increases with increased bandgap (Bower et al., 2002). At room temperature, semiconductors such as GaAs, SiC, GaN and diamond have been successfully used as converter materials in betavoltaic cells by several researchers (Chen et al., 2011; Sciuto et al., 2011; Chandrashekhar et al., 2006; Fitting et al., 2006; Cheng et al., 2012; Bormashov et al., 2015). Experimental studies on the temperature effects on betavoltaic batteries have been also carried out, since nuclear microbatteries can be essential in outer space and deep-sea applications. These environments are under high- or low- temperature conditions and the temperature can vary over a wide range; hence, the electrical performance of a nuclear microbattery should be analysed under different temperatures. Liu et al. (2012) simulated and tested open circuit voltage ($V_{OC}$) and short circuit current ($I_{SC}$) of a betavoltaic cell based on $^{63}$Ni-Si as a function of temperature, they found that $V_{OC}$ became highly sensitive to temperature, and the temperature dependence of $V_{OC}$ can be expressed as an exponential function. Wang et al. (2010) studied the relationship between temperature and electrical performance of two $^{63}$Ni-Si betavoltaic cells; they proved that the electrical performance such as open circuit voltage ($V_{OC}$), maximum output power ($P_{m}$) and conversion efficiency clearly decreased with increase in temperature: in the temperature range 233.15–333.15 K the changing in values of $V_{OC}$ of the two cells were −3.1 and −3.0 mV/K, these changes resulted in decreased $P_{m}$ (8.94 nW at 233.15 K, whilst 2.18 nW at 333.15 K for the best beta cell) and conversion efficiency (1.09% at 233.15 K, whilst 0.265% at 333.15 K for the best beta cell), since $I_{SC}$ varied very little with temperature. Chandrashekhar et al. (2007) investigated the change in $V_{OC}$ of a $^{63}$Ni-4H SiC betavoltaic cell...
in response to temperature; a linear sensitivity of 2.7 mV/K was obtained from 24 to 86 °C. Wang et al. (2015) experimentally and theoretically studied the effect of temperature on the output performance of 63Ni-Si, 63Ni-GaAs, 147Pm-Si, and 147Pm-GaAs microbatteries: in the temperature range 213.15–333.15 K, while the short circuit current (Isc) increased slightly with temperature, the open circuit voltage (Voc) decreased evidently with the increase in temperature and the Voc sensitivities caused by temperature for 63Ni-Si, 63Ni-GaAs, 147Pm-Si, and 147Pm-GaAs microbatteries were −2.57, −5.30, −2.53, and −4.90 mV/K respectively; the conversion efficiency decreased also with the increase in temperature. Tang et al. (2015) demonstrated a radioluminescence nuclear battery that consisted of a 147Pm radioactive isotope, ZnS:Cu phosphor layer and GaAs photovoltaic cell; the effects of temperatures on the microbattery system were studied: experimental and theoretical results indicated that short circuit current density (Jsc) slightly decreased with the increase in temperature, whereas open circuit voltage (Voc) and the maximum output power (Pmax) rapidly decreased with temperature (dVoc/dT = −3.07 mV/K within the low temperature range of 223.15 K to 303.15 K).

GaAs is a III-V wide bandgap material that can give many advantages in a nuclear microbattery. It can be grown with very high crystal quality, its growth and processing techniques are relatively advantageous in a nuclear microbattery. It can be grown with very high temperature range of 223.15 K to 303.15 K. GaAs is a III-V wide bandgap material that can give many advantages in a nuclear microbattery. It can be grown with very high crystal quality, its growth and processing techniques are relatively advantageous in a nuclear microbattery. It can be grown with very high temperature range of 223.15 K to 303.15 K. GaAs is a III-V wide bandgap material that can give many advantages in a nuclear microbattery. It can be grown with very high crystal quality, its growth and processing techniques are relatively advantageous in a nuclear microbattery. It can be grown with very high temperature range of 223.15 K to 303.15 K. GaAs is a III-V wide bandgap material that can give many advantages in a nuclear microbattery. It can be grown with very high crystal quality, its growth and processing techniques are relatively advantageous in a nuclear microbattery. It can be grown with very high temperature range of 223.15 K to 303.15 K.

The wide availability, the relatively low cost, the reduced device damage risk due its endpoint energy of 66 keV, and the need for only comparatively little shielding to protect users make ⁶³Ni attractive option in a nuclear power supply. This paper reports initial characterisation of a GaAs ⁶³Ni radioisotope beta cell, using a custom GaAs photodiode originally intended for soft X-ray photon counting spectroscopy in space science. Beta particles, emitted by the ⁶³Ni radioisotope beta source, were directly converted into electrical energy using the GaAs device. The effect of temperature on the key parameters of the GaAs ⁶³Ni radioisotope beta-photodiode were studied over the temperature range 70 °C to −20 °C. This paper differs from previous studies on GaAs ⁶³Ni betavoltaic batteries (Wang et al., 2015) because of the different structure of the GaAs device used and the temperature range studied.

2. Materials and methods

2.1. Radioactive source and energy conversion device

A standard laboratory ⁶³Ni radioisotope beta source was used to illuminate a GaAs energy conversion device. The beta source was positioned as close as experimentally possible (3 mm) to the top of the GaAs device such to minimize the attenuations of the electrons in the dry nitrogen environment of the temperature test chamber which was used to achieve and maintain the temperatures investigated, and inside which the prototype cell was placed. A 400 µm diameter p⁺-n⁻ unpassivated GaAs mesa photodiode was used to collect the electrons emitted by the beta source. The GaAs epilayer of the device was grown to the Authors’ specifications by metalorganic vapour phase epitaxy (MOVPE) on a commercial GaAs n⁻ substrate at the EPSRC National Centre for III-V Technologies, Sheffield, UK. After growth, the wafer was processed to form mesa structures using wet etching techniques a 1:1:1 H₂PO₄:H₂O:H₂O solution was used followed by 10 s in 1:8:80 H₂SO₄:H₂O:H₂O solution. The device layers, their relative thicknesses and materials are summarised in Table 1. The p⁺-side Ohmic contact covered 33% of the surface of the photodiode studied. Fig. 1 shows schematically the geometry of the ⁶³Ni source and the GaAs detector.

### Table 1

<table>
<thead>
<tr>
<th>Layer</th>
<th>Material</th>
<th>Thickness (µm)</th>
<th>Dopant</th>
<th>Dopant type</th>
<th>Doping density (cm⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Ti</td>
<td>0.02</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Au</td>
<td>0.2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>GaAs</td>
<td>0.5</td>
<td>Be</td>
<td>p⁺</td>
<td>2 × 10¹⁸</td>
</tr>
<tr>
<td>4</td>
<td>GaAs</td>
<td>10</td>
<td></td>
<td>undoped</td>
<td>&lt; 10¹⁵</td>
</tr>
<tr>
<td>5</td>
<td>GaAs</td>
<td>1</td>
<td>Si</td>
<td>n⁺</td>
<td>2 × 10¹⁸</td>
</tr>
<tr>
<td>6</td>
<td>Substrate</td>
<td>n⁺</td>
<td></td>
<td>GaAs</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>Au</td>
<td>0.2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>InGe</td>
<td>0.02</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The absorption of the beta electrons in the GaAs ⁶³Ni radioisotope betavoltaic cell was studied using the Monte Carlo computer modelling package CASINO (version 3.3) (Hovington et al., 1997; Drouin et al., 1997): CASINO allowed the study of where the beta electrons are absorbed in the GaAs structure. At this stage, we were only interested in quantifying the percentage (QE) of the energy of the electrons that were incident on the face of the cell that was absorbed through the GaAs device of specified thickness. Simulations were run such to study the QE of the device at each beta electron energy (1–66 keV); each simulation had 4000 beta particles. The source-device geometry was not studied in these particular Monte Carlo simulations. 66 simulations at different beta electron energies were run and the QE studied. In each case, (i.e. to determine the cell QE for energies from 1 keV to 66 keV in 1 keV steps), the beta particles were simulated as incident on the p⁺-side of the GaAs epilayer; thus beta particle absorption through the device thickness studied. The QE of the GaAs structure as a function of beta particle energy is shown in Fig. 2.

The simulations also showed that for the particular experimental set...
up used, beta particles with energies below 25 keV did not reach the GaAs i-layer primarily because of the attenuation of the particles’ energies in the protective inactive Ni over-layer (1 µm thick) of the radioisotope beta particle source used. The source had this inactive overlayer for laboratory safety reasons; an eventual real world microbattery would use a $^{63}$Ni radioisotope beta source without such a layer. Attenuation through the top contact (covering 33% of the diode’s face) and p+ layer of the GaAs device was a secondary effect. Beta electrons with energy ≥ 25 keV deposited part of their energy in the i-layer (e.g. 53% of the energy of the 66 keV electrons was absorbed in the i-layer). The attenuation of the beta particles in the dry nitrogen gap (3 mm) was also investigated with CASINO and found to be negligible compared to the other losses. The emission spectrum of the $^{63}$Ni (including beta particle self-absorption in the $^{63}$Ni itself) was not taken into account in the simulations; while this was not considered for the calculations of device $Q_E$, it was essential in the estimation of e.g. the theoretical power, as shown below (Eq. (2)).

In the GaAs $^{63}$Ni radioisotope betavoltaic cell, electron-hole pairs were generated in the semiconductor by the beta electrons emitted from the $^{63}$Ni radioisotope. The average energy consumed in the generation of an electron-hole pair is called the electron-hole creation energy (4.18 eV for GaAs (Bertuccio and Maiocchi, 2002)). Because of the combination of built-in and applied electric field, the electrons and holes created in the depletion region, and in the n and p regions near the depletion boundary (i.e. closer than the appropriate carrier’s diffusion length), accelerate in opposite directions and are swept to the p+ -type and n+ -type regions, respectively, generating a photocurrent.

### 2.2. Experiment and measurements

The GaAs $^{63}$Ni radioisotope betavoltaic cell was characterised using a Keithley 6487 picoammeter/voltage source. Forward bias measurements from 0 V to 0.5 V were made in 0.005 V increments in dark conditions and under the illumination of the $^{63}$Ni radioisotope beta source. The uncertainty associated with the current readings was 0.3% of their values plus 400 fA, while the uncertainty associated with the applied biases was 0.1% of their values plus 1 mV (Keithley Instruments, 2013). The GaAs $^{63}$Ni radioisotope betavoltaic cell was characterised in the temperature range 100 °C to −20 °C. To control the temperature and humidity, the beta cell was placed inside a TAS Micro MT climatic cabinet in a dry nitrogen atmosphere (relative humidity < 5%).

Fig. 3 shows typical dark current characteristics as a function of forward bias at different temperatures for the 400 µm diameter GaAs photodiode. At high temperatures, the dark currents through the devices increased due to the greater thermal energy available.

At each temperature, a higher dark current was observed at increased applied biases because of the greater applied electric fields across the photodiode’s depletion region. In a simple p-n diode, the dark current increases exponentially as a function of applied bias according Eq. (1),

$$I = I_0 \exp\{qV/nkT\}$$

where $n$ is the ideality factor, $k$ is the Boltzmann constant and $T$ is the temperature (Sze and Ng, 2007). A linear least squares fit of the natural logarithm of the dark current data as a function of applied bias was used to calculate the saturation current ($I_0$) and the ideality factor ($n$) of the GaAs device at each temperature: Eq. (1) was linearised as $\ln I = A + BV$, with $A = \ln I_0$ and $B = q/(nkT)$, and used linear least square fitting. Fig. 4 and Fig. 5 show the logarithm of the measured saturation current and the ideality factor as functions of temperature, respectively.

The magnitude of the extrapolated natural logarithm of the saturation current decreased with increasing temperatures was in accordance with Butera et al. (2016a), where a similar GaAs structure was used. On the logarithmic scale in Fig. 4, the observed increase was $12.12 \pm 0.02$ between 100 °C and −20 °C (corresponding to an increase in saturation current, $I_{0s}$, of 0.28 pA). This was in remarkable agreement with the expected increase 11.48 (corresponding to an increase in saturation current, $I_{0s}$, of 0.43 pA), computed using the simple assumption that the temperature dependence of the saturation current was proportional to $\exp\{-E_i/(2kT)\}$ (Butera et al., 2016a). At every temperature, an ideality factor close to 2 was observed; this indicated that generation-recombination current was dominant over the diffusion current in the device in the temperature range studied. The small increase in ideality factor as
the temperature decreased (from 1.723 ± 0.004 to 1.820 ± 0.004 between 100 °C and −20 °C) may be attributed to the decreased contribution of the diffusion current when temperature decreased (Sze and Ng, 2007). A similar behaviour has been previously observed for other GaAs structures (Lioliou et al., 2016).

Under the illumination of the 63Ni radioisotope beta source, the current of the GaAs betavoltaic device was measured as a function of applied bias at different temperatures. From these current characteristics, the experimental values of the open circuit voltage (V_{OC}) were obtained as the interception point of the curves on the horizontal axis. Open circuit voltages higher than 0.005 V were observed at temperatures lower than 70 °C. At temperatures above 70 °C the open circuit voltages were found to be lower than the forward bias step size of 0.005 V, and therefore could not be discriminated; such open circuit voltages have been rounded to zero. Fig. 6 shows the illuminated current characteristics as a function of applied bias between 70 °C and −20 °C. At increased temperature, the softness in the knee of the illuminated current as a function of applied forward bias decreased. At −20 °C, a different behaviour, with respect the trend shown by the other temperatures, was observed: the curve at −20 °C almost overlapped the curve at 0 °C indicating that saturation effects from beta particle induced conduction became dominant over the thermal mechanism at increased forward voltages; the beta electrons, loosing energy through the samples, generated electron-hole pairs along their trajectories that decreased the material resistivity. The lower resistivity region decreased at increased forward voltages, the percentage of the lower resistivity region in the depletion region increased and the beta induced conductive mechanism was more evident.

Fig. 7 shows the open circuit current as a function of temperature for the GaAs 63Ni radioisotope betavoltaic cell between −70 °C and 20 °C.

The open circuit voltage (V_{OC}) decreased with increased temperature, reaching a saturation value of 0.2 V at −20 °C. Between 70 °C and 0 °C V_{OC} values in accordance with Butera et al. (2016a) (where similar GaAs structure was used) were observed. The dependence of V_{OC} from temperature agreed with other experimental studies reported in literature (Liu et al., 2012; Tang et al., 2015). This behaviour may be explained considering the dependence of the open circuit voltage from the saturation current (Sze and Ng, 2007). Also similarly to Butera et al. (2016a), a linear relationship between the open circuit voltage and temperature was observed at temperatures between 40 °C and 0 °C. In this temperature range, a linear least squares fit showed that V_{OC} was dependent on temperature (T) according the relation V_{OC} = aT + b with A = (0.0032 ± 0.0001) V °C^{-1} and B = (0.192 ± 0.003) V. At −20 °C, in contrast with Butera et al. (2016a), the open circuit voltage appeared to saturate to a value ≈ 0.2 V; this could be due to the higher carrier density in the semiconductor when it was illuminated with beta electrons compared with X-ray photons previously reported (Butera et al., 2016a). The beta electrons, loosing energy through the samples, generated an increased amount of electron-hole pairs along their trajectories that decreased the material resistivity. The lower resistivity resulted in a lower open circuit voltage compared to Butera et al. (2016a). This mechanism and its effect were evident at low temperature where was dominant over the thermal mechanism. The thermal mechanism consists of the creation of electron-hole pairs due to the thermal energy available. The decrease of the open circuit voltage at increased temperatures for a 63Ni-GaAs betavoltaic battery has been also observed by Wang et al. (2015); V_{OC} temperature dependences of 3.2 mV/°C and 5.30 mV/K were reported here and by Wang (2015), respectively. In Wang et al. (2015), saturation effects are not evident at low temperatures: the open circuit voltage reached a value of ≈ 0.23 V at −20 °C (similar at the value found here at the same temperature) and continued to increase with decreasing the temperatures.

The experimental values of the short circuit current (I_{SC}) were obtained, at each temperature, as the interception point of the curves in Fig. 6 on vertical axis. Fig. 8 shows the I_{SC} as a function of temperature for the GaAs 63Ni radioisotope betavoltaic cell.

The short circuit current magnitude increased with decreasing temperature, between 70 °C and −20 °C the observed increase was 32.6 pA. This behaviour as a function of temperature has been also observed by other researchers (Butera et al., 2016a; Krawczyk et al., 1981). The dependence of the short circuit current with temperature was instead different from the results reported by Wang et al. (2015) for another 63Ni-GaAs microbattery. In contrast to what is reported here, Wang et al. (2015) found a slightly decrease in the short circuit current...
Fig. 9. GaAs $^{63}$Ni radioisotope betavoltaic cell output power as a function of applied forward bias at 70 °C (filled circles), 60 °C (empty circles), 50 °C (filled squares), 40 °C (empty squares), 30 °C (crosses), 20 °C (filled rhombuses), 0 °C (empty rhombuses) and −20 °C (filled triangles).

Fig. 10. Experimental maximum power as a function of temperature for the GaAs $^{63}$Ni radioisotope betavoltaic cell.
where $\eta$ is the internal conversion efficiency, $P_m$ the maximum experimental output power measured at a particular temperature, and $P_P$ the theoretical maximum power obtainable from such system. Slightly higher internal conversion efficiency values would be obtained if we had considered in our calculation a less pure $^{63}$Ni radioactive source: in such circumstances the $^{63}$Ni specific activity would decrease, thus obtaining a lower $P_P$ value and consequently a higher internal conversion efficiency. Considering a specific activity of 11 mCi/mg, an internal conversion efficiency of 9% would have been obtained at $-20^\circ$C. It has also to be noted that the overall system efficiency will be much lower than the calculated internal conversion efficiency.

3. Conclusion

In this paper, a prototype GaAs $^{63}$Ni radioisotope betavoltaic cell was reported: a $^{63}$Ni beta radioisotope was used to illuminate a GaAs converter device such to convert the nuclear energy of the $^{63}$Ni beta particle emissions to electrical energy. Using a $^{63}$Ni beta radioisotope (activity 185 MBq) ensures a long life microbattery with reduced damage risk for the converter device. GaAs growth and processing techniques are relatively cheap and more routinely available than some other nuclear materials. GaAs growth and processing techniques are relatively cheap and more routinely available than some other nuclear materials.

**Data Statement**

Data underlying this work are subject to commercially confidential-