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Gamma ray driven photovoltaic cells: an interface between nuclear and semiconductor physics

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Abstract

We explore the possibility of utilizing low energy γ-photon radiation from single or multiple radioactive sources, for the production of electron–hole pairs in the semiconductors of photovoltaic cells. Our aim is to produce and store electric energy using this mechanism. The radioactive sources in the GRPVC (gamma ray photovoltaic cell) can be either naturally occurring or manufactured nuclear isomers. The γ-photon intensity can be controlled, hence the output and efficiency can also be controlled. A spherical GRPVC of 40 cm diameter, with the nuclear isomer Th*-229 ($E_\gamma = 3.5$ eV, $T_{1/2} = 45$ h) can produce a maximum open circuit voltage $V_{\mathrm{ocmax}} = 2.626$ V, at maximum efficiency $\eta_{\max} = 52.2\%$ initially, which occurs when the semiconductor energy gap, E_g, equals the γ-photon energy, E_γ. This reduces to the lower value $V_{\mathrm{oc}} = 0.5$ V at $\eta = 12\%$ after 188 days. The ultimate efficiency for the GRPVC studied here is about 70%, which occurs for $E_g = E_\gamma \sim 10$ eV, and is above 40% for $E_g = E_\gamma$ (with $E_\gamma \geqslant 3$ eV). It is also shown that nuclear isomers with long half-life can keep the GRPVC operating at high V_{oc} and high efficiency levels for many decades, and even for centuries.

1. Introduction

The AM0 solar spectrum corresponds to an intensity of 1353 W m^{-2} (joules of energy per square metre and per second) which arrives at the free space above the Earth's atmosphere from the sun. When it reaches the surface of the Earth, this spectrum is reduced to AM1 or AM2 due to absorption by the Earth's atmosphere and different angle of incidence, which correspond to the lesser intensity of 925 W m^{-2} and 691 W m^{-2} respectively. This energy is in the form of photons with energies that span the whole range of the electromagnetic spectrum. The prospect of generating electricity from the energy in the solar spectrum has been studied and applied in various ways for several decades. Parabolic mirror thermal devices, semiconductor or organic material based photovoltaic solar cells, are examples which provide the mechanism to convert the energy in the solar photons into electricity. In the case of solar cells, the photons create electron–hole (e–h) pairs in the semiconductors of the cell, and the photo-generated electrons are then collected by special contacts with the semiconductor materials. The efficiency at which the photons do this depends mainly on their energy (frequency), compared to the energy gap of the semiconductors of the cell. A considerable amount of experimental and theoretical work has been expended over the last few decades, in order to improve the conversion efficiency of PVCs (photovoltaic cells) [1–19]. There are other competing mechanisms however, such as Auger and radiative recombination and impact ionization, which take place in the semiconductor materials at varied probabilities. The first two of these affect the efficiency of the PVC negatively, while the latter affects it favourably. Intense theoretical effort has been devoted for a better understanding and utilization of these phenomena, applying a variety of models [9–19].

Thus the operation of a photovoltaic cell is based on an averaging over all photon energies in the solar spectrum and all competing mechanisms taking place in the PVC. This requires the solar panels to be designed so that to minimize losses, for example due to reflection, and the semiconductor parameters to have specific values to balance the e–h generation and recombination in order to optimize the conversion efficiency. Another limiting factor to the efficiency of PVCs is the structure of the solar spectrum itself. Even in theories which take into account the quantum effects of

impact ionization and Auger recombination the efficiency of the photovoltaic solar cell can only be of the order of about 40% [9–17, 26]. Some theoretical work based on combined technologies have reported efficiencies of up to 50% [18]. There are a large number of theoretical efficiency calculations each of which are based on different models and assumptions, and some put the ultimate efficiency to as high as 86%–88% for maximal solar concentration [31, 33]. The experimental efficiency of single junction photovoltaic solar cells however has only reached as high as about 30% [1–4]. More recently, it has been reported that monolithic triple junction solar cells have achieved efficiency up to about 40.6% [35]. Despite the negative contribution made by the electron–hole recombination processes, the efficiency could be enhanced if the spectrum were a narrow one, involving photon energies equal to, or above the semiconductor energy gap. This way all photons in the spectrum would contribute with about the same efficiency in creating e–h pairs. This ideal spectrum can be materialized, independently of the solar spectrum, if we utilize the γ-photons emitted from naturally occurring γ-ray sources for the purpose of driving the operation of photovoltaic cells. It is also possible to utilize laboratory made nuclear isomers [20–24], which may decay by emission of one or more γ-photons. Thus the spectra considered here are either of single energy, i.e. monochromatic light, or very narrow ones as opposed to the infinitely wide solar spectrum. A combined solar–nuclear design can also be materialized. The intensity of the γ-photons can be set to desired levels, by appropriate choice of the GRPVC size and/or the mass of the γ-ray nuclear isomer used. This study involves appropriate formalism for the cell efficiency that holds for discrete spectra. This is obtained by writing the equations for the energy flux density, and the light generated current densities [9, 13–15, 26] to apply to such spectra. The formalism obtained facilitates the study of the cell's response to γ-photons from single sources, i.e. to monochromatic light.

The energy of the γ-photons produced by nuclear isomers can vary from a few eV to several MeV, and even GeV, but in this work our interest lies in the low energy γ-photons. There are several methods for nuclear isomer production usually called: shape or volume excitation; spin traps; and K-isomers. Each of these methods produces isomers that emit γ-photons of various energy and decay rate. Small isomeric nuclei are preferred for this application because the amount of mass in one mole of the material is relatively small; hence making the device reasonably light. Research in isomer nuclear physics has been active for several decades, technology is now available to create them, and isomers emitting γ-photons of various energies can be produced in the laboratory to suit various applications. One interesting application is the construction of very powerful γ-ray laser devices [25]. In these the nucleus is desired to possess two or more excitation energy levels, at least one of which needs to be a metastable state to facilitate population inversion and the lasing transition. Nuclear isomers can be produced through bombardment of a stable nucleus ${}_{Z}\mathrm{X}^{\mathrm{M}}$, with x-rays, synchrotron radiation energy or other particles, depending on the γ-photon emission we wish to achieve. Under irradiation the transition ${}_{Z}\mathrm{X}^{\mathrm{M}} \rightarrow {}_{Z}\mathrm{X}^{\mathrm{M}^*}$ takes place, and subsequently the reverse one ${}_{Z}\mathrm{X}^{\mathrm{M}^*} \rightarrow {}_{Z}\mathrm{X}^{\mathrm{M}} + \gamma$ follows. It is the latter that is of interest in this application.

It is found that this interface between semiconductor and nuclear physics, with the use of isomers of Th-229, produces high V_{oc} and efficiency in GRPVCs. The ultimate efficiency of the system can be attained, provided the semiconductor material energy gap is $E_g = E_\gamma \approx 10$ eV. Manufacturing semiconductors with such large energy gap however, may prove challenging. The production of isomeric states, other than those of Th-229, with such low energy γ-photon emission will also prove challenging to achieve. Possible ways to remedy these difficulties are discussed in the theory and results sections.

2. Theory

When an unstable nucleus decays by emission of either α or $\beta^{\pm}$ particle, the daughter nucleus is usually left in an excited state due to a high energy configuration of its nucleons. In these cases the nucleus will lose the excess energy by a single or multiple γ-photon emission and make a transition to its ground state. Similar γ-photon emission processes take place in laboratory manufactured isomers. In these, protons or neutrons are excited to a higher energy level and then they make a transition to the ground state by emission of γ-photons. In this case the decay does not involve α or $\beta^{\pm}$ particle emission, because the numbers of protons and neutrons in the nucleus are unchanged. Furthermore, given that the γ-photons are desired to be of low energy, the semiconductor materials damage they may cause can be minimal or none [42].

Figure 1 shows three different processes which can lead to an isomeric nuclear state. In figure 1(a) the Co-60 nucleus decays by emission of an electron and an electron–antineutrino, and turns into the isomer Ni*-60. In turn Ni*-60 decays by a cascade emission of two γ-photons [24] both of which are of relatively high energy compared to the energy gap of any semiconductor material available. This is a naturally occurring state of Ni-60, and the indices m1 and m2 distinguish its two isomeric states. With this amount of energy the emitted γ-photons are capable in creating electron–positron pairs. Such high energy γ-photon sources could be used for the proposed GRPVCs, but in this case the γ-photons will need to lose much of their energy via Compton scattering, or some other mechanism, as is discussed below. In figure 1(b) the Th-229 is irradiated by synchrotron energy and is excited to the isomeric state Th*-229 (3.5 eV, $T_{1/2} = 45$ h). The Th*-229 isomer then decays by emission of a soft γ-photon whose energy is only 3.5 eV, which is comparable to the energy gap of semiconductor materials available [27a, 28c, 30]. This is a manufactured isomer of Th-229 nucleus with a half-life of 45 h (1.62×10^5 s) and corresponds to a value of the decay constant $\lambda = 4.28 \times 10^{-6}$ s^{-1}. It is also possible to generate and utilize the newly discovered isomer of Th*-229 (7.6 eV, $T_{1/2} = 5$ h) which also emits low energy γ-photons, of about 7.6 eV, with half-life of about 5 h [34]. A desired isomerization and decay mode is shown in figure 1(c), when $\gamma(E_1)$ and $\gamma(E_2)$ are both low energy γ-photons matching existing semiconductor energy gaps. The use of an isomer of

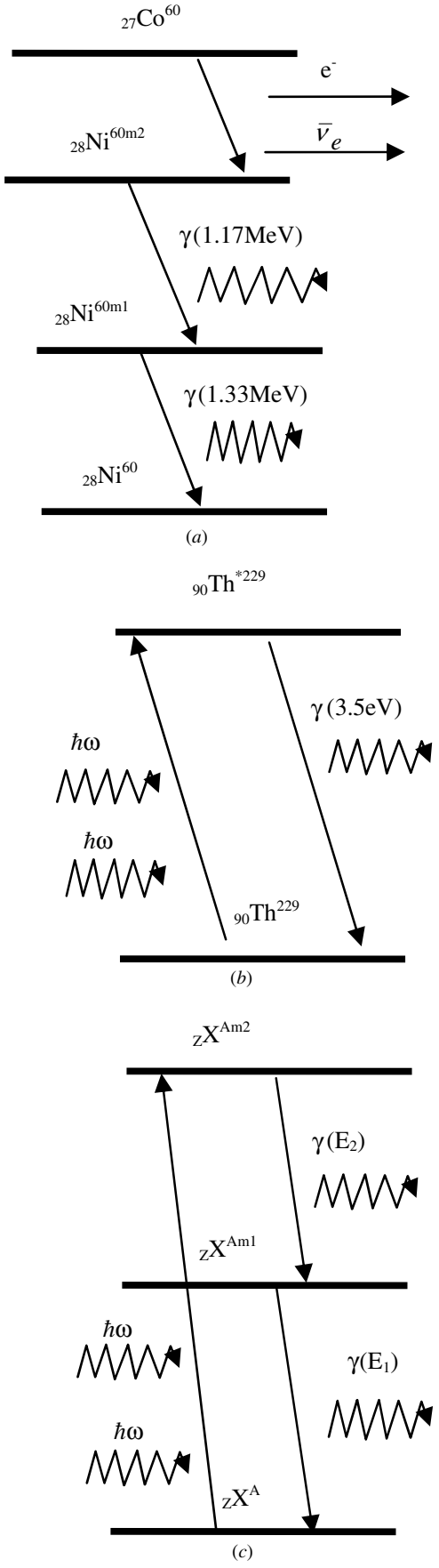

Figure 1. (*a*) Ni-60* emitting two γ-photons, (*b*) decay of isomer of Th-229, (*c*) a preferred decay mode of some isomer X.

this nature may be materialized with the two isomeric states of Th-229. At equilibrium the lower lying isomers decay at a rate equal to that of the parent nucleus.

There are four known processes that can occur when γ-photons interact with matter: (i) Compton scattering, which can occur via collisions of the γ-photon with free electrons in the material; (ii) electron–positron pair creation—this involves γ-photons with energy at least equal to 1.022 MeV; (iii) photoelectron generation and (iv) electron–hole pair creation if the material is a semiconductor. These processes occur with varied probabilities but it is process (iv) that is of interest here. If γ-photons with energy of the order of several hundreds of eV are used, then it is necessary to have these photons suffer multiple Compton scattering in some other material in which the Compton scattering is the dominant interaction process, in order to produce the low energy photons (LEPs) needed for the production of e–h pairs in the semiconductor material of the cell. This is necessary because the probability of the process of multiple e–h pair creation by high energy γ-photons is very low. Thus their effect on the efficiency of the cell is negligible, as we shall see in the results section.

2.1. Possible designs of GRPVCs

It is important that the γ-photon source is placed in the GRPVC in such a way so that to minimize losses. A possible arrangement is shown schematically in the diagrams of figures 2(*a*) and (*b*). Configuration (2a) involves a reflector and a lens for the generation of a parallel beam of γ-photons. In order to minimize γ-photon losses in configuration (2a), one could place reflectors in the form of cylindrical rings, with variable gradient, to ensure that most photons are reflected to the PVC rather than be absorbed outside the GRPVC. Note that in configuration (2b) the cell can absorb up to 100% of the γ-photons emitted by the source, which is placed at the centre of the spherical cell. One advantage of configuration (2b) is that the reflected γ-photons from one part of the cell can be absorbed by another. In addition, the outer surface of the GRPVC can be laid with normal solar cells to operate with sunlight, hence forming a dual PVC.

2.2. Formalism

We assume that at some time t there are n radioactive sources decaying by emission of γ-photons, and let $\{\lambda_1, \lambda_2, \ldots, \lambda_n\}$ and $\{E_1, E_2, \ldots, E_n\}$ be their decay constants and γ-photon energies respectively. The activities of the sources are $\{A_1(t), A_2(t), \ldots, A_n(t)\}$. For a single source we can write

$$A_k(t) = \lambda_k N_k(t), \qquad \text{where} \quad N_k(t) = N_{0k}\, e^{-\lambda_k t}. \tag{1}$$

Thus the total activity is

$$A(t) = \sum_k \lambda_k N_{0k}\, e^{-\lambda_k t}. \tag{2}$$

Also let S be the surface area of the cell, which for the configuration of figure 2(*b*) is $S = 4\pi r^2$. The formalism in this work is developed so that to apply for configuration (2b) and only a minor modification is needed for that of (2a). Since radioactive decay is not thermal emission, there is no need to

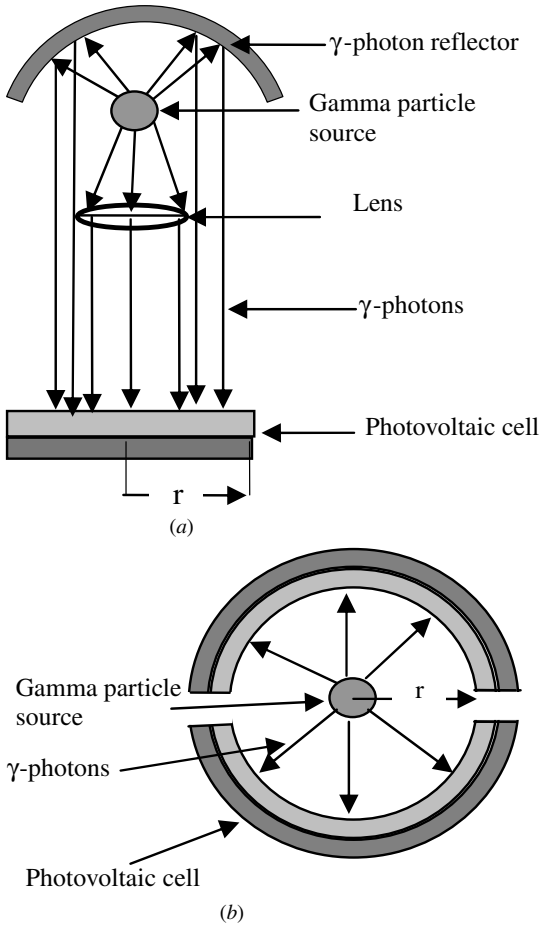

Figure 2. Possible configurations for the GRPVCs design: (*a*) linear, (*b*) spherical.

take into account the Bose–Einstein distribution of the emitted photons. Our analysis will be time dependent in order to facilitate the study of how the efficiency of the cell varies with time. Thus the total γ-photon intensity at time t can be written as

$$I(t) = \frac{1}{4\pi r^2} \sum_k \lambda_k N_{0k}\, \mathrm{e}^{-\lambda_k t}. \tag{3}$$

This can also be written in terms of the amount of mass, M_{0k}, of each source at time $t = 0$ and the atomic mass, M_{Ak}, of the isomers used as follows:

$$I(t) = \frac{N_A}{4\pi r^2} \sum_k \lambda_k \frac{M_{0k}}{M_{A_k}}\, \mathrm{e}^{-\lambda_k t}. \tag{4}$$

If all photons could reach the surface of the cell, then the total energy flux density (W m^{-2}) incident on the cell would be

$$D(t) = e k_B T_p \frac{N_A}{4\pi r^2} \sum_k \lambda_k x_k \frac{M_{0k}}{M_{Ak}}\, \mathrm{e}^{-\lambda_k t} \tag{5}$$

with $x_k = E_k / k_B T_p$.

E_k is the energy, in eV, of the γ-photon from the k source, T_p the temperature of the source, k_B is Boltzmann's constant ($k_B = 1.38 \times 10^{-23}$ J K^{-1}) and N_A is the Avogadro number ($N_A = 6.03 \times 10^{23}$ mol^{-1}). The temperature T_p, of the source, can be the same as that of the cell T_c.

For the sake of generality we shall develop the formalism to hold for heterojunction as well as homojunction cells. The equations for the light generated currents must be written so that to take into account the possibility of the γ-photons encountering the large or the small energy gap first. We assume that the source irradiates the semiconductor labelled 1 first. Let K_1 and K_2 be the absorption probabilities of a photon, $P_{e\text{–}h}(1)$ and $P_{e\text{–}h}(2)$ the probability for a single electron–hole pair creation, E_{g1} and E_{g2} the energy gaps, in material 1 and 2 respectively. If E_k is the energy of the γ-photon from the k nuclear isomer, we define the parameter $n_k(i) = \mathrm{Int}(E_k/E_{gi})$ (the integer part of the ratio E_k/E_{gi}). Then, a high energy photon, with energy $E_k > E_{gi}$, will be able to produce $n_k(i)$ e–h pairs with probability $\sim P_{e\text{–}h}^{n_k(i)}(i)$ where $i = 1, 2$. Thus we can write:

(a) $E_{g1} > E_{g2}$:

$$J_{l1}(t) = \frac{eN_A K_1}{4\pi r^2} \sum_{x_k \geqslant x_{g1}} n_k(1) P_{e\text{–}h}^{n_k(1)}(1) R_k\, \mathrm{e}^{-\lambda_k t}$$

$$J_{l2}(t) = \frac{eN_A K_2}{4\pi r^2} \sum_{x_{g2} \leqslant x_k \leqslant x_{g1}} n_k(2) P_{e\text{–}h}^{n_k(2)}(2) R_k\, \mathrm{e}^{-\lambda_k t} + \frac{eN_A K_2 (1 - K_1)}{4\pi r^2} \times \sum_{x_k \geqslant x_{g1}} n_k(2) P_{e\text{–}h}^{n_k(2)}(2) R_k\, \mathrm{e}^{-\lambda_k t} \tag{6a}$$

(b) $E_{g2} > E_{g1}$:

$$J_{l1}(t) = \frac{eN_A K_1}{4\pi r^2} \sum_{x_k \geqslant x_{g1}} n_k(1) P_{e\text{–}h}^{n_k(1)}(1) R_k\, \mathrm{e}^{-\lambda_k t}$$

$$J_{l2}(t) = \frac{eN_A K_2 (1 - K_1)}{4\pi r^2} \times \sum_{x_k \geqslant x_{g2}} n_k(2) P_{e\text{–}h}^{n_k(2)}(2) R_k\, \mathrm{e}^{-\lambda_k t} \tag{6b}$$

where $R_k = \lambda_k M_{0k}/M_{Ak}$ is the initial molar activity (mol s^{-1}) of the k source. In most applications the first term in the second of equations (6*a*) will be absent since $x_k \geqslant \max(x_{g1}, x_{g2})$. These equations are special cases of equations (2) and (3) in [26] for the continuous solar spectrum. A couple of differences between equations (6) and those in [26] that must be noted are that: (i) equations (6) include explicitly the probability for multiple e–h pair creation; (ii) equations (6) do not contain the impact ionization and Auger recombination terms since we are not concerned with these effects in this work. Thus the total light generated current in the semiconductor materials is $J_l = J_{l1} + J_{l2}$. The heterojunction formalism is in particular useful when a single or two nuclear isomers emitting photons of different energy are used. Such system could be facilitated by the isomers of Th-229 for example. When many different γ-ray isomers are used, a tandem structure for the GRPVC would be more appropriate to use.

For high energy γ-photons, $P_{e\text{–}h}^n$ can be a very small number so that their contribution to light-generated currents can be very small, hence making a very small contribution to the efficiency of the cell. For γ-photons with energy only a few times larger than the energy gap, multiple electron–hole creation can make some small contribution. Thus it

Table 1. Notation of the parameters.

D_e, D_h	Diffusion coefficient of minority carriers
N_{c1}, N_{c2}, N_{v1}, N_{v2},	Effective densities of states
N_D, N_A	Concentration of ionized dopants
τ_e, τ_h	Lifetimes of minority carriers
A_e, A_h	Band-and radiative coefficients
W_n, W_p	Layer widths
T_c, T_p	Cell and radioactive source temperature
x_g, y_g	E_γ/k_BT_p, E_g/k_BT_c

is a matter of choosing between a single e–h creation with high probability, and multiple e–h creation but with much lesser probability. Calculations of the efficiency, η, and open circuit voltage, V_{oc}, for high energy photons and for photons with energy just equal to the energy gap are performed, and comparison is made in the results section.

The dark (or saturation) current is given by standard p–n junction theory as [9, 18]

$$J_s = \alpha_1 \exp(-y_{g1}) + \alpha_2 \exp(-y_{g2}) \tag{7}$$

where

$$\begin{aligned} \alpha_1 &= e\frac{D_e^{1/2}N_{c1}N_{v1}}{\tau_e^{1/2}N_A}\tanh\left(\frac{W_p}{\sqrt{D_e\tau_e}}\right) \\ \alpha_2 &= e\frac{D_h^{1/2}N_{c2}N_{v2}}{\tau_h^{1/2}N_D}\tanh\left(\frac{W_n}{\sqrt{D_h\tau_h}}\right). \end{aligned} \tag{8}$$

The parameters and symbols used in these equations are defined in table 1.

Note that τ_e and τ_h are given in terms of the band and radiative coefficient A_e and A_h through the following equations [4, 26, 30]

$$\tau_e = \frac{1}{A_eN_A}, \qquad \tau_h = \frac{1}{A_hN_D}. \tag{9}$$

Equations (6), (7) and (9) can be generalized to incorporate the effects of impact ioniation and Auger recombination in order to extend this work in a similar way to that in [9, 26]. This will be studied elsewhere.

The J–V equation is used for the calculation of the open circuit voltage V_{oc}, and it is given as [28a]

$$J(V) = J_s[\exp(eV/kT_c) - 1] - J_l. \tag{10}$$

Equation (10) gives the open circuit voltage V_{oc} by letting $J(V) = 0$ as [15, 26, 28b]

$$V_{oc} = \frac{kT_c}{e}\ln\left(1 + \frac{J_l}{J_s}\right). \tag{11}$$

With all the relevant formalism needed for the calculation of the efficiency now in place, we can write the GRPVC cell efficiency as follows:

$$\eta = \eta_1\eta_2\eta_3. \tag{12}$$

η_1, η_2, and η_3 are defined by the equations below [15, 26] where

$$\eta_1 = \frac{x_{g1}J_{l1} + x_{g2}J_{l2}}{D(t)} \tag{13a}$$

is the spectrum factor,

$$\eta_2 = \frac{T_c}{T_p}\frac{J_l}{x_{g1}J_{l1} + x_{g2}J_{l2}}\ln\left(1 + \frac{J_l}{J_s}\right), \tag{13b}$$

Table 2. Values of semiconductor parameters used.

$W_p = W_n = 10^3\ \mu$m	$N_D = N_A = 8 \times 10^{14}$ cm^{-3}
$K_1 = 0.8$, $K_2 = 0.6$,	$D_e = D_h = 300.0$ cm^2 s^{-1}
$P_{e\text{–}h} = 0.8$	$A_e = A_e = 1.12 \times 10^{-11}$ cm^3 s^{-1}
$E_g = 1.47$ eV	

is the voltage factor, and

$$\eta_3 = \frac{v^2\exp(v)}{[(1+v)\exp(v) - 1][v + \ln(1+v)]} \tag{13c}$$

is the fill factor.

Here v satisfies the equation

$$(1+v)\exp(v) - 1 = \frac{J_l}{J_s} \tag{14}$$

in order to maximize the output power [14–16, 26, 28b].

3. Results

For an initial calculation let us consider one mole of the nuclear isomer Th*-229 (3.5 eV, $T_{1/2} = 45$ h) and let the cell energy gap be variable. For this isomer of Th-229 we have found the decay constant, $\lambda = 4.3 \times 10^{-6}$ s^{-1}, and we also assume that the source and the cell are at the same temperature $T_p = T_c = 300$ K. If the surface area of the cell, in configuration (2a) say, is 25 cm^2 (2.5×10^{-3} m^2) the intensity of the γ-photons is about 10^{21} photons m^{-2} s^{-1}. This corresponds to γ-ray intensity $D = 560$ W m^{-2} which is only 41.4% of the light intensity in the AM0 solar spectrum (1353 W m^{-2}), but it is comparable to the spectra AM1 (925 W m^{-2}) and AM2 (691 W m^{-2}). If we take 2.29 kg of Th*-229 however, then the γ-ray intensity will be 5600 W m^{-2}, which is about four times larger than the AM0 spectrum and six times the AM1! For the present calculation we shall only be concerned with a homojunction GRPVC. We shall make use of the semiconductor parameters shown in table 2.

These values of parameters have been taken from table 1; row 5 in table 2; and row 5 in the ninth column of table 4 in [26]. A calculation of the efficiency for a spherical type cell of diameter $d = 40$ cm, when 1 mole of isomer Th*-229 is used at time $t = 0.0 \times T_{1/2}$ gives a disappointing efficiency $\eta = 16.96\%$ and open circuit voltage $V_{oc} = 608$ mV, which are smaller than $\eta = 22.58\%$ and $V_{oc} = 1.04$ V found in [26]. If however the energy gap is increased to about the value of the γ-photon energy, $E_g = 3.5$ eV, then the efficiency improves to $\eta = 52.12\%$ and the open circuit voltage to $V_{oc} = 2.626$ V, both of which are much larger than those in [26]. The response of the GRPVC is much more effective when the energy gap is close or equal to that of the γ-photon. The efficiency η and V_{oc} are shown in table 3 for several E_g values.

The results shown in table 3 can improve further, by reducing the size of the cell, by increasing the number of moles of the radioactive source Th*-229, by 'adjusting' the semiconductor parameters or by doing all these. The use of semiconductor materials with large E_g, in order to match the energy of the γ-photon, serves favourably in that the large

Table 3. η and V_{oc} for $d = 0.40$ m at $t = 0.0 \times T_{1/2}$.

Energy gap, E_g (eV)	Efficiency, η (%)	V_{oc} (V)
1.47	16.96	0.608
2.0	21.13	1.129
2.5	31.43	1.626
3.0	41.79	2.126
3.5	52.19	2.626

Table 4. η and V_{oc} for various multiplicities at $t = T_{1/2}$.

γ-energy (eV)	Multiplicity	η (%)	V_{oc} (V)
3.5	1	51.8	2.608
7.0	2	41.7	2.620
14.0	4	26.8	2.626
21.0	6	17.1	2.625
35.0	10	7.0	2.616
70.0	20	0.74	2.576
140.0	40	8.16×10^{-5}	2.478

energy gap suppresses Auger recombination, as can be seen from the recombination rate equation [27b, 29]

$$W_{\text{Auger}} = \frac{e^4 m_c^*(\mu + m_c^*/m)}{4\pi^{5/2}\varepsilon^2\hbar^3(1+\mu)^{1/2}} \left(\frac{k_B T_c}{E_g}\right)^{3/2} \times \exp\left\{-\frac{(1+\mu)E_g}{k_B T_c}\right\}. \quad (15)$$

Here, m_c^* and m_v^* are the effective mass for an electron in the conduction band and a hole in the valence band, and $\mu = m_c^*/m_v^*$. Thus a large energy gap can reduce the recombination rate by several orders of magnitude.

When nuclear isomers emitting γ-photon with energy several times the E_g are used, then multiple e–h pair creation can take place. But η and V_{oc} will actually decrease significantly for values of the emitted photon many times larger than E_g. Table 4 shows values of η and V_{oc} for various multiplicities, for a value of $P_{e-h} = 0.8$, at time $t = T_{1/2}$ (45 h). It is observed from equations (6) that an important factor determining the efficiency is nP_{e-h}^n which becomes smaller for greater values of γ-photon energy. High energy photons absorbed by the semiconductor will, most likely, loose their energy by multiple Compton scattering with the conduction band electrons, before they can generate e–h pairs, thus contributing to the temperature rise of the material via electron–phonon interactions. It can be seen from table 4 that at large multiplicities the efficiency tends to zero while V_{oc} decreases more slowly after a slight initial increase.

The graph in figure 3 shows the variation of the efficiency, η and V_{oc} as functions of γ-photon energy when $E_g = 3.5$ eV. The peaks of the efficiency correspond to different multiplicities, i.e. when the photon energy becomes a multiple of the energy gap, but we note that they become weaker for large multiplicities. Therefore for higher γ-photon energy η is decreasing smoothly overall. This is in qualitative agreement with the behaviour of the cross section σ_ν for pair production by high energy photons [27c].

The V_{oc} attains a value of about 2.61 V at $E_\gamma = 3.5$ eV and drops very slowly as the γ-photon energy increases. Eventually V_{oc} and η become zero at γ-photon energy of about

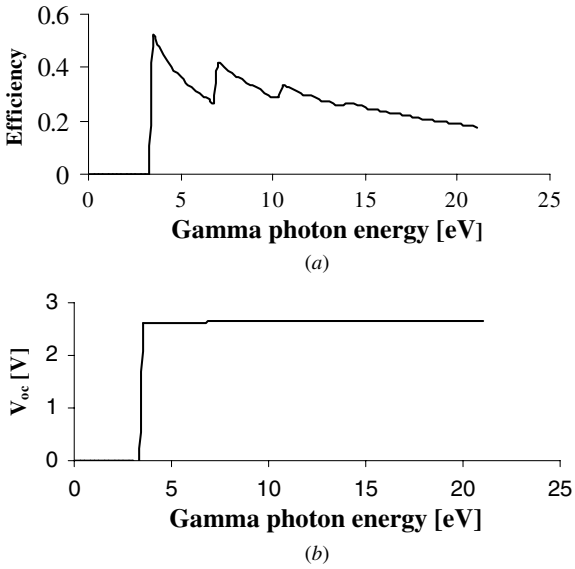

Figure 3. Variation of (*a*) efficiency, η and (*b*) open circuit voltage, V_{oc} for γ-photon energy, E_γ up to $6E_g$, when $E_g = 3.5$ eV and $P_{e-h} = 0.8$.

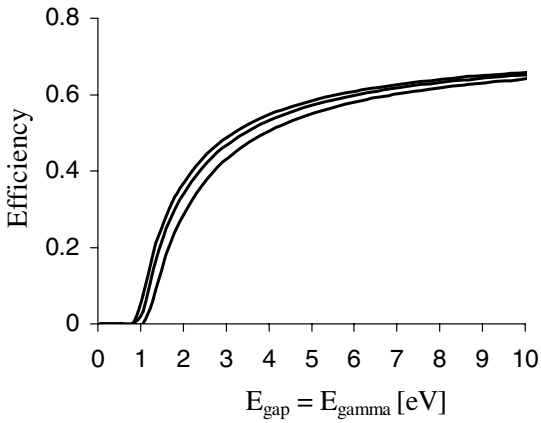

Figure 4. Efficiency attained by a GRPVC with the parameters of table 2, illuminated by 1 mol of isomers with various $T_{1/2}$. Top line: $T_{1/2} = 50$ h; middle line: $T_{1/2} = 50$ d; bottom line: $T_{1/2} = 50$ y.

1.75 keV (multiplicity 500), where the J_l itself becomes very small compared to the dark current J_s. This is a cut-off for the photon energy beyond which the illumination does not produce any measurable output from the GRPVC.

These results indicate that the most appropriate semiconductor material to use with Th^*-229 is one with direct gap $E_g = 3.5$ eV at room temperature. There are several semiconductor materials having energy gap close to or equal to 3.5 eV some of which are: AlAs ($E_{gL} = 2.9$ eV or $E_{g\Gamma} = 3.05$ eV); ZnSe ($E_{gL} = 2.9$ eV) [27a]; In_2O_3 ($E_g = 3.5$ eV); SnO_2 ($E_g = 3.5$ eV) [28c]. This analysis shows that with the use of nuclear isomers, it is possible to design semiconductor materials for GRPVCs, such that η and V_{oc} become optimal. An interesting feature of this system is that the efficiency becomes maximum only when $E_g = E_\gamma$, and it must be noted that only for energy gaps close to about 10 eV the efficiency reaches the saturation value of about 70% (see figure 4).

Table 5. η and V_{oc} for the Th-229 isomers.

$D_e = D_h$ (cm^2 s^{-1})	$N_{ci} = N_{vi}$ (cm^{-3})	Th-229 m1 (3.5 eV, 45 h)	Th-229 m2 (7.6 eV, 5 h)
500	As table 2	$\eta = 52.4\%$ $V_{oc} = 2.672$ V	$\eta = 63.72\%$ $V_{oc} = 6.911$ V
As table 2	3×10^{18}	$\eta = 57.31\%$ $V_{oc} = 2.911$ V	$\eta = 65.72\%$ $V_{oc} = 6.911$ V
As table 2	3×10^{16}	$\eta = 62.20\%$ $V_{oc} = 3.149$ V	$\eta = 68.30\%$ $V_{oc} = 7.149$ V

The requirement of employing large energy gap semiconductors in order to approach the ultimate efficiency of GRPVCs, could prove a challenge to meet. Semiconductors with energy gap of the order of 5 eV or greater must involve materials in the realm of insulators. Thus the question arises: can one dope insulators by donor or acceptor materials in order to produce the large energy gap semiconductors needed for this application? The answer to this question seems to be in the affirmative. Indeed, one intensively studied example is carbon (in the diamond form) which has energy gap 5.47 eV. It is possible to dope diamond with boron (acceptor), with phosphorus or sulfur (donors), to produce p-type and n-type semiconductor materials respectively with large energy gap [36, 37]. With a system of semiconductors of this nature, it will be possible to attain efficiency very close to 60%. Another possibility is the use of A*l*N (aluminium nitride) which has energy gap 6.2 eV [38, 39]. Mg-doped and Si-doped A*l*N produces a p-type and an n-type semiconductor respectively, while other impurities such as oxygen and carbon can also be used to produce n-type and p-type A*l*N-based semiconductors respectively [40, 41]. The deployment of A*l*N in GRPVCs can lead to efficiency in excess of 60%. One should bear in mind, however, that in view of table 5, an appropriate option of semiconductor parameters can also enhance the efficiency further still. On the nuclear physics side, the challenge is to produce relatively long-lived isomeric states, other than those of Th-229 considered here, with γ-photon energy emission close to 10 eV. This is a relatively new and developing area of research in nuclear physics. It is possible however, in principle, to excite isomeric states in one of the known modes: shape excitation, spin-trap excitation (spin mismatch between the excited and the ground states) and K-trap excitation (mismatch of the total angular momentum projection along the axes of nuclear symmetry of the excited and the ground states) [23, 24]. An alternative approach is the one discussed briefly in the theory section. This comprises the stages: (i) select a radioactive material known to decay naturally by γ-photon emission of relatively low energy (several keV or even a few MeV), for example Co-60 as shown in figure 1(*a*) with half-life 5.3 y, or select one of the manufactured higher energy nuclear isomers, (ii) pass the radiation through a thin foil of aluminium to ensure that other charged particle radiation such as α or $\beta^{\pm}$ will be eliminated, (iii) pass the 'high' energy γ-photons through one of the commercially available scintillating materials, which come in various forms: organic; inorganic and gas [43]. An advantage of using scintillators with high energy γ-photons in GRPVCs is that they can yield many thousands of low energy photons, which are appropriate for the GRPVCs, for every MeV of the γ-photons energy entering the scintillator. This will increase the light generated current of the GRPVC, hence it can enhance its V_{oc} and efficiency. The conversion efficiency of the scintillator is as important as its yield. For example the NaI(Tl) (thallium doped sodium iodide) has yield 3.8×10^4 photons/MeV and efficiency 100%, while CsI(Tl) (thalium doped caesium iodide) has yield 6.5×10^4 photons/MeV and conversion efficiency only 45% [43]. It must be stressed however that the photons produced by the scintillator must have energy close to 10 eV, for maximum GRPVC conversion efficiency. Further details on these interesting applications to the GRPVC operation will be discussed elsewhere in future work.

This maximum efficiency constitutes the ultimate efficiency that can be reached by the GRPVC with the prescribed diameter and semiconductor parameters, when illuminated by 1 mole of a gamma-source. Since the value of the maximum efficiency can be improved by reducing the size of the GRPVC and/or by increasing the mass of the gamma-source, the maximum efficiency of a GRPVC does not constitute a representative measure for the ultimate efficiency of all GRPVCs. Note that the half-life of the nuclear source does not affect the ultimate efficiency of the GRPVC. Thus for Th-229 m1 ($E_\gamma = 3.5$ eV, $T_{1/2} = 45$ h) we found $\eta = 52.2\%$ and $V_{oc} = 2.626$ V, and for Th-229 m2 ($E_\gamma = 7.6$ eV, $T_{1/2} = 5$ h) $\eta = 63.4\%$ and $V_{oc} = 6.673$ V. Hence isomers with shorter half-life give larger η and V_{oc} as expected. Table 5 shows how the semiconductor parameters affect the efficiency of the photovoltaic cell. The semiconductor parameters that are not shown in the table are the same as those of table 2. We see from this table that the longest lived isomers have slightly lower values for η and V_{oc}, as expected. It is also interesting to note that the smaller densities of states N_c and N_v favour higher efficiency and V_{oc} in both isomers.

It would be interesting to see how the efficiency and V_{oc} change with time, hence giving us some idea about the operational life of the cell. This is shown in table 6, for the same semiconductor parameters and for the energy gap $E_g = 3.5$ eV, after 1, 10, 20, 40 and 80 half-lives of operation, and in figure 5 for up to 100 half-lives.

Table 6 shows that 75 days later the cell has efficiency $\eta = 37.3\%$ and $V_{oc} = 1.909$ V, and even after 80 half-lives (150 days) the cell's efficiency is about 23% and the $V_{oc} = 1.192$ V.

The graph of figure 5 shows η and V_{oc} for times up to 100 half-lives (4500 h) which is about 188 days. It is interesting that half a year later the efficiency is still above the 10%, and the V_{oc} above the 0.5 V level. The graphs also show a very slow linear rather than exponential decline of η and V_{oc}.

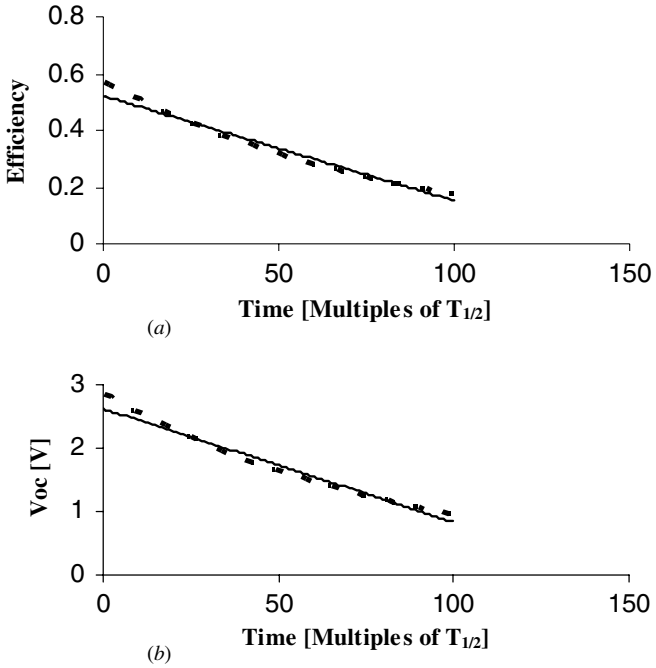

Figure 5. The variation of: (*a*) efficiency, η and (*b*) V_{oc} for times up to $100T_{1/2}$, when $E_g = 3.5$ eV. The broken lines show an exponential decay for comparison.

Table 6. η and V_{oc} for multiples of $T_{1/2}$ ($E_g = 3.5$ eV).

Time (h)	Efficiency η (%)	Open circuit voltage V_{oc} (V)
0.0	52.2	2.626
45.0	51.8	2.608
450	48.5	2.447
900	44.7	2.267
1800	37.3	1.909
3600	22.5	1.192

One can also use nuclear isomers of much longer half-life, which can keep the GRPVC operating for many years at high values of η and V_{oc}. In this case the decay rate will be smaller, leading to smaller γ-photon intensity and hence to reduced light generated currents and slightly less efficiency as seen from figure 4. This, however, can be compensated by the much lower Auger recombination rate due to the larger E_g, as well as by the very long operating life of the GRPVC. For example, an isomer with half-life $T_{1/2} = 50$ days (4.32×10^6 s) has $\lambda = 1.6 \times 10^{-7}$ s^{-1} so that, using the same GRPVC parameters as for Th-229, one mole of the isomer would produce an output $V_{oc} = 2.539$ V at an initial efficiency $\eta = 50.4\%$ which would drop to $V_{oc} = 2.181$ V at $\eta = 42.9\%$, 2.74 years later. Thus the efficiency of such GRPVC would reduce by only $1.5 \times 10^{-4}\%$ per day. If the half-life were 50 years instead, then the initial output would be $V_{oc} = 2.388$ V at $\eta = 47.24\%$ (only 3.16% lower than that for $T_{1/2} = 50$ days). One hundred years later the output would be $V_{oc} = 2.37$ V at $\eta = 46.87\%$. After $100T_{1/2}$, i.e. 5000 y later, the output from the GRPVC will be $V_{oc} = 0.5$ V at $\eta = 10\%$. It is the latter result that suggests the use of GRPVCs with long half-life isomers, for space travel and exploration. These results will certainly change when radiative and Auger recombination, as well as impact ionization processes will be taken into account. However, from previous work on Auger recombination and impact ionization, the reduction in the efficiency of solar PVCs was only by about 3% [26, 32, 33]. In view of the fact that a larger E_g is needed for GRPVCs to work at high efficiency, taking into account equation (15) and the simple model formalism of the impact ionization probability in [32], the effect of these processes on the efficiency of GRPVCs could be less than 3%. That will be discussed in a forthcoming publication.

4. Discussion

We have explored the possibility of utilizing the naturally occurring, or artificially created, nuclear isomers emitting low energy γ-photons in order to drive the operation of photovoltaic devices. The discussion has been focussed in the use of the isomer $_{90}Th^{*229}$ which emits γ-photons of energy 3.5 eV, with half-life of 45 h. Using a 40 cm diameter spherical GRPVC and one mole of $_{90}Th^{*229}$, it has been found that the efficiency η and the V_{oc} of the cell become optimal only when the energy gap of the semiconductor material is close to the energy of the γ-photons emitted by the source. The efficiency and open circuit voltage of the GRPVC have been found in the present work to be surprisingly high at 52% and 2.6 V respectively at half-life of $_{90}Th^{*229}$. Even after one hundred half-lives, about half a year later, the efficiency is greater than 10% and the V_{oc} above 0.5 V. One interesting aspect of GRPVCs is that one has control over the intensity of the γ-photons by increasing the amount of moles of the isomeric material used, and/or making an appropriate geometrical design; reducing the size of the GRPVC for example. Also, since the operation of GRPVCs does not rely on sunlight, a large number of them can be enclosed and packed tightly without the need to occupy large areas of land, which is the case for solar PVCs. It is also possible for this technology to be implemented in space travel where long half-life GRPVCs can be producing electricity for centuries. We consider these to be advantages of GRPVCs over PVCs. The energy of the γ-photons is required to be low in order to be able to match the energy gap of existing semiconductor materials, but the results of this analysis encourage the manufacturing of semiconductors with large energy gaps. Although we have found that the ultimate efficiency that can be reached by the GRPVC we studied in this work is about 70%, this can improve by either reducing the size of the GRPVC or by increasing the amount of the nuclear isomer used. Thus the 70% maximum efficiency found here does not constitute the ultimate efficiency that can be reached by all GRPVCs.

Although large nuclei decaying by α- or β-particle emission can become isomers of the daughter nucleus, emitting high energy γ-photons, it is possible for the nucleus to be excited in the laboratory so as to emit low energy γ-photons and be made appropriate for the GRPVC application. The isomers $_{90}Th^{*229}$ (3.5 eV, $T_{1/2} = 45$ h) and $_{90}Th^{*229}$ (7.6 eV,

$T_{1/2} = 5$ h) can be produced by irradiating $_{90}Th^{229}$ with x-rays or with broad band synchrotron energy from 2.5 to 8 eV so that to be excited to these isomeric states. This is very important because the emission of the γ-photons by $_{90}Th^{*229}$ is not accompanied by the α- or β-particle and its antineutrino. Thus we get the energy of the γ-photons without the hazardous extra radiation and this protects the GRPVC. The isomeric material used, is not necessarily deemed useless at the end of its cycle of operation, it can be recycled in the sense that it can be isomerized and used again, provided the ground state is a stable nucleus. So from this point of view no material is consumed or wasted to cause pollution of the environment. Work is under way currently to extend the present theory in order to incorporate the effects of impact ionization and Auger recombination in homojunction and heterojunction GRPVCs. The study of heterojunctions will investigate the two closely spaced isomeric states of Th-229. A more detailed analysis of the effect of semiconductor parameters, other than the energy gap, on the efficiency of GRPVCs will also be presented.

In conclusion, this work studies an interface between semiconductor and nuclear physics, in an exploration of the possibility of utilizing γ-photons emanating from nuclear isomers, in order to drive the e–h pair generation processes in the semiconductors of photovoltaic cells. The aim is to develop nuclear-energy operated photovoltaic cells for the generation of electrical energy. Although there are semiconductors available with energy gaps matching the Th-229 (3.5 eV, 45 h) isomeric state, there are challenges on both sides. For ultimate GRPVC efficiencies, in semiconductor physics there is need to produce semiconductors with energy gaps close to 10 eV, which is in the realm of insulators. It is possible therefore that doping insulator materials with donors or acceptors, in a similar way to that in small energy gap semiconductors, could produce the n-type and p-type semiconductor materials with large energy gap [36–41], to be made appropriate for the application proposed in this work. In nuclear physics the challenge is to produce long-lived nuclear isomers in one of the modes: shape excitation; spin trap; K-trap type, emitting γ-photons with energy matching the relatively small semiconductor energy gaps. Alternatively, one can use γ-ray scintillators in conjunction with higher energy γ-photons emanating either from naturally occurring radioactive sources or manufactured nuclear isomers.

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