

Recent progress and perspective on batteries made from nuclear waste

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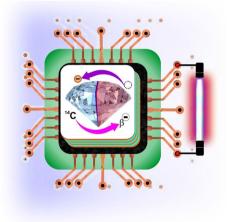
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Abstract

Sustainable energy sources are an immediate need to cope with the imminent issue of climate change the world is facing today. In particular, the long-lasting miniatured power sources that can supply energy continually to power handheld gadgets, sensors, electronic devices, unmanned airborne vehicles in space and extreme mining are some of the examples where this is an acute need. It is known from basic physics that radioactive materials decay over few years and some nuclear materials have their half-life until thousands of years. The past five decades of research have been spent harnessing the decay energy of the radioactive materials to develop batteries that can last until the radioactive reaction continues. Thus, an emergent opportunity of industrial symbiosis to make use of nuclear waste by using radioactive waste as raw material to develop batteries with long shelf life presents a great opportunity for sustainable energy resource development. However, the current canon of research on this topic is scarce. This perspective draws fresh discussions on the topic while highlighting future directions in this wealthy arena of research.

Graphical abstract

A long-lasting miniaturised nuclear battery utilising ¹⁴C radioactive isotope as fuel



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1 Introduction

Miniaturised power sources, especially batteries, are key drivers to attain energy security and to generate wealth in the society to achieve sustainability for human life [1]. In particular, the burning of fossil fuels has already shown the adverse consequences resulting in climate change, triggering newer types of natural calamities, e.g. floods and droughts, wildfire, Tsunamis, changes in land habitation that are emotive of the degrading ecosystem due to the increasing carbon emissions [1, 2]. Newer sources of energy generation are required to run our handheld devices, cell phones, watches and battery-operated vehicles. One key challenge in battery research has been to achieve an infinite lifetime which is acutely required for applications such as the black box of an aircraft to ensure aviation safety as well as space satellites and telecommunications including RFIDs tags of space live satellite/ satellite debris to read the owner's details and to perform damage prognosis. All these components although are a niche area, but instead of cost being a driver, performance here is a strong desirable attribute to set out new scientific ambitions that may dictate the future of human race. On the other hand, everyone wants their cell phone and computers to work for long in a single charging. It is therefore imperative to invest in battery research to maximise the opportunities for energy generation for meeting the growing demands of electronic handheld devices to last longer.

The radioactive materials are denser energy materials and radiate naturally. A small amount of radioactive material is sufficient to cause a controlled fission reaction to produce enormous amount of electricity at an expense of a small amount of nuclear waste. Worldwide, all nuclear energy power plants are producing this nuclear waste, and their management is a challenging task for the scientific community. Nuclear waste's decay takes long time, and during this decaying process, the nuclear waste possesses the potential to harm flora and fauna and it may take ~ 1000 years to become fully neutralised. Various types of nuclear waste can be categorised as uranium tailings (by-product of extraction uranium from ore), transuranic (TRU) waste (by-product during nuclear research and nuclear weapon development), low-level waste (materials containing radioactive contamination or exposed in nuclear fission), intermediate-level waste (nuclear reactor parts, chemical sludges, used reactor's part,), high-level waste (side product of nuclear fission reaction. Exposure to these materials can lead to fatal consequences.

Radioactive materials are unstable atomic nuclei tending to reach stable nucleus via α , β and γ decay. For instance, Carbon-14 (¹⁴C) is a radioactive polymorph of Carbon-12 (¹²C) having a half-life of about 5730 years. ¹⁴C is a by-product of nuclear waste and is currently buried deeply in earth to avoid the release of harmful radiations [3, 4]. Beta decay of ¹⁴C is a highly exothermic process that is complemented by the release of beta particles while carbon transforms into nitrogen which is depicted in Fig. 1a.

During this process, the neutron in the nucleus turns into a proton which releases an electron, or a negative particle called a beta particle. Well-directed engineering can help to use this continuous process for producing batteries with exceptional life. For example, suitable coating or wrapping of radioactive materials with semiconductor materials to channel those electrons can create a battery. As such, there are many β emitters and their specific emission energy is very high and these are, for instance, ¹⁸⁷Re (2.456 keV), ³H (18.59 keV; half-life of 12.32 years), ¹⁴C (155 keV; half-life of 5730 years), and ⁹⁰Sr (546 keV; half-life of 28 years). This speaks for the fact that the energy possessed by the β emitters is very high [5] compared to the solar photon energy (~ 3 eV). More radioactive isotopes are summarised in Fig. 1b to reflect on their β particle energy and their half-life.

Decaying is a steady process which can be well predicted, for instance, one gm of material becomes half a gram at the end of its half-life or in other words, the total energy released from a material reduces to half (Fig. 1a–b). The half-life of ¹⁴C is 5730 years meaning that half amount of ¹⁴C becomes consumed in 5730 years and half of its energy is vanished by then. Additionally, half-life is inversely proportional to the specific power. For example, if 100 atoms of 'A' material (half-life 10 days) have decayed by 50% in 10 days, it means 5 atoms would release 5 β particles per day. On the other hand, if 100 atoms of a 'B' material (half-life 1 day) decay in 1 day, then 50 β particles get released each day. It means that the battery life of material B is shorter compared to the material A.

Semiconductor plays an important role to tap or to convert the energy of the β particles into electrical energy [8]. The β particles collide with the semiconductor interface and generate the electron-hole pairs (Fig. 1c-d). Depending on the type of β particle, there could be as many as 100–1000 electron-hole pairs generated from a single β particle. Until now semiconductor materials have not shown 100% conversion and there is a high probability of leakage of β particles thus, the vulnerability to release the radioactive radiation. By increasing the amount of radioactive material one can

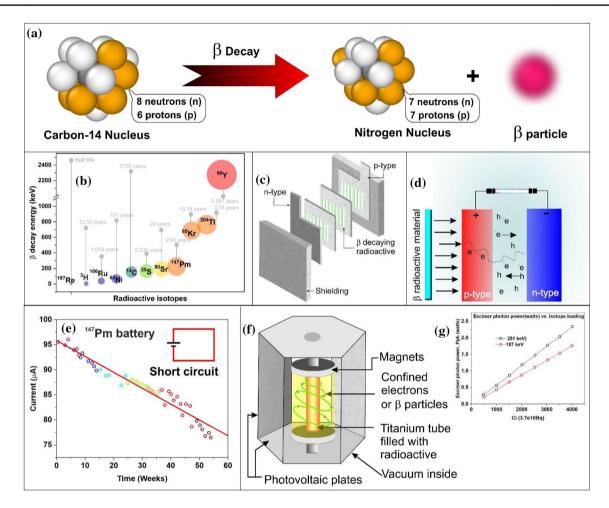


Fig. 1 a β decay reaction of ¹⁴C nucleus, **b** energy release in β - decay in various isotopes and their half-life, **c** a schematic of battery using β -decaying radioactive materials with semiconductor (*p*–*n* junction), **d** schematic conversion of β decay into electric energy by semicon-

ductor, **e** Nuclear battery current decrease in short circuit (Pm halflife is 2.6 years) [6] **f** 3D sketch of a nuclear battery using radioactive fused with noble gas to produce excimer **g** shows the power of an excimer photon [7]. (Color figure online)

enhance the efficiency to a certain threshold but afterward it saturates due to self-absorption of β particles. Increasing the β particles energy by changing radioisotopes can generate more energy, but it increases the internal damage to the semiconducting material especially when the energy from the emitting particles goes past 300 keV [8, 9].

Materials with wide band gap such as diamond $E_{\rm gap} = 5.5$ eV can be used for high energy efficiency conversion and low backscattering due to the low atomic number [9, 10]. Therefore, newer design strategies can immensely benefit the batteries to enhance the conversion efficiency.

Many research groups have attempted to develop nuclear batteries. The pioneering development in this area took place at the Donald W. Douglas Laboratories—a subsidiary of McDonnell Douglas Corp when Olsen et al. used ¹⁴⁷Pm beta isotope [11, 12] to develop the first nuclear battery in 1974. They called this battery a betacel battery and used it in a pacemaker as few of them showed 100

mW and 50 mW power using 147 Pm radioactive materials (half-life = 2.62 years).

The battery's short circuit current decreases at the same rate as half-life of the betacel (decreasing flux) which can be seen in Fig. 1e. In the absence of the radiation damage, the life-time of the battery completely coincided with the half time of the radioactive material [6].

The largest challenge moving forward is that the β decay (high energy electron bombardment) process damages the ordinarily available semiconductor materials, resulting in very low conversion efficiency. The large band gap materials are quite successful in increasing efficiency as well to protect against radiation damage. This is the reason why synthetic diamond with a band gap of 5.5 eV is a better choice over silicon which has a band gap of 1.12 eV [13]. Ayers et al. [7] proposed an improved design of a nuclear battery to increase the battery power from 100 mW to 1 W while reducing the radiation-induced damage

to the semiconductor material. In this design, radioactive material was filled in the thin-walled Ti tube and the β particles emitted into the vacuum through the tube. The β particles start moving in a helical or spiral around the tube due to interaction with the magnetic field placed at the end of the tube. A photon transparent tube containing an excimer precursor gas surrounds the radioisotope tube such that the confined β particles have thousands of interactions with the precursor gas atoms to generate the excimer photons. It resulted in the generation of multiple photons thus, absorbing maximal energy from the electron or β decay. These excimer photons are utilised by the photovoltaic cells which surround the tube and produce electrical energy (Fig. 1f). Nevertheless, the increase in battery power requires either high loading of radioisotopes or high energy β decay isotopes and both need extremely efficient shielding protection to avoid the leakage of nuclear radiation. An improvement in the design can open newer possibilities to enhance efficiency and high-power conversion.

Wang et al. [14] recently formulated a betavoltaic-powered electrochemical cell (BPEC), wherein the TiO₂ nanotubes array was used along with single-wall carbon nanotubes (SWCNTs). These nanotubes act as an anode in the cell (see Fig. 2a). The ⁶³Ni sheets were used as cathodes. The liquid electrolyte (polyiodide solution containing iodide/triiodide (I^{-}/I_{a}^{-}) redox couples) was filled in the tubes for better contact with the nanostructured anode for facilitating the redox reaction to transfer the β -generated carriers. In this way, the energy conversion efficiency achieved was about 8.74%. In another report, TiO_2 nanotube modified with ZrO_2 nanoparticles achieved 9.27% energy conversion efficiency with an open circuit voltage of 0.276 V [15]. Hwang et al. [16] used other strategies using ruthenium-based dye-sensitised TiO₂ as an electrode, where the emitted β -particles were absorbed by the dye. Here, the ¹⁴C works both as the source of β particles as well as the counter electrode.

This phenomenon is attempted to be exploited through several commercial pathways. For instance, a chemist from Bristol named Neil Fox created an enterprise named Arkenlight to develop a prototype using a ¹⁴C beta voltaic battery. Another California-based start-up claimed to produce a nano-diamond battery (NDB) [17] giving an endless battery with an infinite lifetime (estimated 28,000 years). A smartwatch is planned to be powered by NDB and it has been predicted to last for many years without needing any replacement. Bormashov et al. [18] reported a prototype nuclear battery using Schottky barrier diamond diode stacked with ⁶³Ni isotope which provides 0.93 mW power. There are also other companies developing the prototype of nuclear batteries such as BetaBatt, Widetronix, and CityLabs.

To summarise, the nano-diamond refers to a specific material it is being used for different energy applications like an additive in the lithium-ion battery to stop the dendritic growth of lithium (the cause of the explosion of the battery) [19] and is known for its chemical inertness and highest elastic modulus. The nano-diamond doped with boron and nitrogen can act as p and n-type semiconductors, respectively, as well as endowed with supercapacitor performance [20, 21]. As such, synthetic diamond has a higher threshold of radiation damage. Nano-diamonds can act both as a damage-resistant semiconducting material and for shielding from radiation. The doped and undoped diamond interface could be interfacial resistance less. Therefore, all these properties of diamond make it suitable for nuclear battery [22, 23].

2 Factors influencing the performance of a nuclear battery

The performance of a nuclear battery depends on several factors contributing to energy losses such as radiation losses (back scattering, self-absorption), nuclear losses and

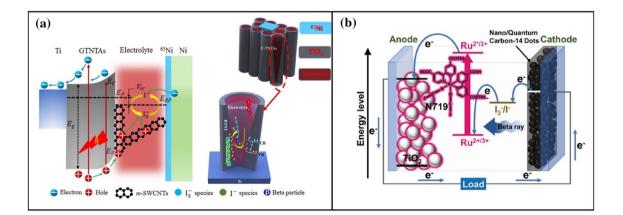


Fig. 2 Different designs of β particles driven cell **a** betavoltaic-powered electrochemical cell (BPEC) with TiO₂ nanotube arrays; **b** dye-sensitized betavoltaic cell with ¹⁴C as counter electrode as well as β emitter. (Color figure online)

electronic energy losses (electrode barrier, recombination, and collection loss). The β particles collision with semiconductor materials results in loss of energy in form of acoustic phonons and optical phonons Fig. 3a, which reduces electron flux density [24]. The efficiency of the nuclear battery also depends on the design of the battery and the process to harness the beta particles for electric energy [25]. Most energy loss during the conversion occurs during the use of different devices like Schottky junction, p-n junction, p-i-n junction, etc., and nano-diamond holds high promise to achieve high conversion efficiency by improving the diamond–metal interface [10, 26].

Liu et al. [27] compared the I-V characteristics of different supporting semiconductor materials or energy conversion systems like Schottky diode (Au–Si) and silicon p-njunctions along with the battery design. Schottky diode performed better for energy conversion compared to the p-njunction when bombarded with the ³H isotope [27]. However, the situation is different with the Ni⁶³ isotope, as shown in Fig. 3b. There are a few prototypes of nuclear batteries studied using Schottky barrier diamond diodes with different isotopes, as shown in Table 1. The ²³⁸Pu nuclear battery uses a source of a red light-emitting diode (15 mA, 1.65 V), but it causes significant lattice damage of the diamond due to the bombardment which limits the lifetime to 50% within 10 h [28]. On the other hand, there was no lattice damage during power reduction from the β source of ⁹⁰Sr, which was observed to last until 1400 h [27, 28]. The transport properties of β particles also affect the performance such as the emission angle of β particles, the penetration depth of Page 5 of 8 33

 Table 1
 Nuclear batteries fabricated with Schottky barrier diamond diodes performance with different isotopes [28]

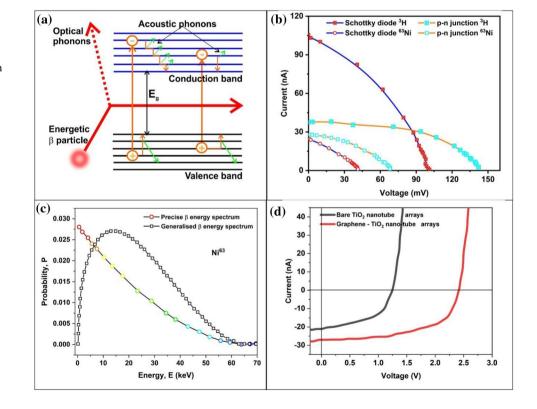
| Isotope | Open circuit voltage | Short circuit current | Output power | Efficiency |
|---|----------------------------|-----------------------------|--------------|------------|
| ⁶³ Ni (β-source) | 1.68 V | 55 nA | 45 nW | 0.6% |
| ¹⁴⁷ Pm (β -source) | 1.81 V | 2.4 mA | 2.4 mW | 1% |
| ⁹⁰ Sr ^{/90} Y (β-source) | 1.75 V | 510 nA | 0.55 mW | 0.004 |
| ²³⁸ Pu (α -source) | 1.85 V | 37 mA | 36 mW | 3.5% |

 β particles, and the self-absorption effect of the radioactive source.

Alam et al. [29] theoretically calculated the performance of ⁶³Ni β particles. The probability of high energy β particles or their distribution is broad and directly affects the performance estimation, as can be seen from Fig. 3c. The energy deposition of electron or electron–hole generation is very high at the surface but decreases as the thickness of semiconducting materials increases. The β particle energy decreases and, in the depth, it is insignificant to generate electron–hole pairs. The depth penetration of ⁶³Ni β particles in Si materials is around 21 mm.

The efficiency of nuclear batteries relies on methods such as increasing the active area or absorptivity of β particles [30]. The nano-porous structure and wide band gap are promising for absorption efficiency. Chen et al. [31] reported beta voltaic devices using TiO₂ nanotube arrays.

Fig. 3 a schematic electron– hole pair generation and energy loss, **b** *I–V* curve of Schottky diode (Au–Si) and silicon *p–n* junction device bombarded with the ³H and ⁶³Ni, [27] **c** theoretically calculated β particle energy distribution from ⁶³Ni [29], **d** *I–V* characteristics of bare Ti and graphene-coated nanotubes array [31]. (Color figure online)



The TiO₂ nanotubes suffer from conductivity losses due to wide bandgap. However, TiO₂ nanotube arrays (TNTAs) can enhance the absorption of beta radiation as well as the transportation of carriers. Graphene was deposited over the top surface of the nanotubes array to increase the conductivity between TNTAs and it achieved a conversion efficiency of 26.55% with 2.38 V open circuit voltage, as shown in Fig. 3d [31]. The protective layer to avoid the radioactive leak is also one of the most required factors for a battery to work efficiently. A Monte Carlo simulation for ⁶³Ni radioactive isotope revealed that the protective layer thickness ranges from 60 to 2 mm for diamond, SiO₂ and SiN semiconducting materials, respectively [32].

The reported specific energy of a nuclear β cell battery (Schottky barrier-based diamond diode) using ⁶³Ni (25% enriched) source is about 3300 mWh/g, which is ten times higher than the commercially available chemical batteries such as the Li- ion battery which has a specific energy of about 100–265 mWh/g [18]. Worth noting is the differences in physical size, for instance, the ⁶³Ni nuclear battery with a size of 5 mm × 5 mm × 3.5 mm attains a maximum power output of 0.93 mW and 0.9 V. The highly enriched ⁶³Ni can produce specific power of 40–50 mW/cm³ [18]. A pacemaker can be operated with 10 µW and the size of 1 cm³. Thus, nuclear batteries are beneficial for use in small sizes to power up devices for long-term needs consuming low power [33, 34].

3 Raw material availability and safety

Nuclear waste materials are hazardous and tackling them is a major issue [35]. The 14 C isotope of carbon is the byproduct of a nuclear plant/reactor. The graphite used as a moderator in the nuclear reactor partially converts into ¹⁴C. The nuclear waste graphite in form of blocks piles up to about ~ 25,000 tonnes worldwide [36] comprising of ¹⁴C isotopes and its extent is growing continually due to the ongoing drive to decommission the second generation nuclear reactors. ¹⁴C can be extracted for fuelling long-lasting batteries. The nuclear graphite waste can be used to extract ¹⁴C by heating and gasification and the residual graphite left for disposal after the process is far less dangerous [36, 37, 38]. The enriched 14 C is the major fuel source for the development of nuclear batteries in different designs. The ¹⁴C can be converted into synthetic diamond or it could also be used for different designs of batteries as discussed in the previous section. The ³H (tritium) isotope of hydrogen as a radioactive material is generally found in natural water, but it can also be obtained as a by-product from nuclear reactors (Nuclear explosion). ⁶³Ni is one of the isotopes of Ni which has a long half-life (100.1 years). However, a nuclear reactor uses many alloys containing ⁵⁸Ni and releases stable ⁶³Ni as a nuclear fission reaction. Promethium (¹⁴⁷Pm) is an artificial element (half-life 2.62 years) and is a fission product of nuclear reactors which occurs via beta decay from neodymium-147.

The ³H and ⁶³Ni are the safest radioactive materials to be utilised in the nuclear battery due to low energy radiation or short range of β particles. The radiation from these radioactive materials can easily be absorbed by plastic containers, protective gloves, etc. However, inhalation of such materials can cause fatal consequences [39]. ¹⁴C β particles have low energy but are sufficient to penetrate the dead layer of the epidermis which makes it dangerous for mankind.

4 Intellectual property and patents

Various components and allied processes of nuclear batteries are patented in several countries (USA, Japan, China, Russia, etc.). A patent claimed the nuclear battery reactor core containing fuel plutonium, carbon, hydrogen and zirconium [40]. The miniaturised nuclear battery with an output power of 0.012 μ W was also claimed [41]. Another patent claimed nuclear battery enabled with the capacity to operate in harsh environments. It was fabricated with a *p*–*n* junction and claimed to use radioactive isotopes -Nickel-63, Tritium, Scandium Tritide, Titanium Tritide and Promethium-147 as fuel. The patent was mostly focused on designing the battery to enhance its efficiency [42]. There are many more patents that claim different types of designs and their components, materials and fuels. However, in this perspective, we do not intend to review these patents in details.

5 End-of-life disposal and safety considerations

The radioactive materials have a spontaneous beta-decaying process which makes them dangerous to human life. The attempt to increase the battery life implies additional safety considerations to be incorporated through regular checks and monitoring of its status. It also entails safe handling and returns to the manufacturer after its complete use. Disposing of nuclear waste is dangerous and needs to be tightly secured in a sealed steel container. If the nuclear waste battery is disposed of normally in a dustbin, it poses the risk of radiation damage to society. In a hypothetical scenario, suppose a battery has been used/run for about 50 years but its manufacturer has gone bankrupt or closed the plant earlier, then it creates problems on who shall safely collect and dispose of the battery at the end-of-their useful life. In such scenarios, one viable option is that a battery licence must be taken from the competent Government body for regulation and their safe return and disposal, and every battery is assigned a wireless remote tracker and a serial number to track its location for

disposing it safely at the end of its life, but enhancement of power generation from such battery remains an issue.

6 Prospects of nuclear battery

- (1) The ideas for developing a nuclear battery or a beta voltaic battery emerged in the 1950s but limited progress has been achieved due to limited R&D in this area and restricted use of radioactive materials in general laboratories. This work has remained confined mostly to focused research communities, especially to nuclear scientists and nuclear physicists, due to safety risks, and this has confined the research to the laboratories equipped with safety considerations. Nuclear waste battery as a concept is less popular across the materials science and electrochemical community which hinders speedy developments in this area. Overall, the nuclear community needs to consider exploiting the hidden energy/potential within the nuclear waste rather than dumping it.
- (2) Nano-diamonds known for their highest bulk modulus, chemical inertness and electrically insulating nature are viable prospects for use as shielding material to surround the radioisotopes, which is an essential fuel of a nuclear battery. Doped diamond can act as a suitable semiconductor, and it is viable to synthesise it using ¹⁴C as a fuel source. Therefore, the ¹⁴C has high prospects for its use in the development of nuclear batteries.
- (3) Filling up the nuclear materials or radioisotopes in the nanotubes enhances the beta particles collection efficiency and increases the self-shielding. At nanoscale, increased surface area plays a vital role and it needs to also be considered as a crucial factor in the future R&D in this area of research.
- (4) There is a requirement to have an improved semiconductor material with long carrier diffusion lengths. The design of battery in such way where the interactions with β-particles with the materials reach to the highest possible level to achieve 100% conversion efficiency or 100% β particles converted into charge is required.
- (5) Gamma radiation emission during nuclear decay is a challenge to shielding compared to α and β particles because these particles can penetrate deeper, and this may need bulk shielding for a tiny battery.
- (6) The mechanisms and processes within the nuclear battery are analogous to photo-voltaic cells and the development of a nuclear battery can fuel the artificial photosynthesis process.
- (7) Integrating nuclear batteries with nanomaterials will play an effective role in developing nanodevices or smart miniatured healthcare devices.

- (8) Devices used for common people integrated with nuclear batteries pose the risk of increased electronic debris with radioactive radiation, which is a matter of grave concern in thinking of the development of this technology. R&D on this front needs to be pursued.
- (9) Nuclear batteries can last longer than the lifetime of their owner which causes a potential problem for safer end of life and disposal.

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