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^{232}U, ^{228}Th, ^{227}Ac, and ^{226}Ra primary radioisotopes: high-power sources for nuclear batteries

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Abstract - The present communication deals with the evaluation of the cumulative energy produced by the sequential α- or α- and β^--decay processes of ^{232}U, ^{228}Th, and ^{226}Ra isotopes, ending in the β^--decaying ^{212}Pb or ^{210}Pb isotopes, and by the β^- of ^{227}Ac, which ends in the ^{211}Pb isotope. The obtained results evidence that the overall decay chains greatly contribute to total specific power and total specific energy released by these radioactive sources. The actual maximum power as well as the total specific energy available at selected times are found from two to four times higher than those expected when, as usual, only the parent-to-daughter decay of ^{232}U, ^{228}Th, ^{226}Ra and ^{227}Ac are taken into consideration. The present analysis enabled to underline the rather complex trends of both total specific power and total specific energy as a function of elapsed time. It has been also evidenced that the decay processes initiated by the alpha-emitting ^{232}U, ^{228}Th, ^{226}Ra and by the beta-emitting ^{227}Ac isotopes occur actually by the sequential emission of different kinds of radiations. These concepts contribute to establish the physical viability of such nuclides for the powering of nuclear batteries and have significant implications on the technology of such devices.

1. Introduction

A long working life and the ability to operate under a range of extreme environmental conditions make the nuclear batteries appropriate systems to electrical power generation in applications for which long-lasting, highly reliable power sources are critical. In this context the nuclear batteries, firstly developed in the 1950's, have demonstrated their suitability to support a range of deep space and planetary missions (see for example: (Cataldo and Bennett, 2011; Ambrosi *et al.,* 2019; Ambrosi *et al*., 2021; Bushell *et al.,* 2021), as well as various terrestrial applications, such as medical devices (Parsonnet *et al.,* 2006), remote scientific installations (Grigoriev *et al.,* 2019) and smoke detectors (Ayodele *et al.,* 2019).

The practical applications of radioisotopes in devices that convert the energy generated from radioactive decays into electrical power (from here on indicated for brevity as radioactivity-to-power conversion) depend on many factors, either technical or physical. Of these two topics, the first is the most extensively tackled. It must be considered indeed that the several types of nuclear batteries up to now proposed operate on the basis of different direct and indirect conversion mechanisms, broadly categorized as thermal and radio-voltaic (Terranova 2022). The need to adapt the structure of the device to the way by which the energy released by a specific radioactive decay is captured and converted in electrical power, explains the existence of a variety of both thermoelectrical and radio-voltaic batteries.

The task to optimize the technical parameters effective for increasing the radioactivity-into-power conversion efficiency is widely addressed, mainly by tailoring innovative converter materials, engineering their structural organization and testing new device architectures (Prelas *et al*., 2014; Terranova, 2022). Conversely, fewer researchers are aimed at studying the physical factors that not only determine the ability to power a nuclear battery, but represent the ultimate limit of its functionality (Tavares and Terranova, 2023).

The fundamental physical factors we are talking are about the type of decay, radiation energy and half-life of the radionuclide under study. Therefore, the identification of radioisotopes best suited to efficiently

power nuclear batteries is presently one of the main topics to be addressed by researchers working in the field of applied nuclear physics (Dustin and Borrelli, 2021).

The present communication deals with the physical viability of nuclear batteries whose power source comes from the average total nuclear energy released by the sequential α- or α- and β^--decay processes starting from the α-decaying ^{232}U, ^{228}Th, and ^{226}Ra parent isotopes, ending in the β^--decaying ^{212}Pb and ^{210}Pb isotopes, and from the β^--decaying ^{227}Ac ending in the ^{211}Pb isotope*, viz*.,

$$^{232}\mathrm{U}(\alpha)\rightarrow {}^{228}\mathrm{Th}(\alpha)\rightarrow {}^{224}\mathrm{Ra}(\alpha)\rightarrow {}^{220}\mathrm{Rn}(\alpha)\rightarrow {}^{216}\mathrm{Po}(\alpha)\rightarrow {}^{212}\mathrm{Pb}, \quad (1)$$

$$^{228}\mathrm{Th}(\alpha)\rightarrow {}^{224}\mathrm{Ra}(\alpha)\rightarrow {}^{220}\mathrm{Rn}(\alpha)\rightarrow {}^{216}\mathrm{Po}(\alpha)\rightarrow {}^{212}\mathrm{Pb}, \quad (2)$$

$$^{226}\mathrm{Ra}(\alpha)\rightarrow {}^{222}\mathrm{Rn}(\alpha)\rightarrow {}^{218}\mathrm{Po}(\alpha)\rightarrow {}^{214}\mathrm{Pb}(\beta^-)\rightarrow {}^{214}\mathrm{Bi}(\beta^-)\rightarrow {}^{214}\mathrm{Po}(\alpha)\rightarrow {}^{210}\mathrm{Pb}, \quad (3)$$

$$^{227}\mathrm{Ac}(\beta^-)\rightarrow {}^{227}\mathrm{Th}(\alpha)\rightarrow {}^{223}\mathrm{Ra}(\alpha)\rightarrow {}^{219}\mathrm{Rn}(\alpha)\rightarrow {}^{215}\mathrm{Po}(\alpha)\rightarrow {}^{211}\mathrm{Pb}. \quad (4)$$

Except for the ^{232}U parent nucleus, all radioisotope members of the above four decay chains belong to the natural ^{238}U, ^{235}U, and ^{232}Th radioactive decay series. As can be noted not only the parent and daughter radionuclides, but also the three, four or five descendant radioisotopes of decay chains (1)– (4), produce power and energy from their decays and therefore contribute to total specific power and energy generated in nuclear batteries based on those parent radionuclides.

To determine the total nuclear energy released by ^{232}U, ^{228}Th, ^{226}Ra and ^{227}Ac parent nuclei by their successive α- and/or β^--decays, we have evaluated the total specific nuclear power, $P_T(t)$, and the total, cumulative specific nuclear energy, $E_T(t)$ (both time-dependent quantities) available at the end of the four chains. This is possible for all the decay chains because the half-life values of the radioisotopes of each chain are progressively decreasing. The only exception is for the pair $^{218}\mathrm{Po}(\alpha) \rightarrow {}^{214}\mathrm{Pb}(\beta^-)$ in the ^{226}Ra decay chain, but the ratio of their half-lives is a little less than nine.

It is worth mentioning that the final ^{212}Pb radioisotope does not contribute at all to specific total power generated by the α-active chains (1)

and (2) because its half-life ($T_{1/2}$) is ~ 2.6 × 10^5 times longer than its precedent α-emitting ^{216}Po. At the same time, the average total energy produced per β^--decay of ^{212}Pb is about eighteen times less than the average total particle energies emitted by all α-active nuclides of chains (1) and (2) (see Table 1).

The same happens with ^{210}Pb and ^{211}Pb isotopes, which end the chains (3) and (4), respectively. Their half-lives are ~ 4.3 × 10^{12} and ~ 1.2 × 10^6 times longer than their respective parents ^{214}Po and ^{215}Po isotopes. Finally, the average total energy produced per β^--decay of ^{210}Pb is ~ 170 times less than the energy of the α-particle emitted by its parent ^{214}Po isotope. This factor drops to ~15 in the case of ^{215}Po(α) → ^{211}Pb(β^-) (see Tables 2 and 3).

2. Total specific power available and total specific energy generated

The basic assumption to obtain the total specific power, $P_T(t)$, and the total specific energy, $E_T(t)$, released by the radioactive sequences (1)–(4) was to start from 1g of an isotopically pure parent radionuclide with mass number A. The law of successive radioactive decay (Bateman, 1910; Kaplan, 1962) allows one to determine the number of each radioisotope, $N_i(t)$ (i = 1, 2, …, n), produced along a chain at any time t (n is the number of members of the chain). The initial conditions (t = 0) are thus $N_1(0) = N_0 = \mathcal{N}/M$, the number of nuclides contained in 1g of pure parent radionuclide, and $N_i(0) = 0$ for i > 1. Here, $\mathcal{N}$ = 6.022 × 10^{23} is Avogadro's constant, and $M \approx A$ is the atomic mass of the parent radioelement expressed in g. $N_i(t)$-values can be obtained from the general expression

$$N_i(t) = N_0 \cdot f_i(t), \quad i = 1, 2, \ldots, n \tag{5}$$

with

$$f_i(t) = \exp(-\lambda_1 t)/C_{i1} + \exp(-\lambda_2 t)/C_{i2} + \exp(-\lambda_3 t)/C_{i3} + \ldots + \exp(-\lambda_i t)/C_{ii} \tag{6}$$

in which the λ's represent the respective decay constants, $C_{11} = 1$, and

$$\Sigma\,(C_{i1}^{-1} + C_{i2}^{-1} + C_{i3}^{-1} + \ldots. + C_{ii}^{-1}) = 0, \quad i = 2, 3, ..., n. \tag{7}$$

Denoting by T_i (i = 1, 2, ..., 6) the half-life-values of radionuclides of chains (1)–(4), and by using $\lambda_i = (\ln 2)/T_i$, the expressions for the dimensionless constant C's in (6, 7) have been evaluated directly from the general solution of Bateman's system of equations that gives the number of active nuclear species with time in a radioactive decay chain (Bateman, 1910):

$$C_{21} = T_1/T_2 - 1;\ C_{22} = -C_{21} \tag{8}$$

$$C_{31} = (1 - T_2/T_1)(T_1/T_3 - 1);\ C_{32} = -C_{21}(T_2/T_3 - 1);\ C_{33} = (T_1/T_3 - 1)(T_2/T_3 - 1) \tag{9}$$

$$C_{41} = (1 - T_2/T_1)(1 - T_3/T_1)(T_1/T_4 - 1);\ C_{42} = -C_{21}(1 - T_3/T_2)(T_2/T_4 - 1) \tag{10a}$$
$$C_{43} = (T_1/T_3 - 1)(T_2/T_3 - 1)(T_3/T_4 - 1);\ C_{44} = -(T_1/T_4 - 1)(T_2/T_4 - 1)(T_3/T_4 - 1) \tag{10b}$$

$$C_{51} = (1 - T_2/T_1)(1 - T_3/T_1)(1 - T_4/T_1)(T_1/T_5 - 1) \tag{11a}$$
$$C_{52} = -C_{21}(1 - T_3/T_2)(1 - T_4/T_2)(T_2/T_5 - 1) \tag{11b}$$
$$C_{53} = (T_1/T_3 - 1)(T_2/T_3 - 1)(1 - T_4/T_3)(T_3/T_5 - 1) \tag{11c}$$
$$C_{54} = -(T_1/T_4 - 1)(T_2/T_4 - 1)(T_3/T_4 - 1)(T_4/T_5 - 1) \tag{11d}$$
$$C_{55} = (T_1/T_5 - 1)(T_2/T_5 - 1)(T_3/T_5 - 1)(T_4/T_5 - 1) \tag{11e}$$

$$C_{61} = (1 - T_2/T_1)\,(1 - T_3/T_1)\,(1 - T_4/T_1)(1 - T_5/T_1)(T_1/T_6 - 1) \tag{12a}$$
$$C_{62} = -C_{21}(1 - T_3/T_2)(1 - T_4/T_2)(1 - T_5/T_2)(T_2/T_6 - 1) \tag{12b}$$
$$C_{63} = (T_1/T_3 - 1)(T_2/T_3 - 1)(1 - T_4/T_3)(1 - T_5/T_3)(T_3/T_6 - 1) \tag{12c}$$
$$C_{64} = -(T_1/T_4 - 1)(T_2/T_4 - 1)(T_3/T_4 - 1)(1 - T_5/T_4)(T_4/T_6 - 1) \tag{12d}$$
$$C_{65} = (T_1/T_5 - 1)(T_2/T_5 - 1)(T_3/T_5 - 1)(T_4/T_5 - 1)(T_5/T_6 - 1) \tag{12e}$$
$$C_{66} = -(T_1/T_6 - 1)(T_2/T_6 - 1)(T_3/T_6 - 1)(T_4/T_6 - 1)(T_5/T_6 - 1). \tag{12f}$$

The initial ($t = 0$) activity of a pure radioisotope of atomic mass A (expressed in g) and half-life T (expressed in annum) is defined by

$$A_0 = \lambda \cdot N_0 = \frac{\ln 2}{T \cdot f} \times \frac{m_0}{A} \cdot \mathcal{N}, \tag{13a}$$

where $\lambda = (\ln 2)/T$ is the decay constant, $f = 3.1557 \times 10^7$ is the annum → second conversion factor, and $N_0 = \mathcal{N}/A$ is the number of radionuclides contained initially in $m_0 = 1$g of the active material. Therefore, it results

$$A_0 = \frac{1.323 \times 10^4}{A \cdot T}\ \text{TBq}, \tag{13b}$$

noting that 1TBq = 10^{12} Bq, and 1 Bq = 1 dps (disintegration per second).

The specific activity of each member of a radioactive chain is given by

$$A_{\mathrm{i}}(t) = 1.323\times10^{4} \cdot \frac{f_i(t)}{A_{\mathrm{p}} \cdot T_i} \ \mathrm{TBq/g}, \tag{13c}$$

and the total specific activity at a time t is evaluated as

$$A_{\mathrm{T}}(t) = \frac{1.323 \times 10^{4}}{A_{\mathrm{p}}} \cdot \sum \frac{f_i(t)}{T_i} \ \mathrm{TBq/g}, \tag{14}$$

where A_{p} is the mass number of the parent radioisotope, T_{i} (i = 1, 2, …, n) is the half-life (expressed in a) of each member of the radioactive chain, and n is the number of radioisotopes of the chain.

To obtain the specific power available for each radioisotope of a chain, $P_{\mathrm{i}}(t)$, it suffices to introduce the factor $\bar{E}_{\mathrm{i}} \cdot F$ into formulas (13, 14), where $\bar{E}_{\mathrm{i}} = \bar{E}_{\alpha,\beta} + \bar{E}_{\gamma}$ (expressed in MeV) is the average total energy released per decay, *i.e.*, the average energy carried out by the α- or β^{-}-particle, $\bar{E}_{\alpha,\beta}$, plus the average energy of the accompanying γ-rays, $\bar{E}_{\gamma}$, and $F = 1.6022 \times 10^{-13}$ is the MeV → J conversion factor. Therefore, one has

$$P_i(t) = \frac{2120}{A_{\mathrm{p}}} \cdot \frac{f_i(t)\bar{\mathrm{E}}_i}{T_i} \ \mathrm{W/g}, \tag{15}$$

and the total specific power made available by a radioactive chain results

$$P_{\mathrm{T}}(t) = \frac{2120}{A_{\mathrm{p}}} \cdot \sum \frac{f_i(t)\bar{\mathrm{E}}_i}{T_i} \ \mathrm{W/g}. \tag{16}$$

Finally, the average total, cumulative, specific nuclear energy generated by a radioactive chain after an elapsed time t_{e} (in a) from preparation of a pure source of the given parent nuclide is evaluated as

$$E_{\mathrm{T}}(t_{\mathrm{e}}) = \frac{26.8}{A_{\mathrm{p}}} \sum_{i=1}^{n} \frac{\bar{\mathrm{E}}_i}{T_i} \int_0^{t_{\mathrm{e}}} f_i(t)\,\mathrm{d}t \ \ \mathrm{MWh/g}. \tag{17}$$

3. The ^{232}U chain: five successive α emissions

The radioisotope ^{232}U was identified for the first time in 1949 by Gofman and Seaborg at the Berkeley 60-in Cyclotron after bombarding a ^{232}Th target with 14-MeV deuterons, and subsequent β^--decay of produced ^{232}Pa (Gofman and Seaborg, 1949; see also Fry and Thoennessen, 2013). Currently, appreciable amounts of ^{232}U isotope are obtained in nuclear reaction chains from $^{234,\ 235,\ 238}$U isotope fuels containing ^{232}Th impurities during reactor operation (Rhodes and Maldonado, 2022). Alternative methods for production of ^{232}U have been reported by Artun (Artun, 2020) and Kulikov (Kulikov *et al.*, 2020). This last paper analyzed also advantages and drawbacks of using ^{232}U to power RTG (Radioisotope Thermoelectric Generator) batteries. The mixing of small amounts of ^{232}U with the long-lived ^{238}Pu and ^{241}Am alpha-emitters has been found to play an important role in increasing the thermal power output of radioisotope power systems (Ambrosi *et al.,* 2021).

The decay data to be used in calculating activities, specific power, and cumulative specific energy generated for each member of ^{232}U and ^{228}Th families, and the total of these quantities, $A_T(t)$, $P_T(t)$, and $E_T(t)$, are displayed in Table 1.

Table 1 - Decay data for radioisotopes of ^{232}U and ^{228}Th decay chains*

i	Isotope (decay mode)	Half-life $T_{½}$[a]	$\bar{E}_{\alpha,\beta}$ (MeV)	$\bar{E}_{\gamma}$ (MeV)	$\bar{E}_i$** (MeV)	decay constant. λ [a^{-1}]
1	^{232}U(α)	68.9	5.286	0.019	5.305	0.01006
2	^{228}Th(α)	1.9125	5.394	0.023	5.417	0.36243
3	^{224}Ra(α)	0.9943×10^{-2}	5.642	0.013	5.685	69.712
4	^{220}Rn(α)	1.76×10^{-6}	6.287	0.0006	6.287	3.938×10^{5}
5	^{216}Po(α)	4.56×10^{-9}	6.778	0	6.778	1.52×10^{8}
6	^{212}Pb(β^-)	1.21×10^{-3}	0.100	0.222	0.322	5.73×10^{2}

*Data have been taken from (Bé *et al*., 2017; Kondev *et al*., 2021).

** $\bar{E}_i = \bar{E}_{\alpha,\beta} + \bar{E}_{\gamma}$ is the average, total energy available per decay.

By applying Eqs. (5–13) and using data of Table 1 we have obtained the trends of specific activity for ^{232}U chain depicted in Fig. 1. It is seen that ^{228}Th, ^{224}Ra, ^{220}Rn, and ^{218}Po are in equilibrium with a maximum specific activity of 0.747 TBq/g at $t = 10.18$ a (Fig. 1-a).

The total specific activity (Fig. 1-b) increases from its initial value of ~ 0.82 TBq/g at $t = 0$ to a maximum of 3.74 TBq/g at $t = 9.56$ a, and the curve $A_T(t)$ has an upward concave trend for $t \geq 20$ a (not shown).

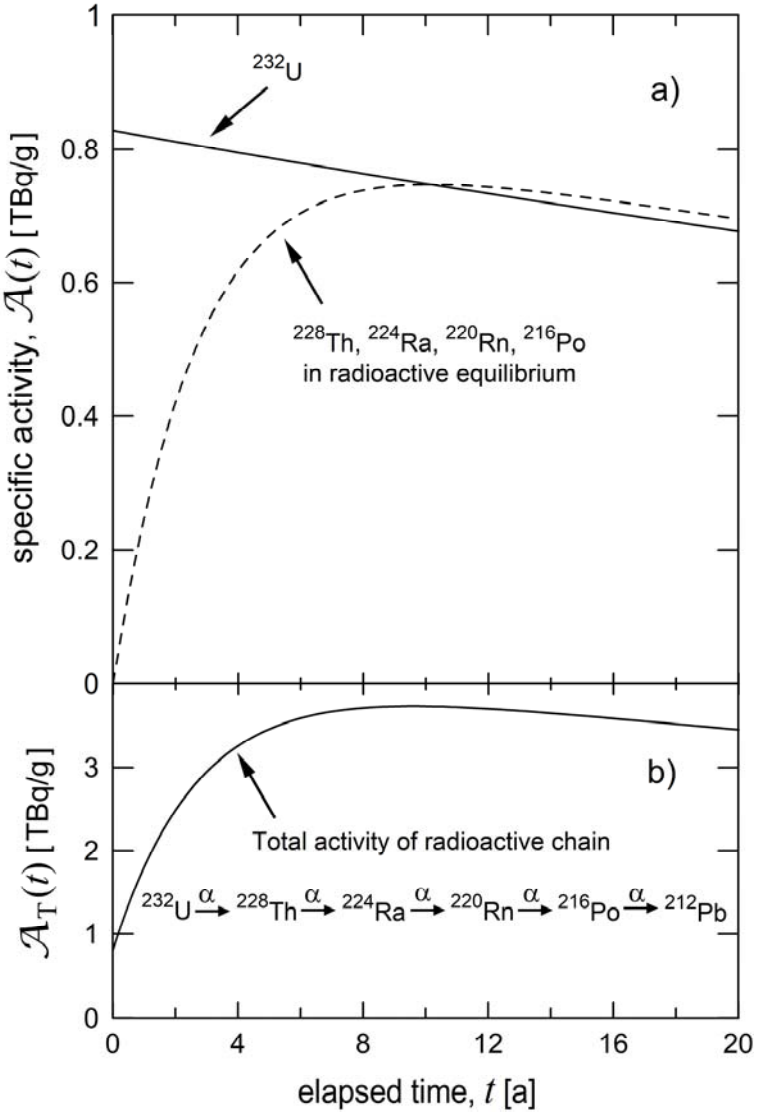

Fig. 1. In part a) the specific activity, $A(t)$, is plotted as a function of time for each radionuclide of ^{232}U decay chain. Part b) shows the total specific activity, $A_T(t)$, which reaches the maximum value 3.74 TBq/g at $t = 9.56$ a.

The trends of specific power available for each radioisotope of the ^{232}U chain, as well as the trend of the total specific power (in W/g), are shown in Fig. 2-a. Except for the ^{232}U parent nucleus, all other curves $P_i(t)$ (i = ^{228}Th, ^{224}Ra, ^{220}Rn, and ^{218}Po) show the maximum at $t \approx 10$ a, with values ranging from ~ 0.65 W/g (^{228}Th) to 0.81 W/g (^{216}Po). The total specific power increases from the initial value 0.7 W/g up to a maximum of 3.53 W/g after an elapsed time of 9.63 a.

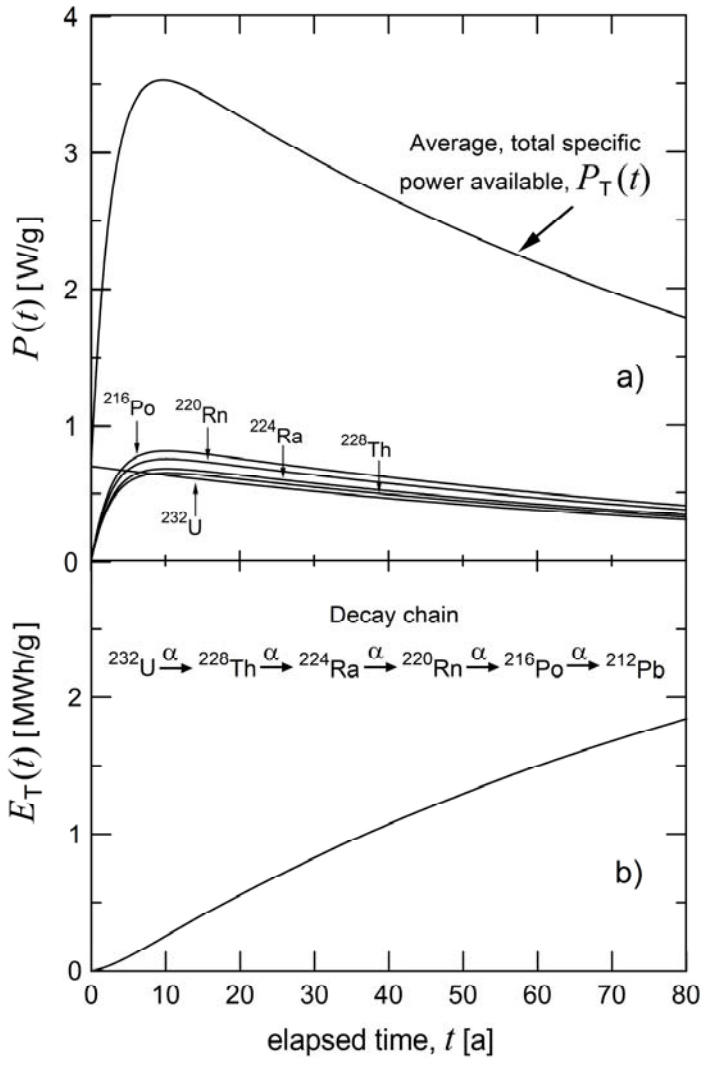

Fig. 2. In part a) the average, specific power available, $P(t)$, is shown for each radioisotope member of ^{232}U decay chain as a function of elapsed time. The upper curve represents the average, total specific power, $P_T(t)$, which has the maximum ~ 3.5 W/g at 9.6 a. In part b) is plotted the total, cumulative, specific energy generated. This curve increases asymptotically, approaching 3.4 MWh/g (not shown).

As regards the total, cumulative, specific energy generated after a time *t* (Fig. 2b), the trend is an increasing curve which approaches asymptotically the value 3.4 MWh/g (not shown), reaching ~ 1 MWh/g after 37 a, and ~ 1.8 MWh/g after 80 a.

4. The ^{228}Th decay chain: four successive alpha emissions

The radioactivity of element Thorium was first detected in 1898 by G. C. Schmidt (Schmidt 1898) and three months later by M. Curie (Curie 1898). The currently known ^{228}Th radioisotope was the one identified by O. Hahn (Hahn 1905) as the great-granddaughter of the natural ^{232}Th decay "family". Presently, large amounts of ^{228}Th have been obtained by high neutron flux irradiation of ^{226}Ra targets at Oak Ridge National Laboratory-ORNL (Kukleva *et al*., 2015).

Following the same procedure applied to ^{232}U, the results for the ^{228}Th chain have been presented in three parts in Fig. 3.

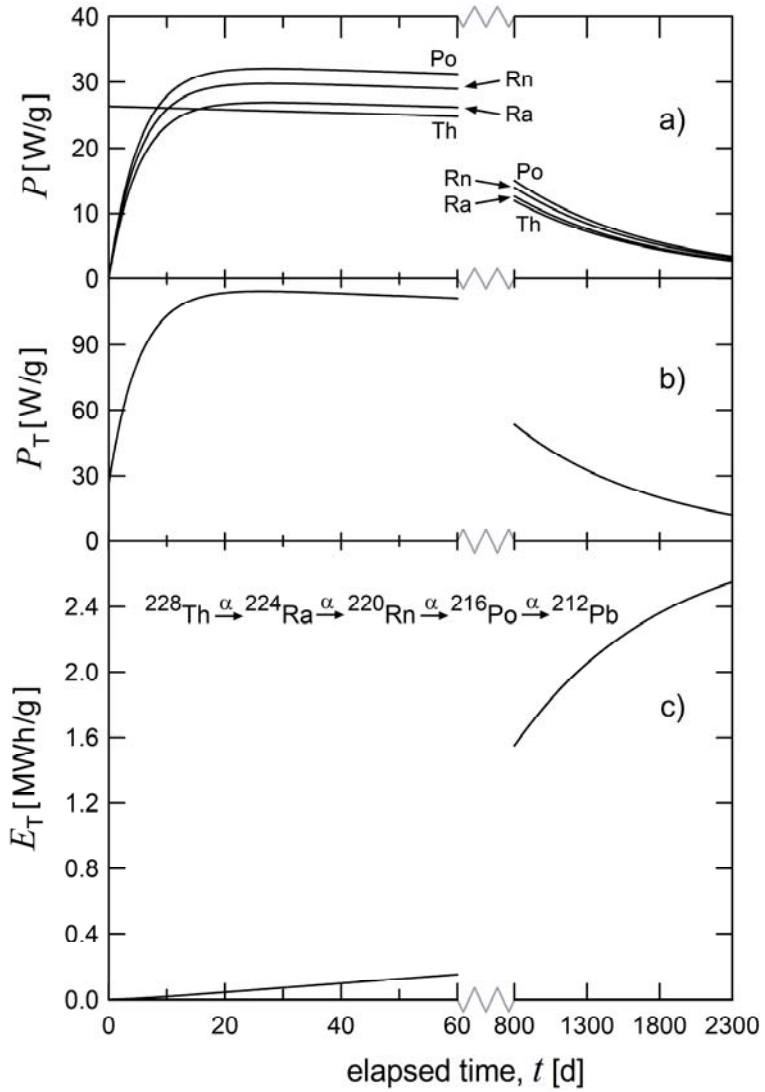

Fig. 3. The ^{228}Th decay chain produces specific power, $P(t)$, total specific power, $P_T(t)$, and total, cumulative, energy generated as function of time, $E_T(t)$, as shown in parts a), b), and c), respectively. Like in Fig. 2a), the increasing contributions to $P_T(t)$ due to the descendant isotopes of ^{228}Th parent nucleus in Fig. 3a) come mainly from the sequential increase of average, total energy released per decay, $\bar{E}_i$, of each radioisotope (see Table 1). Part b) shows that ~ 26 d are needed to reach the maximum of P_T = 114 W/g. In part c) $E_T(t)$ approaches asymptotically to ~ 2.8 MWh/g.

Figure 3-a shows the behavior of the average specific power available at time t for all members of this decay chain. Except for ^{228}Th parent nucleus (exponential decrease with time) the other radioisotopes exhibit a maximum of $P(t)$ at $t \approx 28$ d, whose values have been calculated as ~ 27 W/g for ^{224}Ra, ~ 30 W/g for ^{220}Rn, and ~ 32 W/g for ^{216}Po. From t = 55.4 d on, all $P(t)$ curves become concave upwards, going down to ~ 3 W/g at $t \approx$ 6.3 a. Similar behavior is exhibited by the total specific power, $P_T(t)$, (Fig.

3-b), for which a maximum of ~ 114 W/g occurs at $t \approx 26$ d. Finally, the total, specific energy produced after an elapsed time t (Fig. 3-c) shows an increasing trend from ~ 150 kWh/g after two months to 2.52 MWh/g after ~ 6 a.

5. The ^{227}Ac decay chain: one β^- decay plus four successive α emissions

The radioelement Actinium, known since 1899 (Debierne, 1899), occurs in nature in association with Uranium minerals. The active isotope ^{227}Ac is the great-granddaughter of ^{235}U parent nucleus. Presently, like ^{228}Th the ^{227}Ac isotope has been produced at Oak Ridge National Laboratory-ORNL by neutron irradiation of ^{226}Ra targets (Kukleva *et al*., 2015).

The decay data for the five sequential radioisotopes from ^{227}Ac parent nucleus are reported in Table 2.

Table 2 - Decay data for radioisotopes of ^{227}Ac decay chain*

i	Isotope (decay mode)	Half-life $T_{½}$[a]	$\bar{E}_{\alpha,\beta}$ (MeV)	$\bar{E}_{\gamma}$ (MeV)	$\bar{E}_{i}$** (MeV)	decay const. λ [a^{-1}]
1	^{227}Ac(β^-)	21.772	0.010	0.006	0.016	0.03184
2	^{227}Th(α)	5.12×10^{-2}	5.888	0.137	6.025	13.538
3	^{223}Ra(α)	3.13×10^{-2}	5.669	0.206	5.875	22.145
4	^{219}Rn(α)	1.25×10^{-7}	6.741	0.066	6.807	5.54×10^{6}
5	^{215}Po(α)	5.644×10^{-11}	7.386	0	7.386	1.23×10^{10}
6	^{211}Pb(β^-)	0.69×10^{-4}	0.433[a]	0.597[a]	1.030	1.00×10^{4}

*Data have been taken from (Bé *et al*., 2017; Kondev *et al*., 2021).
**$\bar{E}_i = \bar{E}_{\alpha,\beta} + \bar{E}_{\gamma}$ is the average, total energy available per decay.
[a]Taken from (Singh *et al*., 2013).

Again, by using Eqs. (15)–(17) and the precedent ones (5)–(12), we have obtained the values of $P(t)$ for each radioisotope of the ^{227}Ac "family", the total specific power available, $P_T(t)$, and total specific energy produced

after a time t, E_T(t). Results are presented in Fig. 4, where part a) shows the contributions to $P_T(t)$ due to the four α-emitting nuclides (^{227}Th, ^{223}Ra, ^{219}Rn, and ^{215}Po). The "family patriarch" ^{227}Ac itself does not contribute significantly to total specific power available, since for ^{227}Ac the ratio $\bar{E}_1/T_{½} \approx 0.7\times10^{-3}$ MeV/a is much smaller than the same ratio for any other isotope in Table 2.

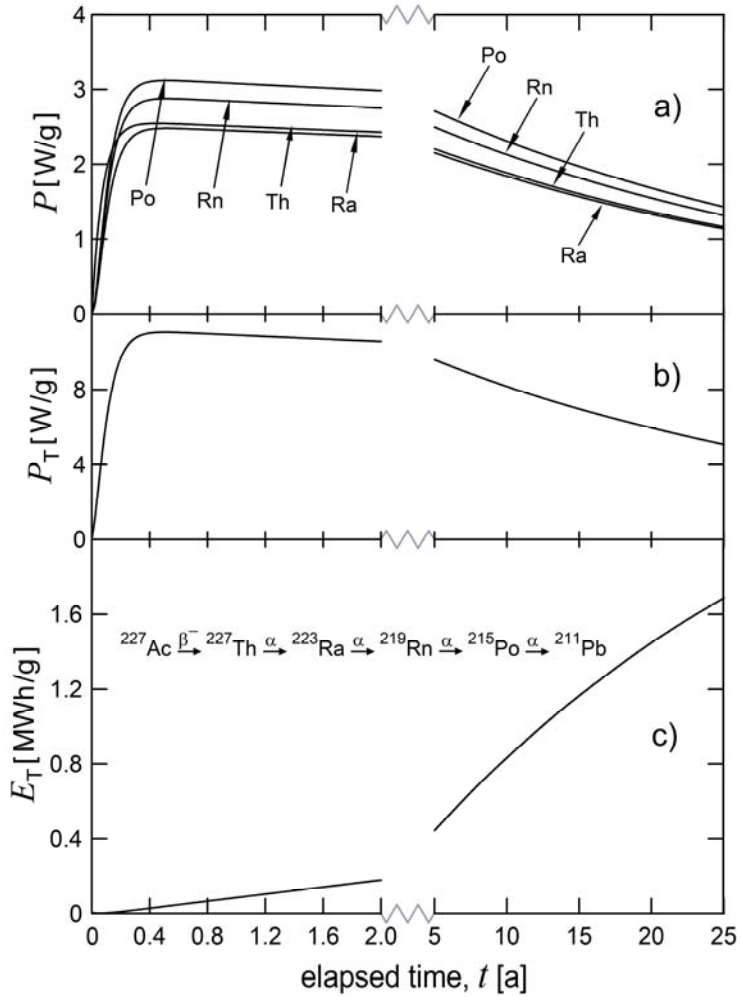

Fig. 4. The same as in Fig. 3 for ^{227}Ac decay chain. Note in Fig. 4 a) the inversion of ^{227}Th and ^{223}Ra for their contributions to average, total specific power, $P_T(t)$. Here the maximum of P_T is 11 W/g which is reached after six months from preparation of a pure ^{227}Ac source (Fig. 4 b)). A total of 1.7 MWh/g is accumulated after an elapsed time of 25 a (Fig.4 c)).

In Fig. 4-a the $P(t)$ curve for ^{227}Th lies above the curve for ^{223}Ra, *i.e.*, these curves appear interchanged in contrast with the positions of ^{227}Th and ^{223}Ra in the ^{227}Ac decay chain. This fact is due to the values of the quantity

$q_i = \bar{E}_i/(T_i \cdot C_{i1})$ (i = 2, 3, 4, 5). Since C_{i1}-values are given by formulas (8)–(11), and for the present ^{227}Ac decay series $T_1 >> T_2 > T_3 >> T_4 >> T_5$, it follows that $q_i = \bar{E}_i / T_1$ (MeV/a). Putting the q-values in descending order one has $q_5 = 0.339$ (^{215}Po), $q_4 = 0.313$ (^{219}Rn), $q_2 = 0.277$ (^{227}Th), $q_3 = 0.270$ (^{223}Ra). The strength of the contributions to total specific power in the present case is dictated by the total energy released per decay of each member of the radioactive "family".

Fig. 4-b shows that the total specific power with time, $P_T(t)$, increases from the initial value of ~ 0.007 W/g up to a maximum of 11 W/g at $t \approx 0.5$ a, then it goes monotonously down to 5 W/g at $t = 25$ a.

The cumulative, total specific energy produced at a time, $E_T(t)$, is depicted in Fig. 4-c). This is an increasing function of t which becomes a curve concave downward from t = 0.5 a onwards, approaching asymptotically the value ~ 3.1 MWh/g (not shown) after being worth 0.18 MWh/g in 2 a, and 1.69 MWh/g in 25 a.

6. The ^{226}Ra decay chain: three successive α emissions, plus two successive β^- emissions, plus one α emission

The radioelement Radium was long ago identified as the granddaughter of the great-granddaughter in the radioactive "family" of natural Uranium (Rutherford and Soddy, 1902). The radioisotope nowadays known as ^{226}Ra was the one discovered in 1898 by P. Curie and M. S. Curie (Curie and Curie, 1898). A quite recent, complete description of advanced methods for preparation, separation, and identification of Radium isotopes has been reported in (Thakur *et al*., 2021).

The decay data for the descendants of ^{226}Ra radioactive "family" are reported in Table 3. These data have been used to evaluate the individual and total specific power at a time t, and the total, cumulative specific energy produced over a time. The results are presented in Fig. 5.

Table 3 - Decay data for radioisotopes of ^{226}Ra decay chain*

i	Isotope (decay mode)	Half-life $T_{½}$[a]	$\bar{E}_{\alpha,\beta}$ (MeV)	$\bar{E}_{\gamma}$ (MeV)	$\bar{E}_{i}$** (MeV)	decay const. λ [a^{-1}]
1	^{226}Ra(α)	1600	4.773	0.011	4.784	4.332×10^{-4}
2	^{222}Rn(α)	1.047×10^{-2}	5.489	0	5.489	66.20
3	^{218}Po(α)	5.888×10^{-6}	6.002	0	6.002	1.177×10^{5}
4	^{214}Pb(β$^-$)	5.095×10^{-5}	0.223	0.300	0.523	1.36×10^{4}
5	^{214}Bi(β$^-$)	3.78×10^{-5}	0.691	1.811	2.502	1.834×10^{4}
6	^{214}Po(α)	5.18×10^{-12}	7.687	0	7.687	1.338×10^{11}
7	^{210}Pb(β$^-$)	22.20	0.007	0.037	0.044	3.12×10^{-2}

*Data have been taken from (Bé *et al*., 2017; Kondev *et al*., 2021).

** $\bar{E}_{i} = \bar{E}_{\alpha,\beta} + \bar{E}_{\gamma}$ is the average, total energy available per decay.

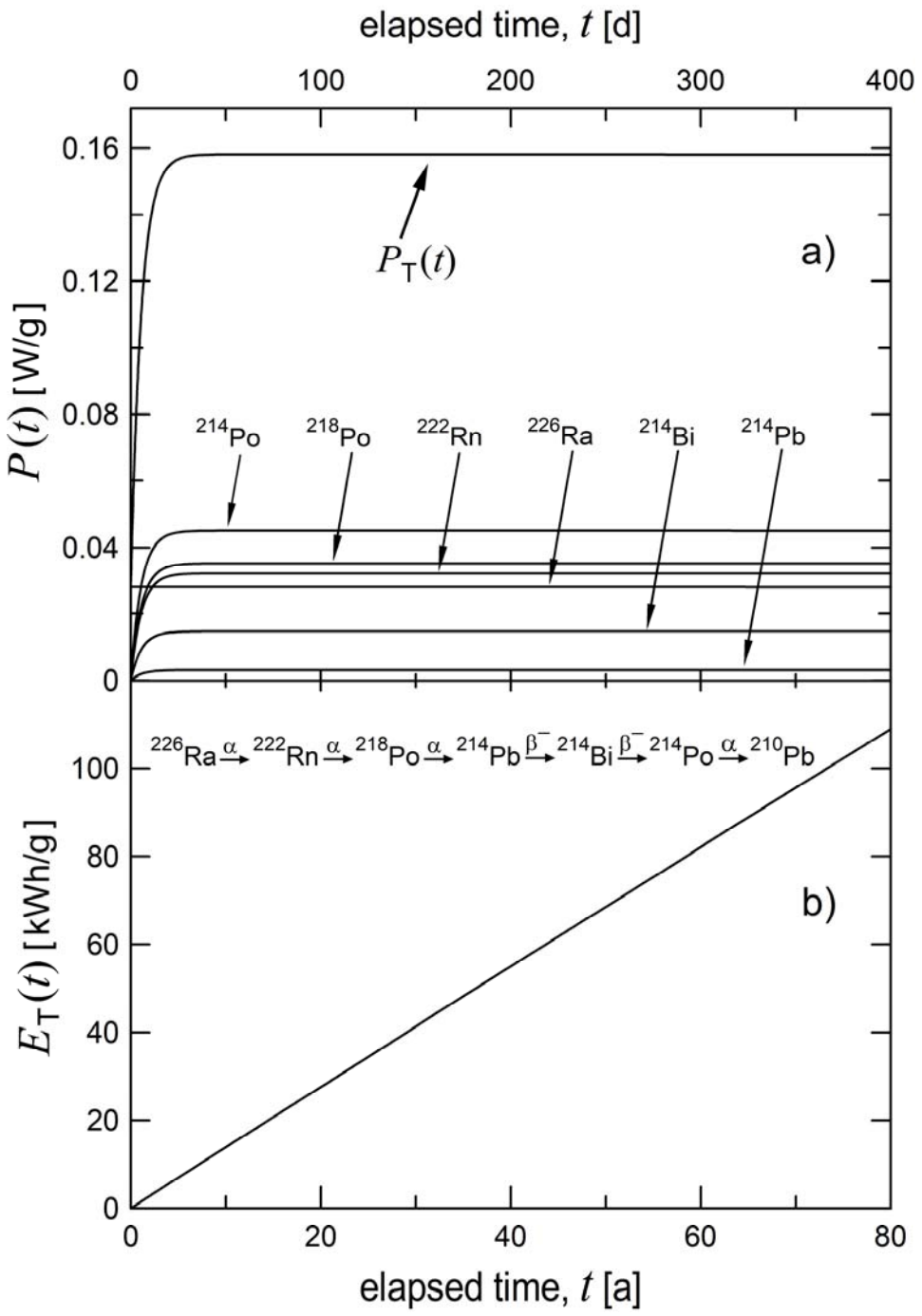

Fig. 5. The same as in precedent figures for ^{226}Ra decay chain. This is the case that makes available less power and energy generated with time. After about three months P_T stabilizes at 0.158 W/g, the main contributions coming (in this order) from ^{218}Po, ^{214}Po, ^{222}Rn, and ^{226}Ra isotopes (Fig. 5 a)). The total, cumulative energy generated, now order of kWh/g, is very approximately proportional to elapsed time in view of the long half-life of ^{226}Ra parent isotope.

Figure 5-a) shows the trend of specific power available at a time t for each member of ^{226}Ra decay "family". The radioisotopes that contribute to

total specific power, $P_T(t)$, do not follow the same sequential order of successive decays starting from ^{226}Ra (see top of Fig. 5 b)). Like in the case of the ^{227}Ac chain, the order of importance for the various radionuclides contributing to $P_T(t)$ is dictated by the ratio $q_i = \bar{E}_i /(T_i \cdot C_{i1})$ (i = 1, 2, …, 6) (see formulas (15) and (8)–(12)). Since $T_1 >> T_i$ (i = 2, 3, …, 6), it follows that $q_i = \bar{E}_i /T_1$. Again, putting these q_i-values (in keV/a) in descending order one has q_6 = 4.8 (^{214}Po), q_3 = 3.7 (^{218}Po), q_2 = 3.4 (^{222}Rn), q_1 = 3.0 (^{226}Ra), q_5 = 1.6 (^{214}Bi), and q_4 = 0.33 (^{214}Pb) (cf. Fig. 5a)). These findings point out that the greater the total energy released per decay, the greater the contribution of the radioisotope to the available total specific power. In addition, calculations have indicated that it takes nearly nine weeks for the total specific power to reach its maximum value of 0.158 W/g, which remains then approximately constant for the next three decades.

The cumulative, total specific energy generated after an elapsed time t (Fig. 5b), upper scale) results quite proportional to t (expressed in a), being described by $E_T(t) \approx 1.38 \times t$ kWh/g, $t \leq 80$ a.

7. Discussion and concluding remarks

The present calculations evidenced that “all” the radioactive members of the decay chains initiated by ^{232}U, ^{228}Th, ^{227}Ac, and ^{226}Ra nuclei, and not only the parent and its daughter radioisotope, remarkably contribute to the available total specific power and to the total, cumulative specific energy released by the primary source.

A critical point regarding the possible use of such valuable isotopes in nuclear batteries is certainly their scarce availability, due to the fact that their production needs complex and expensive processes. However, one would expect that the advantages offered by such radioactive sources will trigger researches towards the development of increasingly efficient nuclear methodologies, able to tackle and solve the present critical issues.

The data reported in Table 4 for ^{232}U and ^{227}Ac, and in Table 5 for ^{228}Th and ^{226}Ra, enable one to better appreciate the contributions to specific power and specific energy arising from radionuclides of the whole decay chains.

In Table 4 and Table 5 one can compare the quantities $P_T(t_M)$ (maximum total specific power at time t_M), and $E_T(t_e)$ (total, cumulative, specific energy produced over an elapsed time t_e), generated by the pair parent/daughter (Mode I) and by all the radioactive members of the decay chain (Mode II).

Table 4 - Values of maximum total specific power available, $P_T(t_M)$ (in W/g), and total, cumulative, specific energy generated at elapsed time t_e, $E_T(t_e)$ (in MWh/g), following two calculation modes for ^{232}U and ^{227}Ac decay chains.[a]

Quantity	^{232}U chain		^{227}Ac chain	
	Mode I	Mode II	Mode I	Mode II
t_M[a]	8.27	9.63	0.45	0.51
$P_T(t_M)$	1.29	3.53	2.55	11.0
t_e[a]	35	35	35	35
$E_T(t_e)$	0.36	0.95	0.48	2.07

[a]Mode I considers only the contributions of the parent and its daughter nuclides; Mode II takes into account all radioactive members of the decay chain.

Table 5 - The same as in Table 4 for ^{228}Th and ^{226}Ra decay chains.[a]

Quantity	^{228}Th chain		^{226}Ra chain	
	Mode I	Mode II	Mode I	Mode II
t_M[d]	24.2	26.4	62.4	64.8
$P_T(t_M)$	52.6	114.3	0.06	0.16
t_e[a]	10	10	10	10
$E_T(t_e)$	1.27	2.76	0.0053	0.0138

[a]Mode I considers only the contributions of the parent and its daughter nuclides; Mode II takes into account all radioactive members of the decay chain.

The most relevant result is the strong increase observed in both the $P_T(t_M)$ and $E_T(t_e)$ values when all the radioactive members of the decay chain are considered. An increase by a factor 2.2 has been found for the ^{228}Th decay chain, by a factor ~2.6, 2.7 in the case of ^{226}Ra and ^{232}U, and by a factor 4.3 in the case of ^{227}Ac decay chain.

As regards the time needed to reach the maximum specific power (t_M), when all the members of the decay chain are taken into account, t_M-values result 4% (^{226}Ra), 9% (^{228}Th), 13% (^{227}Ac), and 16% (^{232}U) higher than when only parent and daughter nuclei are considered. This behavior is partially explained in sections 5 and 6, noting also that in general both the average total energy released per decay, $\bar{E}_i$, and the decay constant, λ_i, increase when one goes from parent and daughter nuclides to granddaughter and great-granddaughter ones.

In short, a little more time to reach the maximum specific power offers the greatest advantage of doubling, tripling or even quadrupling the power available for a variety of applications, *in primis* for fueling nuclear batteries.

The present analysis, based on the physical properties of ^{232}U, ^{228}Th, ^{226}Ra and ^{227}Ac radionuclides, evidenced that for each of them the actual maximum energy that can be produced from the decay chains (1)–(4) is much greater than that expected considering only the parent-to-daughter decay data reported in literature. Moreover, this study has highlighted the complex trends of both specific power and specific, cumulative energy as a function of time. This behavior, due to the overlapping of the different exponential decay rates of the various species originating from an initial radionuclide, makes it possible to adapt the radioactive source to the requirements of any type of nuclear batteries, and to the specific needs in terms of working life.

Beyond evidencing that nuclear batteries powered by ^{232}U, ^{228}Th, ^{226}Ra and ^{227}Ac supply an amount of energy greater than the expected one, the findings of this study can have significant effects also on the technology behind the manufacturing of such devices. Materials and systems utilized to convert the energy released by nuclear decay into electricity are commonly

selected on the basis of type and energy of a given radiation, typically the one emitted by the parent nuclide. Very rarely it is considered that the parent can have a radioactive "progeny" emitting radiations different for type or energy.

Therefore, even if from a physical point of view a parent nuclide that decays producing a series of radioactive nuclides offers the advantage to supply an increased energy, from a technological point of view innovative solutions must be found to address the challenging task of converting the nuclear energy released by more than one type of radiations.

In this context it is hoped that the concepts developed in the present paper could be of some use in driving multidisciplinary efforts to improve the performances of the radiation-based energy sources.

References

Ambrosi R. M., et al. 2019. European Radioisotope Thermoelectric Generators (RTGs) and Radioisotope Heater Units (RHUs) for Space Science and Exploration. Space Sci. Rev. **215**, 55.

Ambrosi, R. M., Kramer, D. P., Watkinson, E. J., Mesalam, R., Barco, A., 2021. A Concept Study on Advanced Radioisotope Solid Solutions and Mixed-Oxide Fuel Forms for Future Space Nuclear Power Systems. Nucl. Tech. **207**, 773.

Artun, O., 2020. Production of energetic ^{232}U, $^{236,\ 238}$Pu, ^{242}Cm, ^{248}Bk, $^{250,\ 252}$Cf and ^{252}Es radioisotopes for use as nuclear battery in thin targets via particle accelerators. App. Radiat. Isot. **166**, 109337.

Ayodele, O. L., Sanusi, K. O., Kahn, M.T.,.2019. Nuclear battery: a source of environmentally friendly energy. J. Eng. Des. Technol. **17**, 172.

Bateman, H., 1910. The Solution of a System of Differential Equations Occurring in the Theory of Radio-active Transformations. Proc. Cambridge Phil. Soc. **15**, 423.

Bé, M.-M, Chisté, V., Dulien, C., et al., 2017. Table of Radionuclides (vol 6 – A = 22 to 242). BIPM Edition, Laboratoire National Henry Becquerel

Bushnell, D. M., Moses, R.W., Choi, S. H., 2021. Frontiers of Space Power and Energy, NASA/TM.0016143.

Cataldo, R.L., Bennett, G.L., 2011. U.S. Space Radioisotope Power Systems and Applications: Past, Present and Future, in Singh., N.(ed), Radioisotopes-Applications in Physical Sciences. London, England: Intech Open; 2011.

Curie, M., 1898. Rayon émis par les composes de l'uranium et du thorium. Compt. Rend. Acad. Sci. **126**, 1101.

Curie, P., Curie, M. S., 1898. On a new radioactive substance contained in Pitchblende. Compt. Rend. **127**, 175

Debierne, A.-L., 1899. Sur un nouvelle matière radio active. Comp. Red. Acad. Sci. **129**, 593.

Dustin, J. S., Borrelli, R. A., 2021. Assessment of alternative radionuclides for use in a radioisotope thermoelectric generator. Nucl. Eng. Des. 385, 111475.

Fry, C., Thoennessen, M., 2013. Discovery of actinium, thorium, protactinium, and uranium isotopes. At. Data Nucl. Data Tables **99**, 345.

Gofman, J. W., Seaborg, G. T., 1949. The Transuranium Elements: Research Papers, Paper 19.14, p. 1427, in: National Nuclear Energy Series IV, 14b, McGraw-Hill, New York

Grigoriev, A., S., Grigoriev S. A., Korolev, A.V., *et al.,* 2019. Small autonomous kW-level power generation based on radioisotope and renewable energy sources for the Arctic zone and the Far East. Atomic Energy. **125**(4), 231.

Hahn, O., 1905. A new radioactive element, which evolves Thorium Emanation. Preliminary Communication. Proc. Roy. Soc. **A76**, 115.

Kaplan I., 1962. Nuclear Physics, second ed. Addison-Wesley, Reading (Mass.) U.S.A., Chap. 10.

Kondev, F. G., et al., 2021. The NUBASE2020 evaluation of nuclear physics properties. Chin. Phys. C **45**, 030001

Kukleva, E., Kozempel, J., Vlk, M., Micolová, P., Volpálka, D., 2015. Preparation of ^{227}Ac/^{223}Ra by neutron irradiation of ^{226}Ra. J. Radioanal. Nucl. Chem. **304**, 263.

Kulikov, G. G., Shmelev, A. N., Apse, V. A., Kulikov, E. G., 2020. Assessment of a possibility to use 232U in radioisotope thermoelectric generators. J. Phys.: Conf. Ser. **1689,** 012035.

Parsonnet, V., Driller, J., Cook, D., Rizvi, S. A., 2006. Thirty-one years of clinical experience with "nuclear-powered"pacemakers. Pace **29**,195.

Prelas, M. A., Weaver, C. L., Watterman, M. L., et al., 2014. A review of nuclear batteries. Prog. Nucl. Energy 75, 117.

Rhodes, J., Maldonado, G. I., 2022. Exploration of producing Uranium-232 for use as a tracer in uranium fuels. Appl. Radiat. Isot. **186**, 110275.

Rutherford, E., Soddy, F., 1902. The cause and nature of radioactivity, Part I. Phil. Mag. **4**, 370.

Schmidt, G. C., 1898. Über die vom thorium und den Thorverbindungens ausgehende Strahlung (About the radiation emitted by thorium and thorium compounds). Ann. Physik **65**, 141.

Singh, B., Abriola, D., Baglin, C., *et al*., 2013. Nuclear Data Sheets for A = 211. Nucl. Data Sheets **114**, 661.

Tavares, O. A. P., Terranova, M. L., 2023. Physical viability for nuclear batteries". J. Rad. Nucl. Chem. **332**(10), 3993.

Terranova, M.L., 2022. Nuclear batteries: Current context and near-term expectations. Int. J. Energy Res. **46**, 19368.

Thakur, P., Ward, A. L., González-Delgado, A. M., 2021. Optimal methods for preparation, separation, and determination of radium isotopes in environmental and biological samples. J. Environ. Radioact. **228**, 106522.

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