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Reuse of Brazilian Mill Tailings Radionuclides as Power Sources

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1. Introduction

Mill tailings in Brazil contain radionuclides of the Uranium-238, Uranium-235 and Thorium-232 decay chains [1]. They are sometimes called "natural series nuclides", composed by the aforementioned radionuclides and their daughter isotopes. In their decay chains, there are some radionuclides that are currently considered as radioactive waste. This work intends to discuss the possibility of reclassifying natural series radionuclides as radioactive resources through the processes of extraction and reuse.

The radionuclides being considered for reuse in this work are Radium-228, Radium-226 and Protactinium-231 and their daughter isotopes, such as Actinium-227, as they have long been investigated as potential substitutes for Plutonium-238 in Radioactive Power Systems (RPS) [2].

While Radium-226 is already used in radiotherapy [3], the other radionuclides have no current widespread use in Brazil. This causes two different issues: the problem of considering those nuclides as radioactive waste that requires management; and the problem of having potential energy sources going to waste. One possible solution could address both issues and also produce a new resource: nuclear batteries.

Over the last sixty years, nuclear batteries have been used both in space missions and on Earth [4]. They are one form RPS, and often are powered by a Radionuclide Thermoelectric Generator (RTG), with Plutonium-238 being the main fuel source. Other radionuclides have been used throughout the years in terrestrial based nuclear batteries, such as Strontium-90 [5].

Still, no attempts have been made with the use of natural series radionuclides.

The main advantage of the RTGs is that they have no moving parts, requiring little to no maintenance [4]. This is almost mandatory for nuclear batteries in space, as there is currently no method of refueling or repairing damage to the systems after they are launched. The drawback is that RTGs have little energetic efficiency, even with the most advanced materials [6].

Other forms of RPS that were considered unsuitable for space but viable on Earth are being studied to access their energetic efficiency, which could enable the use of radionuclides that are still untested in nuclear batteries. Of all proposed alternatives to RTGs, Stirling engines are particularly promising [7].

Terrestrial batteries have some advantages over their space counterparts, mainly the fact that they can be bigger, heavier and have smaller lifetime, as the possibility of refueling their RPS exists. Some batteries based on different RPS are even available for commercial use, such as betavoltaic cells [8].

Alongside alternatives to RTGs, there is also research on new materials that enhance their efficiencies, such as skutterudites [6]. Most research in improving RTGs is made by NASA, as their main focus is on space missions. Recent tests and missions have improved significantly the efficiency of RTGs, achieving values of 7.5% whereas older models had only around 3% [9].

Nonetheless, in order to test the hypothesis of using natural series radionuclides as a reliable RPS fuel source, it is necessary to learn the total electric potential that those nuclides can offer on the entire country. The questions that need to be answered are whether there is enough energy in the mill tailings radioactive waste, for how much time those sources can fuel a battery and if the benefits of reusing those sources are worth the costs.

2. Methodology

The main focus of this work is discussing the energetic potential available at the mill tailings waste. This can only be done thoroughly with an extensive review of the data on uranium and thorium mining in Brazil and their respective radioactive inventories. As most of the data is difficult to obtain, it should take a few years before a full review on the electric potential of the Brazilian mill tailings is finished.

Although obtaining the full data necessary requires a few years, preliminary results can be obtained using the already available data to estimate the electric potential based on the total amount of raw mined U3O8 and Th20 [10].

Obtaining the available thermal energy from the decay of a radionuclide is possible through the use of the compiled tables, such as the ones compiled by the ICRP [11], or calculating the values with equations (1) and (2):

$$\text{Q\_}\alpha = (\text{M\_A} - \text{M\_B} - \text{M\_}\alpha)\text{c}^2 \tag{1}$$

$$\text{Q\_}\beta = (\text{M\_A} - \text{M\_B})\text{c}^2 \tag{2}$$

in which Q is the total decay energy released in MeV, as radionuclide A decays to radionuclide B, M is the atomic mass of the radionuclide in g/mol, Mα is the mass of the helium nucleus in g/mol, and c is the speed of light in MeV/u (931.502 MeV/g/mol).

The specific activity of a radionuclide can be found [12] using equation (3):

$$a = \frac{N_A \ln(2)}{T_{1/2} M} \tag{3}$$

in which a is the radionuclide specific activity in Bq/g, NA is the Avogadro Constant (6.02214086e23 mol-1), T1/2 is the radionuclide half-life in seconds and M is the molar mass of the radionuclide in g/mol.

Since the activity of a radioactive source corresponds to the specific activity of the radionuclide times the mass of the source in grams, equation (3) can be rewritten as:

$$m = \frac{A\, T_{1/2} M}{N_A \ln(2)} \tag{4}$$

in which m is the mass of the radioactive source in grams and A is the activity of the source in Bq.

From the raw mined U3O8, supposing no loss in the separation process, the total amount of uranium-238 can be obtained using the equation:

$$m_{U-238} = \frac{3M_U m_{U3O8}}{\left((3M_U) + (8M_O)\right)} = \frac{714 m_{U3O8}}{842} \tag{5}$$

in which m is the mass, in tons of U3O8 and uranium-238 and M is the molar mass of uranium and oxygen. This can also be done to calculate the total amount of thorium-232 in the Th20 ore mined.

3. Results and Discussion

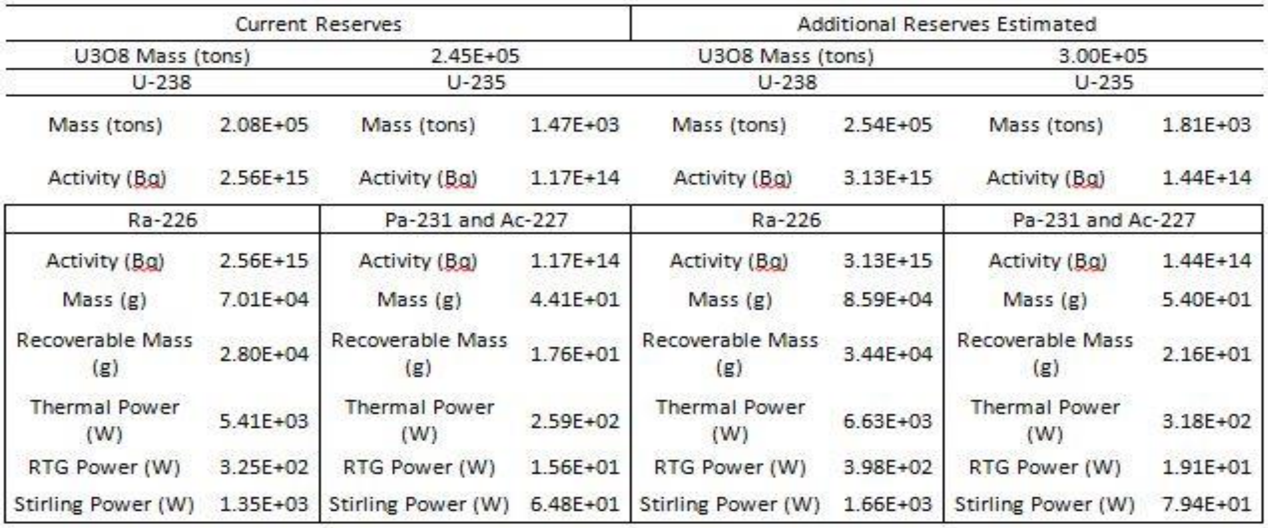
Using the data on raw mined resources and first order approximations, calculation were made in order to access if the investigated radionuclides could provide the required energy to power radionuclide batteries. It is important to note that these estimates and approximations are going to be refined or verified in future calculations in order to obtain the most accurate results as reasonable possible.

Power densities have been obtained using Equations (1) and (2) and are consistent with the listed sources tabulated values[13]. The recovery rate of a radionuclide separated from the radioactive waste was estimated at 40%. Efficiencies in RTG systems and Stirling engines were estimated as 6% and 25% respectively.

These values are nominal efficiencies obtained in experimental tests in laboratory conditions and might not reflect the actual efficiencies on the proper batteries, which is expected to be lower. However, the efficiencies estimated are of older models of RPS, which have increased over the past years. This was done intentionally, in order to obtain a conservative value based on the state of the art technology but without using their nominal efficiencies directly.

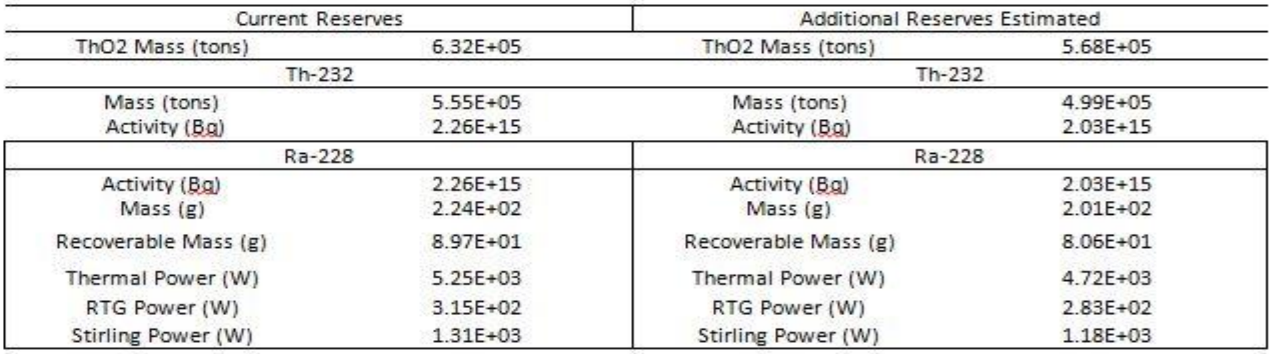
Table 1 summarizes the calculations for the radionuclides that can be found in U_3O_8 and Table 2 does so for radium-228, which can be found in the ThO_2 reserves. Activities and mass were calculated using Equations (3), (4), (5). Thermal Power is calculated simply by multiplying the radionuclide power density with the radionuclide's total available mass.

Table 1 - Available Power in Estimated Uranium Reserves



Current Reserves				Additional Reserves Estimated			
U3O8 Mass (tons)		2.45E+05		U3O8 Mass (tons)		3.00E+05	
U-238		U-235		U-238		U-235	
Mass (tons)	2.08E+05	Mass (tons)	1.47E+03	Mass (tons)	2.54E+05	Mass (tons)	1.81E+03
Activity (Bq)	2.56E+15	Activity (Bq)	1.17E+14	Activity (Bq)	3.13E+15	Activity (Bq)	1.44E+14
Ra-226		Pa-231 and Ac-227		Ra-226		Pa-231 and Ac-227	
Activity (Bq)	2.56E+15	Activity (Bq)	1.17E+14	Activity (Bq)	3.13E+15	Activity (Bq)	1.44E+14
Mass (g)	7.01E+04	Mass (g)	4.41E+01	Mass (g)	8.59E+04	Mass (g)	5.40E+01
Recoverable Mass (g)	2.80E+04	Recoverable Mass (g)	1.76E+01	Recoverable Mass (g)	3.44E+04	Recoverable Mass (g)	2.16E+01
Thermal Power (W)	5.41E+03	Thermal Power (W)	2.59E+02	Thermal Power (W)	6.63E+03	Thermal Power (W)	3.18E+02
RTG Power (W)	3.25E+02	RTG Power (W)	1.56E+01	RTG Power (W)	3.98E+02	RTG Power (W)	1.91E+01
Stirling Power (W)	1.35E+03	Stirling Power (W)	6.48E+01	Stirling Power (W)	1.66E+03	Stirling Power (W)	7.94E+01

Table 2 - Available Power in Estimated Thorium Reserves



Current Reserves		Additional Reserves Estimated	
ThO2 Mass (tons)	6.32E+05	ThO2 Mass (tons)	5.68E+05
Th-232		Th-232	
Mass (tons)	5.55E+05	Mass (tons)	4.99E+05
Activity (Bq)	2.26E+15	Activity (Bq)	2.03E+15
Ra-228		Ra-228	
Activity (Bq)	2.26E+15	Activity (Bq)	2.03E+15
Mass (g)	2.24E+02	Mass (g)	2.01E+02
Recoverable Mass (g)	8.97E+01	Recoverable Mass (g)	8.06E+01
Thermal Power (W)	5.25E+03	Thermal Power (W)	4.72E+03
RTG Power (W)	3.15E+02	RTG Power (W)	2.83E+02
Stirling Power (W)	1.31E+03	Stirling Power (W)	1.18E+03

Considering the possibility of using a multiple isotope fuel, the total amount of power available using those resources alone would be approximately 1.4 kW for the RTG and 5.7 kW for the Stirling engine, indicating the possibility of using those radionuclides as the power sources for the batteries. Further investigation along the development of this work should assess the consistence of the power and performance in the following years.

There is even more available fuel, considering the resources that have already been mined and that can be found in mining companies reports [14] and the unexplored mineable uranium and thorium.

The battery fuel cycle would also be mostly reusable, since the waste product of the battery fuel itself would be the long lived Radium-226, which already is used in radiotherapy. With the exception of a few of the materials and shielding used, almost all of the radioactive sources would have a use in such a setting.

4. Conclusions

Radionuclides that are currently considered radioactive waste have the potential to grow into radioactive resources with the proper management and applications. The early estimates presented in this work highlight the possibility of using the natural series radionuclides in RPS and further reuse them in radiotherapy. The first step towards answering the questions about the reuse of those radionuclides is identifying their total available amount and the amount that is going to be available in the near future.

There already is ongoing research on the subject, and the preliminary results are encouraging. The complete assessment of the natural series radionuclides is necessary in order to answer to remaining questions. Once the total available power has been fully estimated, it is possible to calculate the lifetime and behavior of the energy in the nuclear batteries, which in turn allows for the possibility of estimating the costs and benefits of reusing those resources.

The reuse of radioactive waste can also reduce costs in the management, with the new technologies involved in the entire chain of activities leading to new business opportunities, while simultaneously creating a safer environment and increasing the awareness about practical green uses of nuclear technology.

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