Assessment of a possibility to use $^{232}$U in radioisotope thermoelectric generators

G G Kulikov, A N Shmelev, V A Apse and E G Kulikov

Institute of Nuclear Physics and Engineering, National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe shosse, 31, Moscow, 115409, Russian Federation

GGKulikov@mephi.ru

Abstract. The paper analyzes advantages and drawbacks of the radioisotope thermoelectric generators (RTG) based on application of $^{238}$Pu and $^{232}$U. Currently, the RTG on $^{238}$Pu are widely used for long-term autonomous power supply of spaceships due to the following positive properties of $^{238}$Pu: high specific heat generation rate, long half-life, weak emission of neutrons and $\gamma$-rays. Isotope $^{238}$Pu may be produced by neutron irradiation of $^{237}$Np (main component of transuranium radioactive wastes) in nuclear reactors. The paper considers application of $^{232}$U for the same purpose because of the following positive properties of $^{232}$U: power elevation during initial 10 years of the RTG operation (effect of $\alpha$ and $\beta$-emitters in $^{232}$U decay chain), long half-life (comparable with $^{238}$Pu half-life). Unfortunately, $^{232}$U decay chain includes also intense emitters of high-energy $\gamma$-rays. As a compromise, a possibility is considered to create the RTG based on mixture of $^{238}$Pu with small (below 5%) additive of $^{232}$U. Such RTG will be able to keep long-term stable operation at high power level. The following two-step technology is proposed to produce $^{232}$U: generation of $^{231}$Pa in thorium blanket of hybrid thermonuclear reactors and subsequent neutron irradiation of $^{231}$Pa in nuclear reactors till accumulation of significant $^{232}$U quantities.

1. Introduction

As is known, the RTG are being used to provide long-term autonomous energy supply of the devices installed in remote regions and in spaceships. The relevant characteristics of some radioisotopes, which can be used in the RTG, are presented in table 1. The chosen radioisotopes are constituents of fission products, minor actinides and fissionable nuclides with half-life longer than 10 years and with intense heat generation [1].

Table 1. Characteristics of nuclides, which are candidates for the RTG.

| Nuclide   | $T_{1/2}$ (years) | Total energy of $\alpha$, $\beta$, $\gamma$-radiation of nuclides and their decay products (MeV) | High-energy $\gamma$-rays (MeV) |
|-----------|------------------|-------------------------------------------------------------------------------------------------|-------------------------------|
| $^{90}$Sr | 29               | 2.8                                                                                             | No                            |
| $^{137}$Cs| 30               | 1.2                                                                                             | 0.7                           |
| $^{241}$Am| 433              | 5.5                                                                                             | No                            |
| $^{244}$Cm| 18               | 5.8                                                                                             | No                            |
| $^{238}$Pu| 88               | 5.5                                                                                             | No                            |
| $^{232}$U | 69               | 5.3 ($^{232}$U) + 36.9 (decay products) = 42.2                                                 | 0.6; 2.6 ($^{208}$Tl)         |
At present, isotope $^{238}\text{Pu}$ is mainly used as a heat source in the RTG because, firstly, $^{238}\text{Pu}$ can provide slow relaxation of heat generation rate (figure 1), and, secondly, decay chain of $^{238}\text{Pu}$ does not include sources of high-energy $\gamma$-rays.

![Figure 1. Time dependence of the RTG power based on different radioisotopes.](image)

Half-lives of $^{90}\text{Sr}$ (29 years), $^{137}\text{Cs}$ (30 years) and $^{244}\text{Cm}$ (18 years) are substantially shorter than that of $^{238}\text{Pu}$ (88 years). As a result, more rapid relaxation of heat generation rate will take place in the RTG based on application of these radioisotopes. In addition, decay chain of $^{137}\text{Cs}$ includes some isotopes – emitters of high-energy $\gamma$-rays. If initial nuclei quantities of all these radioisotopes are identical, then initial heat generation rates of $^{90}\text{Sr}$, $^{137}\text{Cs}$ and $^{238}\text{Pu}$ are comparable each other in values while initial heat generation rate of $^{244}\text{Cm}$ is 5 times higher. Thus, $^{244}\text{Cm}$ is the most intense heat source but rather short time of stability. Half-life of $^{244}\text{Cm}$ is about 5 times shorter than that of $^{238}\text{Pu}$. Consequently, heat generation rate of $^{238}\text{Pu}$ is 5 times lower but, on the other hand, $^{238}\text{Pu}$ can provide the longer time period of stable RTG operation. Half-life of $^{241}\text{Am}$ is 5 times longer than that of $^{238}\text{Pu}$. Consequently, heat generation rate of $^{238}\text{Pu}$ is 5 times higher but, on the other hand, $^{238}\text{Pu}$ is not able to provide comparable time period of stable RTG operation. So, there is a possibility to choose radioisotope with proper accounting for heat generation rate and duration of stable RTG operation (the more intense heat generation rate, the shorter time of stable heat generation).

2. **Radioisotope thermoelectric generators and heat units on $^{238}\text{Pu}$**

Papers [2-6] analyse current state, application areas and prospects for the use of the RTG for the outer space investigations. As is noted, $^{238}\text{Pu}$ was chosen for these studies because of high specific heat generation rate (570 W/kg), long half-life (~88 years), low intensity of neutron and $\gamma$-emission. Low level of ionizing radiation makes it possible to fabricate the RTG without bulky radiation shielding, develop small-size and light design. As is known, radioisotope $^{238}\text{Pu}$ can be produced by neutron irradiation of $^{237}\text{Np}$ (main component of transuranium radioactive wastes) in nuclear reactors.

Besides the RTG, some miniature radioisotope heat units on $^{238}\text{Pu}$ have been widely used in the USA space projects. These heat units (thermal power ~1 W) were named LW RHU (Light Weight Radioisotope Heat Units). Small-size (25 mm in diameter, 33 mm in height), low weight (~40 g) RHU have been used to keep nominal temperature conditions in spaceships modules, where research devices
were installed, at extremely negative temperatures of the environment. These conditions are typical for
the outer space investigations because night temperature of the environment can fall down below
minus 100°C. All the USA space projects have applied RHU as heat sources.

The USSR experience in space applications of the RTG was substantially lower than the USA
experience. This fact may be explained by the chosen strategy to use mainly nuclear power facilities
(NPF) on spaceships. Major role has been given to the NPF based on nuclear reactors with direct
(thermolectric or thermos-ionic) conversion of thermal energy to electrical one.

The first spaceships with the RTG have been launched by the USSR in September 1965. Two RTG
(Orion-1 and Orion-2) on 210Po have been put onto near-earth orbits. Isotope 210Po (specific heat
generation rate 0.141 W/g, half-life 138 days) allowed spaceship designers to make small-size RTG.
The RTG lifetime was defined by 210Po half-life and equaled to about 3000 hours. The 210Po-based
RTG have been used in the Moon research vehicles Lunokhod-1 (1969) and Lunokhod-2 (1971) to
keep acceptable temperature conditions in the vehicles.

In the middle of 1970s technological base has been formed in the USSR for full-scale production of
238Pu. Afterwards, 238Pu-based RTG were fabricated only for terrestrial applications (for example, as
energy sources of cardio accelerators after their implantation into human body). By the end of 1990s
the rationality of RTG applications for power supply of spaceships has appeared again. In
particular, RTG have been planned to use for multi-year power supply of the research spaceships
(small autonomous space stations) to be landed on the Mars under the International Project Mars-96.
The RTG must perform the following missions: maintenance of nominal temperature conditions for
reliable operation of the research devices; maintenance of the research devices operation in the duty-
regime; recharging of storage batteries after communications with orbital spaceships.

The RTG design presumes that radiogenic helium (decay product of 238Pu) must be removed from
PuO2 ampoule to a special hermetic cavity and, then, out of the RTG case. Thermal energy is
transferred from surface of radioisotope heat assembly to thermoelectric generator by radiative
transport. Spent heat is removed to the environment through the RTG case.

So, 238Pu-based RTG will be demanded for future long-term planetary and outer space
investigations. As to radioisotope heat units, no practical problems do exist with their any applications.
Reliable and safe operation of RHU is provided by their design features. Low thermal power and small
238Pu mass (below 15 g) create no serious difficulties for safe RHU operation and, if necessary, for
effective heat removal. That is why 238Pu-based RTG and RHU will be used as dominant energy and
heat sources in the nearest future for cosmic investigations of planets and small bodies of the Solar
system.

The Russian Federation and the USA are the largest producers and consumers of radioisotope
238Pu. In 1992 the US Department of Energy has concluded 5-year agreement on the purchase of 238Pu
in Russia at amount of 10 kg with potential extension up to 40 kg. Since 1993, majority of American
spaceships have applied RTG on 238Pu purchased in Russia. In 2009 238Pu deliveries have been broken
off because of reformation of Russian nuclear industry. In total, the Russian Federation has delivered
16 kg 238Pu to the USA in 1993-2009 (about 1 kg per year, in average). In 2009 the US Department of
Energy has inquired funding for renewal of 238Pu production in the USA. In 2013 Oak Ridge National
Laboratory has launched production of 238Pu. Currently, 238Pu production rate in the USA equals to 0.4
kg per year and may be increased up to 1.5 kg per year by 2025.

So, it may be concluded that multi-year experience in the usage of 238Pu-based RTG has
demonstrated that they are the most reliable autonomous sources of thermal and electrical energy for
space investigations. Currently, thermal efficiency of the RTG for spaceships equals to 10-11% and
may be upgraded up to 13-15% by application of new advanced thermoelectric materials. Typical
lifetimes of the RTG and RHU cover the range from 10 to 30 years.

3. Specific peculiarities and advantages of radioisotope 232U
Radioisotope 232U differs profitably from radioisotope 238Pu (table 1) because, at comparable half-life
(~71 years with accounting for 228Th, decay product of 232U), the larger heat amount (by several times
higher) is released in one decay of $^{232}\text{U}$ than that in one decay of $^{238}\text{Pu}$. Such remarkable difference is caused by the presence of five $\alpha$-emitters and three $\beta$-emitters in decay chain of $^{232}\text{U}$ (figure 2) [1].

![Decay chain of $^{232}\text{U}$](image)

**Figure 2.** Decay chain of $^{232}\text{U}$.

Isotope $^{238}\text{Pu}$ decays to $^{234}\text{U}$ (half-life $2.45 \cdot 10^5$ years), i.e. no additional contribution to heat amount will be given. Similar situations take place with isotopes $^{241}\text{Am}$ and $^{244}\text{Cm}$. Isotope $^{241}\text{Am}$ decays to $^{237}\text{Np}$ (half-life $2.1 \cdot 10^6$ years), $^{244}\text{Cm}$ decays to $^{240}\text{Pu}$ (half-life $6.5 \cdot 10^3$ years). Consequently, those decay chains for which $^{238}\text{Pu}$, $^{241}\text{Am}$ and $^{244}\text{Cm}$ are the start isotopes, after burn-up of their start isotopes, remain radioactive chains but cease heat-producing chains.

Isotope $^{232}\text{U}$ is a unique radioisotope which is able to generate significantly larger heat amount than $^{238}\text{Pu}$ can during comparable with $^{238}\text{Pu}$ time period. This effect can be achieved thanks to the longer decay chain. Final isotope of $^{232}\text{U}$ decay chain is a stable nuclide $^{208}\text{Pb}$. Thus, $^{232}\text{U}$ can offer the following two advantages in comparison with $^{238}\text{Pu}$, namely the larger heat generation rate thanks to the presence of $\alpha$ and $\beta$-emitters in its decay chain, and relatively short-term conversion into stable lead isotope.

4. Advantages of $^{232}\text{U}$-based RTG and RHU

One specific peculiarity of $^{232}\text{U}$-based RTG consists in the fact that its power reaches the largest value (substantially higher than the largest power of $^{238}\text{Pu}$-based RTG) only after establishing the equilibrium state in $^{232}\text{U}$ decay chain. The equilibrium state establishes in about 10 years (~5 half-lives of $^{228}\text{Th}$, the longest-lived decay product) after the RTG manufacturing. This peculiarity may be regarded as a positive property because the RTG power increases during initial 10 years and remains within 15%-vicinity of maximal value during 20 years. Power of the RTG based on application of other radioisotopes decreases exponentially with time (figure 3).
It is naturally to expect that mixture of $^{238}$Pu (heat generation rate decreases with time) and $^{232}$U (heat generation rate increases during initial 10 years) can provide stable in time heat generation by the RTG. Deviations of the RTG power from maximal value are presented in table 2 for $^{238}$Pu-based RTG and for ($^{238}$Pu, $^{232}$U)-based RTG as a function of $^{232}$U admixture.

Table 2. Deviations from maximal power of $^{238}$Pu-based RTG ($\delta_1$) and ($^{238}$Pu, $^{232}$U)-based RTG ($\delta_2$).

| RTG operation time (years) | $\delta_1^{(238\text{Pu})}$ (%) | $\delta_2^{(238\text{Pu}, \ 232\text{U})}$ (%) | Effect of $^{232}$U admixture, ($\delta_1-\delta_2$)/$\delta_1$ (%) | $^{232}$U admixture (%) |
|---------------------------|-------------------------------|---------------------------------|-------------------------------------------------|-------------------|
| 10                        | 7.6                           | 3.1                             | 59                                              | 1.0               |
| 15                        | 11.2                          | 6.1                             | 46                                              | 1.5               |
| 20                        | 14.6                          | 9.3                             | 36                                              | 2.0               |
| 30                        | 21.1                          | 16.1                            | 24                                              | 3.2               |
| 40                        | 27.1                          | 22.8                            | 16                                              | 4.2               |
| 50                        | 32.6                          | 29.1                            | 11                                              | 3.2               |

As is seen, $^{232}$U admixture to $^{238}$Pu gives to ($^{238}$Pu, $^{232}$U) mixture property of stable heat generation. Time dependence of the heat generation represents a cupola-shaped form with maximal value in the center. Small deviations $\delta_1$ from maximal value is reached by low $^{232}$U admixtures (only some percent). The shorter RTG lifetime requires the lower $^{232}$U admixture, and this admixture provides the more stable heat generation rate in time.

Another important property of $^{232}$U admixture consists in its larger heat generation potential than that of $^{238}$Pu. Excess heat amounts may be thrown down to the environment. $^{232}$U-based RTG is able to provide stable energy production within 20 years at the power level 8 times higher than that of $^{238}$Pu-based RTG (or within 45 years at the power level 6 times higher than that of $^{238}$Pu-based RTG).

5. Problems of $^{232}$U-based RTG and RHU
The main problem that arises at $^{232}$U application as a heat source is related with high-energy (0.6 MeV and 2.6 MeV) $\gamma$-rays emitted by $^{232}$U decay products. The problem can be resolved by surrounding the RTG with radiation shielding. Thicknesses of lead shielding as dependence on the multiplicity factor of the required attenuation are presented in table 3 [1].
Table 3. Thickness of lead shielding as dependence on the multiplicity factor of high-energy (2.6 MeV) $\gamma$-ray attenuation.

| Multiplicity of attenuation | Thickness of lead shielding (cm) |
|-----------------------------|----------------------------------|
| 10                          | 6.2                              |
| 100                         | 11.8                             |
| 1000                        | 17.3                             |
| $10^4$                      | 22.5                             |
| $10^5$                      | 27.6                             |

It can be seen that $\gamma$-ray attenuation by $10^5$ times requires as thick lead shielding as 27.6 cm. Such a thickness is not very large payment for substantial elevation of the RTG power at replacement of $^{238}$Pu by $^{232}$U.

As is known, the higher atomic number of the shielding material results in the larger attenuation factor of $\gamma$-rays. Indeed, the atomic number defines charge of nucleus and number of electrons in atom. Interactions of $\gamma$-rays with electrons can slow down $\gamma$-rays through photo-effect, Compton-effect and generation of electron-positron pairs. For example, replacement of lead (atomic number 82) by iron (atomic number 26) results in two-fold thickening of radiation shielding at the same attenuation factor [1]. So, if $^{232}$U is diluted by natural uranium (atomic number 92) or natural thorium (atomic number 90), then these heavy materials can provide internal attenuation of high-energy $\gamma$-rays better than external lead shielding.

6. $^{232}$U production and marketing

It seems reasonable to use the following two-step technology for production of isotopically clean $^{232}$U: accumulation of $^{231}$Pa in thorium blanket of hybrid thermonuclear reactor [7-10], and, afterwards, production of $^{232}$U by neutron irradiation of $^{231}$Pa in thermal nuclear reactors. The world annual demands for $^{232}$U are evaluated as several kilograms.

As technology of $^{238}$Pu production through neutron irradiation of $^{233}$Np in nuclear reactors has been already mastered, then cost of $^{232}$U production will be evidently larger than cost of $^{238}$Pu production, at least, at initial stages. Under these conditions it seems reasonable to use $^{232}$U only as a small admixture to $^{238}$Pu for stabilization of heat generation rate. Then, $^{232}$U may be used as a self-dependent isotope in fabrication of the RTG and RHU with substantially higher power. Consequently, according to the problem significance, it seems reasonable to arrange production of strategic $^{231}$Pa stockpiles for further, if necessary, conversion of $^{231}$Pa to $^{232}$U.

The State Corporation Rosatom, the S.P. Korolev Rocket and Space Corporation Energia and some foreign organizations may be potential consumers of $^{232}$U-based RTG and RHU.

7. Conclusion

The paper proposes one out-of-pile application of isotope $^{232}$U as a heat-generating material in the RTG and RHU designs as autonomous energy sources in remote regions and spaceships. Isotope $^{232}$U may be also used as a small admixture to $^{238}$Pu for stabilization of energy generation, or self-dependently as a more intense source of thermal energy.

Acknowledgments

The reported study was funded by RFBR, project number 19-29-02006.

References

[1] Babichev A P et al 1991 Physical values. Reference book (Moscow: Energoatomizdat) p 1232
[2] Pustovalov A A, Gusev V V, Pankin M I and Smetannikov V P 2005 Current state, application areas and prospects for future use of radioisotope thermoelectric generators (RTG) on plutonium-238 for planetary investigations of the Solar system Proceedings of the
International Conference Nuclear power in Space 2005 (Moscow)

[3] Richard M A et al 2019 European Radioisotope Thermoelectric Generators (RTGs) and Radioisotope Heater Units (RHUs) for Space Science and Exploration *SPACE SCIENCE REVIEWS* **215**(8) 55

[4] Xiawa W, Renrong L, Peter F, Walker C and Jun X 2020 Critical design features of thermal-based radioisotope generators: A review of the power solution for polar regions and space *Renewable and Sustainable Energy Reviews* **119** 109572

[5] Nesrine J, Ayda B, Jens M, Brahim M, Fares T and Mohammed I 2019 A comprehensive review of Thermoelectric Generators: Technologies and common applications *Energy Reports* In Press

[6] Alessandra B, Richard M A, Hugo R W and Keith S 2020 Radioisotope power systems in space missions: Overview of the safety aspects and recommendations for the European safety case *Journal of Space Safety Engineering* **7**(2) 137-149

[7] Baatar T and Kulikov E G 2020 Justification of vver-1000 safety when using fuel compositions doped by protactinium and neptunium *Izv. Wysshikh Uchebnikh Zawedeniy, Yad. Energ.* **1** 26–36

[8] Kulikov G G, Shmelev A N, Kulikov E G and Apse V A 2020 Proliferation-protected, ultra-high burn-up reactor fuel produced in the thorium blanket of a fusion neutron source *GLOBAL 2019 - International Nuclear Fuel Cycle Conference and TOP FUEL 2019 - Light Water Reactor Fuel Performance Conference* pp 1088–92

[9] Kulikov G G, Shmelev A N, Geraskin N I, Kulikov E G and Apse V A 2016 Advanced Nuclear Fuel Cycle for the RF Using Actinides Breeding in Thorium Blankets of Fusion Neutron Source *Nuclear Energy and Technology* **2** 147-150

[10] Shmelev A N and Kulikov G G 2016 On the Role of Fusion Neutron Source with Thorium Blanket in Forming the Nuclide Composition of the Nuclear Fuel Cycle of the Russian Federation *Physics of Atomic Nuclei* **79** 1508-1512