Use of Am-241 in RTGs

A V Mikhailov, D O Chernov and V V Korobeinikov
Obninsk Institute for Nuclear Power Engineering
mikhalev.alexandr2017@yandex.ru

Abstract. Currently, transuranic elements are a serious problem. They are formed in spent nuclear fuel as a result of the operation of reactors. These elements have a sufficiently high radioactivity and a huge half-life, thus they remain dangerous for a very long time and must be burned out on specialized reactors. However, not all of them can be classified exclusively as waste. Am-241, which has a low heat release due to its alpha decay, can be used as a radionuclide source for RTGs. However, this nuclide can maintain a fission chain reaction and emits gamma rays as a result of decay. The article investigates some properties of possible devices, as well as an assessment of the radiation protection for such devices.

1. Introduction
In the modern world, autonomous power sources have a great interest. This is primarily due to large-scale space programs developing in various countries, including the United States of America and Russia, involving the exploration of Jupiter, Mars, Saturn, Venus, the Moon and other objects. For the realization of such goals, various devices are used, for example, space nuclear power plants, with a capacity of up to tens of MW [1]. However, the most widely used today are radioisotope thermoelectric generators (RTGs). In 2011, the Curiosity rover was commissioned with a multipurpose radioisotope thermoelectric generator on board, the power of which is about 125 Wte [2]. Pu-238 is an a radioisotope source for it. Due to alpha decays, Pu-238 emits about 560 Wt/kg. It is quite efficient in comparison with other RTG’s materials. In addition, such devices does not require serious protection against ionizing radiation due to the absence of beta and gamma radiation. Recently, the production of plutonium 238 has been suspended in the United States and in the world, which makes it necessary to look for alternative sources for generators.

The strontium-90 isotope is still actively used in RTGs for terrestrial applications. The list of installations using this isotope is presented in table 1 [3,4].

Table 1 (a). Parameters of RTG with Sr-90.

| Type of unit | Initial activity, kCi | Thermal power, Wt | Electric power, Wte | Efficiency, % | Weight, kg | Operation start year |
|--------------|-----------------------|-------------------|---------------------|---------------|------------|---------------------|
| Efir-MA      | 104                   | 720               | 30                  | 4.167         | 1250       | 1976                |
| IEU-1        | 465                   | 2200              | 80                  | 3.64          | 2500       | 1976                |
Table 1 (b). Parameters of RTG with Sr-90.

| Type of unit | Initial activity, kCi | Thermal power, Wt | Electric power, Wte | Efficiency, % | Weight, kg | Operation start year |
|--------------|-----------------------|------------------|-------------------|--------------|-----------|---------------------|
| IEU-2        | 100                   | 580              | 14                | 2.41         | 600       | 1977                |
| Gong         | 47                    | 315              | 18                | 5.714        | 600       | 1983                |
| Gorn         | 185                   | 1100             | 60                | 5.455        | 1050      | 1983                |
| IEU-2M       | 116                   | 690              | 20                | 2.899        | 600       | 1985                |
| Senostav     | 288                   | 1870             | -                 | -            | 1250      | 1989                |
| IEU-1M       | 340                   | 2200             | 120               | 5.455        | 2100      | 1990                |

However, in order to ensure the safety of the environment, such devices require significant dimensions of protection against ionizing radiation, which prevents its use in spacecraft. Under conditions of use on Earth, such settings are not optimal. In particular, in Russia, due to the inadequacy of measures to ensure the nonproliferation of nuclear materials and the protection of radioactive sources from external influences, situations arose with the theft of devices, their depressurization and exposure of the population [5]. In particular, due to an emergency in 1987, an RTG based on strontium-90 was discharged into the water. The source activity for 2014 is 8.9 PBq [6]. Despite the absence of pollution, such accidents pose a huge danger and require increased attention to nuclear safety. Today, a few problems and accidents can be avoided, since much more stringent requirements are imposed on the location and protection of such installations.

2. Possibilities of using Am-241
A possible alternative to the previously used nuclides is Am-241 [7]. Nowadays americium is considered as waste in the spent fuel from thermal and fast nuclear reactors. Therefore, the scenario of burning americium and other minor actinides is being actively considered today. Fast reactors such as the French ASTRID [8] plant are often used for this purpose. Fast reactors also make it possible to completely load the core with americium, which will make it possible to burn off a nuclide in large volumes and simultaneously obtain electricity [9]. Another option is subcritical nuclear plants with an external neutron source, since they have a large excess of high energy neutrons[10]. Also for this purpose can be considered the most common reactors - reactors on thermal neutrons [11]. However it has several useful characteristics, such as a specific heat release of about 0.1 Wt/g and a half-life of 432.6 years. This allows us to consider it as a radioisotope source for generating electricity. In our work we consider two types of Am-241 oxides as a radioisotope: Am₂O₃ and AmO₂ [12,13]. Both have melting point of more than 1500 °C to ensure the required level of safety, in contrast to metallic americium, for which this parameter is 1173 °C [14].

2.1. Device power
The maximum capacity of a facility with Am-241 as a radioisotope source is limited due to the ability of americium to maintain a fission chain reaction. With this in mind, to determine the power, it is necessary to determine the maximum mass of americium oxide that can be loaded into one unit. The permissible mass of americium oxide is the mass at which the effective neutron multiplication factor does not exceed 0.95. These calculations are carried out in the program based on the Monte Carlo method. In the calculations, two types of models are considered, where, in the first case, a sphere of americium oxide is surrounded by a reflector, and the second without a reflector. For illustrative purpose the models are shown in the figures 1 and 2.
Graphite was used as a reflector in the first model. The calculation for two types of models is due to the need to take into account the requirements of nuclear and radiation safety for various scenarios of using the facility. Each model takes into account one of the safety characteristics more conservatively.

The first model surrounds americium oxide with a quality reflector. This model conservatively takes into account the presence of any materials that can be used in the installation (housing materials, radiation protection, environmental objects). However, under real conditions the facility will not be surrounded by a thick layer of high quality reflector. As a result, the mass of americium oxide can be increased, and, as a consequence, the activity of the installation and the radiation load. To determine the radiation protection, we took as a basis model 2 without a reflector. Thus, we can conservatively determine the maximum mass of radioactive material that can be used in absolutely any scenario. The result of calculating is shown in tables 2 and 3.

**Figure 1.** Model with reflector.

**Figure 2.** Model without reflector.
Table 2. The result of calculating the parameters for the first model.

| Material   | $\text{Am}_2\text{O}_3$ | $\text{AmO}_2$ | $\text{AmO}_2$ |
|------------|-------------------------|---------------|---------------|
| Sphere radius, cm | 10.35 | 10.06 | 10.04 |
| Nuclear concentration $10^{-24}$ $1/\text{cm}^3$ | 0.026747 | 0.02576 | 0.02801 |
| Density, g/$\text{cm}^3$ | 11.77 | 11.68 | 12.70 |
| Americium mass, kg | 49.71 | 43.97 | 47.53 |

Table 3. The result of calculating the parameters for the second model.

| Material   | $\text{Am}_2\text{O}_3$ | $\text{AmO}_2$ | $\text{AmO}_2$ |
|------------|-------------------------|---------------|---------------|
| Sphere radius, cm | 12.28 | 12.75 | 11.74 |
| Nuclear concentration $10^{-24}$ $1/\text{cm}^3$ | 0.026747 | 0.02576 | 0.02801 |
| Density, g/$\text{cm}^3$ | 11.77 | 11.68 | 12.70 |
| Americium mass, kg | 83.03 | 89.52 | 75.99 |

For the calculation, we used two versions of $\text{AmO}_2$, since the density of the material is noticeably different in different literature, which directly affects its fissile properties. It should be noted that the maximum possible weight of just the fuel composition of the RTG is large enough and limits the possibility of using such a device. Based on the calculated characteristics, the maximum power was determined for each source option. The result is shown in tables 4 and 5.

Table 4. Maximum power of the device according to model 1.

| Material   | $\text{Am}_2\text{O}_3$ | $\text{AmO}_2$ | $\text{AmO}_2$ |
|------------|-------------------------|---------------|---------------|
| Density, g/$\text{cm}^3$ | 11.77 | 11.68 | 12.7 |
| Thermal power, kWt | 5.27 | 4.66 | 5.04 |
| Electrical power, kWte | 395.21 | 349.58 | 377.85 |
| Electrical power at the end of operational life, Wte | 355.69 | 314.62 | 340.06 |

Table 5. Maximum power of the device according to model 1.

| Material   | $\text{Am}_2\text{O}_3$ | $\text{AmO}_2$ | $\text{AmO}_2$ |
|------------|-------------------------|---------------|---------------|
| Density, g/$\text{cm}^3$ | 11.77 | 11.68 | 12.7 |
| Thermal power, kWt | 8.80 | 9.49 | 8.05 |
| Electrical power, kWte | 660.08 | 711.68 | 604.11 |
| Electrical power at the end of operational life, Wte | 594.07 | 640.51 | 543.70 |

The electric power was calculated taking into account the mass of americium 241 in oxide in each case. The efficiency is 7.5%, which is quite achievable for today's level of technology. Facilities with maximum power, taking into account their weight, can be used exclusively in ground conditions, to provide electricity, for example, autonomous research equipment. However, more compact versions of
the RTG with americium can be used in spacecraft such as satellites or rovers. An important advantage of this nuclide is its long half-life, due to which the drop in power to 90% of the nominal, which, according to GOST [15], is a limitation of the duration of the installation, occurs in 65 years. In real conditions, when the device is used in space or in the harsh conditions of the Far North, the service life may decrease due to the wear of the case material or degradation of other mandatory components of the installation, however, the limitation of the service life of such a "battery" is much higher than that of its competitors (4.37 years for Pu-238 and 13.33 years for Sr-90, taking into account the requirements of regulatory documents [15]).

2.2. Protection against gamma radiation
In addition to the problem of the nuclear chain reaction of Am-241 fission, the presence of soft gamma radiation accompanying the decay of americium should be noted. The yields of gamma quanta and their energy are shown in table 6 [16].

| Isotope | Energy, keV | Yield to decay |
|---------|------------|---------------|
| Am-241  | 59.54      | 0.360         |
|         | 33.20      | 0.001         |
|         | 26.34      | 0.024         |

The importance of considering the problem lies in handling the device at the beginning of its campaign (assembly, transportation, installation), as well as at the end (dismantling and transportation for further storage), which is more typical for ground devices.

For the calculations, three versions of the RTGs were determined, on the basis of which the activity of the sources was determined. The first two options are based on prototypes developed by specialists from the Energia Rocket and Space Corporation and Biapos LLC. The configurations correspond to the minimum and maximum RTG energies discussed in the article, which corresponds to a thermal power of 25 Wte and 400 Wte [13]. The device with the maximum possible amount of americium, which is calculated above (model 2 without a reflector), is adopted as the third configuration. A schematic representation of the calculated problem is shown in figure 3.

![Figure 3. Geometry of the problem for calculating protection.](image-url)
The values of 1 mSv/hour for a point on the surface of the device and 0.05 mSv/hour for a point at a distance of 1 m from the surface of the device are taken as the maximum permissible values of the dose rate. These values are 2 times stricter than those specified in GOST for RTGs, which is associated with a decrease in the possible calculation error [15]. The calculation was carried out according to the formula [17]:

\[
\dot{X} = 4\pi \cdot \Gamma_x \frac{q_S R}{2(r+R)} \cdot (F(\vartheta_1, k) + F(\vartheta_2, k)) \cdot \exp(\mu \cdot d) \cdot B(\mu d) \cdot \delta_{D,Z} \quad (1)
\]

where \(F(\vartheta, k)\) - elliptic integral;
\(\delta(D, Z)\) - barrier correction.

The results are shown in table 7.

**Table 7.** Thickness of protection for installations of different power.

| RTG configuration | Exposure dose rate without protection, mSv/hour | Steel protection thickness, cm |
|-------------------|-----------------------------------------------|-------------------------------|
| 25 Wt             | 2.45E+13                                      | 2.54                          |
| 400 Wt            | 1.35E+14                                      | 2.74                          |
| 9490 Wt           | 1.16E+15                                      | 2.92                          |

The result obtained indicates that there is no need to build a complex and heavy protection against ionizing radiation, which allows the use of such devices for spacecraft.

The exposure dose from a radioisotope source at the end of operation may also be interesting, since during the operation of the RTG, other nuclei are produced as a result of successive decays. For this, the americium decay chain was considered, shown in figure 4. All elements of the chain are subject mainly to alpha and beta decays with insignificant yields of low energy gamma quanta.

![Decay chain of Am-241](Figure 4. Decay chain of Am-241.)

To determine the final isotopic composition of the fuel of the installation, a system of differential equations was compiled and solved:
\begin{equation}
\begin{aligned}
\frac{dn_{Am}[t]}{dt} &= -\lambda_{Am} \cdot n_{Am}[t] \\
\frac{dn_{Np}[t]}{dt} &= \lambda_{Am} \cdot n_{Am}[t] - \lambda_{Np} \cdot n_{Np}[t] \\
\frac{dn_{U}[t]}{dt} &= \lambda_{Np} \cdot n_{Np}[t] - \lambda_{U} \cdot n_{U}[t] \\
\frac{dn_{Th}[t]}{dt} &= \lambda_{U} \cdot n_{U}[t] - \lambda_{Th} \cdot n_{Th}[t] \\
\frac{dn_{Bi}[t]}{dt} &= \lambda_{Th} \cdot n_{Th}[t] - \lambda_{Bi} \cdot n_{Bi}[t]
\end{aligned}
\end{equation}

Nuclides with an insignificant half-life in comparison with other elements are excluded from the system. Their concentration can be taken equal to the concentration of the next isotope in the chain. The result obtained for the example of an installation with a thermal power of 25 W is presented in table 8.

| Isotope | Concentration at a volume of 17.241 cm\(^3\) |
|---------|------------------------------------------|
| Am-241  | 4.557E+23                                |
| Np-237  | 2.859E+22                                |
| U-233   | 1.775E+17                                |
| Th-229  | 9.833E+12                                |
| Bi-213  | 8.854E+09                                |

The result obtained indicates a sharp decrease in the amount of nuclides following Np-237. This indicates an insignificant contribution to the exposure dose from these nuclides. Np-237 itself has two most pronounced gamma lines with energies of 29.37 and 86.48 keV with corresponding yields of 0.14 and 0.12. The low active nuclide and low yield of gamma quanta confirm the low contribution to the exposure dose. This also confirms the calculation according to formula 1. As a result, it can be concluded that the protection built for the start of operation of the installation is also suitable for the end of its operation.

3. Conclusion

The results obtained indicate the sufficient power that can be achieved in RTGs with Am-241, in comparison with installations with other isotopes. Such an installation can be used both for ground-based purposes and for spacecraft for various purposes. The only limitation can be the mass of such a device.

Further the need for protection against ionizing radiation is not a serious limitation, since its thickness does not exceed 3 cm in the most powerful version of devices.

References

[1] Yarygin V I 2013 Nuclear energy of direct transformation in space missions of the XXI century Izv. Vysshikh Uchebnykh Zawedeniy Yad. Energ. 2 pp 5-20
[2] Vorner D F 2015 Another modified version of a multipurpose thermoelectric generator providing power to the self-propelled apparatus Curiosity Thermoelectricity 1 pp 71-82
[3] Bibik I S and Valtseva A I 2015 Radioisotope thermoelectric generators: big in small Energy and resource saving. Power supply. Non-traditional and renewable energy sources pp 351-354
[4] Radioisotope thermoelectric generators [Electronic resource]. URL: https://bellona.ru/2005/04/02/radioizotopnye-termoele (accessed: 18.05.2020).
[5] Atomic energy 2.0 [Electronic resource]. URL: https://www.atomic-energy.ru/articles/2013/03/05/40258 (accessed: 10.05.2020).
[6] Maksimov A A, Gichev D V, Vysotsky V L, Filiprov A S, Tagiltsev A A, Cheranev M Yu, Goncharov R A 2016 Search for an accidentally flooded radioisotope thermoelectric generator according to the thermal field in the bottom layer of sea water Underwater research and robotics 1 pp 56-65

[7] Sarychev G A, Koloskov S A, Skachkov E V, Berdnikov V M, Pustovalov A A, Tsvetkov L A, Magomedbekov E P 2016 Americium-241 promising radionuclide for radioisotope energy sources V Int. Conf. School on Chemical Tech. pp 464-465

[8] Gabrielli F, Rineiski A, Vezzoni B, Maschek W, Fazio C, Salvatores M 2015 ASTRID-like fast reactor cores for burning plutonium and minor actinides Energy Procedia 71 pp 130-139

[9] Korobeinikov V V, Karazhelevskaya Y E, Kolesov V V, Terekhova A M 2019 Investigation of the possibility of Am-241 incineration and transmutation in americium-fueled reactor Izv. Wysshikh Uchebnykh Zawedeniy Yad. Energ. 2019 pp 63-153

[10] Mueller A C 2013 Transmutation of Nuclear Waste and the future MYRRHA Demonstrator Journal of Physics: Conf. Series 420 012059

[11] Kazansky Y A, Ivanov N V, Romanov M I 2016 Results of transmutation of small actinides in the neutron spectrum of thermal and fast neutron reactors Izv. Wysshikh Uchebnykh Zawedeniy Yad. Energ. 2 pp 77-86

[12] Ambrosi R M, Williams H R, Samara-Ratna P, Bannister N P, Vernon D, Crawford T and Konig J 2012 Development and testing of Americium-241 radioisotope thermoelectric generator: concept designs and breadboard system Nuclear and Emerging Technologies for Space

[13] Pustovalov A A, Pankin M I, Prilepo YU P, Rybkin N N and Sinyavskij V V 2016 Space radioisotope thermoelectric generators based on americium-241 Space engineering and technology 1 pp 57-63

[14] SanPin 2.6.1.2749-10 2010 Hygienic requirements for ensuring radiation safety when handling radioisotope thermoelectric generators

[15] Levenec V V, Ome'nik A P and SHChur A A 2007 Spectrometry of gamma and alpha radiation by semiconductor detectors based on CdTe (CdZnTe), manufactured at the NSC KIPT Nuclear Physics and Energy 4 p 22

[16] GOST 18696-90 1991 Radionuclide thermoelectric generators. Types and general specifications (Moscow: Publishing house of standards) p 10

[17] Romantsov V P, Romantsova I V and Tkachenko V V 2012 Collection of problems on dosimetry and protection against ionizing radiation. Tutorial. 2nd edition, supplemented and revised (Obninsk: IATE MEPhI) p 160