Research in Support of European Radioisotope Power System Development at the European Commission’s Joint Research Centre in Karlsruhe

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1 Introduction

The urge to discover the unknown, to explore the unexplored and to broaden our knowledge beyond the limits of the present is inherent to human nature. One of the most interesting and fascinating fields of science is the exploration of the cosmos, either from Earth using telescopes or by sending automated probes to other planets and into the vastness of space.

A basic requirement to operate automated spacecraft successfully over the course of an exploratory mission, is the reliable supply of long-lasting power. If independence of solar radiation is required, e.g. when travelling into deep space or to the dark side of planetary bodies, nuclear energy becomes advantageous compared to other potential sources of energy. The utilisation of nuclear power for applications in space had been considered since the early beginnings of spaceflight in the late 1940s, and the first Radioisotope Power System (RPS) in space was already launched by the U.S. Navy in 1961, onboard the Transit 4A navigational satellite [1,2]. Since then RPS have enabled some of the most spectacular missions in the history of space exploration [1-7], mostly performed by the USA but also by the former Soviet Union, China and Europe (via cooperation with the USA). RPS were used on satellites for navigation, meteorology and communication [8], they have powered scientific instruments on the Moon (Figure 1), and they were used for many of the most famous and exiting exploratory endeavours, such as the Pioneer missions to Saturn and Jupiter [9,10], the Viking missions to Mars [11], and the Voyager 1 & 2 spacecraft, which travelled beyond the boundaries of our solar system and are still delivering scientific results from the interstellar medium, more than 40 years after their launch [12,13]. More recently, the radioisotope-powered missions Galileo, Ulysses and Cassini-Huygens were exploring Jupiter, the Sun and Saturn/Titan, respectively. These more recent missions were performed in collaboration between the National Aeronautics and Space Administration (NASA) and the European Space Agency (ESA) [14]. Historically, this was the only way for Europe to gain access to space nuclear power systems. In 2005 a European Working Group on Nuclear Power Sources for Space identified RPS as a “key enabling technology for future European activities in space” [15], and suggested the establishment of an European safety framework for space nuclear power sources and the development of the technical capabilities to perform nuclear powered missions indendently [16,17]. As a consequence, a research and development programme was launched by ESA for the production of European RPS to satisfy thermal management and electrical power needs for spacecraft [16,17,18,19].

In the past, most RPS for space missions were based on the plutonium isotope Pu-238 [1-5], a radionuclide which is superior to other isotopes, because of its high specific power of 0.567 W/g, low radiation, compatibility with cladding materials and chemical stability as oxide. Pu-238 has a half-life of 87.7 years which enables long-lasting missions [20]. Unfortunately, there is a global shortage of this isotope, the efforts associated with its production are high [21] and there are currently no facilities for its synthesis in Europe. An alternative is the americium isotope Am-241, which is more easily available, since it is produced through decay from Pu-241 and can be extracted isotopically pure from existing stocks of civil plutonium in France or the United Kingdom via chemical extraction [22]. Therefore, ESA has decided to study the use of Am-241 for its RPS development [5,16,17]. However, Am-241 has some disadvantages compared to Pu-238, such as a lower power density of 0.114 W/g and slightly higher radiation levels. In addition, the oxide shows chemical instability at high temperatures, the experience with Am241 is limited and additional research and safety assessment is needed before it can be used for space applications.

Within the ESA research programme, the UK’s National Nuclear Laboratory (NNL) is exploring the cost effective production of Am-241 and the University of Leicester (UoL) is developing a European Radioisotope Heater Unit (RHU) and Radioisotope Thermoelectric Generator (RTG) [16,23,24]. To support these efforts, the European Commission’s Joint Research Centre (JRC) in Karlsruhe is investigating methods to stabilize americium in the oxide form and to establish a safe and reliable pelletizing process [20,25]. In collaboration with UoL, safety relevant properties and behaviour of Americium oxide are assessed under representative conditions for storage on Earth, operations in space as well as hypothetical accident and post-accident environments [16,26],
and the compatibility with the cladding material is tested. Complementary work is performed to develop a qualified welding methodology of the safety encapsulation.

2 Energy Supply in Space

In order to operate spacecraft, a reliable source of power in the form of electricity and heat is required. Electricity is needed to power onboard electronic systems such as navigation and manoeuvring systems, onboard computers, lighting, robotics, scientific instruments and communication systems. In some cases, spacecraft are equipped with electric propulsion systems, and electricity for life support is needed if the mission is manned. Heat is needed in cold environments to keep sensitive spacecraft components at minimum operational or survival temperature, e.g. during lunar nights.

Primary energy sources can be chemical, solar or nuclear. Some of these are limited with respect to their power or energy density, and the profile of each individual mission determines which energy sources can be utilized. Table 1 shows typical energy densities for different energy sources.

Chemical energy sources in the form of solid or liquid fuels can release a large quantity of energy in a very short time, but they are limited with respect to total energy density. For instance, chemical fuels for propulsion can be used whenever high thrust is needed for a short time, e.g. as rocket fuel to overcome the gravity field of earth, but they have shortcomings if long-lasting power or long acceleration times are needed, e.g. to reach the high velocities necessary for interplanetary or even interstellar travel. Chemical energy in the form of batteries or fuel for fuel cells can be used when onboard power is needed for no more than a few weeks, or as rechargeable energy buffer to supply peak loads or to bridge periods without sunlight. For missions, which require continuous power supply for an extended time, only solar or nuclear energy sources are feasible, since the payload associated with chemical fuels or batteries would simply become too high.

Solar energy is principally unlimited, as long as the solar cells, used to convert the radiation energy into electricity, are not degrading, and as long as they can be adjusted in the direction of the sunlight and the spacecraft is not in the shadow of planetary bodies or too far away from the sun. However, their effective area, the conversion efficiency and the intensity of the solar radiation determine the power density of solar cells. This intensity decreases inversely with the square of the distance from the sun, as shown in Figure 2, and if the distance becomes too large solar energy becomes unpractical. For example, while the solar constant is 1.367 kW/m² at the semi-major axis of Earth, at one Astronomical Unit (AU) distance to the sun, it decreases to only 51 W/m² or 3.7 % at the semi-major axis of Jupiter (5.2 AU). Therefore, solar arrays are not an option for all research missions to the outer solar system, but also not if a system is to be operated during long periods of darkness, for example during the lunar nights, which last 14 days.

Nuclear energy has the highest power densities of all possible onboard energy sources, and can deliver reliable power over very long time periods. Most importantly, it is independent of sunlight. There are two types of space nuclear power systems; Reactor power systems (small nuclear reactors), which generate power by controlled fission of fissile isotopes, such as U235 or Pu-239, and Radioisotope Power Sources or Systems (RPS), which obtain their energy from the spontaneous decay of radioactive isotopes. Both types can generate heat for temperature control and/or electricity via additional energy conversion systems.

While nuclear reactors are generally suited for applications, which need significant power levels above 10 kW, RPS are employed whenever a limited amount of solar-independent power, up to 5 kW, is needed for a longer time period. RPS are compact, long-lived, reliable, robust, radiation resistant, solar-independent, maintenance-free, and they have energy densities, which are several orders of magnitude above chemical power sources (Table 1, 1,2,3). Figure 3 shows qualitatively different regimes of power levels and durations, where different energy sources are applicable.

Space power systems, which are based on the decay heat of radioisotopes, can be distinguished into systems which make direct use of the thermal energy, and systems which convert heat into electricity (Figure 4). In both cases the employed radioisotope is the power

<table>
<thead>
<tr>
<th>Energy source</th>
<th>Energy density, MJ/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂ + O₂</td>
<td>13.33</td>
</tr>
<tr>
<td>N₂H₄ + O₂</td>
<td>9.75</td>
</tr>
<tr>
<td>Li + O₂</td>
<td>12.2</td>
</tr>
<tr>
<td>Li-ion battery</td>
<td>0.9</td>
</tr>
<tr>
<td>Fission of U-235**</td>
<td>8.2 · 10⁶</td>
</tr>
<tr>
<td>Decay of Pu-238***</td>
<td>3.3 · 10³</td>
</tr>
<tr>
<td>Decay of Am-241***</td>
<td>7.1 · 10³</td>
</tr>
</tbody>
</table>

*Hydrazine, **total fission, ***over 20 years mission time

[Table 1. Energy densities of typical energy sources.]
source while the heat sink is provided by space. Systems which make direct use of the thermal energy are called Radioisotope Heater Units (RHU). They provide heat to the space craft to keep sensitive electronics warm without using heavy and complicated heat distribution systems, and without creating electromagnetic interference.

Heat-to-electricity conversion systems can be classified into dynamic and static systems. The dynamic systems employ moving parts and use a thermodynamic cycle to convert heat into electricity, e.g. a Stirling engine [27], and show principally the highest conversion efficiencies. The earliest efforts were focussed on the development of dynamic conversion systems, and the first RPS SNAP-1 (SNAP stands for Systems for Nuclear Auxiliary Power) in 1959 was based on a Ce-144 powered mercury Rankine cycle [1]. But despite their high conversion efficiency, dynamic conversion systems have not yet reached the reliability required for the operation of a space probe, which might be on a mission for several decades without the possibility for maintenance or repair, and no dynamic conversion system was ever used in space.

The static heat-to-electricity conversion systems are called Radioisotope Thermal Generators (RTG). RTG have no moving parts and use the thermoelectric principle, also known as the Seebeck effect, to generate electricity. This phenomenon was discovered in 1794 by the Italian scientist Alessandro Volta and, independently, in 1821 by the German physicist Thomas Johann Seebeck. If two dissimilar materials are connected in a closed circuit and a temperature difference is applied over the two junctions, a voltage can be measured and electricity is generated (Figure 5). Such a device is called a thermoelectric couple or thermocouple. In addition, static heat conversion is also possible by thermionic conversion, where a flow of electrons is induced from a hot to a cool surface via thermionic emission.

Thermoelectric conversion is not very efficient and practical RTG systems using SiGe or PbTe/TAGS thermocouples show typical power conversion efficiencies of 6 % to 7 % [5]. To improve efficiency, development of high temperature thermoelectric materials including skutterudites and Zintl-based systems is ongoing [28,29]. However, if operated at low temperatures or under a protective gas cover, existing RTG are very reliable, show low degradation and can provide power over many decades. Because of the importance of reliable and maintenance-free systems for automated space probes, relatively low conversion efficiencies are usually accepted.

The most important design criterion for any space nuclear power system, including all forms of RPS, is safety. If an RPS shall be employed on a space application, it must comply with resolution 47/68 of the United Nations General Assembly on the Principles Relevant to the Use of Nuclear Power Sources in Outer Space. The resolution defines that the use of RPS shall be restricted to those space missions, which cannot be performed by non-nuclear energy sources in a reasonable way. It also states that the design and use of the RPS shall ensure that the hazards during operation and foreseeable accidents are kept below acceptance levels and that radioactive material does not cause a significant contamination of the biosphere and outer space [30].

In order to comply with these requirements, RPS are designed to the meet highest safety standards. The fuel is encapsulated in a cladding of a highly refractory noble metal like iridium or platinum-rhodium alloys (Figure 6), which can withstand the most extreme conditions (e.g. launch pad explosion, Earth re-entry accidents). The cladding is surrounded by thermal insulation, usually made of pyrolytic graphite, which shall protect the cladding from reaching peak temperatures during aerodynamic heating. Finally, the thermal insulation is surrounded by an aeroshell made of carbon-carbon composite (fine-weaved pierced fabric), which provides additional protection from postulated launch vehicle explosions or against impacts on hard surfaces at terminal velocity [1,5,14].

3 Radioisotopes for RPS

The selection of suitable radioisotopes for application in space RPS is based on a number of criteria; among them are a long half-life, high isotopic power and a low level of penetrating radiation. In addition, a chemically stable compound with high density should exist, which can serve as stable host for the decay products and is compatible to the encapsulation materials and the potential operating or post-accident environments. Furthermore, the compound should resist high temperatures and should not disperse into inhalable small particles, in case of an accident. A low solubility in the environment (water) and the human body are also advantageous [4,5].
Since the specific power correlates inversely to the half-life, a compromise has to be found between specific weight and volume of the heat source on one side, and a long-lasting stable power output during the required mission time on the other side. Therefore, the selection of a suitable radioisotope always depends on the concrete application. Alpha emitters tend to be better suited than beta emitters are, because the alpha decay energy is typically in the range between 5 MeV and 6 MeV per decay event, for example compared to 0.546 MeV for the beta decay of Sr-90, and the alpha particles do not generate Bremsstrahlung when stopped in the surrounding matter.

Other important criteria are the availability of the selected isotope, as well as the production costs and necessary infrastructure and effort to process the radioactive material. Table 2 gives an overview over the most common radioisotopes for space RPS, of which Pu-238 is by far the most significant. If not mentioned otherwise, all nuclear data were taken from the JEFF-3.1 nuclear data library [31] via Nucleonica.com [32].

4 Properties of Plutonium-238 & Americium-241

The development of European RPS is based on the strategic decision of ESA to utilize Am-241 and to take advantage of the existing nuclear infrastructure related to the civil reprocessing of spent nuclear fuel in Europe, rather than to establish an expensive production capability for Pu-238 [16,17]. In order to understand the impact of this choice on the RPS design characteristics and the production process, a comparison of both isotopes has to be made. Table 3 summarizes the most important properties of Am-241 compared to Pu-238.

Pu-238 is an isotope of the chemical element plutonium, an actinide which is artificially created in nuclear reactors. It was the first isotope of plutonium, which was discovered by Glenn T. Seaborg in 1940 [33], and it is the most important and most widely adopted radioisotope for space power applications, due to its high power density, long half-life, chemical stability as oxide, and its low neutron and soft gamma emissions. So far, Pu-238 is the only radioisotope used for RPS in space by the USA and China, while the former Soviet Union also utilized Po-210.

Pu-238 has a half-life of 87.7 years and an isotopic power of 0.567 W/g. The isotope decays primarily via alpha decay to U-234, with a decay energy of 5.59 MeV. The main radiation emissions of Pu-238 are alpha particles with an average energy of 5.49 MeV. In addition, the isotope emits soft gamma radiation (main energies: 43.5 keV & 99.85 keV) with low emission probability, and Auger electrons, which in turn generate X-rays at 17.11 keV and 13.61 keV (main lines), also with relatively low emission probabilities. For a sintered and encapsulated pellet, the self-shielding effect and encapsulation are more than sufficient to shield these radiations. Spontaneous fission occurs with a negligible probability of 1.86E-09, and the spontaneous fission reaction creates a neutron yield of circa 2300 n/(s g) (oxide). However, alpha-neutron (α-n) reactions in natural oxygen cause an additional neutron yield of circa 13 400 n/(s g) in plutonium oxide [34]. The (α-n) reactions can only occur in the low abundance isotopes O-17 and O-18, while the threshold energy of O-16 is too high (15.2 MeV) [34]. The neutron yield in pure Pu-238 oxide can be reduced to circa 2700 n/s-g [5] if the oxygen is depleted in O-17 and O-18 by 98 % [21].

Metallic plutonium has a density of 19.77 g/cm³ (Pu-238, α-phase at room temperature and atmospheric pressure), which is the highest density form and would allow a volumetric power density of up to 11.21 W/cm³. However, the metal shows six different structural modifications at ambient pressure in the temperature range from 0 K (-273.15 °C) to 640 °C (melting point), and the phase transitions cause significant dimensional changes as well as alterations of the mechanical and thermal properties (Figure 7). In addition, plutonium metal is burnable, and the powder is extremely pyrophoric. The first RTGs in the 1960s were still fuelled with metallic Pu-238, and designed to burn-up into fine particles below 30 nm and disperse into the atmosphere in case of an re-entry accident. This safety philosophy had to prove itself, when the Transit satellite 5BN-3 failed to achieve orbit in 1964, and the SNAP-9A RTG re-entered the atmosphere, carrying about 1 kg Pu-238 metal. As predicted, the metallic fuel completely burned-up and was completely oxidized, giving rise to a very big explosion and to a significant radiation hazard.

Tab. 2.
Radioisotopes for space applications, [31,32].

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life (y)</th>
<th>Isotopic power (W/g)</th>
<th>Principal decay mechanism</th>
<th>Comment</th>
<th>Shielding (typically)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Am-241</td>
<td>432.8</td>
<td>0.114</td>
<td>alpha</td>
<td>Soft gamma radiation</td>
<td>2 mm lead equivalent</td>
</tr>
<tr>
<td>Cs-137</td>
<td>30.04</td>
<td>0.417</td>
<td>beta</td>
<td>Gamma emitter</td>
<td>Heavy</td>
</tr>
<tr>
<td>Ce-144</td>
<td>285</td>
<td>2.08</td>
<td>beta</td>
<td>Gamma emitter</td>
<td>Heavy</td>
</tr>
<tr>
<td>Cm-242</td>
<td>0.45</td>
<td>122</td>
<td>alpha</td>
<td>Strong neutron emitter</td>
<td>Heavy</td>
</tr>
<tr>
<td>Cm-244</td>
<td>18</td>
<td>2.84</td>
<td>alpha</td>
<td>Strong neutron emitter</td>
<td>Heavy</td>
</tr>
<tr>
<td>Po-208</td>
<td>2.93</td>
<td>18.1</td>
<td>alpha</td>
<td>Short half-life</td>
<td>None</td>
</tr>
<tr>
<td>Po-210</td>
<td>0.38</td>
<td>144</td>
<td>alpha</td>
<td>Short half-life</td>
<td>None</td>
</tr>
<tr>
<td>Pu-238</td>
<td>87.7</td>
<td>0.567</td>
<td>alpha</td>
<td>Very soft gamma radiation</td>
<td>None</td>
</tr>
<tr>
<td>Sr-90</td>
<td>28.79</td>
<td>0.907</td>
<td>beta</td>
<td>Bremsstrahlung</td>
<td>Significant</td>
</tr>
</tbody>
</table>

Tab. 3.
Properties of Pu-238 and Am-241 (Metal and Oxide). *Typical isotopic composition, **Stabilized with 12 % U, ***Sub-stoichiometric.
Fig. 7. Thermal conductivity of metallic plutonium structural modifications [35].

Fig. 8. Production of Am-241 by neutron capture and β-decay.

dispersed into the atmosphere where it was diluted to low concentrations and did not cause any unacceptable health hazard [1,8,14].

After the Transit SB-3 accident and with the upcoming of larger RPS with higher radioactive inventory the dispersion approach was no longer accepted and a new fuel form, which would stay intact at re-entry was needed. Since then, the preferred chemical form of Pu-238 for RPS is plutonium dioxide (PuO₂), safely encapsulated in a cladding of high refractory material which can safely contain the radioactivity under all credible circumstances [8]. PuO₂ is a ceramic material with a high melting point of 2744 °C [36], which can be sintered into stable pellets. The compound crystallizes in the face centred cubic (fcc) structure (space group Fm3m) [37], has a high chemical stability, a low solubility in water and does not react with the typical cladding materials, such as iridium or platinum alloys (e.g. Pt20Rh or Pt30Rh). In addition, it serves as a good host for U-234, the decay product of Pu-238, which is also crystallizing in the fcc structure.

Am-241 is an isotope of the radioactive element americium. It belongs also to the actinides and has the atomic number 95. Like plutonium, americium is an artificial element and was discovered by the group of Glenn T. Seaborg in 1944. Americium is usually created in nuclear reactors by neutron capture and radioactive decay (Figure 8). However, during irradiation not only Am-241 but also other americium isotopes are created, which are unwanted in RPS. Isotopically pure Am-241 is produced continuously in the stocks of civil plutonium through beta decay of Pu-241 (t½=14.33 y), from where it can be separated using chemical methods. A cost efficient separation and purification process (AMPEX) was developed and demonstrated by the UK’s National Nuclear Laboratory (NNL) to separate ingrown Am-241 from plutonium [22,39].

Am-241 has a half-life of 432.8 years and an isotopic power of 0.114 W/g. The isotope decays primarily via alpha decay to Np-237. In a final repository Am-241 is one of the most important drivers for the medium-term heat load, and therefore the required gallery space, and Np-237 (t½=2.14×10⁶ y) is one of the significant isotopes driving long-term radio toxicity [40,41]. The main radiation of Am-241 are alpha particles with an average energy of 5.47 MeV. Unlike Pu-238, Am-241 emits significant gamma radiation at 59.54 keV (main line) with a probability of 36 % and also Auger electrons, which cause X-rays at 14.44 keV (probability 33 %). Even though the radiation is still relatively soft, shielding and remote handling tools are required if larger amounts are to be processed, and especially when the material is in solution and self-shielding is not effective. Once the pellets have been sintered and encapsulated the radiation decreases. However, it remains still significant and adequate radiation protection measures for workers need to be in place. A dose rate of circa 150 µSv/h in 10 cm distance is estimated for a typical full scale RHU containing 26 g AmO₂ encapsulated in 1.8 mm Pt30Rh.

Am-241 has a very low probability for spontaneous fission of 4.3E-12, and the spontaneous fission reaction creates a negligible neutron yield of circa 1.2 n/s/g (oxide). However, as in the case of Pu-238, Am-241 oxide emits additional neutrons from the alpha-neutron (α-n) reaction in natural oxygen. This reaction causes an additional neutron yield of circa 2700 n/(s g) [34]. While this is significantly lower compared to Pu-238 by a factor of six, it has to be considered that for the same thermal power about five times more Am-241 is needed. Therefore, also in the case of Am-241 oxide O-17 and O-18 depletion is needed to reduce the neutron yield to acceptable levels [5].

Metallic americium has similar disadvantages as metallic plutonium for the use in RPS. In addition, its density is relatively low compared to the oxide (13.67 g/cm³ α-phase at room temperature). Elemental americium is a soft metal, which oxidises quickly in the atmosphere forming a protective oxide layer. At room temperature americium forms a stable hexagonal α-phase (space group P6₃/mmc) [35], at 769 °C it changes into the cubic β-phase (space group Fm³m), and at 1077 °C it converts to the γ-phase showing a body-centered cubic structure [35]. The phase transitions cause dimensional changes, however not as significant as in the case of plutonium. The melting point of metallic americium is at 1176 °C. Americium metal powder is also very pyrophoric and metallic americium would definitely burn-up in case of an uncontaminated re-entry accident.

The preferred form of Am-241 for space applications is americium oxide, a ceramic material with a density of 11.68 g/cm³ which can be sintered into pellets [6,16,20,25]. However, there are two major compounds which are relevant for applications in space, the cubic dioxide (AmO₂) (α-phase, space group Fm3m) and the sesquioxide (Am₂O₃) which exists in the hexagonal form (Aphase, space group P3m1) and the cubic form (C-phase, space group Ia3). Unfortunately, the americium-oxygen system shows a complex behaviour. The melting point of AmO₂ is at 2113 °C [42], but at high temperatures and in the vacuum of space americium dioxide loses oxygen. Due to this process, it transforms into substoichiometric AmO₂-x, thereby slowly increasing its volume, until it finally converts into the hexagonal sesquioxide [37,43,44]. This process causes significant dimensional changes and can lead to decomposition of the pellets [25,45]. In addition, the release of oxygen can cause pressure build-up and potentially corrode surrounding
structures. On the other higher, americium sesquioxide is prone to oxidation at lower temperatures and will slowly convert into the dioxide in air, again undergoing significant structural modifications. After several weeks of storage, even under low oxygen partial pressures, this effect, in combination with self-irradiation from \( \alpha \)-decay, will cause Am\(_2\)O\(_3\) pellets to disintegrate into black dioxide powder. Both phenomena, the reduction of Am\(_2\)O at elevated temperatures and the oxidation of Am\(_2\)O\(_3\) at low temperatures, are problematic with respect to the integrity of the fuel pellets and can cause increased dispersion of radioactive material in case of certain accident scenarios \([25]\).

5 The Minor Actinide Laboratory at the Joint Research Centre Karlsruhe

The Joint Research Centre is the European Commission’s science and knowledge service. Its mission is to support EU policies with independent evidence throughout the whole policy cycle. Its work has a direct impact on the lives of citizens by contributing with its research outcomes to a healthy and safe environment, secure energy supplies, sustainable mobility and consumer health and safety. The JRC hosts specialist laboratories and unique research facilities and is home to thousands of scientists working to support EU policy (https://ec.europa.eu/jrc/en).

The JRC in Karlsruhe belongs to the Directorate for Nuclear Safety and Security (Directorate G), where JRC’s nuclear work programme, funded by the EURATOM Research and Training Programme, is carried out. The Directorate contributes to the scientific foundation for the protection of the European citizen against risks associated with the handling and storage of highly radioactive material, and scientific and technical support for the conception, development, implementation and monitoring of community policies related to nuclear energy. Research and policy support activities of Directorate G contribute towards achieving effective safety and safeguards systems for the nuclear fuel cycle, to enhance nuclear security and then contributing to achieving the goal of low carbon energy production.

The JRC supports the ESA research programme on developing a European Radioisotope Heater Unit (RHU) and Radioisotope Thermoelectric Generator (RTG). These activities are focussed on investigating methods to stabilize americium in the oxide form and to establish a safe and reliable pelletizing process \([20,25]\). In collaboration with UoL, safety relevant properties and behaviour of americium oxide are assessed \([16,26]\).

Am-241 emits a significant amount of gamma radiation, and working with Am-241 can result in high dose rates for the operating personnel. Remote operated and shielded equipment is advantageous or even necessary in order to prepare americium-based pellets for RPS. The JRC in Karlsruhe has a unique infrastructure for handling of highly radiative actinide materials, the so-called Minor Actinide Laboratory (MA-Lab) \([46]\). It is of high relevance for safety research on fuels for transmutation in Europe, as it is one of the only dedicated facilities for the synthesis of minor actinide containing materials, either for property measurements or for the preparation of irradiation experiments.

The MA-lab consists of seven glove-boxes with protection walls forming two separate chains. A schematic lay-out of the Ma-Lab is shown in Figure 9. The glove boxes are shielded by 50 cm neutron shielding and 5 cm of lead. Based on the thickness of the water and lead wall, the mass limits have been calculated to 150 g of Am-241 or 5 g of Cm-244. The glove boxes can be accessed manually from the back if radiation levels are low enough to perform experiment preparation or maintenance. In addition, tele-manipulators and remote operated automated equipment can be used for operation at high dose rates.

The glove boxes of the minor actinide laboratory are configured as complete preparation chain for minor actinide containing samples from the base material to the fully encapsulated sample, and the MA-Lab represents an ideal infrastructure for preparation of highly radiating americium pellets and fully qualified fuel pins.

The synthesis of the base material (powder) is performed in the glove box named “infiltration”. The process is dust-free, based on the so-called gel supported precipitation \([47]\) and the porous bead infiltration technique \([48]\). This process is highly flexible and easily adapted to the requirements and specifications of new sample compositions. The next glove box contains a calcination furnace and other equipment for powder preparation. The prepared powders are dust-free, the individual beads typically show heterogeneous size distributions between 30 µm to 120 µm and are ideal for pressing pellets. The ready to press powder can be transferred via an automated channel to the next glove box, where it can be pressed to pellets. After sintering in reducing or oxidizing atmosphere (glove box “Sintering”) the pellets are fully characterized and inserted into a cladding. Finally, pin welding and non-destructive weld examination are performed in the two last, alpha free glove boxes.

6 Stabilisation of Americium Oxide & RHU-Size Prototype Pellet Production

Unlike plutonium oxide, which is stable in a broad range of temperatures and oxygen potentials, americium oxide is prone to phase changes and disintegration in changing environments \([25]\). If americium oxide is sintered under oxidizing conditions into AmO\(_2\), it releases oxygen at elevated temperatures in...
the vacuum of space and changes into sesquioxide \((\text{Am}_2\text{O}_3)\), thereby undergoing strong structural reorganization, density changes and disintegration. If americium oxide is sintered under reductive conditions into \(\text{Am}_2\text{O}_3\), it will transform into the dioxide under accidental conditions, but also under the influence of self-irradiation even at low oxygen potentials, which leads to its total disintegration.

JRC has investigated possibilities to stabilize americium dioxide in its cubic form under a broad range of temperatures, in oxidizing as well as reducing atmospheres. A solution was found by inserting 12 % of uranium into the americium oxide (in addition to 6 % Np and 2 % Pu already present). Thereby, it was possible to stabilize the cubic phase and to sinter a number of discs and pellets, including a prototype pellet in the dimensions of the US LWRHU [49] under moisturized Ar/H2 atmosphere and a larger disc representative for a future European RHU [16] (Figure 10).

Due to the absence of phase changes, the stabilized material showed a good sintering behaviour and it was possible to sinter a number of discs and pellets with good quality and without cracking. After sintering oxidation testing was performed and the material proved stable up to 1000 °C. This result represents a significant improvement with respect to safety of the material against radioactive material dispersion in case of accidental conditions. In addition, the macroscopic and crystallographic swelling was assessed on the small scale prototype pellet (Figure 10) over time. While no macroscopic swelling was observed, only low crystallographic swelling occurred due to self-irradiation, which saturated after circa 60 days. Overall the pellet showed good long-term structural and dimensional stability under self-irradiation conditions (Figure 11) [25].

7 Development of Welding Methodology

In order to be able to meet the launch safety requirements and safely ship RPS sealed sources to sites, where they can be assembled into RHUs or RTGs and subsequently be installed onto a spacecraft, it is necessary to develop containment technologies that meet these requirements. The first layer of containment immediately surrounding the fuel pellets is the cladding, which must ensure the enclosure of radioactivity during storage, normal operation and accident scenarios. The encapsulation has to be performed in a nuclear installation, ideally the manufacturing site, and it has to be ensured that the fueled clads are free of external contamination.

In order to test the feasibility of our welding equipment and to gain experience in the welding of Pt30Rh capsules, two types of Pt30Rh-encapsulation were constructed and welding tests were performed. The first capsule design had similar dimensions as the US LWRHU (Figure 12) and the second capsule design was made according to input by UoL to host \((\text{Am},\text{U})\text{O}_2\) pellets of 15 mm diameter and 20 mm height.

The capsules were welded using established Tungsten Inert Gas welding equipment, which is also used in the frame of qualified welding of fuel rodlets for irradiation experiments [41]. Non-destructive as well as destructive weld examinations were performed and showed that good welding results were achieved; indicating that future welding quality criteria can be met (Figure 13).

8 Conclusions

Radioisotope power sources are a key enabling technology for exploratory missions into deep space or to the dark side of planetary bodies, and the European Space Agency is sponsoring the development of Am-241 based radioisotope power systems. The development of a new RPS based on americium is a challenging task. The optimization of the fuel is a key issue, as the oxide of americium has significantly different properties compared to that of Pu-238, both in terms of...
material engineering and handling. JRC supports the development of a European radioisotope heater unit and radioisotope thermoelectric generator. These activities are focused on investigating methods to stabilize americium in the oxide form and to establish a safe and reliable pelletizing process. Safety relevant properties and behaviour of americium oxide, as well as the compatibility to the cladding material, are assessed in collaboration with the University of Leicester. Prototype pellets of stabilized americium oxide were synthesized in the Minor Actinide Lab of JRC Karlsruhe, and their stability under a broad range of conditions was shown. A welding methodology for the safety encapsulation was developed and demonstrated.

References


