EXPLORATORY RESEARCH ON RADIOISOTOPE THERMOELECTRIC GENERATORS FOR DEEP SPACE MISSIONS

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ABSTRACT

To support the development of a Radioisotope Power Source (RPS) for exploration missions into deep space based on ^{241}Am, the European Commission's Joint Research Centre (JRC) has recently launched an exploratory research project, scheduled for a period of two years. It aims at finding innovative solutions for higher performance, safety and reliability of such a device. In addition, it will serve as a start-up platform for this field of research and enable identification of research areas, where JRC can significantly complement and support the already existing activities in Europe. The project is divided into several subtasks and will investigate

- safety related properties of americium (oxide) as a heat source,
- compatibility of americium compounds with cladding and structural materials,
- properties of alternative americium compounds,
- properties of advanced thermoelectric materials.

The new exploratory research project will be introduced together with an overview on the available facilities and capabilities of JRC in this domain. Alternative americium forms with potential improved stability versus the oxides are discussed and innovative thermoelectric materials based on actinides are introduced.

1. INTRODUCTION

The European Space Agency (ESA) has decided to develop a Radioisotope Heating Unit (RHU) and a Radioisotope Power Source (RPS) for future exploration missions into deep space and contracted several research organisations, mostly in UK, with the implementation [1]. Unlike past ESA missions, which utilized solar panels as energy source, the new device will be powered by ^{241}Am, a transuranic isotope, which generates heat by decay. The heat will be either used directly to keep sensitive equipment at a required temperature or converted to electricity by means of thermoelectric converters (thermocouples). Future concepts might also include more efficient conversion mechanisms, such as Stirling motors [1], [2].

Most of the historical nuclear power sources for exploration missions were either deployed by the United States or the former Soviet Union and were based on ^{238}Pu [2], [3], a radionuclide with a high isotopic power of 0.567 W/g and a half-life of 87.7 years [4]. Unfortunately, there is a global shortage of this isotope and the costs associated with its production are high. Therefore, ESA has decided to alternatively use ^{241}Am, which has a half-life of 432.2 years. ^{241}Am is a decay product of ^{241}Pu ($T_{1/2}$ = 14.35 y) and can be extracted chemically from existing plutonium stocks in Europe. ^{241}Am, however, has some disadvantages compared to ^{238}Pu, such as a relatively low power density of 0.114 W/g, chemical instability at high temperatures and higher radiation levels.

In order to support the development of a European RPS the European Commission's Joint Research Centre (JRC) has launched an exploratory research project. Within the frame of this project JRC will perform an assessment of the safety and reliability relevant properties of americium oxide, which is the currently considered chemical form for application in space batteries, (e.g. measurement of thermal properties, investigation of helium release behaviour, swelling behaviour and oxygen release kinetics, ageing studies, examination of self-irradiation damage) and its compatibility to cladding and structural materials. In addition, alternative americium compounds will be investigated for their thermal and mechanical properties as well as their chemical stability under space mission conditions. Innovative candidate materials for thermo-electric conversion containing actinides will be assessed at high temperatures, utilizing the unique facilities and competences at JRC-ITU and JRC-IET.

2. ^{241}AM AS POWER PROVIDING ISOTOPE

^{241}Am is a transuranic element which is created in nuclear reactors by neutron capture and subsequent decay. Despite the half-life of "only" 432.2 years ^{241}Am is one of the main contributors to the long-term radiotoxicity of spent nuclear fuel, due to its decay product ^{237}Np, which has a half-life of 2.144 million years. It is, therefore, a key isotope for research on partitioning and transmutation with the aim to destroy the long-lived radioactive waste in dedicated irradiation facilities [5]. However, up to now ^{241}Am was never utilized as power providing isotope for space applications (to our knowledge), and some of its

characteristics are different to ^{238}Pu and need to be considered.

Radiation: ^{241}Am decays by alpha decay to ^{237}Np with a specific activity of $1.271 \cdot 10^{11}$ Bq/g and it emits significant gamma radiation (Tab. 1). Therefore, it has to be handled under strict radiation protection measures, and special equipment, such as shielding and remote operation tools, is required. Due to the low energy of the emitted gamma rays (main line at 59 keV) it can be shielded relatively easy with few millimetres of lead (or comparable), but in opposite to ^{238}Pu at least some shielding has to be provided. Therefore, additional payload for a RPS powered by ^{241}Am might be necessary.

Table 1. ^{241}Am Gamma Emission [4].

Type	Energy(keV)	Emission Probability
	JEFF-3.1	JEFF-3.1
γ	59.5412 (± 0.0001)	3.600e-1 (± 1.0e-3)
γ	26.3448 (± 0.0002)	2.430e-2 (± 3.2e-4)
γ	27.040 (± 0.060)	6.26e-3 (± 9.5e-4)
γ	33.1964 (± 0.0003)	1.217e-3 (± 3.9e-5)
γ	43.420 (± 0.020)	6.59e-4 (± 2.3e-5)
γ	42.720 (± 0.040)	3.9e-4 (± 1.1e-4)
γ	98.970 (± 0.020)	2.23e-4 (± 1.1e-5)
γ	102.960 (± 0.020)	2.09e-4 (± 1.1e-5)
γ	55.550 (± 0.040)	1.84e-4 (± 2.2e-5)
γ	125.310 (± 0.020)	4.10e-5 (± 1.2e-6)
γ	69.760 (± 0.020)	2.88e-5 (± 3.0e-6)
γ	28.510 (± 0.050)	2.77e-5 (± 3.0e-6)
γ	123.000 (± 0.040)	1.080e-5 (± 3.9e-7)

Chemical stability: In order to avoid burnup and potential dispersion into the atmosphere during launch abort or re-entry accidents a chemical and mechanical stable form of americium under oxidizing conditions is desired. Therefore, the currently existing RPS designs include dioxide pellets encapsulated in an iridium cladding [2]. Unfortunately, americium exists in different oxidation states and the dioxide is prone to phase changes at elevated temperatures and in vacuum [6]. Fig. 1 illustrates the phase change in vacuum at 1200°C from the dioxide (AmO_2) to the sesquioxide (Am_2O_3) as well as the associated phase change from cubic (alpha) to hexagonal structure.

Because of this behaviour, the oxide form might be not the optimal solution with respect to long term stability of the pellets in the vacuum of space. Alternative compounds, such as $AmAlO_3$, could show a more stable behaviour at high temperatures under both, oxidizing and reducing conditions. Within the frame of the presented exploratory research project JRC will investigate a number of candidate americium compounds for their chemical stability and resistance towards self-irradiation effects.

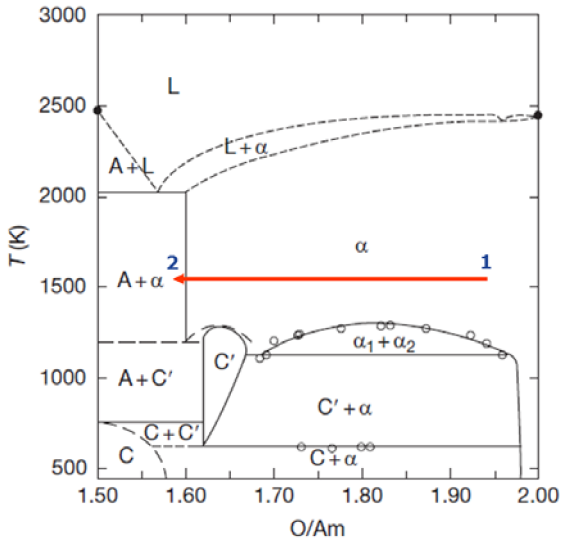

Figure 1. Americium-Oxygen Phase Diagram [6].

Thermal power: Tab. 2 shows a comparison between the thermal properties of the oxides of ^{238}Pu and ^{241}Am. $^{241}AmO_2$ has a power density of 0.101 W/g, which is about one fifth of $^{238}PuO_2$ (0.493 W/g). As a consequence it is necessary to use much more material in order to achieve the same power if americium is chosen. Therefore, it would be very interesting for an americium driven RPS to find possibilities to partly compensate the low power density of ^{241}Am by increased efficiency of the electricity conversion. This could be f.i. achieved by higher operating temperatures, thereby increasing the Carnot efficiency. Therefore, the exploratory research project will look into alternative americium forms, which are long-term resistant to high temperatures, but also candidate thermos-electric materials with high temperature resistance based on actinides.

Table 2. Comparison of ^{238}Pu and ^{241}Am.

	Pu-238	Am-241
Chemical form:	PuO_2	AmO_2
Half life:	87.7 y	432.2 y
Specific decay power (oxide):	0.493 W/g	0.101 W/g
Specific weight (oxide at 95% TD):	10.89 g/cm³	11.10 g/cm³
Thermal conductivity at 1500 K [7]:	~ 2.8 W/m K	~ 2.8 W/m K (~ 1 W/m K for Am_2O_3)

The disadvantage of ^{241}Am low power density, compared to ^{238}Pu, is accompanied by the advantage of

longer half-life, which makes it the ideal power providing isotope for very long lasting missions into the outer region of the solar system or even into the interstellar space and the Oort cloud. Fig. 2 shows the decrease of thermal power of ^{241}Am compared to ^{238}Pu over an period of 200 years. While the plutonium has decayed to circa 20% of the power at BOM, the americium source is still producing about 80% of the initial power. However, it should be noted that a large fraction of the power decrease of an RPS comes from the degeneration of the employed thermoelectric material. Therefore, the exploratory research project is looking into very long-term stability of potential americium compounds as well as alternative actinide based thermo-electric materials.

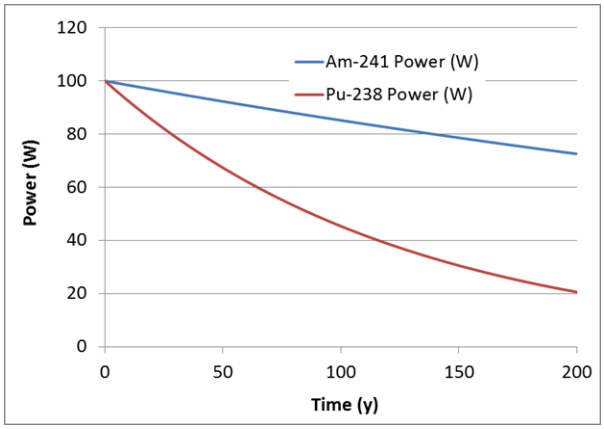

Figure 2. Decrease of ^{241}Am and ^{238}Pu power over 200 y

Thermal conductivity: The thermal conductivity of americium dioxide was found to be slightly lower compared to plutonium dioxide [7]. However, it should be noted that it decreases dramatically with deviation from stoichiometry and the sesquioxide shows a very low thermal conductivity (~ 1 W/m K at 1500 K, see Tab. 2). Since surface temperature, power density and thermal conductivity determine the central pellet temperature during operation, a low thermal conductivity can represent a limiting factor for a desired hot leg temperature of the thermocouples. In addition, a significantly increased thermal conductivity can increase the design flexibility of the RPS and enable relevant space and weight savings. Therefore, we will also investigate alternative americium compounds, and composites, with respect to their thermal properties (including thermal conductivity) at high temperatures.

3. THE MINOR ACTINIDE LABORATORY AT JRC-ITU

Due to strong gamma radiation, working with ^{241}Am can result in high dose rates for the operating personnel. Therefore, remote operated and shielded equipment is advantageous or even necessary in order to prepare americium based pellets for RPS. JRC-ITU possesses a unique infrastructure for handling of highly radiative actinide materials, the so called Minor Actinide Laboratory (MA-Lab) [8]. It is of high importance for safety research on Transmutation fuel 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 test pins for irradiation experiments.

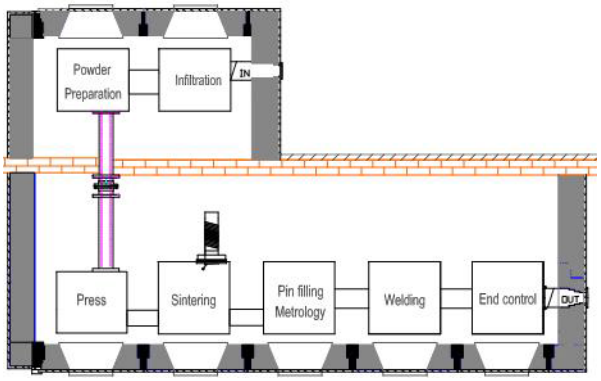

Figure 3. Minor Actinide Laboratory at JRC-ITU [8].

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 Fig. 3. The glove boxes are shielded by 50 cm water 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 ^{241}Am or 5 g of ^{244}Cm. The glove boxes can be accessed from behind manually if radiation levels are low, for 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 [9] and the porous bead infiltration technique [10]. 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.

4. CONCLUSIONS

The European Commission's Joint Research Centre has launched a new exploratory research project in order to find innovative solutions and to improve safety and long-term reliability of ^{241}Am based power sources for future ESA missions into deep space. The project utilizes the unique facilities of JRC for preparing, handling and characterising highly radiating americium compounds. It investigates particular aspects of americium oxide, such as its chemical and self-irradiation stability and looks into promising alternative candidate compounds for americium and new thermo-electric materials containing actinides.

5. REFERENCES

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