

Optimizing Pu-238 Production in Europe

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Abstract

The “Optimus Pro” project explores different ways to achieve a reasonable production rate of the radioisotope Pu-238 in Europe, making use of European infrastructure at an acceptable cost. A prior ESA funded activity showed that it was technically possible to produce about 300g of high quality Pu-238 per annum within Europe at a cost per Watt comparable to the expected cost of Am-241 production. During this project, previous conclusions are further derisked by trying to increase production rate, increase diversification and making the process as efficient as possible. At the same time, some technical gaps are being filled in order to have full confidence on the feasibility. The final objective is to establish a comprehensive roadmap for Pu-238 production from the irradiation of Np-237 targets, including the optimization both in terms of cost and planning of the related production process within the European industrial framework.

1. Introduction

The “Optimus Pro” project, initiated under ESA Contract No. 4000142904/23/NL/KK, is a continuation of the previous Contract No. 4000135477/21/NL/GLC. Both projects were led by Tractebel (Belgium) with SCK-CEN (Belgium) as subcontractor. In the previous contract, Orano (France) also participated as subcontractor. It explores the feasibility of a European Pu-238 production process.

Space missions operating in the inner solar system usually rely on photovoltaic solar cells as primary power source. However, these photovoltaic cells are not appropriate for all space missions, either because the solar radiation is too weak to produce sufficient power within the limitations imposed by today’s solar cell technology, spacecraft mass and volume limitations, or because the timespan to be bridged by the batteries is too long (e.g. lunar, Martian nights, long dust storms, etc.).

Therefore, nuclear power systems have been deployed for heat (Radioisotope Heater Unit or RHU) and electrical power generation (Radioisotope Thermoelectrical Generator or RTG). Both systems rely on radioactive decay as the primary energy source.

Preferred radioisotopes considered by ESA are Americium-241 (Am-241) and Plutonium-238 (Pu-238). Both isotopes are mainly produced in nuclear reactors today, or can be found in spent nuclear fuel following radioactive decay of parent isotopes. Currently, Am-241 has a greater availability in Europe and can be extracted without a further irradiation step, but it has a four to five times lower power density with respect to the Pu-238 isotope. Furthermore, Pu-238 is easier to handle as it emits lower radiation as compared to Am-241 and its oxide is very stable in time and irradiation conditions. Moreover, Pu-238 has a convenient half-life of 87.7 years, which is largely compatible with currently envisaged space missions.

The production of Pu-238 is complicated since it is not extractable as a single isotope. Instead, the production will proceed via neutron irradiation of Neptunium-237 (Np-237), which is created as a by-product in nuclear fission reactors and is currently located in the waste stream of the PUREX process in the La Hague nuclear fuel reprocessing facility in France.

The “Optimus Pro” project explores different ways to achieve a reasonable production rate of Pu-238 in Europe, making use of European infrastructure (irradiation facilities, nuclear workshops, etc.) at an acceptable cost. The

previous project showed that it was technically possible to produce about 300g per year of high quality Pu-238 within Europe at a cost per Watt comparable to the expected cost of Am-241 power. The previous conclusions are further derisked by trying to increase production rate, increase diversification and making the process as efficient as possible. At the same time, some technical gaps are being filled in order to have full confidence on the feasibility.

The final objective is to establish a comprehensive roadmap for Pu-238 production from the irradiation of Np-237 targets including the optimization both in terms of cost and planning of the related production process within the European industrial framework.

2. Production Process

2.1 Source of Pu-238

Even though Pu-238 is present in irradiated nuclear fuel today, its relative concentration with respect to the other plutonium isotopes is very small (maximum a few percentages), whereas it is generally required by the technical specifications of the RTG and RHU to have a relative concentration of at least 82.5% of Pu-238 [1].

Therefore, the production of Pu-238 will proceed via neutron irradiation of neptunium-237 (Np-237), which is also created as a by-product of spent fuel in nuclear fission reactors and is currently found in the waste stream of the PUREX process in the La Hague (France) fuel reprocessing facility, owned and operated by Orano. The PUREX process recovers and purifies uranium and plutonium from irradiated uranium fuel in a continuous solvent extraction process. Bombarding Np-237 atoms with neutrons in a nuclear reactor will temporarily produce Np-238, which will decay to Pu-238 in a few days.

2.2 Neptunium Extraction

In the previous project, Orano showed that it was feasible to extract sufficient quantities of a regular neptunium-rich (predominantly the isotope Np-237 with the longest half-life) waste stream from the La Hague reprocessing facility. However, at the time, it was noted that reaching sufficient quantities would require some modifications to the existing installation.

The extracted waste stream, which is a nitrate solution, will subsequently be purified from remaining fission products. The purification is done by selective ion-exchanging resins and the neptunium is retrieved by elution. The optimal parameters will have to be identified in further studies, determining the final volume and concentration of the solution. For the current and previous study, the reference hypothesis was a concentration of the neptunium nitrate solution of approximately 1.35g neptunium per liter, respecting purification requirements prescribed by the further processes at SCK-CEN.

2.3 Neptunium Transport

In the reference case, the purified neptunium nitrate solution will then be transported from La Hague to the SCK-CEN site in Mol (Belgium) in a dedicated transport cask, which is still to be designed, licensed and manufactured. It is anticipated that such a cask will contain about 100 L of solution.

It should be noted that the regulatory framework for an international transport of neptunium does not exist yet. But as it concerns limited volumes, it is not expected to become an obstacle for the feasibility of the production.

2.4 Neptunium Target Production

At the site of SCK-CEN, as foreseen in the reference case, the front-end operation starts from the neptunium nitrate solution provided by Orano and the neptunium nitrate recycled from subsequent process steps (see §2.6).

The first decay product of Np-237 is protactinium-233 (Pa-233), and is responsible for highly energetic gamma radiation that is a radiological concern for handling the material. Even in initially pure Np-237, after several weeks a secular equilibrium between mother and daughter product is attained. In order to ease the handling of neptunium and reduce the shielding requirements, a separation of the decay product Pa-233 prior to the conversion to neptunium oxide (NpO₂) powder is considered. This operation will temporarily reduce the gamma radiation received from the neptunium

nitrate solution, to a level that allows handling of the neptunium in the target production stage, without excessive shielding.

NpO₂ targets are then designed with a goal for maximizing Pu-238 production, and minimizing peak heat generation rates from local fissions. The considered target design for irradiation in a nuclear reactor is a stack of NpO₂ full-ceramic pellets, clad in a zirconium alloy (Zircaloy), similar to a uranium fuel rod in a nuclear reactor.

2.5 Target Irradiation

In the reference case, the irradiation of the targets in the high neutron flux material test reactor BR2 on the site of SCK-CEN was assumed, even though other European reactors could also be considered for the future actual production. An irradiation campaign in BR2 consists of irradiation cycles with a typical length of about 28 days, and inter-cycle lengths of 28 days for unloading, reloading or reshuffling.

When Np-237 is subject to a neutron flux, in a nuclear reactor, the capture of a neutron creates Np-238, which decays further to Pu-238 with a half-life of approximately 2 days. However, this process has some drawbacks, as competing reactions are unavoidable. The intermediate product Np-238 has a high fission cross-section, risking being broken up into fission fragments before its decay. The Np-238 can also capture a neutron, without causing fission, creating Np-239 which decays to give Pu-239 – which is an unwanted fissile material in this production process. Apart from these two unwanted reactions occurring at relatively high probability, a lot of other reactions can inevitably occur, resulting in unwanted isotopes in the irradiated target.

These adverse effects could be mitigated to some extent by choosing “soft spectrum” reflector positions. It could be beneficial to use a neutron moderating material to thermalize (i.e. slow down) fast neutrons before reaching the target material, achieving a high cross-section of neutron absorption in Np-237, and decreasing the amount of fission products and Pu-236 production. The Pu-236 isotope is responsible for very heavy gamma radiation from the decay of its daughter product thallium-208 (Tl-208) and its concentration is generally limited to 2-3 ppm.

It was calculated that three irradiation cycles would lead to an optimum compromise between a higher amount of plutonium bred on the one hand and a higher quality of the resulting plutonium vector (i.e. the relative concentration of Pu-238 with respect to other plutonium isotopes) on the other hand.

Afterwards, the irradiated targets will need to be cooled for 12-24 months in the reactor pool for fission product decay and for reduction of Pu-236 concentration. Given the half-life of Pu-236 being approximately 2.8 years, this cooling period significantly reduces its presence.

2.6 Irradiated Target Processing

The cooled irradiated targets are subsequently dissolved in nitric acid and the resulting nitrate solution, containing neptunium, plutonium and some fission products, is separated by solvent extraction techniques.

The neptunium stream is purified and reused in the front-end for target fabrication, while the waste stream is stored and evacuated as high activity waste, in a limited volume. It should be noted that because the process source material is nuclear waste already, no additional high activity nuclear waste is created.

The plutonium nitrate solution is then purified and further converted into plutonium oxide (PuO₂) powder, containing approximately 85% of Pu-238. The powder is milled, pressed and sintered into cylindrical pellets which are encapsulated in an iridium based alloy, pending transportation as a final product for use in RTG or RHU.

3. Process Optimisation

The proposed production process described above, resulted from the previous study and demonstrated the technical and economic feasibility of producing Pu-238 in Europe. The current “Optimus Pro” project builds on this foundation by exploring the process in greater depth, addressing some remaining technical gaps and identifying opportunities to enhance efficiency, and reduce ramping-up time and costs.

3.1 Irradiation Facilities

The reference case is the high-flux research reactor BR2 in Mol Belgium, on the site of SCK-CEN. Even though, from a technical point of view, this is an ideal facility to perform the job, alternative facilities are sought for multiple reasons:

- Due to the intensive use of this reactor for medical and material test irradiations, the availability of channels for irradiation of the neptunium targets is limited, potentially reducing the production rate of Pu-238.
- Reactor lifetime of BR2 will not be unlimited, as it operates under open-ended decennial license renewals.
- The irradiation cost, though not fully confirmed yet, is the preponderant cost in the entire Pu-238 production process.
- Using more than one irradiation facility will reduce production risks from unplanned reactor unavailability.

3.1.1 TRIGA II Pitești

A TRIGA reactor is a pool type reactor designed for research and testing, as well as isotope production. It uses a uranium zirconium hydride fuel having a large prompt negative fuel temperature reactivity coefficient, making it an exceptionally safe reactor. There are many TRIGA reactors across the world, most of them commissioned back in the 60s and 70s.

One such TRIGA nuclear facility is located in Pitești, Romania, comprising two reactors: a TRIGA steady state reactor (SSR) of 14 MW, and a TRIGA annular core pulsed reactor (ACPR), commissioned in 1980 and operated by the Institute for Nuclear Research.

According to [2], the flux levels in the TRIGA SSR reactor seem equivalent as for the BR2 reactor, making this reactor a suitable candidate for industrial production. Furthermore, the reactor core structure is designed in a flexible way to allow for a variety of irradiation devices. However, the latest consultation of the IAEA Research Reactor Database indicates that decommissioning of the reactor is scheduled for 2035. If this timing remains unaltered, this reactor will not be a long-term alternative for the production of Pu-238 within Europe.

3.1.2 Maria

The Maria research reactor, situated at the National Center for Nuclear Research (NCBJ) in Świerk-Otwock in Poland, was commissioned in 1974. This pool type reactor operates at a power level of 30 MW, offering neutron flux levels comparable to those of BR2.

Currently, NCBJ is conducting a separate study to explore the opportunities for the production of Pu-238 in the Maria reactor.

In June 2023, the Council of Ministers of Poland adopted a resolution on the Maria reactor modernisation program, to enable its operation beyond 2027. The upgrades aim to extend the reactor's operational life until at least 2050. The Maria reactor's extended lifespan and advantageous neutron flux levels, position it as an excellent choice for the industrial production of Pu-238 in Europe. Additionally, as the reactor is located within the premises of the National Center for Nuclear Research, both front-end and back-end infrastructure could be considered at the same location, streamlining the production process and ensuring efficient management of resources.

3.1.3 Jules Horowitz Reactor (JHR)

The Jules Horowitz Reactor, owned and operated by the French Atomic and Alternatives Energies Commission (CEA), is a material test and isotope production reactor currently under construction at Cadarache in southern France with a reactor power of 70 MW and a high thermal neutron flux. The reactor set-up and its neutron flux level would be suitable for Pu-238 production.

Based on the current construction schedule, the reactor would be available for commercial tests from 2034, which makes it a good alternative for Pu-238 production. Furthermore, its location on one of the largest European nuclear research and development centres, allows the combination with front-end and back-end activities on that site.

3.1.4 Pallas

The NRG Pallas research reactor is currently being built at Petten (the Netherlands) to replace the existing High Flux Reactor (HFR) by the early 2030s. Limited technical information is available yet, but it appears that the neutron flux level and spectrum would be suitable for Pu-238 production.

3.1.5 Pressurized Water Reactor

A Pressurized Water Reactor (PWR) is the most common power production nuclear power plant in the world, and in Europe. In a PWR, light-water is used as both a coolant and a moderator. It operates on fixed cycle lengths of 12 to 18 months, not allowing any fuel manipulation in between two outages.

Target insertion for irradiation could be considered in the guide tubes of the fuel assemblies, for instance by attaching them as rodlets to the thimble plug devices. Other locations are not available in such a compact reactor as a PWR.

The proximity of the targets to the fuel rods will result in a relatively high fast flux component, creating very high unwanted Pu-236 concentrations (more than 10 ppm). Addition of local moderation around the targets to reduce Pu-236 concentration will be difficult, because the diameter of the inner guide tube is small (typically about 1 cm). This will rule out the option of using PWRs for Pu-238 production, unless the limit for allowable Pu-236 can be increased by a factor of 5 or more.

Another issue linked to the use of PWRs for isotope production, is the fixed cycle lengths. The targets will only be able to be extracted during outages, when the vessel is opened, which doesn't allow optimisation of the irradiation duration.

3.1.6 CANDU Reactor

A CANDU reactor is a Canadian designed pressurized heavy-water reactor for electric power production. In Europe, the only CANDU reactors are located in Cernavodă in Romania, namely units 1 and 2, commissioned in respectively 1996 and 2007. The construction of three other CANDU units in Romania has never been completed, but is been considered to resume.

The higher flexibility of a CANDU reactor, with respect to a typical PWR light-water reactor, makes them suitable for isotope irradiation as has been done successfully in Canadian CANDU units for the production of Co-60 (in Pickering and Bruce B plants). There is also an ongoing project for harvesting the medical isotope Mo-99 from the Darlington plant. Recently, in the Bruce plant, the medical isotope Lu-177 has been produced.

There are two options for irradiating isotopes in a CANDU reactor:

- The target could replace a fuel rod within a fuel assembly. The fuel management approach of a CANDU reactor allows for flexible online fuel assembly extraction, not limited to cycle lengths as with typical PWR reactors. The target will need to be removed from the used fuel assembly, which will have an impact on the back-end cost. Furthermore, due to the less moderated flux spectrum within the fuel, the production of the unwanted isotope Pu-236 will be too high.
- The target could be inserted in adjuster rod tubes or in other available positions inside the moderator. The better moderated flux spectrum allows to achieve high quality Pu-238. Even though the neutron flux level is lower compared to BR2, the achievable Pu yield can be significantly higher. In addition, since the moderator-to-fuel ratio of a CANDU is higher than for a PWR, there is more space available for modifications.

The cost of irradiation in a commercial CANDU reactor might be considerably lower, since these reactors need to produce power anyhow. This makes irradiation in a CANDU reactor an attractive alternative, even though it will not necessarily be straightforward to reconcile the commercial operation of a power plant with isotope production needs.

The downside of this option is that front-end and back-end activities will most probably not be compatible with a commercial power production site, requiring the transportation of new and irradiated targets to and from the power plant for other stages of the Pu-238 production process.

3.2 Flow of Materials

3.2.1 Assumptions

Due to the relatively small volumes concerned and due to the nature of the target irradiation, the production process will be performed in a batch-wise manner. For optimizing the production process, and understanding the impact of some options, the previous analysis [3] was used as starting point.

In the old reference scenario, yearly production of the plutonium powder, rich in isotope Pu-238, is calculated taking into account the following assumptions and simplifications:

- The yearly production rate of plutonium is aimed at about 300g, which was based on a rough estimate from SCK-CEN taking into account a typical availability of their irradiation channels in the BR2 reactor.
- The delivery of fresh neptunium is performed at a rate of 1.08kg of neptunium per year, which corresponds to 8 batches of 100 L neptunium nitrate solution at a concentration of 1.35g Np/L, in line with a preliminary feasibility study performed by Orano. Once reaching the aimed yearly plutonium production rate of 300g, the delivery of fresh neptunium is reduced to 3 batches per year.
- The yield of the NpO₂ pellet fabrication is very conservatively estimated at 50%, in the absence of a better estimate. This means that in each target fabrication campaign, only 50% of the available Np material is actually converted to a target, the remainder being available for future campaigns.
- Delays in the target preparation and eventual transportation are not taken into account, but this will be covered by the batch-wise nature of the process, since at most, targets can be inserted in the BR2 reactor once every two months.
- The irradiation of the neptunium targets is performed at BR2, during three cycles of 28 days, with 28 days intercycle. The resulting conversion ratio of neptunium into plutonium is about 6% with an isotopic purity of at least 85% Pu-238.
- The cooling time of the irradiated targets is fixed at 12 months on top of the last intercycle period of 28 days.
- Delays in the separation process of the irradiated targets, powder production, pelletization, encapsulation and transportations are not considered. They would only have a small impact on the total production process, delaying the availability of the Pu by a few months, but not impacting the production rate itself.
- The yield of the PuO₂ pellet fabrication is very conservatively estimated at 50%, in the absence of a better estimate. This means that in each plutonium pellet fabrication campaign, only 50% of the available Pu material is actually converted to a pellet, the remainder being available for future pressing campaigns.
- No process losses of the actinides Np and Pu are considered in this study, even though in reality small process losses will be inevitable.

It is worthwhile to indicate that minimizing neptunium loss in the separation process of the irradiated targets is important. In PUREX technology application, extraction efficiencies of Pu and U can reach very high values, higher than 99.8% [4]. Similar order of magnitudes might be desirable for the separation of Np and Pu. Since the increase and availability of a sufficient amount of neptunium material to irradiate is ensured by multiple recycling of the neptunium material, lower efficiencies will impact the output of the process due to their cumulative nature.

3.2.2 Reference Scenario

The yearly production of plutonium for space applications is shown in Figure 1.

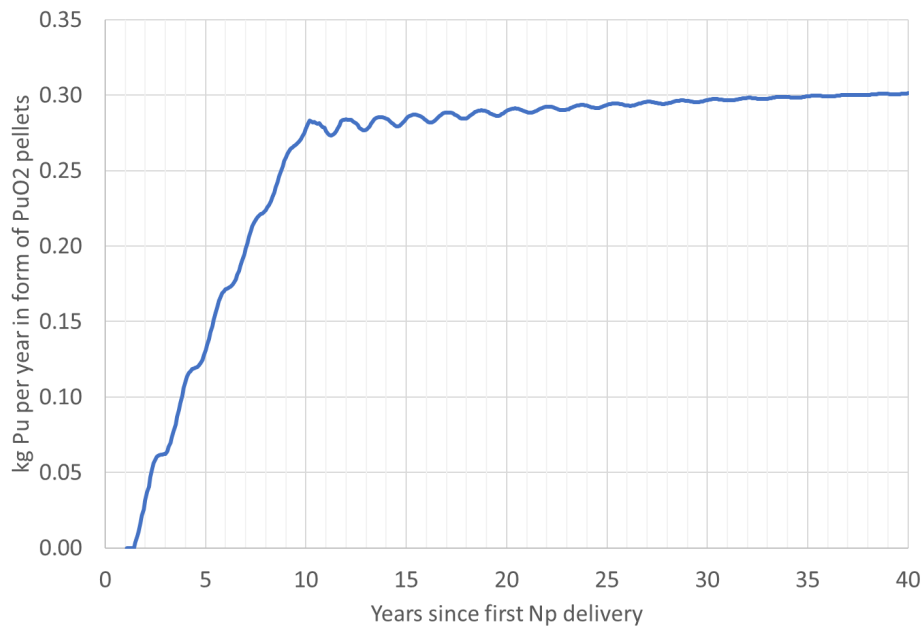


Figure 1: Yearly production of Pu in reference scenario

The cumulative production of plutonium (left axis), together with the cumulative consumption of neptunium (right axis) is shown in Figure 2.

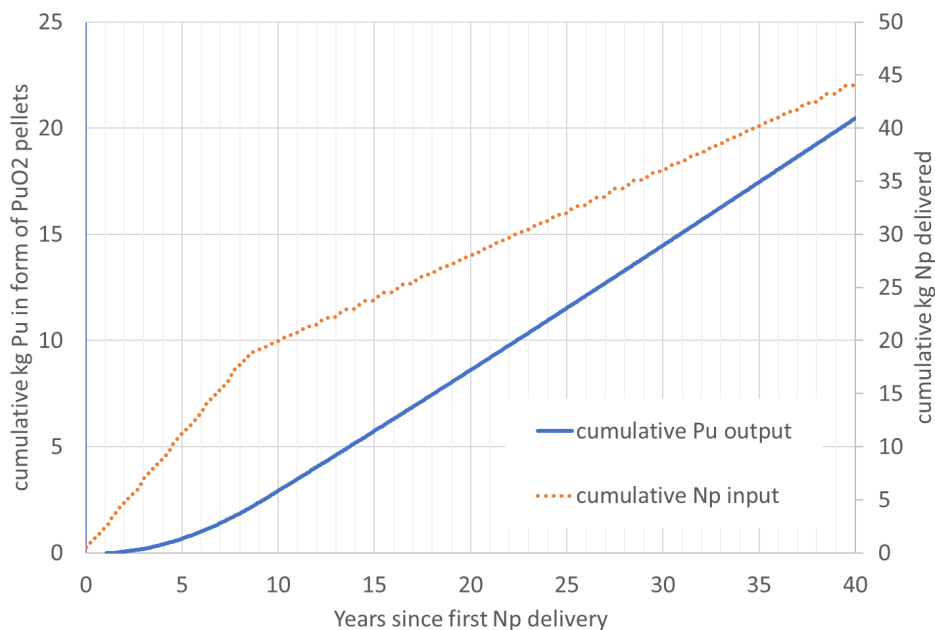


Figure 2: Cumulative production of Pu and consumption of Np in reference scenario.

There are several concerns for this reference scenario, which are the reason for optimization:

- As explained in the previous study [3], the slow ramping-up of the production rate is mainly due to the low Np delivery rate, as well as to the relatively long cooling time of irradiated targets. This cooling time of 1 year is already considered challenging, but the Np delivery rate is something that can be improved.
- Having more than one irradiation facility available will reduce the risks due to temporary unavailability of an irradiation facility (due to maintenance for instance) or to temporary unavailability of irradiation channels as they may be required for medical isotopes or other purposes. It will also more easily allow for an increase of yearly production rate.

3.2.3 Doubling the Neptunium Input

A doubling of the yearly input in neptunium material could be considered, via an increase in concentration (2.70gNp/L instead of 1.35gNp/L) or via an increase of the number of yearly deliveries (16 deliveries per year instead of 8 deliveries per year).

The impact on the production process is a reduction of about 5 years in production ramping-up allowing an earlier availability of the plutonium material. Likewise, the same cumulated amount of plutonium as in the reference case, is obtained about 5 years earlier.

The total gain in production, when looking at the entire plant production, is minimal since the cumulated production is mainly dictated by the maximum yearly production rate of 300g Pu. Increasing the neptunium input will be imperative, however, when considering an increase of the aimed yearly production rate (be it in one single irradiation facility or in a combination of irradiation facilities).

3.2.4 Diversifying Irradiation Facilities

As explained above, diversifying irradiation facilities is a way to increase and derisk production capacity, provided that the neptunium delivery can keep pace with the increased demand. However, diversifying front-end and back-end facilities would not make sense from an economical point of view since it would raise the CAPEX for constructing similar facilities at different locations, not benefitting from the production cost reduction that goes along with a production rate increase.

Therefore, if multiple irradiation facilities will be used, transport cost and timing have to be added to the overall process. This includes transporting neptunium targets from the front-end workshop to the irradiation facility, irradiated targets from the irradiation facility to the back-end workshop and separated untransformed neptunium back to the front-end workshop. Initial evaluations indicate that the total cost increase of the process is minimal with respect to the other costs, as long as the irradiation facilities are situated within mainland Europe, not requiring any maritime transport. Moreover, since the irradiation costs constitute the predominant OPEX, more savings could be achieved by negotiating favourable irradiation price with reactor owners.

3. Conclusions

Though the Optimus Pro project is currently ongoing, the following preliminary conclusions can be drawn:

- So far, there are no significant obstacles that would prevent the production of Pu-238 in Europe.
- Due to the preparation time for constructing and licensing the new facilities and due to the long ramping-up time in production, an early project decision is to be made if ESA would like to use Pu-238 for missions in the next decade.
- Diversifying irradiation facilities will be essential to reduce costs and mitigate risks associated with production.
- Several irradiation alternatives have been identified within Europe, giving confidence that European infrastructure can cope with project goals.
- The unwanted production of Pu-236 and its concentration limit have constrained the potential production methods and reactors. However, increasing the limit in the specifications could open up new alternatives.

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