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POSTER:
NON-POWER INDUSTRIAL
APPLICATIONS

A EUROPEAN RADIOISOTOPE PRODUCTION FACILITY FOR POWER SOURCES IN SPACE

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ABSTRACT

Electrical power sources used in outer planet space missions are a key enabling technology for data acquisition and communications in situations where solar power is inconvenient or inefficient. Power sources currently in use (e.g. Voyager, Cassini, New Horizons) generate electricity from the thermal energy, generated by alpha decay of the radioisotope ^{238}Pu , via thermoelectric conversion. Production of ^{238}Pu requires specialist facilities including a nuclear reactor and reprocessing plants that are expensive to build and operate, so naturally, a more economical alternative is attractive to the industry. With the objective of obtaining a European based independent supply of power sources the European Space Agency (ESA) has commissioned a programme to establish the skills and capability across Europe to fulfil this goal.

Within Europe the radioisotope ^{241}Am is considered a feasible alternative to ^{238}Pu and can provide a heat source for small scale radioisotope thermoelectric generators (RTGs) and radioisotope heating units (RHUs). The ^{241}Am exists in an isotopically pure state within stored civil plutonium on reprocessing sites within the UK and France. A study is underway to design a process that will chemically separate ^{241}Am and at the same time provide a plutonium product that is easier to handle for recycling into new nuclear fuel. This paper will present the process and design of a facility for ^{241}Am production within the UK National Nuclear Laboratory.

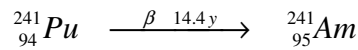
1. Introduction

Plutonium-238 (^{238}Pu) has been used as a power source since the early days of space exploration. It has proven to be an effective source of electrical power where the use of solar generated power is impractical. Its application for use in spacecraft power systems was developed by the USA and the Russian Federation who have spent many \$100s of millions on its development and production. However, for many years ^{238}Pu production ceased and existing worldwide stockpiles have reduced to levels sufficient for only one or two further missions [1]. The USA is currently preparing to restart ^{238}Pu production using the High Flux Isotope Reactor (HFIR) and the Advanced Test Reactor (ATR).

Historically, Europe has relied on collaborations with the USA or Russia to access these nuclear power sources. During 2009, the European Space Agency (ESA) funded a project to examine the cost and practicality of establishing a European sourced material suitable for Radioisotope Power Systems (RPS). The study concluded that a small number of radioisotopes could be used in RPSs and on further review two leading candidates, ^{241}Am and ^{238}Pu , were selected for additional study. Whilst ^{238}Pu was recognized as the isotope of choice for use in RPSs, the costs of setting up a European based production facility was prohibitively expensive compared to ^{241}Am production. If the limitations associated with the lower power output of ^{241}Am can be overcome, or their consequences accepted, then the much reduced production costs make ^{241}Am an attractive option for European based radioisotope power sources.

1.1 Background

Despite its lower power density of $\sim 0.11 \text{ W}_{\text{th}}/\text{g}$, equivalent to around one fifth that of ^{238}Pu , ^{241}Am is viewed as a potential alternative because of its availability in the nuclear fuel cycle in an isotopically pure form. During reprocessing the plutonium in spent nuclear fuel is separated as a chemically pure PuO_2 product containing around 3-11% w/w $^{241}\text{Pu}/\text{Pu}$ isotope and placed into storage. During storage the ^{241}Pu isotope beta decays to ^{241}Am with a half-life of 14.4 years.



In considering the practicality of obtaining sufficient stocks of ^{241}Am or ^{238}Pu radioisotopes for use in RPSs, investment in facilities is required regardless of the chosen radioisotope. However the USA and European positions are very different in this respect. Whereas the USA has existing feed stocks and facilities available for use in ^{238}Pu production, Europe does not. The salient issues are summarised in Table 1.

	^{241}Am Europe	^{238}Pu Europe	^{241}Am USA	^{238}Pu USA
Feed source	Large existing stock of ^{241}Am	Limited stock of ^{237}Np	Limited stock of ^{241}Am -bearing waste	Large existing stock of ^{237}Np
Irradiation of feed	Not required	Limited choice of high flux reactor	Not required	Existing high flux test reactors
Separation of isotope	Conversion of existing facility	New facility required (hot cell)	New facility required	Conversion of existing facility
RPS production	New facility required	New facility required	Existing facility	Existing facility
Flight qualification	Qualification required	Qualification required	Qualification required	Existing qualification in place

Table 1: A comparison of RPS radioisotope manufacturing requirements in the USA and Europe

1.2 Production Requirements

The ESA roadmap [2] for americium fuelled power sources requires a supply of appropriate quantities of separated ^{241}Am . The National Nuclear Laboratory has assessed the feasibility and costs associated with installing a European Radioisotope Production process within its existing facilities at Sellafield.

The existing stocks at Sellafield are more than sufficient for the production of multiple ^{241}Am -based RPSs each year for a number of years provided it can be separated from the plutonium in a cost effective manner. There are substantial advantages in converting existing NNL facilities compared to a new build and these include:

- Facilities on an existing nuclear licensed site
- A license to handle the required amounts of plutonium (kgs)
- Access to PuO_2 stored at the UK Sellafield site
- Existing supporting infrastructure (e.g. security, waste effluent routes etc.)
- More than £250M worth of building infrastructure already built to support plutonium operations.

The processing facility is planned to be housed within the NNL's Central Laboratory in an existing area designed for plutonium active operations. The scope of the overall project is "store to store" i.e. the starting point is aged plutonium currently housed in existing Sellafield stores and the end point is the storage of separated plutonium and americium oxide powders in similar stores. The feedstock for the process is aged Magnox reactor derived plutonium dioxide and will be contained in standard packages,

currently stored on the Sellafield site. The minimum ^{241}Am content of the feed will be 3.5% (w/w HM) and the maximum 4.8% (w/w HM). The required product is americium oxide powder in a package suitable for temporary storage pending fabrication into RPSs. The design basis throughput for the process is 240kg HM/year (i.e. 240kg of aged Pu with Am in-growth). The capability is to be designed for a 15 year operating lifetime, producing on average 8.4kg Am/year.

2. The Process

The conceptual flowsheet for the process of separation and buffer storage of ^{241}Am has been developed:

- Removal of Pu cans from the store at Sellafield
- Transport of the cans to the Central Laboratory import facility
- Dispensing the plutonium
- Ag^{2+} catalytic dissolution in nitric acid
- Am/Pu separation using solvent extraction
- Am/Ag separation using solvent extraction
- Formation (Oxalate precipitation) and packing of a plutonium oxide product
- Formation (Oxalate precipitation) and packing of an americium oxide product
- Ag and solvents recycled
- Storage of the americium oxide product
- Returning purified plutonium oxide back to Sellafield stores

Current work is now focused on underpinning the conceptual flowsheet, using aged plutonium in NNL's PuMA laboratory.

2.1 Plutonium dioxide (PuO_2) dissolution

For many years there has been an interest in the dissolution of plutonium dioxide to recover plutonium from wastes and scraps. The dissolution of PuO_2 in nitric acid is difficult and hence the method of choice for dissolution of PuO_2 and MOx residues is through the use of strong oxidants such as Ce^{4+} or Ag^{2+} . The advantages of these oxidants over others are that they are effective at catalytic concentrations and can be regenerated by electrochemical means. To date only the Ag^{2+} process has been demonstrated on an industrial scale.

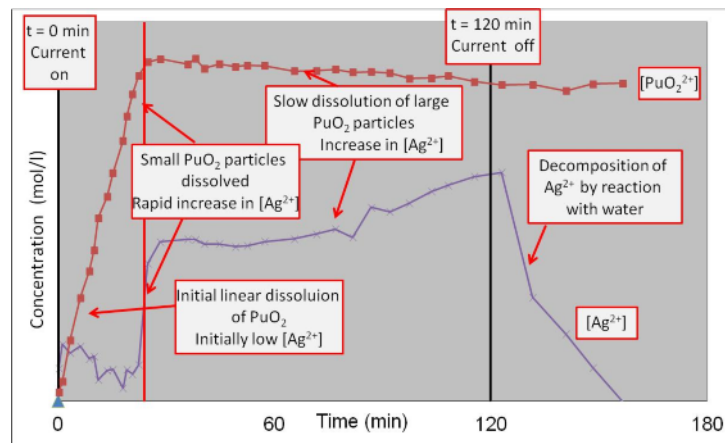


Figure 1. (left) Lab scale PuO_2 dissolver; (right) A typical $[\text{Pu}]$ vs time plot (final $[\text{Pu}] = 100\text{g/l}$)

2.2 Separating plutonium from americium and silver

Once dissolution of the americium containing plutonium has taken place, separation and purification is required. In this process to recover Am there are two solvent systems: TBP/OK to extract Pu and a second solvent system to extract Am from silver impurities. There are, therefore, a number of Decontamination Factors that need to be underpinned to validate the flowsheet; a) rejection of silver from TBP/OK solvent; b) rejection of silver from the second solvent system; c) Extraction of ^{241}Am into the second solvent system; d) effective backwashing of the ^{241}Am from this solvent and; e) effective solvent washing using oxalate to minimise the activity levels in recycled solvent.

The solvent system for Ag,Am/Pu separation is 30% v/v tributylphosphate (TBP) odourless kerosene (OK) mix. This solvent is used on an industrial scale to recover U and Pu from irradiated fuel (PUREX process). A TBP value of 30% has been chosen to cope with the reasonably high metal loading (~30 gPu/l) required during extraction. The Am/Pu separation process will extract the Pu⁴⁺ leaving Am³⁺ and Ag⁺ in the aqueous raffinate. The solvent will be scrubbed with acid before backwashing the plutonium. The raffinate from the Am/Pu separation step is expected to contain 1-3 g/L americium and 0.02-0.1 M silver at ~ 2-3 M HNO₃.

2.2 Separating americium from silver

Purification of the americium stream from silver before precipitation with oxalate is essential because silver oxalate has low solubility ($[Ag] \sim 2.2 \times 10^{-4}M$; $K_{sp} = 5.4 \times 10^{-12}$) and co-precipitates with americium oxalate. This can be achieved by either solvent extraction, selective precipitation, ion exchange chromatography or some other technique; but solvent extraction has been chosen because:

- It is an automated continuous process that requires little hands on intervention, which will be important in keeping the operator dose as low as practicable.
- The small footprint and height of equipment is suitable for glovebox operations.
- The separation factors that can be achieved are much higher than other separation techniques.
- Solid and liquid waste volumes are low if the solvent can be recycled.

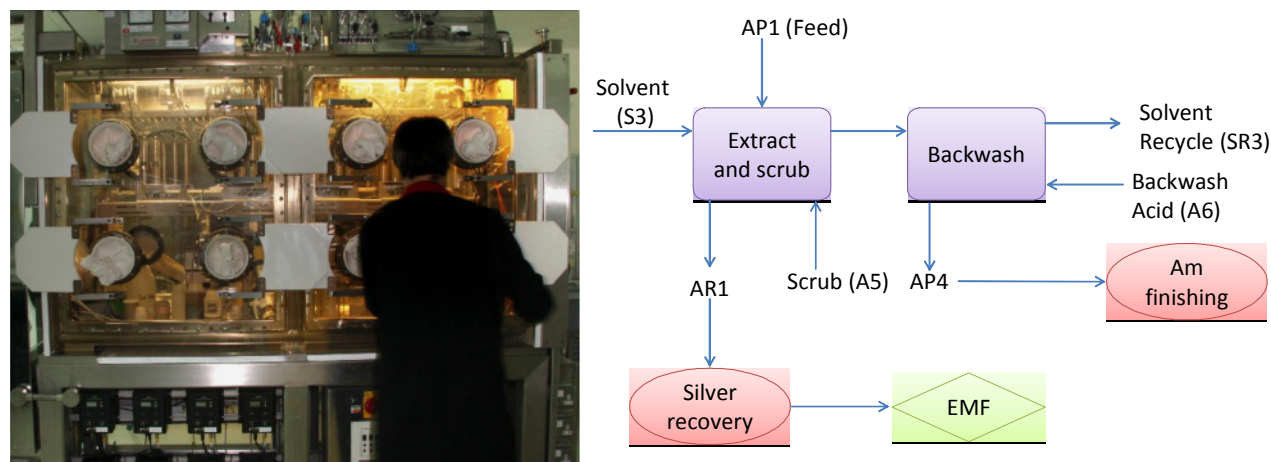


Figure 2. (left) centrifugal contactor rig within the NNL Plutonium and Minor Actinides (PuMA) Laboratory; (right) the flowsheet for separating americium from silver.

In the full scale facility the baseline technology for obtaining a finished americium product will comprise of americium oxalate precipitation probably followed by washing and filtering with a rotary vacuum filter (RVF), drying and calcining. The drier removes excess water and the calciner decomposes the oxalate to an oxide product.

3. Plant design

The processing of americium, with its associated gamma and neutron hazards, requires the use of containment to prevent internal dose to operators, shielding to minimise the external dose to operators, and remote or automatic operations (for example using MSMs) to minimise the contact dose to operators. In addition, the presence of fissile and fissionable materials requires strict criticality control to ensure a failsafe process.

The glove boxes where americium-rich streams are processed will have the operating face fitted with pairs of MSMs and will have gloves on the reverse side of the glove box for maintenance operations. The intention is that the MSMs and automatic equipment inside the glove box will be used for routine operations and the gloves will be used for maintenance operations, where increased dexterity is required, once the dose rate inside the glove box has been proven to be sufficiently low.

A conceptual design of the full scale facility has been established and loaded into a 3D emersion suite to allow visualisation of the plant layout with supporting utilities and the required nuclear safety mechanisms in place (Figure 3).

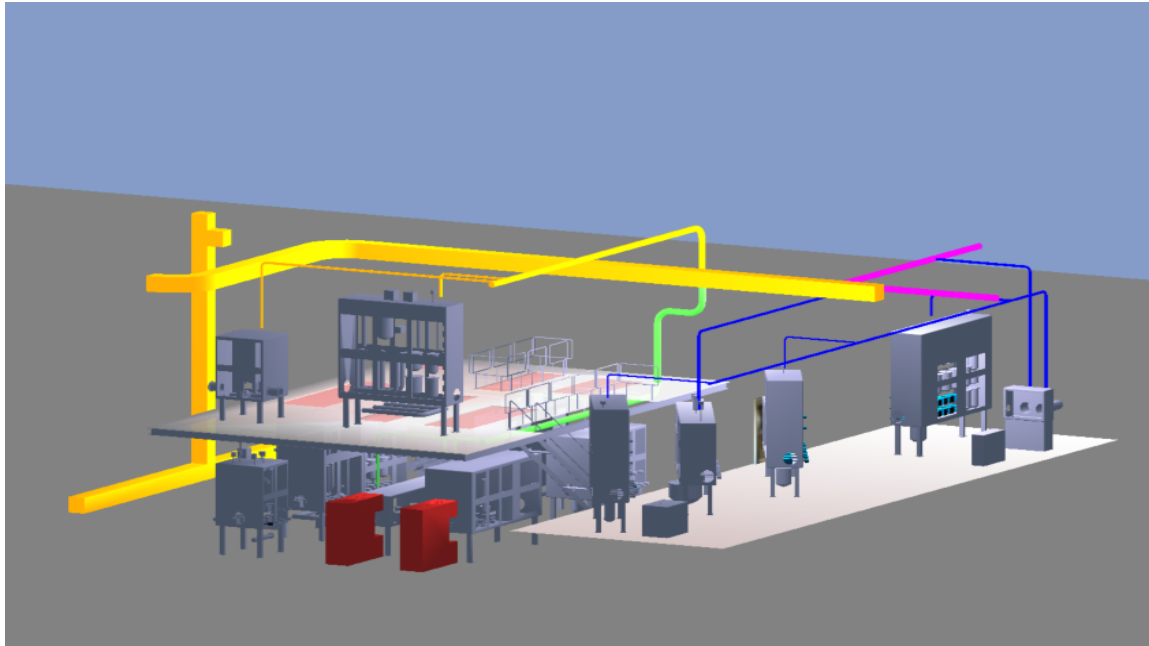


Figure 3. Conceptual design layout of the full scale americium production facility.

4. Conclusion

The potential to use ^{241}Am as an alternative to ^{238}Pu in RPS systems is at an early stage of development. The ESA work plan requires the incremental development of the technology through to a working prototype.

In Europe the cost of producing ^{241}Am is expected to be substantially less than comparative ^{238}Pu production. The reasons for this are that the following facilities would be required:

- A ^{237}Np isolation plant integrated into a current European spent nuclear fuel reprocessing facility (£100Ms).
- A ^{237}Np target production facility (£10Ms).
- A high flux nuclear reactor is required to convert ^{237}Np to ^{238}Pu (£100Ms).
- New dedicated hot cells would be required to process the irradiated ^{237}Np targets and separate ^{238}Pu (£10Ms).

Americium is attractive because:

- Over 1000 kg of ^{241}Am exists in the civil PuO_2 stockpile of the UK and France.
- Technology exists that can be applied to a ^{241}Am production facility.
- The UK has facilities that can be modified to produce ^{241}Am at a much reduced cost.

A design of a plant suitable for production of kg quantities of ^{241}Am from separated civil plutonium has been developed, and experimental work to validate the flowsheet is in progress. This process has the added benefit of producing PuO_2 with a lower gamma contact dose making the material easier to fabricate into new mixed oxide (MOx) nuclear fuel.

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