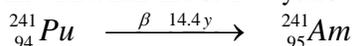


²⁴¹Am Production for Use in Radioisotope Power Systems. M.J. Sarsfield,¹ K. Bell,¹ C.J. Maher,¹ M.J. Carrott,¹ C. Gregson,¹ J. Brown,¹ D.A. Woodhead,¹ S.R. Baker,¹ R.J. Taylor,¹ T.P. Tinsley,¹ T.G. Rice,¹ C.J. Rhodes,¹ M. Clough¹, K. Stephenson². ¹Fuel Cycle Solutions, National Nuclear Laboratory, Central Laboratory, Sellafield, Seascale, CA20 1PG, United Kingdom, ²European Space Agency, ESTEC TEC-EPS, PO Box 299 - 2200 AG Noordwijk, The Netherlands. mark.sarsfield@nnl.co.uk

Introduction: During the reprocessing of spent nuclear fuel, plutonium is separated as a chemically pure plutonium dioxide (PuO₂) product, containing around 3-11% w/w of the ²⁴¹Pu isotope, and placed into storage. During storage the ²⁴¹Pu isotope beta decays to ²⁴¹Am with a half-life of 14.4 years.



This process has resulted in the creation of hundreds of kg of ²⁴¹Am growing into the civil plutonium stockpile in the UK; material that is of value in generating power sources. Over the past two years a process for separating ²⁴¹Am from the PuO₂ has been defined and a concept production plant designed based on a mass balance flowsheet and an outline of the process is shown in Figure 1. Experimental underpinning of the flowsheet was required to provide proof of concept and take the technology to TRL 3. The results of that underpinning will be reported in this paper.

The Chemical Separation Process:

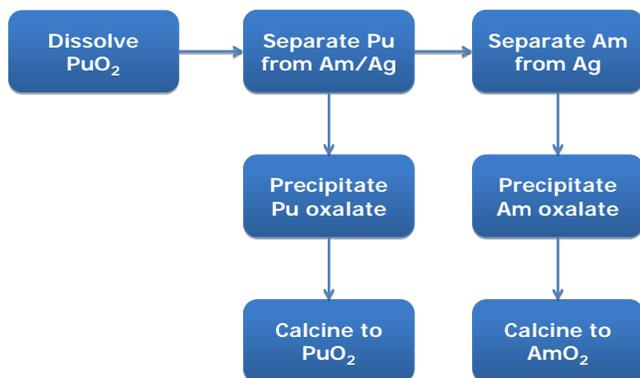


Figure 1: Outline of the process for isolating ²⁴¹Am from PuO₂.

The full scale process involves the dissolution of PuO₂, which is very insoluble even in concentrated nitric acid. To catalyse the dissolution silver (Ag) nitrate is added to the system and electrochemically oxidized to silver (II). The silver (II) reacts with PuO₂ oxidising the plutonium to the more soluble plutonium (VI) [1]. This then needs to be conditioned to the more extractable plutonium (IV) nitrate by bubbling NO_x gas through the solution before feeding to a solvent extraction process. The solvent extraction process uses tributyl phosphate (TBP) in odourless kerosene (OK)

to extract the plutonium into an immiscible solvent phase while leaving the americium and silver in the aqueous phase. The plutonium is then recovered from the solvent by reducing with hydroxylamine nitrate to plutonium (III) nitrate which is not extractable in TBP/OK (Figure 2). The plutonium is recovered in > 99.99% yield and > 99% purity, which is important when complying with the strict requirements for returning the material back to storage.

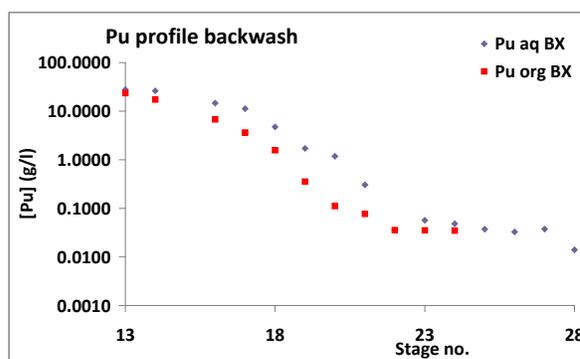


Figure 2: Example of the plutonium concentration profile across each of the centrifugal contactors used to recover the plutonium from the TBP/OK organic solvent. The aqueous stream is collected at stage 13 while the organic phase is collected at stage 28.

The separated americium left in the nitric acid is contaminated with silver, which needs to be removed. The most effective and practical method for separating the americium to the desired purity level is achieved using a second solvent extraction cycle. This employs a solvent mixture that selectively extracts americium and leaves the silver in the nitric acid aqueous phase. The solvent is then treated with fresh dilute nitric acid to recover the extracted americium.

In both solvent extraction cycles a counter current process using centrifugal contactors ensures efficient mixing and separating of the aqueous and organic phases. This is evident in the final purity of the americium nitrate solution with > 99.99% recovery and > 99% purity based on the impurities analysed (Table 1). The full paper will discuss how the different elements are distributed across the flowsheet and comment on potential process improvements.

Table 1. Calculated impurity levels in the final americium nitrate liquor for the full scale flowsheet

	Element concentration (g/l)	Final assay %w/w
Np	0.0049	0.20%
U	0.0004	0.02%
Pu	0.0023	0.09%
Am	2.4999	99.60%
Ag	9.67E-05	0.004%
Fe	2.13E-03	0.08%
Ni	9.82E-05	0.004%
Cr	<LLD	0.00%

The americium is recovered from this liquor by precipitating americium oxalate as an insoluble solid. Experiments have been performed to validate existing solubility models in the open literature and these results will be presented (Figure 3) [2].

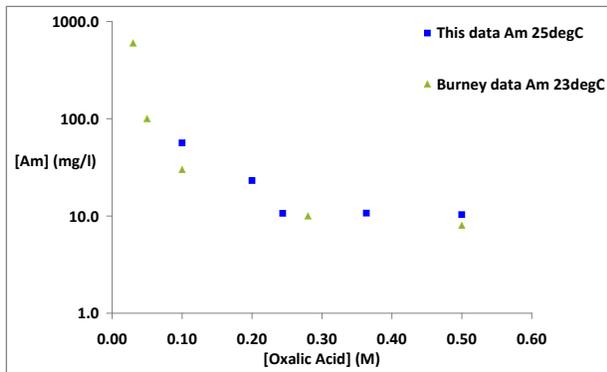


Figure 3: Americium oxalate precipitation. Soluble americium in the mother liquor; $[\text{HNO}_3] = 1\text{M}$.

The americium oxalate is then calcined at temperatures between 500 and 700°C in an air atmosphere to give americium dioxide. The solid isolated was analysed by calorimetry and thermogravimetric analysis and shown to be of the expected purity, although material calcined at lower temperatures was found to contain < 3% w/w impurities believed to be non-oxidized carbon.

Conceptual Plant Design:

Based on the above flowsheet each of the process steps requires alpha active gloveboxes with all the required criticality and radiological controls in addition to support operations such as analysis, waste treatment steps, off-gas abatement and buffer storage. By generating a 3D virtual image of the plant layout we have established the footprint and relative positions of vent systems with utility supplies and waste routes to ensure

that the available space within the National Nuclear Laboratory, Central Lab, can accommodate a full scale production facility that is capable of producing around 8.4kg of ^{241}Am per year (Figure 4).

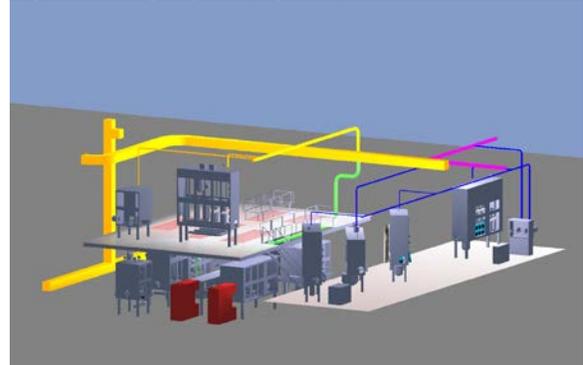


Figure 4: Design of the full scale production facility

The next stage of the process is to establish a route to densified americium oxide ceramic material to be used as heat sources. There are a number of challenges involved that will be addressed over the next 2.5 years such as:

Defining the correct Am:O ratio in AmO_{2-x} to ensure that there is no volatility of americium under sintering conditions.

Define the correct Am:O ratio in AmO_{2-x} to ensure a suitable crystal phase (i.e. cubic) will be formed during sintering that will be stable to the final encapsulation process.

Conclusions:

A separation process for the extraction of ^{241}Am from civil plutonium dioxide tested on a laboratory scale gives pure americium and plutonium products in > 99% purity with > 99.99% recovery.

Acknowledgements:

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