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## The separation of $^{241}\text{Am}$ from aged plutonium dioxide for use in radioisotope power systems using the AMPPEX process

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### Abstract

Electrical power sources used in outer planet missions are a key enabling technology for data acquisition and communications. State-of-the-art power sources generate electricity from alpha decay of  $^{238}\text{Pu}$  via thermoelectric conversion. However, production of  $^{238}\text{Pu}$  requires specialist facilities including a nuclear reactor, a source of  $^{237}\text{Np}$  for target irradiation and hotcells to chemically separate neptunium and plutonium within the irradiated targets. These specialist facilities are expensive to build and operate, so naturally, a more economical alternative is attractive to the space industry. Within Europe  $^{241}\text{Am}$  is considered a promising alternative heat source for radioisotope thermoelectric generators (RTGs) and radioisotope heating units (RHUs) since, as a daughter product of  $^{241}\text{Pu}$  decay,  $^{241}\text{Am}$  exists in 1000 kgs quantities within the UK civil plutonium stockpile. A chemical separation process is required to extract the  $^{241}\text{Am}$  in a pure form and this paper describes the AMPPEX process (Americium and Plutonium Purification by Extraction), successfully developed over the past five years to isolate  $^{241}\text{Am}$  in high yield (> 99%) and to a high purity (> 99%). The process starts by dissolving plutonium dioxide in nitric acid with the aid of a silver(II) catalyst, which is generated electrochemically. The solution is then conditioned and fed to a PUREX type solvent extraction process, where the plutonium is separated from the americium and silver. The plutonium is converted back to plutonium dioxide and the americium is fed forward to a second solvent extraction step. Here the americium is selectively extracted leaving the silver in the aqueous phase. The americium is stripped from the solvent and recovered from solution as americium oxalate, which is calcined to give americium dioxide as the final product. This paper describes the development of the AMPPEX process over a series of six solvent extraction separation trials using centrifugal contactors. The technical underpinning reported here has allowed the design of a facility capable of producing 25-50 g of americium per day.

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## 1. Introduction

Our knowledge of the solar system has increased dramatically over the last 50 years mainly as a consequence of the success of space probes, such as Voyager and Cassini and rovers such as Mars Curiosity, that use electrical power and heat generated by radioisotope decay [1]. Current state-of-the-art radioisotope thermoelectric generators (RTGs) and radioisotope heating units (RHUs) are powered by the alpha decay of  $^{238}\text{Pu}$ , which has a half-life of 87.7 years and a specific thermal power output of  $0.568 \text{ W}_{\text{th}}/\text{g}$ . The decay heat is converted to electrical energy, by the thermoelectric effect, or used to heat instrumentation. Production of  $^{238}\text{Pu}$  has mainly occurred in the USA and Russia and global stocks are depleting, although a programme of work has begun in the USA to restart  $^{238}\text{Pu}$  production [2].

Over the last few years there has been an interest by the European Space Agency (ESA) to develop power systems for space missions [3] and a study was initiated to explore the development of radioisotope powered systems suitable for producing heat and electrical power within Europe. A wide range of isotopes were considered and while  $^{238}\text{Pu}$  was the most attractive from a performance perspective, the most accessible and economically viable isotope in Europe proved to be 241-amerium ( $^{241}\text{Am}$ ) with a half-life of 432.7 years and a specific thermal power output of  $0.1146 \text{ W}_{\text{th}}/\text{g}$ .

The radioisotope  $^{241}\text{Am}$  is a by-product of the civil nuclear energy industry. Over the last few decades France and the UK have provided a service to reprocess spent nuclear fuel from nuclear reactor sites, separating the uranium and plutonium and storing these products as the oxides ready for reuse in new fuel. In the UK some of the plutonium has been stored for many decades and one of the isotopes ( $^{241}\text{Pu}$ ), present within the plutonium, has beta decayed to the isotopically pure  $^{241}\text{Am}$ . A programme of work was initiated by ESA in 2009 to develop a process to isolate  $^{241}\text{Am}$  from plutonium dioxide ( $\text{PuO}_2$ ) [4, 5], while separate parallel studies established how the isotope would be incorporated into power systems [6].

This paper provides an overview of the radioisotope production aspects of the programme.

## 2. The full scale chemical separation process

There are many ways to recover americium from nuclear material [7] but not all provide the product quality required (> 99% purity) and product recovery (> 99%) whilst minimising the amount of waste volumes and radioactivity produced. An outline of the AMPPEX process developed is shown in Fig. 1. In the full scale process 300 gHM (Heavy Metal) Pu as  $\text{PuO}_2$  powder will be dissolved in nitric acid using a silver(II) catalyst, that is continuously generated electrochemically, to provide a 150 gPu/L solution of Pu(VI) nitrate. This solution is sparged with  $\text{NO}_x$  at  $50^\circ\text{C}$  to provide a Pu(IV) nitrate solution and then conditioned to 4.5 mol/L nitric acid 100 gPu/L solution. The solution is filtered and then fed to a solvent extraction process based on well-established PUREX technology using 30% tributyl phosphate (TBP) diluted in odourless kerosene (OK). The plutonium is extracted and separated from the americium and silver before reductive backwashing as Pu(III). The plutonium is recovered as a 30 gPu/L solution, which is conditioned to Pu(IV) before oxalate precipitation and calcination to the oxide  $\text{PuO}_2$ .

The product solution containing americium and silver is then fed to another solvent extraction process where americium is separated from silver. The solvent used is a mixture of TODGA (N,N,N',N'-Tetraoctyl diglycolamide) and DHOA (N, N-dihexyl octanamide) developed to provide the optimum extraction at the acid concentration of the americium/silver solution coming from the first solvent extraction step and to provide adequate recovery of americium from the solvent using a simple low acid stripping agent. The silver and solvents are recycled and reused.

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