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Isolation of high purity americium metal via distillation

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ABSTRACT

Pure americium metal is a crucial component for the fabrication of transmutation fuels. Unfortunately, americium in pure metal form is not available; however, a number of mixed metals and mixed oxides that include americium are available. In this manuscript a method is described to obtain high purity americium metal from a mixture of americium and neptunium metals with lead impurity via distillation.

1. Introduction

The disposal of spent nuclear fuel is one of the main challenges facing the nuclear power industry today due to the long term health and safety hazards of storing the material once it is removed from the reactor. A key factor in reducing the hazards associated with long term storage is the reduction or elimination of minor actinides in fuel such as neptunium, americium and curium, which have relatively long half-lives. Transmutation is one method currently under investigation to resolve this issue. Transmutation aims to incorporate the long lived actinides into new fuel which can be placed in a fast reactor where the elements of concern will fission into products which have shorter half-lives. In order to develop transmutation fuels on a large scale it is necessary to first fabricate small quantities of fuel with minor actinide additives on which to perform thorough characterization and irradiation testing. In addition, the thermal characterization of pure americium metal is incomplete and pure americium metal is needed in order to perform measurements to gain a better understanding of its thermal properties. Since americium is needed for characterization as well as in the fabrication of transmutation fuels and, currently, there is very little of this material in existence as pure metal it is necessary to develop methods to isolate it from the feedstock material that is available. The majority of the americium bearing feedstock available is in oxide or mixed metal form. Processes were developed in the past to isolate americium from americium oxide using metallothermic reduction techniques [1–3] as well as from mixed metals using distillation [1,3–5]. Ion exchange columns have also been used to produce high purity americium by separating it from other actinides [6]. Due to the numerous types of americium bearing feedstock available, many of which are in small quantities, methods for isolating the material must be continuously developed. This manuscript describes a process for separating americium metal from neptunium metal using a unique distillation setup. Parameters described herein may be of use in the development of distillation separation processes for other metal mixtures containing americium. Also described in this publication is a method to protect the isolated metal from rapid oxidation.

2. Materials and methods

The furnace setup for the distillation process is shown outside the glovebox in Fig. 1A and inside the glovebox in Fig. 1B. The system was originally developed and tested on a benchtop using manganese metal as a surrogate for americium and alumina crucibles. It was then installed in an argon atmosphere radiological glovebox. The glovebox is designed with leaded glass windows and bismuth lined gloves to protect workers from radiation exposure. This is an important consideration as the americium bearing material had radiological readings of up to 5 Rad on contact. The glovebox was maintained at less than 10 ppm oxygen any time this work was being performed.

The experimental setup consisted of a furnace with a tantalum crucible with an OD of 0.375 inches and an ID of 0.295 inches (Metal Technology, Albany, OR). In addition, a gamma detector was used to locate the area of material deposition and had the potential to monitor distillation progress by observation of the 59 keV gamma ray from ^{241}Am. The crucible design incorporated a tantalum endcap cooled by a copper tube for collection of the vaporized americium. Fig. 2 shows schematics of the inside of the furnace and the cold finger design. The copper rod was cooled passively and extended down into the crucible to a level just beneath the furnace

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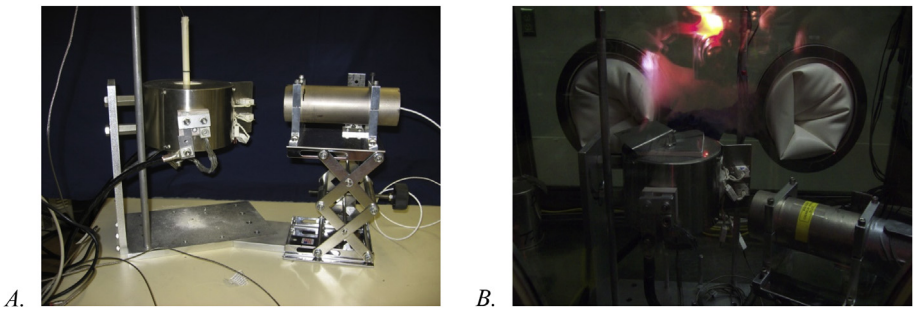

Fig. 1. A. The furnace and detector setup with an alumina crucible is shown outside the glovebox. B. The furnace and detector setup is shown inside the glovebox without a crucible.

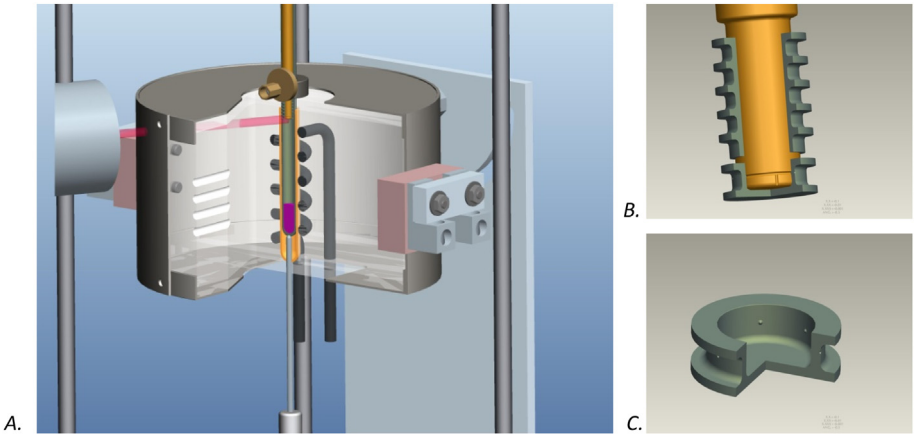

Fig. 2. Schematics of the americium distillation furnace and cold finger. A. The inside of the furnace containing a tantalum crucible (dark grey) loaded with starting material (purple) and the copper cold finger. B. The tantalum endcap on the copper cold finger. C. The tantalum endcap. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

surface. Originally the material was removed by hand drilling, but retrieval was later improved by the addition of a modified lathe (Sherline 4000A/4100A Vista, CA). Fig. 3 shows the modified lathe with a stand and catch plate. The crucible was clamped into the lathe and drill bits of differing diameters (0.27 inch, 0.28 inch, 0.29 inch) were used to remove material deposited on the crucible walls.

3. Experimental

For most experiments the starting material containing 96.2 wt% (±5 wt%) americium metal and 4.6 wt% (±0.23 wt%) neptunium metal and trace lead and oxygen impurities was loaded into the bottom of the tantalum crucible. In some cases material retrieved from the bottom of the crucibles of previous experiments was used as starting material to ensure as much americium as possible was retrieved from the available material. In these experiments the starting material was depleted of americium, but based on the amounts of americium metal retrieved from the initial experiments, it was deemed worthwhile to redistill the material that remained in the bottom in an attempt to retrieve americium left behind after the initial distillation. The cold finger was then inserted into the top of the crucible and the set-up was connected to a rotary vein roughing pump. The system was pumped down to approximately 30 mtorr (measured at the glovebox interface). The gamma detector was aligned with the location of the cold finger inside the crucible to monitor the 59 keV ^{241}Am line and the progress of the distillation. The control system was set to heat at 25°C per minute to 1000°C. As the temperature approached 1000°C the ramp rate and set point temperature were adjusted manually to maintain a controlled heat while not overshooting the desired temperature of 1170°C–1190°C. The 59 keV ^{241}Am line was monitored to determine when the material of interest consolidated on or below the cold finger. It is important to note that once the americium background within the glovebox became too high due to the various americium processes that the box is used for, the

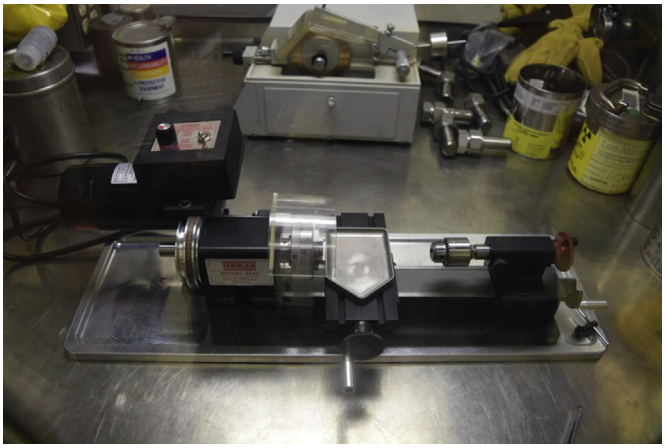

Fig. 3. The modified lathe with the addition of a catch pan for collection of retrieved metal shavings and a stand to allow it to stand upright when not in use and conserve glovebox floor space.