The Need for Minor Actinide Production*

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ABSTRACT

The US Department of Energy (DOE) manages an inventory of materials that contains a range of long-lived radioactive isotopes that were produced from the 1960s through the 1980s by irradiating targets in production reactors to produce special heavy isotopes for DOE programmatic use, scientific research, and industrial and medical applications. Oak Ridge National Laboratory (ORNL) used many of these materials as feedstock in the calutron Electromagnetic Isotope Enrichment Facility (EIEF), one of only two facilities in the world with capabilities to enrich radioisotopes in multigram quantities. Both the production reactors and enrichment facilities have been shut down, and many of these unique materials will never be produced again. ORNL maintains a major portion of the United States' inventory of these materials and uses them in DOE's center for production, storage, and distribution of transuranium isotopes (plutonium through californium) to the user community. The US inventory of enriched actinides is being depleted, and attempts for over a decade to obtain critical supplies from foreign sources have been unsuccessful. Significant strides have been made recently in the development of new and improved enrichment technologies to replace the previous electromagnetic separations techniques. This paper summarizes the US's need for production of high-purity enriched actinides by national security, nuclear nonproliferation, and basic research programs.

INTRODUCTION

The actinide radioisotopes are 15 chemical elements with atomic numbers ranging from 89 to 103 (actinium through lawrencium). With the exception of ^{232}Th, ^{235}U, and ^{238}U, which occur naturally in substantial quantities, the actinide radioisotopes are synthetically produced by irradiation in nuclear reactors, generally with low isotopic purity. Many isotopes of interest must then be enriched to obtain the isotopic purity required for user applications, often with isotopic purities in the 99–99.99% range.

There have been only two known sources of enriched radioisotopes produced in multigram quantities in the world: (1) the EIEF located in Oak Ridge, Tennessee, and (2) Russian facilities,

primarily the Russian Federal Nuclear Center – All-Russian Scientific Research Institute of Experimental Physics Enrichment Facility (RFNC-VNIIEF) [1]. These facilities used electromagnetic isotope separation (EMIS) to produce both stable and radioactive enriched isotopes in large quantities. The EIEF produced gram quantities of numerous valuable enriched actinide isotopes (up to 99.999% purity) prior to being shut down in 1979. The elements separated in the EIEF included lithium, boron, carbon, magnesium, silicon, sulfur, chlorine, potassium, calcium, titanium, vanadium, chromium, iron, nickel, copper, zinc, gallium, germanium, selenium, bromine, rubidium, strontium, zirconium, molybdenum, ruthenium, palladium, silver, cadmium, indium, tin, antimony, tellurium, barium, lanthanum, hafnium, tantalum, tungsten, rhenium, osmium, iridium, platinum, mercury, thallium, lead, cerium, neodymium, samarium, europium, gadolinium, dysprosium, erbium, ytterbium, lutetium, thorium, uranium, plutonium, americium, and curium [2]. The RFNC-VNIIEF has produced enriched plutonium, americium, and curium using calutron technology similar to the EIEF, primarily for its country's domestic use [3].

Because the capability to enrich milligram-to-gram quantities of actinide radioisotopes no longer exists in the United States, the enriched actinide needs of the nation are currently being met using existing, but diminishing, US inventories and foreign (mostly Russian) sources, where available. The dwindling US inventory and the unavailability of future domestic and international sources are of concern to both suppliers and users. For example, inventory of enriched ^{244}Pu is in short supply, and several standards organizations, such as New Brunswick Laboratory and the Institute for Reference Materials and Measurements, stopped or severely limited distribution of their ^{244}Pu standards to conserve the remaining small quantities (milligram level) for very-high-priority needs. Without additional supplies of enriched ^{244}Pu, the United States, the International Atomic Energy Association (IAEA), and others risked losing certain measurement capabilities that are essential in maintaining an active nuclear forensics and safeguards posture in current and future world affairs.

This shortage prompted the IAEA to initiate a project in the 1990s to have the RFNC-VNIIEF enrich 5 g of ^{244}Pu that would be supplied by ORNL. The goal was to produce 100 mg of 99.5% ^{244}Pu and 1 mg of 99.99% ^{244}Pu. Three decades after the project was initiated, the effort was put on indefinite hold after a limited demonstration in which only a small test quantity of ^{244}Pu was enriched. The Russians completed an enrichment demonstration in 2012 using a 0.5 g sample of ORNL-supplied feedstock in a two-stage electromagnetic separation [4]. The first separation yielded ~10 mg plutonium with ~98.86% ^{244}Pu. The second separation yielded 0.88 mg plutonium with 99.98% ^{244}Pu**.** Although the demonstration did not meet the desired quantity or enrichment goals, the final separation product was returned to the United States where it was certified as a reference material for isotope dilution mass spectrometry to meet the needs of the IAEA Network of Analytical Laboratories for the immediate future.

In response to the growing scarcity of enriched stable isotopes and concerns about the reliability of supply, the National Science Foundation Nuclear Science Advisory Committee's (NSAC's) Isotope Subcommittee concluded in its August 2009 report that it is essential that the US reestablish the base production capability for stable isotope separation [5]. To meet these needs, ORNL, in conjunction with the DOE Office of Nuclear Physics (DOE-NP) Isotope Program, developed a modern high-resolution, high-throughput EMIS for stable isotope enrichment. The system has the capability of producing a milligram to tens of grams of enriched stable isotopes and went into production operations in 2017 [6–8].

DOE/NSAC also recommended reestablishing the domestic capability for production of high-purity radioactive isotopes to support research needs [9]. The present effort was undertaken to help determine the need for high-purity radioactive isotope enrichment.

THE NEED

A list of user needs for actinide radioisotopes was obtained through a variety of sources, including open literature technical documents and canvasing of users with science and technology needs. The list focuses on the desires for high-purity enriched actinides by US national security, nuclear nonproliferation, and basic research programs; enrichment of ^{235}U is not included in this study [2]. The existing inventories of enriched actinides and/or the existing production capabilities for enriched actinides were evaluated to determine if they could meet anticipated user demand. The needs that could not be met through existing inventories and/or existing production capabilities were used as the basis of this actinide-enrichment-requirements evaluation. A list of uses of enriched actinide radioisotopes is given in Table 1, and a list of the enrichment projections is given in Table 2.

Both ^{242}Pu and ^{244}Pu can be used as nuclear forensic tracers. As the world continues to run nuclear power plants and recycled fuels, the amount of ^{242}Pu in the environment will increase, thus decreasing the suitability of ^{242}Pu for nuclear forensics. Plutonium-244, however, is not produced in appreciable quantities in standard power reactor fuel; therefore, the demand for the very limited existing supply of enriched ^{244}Pu is predicted to increase in the future.

Enriched ^{244}Pu material is also needed as target material for heavy ion bombardment for studies of transactinide elements, which are used in the production and discovery of the super heavy elements. In December 1998, for example, a team of scientists from Lawrence Livermore National Laboratory collaborated with Russian scientists at the Joint Institute for Nuclear Research in Dubna, Russia, on the discovery of element 114 using a heavy ion cyclotron to bombard a film of ^{244}Pu with ^{48}Ca ions for 40 days (a discovery that was recognized by *Popular Science* as one of the year's 100 greatest achievements in science and technology) [10].

Table 1. Uses of Enriched Actinide Radioisotopes

Enriched Actinide	Use	Users
^{236}Np	Nuclear forensics	Department of Homeland Security Department of Energy Defense Threat Reduction Agency Intelligence Community International Atomic Energy Agency
^{242}Pu	Plutonium research	Department of Energy National Nuclear Security Administration
^{242}Pu	Superheavy element (SHE) research	Department of Energy SHE Research Community
^{244}Pu	SHE research/nuclear forensics	Department of Homeland Security Department of State Defense Threat Reduction Agency Intelligence Community International Atomic Energy Agency Department of Energy SHE Research Community
^{244}Cm	SHE research	Department of Energy SHE Research Community
^{248}Cm	SHE research	Department of Energy SHE Research Community
^{251}Cf	SHE research	Department of Energy SHE Research Community

Table 2. Actinide Enrichment Projections

Enriched Actinide	Isotopic Purity Desired	Maximum Quantity Desired over 5 Year Period
^{236}Np	99.99%	2 mg
^{242}Pu	99%	To be determined
^{242}Pu	99.9%	200 mg
^{244}Pu	99.9%	1,000 mg
^{244}Pu	99.99%	2–5 g
^{244}Cm	99.9%	500 mg
^{248}Cm	99.9%	650 mg
^{251}Cf	99.9%	650 mg

In addition, ^{244}Pu has a half-life that is more than 200 times longer than any other plutonium isotope and a low specific activity (1.9E-5 Ci/g), making it extremely useful in studies that attempt to understand the fundamental thermodynamics of plutonium in either the solution or solid state. Plutonium-244 is also the prime isotope for plutonium tracing (e.g., in the marine environment).

Like ^{244}Pu, ^{242}Pu has a long half-life and a specific activity (0.004 Ci/g) much lower than other common isotopes of plutonium (^{239}Pu, ^{240}Pu, and ^{241}Pu). Therefore, highly enriched ^{242}Pu can be handled in a facility with fewer safety requirements. It is also available in larger quantities than ^{244}Pu.

Neptunium-236 is of interest for use as an Integrated Database Management System tracer for the radiochemical analysis of ^{237}Np, which is significant in a number of fields of research such as nuclear forensics, environmental analysis, and the nuclear fuel cycle. Although various radioactive tracers have been used, including ^{235}Np, ^{239}Np, and ^{236}Pu, none have been found to be satisfactory. Neptunium-236 is a potential candidate as a neptunium yield tracer because its 1.55×10^5 year radioactive half-life allows it to be used for both radiometric and mass spectrometric measurements.

Much of the recent progress in superheavy element (SHE) research has been accomplished using the hot fusion method in which neutron-rich actinide targets are bombarded with intense beams of neutron-rich ^{48}Ca ions. ORNL has provided actinide target materials that have been used for the last 20 years for the synthesis of all SHEs above element 112. For example, ORNL-produced ^{249}Bk target material was used for the discovery of element 117 in a US–Russian experiment at the Joint Institute for Nuclear Research in Dubna, Russia. This discovery was confirmed using ORNL-made ^{249}Bk at the Gesellschaft für Schwerionenforschung Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany. Element 117 officially joined the periodic table in 2016 as tennessine (Ts).

Enriched long-lived actinide materials, including ^{244}Cm, ^{248}Cm, ^{242}Pu, and ^{244}Pu, are required for use in SHE research to connect the proposed hot fusion island of stability to the nuclear mainland, to study decay properties including fission probabilities, and to investigate reaction mechanisms [11–14]. Californium-251, ^{244}Cm, and ^{248}Cm are used in SHE discovery research. Californium-251 was used in the recent discovery of element 118 and is now being used in SHEs for the element's synthesis. It is also being considered for experimental studies to discover elements beyond element 118, including elements 119, 120, and more. Curium-248 is also a very desirable research isotope for new element discovery because of its high neutron-to-proton ratio and is the isotope of choice for curium chemistry studies because of its long half-life (3.4×10^5 years) and relatively low specific activity (0.00424 Ci/g). It has been identified for use in discovery research for element 120. Curium-248 is also used as the primary feedstock in the production of ^{252}Cf and the coproduction of ^{249}Bk, which was used in the discovery and

verification of element 117. The goal of reaching the long-predicted new element 124 will require a highly enriched ^{251}Cf target.

PRODUCTION METHODS

EMIS is the only technology reported to have been used in the past to enrich multigram quantities of actinides, and irradiation technology has been used to produce small quantities of a limited number of isotopes. Other enrichment technologies have reportedly only been used for production of stable isotopes. All of these technologies could be considered for implementation of future actinide enrichment capabilities, however EMIS has the most advantages for this application. The attributes of these potential enrichment processes are listed in Table 3 [2, 6].

Table 3. Comparison of Enrichment Technologies

	Typical Quantities	**Enrichment per Pass**	**Isotope Collection**	**Separation Medium**	**Research Required for New Isotopes**
Electromagnetic Isotope Separation	mg to g	High	All simultaneous	Ions	Low
Gas Centrifuge Isotope Separation	kg	Low	End isotopes	Gas	Medium
Gas Diffusion	kg	Very low	End isotopes	Gas	High
Plasma Separation Process	kg	Moderate	Single, targeted	Ions	Medium
Atomic Vapor Laser Isotope Separation	kg	Moderate	Single, targeted	Vapor ions	High
Chemical and Physical Processes	kg	Varies	Varies	Varies	High
Irradiation	mg to g	Low	End isotopes	Varies	Low

EMIS technology is the most versatile known means of separating isotopes [15] and has been used to enrich most of the elements on the periodic table. It has a high first-pass enrichment capability (~10% natural abundance to ~99% enrichment in one pass), can enrich all isotopes of

an element simultaneously, and has a throughput on the order of milligrams to grams per hour. ORNL, in conjunction with the DOE-NP Isotope Program, developed a modern high-resolution, high-throughput EMIS for stable isotope enrichment to replace the World War II–era calutron EMIS. The system has the capability of producing a milligram to tens of grams of enriched stable isotopes and went into production operations in 2017 [6–8]. The first isotope to be produced was ^{96}Ru needed for experiments at Brookhaven National Laboratory's Relativistic Heavy Ion Collider (RHIC). The system produced 448.6 mg of enriched ^{96}Ru for the RHIC experiments and 99.2% enrichment of ^{102}Ru at 1.35 mA average for 1,000 hours.

Irradiation is applicable only when the desired radioisotope has significantly lower fission cross sections relative to other isotopes, allowing the other isotopes to be "burned out" by fissioning, thus enriching the desired isotope. Patton et al. [16, 17] evaluated enrichment by irradiation in the ORNL High Flux Isotope Reactor and concluded that enrichment by irradiation could be successful in only a limited number of special cases. For example, material containing 5% ^{244}Pu could be enriched to 13% with 18 irradiation cycles, and material containing ~50% ^{242}Pu could be enriched to ~90% with 18 irradiation cycles. Engle et al. [18] calculated that radioisotopic purities of ^{242}Pu in excess of 99.5% could be achieved by irradiating plutonium materials with an initial ^{242}Pu concentration of 95%.

Other enrichment methods provide much higher throughput than required to meet the user demands identified in this study, and they require multistaging to obtain high enrichments and are either isotope selective or alter the feed abundance to enhance either the heaviest or lightest isotope [19].

SUMMARY

Because the capability to enrich milligram-to-gram quantities of actinide radioisotopes no longer exists in the United States, the enriched actinide needs of the nation are currently being met using existing, but diminishing, US inventories and foreign (mostly Russian) sources where available. DOE/NSAC has recommended reestablishing the domestic capability for production of high-purity radioactive isotopes to support research needs [9]. The review of actinide-enrichment needs summarized in this paper revealed that milligrams to hundreds of milligrams highly enriched neptunium, plutonium, curium, and californium are desired. The accompanying review of enrichment technologies indicated that EMIS, potentially coupled with irradiation in some cases, is likely to be the most appropriate enrichment process for the quantity and range of isotopes needed.

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