US Heavy Actinide Production at Oak Ridge National Laboratory—Ongoing Actinide Partitioning and Transmutation Programs

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Summary

Actinide partitioning and transmutation (P&T) processes for heavy element production are currently being done at Oak Ridge National Laboratory (ORNL) using similar technologies that are of interest for commercial power reactor spent nuclear fuel actinide P&T. Such reprocessing and recycle methods are of interest to shorten the period of spent nuclear fuel management by converting long-lived actinides to shorter-lived fission products. Greater amounts of neutron-emitting Cm and Cf are being used at ORNL than in most P&T research and development (R&D) programs.

Introduction

Actinide P&T programs, with continuous recycle of feedstocks, are being operated at ORNL for heavy element isotope production at the multigram-to-kilogram scale (Collins et al., 2012). These P&T processes are similar to those being developed for separation and destruction of the long-lived actinides in high-level nuclear waste from nuclear power reactors. Therefore, these ongoing isotope production programs offer opportunities to improve, develop, and demonstrate the methods needed for actinide P&T. Greater quantities of neutron-emitting curium and californium are being used than in most worldwide P&T R&D studies.

Multigram amounts of ^{242}Pu, ^{243}Am, and ^{244}Cm have been transmuted to produce heavier Cm isotopes and transcurium element actinides, Bk and Cf in multimilligram amounts, Es in multimicrogram-to-milligram amounts, and Fm in atoms to picogram amounts. These materials are used for (1) exploration of the nuclear and chemical properties, (2) creation of particle accelerator targets for production of super heavy elements, and (3) development of practical uses for the actinides and their isotopes. This program has been the basis of the transuranium element isotope production program since the High Flux Isotope Reactor and Radiochemical Engineering Development Center were built in the mid-1960s (King et al., 1980). Practical uses for ^{252}Cf as a portable neutron source, due to its significant spontaneous fission, have been developed for various neutron radiography and neutron activation analysis applications (Boulogne, 1981). Funding from ^{252}Cf neutron source fabricators is now sustaining the program.

More recently (in the last decade) ^{238}Pu production has been reestablished for use by the National Aeronautics and Space Administration (Wham et al., 2015). The goal is to reach an annual production rate of 1.5 kg/y or greater of heat source plutonium oxide, containing a Pu elemental mass composition with >82% of the isotope, ^{238}Pu. This isotope has an 88-y half-life and 0.57 W/g decay heat, characteristics that make it the choice for use in radioisotope thermoelectric generators used to power the instrumentation package on deep space missions. The feedstock to

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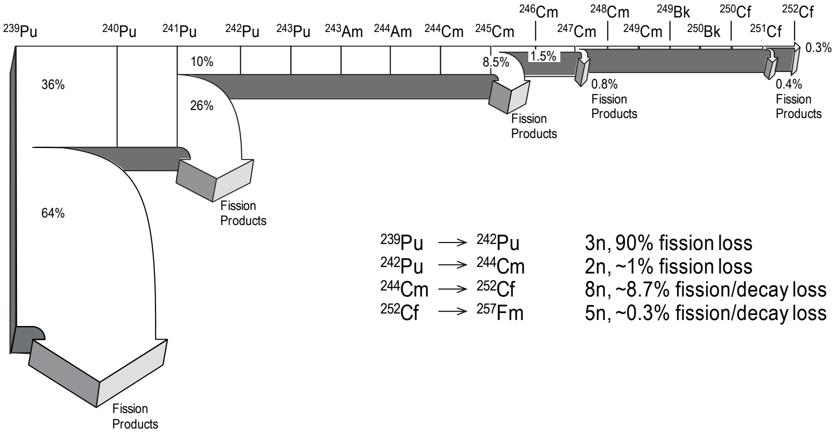
this P&T production process is 12–25 kg/y of ^{237}Np, and the fractional conversion to ^{238}Pu is 5–15% per P&T cycle.

Transmutation

The transmutation path for production of the heavy actinide element isotopes is a series of *n*/*γ* and ß decay reactions, as shown in Figure 1 for average thermal neutron cross sections. As described above, feedstocks have been either ^{242}Pu or ^{243}Am and ^{244}Cm, which were produced by transmutation of Pu in large production reactors at the Savannah River Site. The heavy element transcurium products are Bk, Cf, Es, and Fm (Figure 3).

Figure 1: Transmutation path for production of heavy actinides



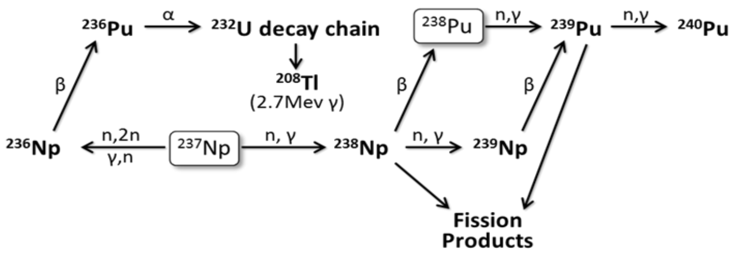
Fission of isotopes along the transmutation path is a significant loss in the production yield; however, this very phenomenon is a desirable attribute for destruction of long-lived actinides in nuclear waste.

In the three neutron captures required for the transmutation of ^{239}Pu to ^{242}Pu, 90% of the mass is transmuted to fission products in thermal spectrum reactors. If the "minor actinides" ^{243}Am and ^{244}Cm are recycled and transmuted, the conversion to fission products is >99% because of the large fission cross sections for ^{245}Cm and ^{247}Cm.

Reactor physics modeling calculations (Renier, 2008) show that the Am–Cm isotopes from typical light water reactors with burnup of 45 GWd/metric ton of initial heavy metal (MTHM) and 10 y of decay after reactor discharge, contain 0.78 kg of Am and 0.028 kg of Cm per MTHM. The mass of the Am is 86% ^{241}Am and 14% ^{243}Am, whereas the mass of Cm is 90.6% ^{244}Cm, 8.0% ^{245}Cm, 1.1% ^{243}Cm, and 0.7% ^{246}Cm. Further P&T of the Am would produce primarily ^{238}Pu and ^{242}Pu plus fission products from the ^{241}Am component, and the ^{243}Am and ^{244}Cm would produce primarily fission products and heavier isotopes of Cm, as indicated in Fig. 1. Partitioning of the transmuted actinides via well-known solvent extraction processes would easily separate the Pu and Cm fractions. That method is how the feedstock for heavy actinide production has been generated.

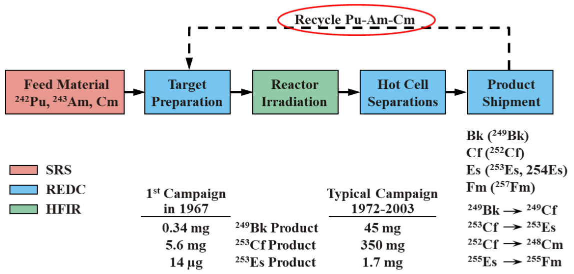
The pathway for ^{238}Pu production (Figure 2) via transmutation of ^{237}Np, is much shorter, but it is still complex because of the fission of ^{238}Np, the unwanted transmutation of ^{238}Pu to ^{239}Pu, and the side reactions of ^{237}Np to ^{236}Pu, which is undesirable because of its decay to ^{232}U and its high radiation energy decay daughters. Because the neutron capture cross section for ^{238}Pu is three times greater than that for ^{237}Np, the conversion per P&T cycle is limited to <20%.

Figure 2: Transmutation pathway for production of ^{238}Pu



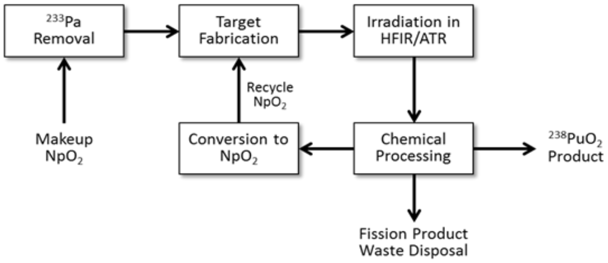
Functional diagrams for the two ongoing isotope production programs are shown in Figure 3 for heavy actinides and in Figure 4 for ^{238}Pu.

Figure 3: Heavy element production process steps



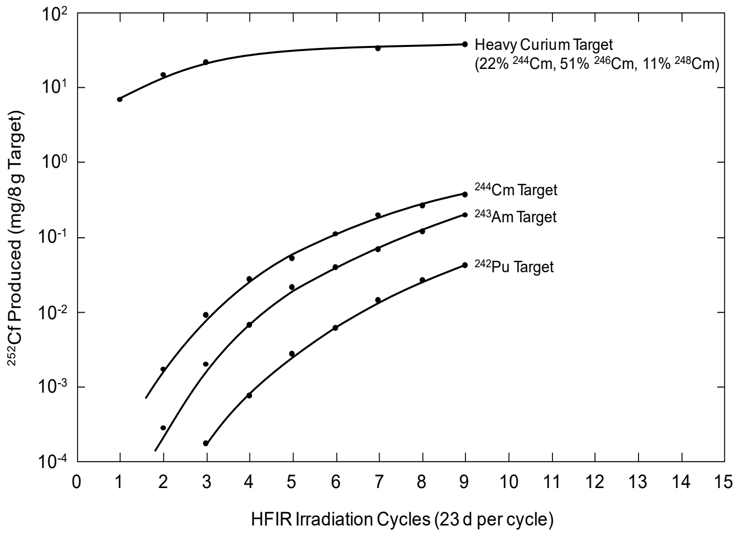
1st Campaign in 1967		Typical Campaign 1972-2003
0.34 mg	^{249}Bk Product	45 mg
5.6 mg	^{253}Cf Product	350 mg
14 µg	^{253}Es Product	1.7 mg

Figure 4: Plutonium-238 production process steps



Recycle of the lighter actinides is necessary in heavy actinide production to increase the heavy Cm isotope (^{246}Cm and ^{248}Cm) concentrations in the targets fed to the transmutation irradiation step, and thus, to increase the production rate of ^{252}Cf and other transcurium actinides. This is an exponential effect is shown in Figure 5. In the production of ^{238}Pu, recycle of ^{237}Np is necessary to conserve the feedstock ^{237}Np because its conversion to ^{238}Pu is limited to <20% per P&T cycle.

Figure 5: Rate of ^{252}Cf and other transcurium actinides



Partitioning

After dissolution of the oxide components (actinide and fission product elements), the feed solutions for partitioning have equally high or greater concentrations for the isotope production processes than for reprocessing minor actinides from spent nuclear fuel, although the overall volumes of spent fuel reprocessing solutions are much greater. Similar chemical separations processes can be used.

The first step in the partitioning flowsheet is the selective extraction of the transplutonium actinides and lanthanide fission products, thereby removing non-lanthanide fission products, such as Cs, Sr, Ru, Zr–Nb, and Ce (as trivalent Ce). For this step, spent nuclear fuel reprocessing R&D programs have used the PUREX (Benedict et al., 1981) TRUEX (Bond and Leuze, 1980), and TALSPEAK (Weaver and Kappelmann, 1968) processes; in addition, the heavy actinide element production program has used the CLEANEX process (Bigelow, 1980). All of these processes use metal (phosphorus)-based ligands, such as n-tributyl phosphate (TBP), carbamoyl methyl phosphonate (CMPO), or di-2-ethylhexyl phosphoric acid (HDEHP), as the extractant. R&D programs are now using phosphorus-free, soft-donor extractants containing only C, H, O, and N ligands, such as the mono- and di-amides and pyridines.

In heavy actinide production, the CLEANEX process has been used successfully for many years, but the HDEHP must be disposed as liquid waste after use because it cannot be cleaned and recycled. Therefore, recent R&D tests have been done to substitute tetra-octyl di-glycol amide (TODGA) as the extractant for future operations (Delmau, 2017). This substitution will enable solvent cleanup and recycle. Also, it will provide a less complex feed adjustment because an aqueous feed solution with higher nitric acid concentration can be used.

The subsequent partitioning process in both the heavy element production program and in spent nuclear fuel reprocessing is done to separate the actinides from the lanthanide fission products. The early and still currently used process for heavy element production is the LiCl-based anion exchange chromatography process (Collins et al., 1981). However, this process requires the use of chloride corrosion–resistant equipment, which is more expensive than stainless steel, and requires time-consuming operations to switch from nitric to chloride media and then back to nitric media for subsequent partitioning separations and conversion steps.

Trials using the TALSPEAK solvent extraction process have been made, but separations efficiency was not as good as with the anion exchange chromatographic process. Also, the TALSPEAK process uses HDEHP as the extractant. New R&D will consider the use of bis-triazyl pyridine (BTP) derivatives, such as the camphor derivative.

Another option to be considered is the nitrate-based, cation exchange chromatographic process used successfully in the past at the Savannah River National Laboratory (Harbour et al., (2000). That process is similar, but operated at larger scale, than the high-pressure cation exchange chromatographic process currently used to elute and separate the heavy elements with alpha hydroxyl isobutyrate at different pH levels (Benker et al., 1981).

In the processing of Np–Pu–fission product solutions for the recovery of ^{238}Pu, the TBP solvent is cleaned and reused, but the strong alpha radiolysis from extracted ^{238}Pu causes greater solvent degradation, resulting in excessive concentrations of monobutyl and inorganic phosphates in the separated aqueous Pu and Np product solutions. This is one of the reasons that the anion exchange purification steps have been necessary. Testing is in progress to examine the use of monoamides to replace TBP in the partitioning solvent extraction.

Summary and Conclusions

Several examples of actinide P&T processes for heavy element production are currently being done at ORNL using similar technologies that are of interest for commercial power reactor spent nuclear fuel actinide P&T. Such reprocessing and recycle methods are of interest to shorten the period of spent nuclear fuel management by converting long-lived actinides to shorter-lived fission products. Moreover, greater amounts of neutron-emitting Cm and Cf are being used at ORNL than in most P&T R&D programs. Opportunities for collaboration exist to expand reprocessing and spent nuclear fuel component recycle R&D in the future.

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