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Purification of ^{242}Pu by irradiation with thermal neutrons

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ABSTRACT

The feasibility of purifying a radioisotopically impure sample of ^{242}Pu using neutron irradiation to "burn up" the plutonium isotopic contaminants ^{238,239,240,241}Pu is investigated. A starting point of 95% ^{242}Pu is assumed, and different neutron energy spectra, fluxes, and irradiation periods representative of the conditions found at available reactors in North America were assessed with Monte Carlo simulation methods to predict the maximum obtainable enrichment of ^{242}Pu. Calculations indicate that radioisotopic purities of ^{242}Pu in excess of 99.5% are achievable with neutron fluxes at Oak Ridge National Laboratory's High Flux Isotope Reactor, pointing encouragingly to experimental verification of these estimates.

1. Introduction

Isotopically pure samples of a variety of actinides are useful for applications in nuclear forensics and research into the fundamental physics and chemistry of these elements; in particular, isotope dilution mass spectrometry methods for sensitive detection of trace levels of actinide isotopes require high purity isotope standards [1]. In this regard, a supply of isotopically pure ^{242}Pu ($t_{1/2}$ = 3.75E5 y, 100% α) would be attractive, especially given its relatively long half life and separation from other similarly persistent plutonium isotopes by two mass numbers. In addition to electromagnetic separation for milligram-scale production [2], the purification of isotopically mixed samples of plutonium by neutron irradiation has been proposed [3], and neutron irradiation has been used to obtain transactinide elements at large scales [4]. However, to the best of our knowledge neither feasibility studies nor experimental results of an effort to purify ^{242}Pu are described in the open literature.

Nuclear data available for consideration are, in our experience, commonly used to describe the formation of desired radionuclides. The measured energetic dependence of the formation rate of residual nuclei is described with nuclear production cross sections, which are a measure of the likelihood that a bombarding particle will interact with target nuclei in the desired manner to produce the chosen product nucleus. To ascertain the feasibility of ^{242}Pu purification described here, it is necessary to distinguish between these "production" cross sections and "burnup" cross sections. In our proposed irradiation scheme, nuclear reactions described by burnup cross sections preferentially eliminate isotopic impurities in the ^{242}Pu sample through reactions which lead to an indiscriminate array of radioisotopic products, e.g. through well-known channels such as (n,fission) and (n,pxn). These reactions proceed in tandem with the formation of desired and undesired plutonium isotopes through (n,γ) reactions on lower-mass plutonium isotopic targets, the decay of residual nuclei, and additional reactions with decay products as new targets for the incoming neutron flux. The differential rate of formation and burnup of plutonium isotopes is the focus of this investigation, and is quantified by these reported results in terms of the final plutonium isotopic content in the irradiation sample expressed as a mass percentage.

Qualitative evaluation of cross section data in the ENDF/B-VII.1 repository provides some rationale for the preferential burnup of lower mass plutonium isotopes in an attempt to purify ^{242}Pu (see Figs. 1 and 2 below) [5]. The relevant burnup cross section of ^{242}Pu for thermal neutrons are predominantly (n,γ) and (n,fission) reactions initiated by sub-eV neutrons. These burnup reactions' cross sections are at least an order of magnitude lower than the cross sections for the same reactions on $^{238\text{-}241}$Pu targets. For ^{239}Pu in particular, the largest plutonium isotopic component in

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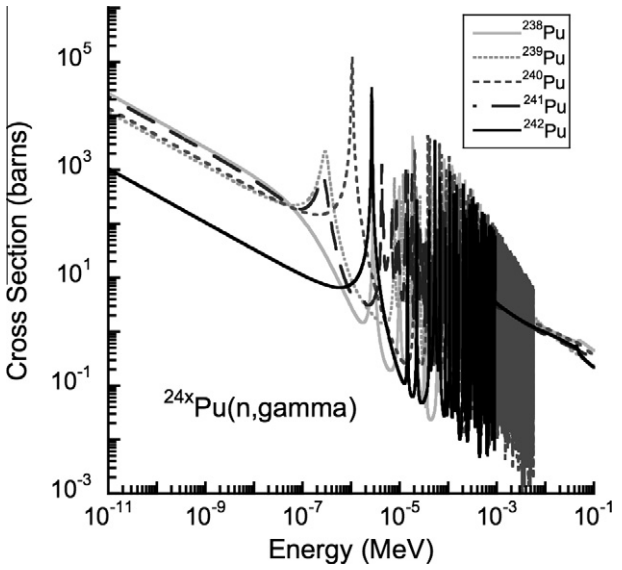

Fig. 1. Comparison of ENDF/B-VII1.0 evaluated (n,γ) cross sections for $^{238-242}$Pu.

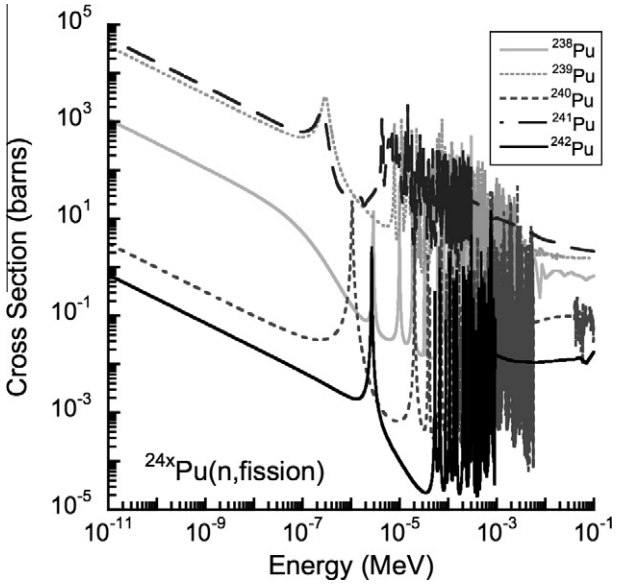

Fig. 2. Comparison of ENDF/B-VII1.0 evaluated (n,fission) cross sections for $^{238-242}$Pu.

the vast majority of spent nuclear fuel samples, the (n,fission) burnup cross sections' magnitude for cold neutrons extends into the kbarn range, almost five orders of magnitude larger than the burnup cross sections for the same reaction on ^{242}Pu.

Predicting the results of reactor irradiation of plutonium samples is complicated, however, by the varying lifetimes of the relevant plutonium isotopes and by the presumed co-production of americium and curium isotopes in a given neutron flux. Fig. 3 shows simplified schematic representations of ^{238}Pu and ^{240}Pu production via successive neutron capture reactions and β^- decay beginning with ^{241}Pu and ^{243}Pu, respectively. Because the half lives of ^{243}Pu and ^{244}Am in particular are only hours-long, (n,γ) reactions on ^{242}Pu in particular might be expected to contribute significant ^{240}Pu impurity, especially if desired final sample concentrations of ^{242}Pu are very high (e.g., >99%). Additionally, the magnitude of capture cross sections is similar to that of (n,fission) cross sections for the relevant plutonium and americium isotopes, complicating estimates of the relevant contribution of competing reaction channels.

The complexity of the proposed irradiation scheme's isotopic transmutations, combined with the challenging logistics of irradiating a sample composed of special nuclear material, necessitated the use of nuclear codes available through the Radiation Safety Information Computational Center to achieve a feasibility assessment. This study is described below.

2. Simulations and calculations

In order to obtain an understanding of how plutonium isotopic compositions change with irradiation in representative neutron energy spectra, calculations were performed with the isotope depletion and production codes ORIGEN2 [6] and CINDER90 [7]. The input to these codes includes starting material compositions, reactor operating parameters, and cross sections representing a particular reactor type. CINDER90 uses a 63-group input for cross sections, which is collapsed into a single group for further calculations. The output used for the calculations described here is material composition as a function of irradiation time. The starting plutonium isotopic content of the sample was set at 0.5% ^{238}Pu, 1% ^{239}Pu, 3% ^{240}Pu, 0.5% ^{241}Pu, and 95% ^{242}Pu by mass; this isotopic distribution was used as the starting point for all computations described below. The assumed chemical form of the sample used for computation is plutonium oxide with a density of 10.9 g/cm^3 (95% of theoretical density); sample masses were chosen to fill the available cell space. Actual irradiations will require thermal description of target quantities and materials, as well as consideration of the effects of especially large samples upon the criticality of the reactor facility.

Three reactors types were considered. Reactors that could potentially irradiate a small sample of the material include the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL), the Massachusetts Institute of Technology's Nuclear Research Reactor (MITR), or the Advanced Test Reactor (ATR) at Idaho National Laboratory. As described below, initial computational experiments indicated the flux at HFIR proved to be the most potentially advantageous, and HIFR was therefore chosen for more detailed analyses. To capture the details of the flux spectrum and resulting cross sections representing HFIR, a detailed geometric model of the reactor using the Monte Carlo transport code MCNP6 was obtained [8]. The cycle described in the MCNP6 input files represents the geometry from cycle 400, which ran between April 27, 2004 and May 21, 2004 (24.6 days) and has a run cycle of typical duration for HFIR [9]. As the composition of fuel rods in the core changes during the run cycle, the energetics of the neutron fluence also evolve. For this reason, two files, one pertaining to the reactor characteristics at the beginning of the cycle (BOC) and one from the end (EOC) are provided by HFIR staff at ORNL and were used comparatively for the calculations discussed here. Cross sections at a temperature of 450 K were used for the fuel areas and 350 K for the water using ENDF/B-VII processed data [5].

The linkage code *Monteburns* was then run to coordinate interaction between the transport and depletion calculations. *Monteburns* is a Monte Carlo burnup code that links the Monte Carlo transport code MCNP to an isotope depletion and production code, such as ORIGEN2 or CINDER90. Results using both depletion codes CINDER90 and ORIGEN2 were compared and determined to be similar; further results are reported using CINDER90 because those isotopic compositions were directly fed into further decay calculations.

Monteburns is beneficial for analyzing irradiation behavior of specific reactors because MCNP uses continuous-energy cross