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Preconceptual Feasibility Study to Evaluate Alternative Means to Produce Plutonium-238

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Abstract. There is currently no large-scale production of ^{238}Pu in the United States. Feasibility studies were performed at the Idaho National Laboratory to assess the capability of developing alternative ^{238}Pu production strategies. Initial investigations indicate potential capability to provision radioisotope-powered systems for future space exploration endeavors. For the short term production of ^{238}Pu, sealed canisters of dilute ^{237}Np solution in nitric acid could be irradiated in the Advanced Test Reactor (ATR). Targets in the large and medium "I" positions of the ATR were irradiated over a simulated period of 306 days and analyzed using MCNP5 and ORIGEN2.2. Approximately 0.5 kg of ^{238}Pu could be produced annually in the ATR with purity greater than 92%. Optimization of the irradiation cycles could further increase the purity to greater than 98%. Whereas the typical purity of space batteries is between 80 to 85%, the higher purity ^{238}Pu produced in the ATR could be blended with existing lower-purity inventory to produce useable material. Development of irradiation methods in the ATR provides the fastest alterative to restart United States ^{238}Pu production. The analysis of ^{238}Pu production in the ATR provides the technical basis for production using TRIGA® (Training, Research, Isotopes, General Atomics) nuclear reactors. Preliminary analyses envisage a production rate of approximately 0.7 kg annually using a single dedicated 5-MW TRIGA reactor with continuous flow loops to achieve high purity product. Two TRIGA reactors represent a robust means of providing at over 1 kg/yr of ^{238}Pu annually using dilute solution targets of ^{237}Np in nitric acid. Further collaboration and optimization of reactor design, radiochemical methods, and systems analyses would further increase annual ^{238}Pu throughput, while reducing the currently evaluated reactor requirements.

Keywords: ^{238}Pu, ATR, high purity, TRIGA, production.

INTRODUCTION

Historically, ^{238}Pu has been proven to be the best radioisotope for the provision of space nuclear power because of its long half-life, low radiation levels, high power density, and stable fuel form at high temperatures. Typically 7 to 8 kilograms are required to power an explorer-class mission such as the National Aeronautics and Space Administration's (NASA) New Horizons mission to Pluto.[1] Missions such as the Mars Science Laboratory which use the Multi-Mission Radioisotope Thermoelectric Generator (MMRTG) are to use less than 5 kg ^{238}Pu, effectively implementing just more than half of what a General Purpose Heat Source Radioisotope Thermoelectric Generator (GPHS RTG) would need.[2-4] Intentions to develop an Advanced Stirling Radioisotope Generator (ASRG) that uses less than 1 kg ^{238}Pu are still in progress.[5]

Currently there is no large-scale production of ^{238}Pu in the United States. The K-Reactor at the Savannah River Site in South Carolina, which was shut down in 1996, was the last reactor to produce significant quantities of ^{238}Pu. Approximately 40 kg of ^{238}Pu has been purchased from Russia over the past two decades to augment the dwindling supply already stockpiled in the United States. Unfortunately, Russia has also lost its capability to produce new ^{238}Pu. Because the Russian plutonium cannot be used for national security missions, much of the domestic supply has been consumed for these purposes.[1]

The traditional approach to producing and recovering ^{238}Pu is shown in Figure 1. Solid pellet targets of $^{237}NpO_2$ in aluminum are irradiated to produce ^{238}Np via neutron capture. The ^{238}Np then beta decays into the desired ^{238}Pu product. Chemical post processing of the target can separate the ^{238}Pu product and purify the recovered ^{237}Np for recycling into another target. The isotopic content of ^{238}Pu is greater than 80%. Each production cycle typically converts between 10-15% ^{237}Np; the remainder is recycled.[1,6]

A previous feasibility assessment has been performed for the production of ^{238}Pu using solid $^{237}NpO_2$ pins in the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL). For the reactor operating at 200 MW, using a significant portion of the reactors flux traps, a total of 13 kg ^{238}Pu could be produced with a purity of 88.3% using 87 kg ^{237}Np.[7-8]

In the late 1950s, it was proposed to replace the conventional solid target irradiation scheme with that of a liquid loop system coupled in a nuclear reactor coupled with a chemical separations facility. Such a concept would reduce irradiation and decay time between chemical processing steps, increase the annual ^{238}Pu production rate, decrease the loss of ^{237}Np feedstock, and improve the overall ^{238}Pu purity to over 98%.[9]

The purpose of this investigation is to provide a feasibility analysis of the irradiation of encapsulated ^{237}Np liquid-solution targets in a light-water reactor to determine the effective annual production rate and purity of ^{238}Pu. These canisters could be irradiated for a short period of time, removed from the reactor, and their contents can be processed to obtain the ^{238}Pu-product and ^{237}Np-recycle materials. Both short- and long-term strategies were initiated in an endeavor to provide a long-term production rate of at least 1 kg of ^{238}Pu per annum.

NEAR TERM ^{238}PU PRODUCTION FEASIBILITY

The Advanced Test Reactor

For the short term production of ^{238}Pu, sealed canisters of ^{237}Np solution in nitric acid can be irradiated in the ATR at the Idaho National Laboratory. The ATR has been continuously in operation since 1967 with its primary mission in support of the development and refinement of nuclear propulsion systems for the U.S. Navy. Now it serves a range of customers internationally as a National Scientific User Facility (NSUF). Although it only operates at approximately 60% of its experiment loading capacity, there is a high demand for many of the primary flux trap positions.[10] The reactor is licensed for a thermal power rating of 250 MW but typically operates between 110 and 140 MW with a maximum rating during experiments of 200 MW. A diagram of the ATR is shown in Figure 2.

The medium and large "I" positions in the ATR were assessed in this feasibility analysis. The four large I positions (1, 6, 11, and 16) are seen along the outer edge of the core with two medium I positions on each side of the large I positions, as seen in Figure 2. These positions were selected because of their low demand for use. Thus they can be utilized continuously throughout the year with canisters. The canisters could then be changed between normal irradiation cycles in the ATR.

Some basic assumptions used in this analysis are as follows: 1) the analysis was performed using the 19 fuel plate, full-core benchmark model of the ATR provided in the *International Handbook of Evaluated Criticality Safety Benchmark Experiments*[11]; 2) the average neutron flux in the solution will change over time because of composition changes caused by neutron capture and subsequent transmutation of the isotopes present; 3) the entire solution is exposed to the average neutron flux calculated for a given position. Although the flux distribution throughout the solution is uneven, diffusion of the various isotopes produced from neutron capture away from the higher concentrations regions, which will also be higher flux region, will distribute the flux evenly throughout the solution; 4) sufficient cooling is provided by the primary coolant flow such that boiling within the solution does not occur; and, 5) a full ATR cycle is set to 50 days and the shutdown period between cycles is set to 14 days for the purpose of this evaluation. Actual irradiation cycles would vary in length.

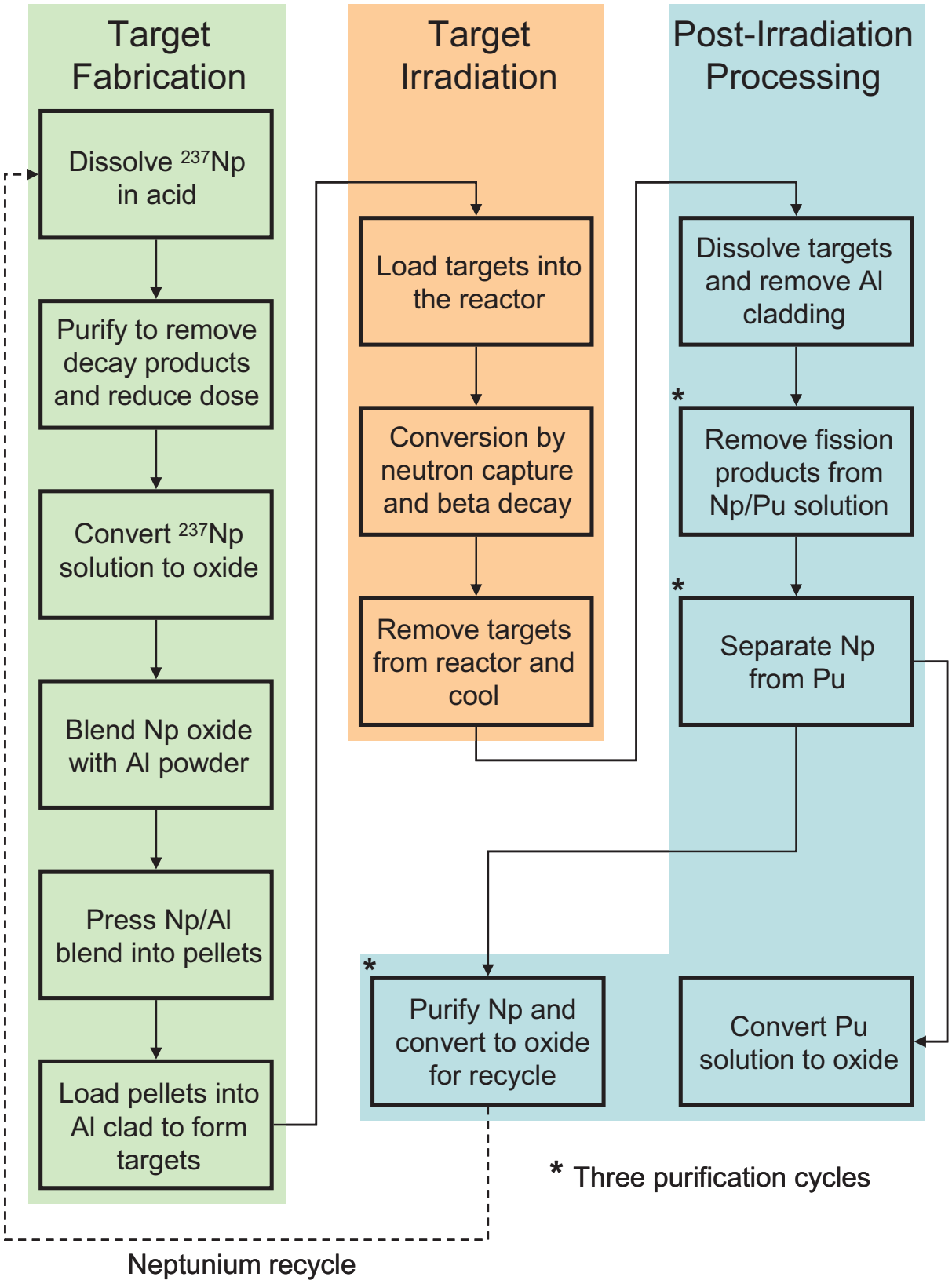

FIGURE 1. Historic Process for ^{238}Pu Production and Recovery. The Final Product is a Plutonium Dioxide Powder with an Isotopic Content of >80% ^{238}Pu. Each Production Cycle Converts 10-15% ^{237}Np to ^{238}Pu with the Remaining Np Recycled.[6]

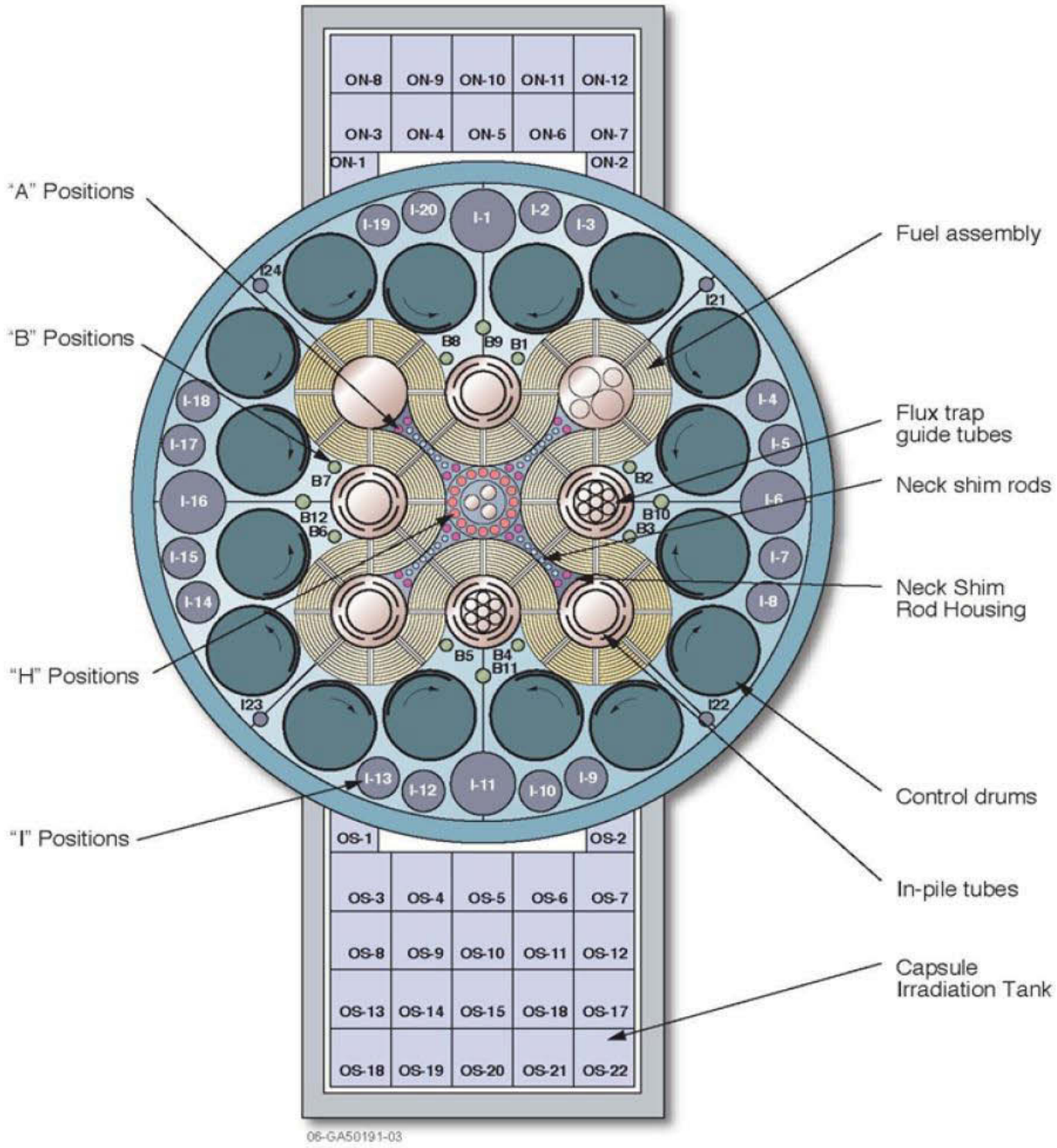

FIGURE 2. Cross Section View of the Advanced Test Reactor.

^{238}Pu Production Analysis

The benchmark model was analyzed using Monte Carlo N-Particle (MCNP) version 5-1.4 [Ref. 12] with the ENDF/B-VII.0 neutron cross-section data library.[13]

The ^{237}Np solution is placed in a double canister. The double canister design consists of two 0.125-in.-thick SS 304 canisters; the gap separating the smaller canister from the larger was set at 0.05 in. and filled with helium. A 0.25 in. gap between the outer diameter of the larger canister and the surrounding beryllium for all I positions was maintained to allow for sufficient cooling of the canister and solution.

Since the medium and large I position diameters varied, two different designs had to be set for the double canisters. For the large I positions, the inner diameter of the smaller canister is set at 3.9 in. and the larger canister's inner diameter is set at 4.25 in. For the medium I positions, the inner diameter of the smaller canister is set at 2.15 in. and the larger canister's inner diameter is set at 2.5 in. For both the medium and large I positions, the total axial length of the smaller canister is set at 48.5 in. to allow for the solution to be exposed to the full length of the ATR fuel meat and the surrounding canister's total axial length is set at 48.85 in. For the purposes of establishing the feasibility of ^{238}Pu production, two different solutions were defined with 200 and 300 g/L of ^{237}Np dissolved in 2 M nitric acid (HNO_3).

MCNP and ORIGEN2.2 [Ref. 14] are used together to determine the total quantity of ^{238}Pu produced in the various I positions in the ATR. The average flux for the ^{237}Np solution was calculated using a cell averaged flux tally in MCNP along with an (n,γ) reaction rate multiplier card to calculate the microscopic cross sections of ^{237}Np, ^{238}Np, ^{238}Pu, and ^{239}Pu. The calculated flux values and cross sections were then passed into ORIGEN2.2 with the correct isotope composition of the solution and then analyzed for neutron irradiation over the course of five 50-day ATR cycles with intermediary, two-week, shutdown periods. The total in-core irradiation time is 306 days. The total ^{238}Pu amount in grams was calculated after the last 50-day ATR cycle had been completed. The operating power of the ATR was 110 MW.

The final calculated production capability in the ATR is approximately 456 g/yr of ^{238}Pu using the medium and large I positions containing 200 g/L ^{237}Np solution. Increasing the ^{237}Np concentration to 300 g/L could boost the ^{238}Pu production rate up to approximately 545 g/yr. Concentration in the feed solution was not optimized; further testing of higher concentration should be assessed to further increase annual production rates.

Purity is another important factor in determining the feasibility of ^{238}Pu production. Quantities of other plutonium isotopes such as ^{239}Pu or ^{241}Pu would increase the necessity for additional radiological protection and shielding when handling the plutonium material or using it as a heat source. Therefore the total plutonium impurities present within the system were tracked over the total simulation time. Purity as a function of simulation time is shown in Figure 3.

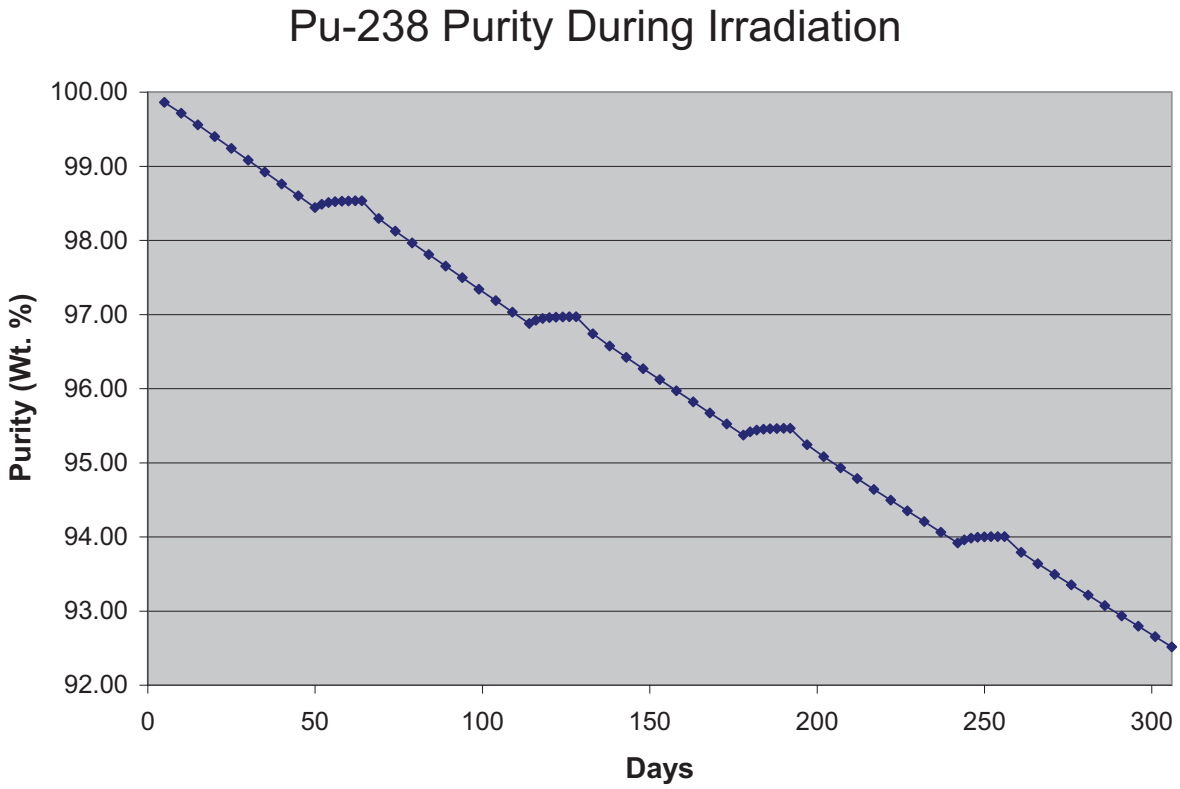

FIGURE 3. Purity of ^{238}Pu during Simulated Irradiation. The Temporary Jumps in Purity are Due to Beta-Decay of ^{238}Np during Decay Periods. The Purity of the Solution Drops by Approximately 1.5% per Cycle.

LONG TERM ^{238}PU PRODUCTION STRATEGY

The TRIGA Reactor

The analysis of ^{238}Pu production in the ATR provides the technical basis for production using a TRIGA® (Training, Research, Isotopes, General Atomics) nuclear reactor.[15] Development of a long-term production strategy will require implementation of nuclear reactor(s) with sufficient capabilities to provide uninterrupted irradiation services.

The TRIGA reactor is a pool-type reactor with light-water coolant. These reactors are use for non-power application and are fueled with uranium zirconium hydride (UZrH) fuel. TRIGA reactors are available through General Atomics. There are generally two TRIGA designs: an annular- or hexagonal-pitch lattice and a square-pitch lattice. The annular- and hexagonal- pitch cores are typical licensed to operate at thermal steady-state power outputs of less than 5 MW. The square-pitch cores, like the one currently in operation in Romania,[16] operate at 14 MW. Some TRIGA reactors have pulsing capabilities, which are not currently of interest in this application.

Some basic assumptions used in this analysis are as follows: 1) the analysis was performed using the simplified 1- and 14-MW TRIGA reactor designs; 2) the target locations within the reactor represent continuous flow loops to achieve high purity product; 3) sufficient cooling is provided by the primary coolant flow such that boiling within the solution does not occur; and, 4) a full production period is established at 365 days of continuous operation. Actual results will vary depending on the quantity and length of shutdown periods.

^{238}Pu Production Investigation

Two TRIGA reactor types are modeled in this study, a 1-MW hexagonal-pitch core and a 14-MW square-pitch core. Five configurations of the hexagonal-pitch core and three configurations of the square-pitch core were studied, varying the mass of uranium per fuel rod, location, and type of target: annular tank vs. cylindrical targets. The Monte Carlo design of the 1-MW TRIGA model is based off of information and data found elsewhere.[17-22] The most productive design for the hexagonal-pitch core uses six control rods and 45/20 UZrH fuel (Figure 4). The total in-core volume of ^{237}Np solution is 40.20 L, with a concentration of 200 g/L.

The 14-Mw square-pitch core uses 45/20 UZrH fuel placed in 5 × 5 lattices within fuel assemblies. The optimum design for this study uses 64 canisters placed surrounding the fueled zone with 100 to 200 g/L of ^{237}Np solution, again simulating continuous flow loops (Figure 5). The Monte Carlo design of the 14-MW TRIGA model is based off of information and data found elsewhere.[20-22]

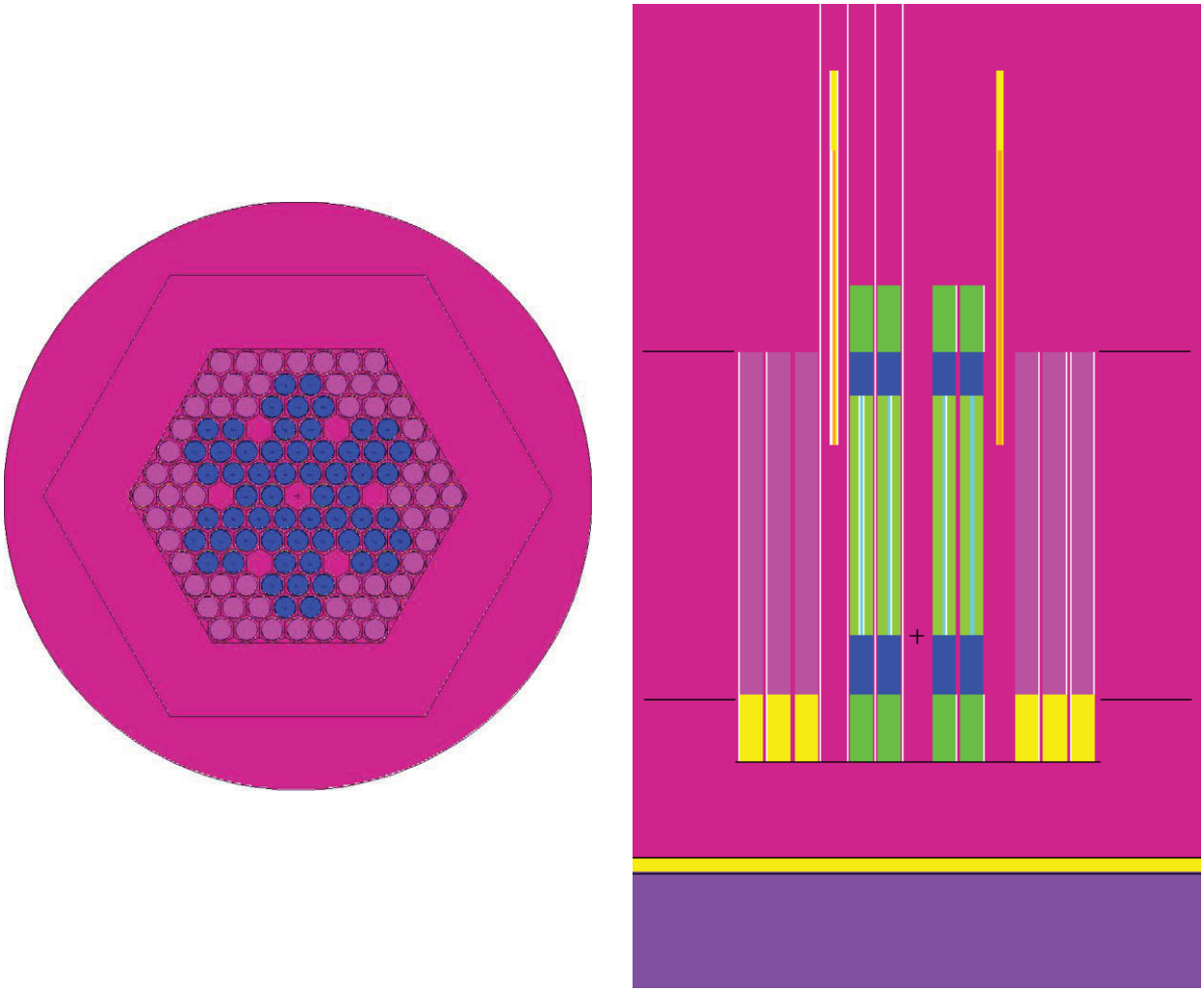

FIGURE 4. 1-MW TRIGA Core using 45/20 Fuel and ^{237}Np Targets Simulating Continuous Flow Loops.

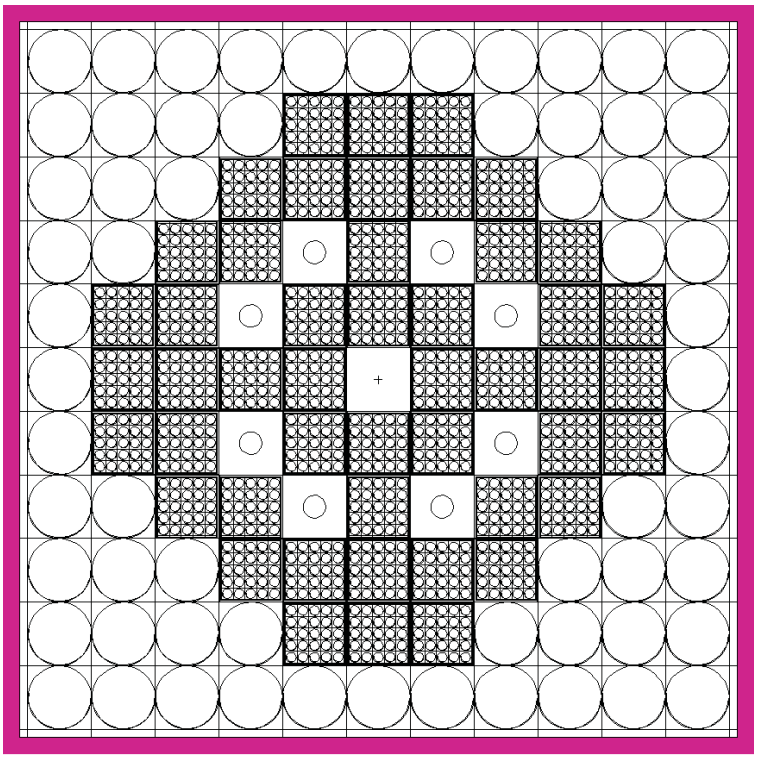

FIGURE 5. 14-MW TRIGA Core using 45/20 Fuel and ^{237}Np Targets Simulating Continuous Flow Loops.

It was estimated that for a 1-MW hexagonal-pitch TRIGA reactor using the design shown in Figure 4, a ^{238}Pu production rate of ~77 g/yr could be achieved. Increasing the solution concentration of ^{237}Np from 100 to 300 g/L can increase the production rate by an additional ~80%. Scaling to a 5 MW reactor with the same core design, without accounting for temperature or power effects, could achieve a high purity ^{238}Pu production rate of ~690 g/yr. Two 5 MW TRIGA reactors could easily provide 1 kg/yr of ^{238}Pu.

Estimates with the 14-MW square-pitch TRIGA reactor design shown in Figure 5 provided a ^{238}Pu production rate of ~578 g/yr using a ^{237}Np solution concentration of 100 g/L. Increasing the Np concentration to 200 g/L actually reduced the ^{238}Pu production rate to ~380 g/yr. To generate ~1 kg/yr of high-purity ^{238}Pu with a single reactor, a square-pitch TRIGA core could be licensed and operated up to 25 MW.[16] Coordinated efforts with General Atomics would be necessary to develop and analyze more detailed models of these high-power TRIGA reactors. Further optimization of target placement and design in and around the core may also serve to increase the annual product yield.

CONCLUSION

Both short- and long-term strategies have been investigated for restarting ^{238}Pu production in the United States. Further investigation is needed to optimize the long-term strategy for final implementation. However, the annual production rate of 1 kg ^{238}Pu is achievable using existing TRIGA reactor technology. Approximately 0.5 kg of ^{238}Pu could be produced annually in the ATR with purity greater than 92%. Optimization of the irradiation cycles could further increase the purity to greater than 98%. Whereas the typical purity of space batteries is between 80 to 85%, the higher purity ^{238}Pu produced in the ATR could be blended with existing lower-purity inventory to produce useable material. Development of irradiation methods in the ATR provides the fastest alterative to restart United States ^{238}Pu production. Preliminary analyses envisage a production rate of approximately 0.7 kg annually using a

single dedicated 5-MW TRIGA reactor with continuous flow loops to achieve high purity product. Two TRIGA reactors represent a robust means of providing at over 1 kg/yr of ^{238}Pu annually. Further collaboration and optimization of reactor design, radiochemical methods, and systems analyses would further increase annual ^{238}Pu throughput, while reducing the currently evaluated reactor requirements.

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