Isotopic Analysis of the In-Zinerator Actinide Management System

P. Phruksarojanakun, P.P.H. Wilson, B.B. Cipiti, R.M. Grady

November 2006

UWFDM-1307

Presented at the 17th ANS Topical Meeting on Fusion Energy, 13-15 November 2006, Albuquerque NM.

FUSION TECHNOLOGY INSTITUTE

UNIVERSITY OF WISCONSIN

MADISON WISCONSIN

Isotopic Analysis of the In-Zinerator Actinide Management System

P. Phruksarojanakun, P.P.H. Wilson, B.B. Cipiti, R.M. Grady

Fusion Technology Institute
University of Wisconsin
1500 Engineering Drive
Madison, WI 53706

http://fti.neep.wisc.edu

November 2006

UWFDM-1307

Presented at the 17th ANS Topical Meeting on Fusion Energy, 13-15 November 2006, Albuquerque NM.

ISOTOPIC ANALYSIS OF THE *IN-ZINERATOR* ACTINIDE MANAGEMENT SYSTEM

P. Phruksarojanakun[1], P.P.H. Wilson[1], B.B. Cipiti[2], R. M. Grady[1]

[1]*Fusion Tech. Inst., UW-Madison, 1500 Engineering Dr, Madison, WI 53706, wilsonp@engr.wisc.edu*
[2]*Sandia National Laboratories, Albuquerque, NM*

Efficient burn up of minor actinides is one of the most promising alternatives for minimizing waste in advanced nuclear fuel cycles. This work examines the concept of employing Z-pinch driven fusion source in a sub-critical transmutation reactor designed to burn up actinides and generate constant power. Its fuel cycle is designed to allow on-line fission product removal and fuel replenishment. The variation of the actinide inventory is an essential quantity used to calculate the energy multiplications and neutron spectrum, as well as to design an appropriate reactivity control mechanism.

In this paper we develop a method to calculate time-dependent isotopic distributions, fuel feeding rate and fission product removal rate necessary to obtain a constant power level. The calculation is performed by using both MCise, a Monte Carlo isotopic inventory code, and MCNP5. An important feature of MCise for this system is the ability to simulate the on-line removal of fission products from the actinide mixture.

In addition to reporting the actinide inventory and burn rates, the impact of the actinide inventory on the fission/fusion energy multiplication will be examined.

I. INTRODUCTION

The *In-Zinerator* is a novel power plant design, which is expected to transmute about 1200 kg/FPY of long-lived transuranic (TRU) actinides from LWRs and produce 3000 MWth (Ref 1.). While a number of engineering challenges must be resolved to assess the feasibility of the *In-Zinerator* concept[2], its ability to meet the fundamental goal of actinide management must be considered early in the design process. This work develops a methodology for studying the dynamic actinide inventory of the *In-Zinerator* system to determine its suitability in the role of a transmutation system.

Primarily based on the Z-pinch power plant[3] design, it is modified to operate with the sub-critical blanket. A 200 MJ D-T fusion target, producing a point source of 14.1 MeV neutrons, is ignited every ten seconds providing what can be viewed as a 20-MW fusion source for the blanket. Fig. 1 shows a cross-section of the In-Zinerator geometry. The fusion target is located in the center of the 5 meter tall cylindrical chamber, 2 meters from the 5 cm thick Hasteloy-N first wall. Beyond the first wall is a 107 cm thick liquid lead blanket region contained in a 5 cm thick Hasteloy-N vessel. The first 57 cm contains 1150 tubes of the actinide mixture while the remaining 50 cm acts as a reflector. Each tube is made of 0.1 cm thick Hasteloy-N and has an outer radius of 2.4 cm, respectively.

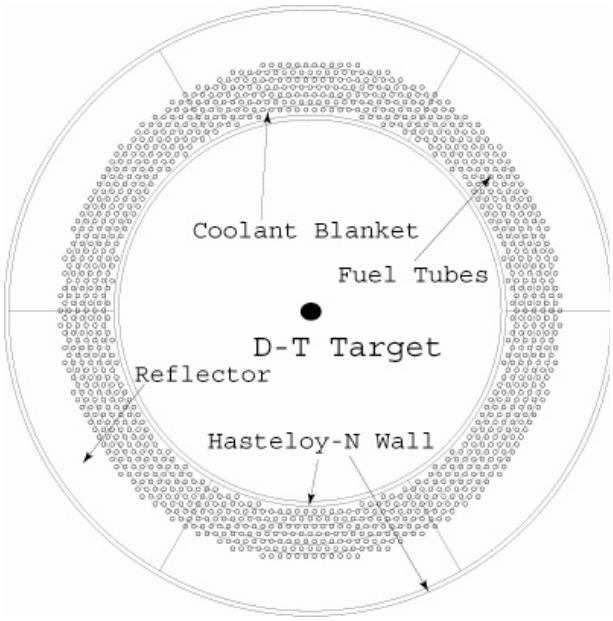

Fig. 1. Cross-section of In-Zinerator geometry.

The fluid fuel is a eutectic at 675 °C formed by LiF and AnF_3 (actinide fluoride) with a molecular ratio of 3 to 1. Using a liquid fuel allows a portion of the fuel to be processed on-line to remove fission products, replenishing the fuel with fresh TRU to maintain constant total inventories.

The Monte Carlo Inventory Simulation Engine (MCise) (ref. 4) is implemented with capabilities to handle online material addition into and extraction out of the system. The goal of this paper is to use MCise to calculate detailed isotopic distributions at all operation times.

II. MCISE TRANSMUTATION MODEL

II.A. Background

MCise has been developed and implemented for modeling activation of materials with complex processes and irradiation histories. This tool is specifically aimed at systems with flows that separate into multiple streams, each one subject to different processes or irradiation environments before rejoining into a common stream. Monte Carlo (MC) techniques based on following the history of individual atoms allows these atoms to (a) follow randomly determined flow paths, (b) enter or leave the system at arbitrary locations and (c) be subjected to radiation or chemical processes at different points in the flow path. Many elements of the methodology for MC inventory analysis have direct analogs to neutral particle MC radiation transport, where neutral particles traveling through space and changing their energy are replaced by isotopes traveling through time and changing their isotopic identity.

The current implementation of MCise includes the capability to simulate simple, complex, loop flows, and any combination of these. These advanced capabilities can later be used to implement features of real systems including sources, sinks, post-irradiation decay and extraction processes. In addition, some basic variance reduction techniques have been employed to enhance the analog simulation. These capabilities make MCise a suitable tool for a transmutation calculation of the eutectic fuel in the *In-Zinerator* because of the need to account for the constant addition of fresh fuel and extraction of fission products.

The basic methodology of MCise can be summarized as follows. The total effective reaction rate coefficient, λ_{eff}, of an isotope at an arbitrary time can be determined by collapsing the total transmutation cross-section with the neutron flux and adding the decay constant,

$$\lambda_{eff} = \lambda + \int \phi(E)\,\sigma_{tot}(E)\,dE\,. \tag{1}$$

The probability of this isotope undergoing a reaction within time dt is

$$p(t)\,dt = e^{-\lambda_{eff} t}\,dt\,. \tag{2}$$

Using the inverse transformation of the cumulative distribution of $p(t)$, the time until the next reaction, defined as t_{rxn}, can be determined from a random variable ξ by

$$t_{rxn} = \frac{-\ln(1-\xi)}{\lambda_{eff}} = n_{rxn}\,\tau_{eff}\,, \tag{3}$$

where τ_{eff} is the mean reaction time ($1/\lambda_{eff}$) and n_{rxn} is the randomly determined number of mean reaction times until the next reaction, if ξ is uniformly distributed between 0 and 1. If the remaining amount of time in the current irradiation environment, expressed in a unit of the number of mean reaction times

$$n_{rem} = t_{rem}\,\lambda_{eff}\,, \tag{4}$$

is more than n_{rxn}, a new isotope is sampled from the possible reaction pathways. The relative probability of the new isotope is calculated from the individual pathway cross-sections weighted by the current neutron flux and/or decay rates. Also, the amount of the remaining time is decremented appropriately by

$$t_{rem} = t_{rem} - t_{rxn}\,. \tag{5}$$

The new isotope is then followed in the same way as the previous isotope. On the other hand, if the remaining amount of time is less than n_{rxn}, the particle moves to another environment and λ_{eff} is updated for the flux at this new point. The number of mean reaction times until the next reaction is decremented to,

$$n_{rxn} = n_{rxn} - n_{rem}\,. \tag{6}$$

For a calculation in a single irradiation environment, this second condition indicates the end of the history of an atom and a new atom is sampled. By appropriately counting the isotopic species each history represents at a given time of interest, an expected value of the isotopic composition can be determined.

MCise was specifically designed to simulate systems such as sources, sinks, post-irradiation decay and extraction processes. These capabilities make MCise a suitable tool for an activation and burnup calculation of the eutectic fuel in the *In-Zinerator* because of the need to account for the on-line time-dependent addition of fresh fuel and extraction of fission products.

II.B. MCise Modeling Requirements

A fully specified MCise model includes a description of the network of irradiation environments and flow paths between them and also the sources of atoms in that network.

An irradiation environment in MCise is described by *a control volume,* characterized by a constant multi-group neutron flux spectrum and a residence time, t_r. The residence time represents the average amount of time any atom spends in the control volume and is defined by the engineering performance requirements of the system. For example, it can be based on a minimum required flow rate through a system for adequate heat removal or on a maximum flow rate through a chemical extraction system.

Since the flow rate leaving a control volume is defined by its residence time, the flow paths between control volumes are defined only by the relative

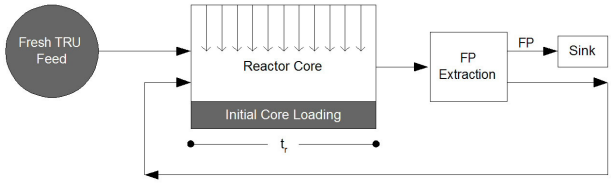

Fig. 2. Schematic of In-Zinerator MCise model, showing sources in red and sinks in blue.

distribution of the flow to downstream control volumes using a simple discrete probability distribution function (PDF). The flow distribution can act on all isotopes equally, representing a bulk material flow, or can act differentially on specific isotopic or atomic species, representing extraction/separation processes.

Another necessary component of an MCise model is a set of one or more atom sources. In MCise, each atom source is associated with a single control volume and has a time-invariant isotopic composition and a time-dependent the source strength, $r(t_{sim})$. The total strength of each source, R_s, is defined by integrating $r(t_{sim})$ over the total simulation time T_{sim}, i.e.,

$$R_s = \int_0^{T_{sim}} r\left(t_{sim}\right) dt_{sim}. \quad (7)$$

The set of total source strengths defines a discrete PDF which can be sampled to determine from which source a new atom comes. Once a particular source is chosen, its initial control volume is explicitly defined. Its isotopic identity can be randomly sampled from the discrete PDF representing the isotopic mix and its birth time can be randomly sampled from the continuous PDF, $r(t_{sim})/R_s$.

Another initial parameter for new atoms is the remaining time in its initial control volume. Some sources introduce atoms to the system at the beginning of the control volume, representing a flow from outside the network of control volumes into the system. Other sources represent inventories of atoms that are initially present in the system, some of which may leave their initial control volume immediately and others of which are resident in their initial control volume for the full residence time of that control volume. PDFs can easily be constructed to be sampled for the initial remaining residence time, t_{rem}.

II.C. *In-Zinerator* MCise Model

A schematic of MCise model of the *In-Zinerator* is illustrated in Fig. 2. The two control volumes in this schematic are the reactor core and fission product extraction environments. The reactor core is characterized by the average neutron flux that the eutectic fuel experiences while the second control volume has a neutron flux of zero. For the purpose of this analysis, a residence time of 100 days in the reactor was chosen corresponding the processing of 1% of the total inventory per day in the fission product extraction step. The residence time of the FP extraction represents a processing period of the fuel and was chosen to be zero for the purposes of this initial study.

All of the flow leaving the reactor core goes to the FP extraction process, but the flow leaving the FP extraction process is divided into two streams based on the atomic species. All fission products flow to the sink and all actinides (and Li and F) are returned to the reactor core. This model was chosen to represent an ideal separations process and the flow distribution of each species can be adjusted to represent the real separations efficiencies.

There are two atom sources in the *In-Zinerator*

TABLE I. Isotopic composition of initial core loading and TRU feed stream

Isotope	**Initial Core Loading** (atomic fraction)	**Feed Stream** (atomic fraction)
Li-6	1.2501E-02	-
Li-7	2.3753E-01	-
F-19	6.2507E-01	-
Np-236	9.0012E-09	7.3147e-08
Np-237	9.0761E-03	7.3435E-02
Np-238	-	3.1085E-11
Np-239	-	1.7966E-08
Pu-236	-	1.4384E-12
Pu-238	3.3129E-03	2.6630E-02
Pu-239	5.6632E-02	4.5452E-01
Pu-240	3.2879E-02	2.6288E-01
Pu-241	1.1576E-03	9.2095E-03
Pu-242	7.2382E-03	5.7352E-02
Pu-243	1.4127E-06	1.3491E-16
Pu-244	-	1.1055E-05
Am-241	1.1576E-02	9.2075E-02
Am-242	2.0627E-05	1.9542E-09
Am-242m	-	1.6305E-04
Am-243	2.6003E-03	2.0530E-02
Am-244		1.5457E-20
Cm-242	4.9754E-08	3.9434E-07
Cm-243	3.5004E-06	2.7573E-05
Cm-244	2.6878E-04	2.1100E-03
Cm-245	1.1051E-04	8.6423E-04
Cm-246	2.4878E-05	1.9336E-04
Cm-247	4.8005E-07	3.7225E-06
Cm-248	5.5381E-08	4.2804E-07
Bk-249	1.2501E-10	-
Cf-249	1.2501E-10	-
Cf-250	1.2501E-10	-

model. Isotopic distributions of both sources are summarized in Table I.

The first source represents the isotopic mix of the initial core loading and is assigned to the reactor core control volume. Mathematically, its time-dependent source strength is defined as

$$r_1\left(t_{sim}\right) = R_1\delta\left(0\right), \tag{8}$$

where R_1 is the total number of atoms at the initial core loading. Since the atoms sampled from this source would start their history uniformly inside the reactor core, a PDF describing their remaining residence time is given by:

$$p_1\left(t_{rem}\right) = 1/t_r \tag{9}$$

The other source accounts for the addition of fresh fuel (TRU) to replace the consumed fuel. For this model, it was assumed that actinides could only be added as rapidly as fission products were being removed, to maintain a constant inventory in the reactor. More precisely, since each actinide fission results in two fission products, the rate of addition of actinides should be ½ the rate of removal of fission products. The fission product removal rate is determined by the inventory of fission products in the system. Under the assumption of a constant power level in the reactor, this can be calculated analytically:

$$\begin{aligned} \dot{F} &= \kappa P - \frac{\varepsilon \dot{C}}{I}F, \\ F\left(0\right) &= 0, \end{aligned} \tag{10}$$

where

F = total inventory of fission products [atoms]
P = desired power level [W]
κ = number of fission products produced for an average fission energy released $\approx 2\ atoms/180MeV$
ε = an efficiency of a fission product separation process
$\dot{C}$ = a processing capacity rate [atoms/day]
I = a total initial inventory [atoms]

Therefore,

$$F\left(t\right) = \frac{\kappa P I}{\varepsilon \dot{C}}\left(1 - e^{-\varepsilon \dot{C}/I}\right). \tag{11}$$

The feed rate of fresh TRU needed is also equivalent to the time-dependent source strength of the second source in the MCise simulation and given by:

$$\begin{aligned} r_2\left(t_{sim}\right) &= \frac{1}{2}\frac{\varepsilon \dot{C}}{I}F\left(t_{sim}\right) = \frac{\kappa P}{2}\left(1 - e^{-\frac{\varepsilon \dot{C} t_{sim}}{I}}\right), \\ 0 &< t_{sim} < T_{sim}. \end{aligned} \tag{12}$$

Since the feed stream always enters at the beginning of the reactor control volume, a PDF describing the remaining residence time of the feed is defined with a delta function,

$$p_2\left(t_{rem}\right) = \delta\left(t_r\right). \tag{13}$$

The CINDER90 nuclear data library used in this study has a 63 group energy structure and includes both transmutation reactions and fission reactions with fission product yields. The fission product yields are not explicitly dependent on the neutron flux spectrum, but are defined for a number of representative spectrum types – thermal, fast and high-energy – with the fast spectrum set used for this study. For some isotopes, spontaneous fission product yields are also given. CINDER does not provide fission product yields for all possible fission reactions. In such cases, when a fission reaction occurs, the product isotopes are assigned a placeholder isotopic identity, *unknown fission product*. This isotope is stable and neutronically transparent and will accumulate. An accumulation of this isotope could result in underestimating decay heat and specific activity of the system.

With all the necessary components defined, MCise employs the following algorithm to generate results.

1. A source is randomly chosen between two specified sources.
2. An initial atom is randomly sampled from a prescribed isotopic composition. Its entry time to and remaining residence time in the reactor core are determined from their respective PDF.

TABLE II. Model parameters used to determine feed rate of fresh TRU.

Parameter	Value	Description
P [MW_{th}]	3723	A F6 tally in MCNP is used to detect energy absorption in the reactor structure. The fusion source strength is taken to be 7.1e18 s^{-1}.
I	2.8293e29	A total number of atoms from the initial core loading
ε	100%	Assumed
$\dot{C}$ [day^{-1}]	I/100	Assumed
t_{sim} [days]	20,000	Assumed
t_r [days]	100	I/$\dot{C}$

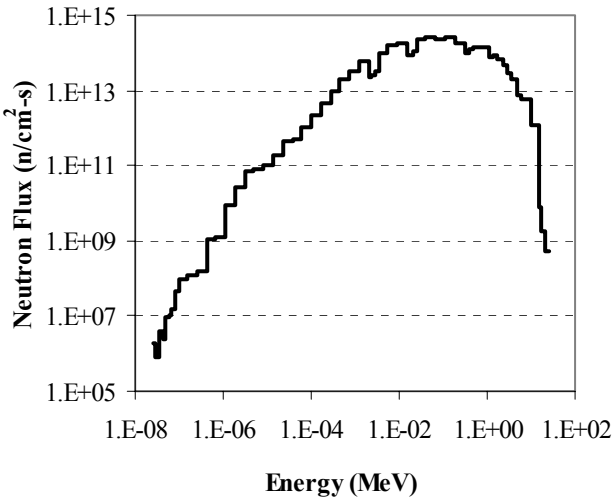

Fig. 3. Neutron energy spectrum with initial core loading isotopics.

3. As the simulated atom travels through the reactor core, its history is tracked according to the methodology described in the Background section. The history ends when the total simulation time is reached.

Upon exiting the reactor core, the simulated atoms go into the fission product extractor. All fission product atoms enter the sink and have their histories terminated. The other atoms continue their histories in Step 3.

II.D. Analysis Methodology

As in any fissile system, a calculation of the long term isotopics requires a tight coupling between the neutron transport calculation and the changing isotopics. In this system, justified in part by the constant replenishment of TRU fuel, the system was modeled with a constant neutron flux, both magnitude and energy spectrum, and assumed to have a constant power level. The validity of those assumptions as well as improvements for the analysis methodology in the next phase of this study will be discussed later.

To model neutron transport through the system, MCNP5 was used in a fixed source mode, with an isotropic neutron source positioned at the location of the z-pinch driven fusion source. This model implicitly accounts for the source neutrons that are lost through the upper and lower surfaces of the chamber and all results shown here include the effect of this neutron source loss.

In addition to these assumptions, several key parameters must be assumed to initiate an MCise simulation. Those parameters are summarized in Table II. The neutron flux at the initial core loading can then be obtained from MCNP in a subcritical mode and is shown in Fig. 3 using the same 63 energy groups defined by the CINDER data library.

Based on these assumptions and parameters, an MCise simulation was performed with a constant neutron flux in the reactor core for 20000 days of operation, with isotopic inventory results recorded every 20 days.

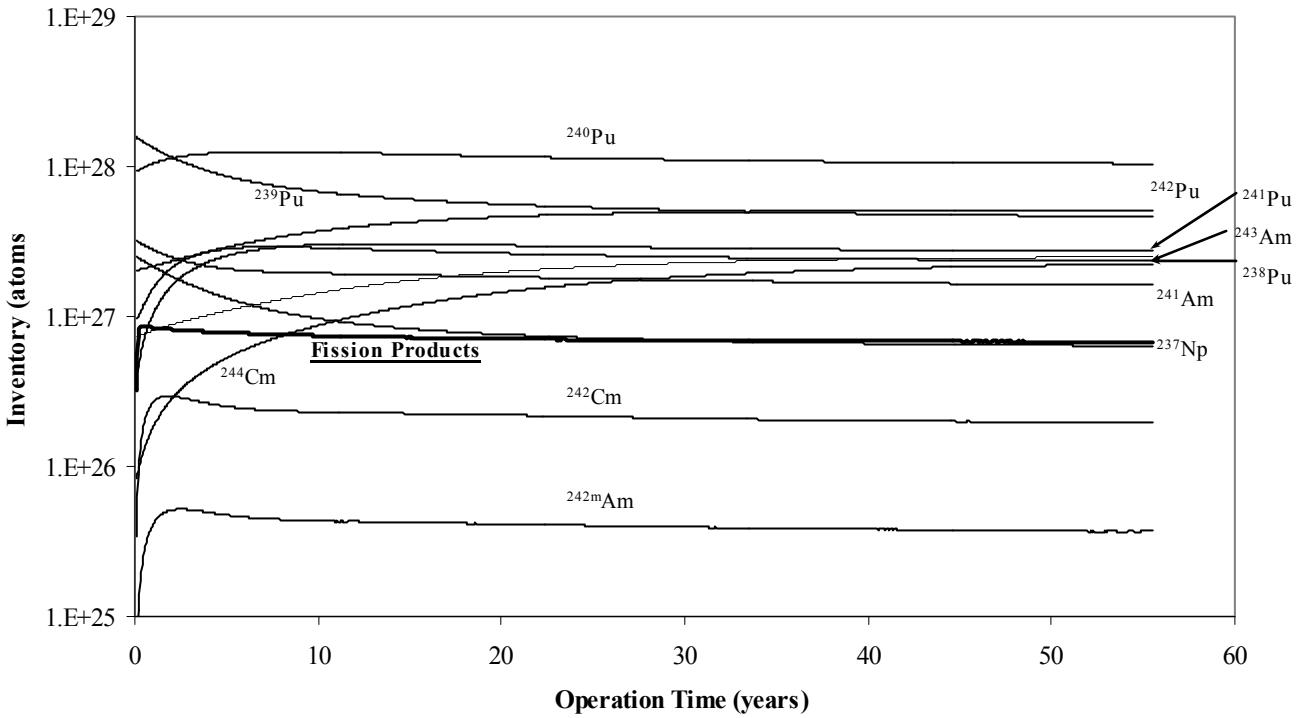

Fig. 4. Inventories of 10 most abundant isotopes and fission products.

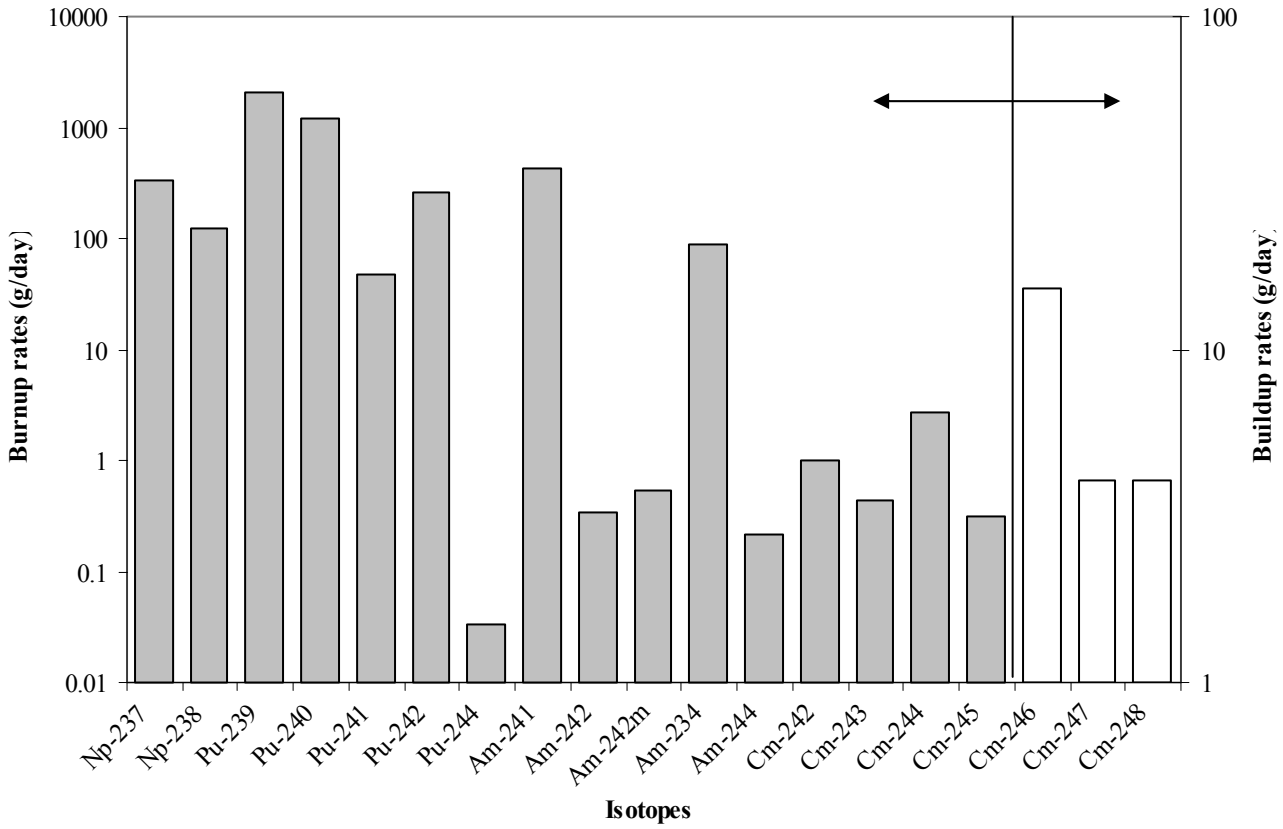

Fig. 5. Actinide Burnup/Buildup after 50 years of operation.

III. RESULTS

Results for the 11 isotopes with the highest concentrations are shown in Fig. 4. Fission products build up very rapidly at the beginning of the system's life. Many actinides approach equilibrium levels after about 10 years of operation. Other actinides gradually reach the equilibrium at times closer to 30 years. Instantaneous burnup and buildup rates of isotopes after 50 years of operation are considered to estimate the equilibrium plant performance at burning actinides (see Fig. 5).

The changes in actinide inventory result in variations in other important system parameters, specifically the multiplication factor, energy multiplication, and tritium breeding ratio (TBR) (see Fig. 6 through Figure 8). In the absence of a reactivity control strategy, this means that the total system power will vary over the operation lifetime of the system. This effect suggests an important limitation of these results. Although a constant neutron flux spectrum was used throughout the isotopic inventory calculation, any reactivity control strategy would result in a change in flux magnitude and flux spectrum in order to maintain a constant power level.

For example, the burnup/buildup rates shown in Fig. 5 total 4540 g/full-power day which represents a power level of approximately 4.5 GW_{th}. This can be scaled linearly to approximately 1100 kg/FPY at 3 GW_{th}, but the distribution of the burnup among the actinides may be different due to a changing flux spectrum. In terms of energy efficiency, about 13.5 MW is needed to operate for 20-MW z-pinch driven fusion source. Assuming a 33-percent thermal efficiency of the *In-Zinerator*, this inducing fusion power is about 1% of the output power.

Fig. 6 shows the variation in the multiplication factor due to changes in the isotopic inventory. Error bars represent 1σ statistical errors calculated by MCNP with material compositions based on the results of MCise. The rapid increase in fission products causes the multiplication factor to drop over the first 200 days. If the ^{6}Li inventory of the molten salt eutectic is not replenished, the ^{6}Li depletion results in a rapid increase in the multiplication factor. With ^{6}Li replenishment there is a slow increase in the multiplication factor.

Fig. 7 shows a similar trend for the energy multiplication of the system. Energy multiplication is defined as the total thermal power, measured in MeV per source neutron (total neutron and photon heating in entire problem geometry) divided by the average neutron energy

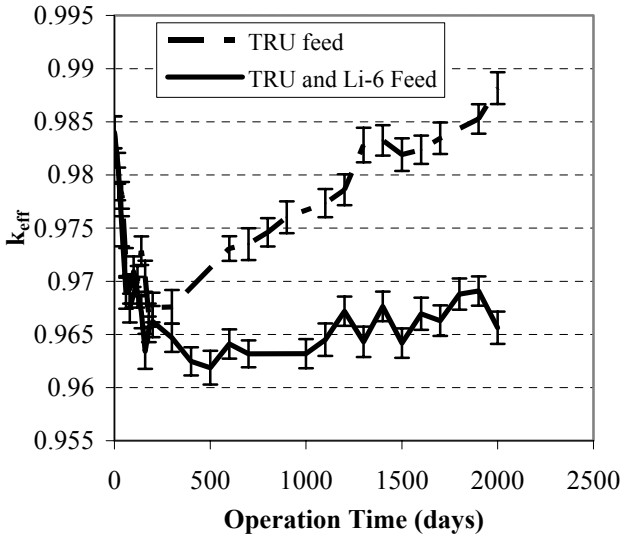

Fig. 6. Total system multiplication factor with and without replenishment of ^{6}Li.

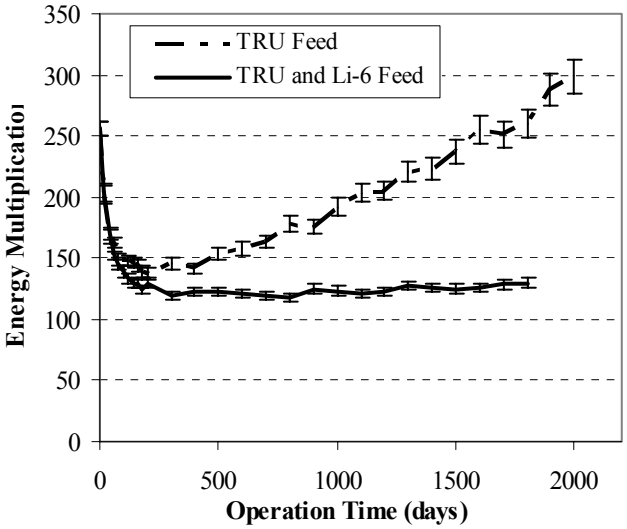

Fig. 7. Energy multiplication for whole system with and without replenishment of ^{6}Li.

as it leaves the target region (12.8 MeV per source neutron). ^{6}Li replenishment stabilizes the energy multiplication in the long term, although at ½ of the initial value. As mentioned above, reactivity control mechanisms must be introduced to ensure a constant system thermal power level. The changing power levels in this analysis result in an inconsistency with the TRU source function derived above. Since the power level is not constant, the fission product production rate is not constant. Fig. 4 shows that the fission product inventory initially reaches a level that is higher than the long term equilibrium level and must drop to that level.

A final system performance metric is the tritium breeding ratio (TBR). Once again, the changing isotopic inventory has a dramatic effect on the TBR, causing it to reach a level that is substantially below the level for tritium self-sufficiency (~1.1). It is clear that this drop is not due to the depletion of ^{6}Li since replenishment of this isotope does not restore the TBR to sufficient levels.

IV. FUTURE WORK

These results demonstrate that reactivity control mechanism will be necessary to ensure a constant energy multiplication over the life of the system. If such a mechanism were to preserve the neutron energy spectrum and magnitude, then the modeling assumptions made here would continue to be valid. However, preliminary results show that this may not be the case for all reactivity control options, requiring a tighter coupling between the neutronics, isotopics, and reactivity configuration of the system. One promising reactivity control mechanism for this system is to vary the effective thickness of the reflector. This can be done by the implementation of air-filled rods that enhance leakage of neutrons from the system. These rods are inserted into or pulled out of the reflector region to decrease or increase the reactivity, respectively. Once the reactivity control mechanism is established, an iterative calculation between MCNP and MCise must be employed to accurately determine burnup and activation of the materials. The following iterative scheme is proposed for the future work.

1. At time, t_i, use MCNP to ***iteratively*** find the reactivity control state, S_i, that achieves a desired energy multiplication for the current isotopic inventory state, I_i,
2. Use the neutron flux from step 1 with MCise to determine isotopic distributions for the next time step, I_{i+1}.

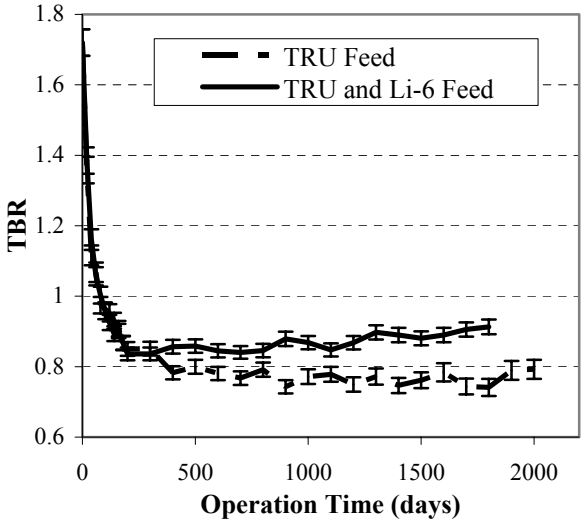

Fig. 8. Tritium breeding ratio (TBR) for whole system, with and without replenishment of ^{6}Li.

ACKNOWLEDGEMENTS

This work was funded, in part, by a grant from Sandia National Laboratories (Contract# 505658).

REFERENCES

1. B. B. CIPITI et al., "Fusion Transmutation of Waste: Design and Analysis of the In-Zinerator Concept", SAND2006-6590, November 2006.
2. L. EL-GUEBALY, B. CIPITI, P. WILSON, P. PHRUKSAROJANAKUN, R. GRADY, and I. SVIATOSLAVSKY, "Engineering Issues Facing Transmutation of Actinides in Z-Pinch Fusion Power Plant," these proceedings.
3. C.L. OLSON et al., "Z-pinch IFE Program Final Report for FY04", SAND2005-2742, April 2005.
4. P.P.H. WILSON and P. PHRUKSAROJANAKUN, "Analog Monte Carlo methods for simulating isotopic inventories in complex systems." *Nucl.Sci.Eng.*, **152**, 243-255. (2006)