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Preliminary Study of Time-Dependent Isotopic Inventory of the *In-Zinerator* Actinide Management System

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Efficient burnup 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 a 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 actinide inventory in the system is an essential input used to calculate the energy multiplications and neutron spectrum, as well as to design its 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 MCNP. 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.

Background

A Monte Carlo inventory simulation engine (MCise) has been developed and implemented for modeling activation of materials with complex processes and irradiation histories[1]. 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 an activation calculation of the eutectic fuel in the *In-Zinerator* because of the need to account for the on-line 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,

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

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

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

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

$$t_{rxn} = \frac{-\ln \xi}{\lambda_{eff}} = n_{rxn}\tau_{eff} ,$$

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

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

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

$$t_{rem} = t_{rem} - t_{rxn} .$$

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

$$n_{rxn} = n_{rxn} - n_{rem} .$$

For a calculation at a single time point, 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 Modeling Requirements

To fully specify an MCise model of a nuclear system requires describing the network of irradiation environments and flow paths between them and then including the sources of atoms.

An irradiation environment in MCise is defined by a *control volume.* The control volume is characterized by a neutron flux and a residence time, t_r. The neutron flux is expressed as a multi-group spectrum and is assumed to be constant throughout the control volume. The number of energy groups and energy group structure for the neutron flux must conform with those of the nuclear data format. 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 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 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-independent isotopic composition and a time-dependent function $r(t_{sim})$ characterizing the source strength. The total strength of each source, R_s, is defined by integrating $r(t_{sim})$ over the total simulation time T_{sim}, i.e.,

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

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 as 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}.

In-Zinerator MCise Model

A schematic of the MCise model of the *In-Zinerator* is illustrated in Figure 1. 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 to 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 during the initial stages of this 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 separation process and the flow distribution of each species can be adjusted to represent the real separation efficiencies.

There are two atom sources in the *In-Zinerator* model. Isotopic distributions of both sources are summarized in Table I.

The first one is the source representing 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

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

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:

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

The other source accounts for the addition of fresh fuel (TRU) to replace the consumed fuel.
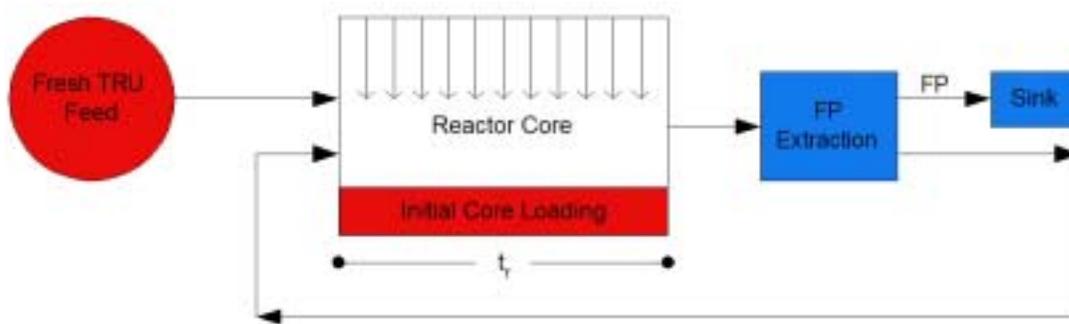
Figure 1. Schematic of In-Zinerator MCise model, showing sources in red and sinks in blue.

Table I

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	-

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 half 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:

$$\dot{F} = \kappa P - \frac{\varepsilon \dot{C}}{I} F ,$$
$$F(0) = 0,$$

where

$F =$ a total inventory of fission products [atoms]

$P =$ a desired power level [energy/time]

$\kappa =$ a number of fission products produced for an average fission energy released

$\approx \dfrac{2}{180 MeV}$ [atoms/energy]

$\varepsilon =$ an efficiency of a fission product separation process [dimensionless]

$\dot{C} =$ a processing capacity rate [atoms/time]

$I =$ a total initial inventory [atoms]

Therefore,

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

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:

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

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,

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

The CINDER90 nuclear data library is used in this study. It uses 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. For some isotopes, spontaneous fission product yields are also given. CINDER does not provide fission yields for all possible fission reactions. In such cases, when a fission reaction occurs, the product isotopes will be 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.

Now that all necessary MCise components are 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 and remaining residence time in the reactor core are determined from their respective PDF.
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.

Table II. Analysis Parameters

Parameter	Value	Description
P	3.7233e3 MW$_{th}$	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
$\dot{C}$	I/100 day^{-1}	Assumed
t_{sim}	20,000 days	Assumed
t_r	100 days	I/$\dot{C}$

4. 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.

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.

In addition to these assumptions, several key parameters must be assumed to initiate an MCise simulation. Those parameters are summarized in Table II. The detailed design description of the *In-Zinerator* can be found in [2]. The neutron flux at the initial core loading can be obtained from MCNP and is shown in Figure 2 using the same 63 energy groups defined by the CINDER data library.

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

Results

Results for the 10 isotopes with the highest concentrations are shown in Figure 3. 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 equilibrium at times closer to 30 years.

The tritium breeding ratio (TBR), energy multiplication and k_{eff} are calculated as a function of time and shown in Figure 4 to 6, respectively. Energy multiplication is calculated by dividing the total thermal power, measured in MeV per source neutron (total neutron and photon heating in entire problem geometry) by 12.8 MeV per source neutron (the average

neutron energy as it leaves the target region). Error bars in the figures represent 1σ statistical errors. These are calculated using MCNP with material compositions based on the results of MCise.

Original results based on a replenishment of only TRU do not take into account the substantial depletion of ^{6}Li during the operation. This depletion is partly responsible for a decrease in tritium breeding ratio and increase in energy multiplication. Results are also shown with a constant ^{6}Li inventory, based on the same MCise isotopic inventory results but with the ^{6}Li inventory maintained in the MCNP input file. As expected, the tritium breeding ratio increases and equilibrium is reached within about 1 year based on the energy multiplication and multiplication factor.

The rapid drop in energy multiplication at early times affects the fission product accumulation. At early times, the fission product inventory increases towards an equilibrium value that is a function of the power level. Since the power level drops on the same time scale that the fission product inventory is accumulating, the fission product inventory reaches a level that is higher than the long term equilibrium level and must drop to reach that level.

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, requiring a tighter coupling between the neutronics, isotopics, and reactivity configuration of the system.

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}.

References
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2. B. B. Cipiti, "Fusion transmutation of waste and the role of the In-Zinerator in the nuclear fuel cycle," technical report SAND2006-3522, Nuclear and Risk Technologies, Sandia National Laboratories.
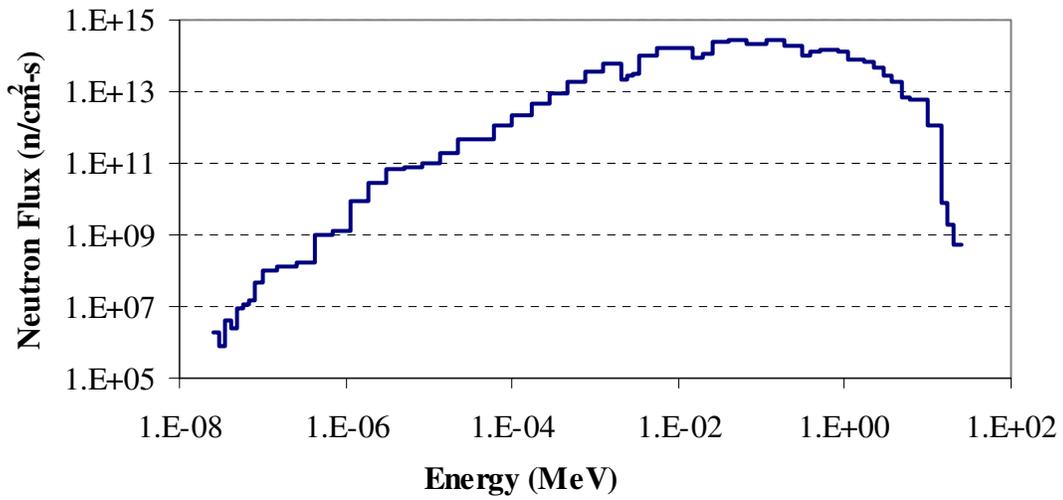
Figure 2. Neutron energy spectrum with initial core loading isotopics.
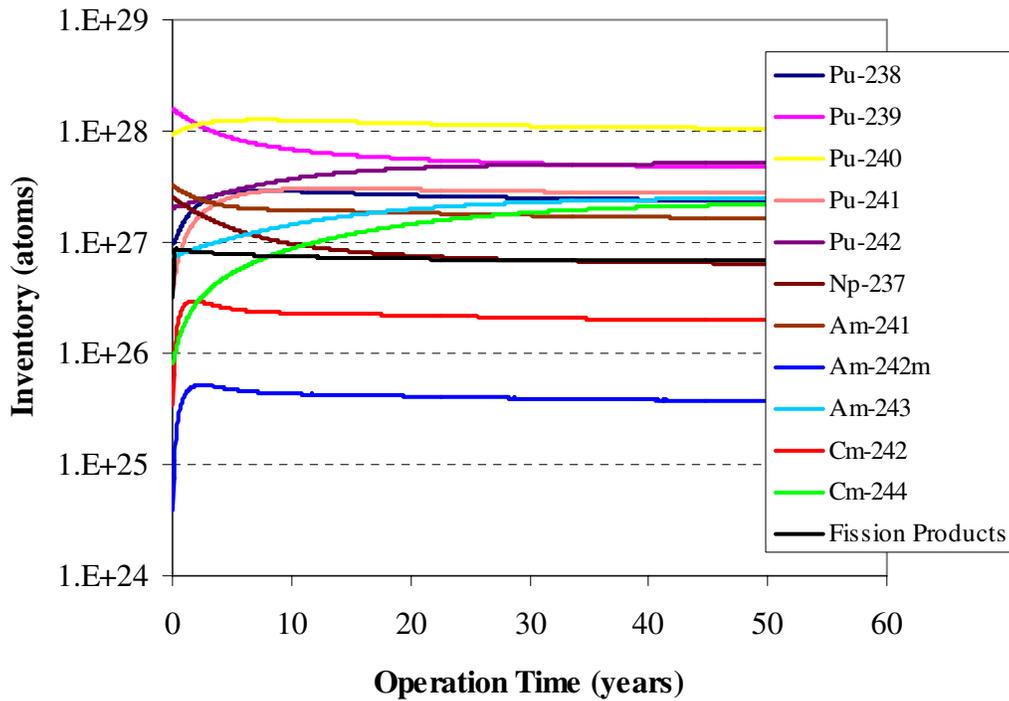
Figure 3. Inventories of 10 most abundant isotopes and fission products.
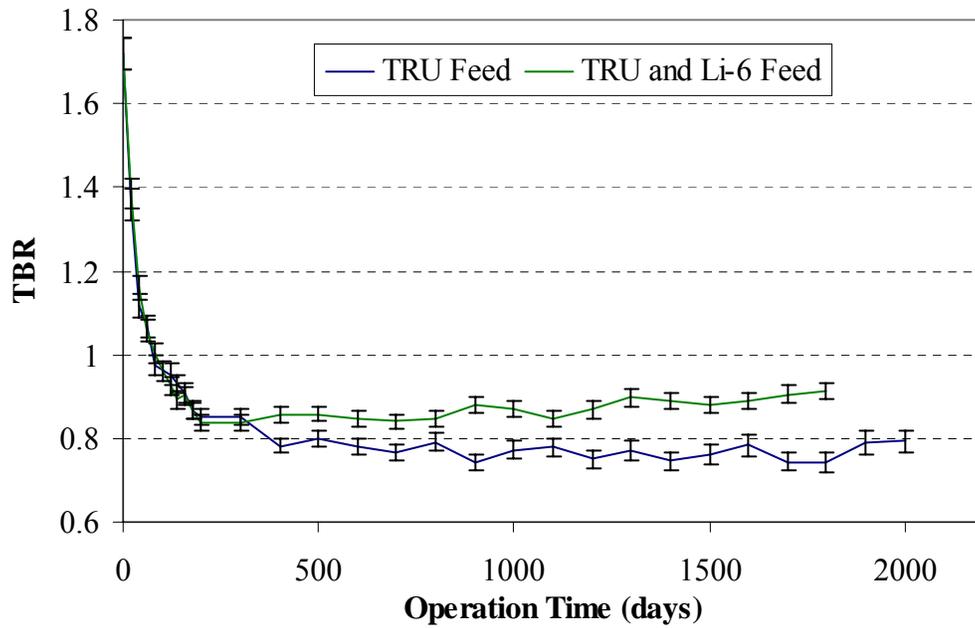
Figure 4. Tritium breeding ratio (TBR) for whole system, with and without replenishment of ^{6}Li.
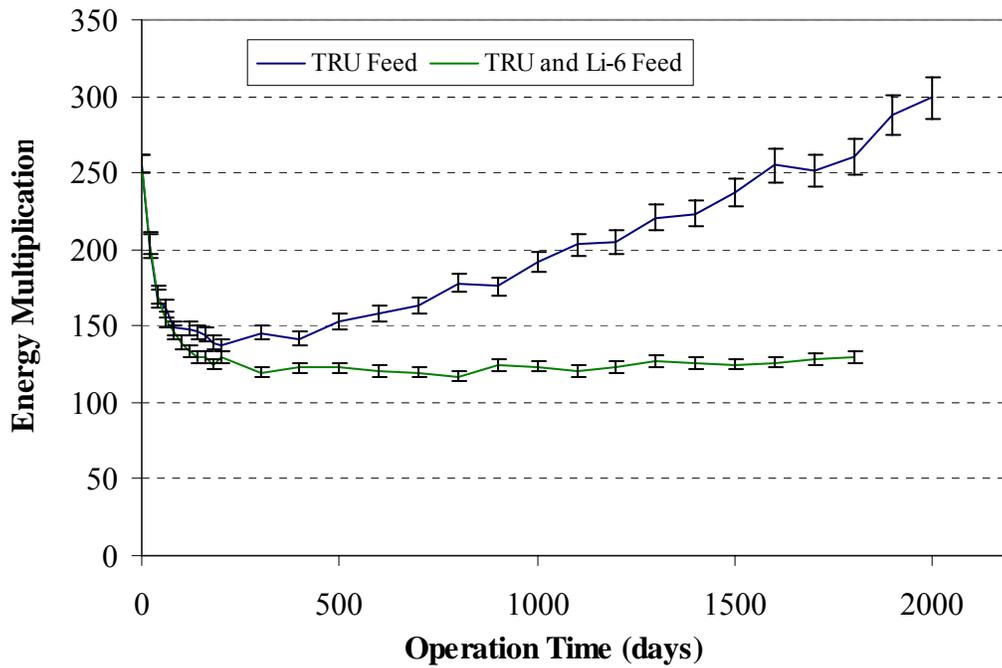
Figure 5. Energy multiplication for whole system with and without replenishment of ^{6}Li.
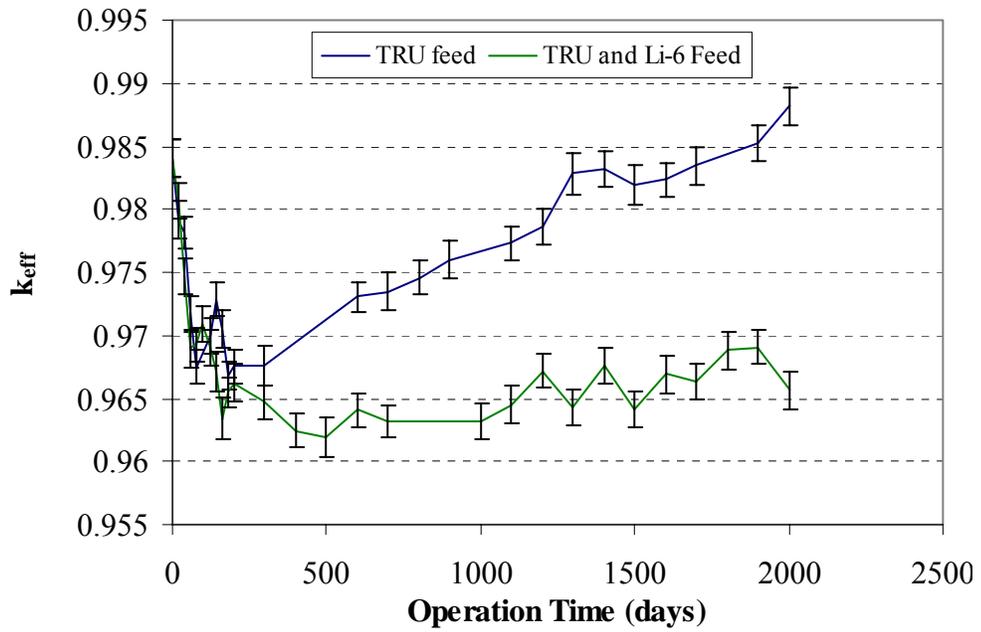
Figure 6. Total system multiplication factor with and without replenishment of ^{6}Li.