CONF-9509162--6

LA-UR- 95-1828

Title: A Repository Released-Dose Model for the Evaluation of Long-Lived Fission Product Transmutation Effectiveness

Author(s): J. Wiley Davidson

Submitted to: Global 1995, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems
September 11-14, 1995
Versailles - France - Palais Des Congres

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Los Alamos
NATIONAL LABORATORY

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

Form No 836 R5
ST 2629 10/91

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

A REPOSITORY RELEASED-DOSE MODEL FOR THE EVALUATION OF LONG-LIVED FISSION PRODUCT TRANSMUTATION EFFECTIVENESS

J. Wiley Davidson

Los Alamos National Laboratory
Systems Engineering & Integration Group
P.O. Box 1663, MS F607
Los Alamos, NM 87545

ABSTRACT

A methodology has been developed to quantify the total integrated dose due to a radionuclide species i emplaced in a geologic repository; the focus is on the seven long-lived fission products (LLFPs) . The methodology assumes continuous exposure water contaminated with species i at the accessible environment (i.e., just beyond the geologic barrier afforded by the geologic repository). The dose integration is performed out to a reference post-release time. The integrated dose is a function of the total initial inventory of radionuclide i the repository, the time at which complete and instantaneous failure of the engineered barrier (e.g., waste canister) in a geologic repository occurs, the fractional dissolution rate (from waste solid form) of radionuclide i in ground water, the ground water travel time to the accessible environment, the retardation factor (sorption on the geologic media) for radionuclide i , the time after radionuclide begins to enter the biosphere. In order to assess relative dose, the ratio of total integrated dose to that for a reference LLFP species j (e.g., ^{99}Tc) was defined. This ratio is a measure of the relative benefit of transmutation of other LLFPs compared to ^{99}Tc. This methodology was further developed in order to quantify the integrated dose reduction per neutron utilized for LLFP transmutation in accelerator-driven transmutation technologies (ADTT). This measure of effectiveness is a function of the integrated dose due to LLFP species i, the number of total captures in LLFP species i chain per LLFP nuclide fed to the chain at equilibrium, and the number of total captures in related transmutation product (TP) chains per capture in the LLFP species i chain. To assess relative transmutation effectiveness, the ratio of integrated dose reduction per neutron utilization to that for a reference LLFP species j (e.g., ^{99}Tc) was defined. This relative measure of effectiveness was evaluated for an example LLFP transmutation strategy.

INTRODUCTION ✓

Transmutation of the long-lived fission products (LLFPs) produced in uranium or plutonium burning as well as those present in spent reactor fuel can require a significant neutron excess in a fissioning system. Such an excess is difficult to obtain in a critical system; however, an accelerator-driven subcitical system allows the available neutron excess to vary with the degree of subcriticality. This excess is proportional to the fraction of the total neutron production which does not come from fission and thus, is proportional to the proton induced spallation neutron source. The cost impact of this neutron excess is large and therefore, a quantitative measure of the neutron requirements and transmutation effectiveness is necessary to evaluate the cost and performance trade-offs for the various LLFP transmutation strategies.

The fission multiplication factor for the system, k_{eff} is fundamentally an neutron economy parameter which measures the fraction of excess neutrons produced in fission which are required to sustain the fission reaction rate. For a critical system, k_{eff} must equal 1.0. k_{eff} can be expressed as

$$k_{eff} = \frac{\nu p_{NL}}{1 + \alpha_F + \alpha_{S/T} + \alpha_{FP} + \alpha_{LL}} \quad (1)$$

where

ν is the average number of neutrons liberated per fission,

p_{NL} is the system neutron non-leakage probability,

α_F is the number of system neutron captures in the fuel per fission in the fuel,

$\alpha_{S/T}$ is the number of system neutron captures in the structure and target per fission in the fuel,

α_{FP} is the number of system neutron captures in the parasitic fission products per fission in the fuel, and

α_{LL} is the number of system neutron captures in the long-lived fission products per fission in the fuel.

The parameters in this expression are system values and can either describe the neutron economy in replicated, multi-function target-blanket modules or the neutron economy averaged over target-blanket systems with different functions (e.g., thorium-uranium energy production, plutonium burning, LLFP transmutation only, etc.). The parameter α_F is characteristic of the nuclear fuel and spectrum while $\alpha_{S/T}$ is strongly dependent on the target-blanket design(s). The parameters α_{FP} and α_{LL} determined by the fuel processing scheme and long-lived fission product transmutation strategy, respectively.

Methodology for Calculating Relative Doses for Radionuclides Released to the Accessible Environment

If complete and instantaneous failure of the engineered barrier (e.g., waste canister) in a geologic repository occurs at time t_E after emplacement, the amount of radionuclide species i, I_i^E, available at that time to enter groundwater which may have intruded into the repository is given by

$$I_i^E = I_i^o \, e^{-\lambda_i t_E} \quad (2)$$

where

I_i^o is the total initial inventory (nuclides) of radionuclide i the repository,

λ_i is the decay constant for radionuclide i (sec^{-1}), and

t_E is the time at which the engineered barrier (e.g., waste canister) fails (sec).

The rate at which a radionuclide i enters groundwater within the repository at post-failure time $t\leq$ after the failure of the engineered barrier $R_i^E(t\leq)$ (nuclides/second) is given by

$$R_i^E(t'') = I_i^o \, e^{-\lambda_i t_E} f_i \, e^{-\lambda_i t''} \qquad (t_E < t\leq < t_E + t_{Di}) \quad (3)$$

where

f_i is the fractional dissolution rate of radionuclide i in ground water (sec^{-1}),

$t\leq$ ($t\leq = t-t_E$) is the time after failure of the engineered barrier (sec).

The value of f_i is given by $1/t_{Di}$ where t_{Di} is the total dissolution time for species i. The water solubility of species i (in its preferred chemical form) and groundwater exposure rates are two of the most important parameters determining t_{Di}.

During the transport of the radionuclide in the ground water, decay and sorption on the geologic media decrease the amount available for release to the biosphere. Thus the release rate (nuclides/second) at post-release time t' of radionuclide i from a geologic repository $R_i^R(t')$ can be expressed as

$$R_i^R(t') = I_i^o\, e^{-\lambda_i t_E}\, f_i\, e^{-\lambda_i K_i t_W}\, e^{-\lambda_i t'} \quad (t_E+t_W < t' < t_E+t_W+t_{Di}) \tag{4}$$

where

t_W is the ground water travel time to the accessible environment (sec),

K_i is the retardation factor for radionuclide i (unitless), and

t' ($t' = t-t_E-t_W$) is the time after radionuclide i begins reaching accessible environment (sec).

At post-release times greater than the sum of the dissolution time plus the water travel time (t_W+t_{Di}), the release rate $R_i^R(t')$ is zero.

The total amount of radionuclide i (nuclides) in the accessible environment $I_i(t')$ at post-release time t' ($t_E+t_W < t' < t_E+t_W+t_{Di}$) is given by the solution to the equation

$$\frac{d[I_i(t')]}{dt'} = R_i^R(t') - \lambda_i I_i(t') \tag{5}$$

The solution to Eq. 5 is

$$I_i(t') = I_i^o\, e^{-\lambda_i t_E}\, f_i\, e^{-\lambda_i K_i t_W}\, t'\, e^{-\lambda_i t'} \tag{6}$$

Similarly at post-release time t' ($t' > t_E+t_W+t_D$), $I(t')$ is given by

$$I_i(t') = I_i^R\, e^{-\lambda_i t'} \tag{7}$$

The parameter R_i^R is the total amount of radionuclide i in the accessible environment at the time the release of that species is complete. It is given by

$$I_i^R = I_i^o\, e^{-\lambda_i t_E}\, f_i\, e^{-\lambda_i K_i t_W}\, t_{Di}\, e^{-\lambda_i t_{Di}} \tag{8}$$

A volume (m^3), of water V^{di} which represents the dilution (and dispersion) of radionuclide i at release ($t = t_E+t_W$) to the biosphere, is used to calculate the concentration of species i in contaminated water in the accessible environment. No further dilution is assumed beyond time and thus calculated concentrations represent upper bounds. The total number of decays or disintegrations per unit volume (dis/m^3) of contaminated water in the accessible environment at "reference" post-release time $t' = t_R$ due to radionuclide i, $r_i(t_R)$, is given by

$$r_i(t_R) = \int_0^{t_R} \frac{\lambda_i \, I_i(t')}{V^d_i} \tag{9}$$

For $t_R < t_{Di}$ and $t_R > t_{Di}$, Eq. 9 becomes, respectively,

$$\begin{aligned} r_i(t_R) &= \frac{\lambda_i}{V^d_i} \, I_i^o \, e^{-\lambda_i t_E} \, f_i \, e^{-\lambda_i K_i t_W} \int_0^{t_R} t' \, e^{-\lambda_i t'} dt' \quad t_R < t_{Di} \\ &= \frac{\lambda_i}{V^d_i} \, I_i^o \, e^{-\lambda_i t_E} \, f_i \, e^{-\lambda_i K_i t_W} \left[\int_0^{t_{Di}} t' \, e^{-\lambda_i t'} dt' + \int_{t_{Di}}^{t_R} t_{Di} \, e^{-\lambda_i t'} dt' \right] \quad t_R > t_{Di} \end{aligned} \tag{10}$$

The analytic solution to Eq. 10 is given by

$$\begin{aligned} r_i(t_R) &= \frac{I_i^o}{\lambda_i \, V^d_i} \, e^{-\lambda_i t_E} \, f_i \, e^{-\lambda_i K_i t_W} \left[1 - e^{-\lambda_i t_R} - \lambda_i t_R \, e^{-\lambda_i t_R} \right] \quad t_R < t_{Di} \\ &= \frac{I_i^o}{\lambda \, V^d} \, e^{-\lambda_i t_E} \, f_i \, e^{-\lambda_i K_i t_W} \left[1 - e^{-\lambda_i t_{Di}} - \lambda_i t_{Di} \, e^{-\lambda_i t_R} \right] \quad t_R > t_{Di} \end{aligned} \tag{11}$$

These expressions can be simplified to give

$$r_i(t_R) = \frac{1}{\lambda_i} \, \beta_i(t_E) \, \gamma_i(t_W) \, \delta_i(t_R) \, I_i^o \tag{12}$$

where

$$\beta_i(t_E) = f_i \, e^{-\lambda_i t_E} \quad , \tag{13}$$

$$\gamma_i(t_W) = \frac{e^{-\lambda_i K_i t_W}}{V^d_i} \quad \text{, and} \tag{14}$$

$$\begin{aligned} \delta_i(t_R) &= 1 - e^{-\lambda_i t_R} - \lambda_i t_R \, e^{-\lambda_i t_R} \quad t_R < t_{Di} \\ &= 1 - e^{-\lambda_i t_{Di}} - \lambda_i t_{Di} \, e^{-\lambda_i t_R} \quad t_R > t_{Di} \end{aligned} \tag{15}$$

The total integrated dose due to continuous exposure to contaminated water at the accessible environment at reference post-release time t_R due to radionuclide i $D_i(t_R)$ (rems) is simply

$$D_i(t_R) = c_i \, r_i(t_R) \tag{16}$$

where

c_i is the dose conversion factor for radionuclide i (rem per dis/m^3).

If Eqs. 12 and 16 are combined, the resulting expression is

$$D_i(t_R) = \frac{c_i}{\lambda_i} \beta_i(t_E) \gamma_i(t_W) \delta_i(t_R) I_i^o \quad . \tag{17}$$

The values of the radionuclide-dependent parameters for the seven LLFPs are given in Table 1. The values of f_i and K_i for ^{99}Tc, ^{129}I, and ^{135}Cs were taken from a paper by Pigford[3] which focused on these three radionuclides (in addition to the actinides) because of their large solubilities in water. The values of f_i for the other four LLFPs were taken to be the same as that for highly soluble ^{99}Tc while the values of K_i were assumed to be those of highly mobile ^{129}I. The values represent very conservative assumptions for ^{79}Se, ^{93}Zr, ^{107}Pd, and ^{126}Sn release. Note that values of the parameter V^d_i are not available and subsequent development of this methodology assumes that the ratio of this parameter for any two LLFP species is approximately equal to 1.0 based on Pigford's paper.

Table 1 Representative radionuclide-dependent parameters for the integrated dose model with data from Pigford.

	I_i^o (g-a/MTHM)	λ_i (sec^{-1})	f_i (sec^{-1})	K_i
^{79}Se	7.43E-02	3.38E-13	7.92E-12*	1.00E+00†
^{93}Zr	7.73E+00	1.46E-14	7.92E-12*	1.00E+00†
^{99}Tc	7.79E+00	1.03E-13	7.92E-12	1.60E+00
^{107}Pd	2.04E+00	3.38E-15	7.92E-12*	1.00E+00†
^{126}Sn	2.17E-01	2.20E-13	7.92E-12*	1.00E+00†
^{129}I	1.39E+00	1.40E-15	7.92E-12	1.00E+00
^{135}Cs	2.22E+00	9.55E-15	7.92E-12	6.10E+02
* conservatively assumed to be same as technetium † conservatively assumed to be same as iodine				

The dose conversion factor c_i used in this analysis is calculated as the absorbed dose in an infinite volume of water per unit concentration of radionuclide species i. The average energy per decay includes beta and gamma energies of parent and daughters assuming an RBE of 1.0. Biological consolidation or retention is not included. The dose conversion factors used for the LLFPs are presented in Table 2.

Table 2 Dose conversion factors based on total absorbed decay energy in an infinite volume of water.

LLFP	c_i (rem per dis/m^3)	E_{avg} (MeV)	Contributing Daughters
^{79}Se	8.55E-16	0.0533	None
^{93}Zr	8.08E-16	0.0504	^{93m}Nb
^{99}Tc	1.56E-15	0.0973	None
^{107}Pd	2.14E-16	0.0133	None
^{126}Sn	4.34E-14	2.7074	^{126m}Sb, ^{126}Sb
^{129}I	1.40E-15	0.0876	None
^{135}Cs	1.12E-15	0.0700	None

In order to assess relative dose, the ratio of total integrated dose $D_i(t_R)$ to that for a reference LLFP species j (e.g., ^{99}Tc) is defined. This ratio $H_{iTc}(t_R)$ is a measure of the relative benefit of management of other LLFPs compared to ^{99}Tc. The ratio is given by

$$H_{ij}(t_R) = \frac{c_i\, \beta_i(t_E)\, \gamma_i(t_w)\, \delta_i(t_R)}{c_j\, \beta_j(t_E)\, \gamma_j(t_w)\, \delta_j(t_R)}\, \frac{\lambda_j}{\lambda_i}\, \frac{I_i^o}{I_j^o} \quad . \tag{18}$$

Species for which decay daughter chains, particularly the actinides are large contributors to the total dose assigned to the original precursor to not lend themselves to this treatment. For LLFPs with a single significant daughter in equilibrium (e.g. ^{126}Sb with ^{126}Sn and ^{93m}Nb with ^{93}Zr), c_i (Table 2) accounts for the dose multiplier associated with the daughter.

Pigford[3] defined a similar "relative dose index" parameter as a measure of the dose rate of a released radionuclide species relative to that of the dominant LLFP species ^{99}Tc; the dose rate was taken at the boundary to the biosphere and at the time of initial release. The parameter accounts for both decay and sorption on rock as the species migrate from the repository and thus is a function of the water travel time to the biosphere boundary. The "relative dose index" has the form

$$\frac{D_i}{D_j} = \frac{M_i\, f_i\, e^{-\lambda_i K_i t_w}\, C_i\, W_i}{M_j\, f_j\, e^{-\lambda_j K_j t_w}\, C_j\, W_j} \quad . \tag{19}$$

where f_i, t_W, and K_i are defined as above and

D_i is a dose rate contribution (rem/yr) from radionuclide species i,

M_i is the inventory (Ci) of radionuclide species i emplaced in the repository,

C_i is a dose conversion factor (average rem/yr per Ci/m^3 in H_2O) from radionuclide species i, and

W_i is a dilution/dispersion rate (m^3/yr) of radionuclide species i as it migrates to the biosphere.

This ratio is a measure of the relative dose rate of radionuclide species at the boundary to the biosphere and at the time of initial release. It does not reflect either the time-dependent dose rate or integral dose.

Pigford used the "relative dose index", in a recent assessment of the impact of actinide and fission product transmutation on waste repositiory performance[3], to show that some of the LLFPs were much more significant dose contributors to the biosphere in releases from a repository than the major actinides. These results were largely based on the high solubilities of the these LLFPs with respect to the actinides. The actinide solubilities used by Pigford were based on actinide metals; however, recent Yucca Mountain peformance assessments[4] recognize that the oxide solubilites are the relavant values. These values are from 10^3 to 10^5 larger than those use by Pigford (see Table 3).

Table 3. Actinide Solubilities (moles/liter) Assumed for Repository Assessments

Material	1991 Value	1993 Value
Uranium	$10^{-7.7}$	$10^{-4.5}$
Neptunium	10^{-9}	10^{-4}
Plutonium	$10^{-12.4}$	10^{-7}
Americium	$10^{-8.2}$	10^{-7}

If Pigford's "relative dose index" methodology is modified for these more recent actinide solubilities, the actinides become larger contributers to the repository release dose, but the LLFPs still remain as equally significant contributors. The results of this modification to the Pigford analyses are presented in Fig. 1.

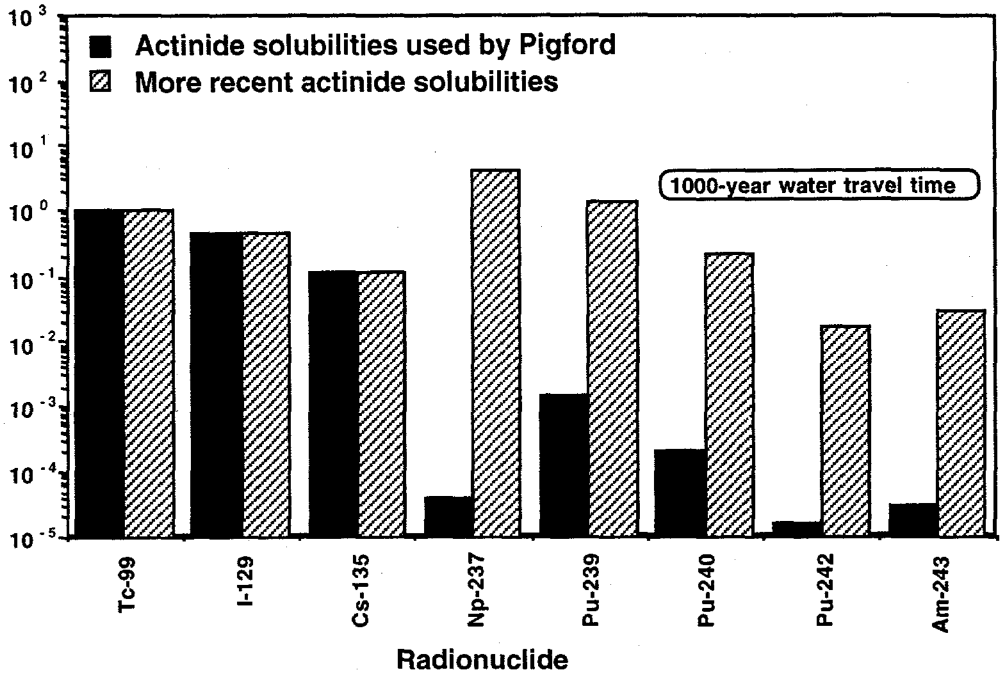

Figure 1. Relative Dose Index for repository release (and a water travel time of 1000 years) of dominant actinides and LLFPs calculated by Pigford and modifed for newer actinide solubilities.

Contributions to Reactivity in an Accelerator-Driven Subcritical Transmutation System

For a system fission power P_f expressed in fissions per second, the accelerator beam current requirement I_b expressed in protons per second is given by

$$I_b = \frac{P_f \bar{\nu}}{y_{n/p} M_S} \tag{20}$$

where

ν is the average number of neutrons liberated per fission,

$y_{n/p}$ is the net spallation target yield (neutrons/proton), and

M_S is the system source neutron multiplication.

The source neutron multiplication M_S is given by

$$M_S = \frac{k_{eff}}{1 - k_{eff}} \tag{21}$$

where k_{eff} is fission multiplication factor for the system can be expressed as

$$k_{eff} = \frac{\nu p_{NL}}{1 + \alpha_F + \alpha_{S/T} + \alpha_{FP} + \alpha_{LL}} \tag{22}$$

where

ν the number of neutrons produced per fission,

p_{NL} is the system neutron non-leakage probability,

α_F is the number of system neutron captures in the fuel per fission in the fuel,

$\alpha_{S/T}$ is the number of system neutron captures in the structure and target per fission in the fuel,

α_{FP} is the number of system neutron captures in the parasitic fission products per fission in the fuel, and

α_{LL} is the number of system neutron captures in the long-lived fission products per fission in the fuel.

The parameters in this expression are system values and can either describe the neutron economy in replicated, multi-function target-blanket module or averaged over target-blanket systems with different functions (e.g., plutonium burning, LLFP transmutation, etc.). The parameters α_F is characteristic of the nuclear fuel and spectrum while $\alpha_{S/T}$ is strongly dependent on the target-blanket design(s). The parameters α_{FP} and α_{LL} determined by the fuel processing scheme and long-lived fission product transmutation strategy, respectively.

F.1 Contribution of Parasitic Fission Product Absorption to Reactivity

The parameter a_{FP} is determined by the fission product cross sections and the relative rates of fission product species production from fission and recovery in processing; it varies as

$$\alpha_{FP} \propto \phi \, \tau \, \bar{\sigma}^a_{FP} \tag{23}$$

where

ϕ is the average neutron flux seen by the fission product containing fuel,

$\sigma^a{}_{FP}$ is an averaged 'lump" fission product absorption cross section, and

τ is a characteristic removal processing time for the fission products.

The value of α_{FP} varies as the fission product isotopics, and hence $\sigma^a{}_{FP}$, shift with time; however, an equilibrium value is reached in times which are short with respect to blanket module lifetimes. The characteristic fission product processing time τ is defined as

$$\frac{1}{\tau} = \sum_{i} \frac{f_i}{\tau_i} \qquad (24)$$

where

f_i is the fraction of the fission product concentration which is element species i, and

τ_i is the processing time for element species i.

Contribution of Long-Lived Fission Product Transmutation to Reactivity

The values of α^i_{LL} are determined by the relative isotopic feeds and activation chains for each species. These chains are illustrated in the following figure for major long-lived fission products.

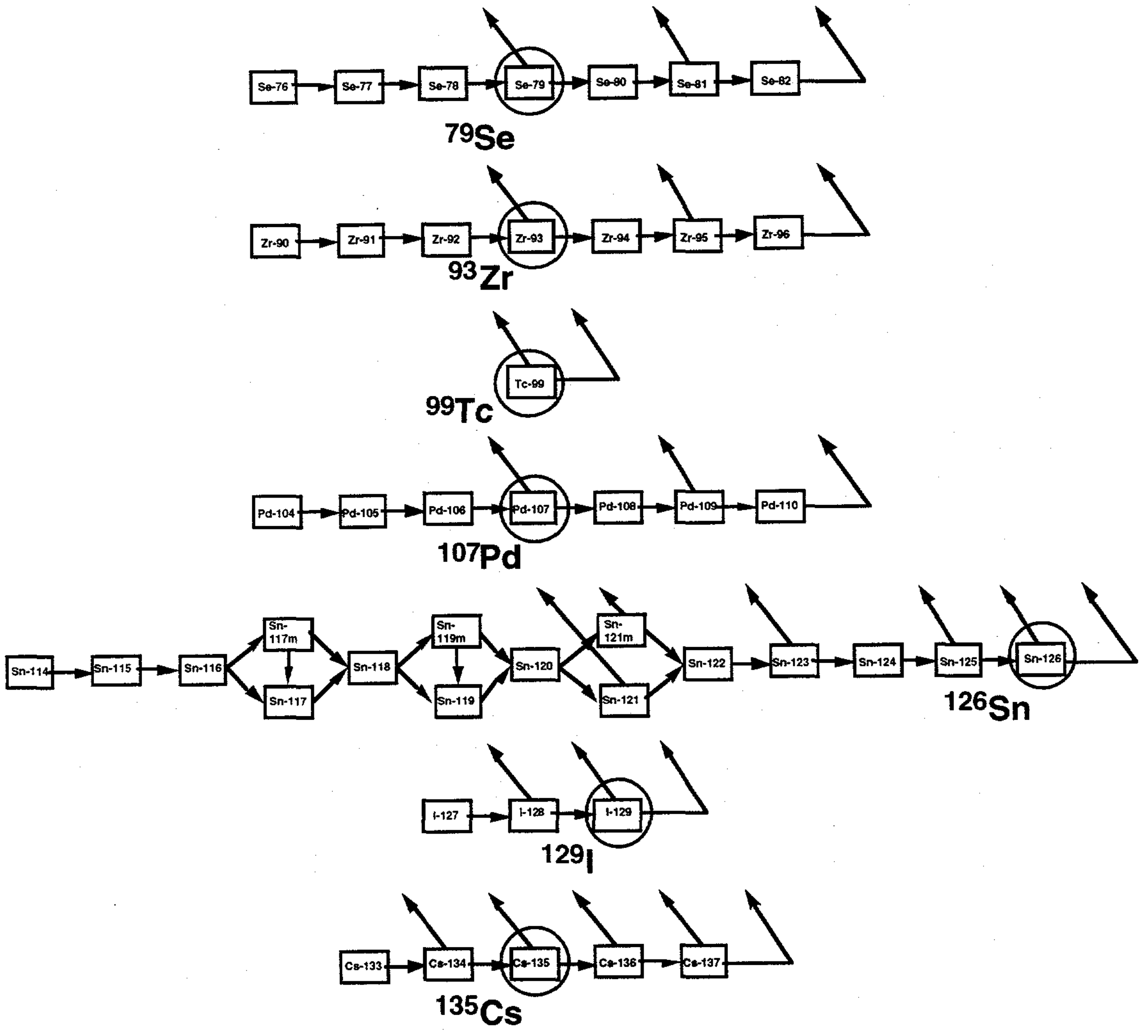

Figure 2. Long-Lived Fission Product Chains

Long-lived fission products can be introduced into the ABC/ATW system via internal feed (resulting from fissioning of plutonium and other actinides in the system) and/or through external feed of selected products. Representative isotopic and elemental feeds for such long-lived fission products are determined through ORIGEN2 calculations. The spent fuel concentrations for each LLFP species were those in PWR/U fuel exposed for 33 GW-d and cooled for 10 years. Isotopic feed is then expressed as moles of spent-fuel LLFP chain species i per mole of spent-fuel actinide fission in a system burning both actinides and LLFPs in equilibrium. The internal production isotopics for each LLFP species were calculated as a function of flux level and characteristic processing time in an equilibrium ATW actinide fuel where over 85% of the fission occurs in ^{239}Pu. Isotopic feed is then expressed as moles of LLFP chain species i recovered in processing per mole of actinide fission in a system burning actinides at equilibrium. The total calculated chain feed for both elemental and isotopic (100% enriched) feeds are presented in Table 4.

Table 4. Total chain feed for equilibrium LLFP chain calculations.

Isotopic Feed (moles/mole of fission)			
LLFP Isotope	Internal Only	External Only	External+ Internal
^{79}Se	3.89E-04	1.83E-03	2.22E-03
^{93}Zr	3.56E-02	1.90E-01	2.26E-01
^{99}Tc	4.83E-02	1.92E-01	2.40E-01
^{107}Pd	3.38E-02	5.02E-02	8.40E-02
^{126}Sn	1.69E-03	5.34E-03	7.02E-03
^{129}I	9.81E-03	3.41E-02	4.39E-02
^{135}Cs	1.00E-02	5.47E-02	6.47E-02
Elemental Feed (moles/mole of fission)			
LLFP Element	Internal Only	External Only	External+ Internal
selenium	4.31E-03	2.15E-02	2.58E-02
zirconium	3.32E-01	2.11E+00	2.45E+00
technetium	4.83E-02	1.92E-01	2.40E-01
palladium	4.96E-01	1.01E+00	1.50E+00
tin	7.71E-03	5.19E-02	5.96E-02
iodine	1.36E-02	4.49E-02	5.85E-02
cesium	1.37E-01	5.75E-01	7.13E-01

The value of α^{i}_{LL} for internal recycle is strongly dependent on the relative values of the average neutron flux seen by the fission product containing fuel and of the characteristic removal processing time for the LLFP. At long relative residence times, significant transmutation of the LLFP occurs in the fuel before recovery and appears as a contribution to α_{FP} rather than to α_{LL}. The total impact on reactivity for LLFP species i is given by the sum of α^{i}_{FP} and α^{i}_{LL}.

Neutron Economy Impacts Associated with LLFP Integrated Dose Reduction

The total integrated dose $d_i(t_R)$ per nuclide emplaced in a geologic repository per unit volume of contaminated water released to the accessible environment at reference post-release time t_R due to species i (rems/nuclide) can be expressed as

$$d_i(t_R) = \frac{D_i(t_R)}{I_i^o} = \frac{c_i}{\lambda_i}\,\beta_i(t_E)\,\gamma_i(t_W)\,\delta_i(t_R) \qquad (25)$$

form Eq. 16. A quantity w_i can be defined as the integrated LLFP dose reduction per neutron capture per transmutation of LLFP species i given by

$$w_i = \frac{d_i(t_R)}{\chi^i_{LL}} \qquad (26)$$

where the parameter χ^i_{LL} is defined as the number of neutron captures required for transmutation of LLFP species i. This latter value is not 1.0 because other isotopes of species i as well as other elemental transmutation products (TPs) of species i may be present as additional absorbers. The parameter χ^i_{LL} is given by

$$\chi^i_{LL} = g^i_{LL}\left(1+g^i_{TP}\right) \qquad (27)$$

where

g^i_{LL} is the number of total captures in LLFP species i chain per LLFP nuclide fed to the chain at equilibrium and

g^i_{TP} is the number of total captures in related TP chains per capture in the LLFP species i chain.

This is value of g^i_{LL} is illustrated in Figs. 3 and 4 for transmutation of the LLFP ^{135}Cs. Note that for elemental fission-product cesium transmutation (Fig.4), a total of 3.789 ^{135}Cs nuclides of are "burned" per ^{135}Cs nuclide "fed" to the system. Also, 10.518 neutron captures in the equilibrium cesium chain are required per ^{135}Cs nuclide "fed" to the system. For isotopically pure feed (Fig. 4), only 1.0007 neutron captures in the equilibrium cesium chain are required per ^{135}Cs nuclide "fed" to the system.

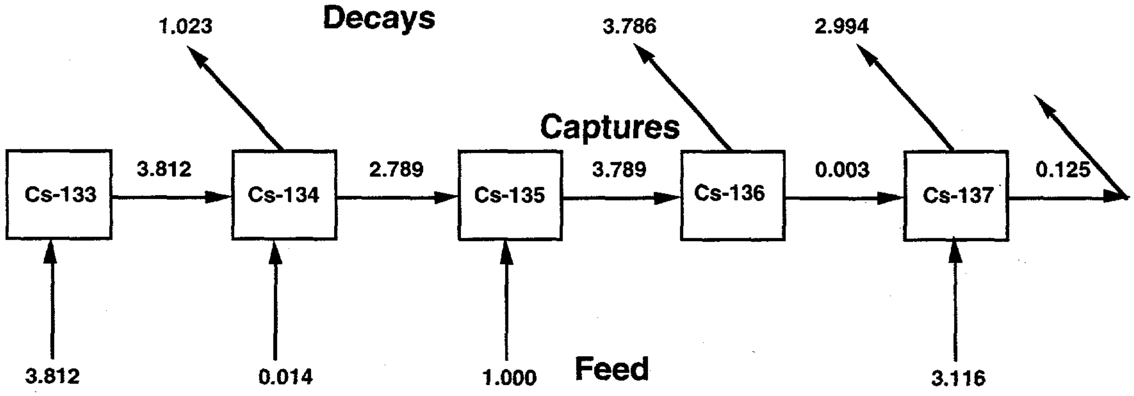

Figure 3 Equilibrium transmutation chain for ^{135}Cs assuming spent-fuel elemental fission-product cesium as feed to transmutation system.

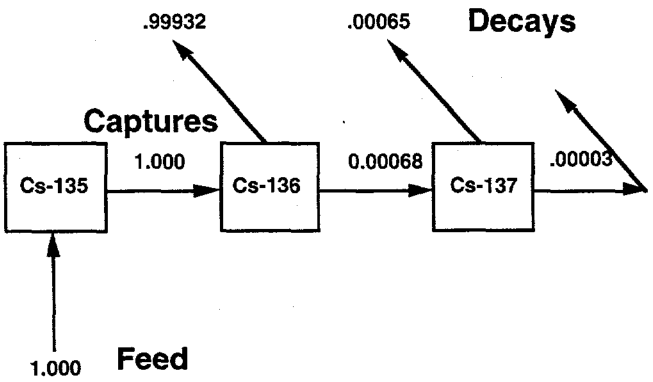

Figure 4 Equilibrium transmutation chain for ^{135}Cs assuming 100% isotopically enriched ^{135}Cs as feed to transmutation system.

Values of g^i_{LL} for spent-fuel fission-product feed are presented in Table 5.

Table 5 The number of neutron captures required for transmutation of LLFP species i for spent fuel (10-year-old, 33 GW-d) feed isotopics, a neutron flux level of $4x10^{14}$, and a CANDU thermal spectrum.

	g^i_{TP} Isotopic Feed	g^i_{TP} Elemental Feed
^{79}Se	1.999	11.740
^{93}Zr	2.002	11.110
^{99}Tc	1.000	1.000
^{107}Pd	2.000	20.060
^{126}Sn	1.000	9.696
^{129}I	1.000	1.316
^{135}Cs	1.001	10.520

The parameter g^i_{TP} accounts for the absorption in transmutation products (TPs) of species i which are not the same element. This allows separate recovery and recycle processing for the LLFP and TP elements. The TPs can contribute significantly to the total absorption associated with elimination of species i; this absorption is strongly dependent on the rate at which the LLFP transmutation loops are processed for TP removal relative to their production rate or neutron flux level. The predominant absorption is in the element of next higher Z (e.g. Ruthenium for Technetium). The value of g^i_{TP} for ^{99}Tc transmutation is presented in Fig. 5.

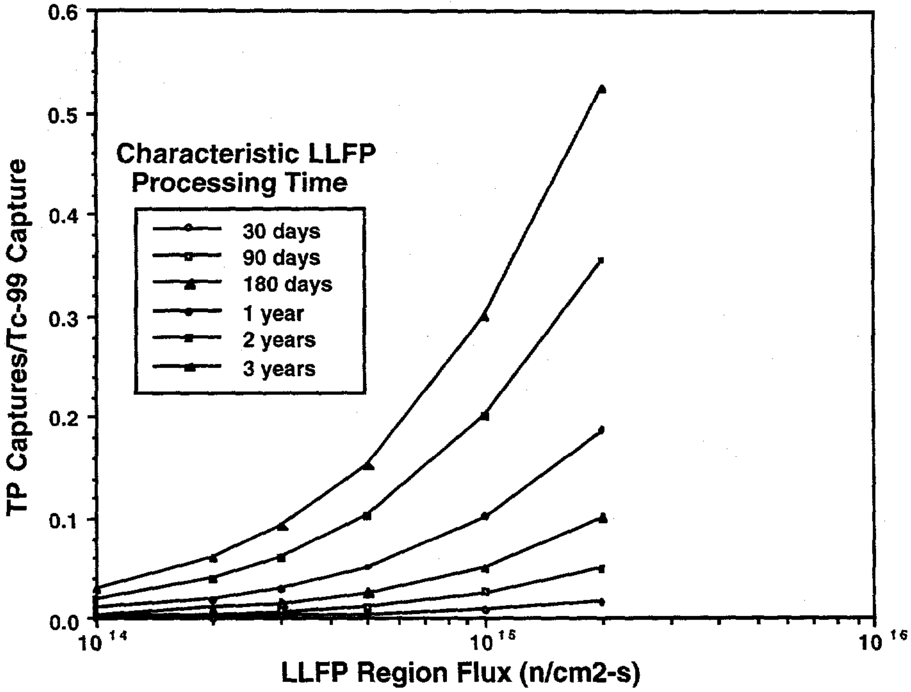

Figure 5 Captures (per ^{99}Tc capture) in ruthenium and other transmutation products as a function of processing time and flux level.

Values of g^i_{TP} for spent-fuel fission-product feed are presented in Table 6.

Table 6 The number of neutron captures in transmutation products per absorption in the LLFP equilibrium chain at a neutron flux level of 4x10^{14}, in a CANDU thermal spectrum, and with a LLFP-TP separation processing characteristic time of 180 d.

	g^i_{LL} Isotopic Feed	g^i_{LL} Elemental Feed
^{79}Se	0.128	0.007
^{93}Zr	0.014	0.013
^{99}Tc	0.003	0.003
^{107}Pd	1.009	0.027
^{126}Sn	0.006	0.002
^{129}I	0.004	0.004
^{135}Cs	0.001	0.001

The quantity w_i defined in Eq. 26 is the integrated LLFP dose reduction per neutron capture per transmutation of LLFP species i only. In order to compare LLFP transmutation strategies, a parameter W_i is defined which accounts for the relative LLFP production rates from fission or concentrations in spent fuel. W_i is the integrated LLFP dose reduction (due to transmutation of LLFP species i) per neutron capture per LLFP and is given by

$$W_i = \frac{w_i}{y^i_{LL}} \qquad (28)$$

where

y^i_{LL} is the species i fraction (isotopic or elemental) of the total LLFPs (isotopic or elemental).

W_i can be expressed in terms of the integrated dose reduction per nuclide $d_i(t_R)$ and the various chain efficiency parameters g^i_{LL}, g^i_{TP}, and y^i_{LL} as

$$W_i = \frac{d_i(t_R)}{g^i_{LL}\left(1+g^i_{TP}\right) y^i_{LL}} \tag{29}$$

Whether the LLFPs are in isotopic or elemental form is determined by the LLFP transmutation strategy adopted which in turn, is based on evaluation of dose reduction effectiveness versus cost. The values of y^i_{LL} are presented in Tables 7 for all isotopic strategy and an elemental strategy, respectively.

Table 7 Values of y^i_{LL} for external feed and internal recycle.

Isotopic Feed Fraction (moles/mole of total feed)			
LLFP Isotope	Internal Only	External Only	External+ Internal
^{79}Se	0.003	0.003	0.003
^{93}Zr	0.360	0.255	0.338
^{99}Tc	0.363	0.346	0.359
^{107}Pd	0.095	0.242	0.126
^{126}Sn	0.010	0.012	0.011
^{129}I	0.065	0.070	0.066
^{135}Cs	0.104	0.072	0.097
Elemental Feed Fraction (moles/mole of total feed)			
LLFP Element	Internal Only	External Only	External+ Internal
selenium	0.009	0.006	0.008
zirconium	0.480	0.333	0.448
technetium	0.096	0.088	0.094
palladium	0.158	0.309	0.191
tin	0.017	0.008	0.015
iodine	0.022	0.025	0.023
cesium	0.218	0.231	0.221

In order to assess the relative dose reduction effectiveness per neutron utilized, the ratio of W_i to that for a reference LLFP species j (e.g., ^{99}Tc) is defined. This ratio E_{iTc} is a measure of the neutron dose-reduction effectiveness relative to that for ^{99}Tc. The LLFP is selected both because of its dominance in repository release but also because of simple one-isotope, large cross-section impact on neutron economy. The ratio is given by

$$E_{ij} = \frac{d_i(t_R)\, g^j{}_{LL}\left(1+g^j{}_{TP}\right) y^j{}_{LL}}{d_j(t_R)\, g^i{}_{LL}\left(1+g^i{}_{TP}\right) y^i{}_{LL}} = \frac{c_i\, \beta_i(t_E)\, \gamma_i(t_W)\, \delta_i(t_R)\, \lambda_j\, g^j{}_{LL}\left(1+g^j{}_{TP}\right) y^j{}_{LL}}{c_j\, \beta_j(t_E)\, \gamma_j(t_W)\, \delta_j(t_R)\, \lambda_i\, g^i{}_{LL}\left(1+g^i{}_{TP}\right) y^i{}_{LL}} \tag{30}$$

The parameter E_{iTc} is a mearue of relative LLFP transmutation effectiveness with respect to accelerator-produced neutron utilization. It can be used to evaluate LLFP transmutation strategies as well as to identify critical cost-performance trades.

Assessment Of The Neutron-Induced Dose-Reduction Effectiveness Of LLFPTransmutation Strategies

The measure of neutron-induced dose reduction effectiveness E_{iTc} defined above requires the additional definition of a LLFP transmutation strategy (i.e., the number and isotopics of LLFPs to be transmuted as well as whether externally and/or internally generated LLFPs are to be transmuted). To illustrate the method, a strategy is defined which is based on an ADEP system which burns all of its internally generated LLFPs. The isotopic composition of the LLFPs recycled is based on crude assumptions about cost impacts of enrichment. The LLFPs ^{99}Tc and ^{129}I are recycled in their elemental for because technetium has a single isotope and iodine has only two but is dominated by ^{129}I. The LLFPs ^{126}Sn and ^{135}Cs are recycled as 100% enriched because these nuclides are on the end of their respective chains and require only a single enrichment following recovery from the thorium/uranium fuel. The LLFPs ^{79}Se, ^{93}Zr, and ^{107}Pd are recycled in elemental form because they are in the middle of their respective chains and would require continuous enrichment as part of the processing to recover the transmutation products. The values of $g^i{}_{LL}$, $g^i{}_{TP}$, and $y^i{}_{LL}$ are presented in Table 8 for this strategy. These values assume a flux level of 4×10^{14} in both the fuel and LLFP transmutation regions, a characteristic fuel processing time of 30 days for recovery of the LLFPs, and a characteristic processing time of 180 days for removal of the TPs for the LLFP loops.

Table 8 Values of $g^i{}_{LL}$, $g^i{}_{TP}$, and $y^i{}_{LL}$ for an example ADEP LLFP transmutation strategy.

LLFP Form	$g^i{}_{LL}$ (moles/mole)	$g^i{}_{TP}$ (moles/mole)	$y^i{}_{LL}$ (moles/mole)
selenium	10.960	0.033	0.008
zirconium	8.660	0.086	0.453
technetium	1.000	0.020	0.133
palladium	9.524	0.125	0.352
^{126}Sn	1.000	0.037	0.004
iodine	1.310	0.025	0.037
^{135}Cs	1.001	0.002	0.012

Appendix H: Impact Of LLFP Transmutation on Neutron Economy

The impact of LLFP transmutation on neutron economy may also be assessed in terms of dose reduction. Using the values of α_{LL} for the LLFP transmutation strategy described above (see Figure 8), a value of the cummulative dose reduction (relative to ^{99}Tc) as a function of the Δk_{eff} can be calculated. A reference value of k_{eff} for burning ^{99}Tc (without internal recycle) and actinides present in spent fuel as well as the representative values of

ν, p_{NL}, α_F, $\alpha_{S/T}$, and α_{FP} (based on equilibrium burning of spent-fuel actinides in a well-moderated target-blanket neutron flux spectrum) are presented in Table 9.

Table 9 Values of a_{LL} for the LLFP transmutation strategy presented above.

LLFP isotope or element	Equilibrium α_{LL} (external feed)
selenium	0.0258
zirconium	2.4450
technetium	0.2400
palladium	1.5020
sn-126	0.0070
iodine	0.0585
cs-135	0.0647

Table 10 Reference neutron economy parameters for the LLFP transmutation strategy presented above

Neutron economy paramter	Equilibrium value
ν	3.0451
p_{NL}	0.9842
$\alpha_{S/T}$	0.1752
α_F	1.6033
α_{FP}	0.1521
α_{Tc}	0.2400
k_{eff}	0.9453

The cummulative relative dose reduction for the LLFPs is calculated based on an order determined by decreasing incremental values (i.e., the first LLFP to be transmuted beyond ^{99}Tc is the one with the largest relative dose reduction). Since operation at large Δk_{eff} ($k_{eff} < 0.9$) translates into large accelerator requirements, and hence cost, there is a stong incentive to identify other management strategies for the LLFPs with minimal dose impacts.