

Isotopic depletion

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Burnup and decay reactions

- The exposure of an isotope ${}^A_Z X$ to neutron flux produces nuclear reactions that may modify its nuclear characteristics.

Radiative capture (n,γ)	${}^A_Z X + {}^1_0 n \longrightarrow {}^{A+1}_Z X$
Fission (n,f)	${}^A_Z X + {}^1_0 n \longrightarrow {}^C_D Y + {}^{A+1-C-\nu}_{Z-D} Z + \nu {}^1_0 n$
(n,xn) reaction	${}^A_Z X + {}^1_0 n \longrightarrow {}^{A+1-x}_Z X + x {}^1_0 n$
(n,α) transmutation	${}^A_Z X + {}^1_0 n \longrightarrow {}^{A-3}_{Z-2} Y + {}^4_2 \text{He}$
(n,p) transmutation	${}^A_Z X + {}^1_0 n \longrightarrow {}^A_{Z-1} Y + {}^1_1 \text{H}$

- Some isotopes may be unstable, even at ground state, and are subject to **radioactive decay**.

Alpha decay (α)	${}^A_Z X \longrightarrow {}^{A-4}_{Z-2} Y + {}^4_2 \text{He}$
Negative beta decay (β ⁻)	${}^A_Z X \longrightarrow {}^A_{Z+1} Y + {}^0_{-1} e$
Positive beta decay (β ⁺)	${}^A_Z X \longrightarrow {}^A_{Z-1} Y + {}^0_1 e$
Isomeric decay	${}^A_Z X^m \longrightarrow {}^A_Z X$
Delayed neutron decay	${}^A_Z X \longrightarrow {}^{A-1}_{Z+1} Y + {}^0_{-1} e + {}^1_0 n$

- The secondary nucleus can also be left into a metastable state, called **isomeric state**, and denoted Y^m .

The time-dependent number density $N(t)$ of the nuclides of type ${}^A_Z X$ is decreasing at a rate that is proportional to $N(t)$ and to the lethargy-integrated microscopic absorption rate $\langle \sigma_a \phi \rangle$:

$$(1) \quad \frac{dN}{dt} = -N(t) \langle \sigma_a \phi(t) \rangle$$

with

$$(2) \quad \langle \sigma_a \phi(t) \rangle = \int_0^\infty du \sigma_a(u) \phi(u, t)$$

where the **absorption cross section** of isotope ${}^A_Z X$ is given in terms of the total and scattering cross sections using $\sigma_a(u) = \sigma(u) - \sigma_e(u) - \sigma_{in}(u)$.

In absence of sources, an initial quantity $N(t_0)$ of isotopes ${}^A_Z X$ is going to decrease at an exponential rate, according to

$$(3) \quad N(t) = N(t_0) e^{-\int_{t_0}^t dt' \langle \sigma_a \phi(t') \rangle} .$$

We see that the wear of a material due to neutron-induced reactions is function of the time-integral of the neutron flux over the irradiation period. We define the **neutron exposure** $\omega(t)$ by the relation

$$(4) \quad \omega(t) = \int_0^t dt' \langle \phi(t') \rangle .$$

Another measure of wear is the **burnup** $B(t)$ defined as the time-integrated power (or energy) per initial unit mass. It is written

$$(5) \quad B(t) = \frac{V}{W} \int_0^t dt' \langle H\phi(t') \rangle .$$

where V is the volume of the fuel, W is the mass of heavy isotopes at time $t = 0$ and $H(E)$ is the **H-factor**, used to compute the recoverable energy from neutron-induced reactions.

- The main component of $H(E)$ is $\kappa\Sigma_f(E)$ where κ is the energy produced by fission (around 200 MeV) and $\Sigma_f(E)$ is the macroscopic fission cross section.
- Other neutron-induced reactions may produce energy and their contributions must be included in $H(E)$.
- The unit of the neutron exposure is the neutron per kilo-barn (n/kb) and the unit of the burnup is the mega-watt-day per metric tonne of initial heavy isotopes (MWd/Mg).

Radioactive decay

A second cause of isotopic depletion is the radioactive decay of an unstable nucleus ${}^A_Z X$. Isomeric states are always unstable and decay toward the underlying ground state.

The positive beta decay is often combined with the **electronic capture decay** which can be written ${}^A_Z X + {}_{-1}^0 e \longrightarrow {}^A_{Z-1} Y$.

The time-dependent number density $N(t)$ of the nuclides of type ${}^A_Z X$ is decreasing at a rate that is proportional to $N(t)$ and to the **radioactive decay constant** λ , so that

$$(6) \quad \frac{dN}{dt} = -\lambda N(t) .$$

The radioactive decay constant is different for each type of isotope (or isomeric state) and each mode of decay. In absence of sources, an initial quantity $N(t_0)$ of isotopes ${}^A_Z X$ is going to decrease at an exponential rate, according to

$$(7) \quad N(t) = N(t_0) e^{-\lambda (t-t_0)} .$$

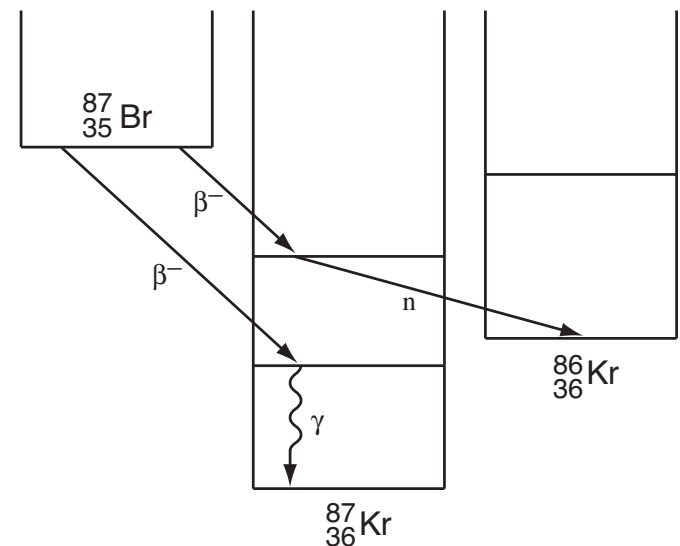
The **half-life** $T_{1/2}$ of an unstable isotope is the period of time required to decay $N(t_0)/2$ nuclides. It is written

$$(8) \quad T_{1/2} = \frac{\ln 2}{\lambda} .$$

Radioactive decay

- The **negative beta decay** is efficient in reducing the neutron/proton ratio (or $(A - Z)/Z$) in fission products.
- In case where $A - Z \gg Z$, **delayed neutron decay** may become possible. This mode of decay is a negative beta decay of specific half-life, producing a daughter nucleus with a level of excitation sufficient to emit a neutron.
 - The half-lives of the **delayed neutron precursors** are smaller than one minute.

- The delayed neutrons allow to reach $K_{\text{eff}} = 1$ with a larger time scale.



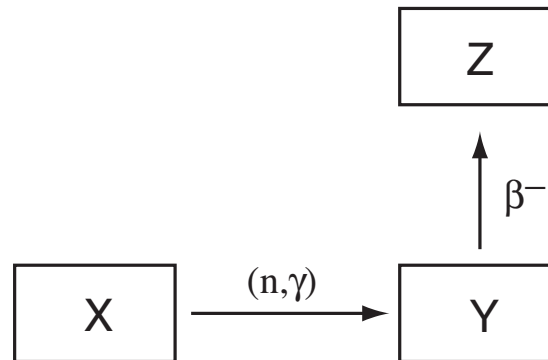
- The **delayed neutron fraction** $\nu^{\text{del}} / (\nu^{\text{pr}} + \nu^{\text{del}})$ is of the order of 0.0065 for a fission of U-235 and of the order of 0.002 for a fission of Pu-239.
- A nucleus may have a non-zero probability to decay accordingly to different channels. The total radioactive decay constant is the sum of all decay constants:

(9)

$$\lambda = \lambda_{\alpha} + \lambda_{\beta^-} + \lambda_{\beta^+} + \dots$$

Depletion equations

Some isotopes produced by neutron-induced reactions are themselves going to decay or to undergo another neutron-induced reaction. Equations (1) and (6) must be combined in accordance to the **depletion chain** describing the father-daughter relations:



Assuming $X(0) = X_0$ and $Y(0) = Z(0) = 0$ as initial conditions, and assuming that the neutron flux is constant and equal to ϕ , we obtain

$$\begin{aligned}
 \frac{dX}{dt} + \langle \sigma_{\gamma} \phi \rangle X(t) &= 0 ; \quad X(0) = X_0 \\
 \frac{dY}{dt} + \lambda_{\beta^-} Y(t) &= \langle \sigma_{\gamma} \phi \rangle X(t) ; \quad Y(0) = 0 \\
 \frac{dZ}{dt} &= \lambda_{\beta^-} Y(t) ; \quad Z(0) = 0 .
 \end{aligned}
 \tag{10}$$

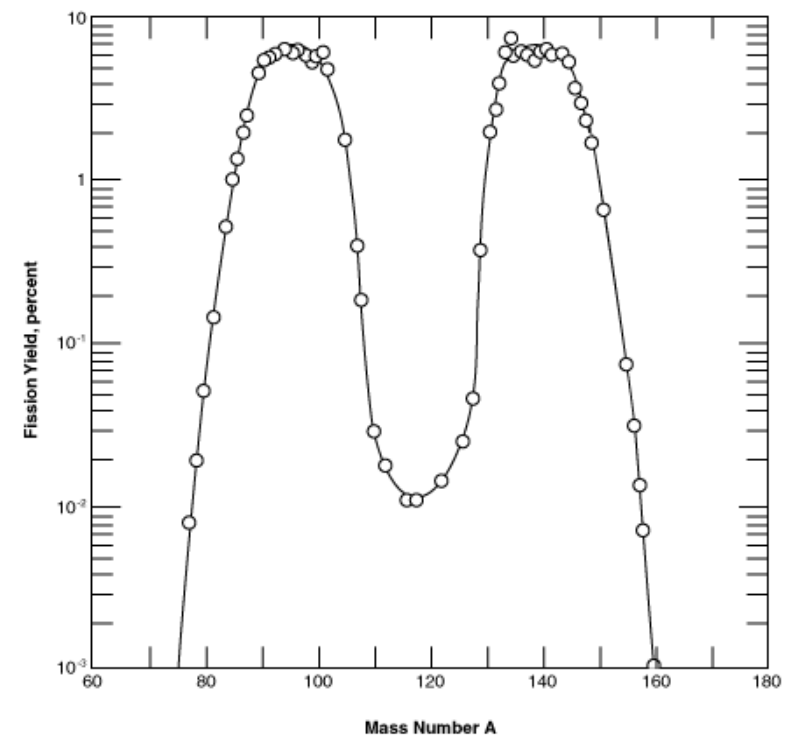
Depletion equations

We obtain a coupled system of K first-order ordinary differential equations with initial conditions, where K is the number of depleting nuclides. These equations are also known as the **Bateman equations**.

- Many techniques are available to solve the resulting system, such as the Laplace transform method, the **Runge-Kutta** family of numerical methods, and the **integration factor** method.
- For in-core depletion calculations, one assumes linear flux variation over each irradiation period, or **time stage**. The initial (and possibly final) flux distributions are recovered from previous neutron flux calculations. In-core depletion can be performed at constant flux or constant power

(expressed in MW/tonne of initial heavy elements) but these values can undergo step variations from one time stage to another.

- Figure represents $Y_{k,l}$, the fission yield for production of fission product k by fissile isotope l .



In each burnup mixture of the unit cell, the depletion of K isotopes over a time stage (t_0, t_f) follows the following equation:

$$(11) \quad \frac{dN_k}{dt} + \Lambda_k(t) N_k(t) = S_k(t) \quad ; \quad k = 1, K$$

$$(12) \quad \Lambda_k(t) = \lambda_k + \langle \sigma_{a,k}(t) \phi(t) \rangle ,$$

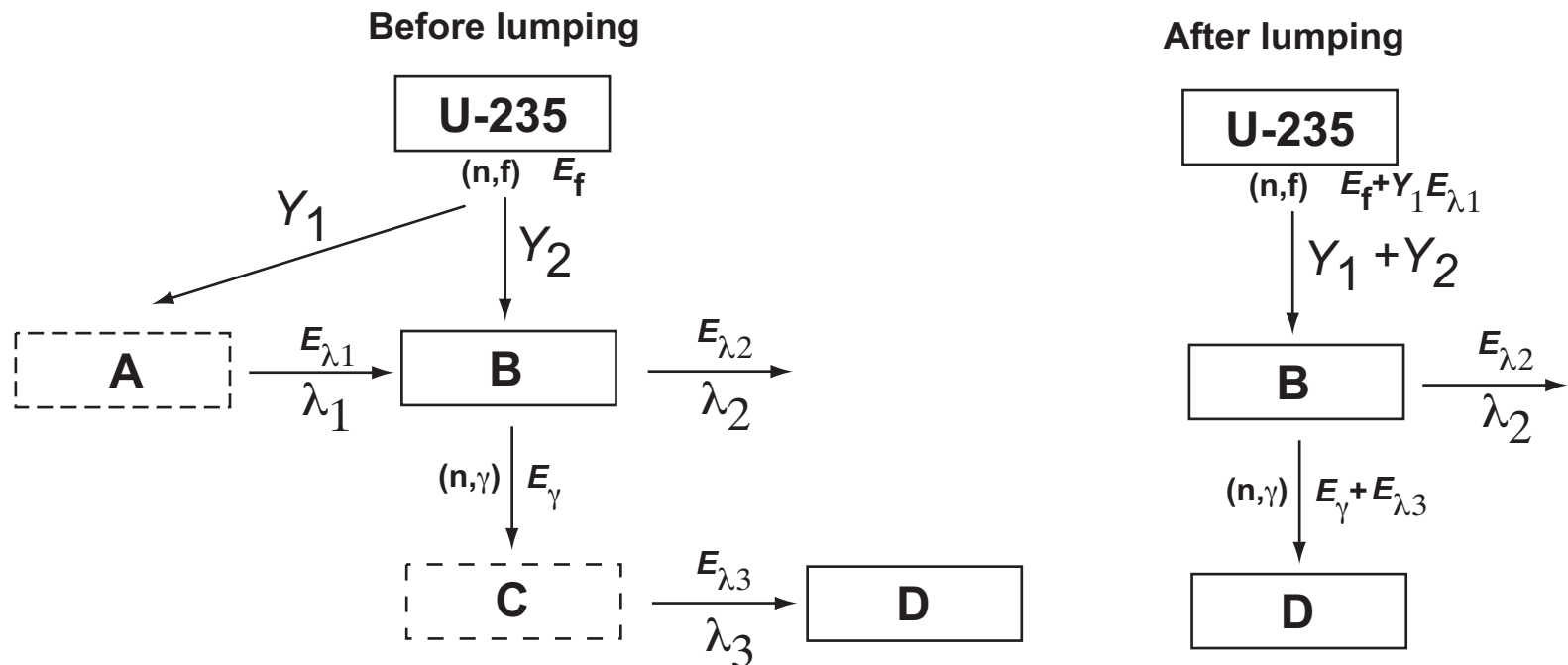
$$(13) \quad S_k(t) = \sum_{l=1}^L Y_{k,l} \langle \sigma_{f,l}(t) \phi(t) \rangle N_l(t) + \sum_{\ell=1}^K m_{k,\ell}(t) N_\ell(t) ,$$

$$(14) \quad \langle \sigma_{x,l}(t) \phi(t) \rangle = \int_0^\infty du \sigma_{x,l}(u) \phi(t, u)$$

$$(15) \quad \begin{aligned} \sigma_{x,k}(t, u) \phi(t, u) &= \sigma_{x,k}(t_0, u) \phi(t_0, u) \\ &+ \frac{\sigma_{x,k}(t_f, u) \phi(t_f, u) - \sigma_{x,k}(t_0, u) \phi(t_0, u)}{t_f - t_0} (t - t_0) \end{aligned}$$

Depletion equations

Some short-lived isotopes (isotopes with high values of Λ) can be lumped in the cross-section processing stage (NJOY stage), before constructing the multigroup library used by the lattice code. It is not possible to recover number-densities for these isotopes in the lattice code.



The power normalization

We note that the simultaneous presence of a radioactive decay constant and of a microscopic reaction rate in Λ_k requires the correct power normalization of the neutron flux before any attempt to solve the depletion equations.

1. Constant flux depletion. In this case, the lethargy integrated fluxes at beginning-of-stage and end-of-stage are set to a constant F :

$$(16) \quad \int_0^\infty \phi(t_0, u) du = \int_0^\infty \phi(t_f, u) du = F$$

2. Constant power depletion. In this case, the power released per initial heavy element at beginning-of-stage and end-of-stage are set to a constant W .

$$(17) \quad \sum_{k=1}^L \left[\kappa_{f,k} \langle \sigma_{f,k}(t_0) \phi(t_0) \rangle + \kappa_{\gamma,k} \langle \sigma_{\gamma,k}(t_0) \phi(t_0) \rangle \right] N_k(t_0) =$$

$$\sum_{k=1}^L \left[\kappa_{f,k} \langle \sigma_{f,k}(t_f) \phi(t_f) \rangle + \kappa_{\gamma,k} \langle \sigma_{\gamma,k}(t_f) \phi(t_f) \rangle \right] N_k(t_f) = C_0 W$$

The end-of-stage power is function of the number densities $N_k(t_f)$; iterations will therefore be required for the end-of-stage power can be set equal to the desired value.

The saturation model

Equations (11) are said to form a **stiff system of equations** when they feature very high and very low values of Λ_k . Numerical instabilities can be avoided in two different ways:

- by adopting a numerical method that has the capability to deal with stiff systems of equations. The **Kaps-Renthrop** algorithm and **integration factor** method are numerical methods with such capability.
- by eliminating the depletion equations with high values of Λ_k .
 - These equations can be lumped at the origin of the cross-section library creation.
 - They can also be lumped in the depletion module, using a **saturation model**, as presented in this section. The latter approach is preferred in cases where the knowledge of the number densities for the lumped isotopes is required.
 - Once the lumping operation has been completed, the remaining depletion equations can be solved using a classical numerical method.

Depleting isotopes with $\Lambda_k(t_0) [t_f - t_0] \geq V_{\max}$ and $\Lambda_k(t_f) [t_f - t_0] \geq V_{\max}$, with V_{\max} set to an arbitrary large value (= 80 is fine), are considered to be at saturation. They are described by making $dN_k/dt = 0$ in Eq. (11) to obtain

$$(18) \quad N_k(t) = \frac{S_k(t)}{\Lambda_k(t)} \quad ; \text{ if } k \text{ is at saturation.}$$

The saturation model

Beginning-of-stage and end-of-stage Dirac contributions can be added:

$$(19) \quad N_k(t) = \frac{1}{\Lambda_k(t)} [a\delta(t - t_0) + S_k(t) + b\delta(t - t_f)] \quad ; \text{ if } k \text{ is at saturation}$$

where a and b are chosen in order to satisfy the time integral of Eq. (11):

$$(20) \quad N_k(t_f^+) - N_k(t_0^-) + \int_{t_0^-}^{t_f^+} N_k(t) \Lambda_k(t) dt = \int_{t_0^-}^{t_f^+} S_k(t) dt$$

A first possibility consists to set the following values of a and b :

$$(21) \quad a = N_k(t_0^-) - \frac{S_k(t_f^+)}{\Lambda_k(t_f^+)} \quad \text{and} \quad b = 0 .$$

This approach is preferred in cases where the matrix $m_{k,\ell}$ is triangular ($m_{k,\ell} = 0$ if $k < \ell$). If this is not the case, it is numerically convenient to chose the following values of a and b :

$$(22) \quad a = N_k(t_0^-) - \frac{S_k(t_0^+)}{\Lambda_k(t_0^+)} \quad \text{and} \quad b = \frac{S_k(t_0^+)}{\Lambda_k(t_0^+)} - \frac{S_k(t_f^+)}{\Lambda_k(t_f^+)}$$

The numerical solution techniques used in the isotopic depletion module of the lattice code are the following:

- Isotopes with very short half-life are taken at saturation and are solved apart from non-saturating isotopes.
- In the lattice code DRAGON, the lumped depletion matrix system containing the non-saturating isotopes is solved using either a fifth order Cash-Karp algorithm or a fourth order Kaps-Rentrop algorithm, taking care to perform all matrix operations in sparse matrix algebra.
- Matrices $\mathbb{M}_{kl}(t_0)$ and $\mathbb{M}_{kl}(t_f)$ are therefore represented in diagonal banded storage and kept apart from the yield matrix \mathbb{Y}_{kl} .
- Every matrix multiplication or linear system solution is obtained via the LU algorithm.

The integration factor method is an analytical solution technique.

- The time domain is divided into steps, over which the neutron flux variation is assumed to be known.
- An exact analytical integration of each depletion equation is used in order to compute the isotopic number densities at the end of a given time step.
- Power series are used in order to represent exponential terms with small arguments. This approach is adapted to cases where the source term in Eq. (13) can be expressed only in terms of already known isotopic densities, so that

$$(23) \quad S_k(t) = \sum_{l=1}^L Y_{k,l} \langle \sigma_{f,l}(t) \phi(t) \rangle N_l(t) + \sum_{\ell=1}^{k-1} m_{k,\ell}(t) N_\ell(t) \ .$$

This condition is not satisfied with the heavy elements and can only be used to study fission products. Assuming constant flux and constant cross sections, Eq. (11) can now be written

$$(24) \quad \frac{dN_k}{dt} + \Lambda_k N_k(t) = \sum_{l=1}^L Y_{k,l} \langle \sigma_{f,l} \phi \rangle N_l(t) + \sum_{\ell=1}^{k-1} m_{k,\ell} N_\ell(t) \ ; \ k = 1, K \ .$$

The RHS of Eq. (24) is rewritten as a sum of exponential terms of the form

$$(25) \quad \sum_{l=1}^L Y_{k,l} \langle \sigma_{f,l} \phi \rangle N_l(t) + \sum_{\ell=1}^{k-1} m_{k,\ell} N_{\ell}(t) = \sum_{j=1}^J a_{k,j} t^{n_j} e^{-\nu_j t}$$

where n_j is an integer ≥ 0 . The first 35 terms of the RHS summation are a pure power series with

$$(26) \quad \nu_j = 0 \quad \text{and} \quad n_j = j - 1 \quad ; \quad j \leq 35 \quad .$$

These 35 terms are useful to eliminate any exponential function with $\nu_j \leq 7$ from the solution. Such exponential functions may arise in presence of nuclides with very long half-lives. For the sake of efficiency of computation, each time an exponential is found to be greater than e^{-7} , it is replaced by a 35-term power series of the form

$$(27) \quad e^{-\nu t} \simeq \sum_{\ell=1}^{35} \frac{(-\nu t)^{\ell-1}}{(\ell-1)!} \quad \text{if} \quad \nu t \leq 7 \quad .$$

Equation (24) is multiplied by an integration factor and is integrated between 0 and t . The LHS leads to

$$(28) \quad \int_0^t dt' e^{\Lambda_k t'} \left[\frac{dN_k}{dt'} + \Lambda_k N_k(t') \right] = N_k(t) e^{\Lambda_k t} - N_k(0^+)$$

so that

$$(29) \quad \begin{aligned} N_k(t) &= e^{-\Lambda_k t} \left[N_k(0^+) + \sum_{j=1}^J \int_0^t dt' a_{i,j} t'^{n_j} e^{(\Lambda_k - \nu_j)t'} \right] \\ &= N_k(0^+) e^{-\Lambda_k t} + \sum_{j=1}^J \mathcal{I}_{k,j}(t) \end{aligned}$$

The value of $\mathcal{I}_{k,j}(t)$ is given by one of the following three formulas:

$$(30) \quad \mathcal{I}_{k,j}(t) = \frac{a_{k,j}}{\Lambda_k - \mathcal{V}_j} \left[e^{-\mathcal{V}_j t} - e^{-\Lambda_k t} \right]$$

if $n_j = 0$ and $\Lambda_k \neq \mathcal{V}_j$.

$$(31) \quad \mathcal{I}_{k,j}(t) = \frac{a_{k,j}}{\Lambda_k - \mathcal{V}_j} \left[t^{n_j} e^{-\mathcal{V}_j t} - n_j e^{-\Lambda_k t} \int_0^t dt' t'^{n_j-1} e^{(\Lambda_k - \mathcal{V}_j)t'} \right],$$

if $n_j \neq 0$ and $\Lambda_k \neq \mathcal{V}_j$, or

$$(32) \quad \mathcal{I}_{k,j}(t) = \frac{a_{k,j}}{n_j + 1} t^{n_j+1} e^{-\mathcal{V}_j t} \quad \text{if } \Lambda_k = \mathcal{V}_j.$$

Collecting together terms with the same dependence on time, Eq. (29) can now be written as

$$(33) \quad N_k(t) = \sum_{j=1}^{J+1} c_{i,j} t^{n_j} e^{-\mathcal{V}_j t}$$

so that

$$(34) \quad a_{k+1,j} = \sum_{\ell=1}^k m_{k+1,\ell} c_{\ell,j}$$

will be used by nuclide $k + 1$ as a source contribution in Eq. (25).

An analytic solution over a time interval Δt , will be possible if

1. The neutron flux $\phi(t)$ is constant within Δt . However, the method can be adapted to the case where the neutron flux varies linearly within Δt .
2. The variation of the fission rates within Δt is assumed to be given by an expression as

$$(35) \quad \langle \sigma_{f,l} \phi \rangle = f_{l,0} \delta(t) + \sum_{j=1}^{J^f} f_{l,j} t^{n_j} e^{-\nu_j t}$$

where $\delta(t)$ is a Dirac delta distribution. This component is used as initial condition in the solution of Eq. (24), so that

$$(36) \quad N_k(0^+) = N_k(0^-) + \sum_{l=1}^L Y_{k,l} f_{l,0} \quad ; \quad k = 1, K \quad .$$

3. The matrix $m_{k,\ell}$ is triangular ($m_{k,\ell} = 0$ if $k < \ell$).
4. The resonance self-shielding of the heavy isotopes is constant within Δt .

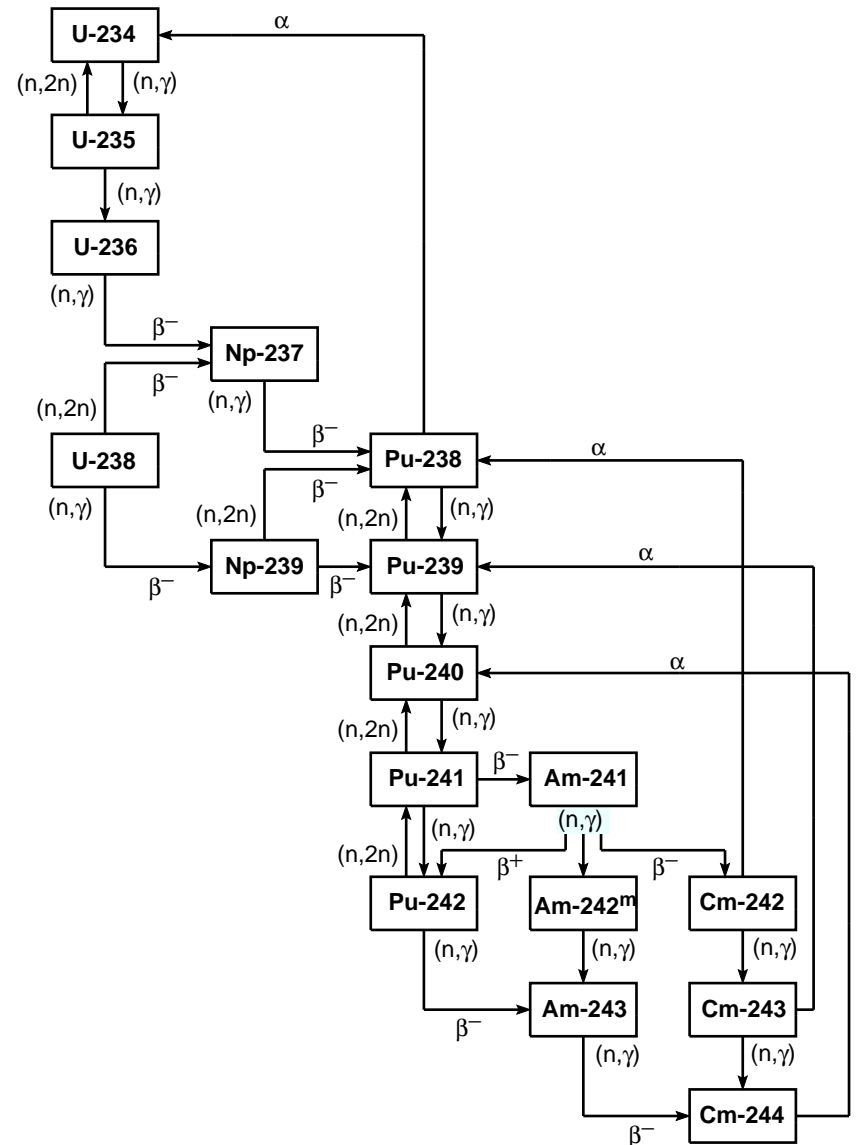
Otherwise, a numerical solution approach, similar to the Runge-Kutta method, will be used.

Depletion of heavy isotopes

The burnup chain of the heavy isotopes in the uranium cycle is represented in figure.

When the radioactive decay constant of an isotope is big in comparison of the microscopic absorption rate $\langle \sigma_a \phi \rangle$, we may assume that the decay is instantaneous and remove this isotope from the burnup chain. This is the case of the radiative capture of a neutron in U-238 which can be considered to produce directly a Np-239 nucleus.

When a U-235 nuclide absorbs a neutron, the most likely reaction is fission. However, a fraction of the absorptions will result in the production of a U-236 nuclide. The absorption of a neutron in U-236 will produce a U-237 nuclide that will decay immediately in Np-237, a more stable isotope. Another decay of Np-237 will produce a Pu-238 nuclide.



Depletion of heavy isotopes

- The uranium cycle corresponds to the majority of applications of nuclear energy.
- It is used in many reactor systems such as
 - the **pressurized water reactor** (PWR),
 - the **boiling water reactor** (BWR),
 - the **Canada deuterium uranium reactor** (CANDU) and
 - the **liquid-metal fast-breeder reactor** (LMFBR).
- The **conversion ratio** is the ratio of the number of fissile nuclides produced per unit time over the number of fissile nuclides burned per unit time. A nuclear reactor with a conversion ratio greater than one is referred as a **breeder**.

Reactor system	Enrichment	Conversion ratio
CANDU	U-235 at 0.711%	$\simeq 0.8$
PWR	U-235 at 3.3 %	$\simeq 0.5$
BWR	U-235 at 2.6 %	$\simeq 0.5$
LMFBR (Super-Phoenix)	Pu-239 at 16.0 %	> 1