

Fuel Cycle Physics for Nuclear Fission Reactors

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Section 1

Fuel Cycle Physics - Introduction

► Fuel Cycle Physics

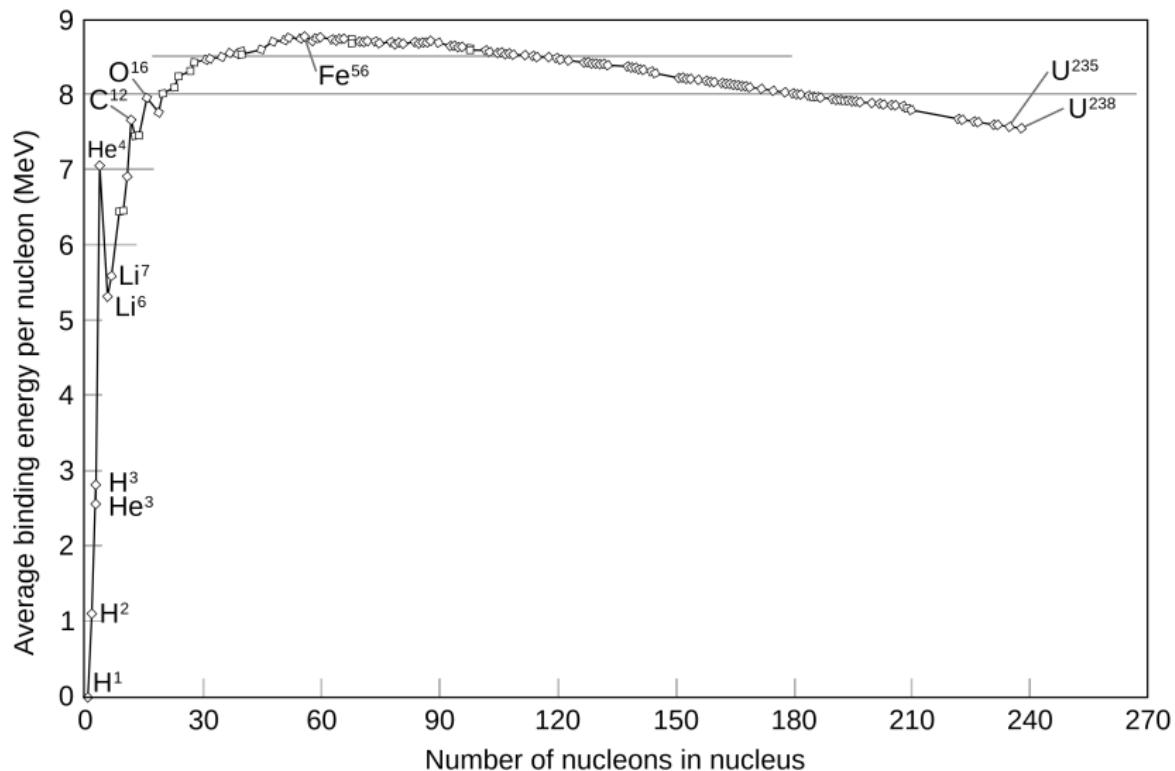
During the operations of a fission based power plant nuclear fuel is subject to change in concentration of isotopes due to

- intrinsic nature of nuclear energy production (heavy nuclei subject to fission)
- conditions under which the components of the core are (neutron bombardment of the nuclei)

Generally a nucleus ${}^A_Z X$ will be subject to

- disintegration
- capture/neutron induced phenomena

► Binding Energy



► Nuclear Fission Materials

Fission fuel isotopes may be divided according to how they can undergo fission

- Fissile materials, can undergo fission with neutrons of any energy (applications typically with thermal neutrons); ^{235}U , ^{239}Pu
- Fissionable materials, require high energy (MeV) neutrons in order to undergo fission; ^{238}U , ^{232}Th
- Fertile materials, absorbing neutrons they become fissile isotopes; ^{234}U , ^{238}U , ^{232}Th

Section 2

Reactions

► Disintegration Reactions

Principal disintegration reactions:

- (Spontaneous) Fission ($\rightarrow Y + Z + n + \gamma + \dots$)
- α decay ($\rightarrow_{Z-2}^{A-4} Y$)
- β^\pm decay ($\rightarrow_{Z\mp 1}^A Y + (\dots)$)
- ν emission ($\rightarrow_{Z-1}^{A-1} Y$)
- delayed neutron emission
- γ decay/isomeric transition
- + others

► Neutron Induced Reactions

Reactions induced by neutron capture:

- (n,γ) (radiative capture)
- (n,xn)
- $(n,fission)$
- $(n,others)$

► Bateman Equations (Introduction)

Bateman equations are differential equations that describe the variation of populations of atomic nuclei in decay chains.

For nuclear reactor fuel we find 3 Bateman terms describing:

- Heavy Nuclei
- Fission Products
- Activation Products

► Heavy Nuclei

Usually the term heavy nuclei requires a bit of clarification since it may refer to:

- Elements heavier than lead
- Elements that cannot be produced by fission
- Rarely: elements like Gadolinium ($Z=64$)

Bateman equations for heavy nuclei include

- activation terms of the form $\Phi(\sigma_{(n,x)}N(t))$, where Φ represents the neutron flux during depletion
- decay terms in the form $\lambda_x N(t)$

Both of these may proceed in both directions

► Heavy Nuclei (2)

$$\frac{dN}{dt}_{(A,Z)} = \sum_i \Phi(\sigma_{(n,x)} N_i(t)) + \sum_i \lambda_x N_i(t) + \\ -\Phi((\sigma_{(n,x)} + \sigma_{capture} + \sigma_{fission} + \dots) N_{(A,Z)}(t)) - \lambda_{tot} N_{(A,Z)}(t)$$

λ_{tot} includes all decay contributions

$$\lambda_{tot} = \lambda_\alpha + \lambda_{\beta^\pm} + \dots + \lambda_{\text{spontaneous fission}}$$

and lets us define branching fractions Γ as

$$\Gamma_i = \frac{\lambda_i}{\lambda_{tot}}$$

► Fission Products

Fission products also include a fission production term

$$\frac{dN}{dt}_{(A,Z)} = \sum_i \Phi(\sigma_{(n,x)} N_i(t)) + \sum_i \lambda_x N_i(t) + \\ -\Phi((\sigma_{(n,\gamma)} + \dots) N_{(A,Z)}(t)) - \lambda_{tot} N_{(A,Z)}(t) + Y_{G_{(A,Z)}}$$

where $Y_{G_{(A,Z)}}$ is the total fission yield given by

$$Y_{G_{(A,Z)}} = \sum_{F \text{ fissile nuclei}} \gamma_{(A,Z)}^F \tau_F$$

- τ_F fission rate of the F isotope
- $\gamma_{(A,Z)}^F$ fission yield of the F isotope into ${}^A_Z X$

► Activation Products

Some radioactive nuclei may be produced inside of nuclear reactors thanks to capture phenomena. Similarly to heavy nuclei we get:

$$\frac{dN}{dt}_{(A,Z)} = \sum_i \Phi(\sigma_{(n,x)} N_i(t)) + \sum_i \lambda_x N_i(t) + \\ -\Phi((\sigma_{(n,\gamma)} + \dots) N_{(A,Z)}(t)) - \lambda_{tot} N_{(A,Z)}(t)$$

It is worth noting that typical activation processes are threshold reactions.

For example ^{60}Co , a radioactive isotope which finds several industrial/medical applications, is produced via activation reactions in nuclear reactors.

Section 3

Relevant Quantities for the Fuel Cycle

► Relevant Quantities for the Fuel Cycle

The most relevant quantities for the fuel cycle are

- Mass Balance
- Burn-up
- Activity
- Decay Heat
- γ/n^0 Dose

► Mass Balance

Mass balance is a nuclide count for the specie of interest.

$$M[g/cm^3] = \frac{N[\#/cm^3]}{N_A[\#/mol]} A[g/mol]$$

It is useful to quantify fuel composition at different stages of the burn-up.

► Burn-up

Burn-up is a parameter that quantifies how much energy (typically in MegaWatt·day or GigaWatt·day) has been produced by the heavy metals in the fuel with respect to their original mass (in tonnes). Only the mass of metals is considered, for example using UO_2 fuel only the mass of uranium is considered.

$$\text{burnup[GW} \cdot \text{d/t]} = \frac{E[\text{GW} \cdot \text{d}]}{M[\text{t}]}$$

This definition is chosen because

- masses of heavy metals in the fuel vary during reactor operations
- it is easy to compare energy outputs of different sources/different fuel compositions (especially on industrial scales)
- it is easy to retrieve power from the number of operating days

► Thermal Burn-up

For thermal neutron (10^{-2} eV) reactors the burn-up is typically expressed as thermal burn-up

$$\text{Thermal burn-up[GW d/t]} = \frac{\sum_F \kappa_F N_F \sigma_F \Phi[\text{GW}]}{M[t]} \Delta t[\text{d}]$$

where the sum is on all fissile nuclei F.

The terms of the expression are all relative to the reactor operations:

- $\kappa_F \approx 200$ MeV/fission, energy released for each fission of the nuclear fuel (+energy conversion terms MeV → J)
- N_F concentration of the F nuclide
- $\sigma_F \Phi$ fission rate [fissions/time]
- Δt operating time of the reactor

Thermal burn-up does not include the energy released by other neutron induced reactions (*capturing*), which is accounted for in the effective power produced.

► Fission Burn-up

Fission burn up is more related to the true physical phenomena happening inside the reactor.

There are two principal definitions, mostly used for fast fission reactors, both dimensionless and typically expressed in %:

- Fission per Invested Fissile Atom (FIFA):

Number of fissions over the total number of fissile atoms ($N_f(t = 0)$)

$$\text{FIFA} = \frac{\int \Sigma_f \Phi dt}{N_f(0)}$$

- Fission per Invested Metal Atom (FIMA):

Number of fissions over the total number of heavy atoms ($N(t = 0)$)

$$\text{FIMA} = \frac{\int \Sigma_f \Phi dt}{N(0)}$$

Typically FIMA is preferred since it provides more accurate estimates (fertile nuclides can later undergo fission).

Pure ^{235}U has a 100% FIMA of 1000 GWd/t (1% FIMA is 10 GWd/t).

► Burn-up and Depletion

During operations the dominant term of the Bateman equation becomes the capture rate compared to spontaneous decay ($\Sigma\Phi \gg \lambda N$)

$$\frac{dN}{dt} = -\lambda N + \Sigma\Phi \xrightarrow{\Sigma\Phi \gg \lambda N} \Delta N = \Sigma\Phi\Delta t$$

where Φt is a neutron fluence [$\#\text{neutrons}/\text{kilobarn}$] and $\Phi\Delta t$ a fluence increment.

Fuel subject to the same fluence will be at the same burn-up; this may be achieved by keeping a constant product between power and operating time. For this reason in first approximation (decay term of Bateman equation is negligible) heavy nuclide concentration is dependent on burn-up and not on time.

► Activity

Activity is the number of disintegrations per second

$$A[\text{Bq}] = \lambda N[\#\text{atoms}/\text{s}]$$

A becquerel (Bq, 1 disintegration per second) is usually too small for any application, therefore the curie (Ci) is more commonly used (1 Ci=3.7*10¹⁰ Bq, the activity of one gram of ²²⁶Ra).

► Decay Heat

Decay heat is the heat released due to radioactive decay and its calculation is a crucial parameter to keep under control for reactor safety since fuel will continue emitting heat even after the reactor has stopped its operations. This heat production leads to the requirement of cooling systems for the fuel, even the spent one (cooling pools).

The power produced by the fuel after shutdown (t , in seconds) is described by the Way-Wigner empirical formula

$$P(t) = P_0 \cdot 0.0622(t^{-0.2} - t_{start}^{-0.2})$$

where P_0 is the power of the reactor before the shutdown and t_{start} [s] is the time passed since the starting of the reactor operations.

► Decay Heat: Summation Method

The total decay heat (DH) is given by the contribution of each isotope involved via

$$DH_{tot} = \sum_{i \text{ isotopes}} DH_i$$

where the singular contributions to decay heat is the product of activity and the energy freed in the decay

$$DH_i[\text{W}] = A[\#\text{decays/s}] \cdot \bar{E}_{tot}[\text{J/decay}] = \lambda N \sum_{j \text{ decay type}} \bar{E}_j$$

Notice that the energy of α and β decays also include the following emission of γ (\bar{E}_α includes both the α emission and connected chains of γ transitions).

DH is also decomposed as

$$DH_{tot} = DH_{\text{delayed } n^0 (t < 10 \text{ min})} + DH_{\text{fission products}(\beta)} + DH_{\text{heavy nuclides}(\alpha, \gamma)}$$

► Decay Heat: Summation Method (2)

Summation method is exact but requires precise knowledge of the isotope populations in the reactor, which may be difficult or even impossible at times to either get or calculate.

Adopted solution is to reduce the considered number of isotopes to just the most dominant ones up to the required precision.

Notice that the most relevant isotopes depend also on the timescale: for long timescales fewer nuclides may be considered, on the contrary for short timescale (immediately after reactor shutdown) many different isotopes must be considered.

Laugier curves (next slides) are a good way to represent how many isotopes must be considered to achieve the required precision on the desired timescales ($1 \text{ day} \approx 10^5 \text{ s}$).

► Decay Heat: Laugier Curves

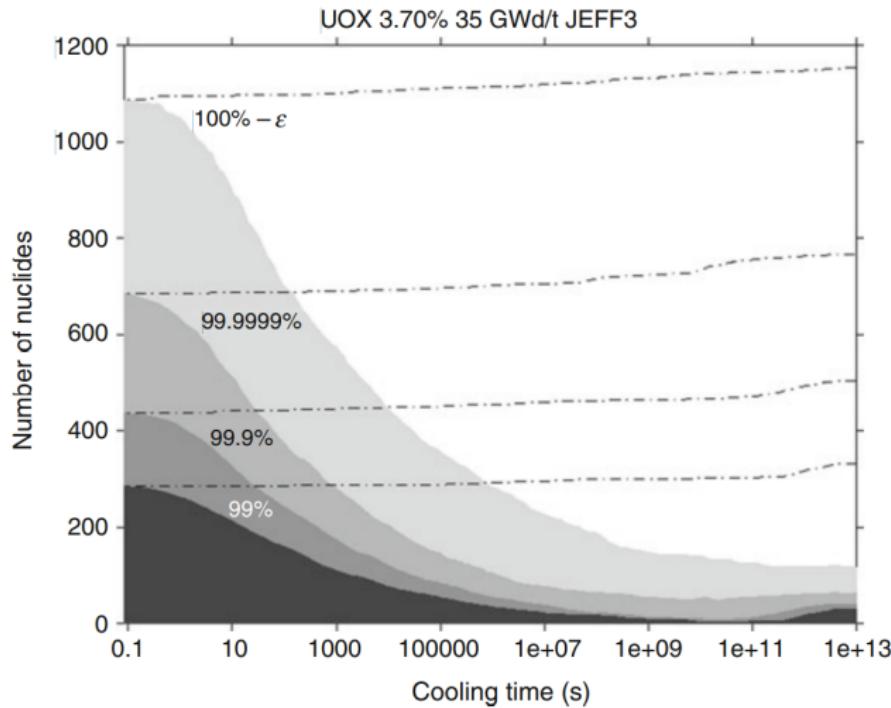


Fig. 15.4 Laugier curves for short cooling times

► Decay Heat: Laugier Curves (2)

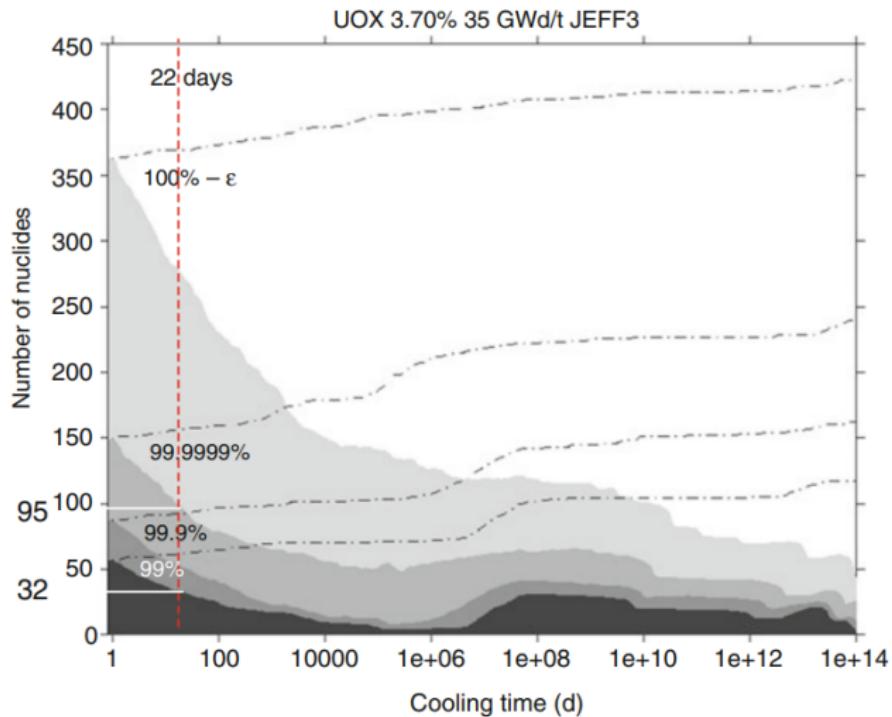


Fig. 15.5 Laugier curves for geological times

► Decay Heat Burst Function (DHBF)

Decay heat calculation via previously given definition have the strong requirement of having knowledge of isotope concentrations in order to carry out the computations. We can find a faster way to compute DH via the use of the Decay Heat Power Burst Function (DHBF, one fission per heavy nuclide) leading to a DH term computed for each fissile isotope (F) in the fuel as

$$DH_F(t) = \int_0^T N_F(\tau) \sigma_f^F(\tau) \Phi(\tau) DHBF_F(T + t - \tau) d\tau$$
$$DH(t) = \sum_F DH_F(t)$$

DHBF can be experimentally measured via irradiation of samples.

DHBF approach simplifies computations at the expenses of requiring correction terms to take into account DH of fission products during irradiation (DHBF obtained in absence of neutron flux).

► Decay Heat: Elementary Value Curves (EVC)

Elementary Value Curve approach consists in calculating fuel composition before irradiation and using a simplified depletion chain and calculating decay heat from the response of each isotope in the simplified chain

$$DH(t) = \sum_{i \text{ isotope}} EVC_i(t)$$

where EVC_i is given by the decay heat (dh) of i isotope and all the j isotopes in its decay chain according to

$$EVC_i(t) = dh_i(t) + \sum_{i \rightarrow \dots \rightarrow j} dh_j(t)$$

This definition leads to having non-zero EVC for isotopes that may be exhausted in the fuel since their daughter nuclei may still be contributing to the decay heat.

This method is very fast but requires calculation of EVC beforehand.

► Dose Calculation

The dose is the energy deposited over a considered mass by the action of ionising particles

$$H[\text{Gy}] = \frac{E[\text{J}]}{M[\text{Kg}]}$$

Different units are used in the field

- Gray, 1 Gy = 1 J/Kg (Standard SI unit)
- erg/g, 1 erg/g = 10^{-4} J/Kg (CGS unit)
- rad, 1 rad = 100 erg/g (later adopted CGS unit)

► γ/n^0 Dose Calculation

Principal mechanisms of energy deposition include

- γ pair production of e^\pm pairs in presence of electromagnetic fields of atoms or electrons (+compton and photoelectric effect)
- n^0 elastic and inelastic scattering with atoms of matter

Neutral particles may travel far from where they were originally produced and therefore it is important to know well how to shield and monitor their radiation.

► γ/n^0 Dose Calculation: Kerma

One parameter used in dosimetry when dealing with neutral particles is the Kerma, Kinetic Energy Released in Matter, corresponding to the kinetic energy of the charged particles released in the interaction.

$$\text{Kerma[Gy]} = \frac{dE_{kin}}{dm} = \frac{1}{\rho} \frac{dE_{kin}}{dV}$$

At low energies (x-rays) Kerma is equivalent to the dose but for higher energies (γ -rays leading to deep photoelectric transitions or compton scattering) the scattered particles may leave the scattering region, leading to a Kerma (in this case \approx energy of scattered e^-) higher than the absorbed dose.

► γ/n^0 Dose Calculation: Equivalent Dose

Sometimes it is more useful to quantify how much biological damage some radiation caused. For this reason in dosimetry the equivalent dose (H_T) is more commonly used rather than the dose (H) itself.

The equivalent dose is given by the dose weighted by a quality factor (Q) according to

$$H_T[\text{Sv}] = H[\text{Gy}] \cdot Q[\text{Sv/Gy}]$$

Photons are taken as reference ($Q_\gamma = 1$), therefore a dose of 1 Gy of photons delivers an equivalent dose of 1 Sv (sometimes also rems are used, where $1 \text{ Sv} = 100 \text{ rem}$).

Quality factors vary from 1 for γ and e^\pm radiation or 2 for protons and can go up to 20 for heavy ions (hadrontherapy applications).

Dividing over time of exposure it is also possible to obtain (equivalent) dose rates, used especially to put limits on exposures.

Section 4

Depletion Chains - Bateman Equations

► Isotopic Depletion Calculation

For practical purposes it may be worth to solve analytically some simplified decay chain models.

For a chain of n isotopes N_i with decay constants λ_i ,

$$N_1 \rightarrow N_2 \rightarrow \dots \rightarrow N_j \rightarrow N_{j+1} \rightarrow \dots \rightarrow N_n \text{ stable}$$

Bateman equations become

$$\begin{cases} \frac{dN_1(t)}{dt} = -\lambda_1 N_1(t) \\ \frac{dN_i(t)}{dt} = +\lambda_{i-1} N_{i-1}(t) - \lambda_i N_i(t) & 1 < i < n \\ \frac{dN_n(t)}{dt} = +\lambda_{n-1} N_{n-1}(t) \end{cases}$$

where at $t = 0$ only $N_1(t) \neq 0$

► Isotopic Depletion Calculation (2)

Via a Laplace transform we end up with the generic solution for the isotope $N_j(t)$

$$N_j(t) = N_1(t=0) \cdot \prod_{k=1}^{j-1} \lambda_k \cdot \sum_{i=1}^j \left(\frac{e^{-\lambda_i t}}{\prod_{k=1, k \neq i}^j (\lambda_k - \lambda_i)} \right)$$

This solution poses an extra requirement on isotopes such that two elements of the chain do not have the same λ decay constant, which rarely happens (still problematic for implementations since similar λ may still lead to computational errors in computer codes).

There is also a second constraint on having a loopless chain, since it would go back to the previous case of $\lambda_i = \lambda_j$ for $i \neq j$.

► Isotopic Depletion Calculation (3)

The formula can be extended to the more realistic case of all $N_j(t = 0) \neq 0$ where for each element N_m can be added a contribution term of the form

$$N_{j,m}(t) = N_m(t = 0) \cdot \prod_{k=m}^{j-1} \lambda_k \cdot \sum_{i=m}^j \left(\frac{e^{-\lambda_i t}}{\prod_{k=m, k \neq i}^j (\lambda_k - \lambda_i)} \right)$$

finally leading to the general solution

$$N_j = \sum_{m=1}^{m=j-1} N_{j,m} = \sum_{m=1}^{m=j-1} N_m^0 \cdot \prod_{k=m}^{j-1} \lambda_k \cdot \sum_{i=m}^j \left(\frac{e^{-\lambda_i t}}{\prod_{k=m, k \neq i}^j (\lambda_k - \lambda_i)} \right)$$

► Heavy Nuclides Case

For heavy nuclides the solution resorts to vector form of the Bateman equations

$$\frac{d\vec{N}(t)}{dt} = A\vec{N}(t)$$

which may be solved by numerical methods (Runge-Kutta) or by matrix exponential

$$\vec{N}(t) = e^{At} \vec{N}(t=0)$$

where the problem reduces to the evaluation of the exponential matrix

$$e^{At} = \sum_{k=0}^{\infty} \frac{(At)^k}{k!} \approx \mathbb{1} + At + \frac{(At)^2}{2!} + \dots + \frac{(At)^m}{m!}$$

In the case of fission extra terms appear taking them into account.

Section 5

Decay Chains

► Decay Chains: Reduction Principle

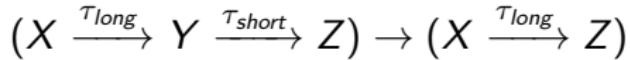
Full analysis of nuclear fuel would be unpractical and computationally unfeasible, for this reason decay/reaction chains are reduced to take only most significant phenomena into account, leading to simplified chains.

Significance of an isotope in a chain is a function of

- abundance
- activity
- considered timescale

In the fuel analysis it is crucial to take these factors into account.

One typical approach is to remove fast decays following slow decays, an example being



In any case the reduction process must be carried out with attention to the specific case under study, with particular care in the presence of branching.

► ⁷⁹Se Reduced Chain

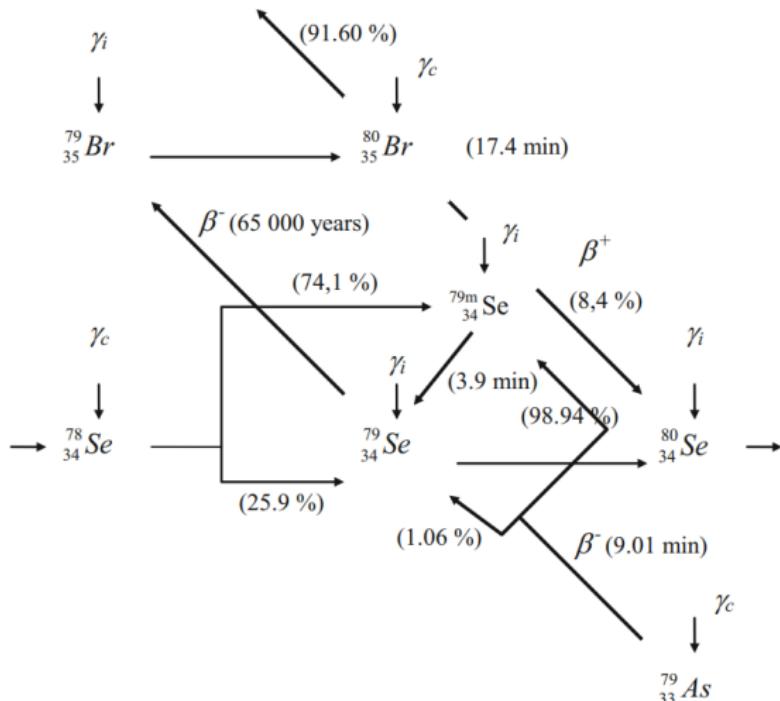


Fig. 15.26 Reference chain of $^{79}_{34}Se$

► ⁷⁹Se Reduced Chain (2)

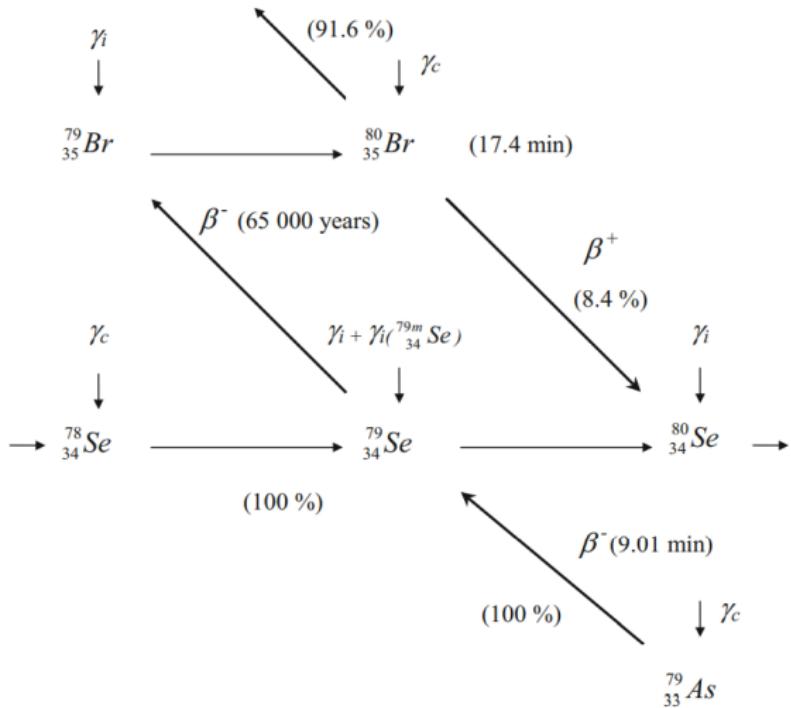


Fig. 15.27 Reduced chain of $^{79}_{34}\text{Se}$ (1st step)

► ^{79}Se Reduced Chain (3)

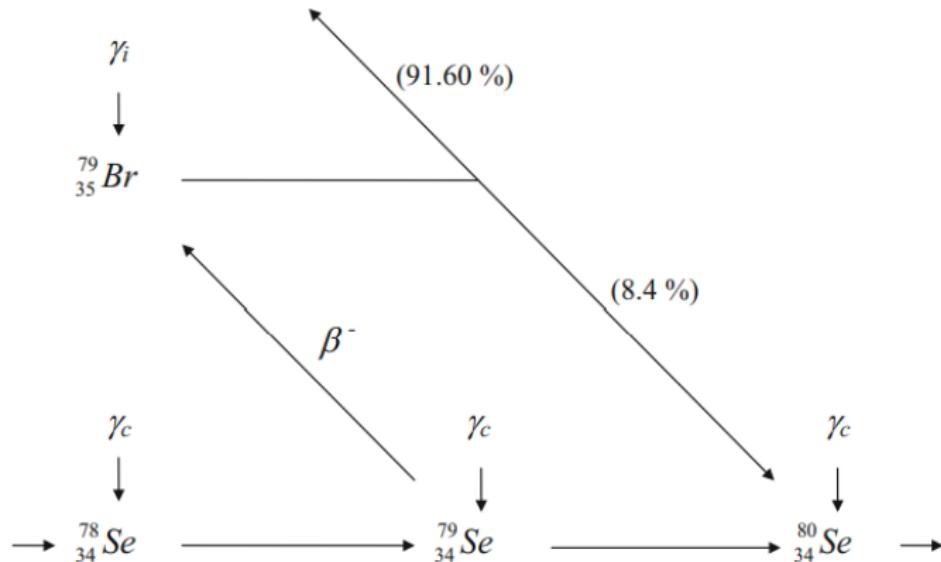


Fig. 15.28 Reduced chain of $^{79}_{34}\text{Se}$ (2nd step)

Section 6

Poison Elements

► Poison Elements

Poison elements are elements that inhibit the development of the chain reactions.

There are 3 types of poisons

- Xenon-like poisons (short term poisoning)
- Samarium-like poisons (long term poisoning)
- Burnable poisons

► Xenon Poisoning

$^{135}_{54}\text{Xe}$ is the highest thermal neutron absorbing isotope currently known ($\sigma \approx 2.65 \cdot 10^6 \text{ b}$). It is produced in fission reactors mainly as a fission product and more importantly via β^- decay of $^{135}_{53}\text{I}$ (which in turn is the decay product of ^{135}Te which is very short lived and treated via chain reduction techniques).

The half life of $^{135}_{54}\text{Xe}$ is $\approx 9 \text{ h}$, before it decays via $\beta^- (\rightarrow ^{135}_{55}\text{Cs})$ decay. After stopping reactor operations the population of ^{135}Xe increases over time until it reaches a peak called the Xenon Peak/Pit (or Iodine Hole). This increase in ^{135}Xe population may inhibit the restarting of reactor operations due to its huge neutron capture cross section. After the peak the concentration of ^{135}Xe decreases and the reactor may power up again.

^{135}Xe yields may vary considerably (0.01% to 2%) depending on several factors including fuel and reactor design while the ^{135}I is rather constant (typically 5%).

Xenon Chain

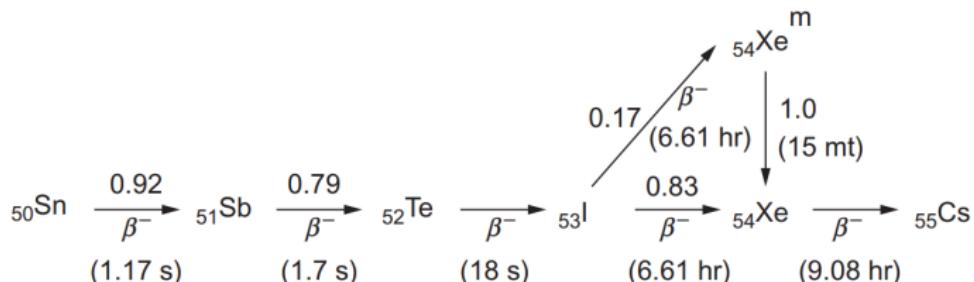


FIG. 6.2 Chain of ${}^{135}\text{Xe}$ formation and decay.

Short timescales of previous elements of the chain clearly allow for reduction chain techniques. (${}^{135}\text{Xe}^m$ decays in 15 minutes)

► Xenon Poisoning (2)

The treatment of Xenon effects can be simplified via chain reduction considering only ^{135}I and ^{135}Xe . Using $\Sigma\phi$ as fission rate, $\gamma_{I/X}$ as fission product yields and I/X as isotope concentrations we get

$$\frac{d}{dt}I(t) = +\gamma_I\Sigma\phi - \lambda_I I(t)$$

$$\frac{d}{dt}X(t) = +\gamma_X\Sigma\phi - \lambda_X X(t) + \lambda_I I(t) - \sigma_X X(t)\phi$$

noticing that the σ_I neutron absorption term is omitted being negligible compared to the decay rate of I.

We are led to the infinite time equilibrium condition

$$I_\infty = \frac{\gamma_I\Sigma\phi}{\lambda_I}$$

$$X_\infty = \frac{(\gamma_I + \gamma_X)\Sigma\phi}{\lambda_X + \sigma_X\phi}$$

Under high fluxes the decay term becomes negligible.

Xenon Poisoning (3)

For instantaneous shutdown we may assume the flux going to zero¹ leading to a solution for the system in the form

$$I(t) = I_0 e^{-\lambda_I t}$$

$$X(t) = X_0 e^{\lambda_X t} + \frac{\lambda_I}{\lambda_I - \lambda_X} I_0 (e^{-\lambda_X t} - e^{-\lambda_I t})$$

which using $I_0 = I_\infty$, $X_0 = X_\infty$ can finally lead to

$$X(t) = \Sigma \phi \left[\frac{\gamma_I + \gamma_X}{\lambda_X + \sigma_X \phi} e^{-\lambda_X t} + \frac{\gamma_I}{\lambda_I - \lambda_X} (e^{-\lambda_X t} - e^{-\lambda_I t}) \right]$$

The final effect is a decrease in reactivity proportional in first approximation to $\sigma_X X(t)$.

¹In the final formulas (also for next slides) ϕ refers to the flux value before shutdown.

Xenon Peak

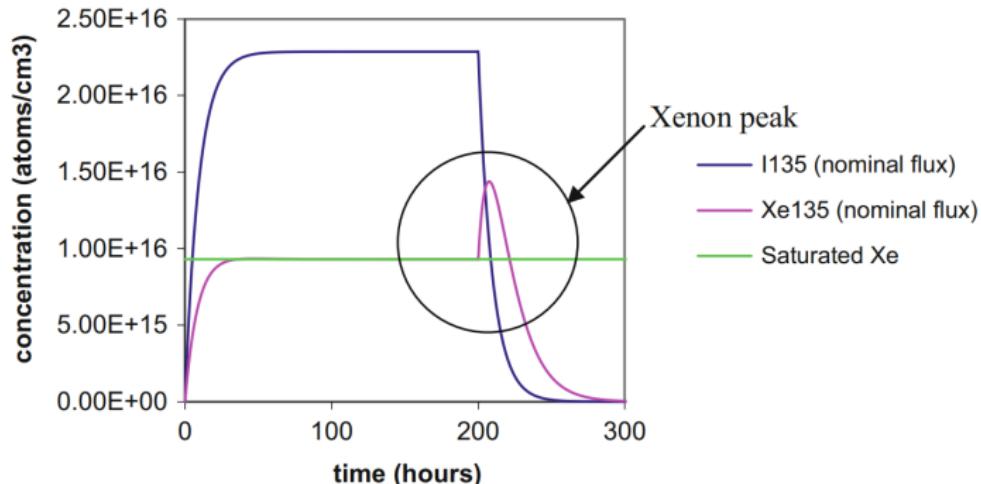


Fig. 15.33 Iodine 135 and xenon 135 transients following instantaneous reactor shutdown after 200 operating hours at nominal power

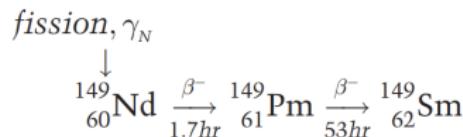
The Xenon peak impacts the reactivity, possibly stopping the ability to restart the chain reactions, anti-reactivity may be counterbalanced by reduction of control bars insertion (Chernobyl). Safer alternative is waiting for ¹³⁵Xe to decay.

► Samarium Poisoning

$^{149}_{62}\text{Sm}$ is another important thermal neutron absorber. It is stable and therefore it keeps poisoning the fuel even after long shutdowns.

Production mechanisms of ^{149}Sm include neutron capture from ^{148}Sm , $^{149}_{61}\text{Pm}$ decay and as a fission product. ^{149}Sm does not reach its saturation value, but due to its stable nature it increases in concentration with operation time. It is sometimes already included in control bars.

By similar arguments to those used for the concentration of ^{135}Xe we can retrieve the equation for $S(t)$ taking $\lambda_S = 0$ since it is stable and γ_S from the reduction of the ^{149}Pm - ^{149}Sm chain.



$$S(t) = \gamma_P \Sigma \phi \left[\frac{1}{\sigma_S \phi} + \frac{1}{\lambda_P} (1 - e^{-\lambda_P t}) \right]$$

► Samarium Curve

Evolution of samarium population after shutdown described by:

$$S(t) = \gamma_P \Sigma \phi \left[\frac{1}{\sigma_S \phi} + \frac{1}{\lambda_P} (1 - e^{-\lambda_P t}) \right]$$

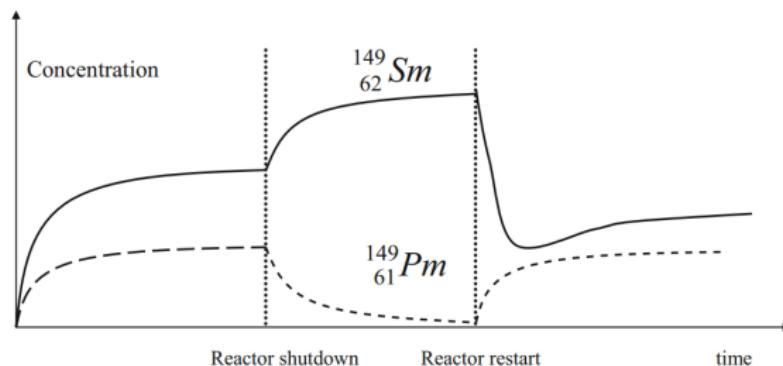


Fig. 15.36 Evolution of samarium concentration after reactor shutdown

► Gadolinium Poisoning

Gadolinium is an element with several neutron absorbing isotopes, ^{155}Gd and ^{157}Gd having the highest capture cross sections. It serves as burnable absorber, typically in the Gd_2O_3 form, so that the reactor can reach gradually operating conditions, with absorbing isotopes disappearing during the cycle. Burnable poison concentrations follow simply

$$\frac{d}{dt}N^{BP}(t) = -N^{BP}(t)\sigma\phi(t)$$
$$N^{BP}(t) = N_0^{BP} e^{-\sigma\phi(t)}$$

Burnable poison distribution can also help reduce localised power peaks and dishomogeneities in the reactor.

Gadolinium Curve

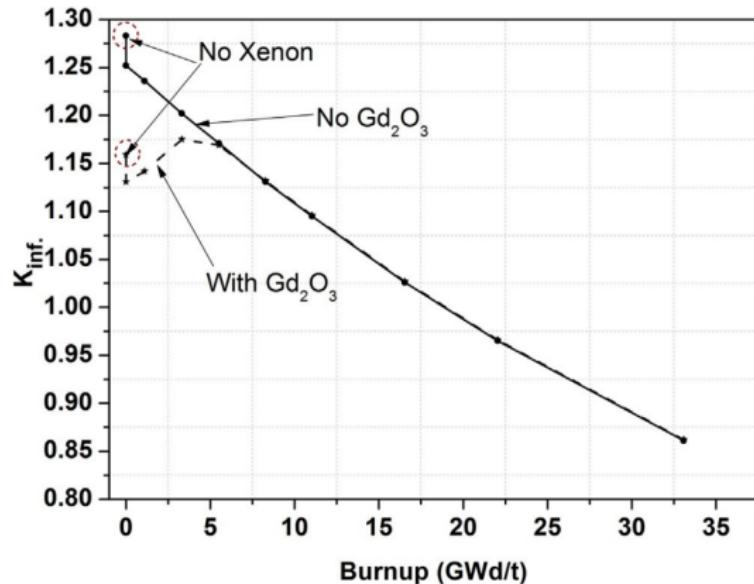


FIG. 6.4 Variation of k_{∞} of 2.4% enriched U oxide BWR fuel assembly without and with 1.5% Gd_2O_3 in two pins.

Flatter curve in the beginning of the multiplication process, leads to easier control of the reactor during starting of operations.

► Burnable Boron

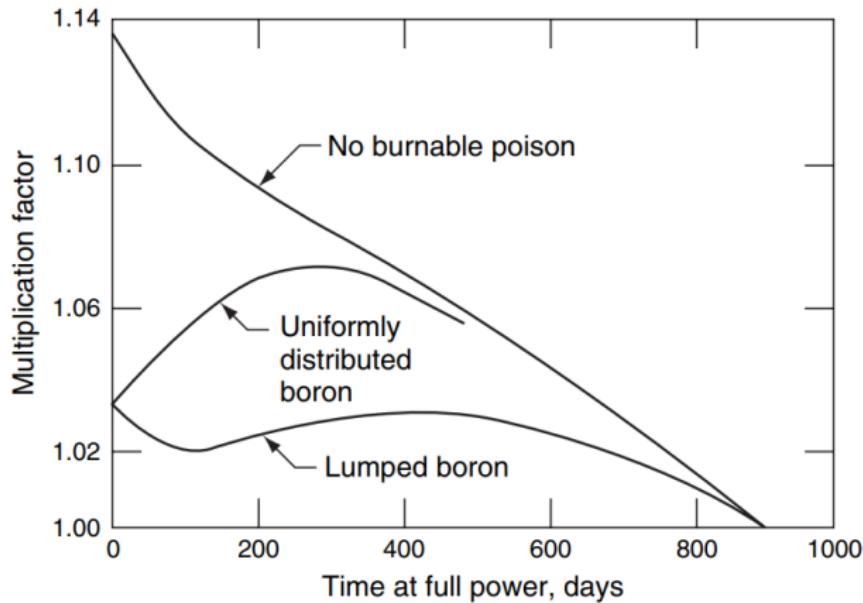


FIGURE 10.4 Effect of burnable poisons on reactor multiplication (adapted from "Kinetics of Solid-Moderator Reactors," by H. B. Steward and M. H. Merrill, *Technology of Nuclear Reactor Safety*, Vol. 1, 1965, T. J. Thompson and J. G. Beckerley, Eds. Courtesy of the MIT Press).

Section 7

Industrial Fuel Cycle

► Industrial Fuel Cycle

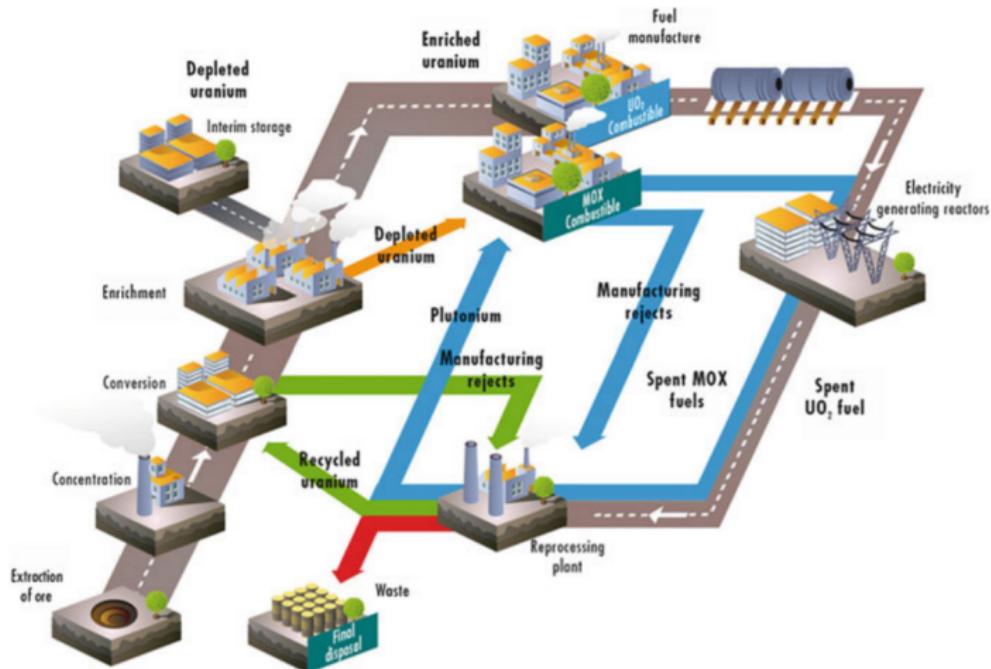


Figure: Typical fuel cycle with recycling.

► Uranium Extraction

Uranium may come from

- land sites (today most common origin)
- ocean water (3 mg/ton of water, may be relevant in the future)

Reports on Uranium resources and production are periodically issued by the Nuclear Energy Agency and the International Atomic Energy Agency².

²NEA/IAEA (2021), Uranium 2020: Resources, Production and Demand, OECD Publishing, Paris, <https://doi.org/10.1787/d82388ab-en>

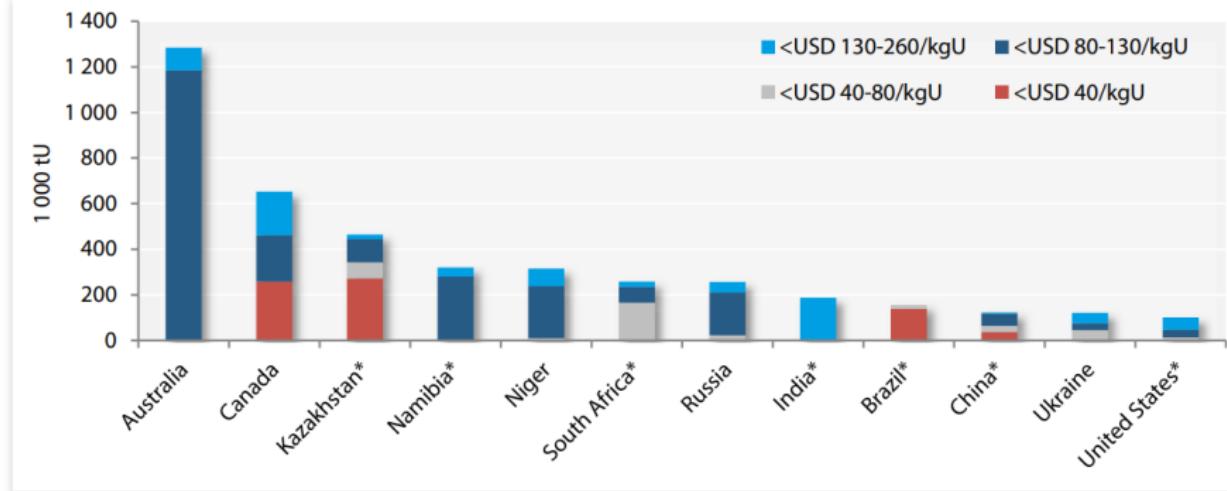
► Uranium Natural Reserves

Figure 1.1. Global distribution of identified resources
($\text{USD } 130/\text{kgU}$ as of 1 January 2019)



► Uranium Natural Reserves (2)

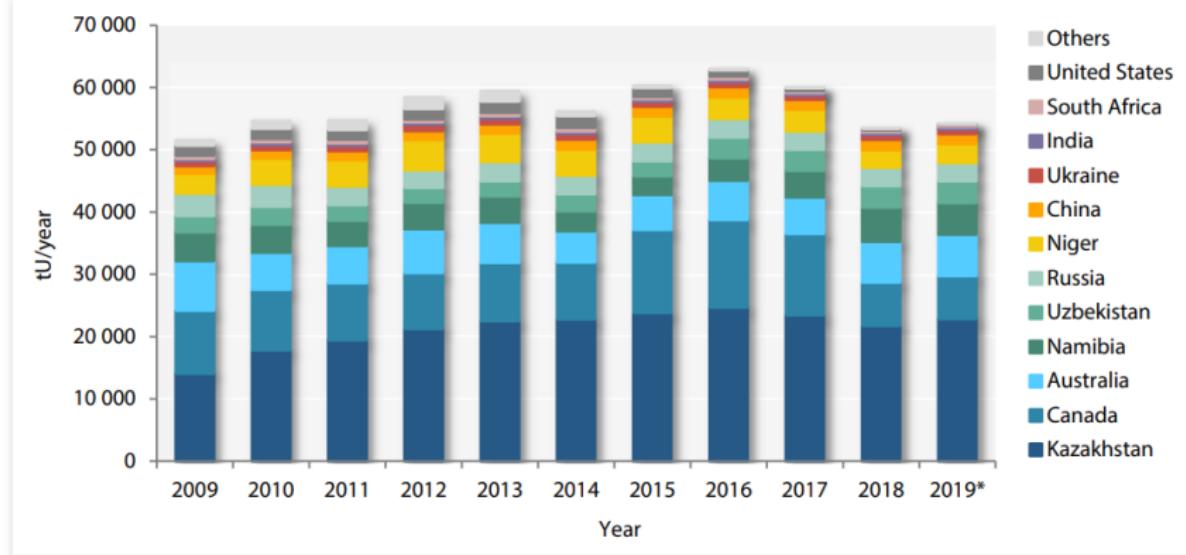
Figure 1.2. Distribution of reasonably assured resources (RAR) among countries with a significant share of resources



* Secretariat estimate or partial estimate.

► Uranium Extraction

Figure 1.6. Recent world uranium production (tU/year)



"Others" includes the remaining producers (see Table 1.18 and previous Red Book editions).

* NEA/IAEA estimate.

► Uranium Processing



Figure: Yellowcake ($\text{Na}_2\text{U}_2\text{O}_7$ Sodium Diuranate) is the final product of extraction.

The yellowcake is then converted via a process called calcination to UO_3 which in turn reduces to UO_2 .

If the Uranium is intended as fuel for CANDU heavy-water moderated reactors no further processing is required.

If the fuel instead is intended for reactors working with enriched uranium, typically light-water moderated reactors, some further processing is required. First UO_2 is hyper-fluorinated to UF_4 and then fluorinated to UF_6 .

► Uranium Enrichment: Diffusion Cascade

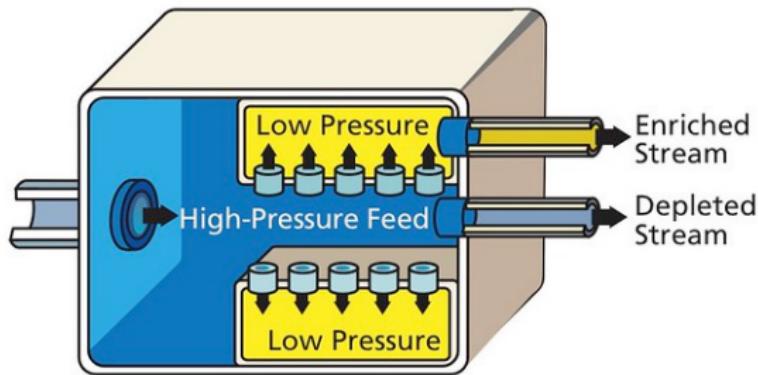
UF_6 is solid at room temperature but it sublimates already at $\approx 56\text{ }^\circ\text{C}$, the gaseous form allows the enrichment process.

The first enrichment procedure ever developed was based on Graham's diffusion law and allowed the construction of the Little Boy atomic bomb. The facility built for the Manhattan Project was called K-25, located in Oak Ridge, Tennessee. It employed more than 3000 diffusers.

Graham's law states that fluency rate is inversely proportional to the mass of the effusing molecule. By this argument it can be seen that UF_6 will diffuse differently depending on the isotope of U present in the molecule (note that fluorine is almost 100% pure in the ^{19}F isotope)

$$\frac{\text{Rate}_{235\text{UF}_6}}{\text{Rate}_{238\text{UF}_6}} = \sqrt{\frac{M_{238\text{UF}_6}}{M_{235\text{UF}_6}}} (= \sqrt{\frac{352}{349}} \approx 1.0043)$$

► Uranium Enrichment: Diffusion Cascade (2)



The depleted stream gets injected into the previous chamber while the enriched goes into the next one.

The main issue of this system is its enormous energy cost, and as of today this technique is considered obsolete.

► Uranium Enrichment: Ultracentrifugation

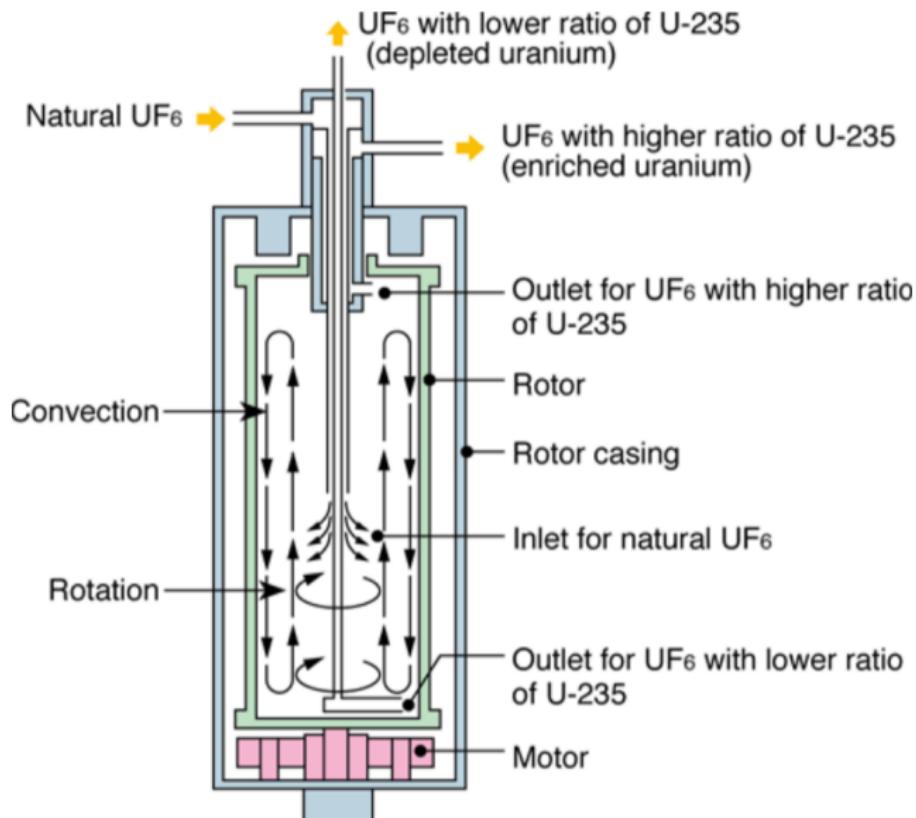
A better approach to enrichment of U is ultracentrifugation.

Since heavier $^{238}\text{UF}_6$ tends to move outwards in a centrifuge compared to lighter $^{235}\text{UF}_6$ which in turn will tend to remain closer to the centrifugation axis it is possible to construct a long chain of centrifuges spinning at about 50 krpm.

With this approach it is still possible to enrich uranium to the desired concentration of ^{235}U at a fraction of the energy cost ($\approx -95\%$) required by diffusion cascade techniques.

Other enrichment techniques exist but do not find industrial scale implementation.

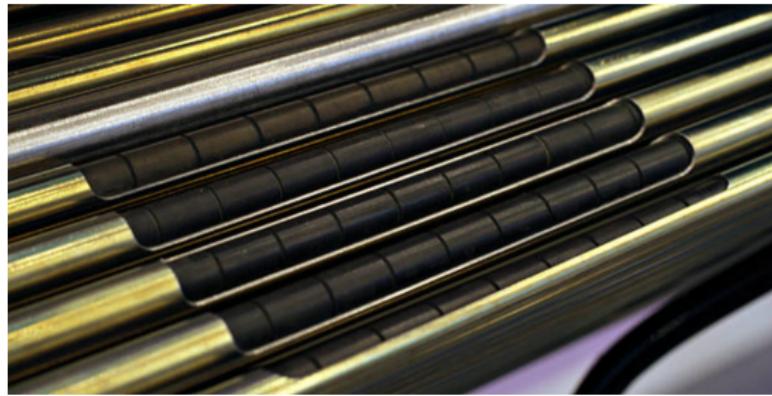
► Uranium Enrichment: Ultracentrifugation (2)



► Uranium Pellets and Bars

Upon reaching the target enrichment UF_6 gets converted into UO_2 powder, which is then turned via sintering into fuel pellets.

UO_2 are then placed inside Zircaloy ($>90\%$ Zirconium) tubes which will constitute the fuel assembly. An assembly contains around 500 Kg of Uranium and a fuel loading of a nuclear plant requires around 50 assemblies. The loading will provide approximately 4 years of energy before fuel change.

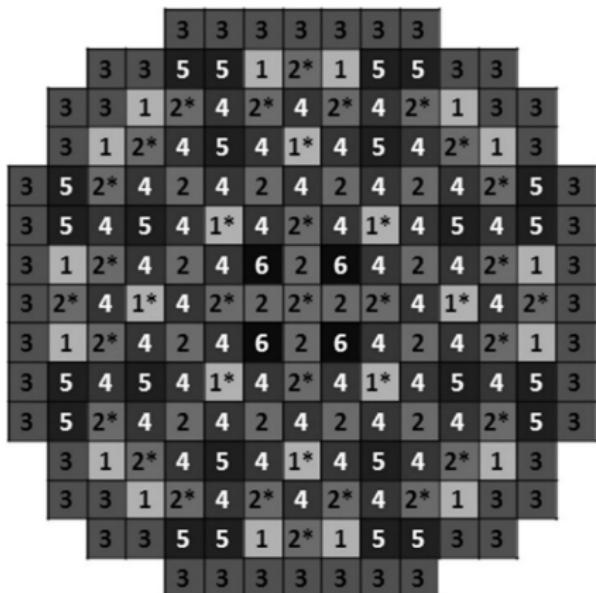


► Fuel Management

Fuel management and loading are very reactor dependant

- Burn up levels
- Loading configurations:
 - Out-to-in (fresh fuel far from centre) →
 - + easier to control,
 - higher neutron leakage
 - In-to-out (fresh fuel in the centre) →
 - + lower neutron leakage,
 - power peaked region in the centre
 - Scattered distribution (see next slide) →
Try to control both flux/leakage and power peaks

► Scattered Load Pattern

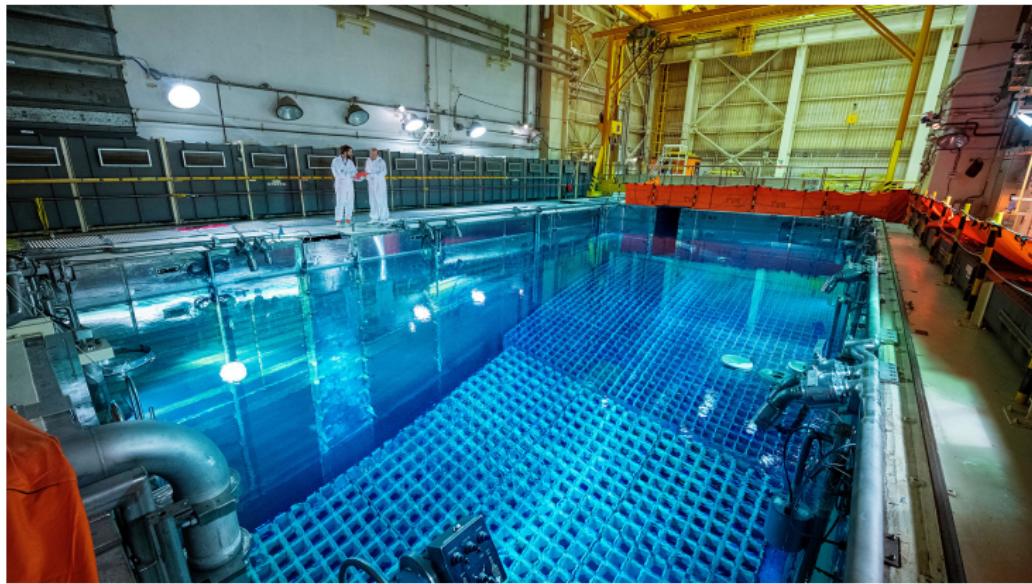


	ASSEMBLY TYPE
1	Fresh
2	Once Burned
3	Twice Burned
4	Thrice Burned
5	Fresh + Burnable Absorber
6	Once Burned + Burnable Absorber
*	Assemblies with Control Rods

FIG. 6.11 A typical equilibrium loading pattern in a PWR with multiple cycles.

► Cooling Pools

After unloading spent fuel is cooled in cooling pools for some time (months to years) in order to reduce the power produced, given by the decay of short and medium lived isotopes.



► Fuel Treatment and Recycling

After cooling assemblies are moved to special facilities. Here typically bars are pretreated. Pretreatment applies a sort of primary separation of components, trying to simplify (and reduce the cost of) the later steps of the process. Pretreatment may include, but is not limited to, shredding or cutting parts of the assembly.

After pretreatment the waste is ready for treatment, which aims to separate radioactive components via dissolution and centrifugation. Typically in the first phase fission products are separated. In the industrial lines in which fuel recycling is implemented after this phase there is an extra phase in which uranium and plutonium are extracted and separated from the rest of the fuel.

Finally highly radioactive by-products undergo a conditioning phase, in which they are solidified into cement or glass and then sealed in containers, which get buried in underground permanent storage facilities.

Conclusions

In this presentation we analysed what happens during the life-cycle of nuclear fuel, first by analysing the **reactions** which it undergoes and the **equations** describing the evolution of its components.

Then some **techniques** implemented in the study of fuel isotope populations were presented, with also some hints on the effects of the major **poison elements**.

Finally a typical **industrial fuel life-cycle** was presented.

Thank you for your attention.

Section 8

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