



Aalto University
School of Science
and Technology

PHYS-E0562 Nuclear Engineering, advanced course

Lecture 5 – Burnup calculation

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Outline

Background

Effects of burnup on neutronics and reactor operation:

- ▶ Reactivity
- ▶ Reactor safety
- ▶ Fission product poisons
- ▶ Decay heat and radioactive inventory

Formulation of the depletion problem:

- ▶ Radioactive decay and neutron-induced transmutation reactions
- ▶ Formulation of the Bateman depletion equations
- ▶ Linear chains solution
- ▶ Matrix exponential solution

Burnup algorithms:

- ▶ Burnup calculation as an example of a non-linear coupled problem
- ▶ Explicit Euler and predictor-corrector methods

Background

Nuclear fuel in light water reactors is loaded in the reactor core for the duration of the entire operating cycle, which is typically 12 or 18 months. After the cycle is completed, 1/3 or 1/4 of the fuel assemblies are removed and replaced with fresh fuel, and the core is shuffled to re-optimize the loading pattern.

This means that a single fuel assembly spends 3 to 4 years in the reactor, and each core loading has a history extending several years in the past. During this time, the neutronics properties of the fuel change due to various changes in the isotopic composition:¹

- ▶ ^{235}U is depleted and replaced by ^{239}Pu as the primary fissile isotope
- ▶ Non-fissile plutonium and minor actinides and fission products are accumulated in the fuel
- ▶ Burnable absorber used for passive reactivity control is depleted

These changes are directly reflected in reactivity and the safety parameters of the reactor core. The accumulation of radioactive fission products also forms the source term for accident analyses and final disposal of spent fuel.

Fuel utilization is measured in units of burnup, which refers to the amount of extracted energy per uranium or heavy metal mass (for example, 40 MWd/kgU).

¹The effects of fuel burnup covered in this lecture are focused on neutronics, but exposure to high temperature and neutron irradiation also causes significant changes in heat transfer and mechanical properties of the materials. These topics will be covered in a separate lecture.

Background

Simulating the changes in the neutronics properties of fuel during irradiation requires burnup calculation, which essentially implies tracking the nuclide compositions from fresh fuel to discharge. Since macroscopic cross sections depend on fuel composition, accounting for these changes means that the transport problem becomes non-linear.

In practice, this non-linearity is handled by linearizing the problem over some time interval and obtaining the coupled solution by iterating between the solvers. In reactor analysis, the term “burnup calculation” may refer to different procedures, depending on the context:

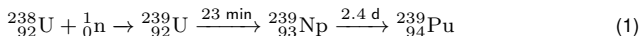
- ▶ In assembly-level calculations (spatial homogenization) the term refers to assembly burnup calculation, in which the changes in the isotopic compositions of each depletion zone, fuel pin or pin type are tracked at the microscopic level, and group constants generated for a representative set of burnup points.
- ▶ In core-level calculations (fuel cycle simulation), the information on the concentrations of (most) individual nuclides is lost in the process of homogenization, and the changes are instead reflected in the parametrization of the group constant data.²

This lecture is mainly focused on burnup calculations at the microscopic level, and parametrization of group constants is left for Lecture 8. The qualitative effects of burnup on reactor operation and radioactive source terms is covered before going into the formulation and solution of the depletion equations.

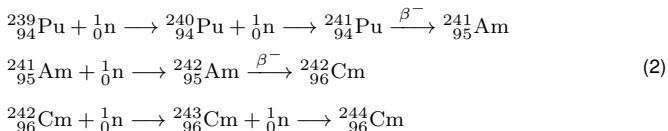
²Fission product poisons ^{135}Xe and ^{135}Sm are often handled explicitly in core-level calculations as well.

Effects of burnup on neutronics: reactivity

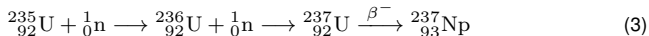
Changes in the composition of fissile isotopes is directly reflected in reactivity. The fraction of fissile uranium (^{235}U) falls from $\sim 3\text{-}5\%$ in fresh fuel to below 1% at discharge. Fissile plutonium is simultaneously produced from neutron capture in ^{238}U :



The transmutation chain continues to ${}^{240}\text{Pu}$, ${}^{241}\text{Pu}$, etc., and via beta-decay to isotopes of americium and curium:



In addition, ${}^{237}\text{Np}$ is produced from ${}^{235}\text{U}$ via neutron capture and beta decay:³



³Another production path to ${}^{237}\text{Np}$ is via β -decay of ${}^{241}\text{Am}$.

Effects of burnup on neutronics: reactivity

Uranium and plutonium isotopes are referred to as the major actinides, and neptunium, americium and curium correspondingly as the minor actinides.

As noted in Lecture 1, the fission probability of actinides is strongly affected by the parity effect. When the compound nucleus is formed, the absorbed neutron brings in:

- 1) Binding energy
- 2) Kinetic energy

the binding energy component depends on the nucleon configuration, and nuclides with even number of protons (even-Z) or neutrons (even-N) tend to be more tightly bound.

The fission barrier is typically in the order of 5-6 MeV, and the parity effect is seen in that:

- ▶ Odd-N actinides (^{235}U , ^{239}Pu , ^{242}Am , etc.) are usually fissile, i.e. they can undergo fission regardless of the neutron energy (with the binding energy alone)
- ▶ Even-N actinides (^{232}Th , ^{238}U , ^{237}Np , ^{240}Pu , ^{241}Am) are fissionable, i.e. they require excess kinetic energy (~ 1 MeV) for fission to occur

The low fission probability of even-N actinides in LWR flux spectrum leads to the accumulation of these isotopes, which together with fission products adds parasitic absorption and reduces reactivity.

Effects of burnup on neutronics: reactivity

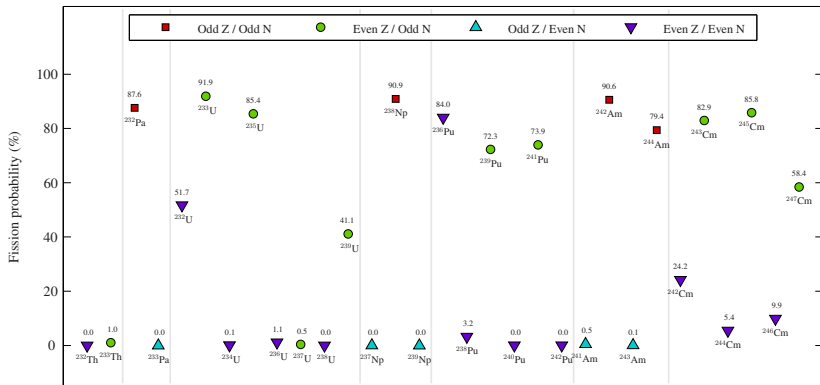


Figure 1 : Probability of fission induced by thermal neutrons (Maxwellian energy distribution corresponding to room temperature). If the binding energy released in neutron absorption exceeds the fission barrier, the nuclide can be fissioned by low-energy neutrons. This is more likely to happen with odd-N nuclides, for which the energy release is higher because of the parity effect.

Effects of burnup on neutronics: reactivity

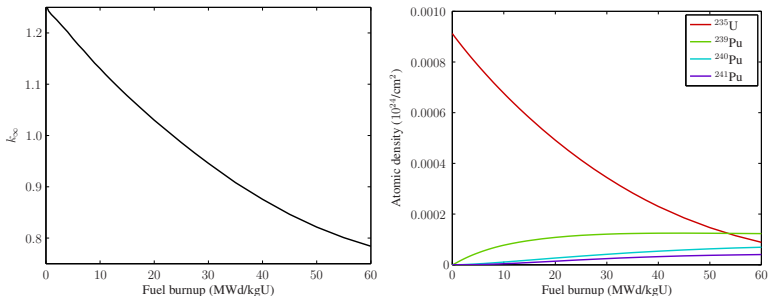


Figure 2 : The composition of nuclear fuel changes along with the reactor operating cycle. The depletion of uranium and build-up of plutonium, minor actinides and fission products changes the neutronics characteristics of the core. Left: Infinite multiplication factor as function of fuel burnup. Right: depletion of ^{235}U and build-up of plutonium isotopes in PWR fuel as function of burnup.

Effects of burnup on neutronics: reactivity

Maintaining criticality over the operating cycle requires sufficient amount of excess positive reactivity, and an equal amount of negative reactivity in the capacity of control systems (control rods and boron shim) to compensate for it at beginning of cycle.

But there are also other limitations:

- ▶ Too much boron in the coolant results in positive moderator void coefficient
- ▶ Too much absorption in control rods leads to prohibitively high reactivity insertion in rod ejection (PWR) / drop (BWR) transients

The amount of excess reactivity can be compensated for by passive reactivity control using burnable absorbers, i.e. absorbers that are slowly depleted by neutron irradiation. Typical burnable absorbers include:

- ▶ Boron silicate glass rods inserted in the control rod guide tubes of selected fuel assemblies (used in PWR's, absorber isotope ^{10}B)
- ▶ Gadolinium oxide mixed with uranium oxide in selected fuel pins (used in PWR's and BWR's, absorber isotopes ^{155}Gd and ^{157}Gd)

The absorber is usually fully depleted well before the end of the assembly's life.

Effects of burnup on neutronics: reactivity

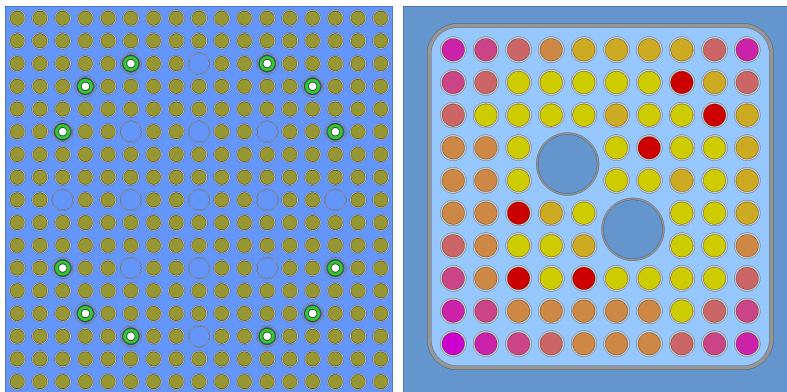


Figure 3 : Illustration of burnable absorbers in LWR fuel assemblies. Left: 12 boron silicate glass rods inserted in the control rod guide tubes of PWR fuel assembly. Right: 6 gadolinium-doped fuel pins (red) in a BWR assembly.

Effects of burnup on neutronics: reactivity

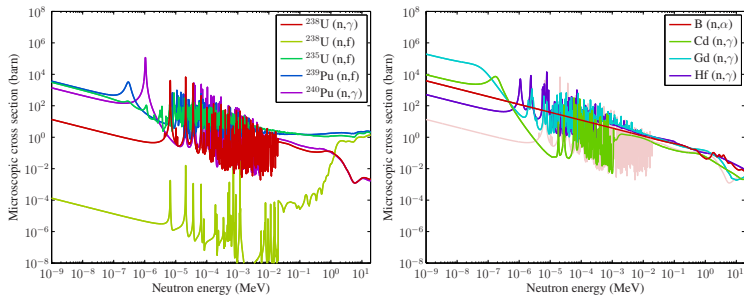


Figure 4 : Microscopic cross sections of fuel and absorber isotopes. Left: fission and radiative capture cross sections of actinides. Right: absorbers used for active and passive reactivity control. Capture cross section of ^{238}U is plotted in the background for comparison.

Effects of burnup on neutronics: reactivity

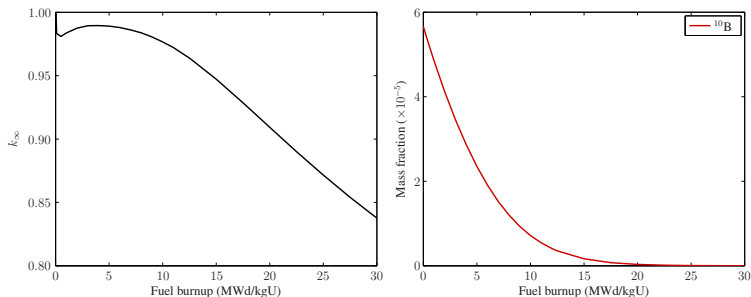


Figure 5 : Depletion of a “gray” boron silicate glass burnable absorber in a PWR fuel assembly calculation. Left: Infinite multiplication factor, Right: Absorber concentration. The absorber is natural boron (20% ^{10}B , 80% ^{11}B), but only the high-absorbing isotope is depleted.

Effects of burnup on neutronics: reactivity

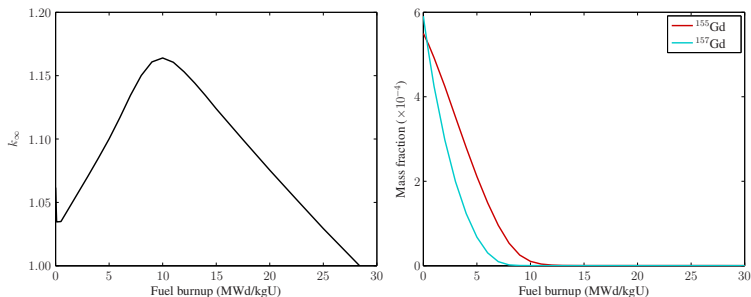
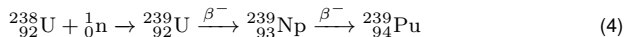


Figure 6 : Depletion of a gadolinium oxide burnable absorber in a BWR fuel assembly calculation. Left: Infinite multiplication factor, Right: Absorber concentrations. The absorber is natural gadolinium (0.2% ^{152}Gd , 2.1% ^{154}Gd , 14.8% ^{155}Gd , 20.6% ^{156}Gd , 15.7% ^{157}Gd , 24.8% ^{158}Gd , 21.8% ^{160}Gd), but only the two high-absorbing isotopes are depleted.

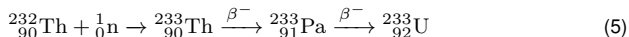
Effects of burnup on neutronics: reactivity

As mentioned above, the reactor not only consumes, but also produces new fissile material, which compensates for the loss of reactivity during irradiation. In uranium-fueled reactors the process is based on the conversion of fertile ^{238}U to fissile ^{239}Pu (and ^{241}Pu after two more neutron captures):



^{239}Pu becomes the main fissile isotope in LWR fuel during the second half of the assembly's life time. In high-burnup fuel, more than 80% of fission power can be produced from plutonium isotopes.

Similar fertile-to-fissile conversion occurs in thorium-fueled reactors:



Conversion ratio, c determines the ratio between the production and depletion rate of fissile material. When $c > 1$, the process is referred to as breeding, and the reactor produces more fissile material than it consumes.

Achieving breeding in the $^{238}\text{U} - ^{239}\text{Pu}$ cycle requires a fast spectrum system, but in the $^{232}\text{Th} - ^{233}\text{U}$ cycle breeding can be achieved in thermal reactors. In practice, breeder reactor cycles also require reprocessing of the spent fuel before the next operating cycle.

Effects of burnup on neutronics: reactor safety

Since ^{239}Pu is produced from ^{238}U , mainly from captures in the low resonance range above thermal region, the amount of moderation has a significant impact in plutonium production. The difference is seen in BWR's, where more plutonium is produced for given fuel burnup in the upper part of the assemblies, where the spectrum is harder.

The cross sections of plutonium isotopes are higher compared to uranium in the thermal energy range. Increasing parasitic absorption and the contribution of ^{239}Pu in total fission rate is also reflected in the safety characteristics:

- ▶ The accumulation of non-fissile plutonium, minor actinides and fission products increases spatial self-shielding and hardens the flux spectrum, which decreases the reactivity worth of control rods and soluble boron.
- ▶ Average number of prompt neutrons emitted in ^{239}Pu thermal fission is about 2.8, compared to 2.4 for ^{235}U , which adds to its reactivity value. Delayed neutron yield is considerably lower, and high ^{239}Pu content shortens reactor time constants and lowers the margin to prompt super-criticality.
- ▶ The changes in spectrum and isotopic composition can also affect the magnitude of reactivity feedback coefficients, but the underlying mechanisms are not straightforward.⁴

⁴The overall effect is the sum of multiple factors. ^{240}Pu has a high capture resonance at 1 eV, which is reflected in Doppler feedback. Feedback effects are also affected by the build-up of ^{239}Pu and the hardening of the flux spectrum. In PWR's the reduced coolant boron concentration also plays a major role.

Effects of burnup on neutronics: reactor safety

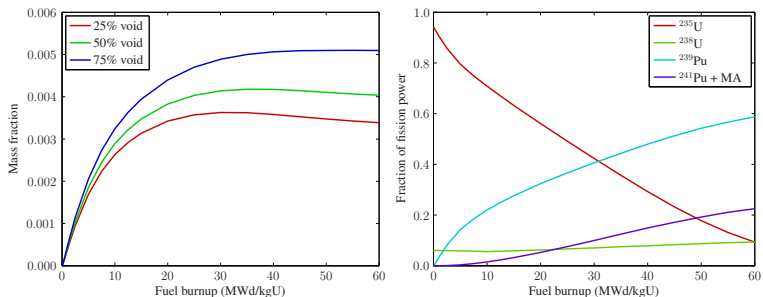


Figure 7 : Left: Plutonium production at different coolant void fractions in a BWR fuel assembly. Right: Contributions of ^{235}U , ^{239}Pu and ^{241}Pu in power production in a BWR assembly (25% void fraction) as function of burnup.

Effects of burnup on neutronics: reactor safety

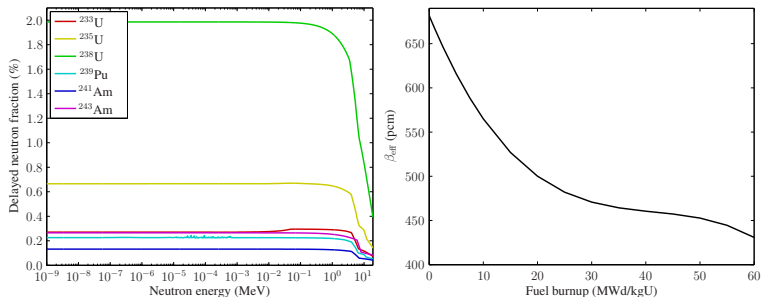


Figure 8 : Left: delayed neutron fraction of selected actinides as function of neutron energy. The yield depends on the probability of producing precursor isotopes, which depends on the fission product distribution of the actinide and the neutron energy. The fraction additionally depends on the prompt neutron yield, which varies from nuclide to nuclide and increases practically linearly as function of neutron energy. Right: Effective delayed neutron fraction in PWR fuel assembly as function of fuel burnup (1 pcm = 0.001%). Large plutonium and minor actinide content leads to lower delayed neutron fraction compared to fresh uranium fuel.

Effects of burnup on neutronics: reactor safety

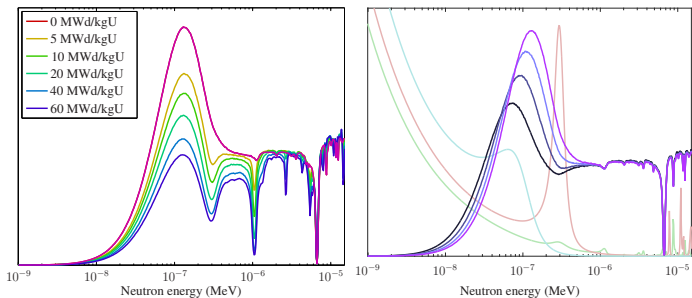


Figure 9 : Left: Effect on increasing burnup on the thermal end of flux spectrum in a BWR fuel pin (40% coolant void fraction). The shape of ^{239}Pu and ^{240}Pu cross sections (See Fig. 4) are clearly reflected in the results. Increasing burnup leads to harder spectrum, which reduces the reactivity worths of absorbers and affects feedback coefficients. Right: Effect of coolant temperature on the distribution of thermalized neutrons. Shapes of ^{235}U , ^{239}Pu fission and ^{135}Xe capture cross sections are plotted on the background in green, red and cyan, respectively (not to scale). Increasing the temperature moves the thermal peak upwards on the energy scale. This reduces the fission rate of ^{235}U . For ^{239}Pu the impact is opposite, because of the peak located right above the distribution. Also the capture rates of high-absorbing fission products are reduced, which results in a positive reactivity effect. The effect of spectral shift is usually not as strong as the moderator density effect, which in LWR's causes a negative feedback. (See Lecture 1).

Effects of burnup on neutronics: fission product poisons

Hundreds of intermediate-mass nuclides are produced in the fuel during irradiation by fission reactions. The accumulation of all fission product isotopes increases parasitic neutron absorption in fuel, which leads to similar deterioration of reactivity and spectrum-hardening as the accumulation of non-fissile actinides.

Two isotopes, commonly referred to as fission product poisons: ^{149}Sm and ^{135}Xe have exceptionally high capture cross sections in the thermal region.⁵ These isotopes have a direct impact in neutronics, even at low concentrations.

Tracking the concentrations of ^{149}Sm and ^{135}Xe is complicated by the fact that the two nuclides are produced not only in fission, but also in the decay chains of their precursor isotopes, which means that their concentrations follow changes in fission rate with a considerable delay.

^{135}Xe is a radioactive nuclide that decays into low-absorbing ^{135}Cs with a half-life of 9 hours after reactor shut-down. During reactor operation, the dominant removal mechanism is neutron capture. ^{149}Sm is stable, and only removed by capture.

⁵The capture cross section of ^{135}Xe is about a factor of 10^6 larger than that of ^{238}U .

Effects of burnup on neutronics: fission product poisons

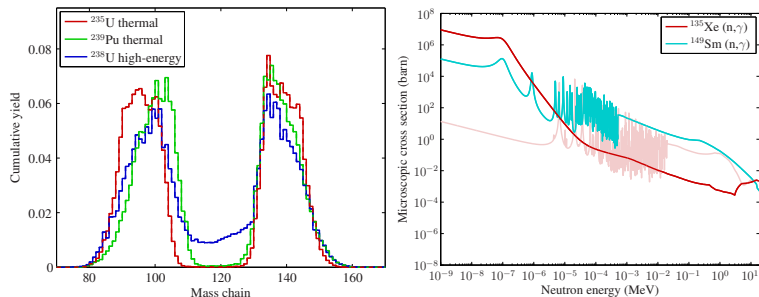


Figure 10 : Left: The double-peaked fission product distribution. The yields are calculated as cumulative values over each mass chain. The lower peak shifts to the right as the mass of the fissioned actinide increases. High-energy fission for ^{238}U refers to reaction caused by 14 MeV neutrons. Increasing neutron energy lifts the distribution between the two peaks. Right: Microscopic cross sections of fission product poisons ^{135}Xe and ^{140}Sm Capture cross section of ^{238}U is plotted in the background for comparison.

Effects of burnup on neutronics: fission product poisons

Samarium- and xenon-poisoning are characterized by time constants that depend on the production of the isotopes in their precursor chains and removal by radioactive decay (^{135}Xe only) and neutron capture. The relevant time-scale is measured in several days.

Samarium poisoning

Samarium-149:

- ▶ $\sigma_{\gamma} = 4.0 \cdot 10^4$ barn (thermal)
- ▶ Stable nuclide

Precursor decay chain:



Fission yields: (Thermal ^{235}U fission)

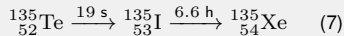
- ▶ ${}^{149}\text{Nd}$: 0.011
- ▶ ${}^{149}\text{Pm}$: ~ 0
- ▶ ${}^{149}\text{Sm}$: ~ 0

Xenon poisoning

Xenon-135:

- ▶ $\sigma_{\gamma} = 2.6 \cdot 10^6$ barn (thermal)
- ▶ $T_{1/2} = 9.1 \text{ h}$

Precursor decay chain:



Fission yields: (Thermal ^{235}U fission)

- ▶ ${}^{135}\text{Te} + {}^{135}\text{I}$: 0.0620
- ▶ ${}^{135}\text{Xe} + {}^{135\text{m}}\text{Xe}$: 0.0026

Effects of burnup on neutronics: fission product poisons

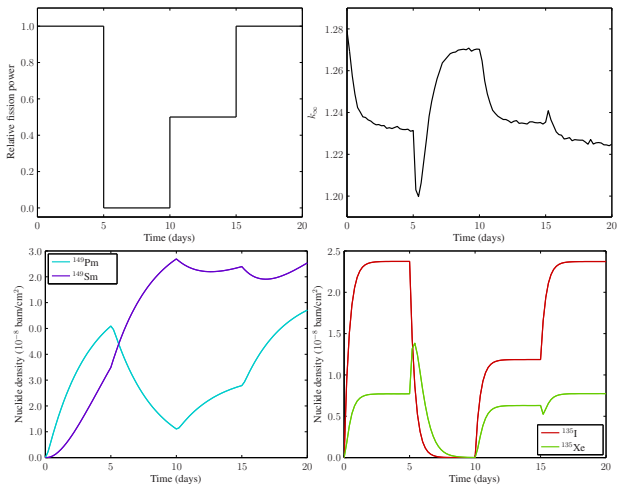


Figure 11 : Concentrations of ¹⁴⁹Sm (bottom left) and ¹³⁵Xe (bottom right) and their precursors following the changes in power level (top left). The changes in absorption rate are reflected in k_{∞} (top right).

Effects of burnup on neutronics: fission product poisons

The behavior of ^{135}Xe concentration in fuel is characterized by two physical factors:

- 1) The source term is dominated by the decay of ^{135}I , which means that the production rate of ^{135}Xe follows changes in power level with a delay characterized by the precursor half-life (6.6 h)
- 2) The loss term is dominated by neutron capture, which means that the loss rate of ^{135}Xe follows changes in power level instantaneously

The result is a positive feedback-effect, that tends to amplify distortions in the distribution of ^{135}Xe .

The positive feedback and the fact that the concentration of ^{135}Xe keeps increasing after reactor shutdown may lead to a condition where the negative reactivity exceeds the positive reactivity reserve, making start-up impossible until the isotope has decayed from the fuel. This period is known as the xenon dead time.⁶

Xenon poisoning played a crucial role in the events leading to the runaway reactivity excursion in the Chernobyl accident in 1986.

⁶Xenon poisoning was discovered during the Manhattan project, when the first plutonium production reactors were built in Hanford, Washington. An unidentified phenomenon forced the reactors to shut down after a relatively short period of operation. Before that time, all reactor experiments were performed at low power, and the build-up of fission products was not an issue.

Effects of burnup on neutronics: fission product poisons

In large loosely-coupled systems the positive feedback between power and xenon concentration may also lead to the following sequence of events:⁷

- 1) When the reactor is operated at constant power, a disruption in the equilibrium power distribution leads to increasing xenon concentration in a region of the core where power is suddenly lowered.
- 2) The flux is pushed to another part of the core. The effect is amplified by the increased xenon burnout in the high flux region.
- 3) Increase in flux leads to increase in power and the production of ^{135}I . The concentration of ^{135}Xe follows behind with a delay.
- 4) When the xenon concentration starts to increase, the flux is again pushed to another location, and the cycle repeats itself.

The result is a spatial oscillation, with a period of about 15 hours. Xenon oscillations are damped by strong negative temperature feedbacks, and managed by core design and active reactivity control. The onset of xenon oscillations also depends on flux spectrum, as ^{135}Xe absorbs mainly low-energy neutrons.

⁷In practice, a loosely coupled system refers to a reactor in which the neutron migration distance is short compared to the dimensions, for example, a large LWR core.

Effects of burnup on radioactive inventory

Reactor fuel comprises the vast majority of radioactive inventory produced during reactor operation. Compared to spent fuel, the activation of coolant and structural materials is practically negligible.⁸

Burnup calculation provides the source terms for accident and final disposal analyses. Since activity is inversely proportional to nuclide life time, different fission product and actinide isotopes dominate the total activity at different time periods.

As discussed later on, short-lived nuclides reach their equilibrium concentrations soon after reactor start-up, while the concentrations of long-lived isotopes depend on fuel burnup. From the viewpoint of safety analyses the source term can be divided into:

- 1) Decay heat, i.e. heat production after reactor shut-down, which is determined by the decay rate of the nuclide and the energy release per decay reaction
- 2) Radiotoxicity, which is determined by the decay rate of the nuclide, energy release, and a number of physiological factors (decay mode, accumulation, biological half-life, etc.)

Both decay heat and radiotoxicity of a mixture of several nuclides can be considered proportional to activity, although there are differences in the decay energies and physiological accumulation of individual nuclides.

⁸The source term for final disposal has to account for certain activation products, such as ^{14}C , which form mobile compounds.

Effects of burnup on radioactive inventory

The main contributors to decay heat during reactor operation and immediately after shut-down are short-lived nuclides, such as:⁹

- ▶ ^{239}U ($T_{1/2} = 23$ minutes)
- ▶ ^{239}Np ($T_{1/2} = 2.4$ days)
- ▶ ^{134}I ($T_{1/2} = 53$ minutes)
- ▶ ^{138}Cs ($T_{1/2} = 32$ minutes)
- ▶ ^{140}Cs ($T_{1/2} = 1.1$ minutes)

The level is saturated to 5-7% of fission power very soon after reactor start-up, and decays exponentially after shutdown.

Heat production from radioactive decay is the most significant threat to fuel integrity during operation and long after the assemblies have been discharged from the reactor. Loss of coolant flow and incapability to remove decay heat lead to the over-heating and melt-down of three reactor cores in the Fukushima accident in 2011.

⁹There are dozens of short-lived nuclides without any clearly dominating isotopes.

Effects of burnup on radioactive inventory

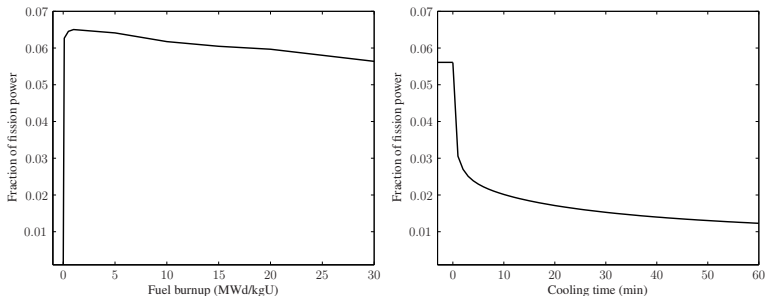


Figure 12 : Decay heat relative to fission power as function of burnup during reactor operation (left), and during the first hour (right) after reactor shutdown. Heat production falls to about 0.5% during the first week, after which the level remains relatively high for a long time.

Effects of burnup on radioactive inventory

Short-lived fission products that are released at low temperature form the most significant radiological source term in nuclear accidents, in particular:

- ▶ Noble gases (released from the gas gap once the cladding is burst, and not easily contained)
- ▶ ^{131}I (some iodine compounds are gaseous at 240°C , but the chemistry is extremely complicated)
- ▶ ^{137}Cs (gaseous compounds at 1300°C)

Melting of fuel at about $2600\text{-}3000^\circ\text{C}$ releases ^{90}Sr and isotopes of barium, ruthenium and lanthanum.

The most significant contributor to radiation dose for inhabitants living within the fallout zone is ^{131}I ($T_{1/2} = 8$ days). Long-term exposure and limitations to land cultivation are mainly due to contamination by ^{137}Cs ($T_{1/2} = 30$ years).

The activity of ^{131}I in the fuel saturates within 30 days of continuous reactor operation. The saturation of ^{137}Cs takes much longer than the fuel is irradiated in the reactor, which means that the core inventory depends on burnup.

Effects of burnup on radioactive inventory

Long-lived radionuclides in spent fuel form the radiological hazard in nuclear waste. Important contributors include:

- ▶ Plutonium isotopes: ^{239}Pu ($T_{1/2} = 24,000$ years), ^{240}Pu ($T_{1/2} = 6500$ years)
- ▶ Minor actinides: ^{237}Np ($T_{1/2} = 2$ million years), ^{241}Am ($T_{1/2} = 430$ years), ^{243}Am ($T_{1/2} = 7400$ years)
- ▶ Long-lived fission products: ^{99}Tc ($T_{1/2} = 210,000$ years), ^{129}I ($T_{1/2} = 15$ million years), ^{135}Cs ($T_{1/2} = 2$ million years)

The contribution of uranium decay products becomes significant after about 100,000 years.

Decay heat must be taken into account in intermediate storage and the beginning of final disposal. For the first few centuries after discharge the heat production is dominated by fission products, such as ^{137}Cs and ^{90}Sr , together with ^{238}Pu ($T_{1/2} = 88$ years).¹⁰

NOTE: *Radioactive inventory forms only the source term for final disposal analyses. The radiological hazard is determined by how these nuclides are transported to ground water and food chain.*

¹⁰Plutonium-238 is produced from ^{237}Np by neutron capture and β -decay and from ^{239}Pu by (n,2n) reactions.

Effects of burnup on radioactive inventory

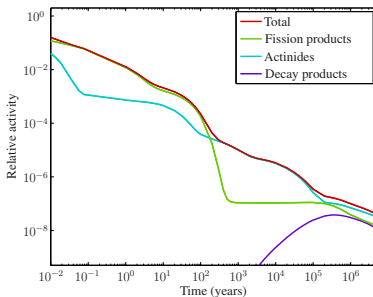


Figure 13: Spent LWR fuel activity after disposal (relative to time of reactor shutdown). The activity is dominated by fission products for the first few centuries after shut-down, followed by actinides (^{241}Am , ^{240}Pu , ^{239}Pu). Long-lived fission products and uranium decay products become significant in the 100,000+ years time scale.

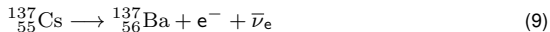
Radioactive decay: decay modes

The most common decay modes are beta-decay and alpha-decay. In beta-decay, a neutron is converted into proton or vice versa.

β^- decay is written as:



for example:



β^+ decay and electron capture are written as:



for example:



The nuclide mass number (A) is preserved in beta-decay, only the proton number (Z) changes. The reaction is typical for light elements and neutron-rich fission products.

The decay energy is divided between the daughter nuclide, the electron / positron and the anti-neutrino (β^- decay) / neutrino (β^+ decay). The emission spectrum of beta-decay is continuous.

Radioactive decay: decay modes

In alpha-decay, the nuclide emits a He-4 nucleus:



for example:



Both the mass (A) and the proton number (Z) change in alpha decay. The reaction is typical for heavy elements (above lead). The decay energy is divided between the two products and the emission spectrum consists of discrete lines.

Other decay modes include:

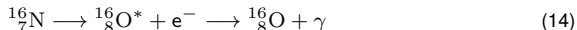
- ▶ Spontaneous fission, in which the nucleus is spontaneously split in two daughter nuclides and a number of neutrons.¹¹
- ▶ Proton and neutron emission, which competes with beta-decay in nuclides with large excess of protons/neutrons.

Even though these decay modes are seemingly rare, they are relatively common within the exotic nuclide composition of spent nuclear fuel, and must be taken into account in burnup calculations.

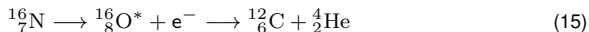
¹¹This decay mode becomes significant for higher actinides, especially some curium isotopes. Spontaneous fission forms a background neutron source in irradiated fuel.

Radioactive decay: decay modes

When the nucleon configuration changes, the nucleus is typically left in an excited state. The excess energy is usually released by gamma emission, e.g.:



This means that radioactive decay is usually accompanied by gamma radiation. If the nucleus is at a very high excited state, it can also undergo another radioactive decay almost immediately after the first one, e.g.:



The emission of delayed neutrons after fission is based on a similar process:



The decay of ${}^{87}\text{Kr}$ is practically instantaneous, but the half-life of the precursor ${}^{87}\text{Br}$ is 55.7 seconds.

It is also possible that the daughter nucleus is left at the ground state, in which case no gamma radiation is emitted (e.g. β^- decay of ${}^3\text{H}$ and ${}^{90}\text{Sr}$).

Radioactive decay: exponential decay law

As mentioned above, radioactive decay is a stochastic process and independent of external factors. The probability of a single nuclide decaying per unit time is characterized by the decay constant λ :

$$\frac{dP}{dt} = \lambda \quad (17)$$

The number of radioactive nuclides with decay constant λ in a sample follows an exponential law:¹²

$$N(t) = N_0 e^{-\lambda t} \quad (18)$$

where N_0 is the number of nuclides at $t = 0$. The activity of a sample of nuclides is defined as:

$$A = \lambda N \quad (19)$$

and it follows the same exponential decay law:

$$A(t) = A_0 e^{-\lambda t} \quad (20)$$

The exponential decay law leads to the definition of two time constants:

- ▶ The average life time of the nucleus: $T_{\text{ave}} = 1/\lambda$
- ▶ The half-life of the nucleus: $T_{1/2} = \ln(2)/\lambda$

¹²For the derivation of this law, see similar dependence between neutron free path length and macroscopic total cross section in Lecture 3.

Neutron-induced reactions

Nuclides can also be transmuted into other species by various nuclear reactions. The high flux level in operating nuclear reactors means that all non-neutron reactions can be ignored without inflicting significant errors in the results.

The probability of a single nuclide undergoing a neutron-induced reaction per unit time depends on the microscopic cross section σ :

$$\frac{dP}{dt} = \phi\sigma \quad (21)$$

where ϕ is the integral flux and σ is obtained as the flux-volume weighted average value of the corresponding continuous-energy cross section:¹³

$$\sigma = \frac{\int_V \int_E \phi(\mathbf{r}, E)\sigma(E)dV dE}{\int_V \int_E \phi(\mathbf{r}, E)dV dE} \quad (22)$$

using the flux solution from the transport calculation.

Since Eq. (21) is of similar form as Eq. (17) for radioactive decay, a similar exponential law can be derived for the number of nuclides in a sample subjected to neutron irradiation:

$$N(t) = N_0 e^{-\phi\sigma t} \quad (23)$$

¹³See similar energy condensation for macroscopic cross sections by preserving reaction rate balance in the derivation of the multi-group transport equation in Lecture 4.

Neutron-induced reactions: reaction types

Typical neutron-induced transmutation reactions include (n,γ) reaction (radiative capture):

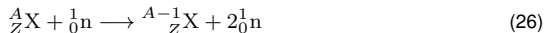


for example:

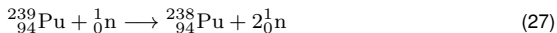


which is possible for all nuclides at all energies.¹⁴

Actinides typically also undergo $(n,2n)$ reactions:



for example:

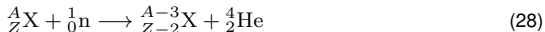


which is a threshold reaction ($E_{\min} \sim 5$ MeV). Similar neutron-multiplying transmutation reactions include $(n,3n)$ and $(n,4n)$, for which the threshold energies are much higher.

¹⁴Helium-4 has no measured (n,γ) cross section. The product nuclide ⁵He decays by neutron emission with a half-life of $\sim 10^{-24}$ s, which makes the capture reaction of ⁴He indistinguishable from inelastic scattering.

Neutron-induced reactions: reaction types

A typical transmutation reaction for light nuclides is (n,α) :



for example:



This reaction mode is a threshold reaction for some nuclides (${}^{16}\text{O}$), but possible at all energies for other (${}^{10}\text{B}$).

In practice there is a multitude of other reaction modes: (n,p) , (n,d) , (n,t) , $(n,{}^3\text{He})$, $(n,n\alpha)$, ... but the list above covers the most important reactions causing changes in the composition of irradiated fuel.

Fission is essentially a neutron-induced decay reaction, in which the target nucleus is split in two parts. The division is asymmetric, with two peaks centered at around $A\sim 100$ and $A\sim 140$ (see Fig. 10). The fission yield libraries in evaluated nuclear data files list hundreds or even thousands of fission product nuclides, and the distribution depends on the actinide isotope and neutron energy.

Due to the excess number of available neutrons, fission products are typically radioactive and decay by β^- reaction. Since the decay mode preserves nuclide mass, the decay path is limited to a single mass chain. Fission yields for individual isotopes can be given as independent or cumulative (cumulative sum over mass chain).

Bateman depletion equations

When a material consisting of several nuclides is subjected to neutron irradiation, the changes in the nuclide composition over time are characterized by the Bateman depletion equations:

$$\frac{dN_j}{dt} = \underbrace{\sum_{i \neq j} S_{i \rightarrow j}}_{(A)} - \underbrace{\lambda_j N_j}_{(B)} - \underbrace{\phi \sigma_j N_j}_{(C)} \quad (30)$$

where:

(A) is the production rate of nuclide j from nuclide i by decay, transmutation and fission

(B) is the decay rate of nuclide j

(C) is the total transmutation and fission rate of nuclide j ¹⁵

The source term can be written as the sum of decay, transmutation and fission terms:

$$S_{i \rightarrow j} = \lambda_{i \rightarrow j} N_i + \phi \sigma_{i \rightarrow j} N_i + \phi \gamma_{i \rightarrow j} \Sigma_{f,i} \quad (31)$$

where the decay constant $\lambda_{i \rightarrow j}$, transmutation cross section $\sigma_{i \rightarrow j}$ and fission yield $\gamma_{i \rightarrow j}$ couple the production rate of nuclide j to the corresponding loss terms of nuclide i .

Obtaining the neutron-induced source and loss terms requires resolving the neutron flux, which couples the depletion problem to the transport problem, as will be discussed later on.

¹⁵Cross section σ_j includes here all neutron reactions except elastic and inelastic (non-multiplying) scattering.

Bateman depletion equations

Some important results characterizing the behavior of nuclide concentrations can be obtained by assuming the coefficient terms in Eq. (30) as constants. In such case the concentration tends towards an asymptotic level, which can be calculated by setting the time derivative at infinity to zero:

$$\lim_{t \rightarrow \infty} \frac{dN_j(t)}{dt} = \lim_{t \rightarrow \infty} \left[\sum_{i \neq j} S_{i \rightarrow j}(t) - \lambda_j N_j(t) - \phi \sigma_j N_j(t) \right] = 0 \quad (32)$$

For example, for constant production and radioactive decay ($\phi \sigma_j \ll \lambda_j$):

$$S_j = \lambda_j \lim_{t \rightarrow \infty} N_j(t) \iff \lim_{t \rightarrow \infty} A_j(t) = S_j \quad (33)$$

which means that that the nuclide concentration is at equilibrium when activity is equal to the production rate.¹⁶ The saturation of activity follows equation:

$$A_j(t) = S_j \left(1 - e^{-\lambda_j t} \right) \quad (34)$$

For example, 95% of saturation concentration ($A_j(t) = 0.95 S_j$) is reached after time:

$$t = -\log(0.05)/\lambda \approx 3/\lambda \approx 4.3 T_{1/2} \quad (35)$$

Two simple examples of Bateman equations are given in the following.

¹⁶The saturation of activity explains, for example, why the inventory of short-lived ¹³¹I does not significantly depend on fuel burnup but that of ¹³⁷Cs does.

Bateman depletion equations: example 1

Example 1: Equilibrium ^{135}Xe

As discussed above, the production chain of fission product poison ^{135}Xe is written as:



By lumping the short-lived precursors in the fission yield of ^{135}I and assuming that the capture rate of this nuclide is negligible compared to radioactive decay, the Bateman equations can be written for iodine (I) and xenon (X) concentration as:

$$\begin{aligned} \frac{dI}{dt} &= \phi\gamma_I\Sigma_f - \lambda_I I \\ \frac{dX}{dt} &= \phi\gamma_X\Sigma_f + \lambda_I I - (\lambda_X + \phi\sigma_X) X \end{aligned} \quad (37)$$

where the fission cross sections and yields represent the material-wise average.

Bateman depletion equations: example 1

Example 1: Equilibrium ^{135}Xe

If it is assumed that the fission source terms remain constant for a sufficiently long time, setting the derivatives to zero gives for the equilibrium concentrations:

$$\begin{aligned}\lim_{t \rightarrow \infty} I(t) &= \frac{\phi \gamma_I \Sigma_f}{\lambda_I} \\ \lim_{t \rightarrow \infty} X(t) &= \frac{\phi \Sigma_f (\gamma_X + \gamma_I)}{\lambda_X + \phi \sigma_X}\end{aligned}\tag{38}$$

This is a very good approximation for the poison concentration when the reactor has operated at constant power for several days.

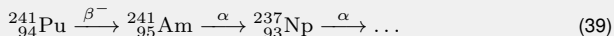
Reactor core simulator codes typically read the fission yields of ^{135}I and ^{135}Xe and the microscopic one-group cross section ^{135}Xe as part of the group constant input, in order to calculate the equilibrium concentration explicitly, and its effect in total absorption.

Similar equations can be derived for ^{149}Sm and its precursors.

Bateman depletion equations: example 2

Example 2: radioactive decay chain with multiple nuclides

The beginning of ^{241}Pu decay chain is written as:



where the half-lives of ^{241}Pu , ^{241}Am and ^{237}Np are 14.4, 432 and 2.1 million years, respectively. The depletion (in this case, decay) equations characterizing the system are:

$$\begin{aligned} \frac{dN_{41}}{dt} &= -\lambda_{41} N_{41} \\ \frac{dN_{51}}{dt} &= \lambda_{41} N_{41} - \lambda_{51} N_{51} \\ \frac{dN_{37}}{dt} &= \lambda_{51} N_{51} - \lambda_{37} N_{37} \end{aligned} \quad (40)$$

The source term of ^{241}Pu is assumed zero, and ^{241}Am and ^{237}Np are produced only in radioactive decay from the nuclide higher in the chain.

Bateman depletion equations: example 2

Example 2: radioactive decay chain with multiple nuclides

The solution for ^{241}Pu in Eq. (40) results:

$$N_{41}(t) = N_{41}^0 e^{-\lambda_{41}t} \quad (41)$$

The solution is substituted in the equation for ^{241}Am , and the resulting equation resolved into:

$$N_{51}(t) = N_{41}^0 \frac{\lambda_{41}}{\lambda_{41} - \lambda_{51}} (e^{-\lambda_{51}t} - e^{-\lambda_{41}t}) + N_{51}^0 e^{-\lambda_{51}t} \quad (42)$$

If it is assumed that ^{237}Np can be considered stable within the observed time frame (half-life 2.1 million years), the substitution of the previous solution results in an equation with solution:

$$N_{37}(t) = N_{41}^0 \left(1 + \frac{\lambda_{51}}{\lambda_{41} - \lambda_{51}} e^{-\lambda_{41}t} - \frac{\lambda_{41}}{\lambda_{41} - \lambda_{51}} e^{-\lambda_{51}t} \right) + N_{51}^0 (1 - e^{-\lambda_{51}t}) + N_{37}^0 \quad (43)$$

Already with three nuclides it is seen how the the complexity of the solution increases along with the decay chain length. Neutron-induced reactions increase the complexity by adding terms and branches in the chains.

Bateman depletion equations: example 2

Example 2: radioactive decay chain with multiple nuclides

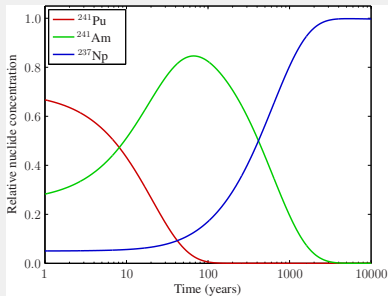


Figure 15 : Concentrations of ^{241}Pu , ^{241}Am and ^{237}Np as function of time. The initial concentrations are chosen arbitrarily.

Bateman depletion equations: systematic solution

When the Bateman depletion equations (30) are written for a complete system of nuclides, the result is an equal number of coupled first-order differential equations. In burnup calculations, the number can reach 1500 - 2000.

The transmutation and decay chains starting from nuclide j can be thought of as a complicated tree-structure, where the path is divided into multiple branches for every nuclide with multiple reaction modes. The branching considerably complicates the system of equations, and makes it impossible in practice to write an analytical solution to the problem in closed form.

Even though the problem is of a relatively simple form, its complexity and difficult numerical characteristics makes the solution a non-trivial task.

There are various solution methods to the problem, and two popular categories are introduced here:

- 1) Recursive analytical solution of linearized chains
- 2) Matrix exponential solutions

NOTE: *The prerequisite of both introduced methods is that all reaction rates remain constant over the time interval i.e. that the coupled problem is linearized.*

Linear chains method

The linear chains method is based on the decomposition of this tree structure into multiple independent chains:

- ▶ The structure is followed recursively, in such way that each branch starts a new chain.
- ▶ The result is a large number of linear chains, which can be solved analytically.
- ▶ The final result is obtained by summing over the solutions of all linear chains.

The concentration of k :th nuclide in a chain starting from nuclide 1 is obtained from equation:¹⁷

$$N_k(t) = \frac{N_1(0)}{\lambda_k} \sum_{i=1}^k \left[\lambda_i \prod_{j \neq i}^k \left(\frac{\lambda_j}{\lambda_j - \lambda_i} \right) \exp(-\lambda_i t) \right] \quad (44)$$

where λ :s are the generalized decay / transmutation constants The solution of the equation is not based on any approximations, but the practical implementation is subject to two problems:

- ▶ Transmutation chains starting from actinides can become extremely long, which translates into high computational cost
- ▶ Closed chains (e.g. consecutive (n,γ) and $(n,2n)$ reactions) form singularities in Eq. (44), as the same nuclide is included in the same chain multiple times.

In practice, the linear chains method requires a cut-off criterion for long chains.

¹⁷Note: the indexing is not fixed to the nuclide species, but the order of nuclides in the chain.

Matrix exponential methods

The Bateman depletion equations (30) form a system of linear differential equations, which can be written in matrix form:

$$\mathbf{n}' = \mathbf{A}\mathbf{n}, \quad \mathbf{n}(0) = \mathbf{n}_0 \quad (45)$$

where matrix \mathbf{A} contains the coefficients (loss terms on the diagonal, production terms off-diagonal) and vector \mathbf{n} contains the nuclide concentrations.

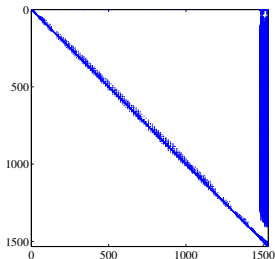


Figure 16: Non-zero elements in a coefficient matrix for a system of 1500 nuclides, indexed according to isotope mass. The vertical columns on the right are formed by the fission product distributions of actinides.

Matrix exponential methods

The solution of the Bateman depletion equations in matrix form can be written formally as:

$$\mathbf{n}(t) = e^{\mathbf{A}t} \mathbf{n}_0 \quad (46)$$

which reduces the problem to the calculation of the matrix exponential function.¹⁸ The problem is that the coefficient matrix \mathbf{A} has very difficult numerical characteristics:

- ▶ Size up to $\sim 1700 \times 1700$
- ▶ Norm: $\sim 10^{21}$
- ▶ Eigenvalues: $|\lambda| \in [0, 10^{21}]$
- ▶ Time steps: $t \sim 10^1 \dots 10^6$ s

which is why most solution methods resort to reducing the size of the matrix, for example, by handling short-lived nuclides separately.¹⁹

¹⁸Obviously, the matrix exponential function is not equivalent with taking the element-wise exponentials.

¹⁹A novel matrix exponential solution based on the Chebyshev Rational Approximation Method (CRAM) was developed at VTT for the Serpent Monte Carlo reactor physics burnup calculation code. The method takes advantage of the fact that the eigenvalues of the coefficient matrix are concentrated near the negative real axis, which allows using a very accurate rational approximation for the solution. The CRAM method can handle the full system of nuclides without approximations or practical limitations regarding time steps.

Non-linearity of the coupled depletion problem

In the formulation of the Bateman depletion equations it was assumed that neutron-induced (microscopic) reaction rates remain constant in time. A similar assumption is made for the (macroscopic) cross sections in the formulation of the neutron transport equation. In reality, neither is true, because:

- 1) Changes in flux spectrum are reflected in the spectrum-averaged one-group cross sections used for solving the depletion problem.
- 2) Changes in nuclide concentrations are reflected in the macroscopic cross sections used for solving the transport problem.

The result is that the coupling of two linear problems forms a non-linear system.²⁰

There exists methods for solving the coupled equations as a single problem, but in practice the usual approach is to divide the time interval into discrete depletion steps and apply operator splitting:

- ▶ Transport problem is solved assuming that reaction rates remain constant over the time interval. The calculation produces flux spectrum, which is used for calculating the microscopic transmutation cross sections for the depletion equations.
- ▶ Depletion problem is solved assuming that flux spectrum remains constant over the time interval. The calculation produces material compositions for the next transport solution.

²⁰ Similar coupling between two solvers is encountered when modeling thermal-hydraulic feedback, only the time-scale is considerably shorter.

Non-linearity of the coupled depletion problem

How well the approximation of linearity applies in reality depends on several factors, in particular the rate of change of nuclide reaction rates relative to the length of the time interval. The rate of change of reaction rates in turn depends on self-shielding and production and loss terms.

Selection of depletion step length is a compromise between accuracy and computational cost, and optimization becomes important especially with Monte Carlo based depletion schemes, where the transport solution can be expensive to obtain.

The selection of step length should reflect at least the following physical factors:

- ▶ Reactor poison ^{135}Xe saturates within 48 hours of constant power operation. During this period, flux and reactivity are in continuous change, which calls for shorter steps at the beginning of the irradiation cycle. The same applies to any changes in reactor power level.
- ▶ Burnable absorber (gadolinium) is usually depleted by 10-15 MWd/kgU burnup. Shorter steps should be used until the absorber is completely gone.
- ▶ Reactivity changes are caused by the depletion of the primary fissile isotope (^{235}U) and burnable absorbers, together with the build-up of plutonium and fission products. These changes do not reflect the accumulation of weak absorbers, such as ^{131}I or ^{137}Cs , which may be important for some other reason (source terms for accident analysis, etc.).

In addition to time discretization, also spatial discretization causes error in the results by assuming uniform reaction rates over large material regions. To reduce the error, burnable materials are usually sub-divided into multiple depletion zones. This is particularly important for burnable absorber pins, which are subject to strong spatial self-shielding. (see Lecture5_anim1.gif)

Depletion algorithms

In practice, all depletion algorithms rely on time-step discretization and either explicit or implicit iteration:

- ▶ Explicit methods are based on sequential calls to transport and depletion solvers while proceeding to new steps
- ▶ Implicit methods perform inner iterations to converge the two solutions before moving to the next step

Explicit methods are computationally less expensive, but subject to errors and numerical instabilities when the depletion step is chosen too long. Since assembly burnup calculations are traditionally carried out in 2D geometries, explicit methods are usually considered sufficient for their solution.²¹

The simplest depletion algorithm is the explicit Euler method, in which the beginning-of-step (BOS) flux spectrum and reaction rates are used directly for solving the depletion equations.

The changes in the neutron-induced reaction rates within the step can be accounted for to some extent by using constant values that represent the average behavior, rather than the BOS state. These values are calculated by assuming some functional behavior for the cross sections, typically linear dependence between two points. This is the basis of predictor-corrector methods.

²¹This is no longer the case when the calculations are expanded to 3D.

Depletion algorithms

It is important to realize that, even though flux and cross sections can be assumed to exhibit a continuous behavior over each burnup interval $[t_i, t_{i+1}]$, the formulation of the depletion equations necessitates the use of constant values.

The solution is therefore subject to two approximations:

- 1) The behavior is approximated by a continuous analytical function $f(t)$, which is formed using discrete values calculated for one or several burnup steps.
- 2) The continuous behavior is further approximated by a constant value, defined by averaging the continuous function over the length of the depletion step.

$$\frac{1}{t_{i+1} - t_i} \int_{t_i}^{t_{i+1}} f(t) dt \quad (47)$$

The Euler method and conventional predictor-corrector calculation with linear interpolation are demonstrated in the following by an example.

Depletion algorithms: example

Example 3: burnup calculation with Euler method and predictor corrector

The test case used for demonstration is a simplified PWR fuel assembly model with a single burnable absorber pin surrounded by 15 UO₂ pins. The uranium pins are treated as a single burnable material, while the absorber is divided into 10 annular regions with equal volume.

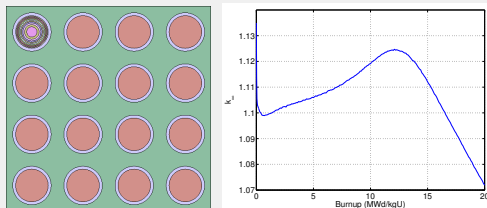


Figure 17 : Left: Geometry model of the test case. Right: k_{∞} as function of burnup.

The following slides show the depletion of Gd-155 in the outermost ring of the burnable absorber pin, using two different burnup algorithms. To emphasize the differences, the step length is set to 2 MWd/kgU, which would normally be considered too long for this type of calculation.

Depletion algorithms: example

Example 3: burnup calculation with Euler method and predictor corrector

The Euler method is the simplest depletion algorithm, that assumes that the flux and cross sections remain at their beginning-of-step (BOS) values throughout the step.

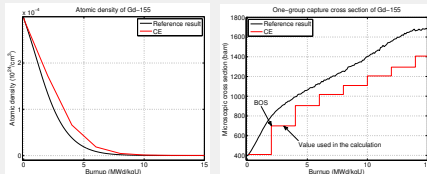


Figure 18 : Depletion with Euler method. Left: Atomic density of Gd-155 as function of burnup. Right: one-group capture cross section of Gd-155 as function of burnup.

If the cross section is an increasing function of burnup, which is often the case with burnable absorbers due to reduced self-shielding, the BOS value under-estimates the average cross section over the step. The result is that the depletion rate is also under-estimated, which, in turn, results in the over-prediction of the nuclide concentration at the end. The same problem occurs in the next step, and the algorithm simply cannot keep up with the actual depletion rate. The error can be reduced by shortening the step length, but this is often not the optimal solution to the problem.

Depletion algorithms: example

Example 3: burnup calculation with Euler method and predictor corrector

The simplest predictor-corrector method applies two transport calculations per depletion step, in order to more accurately represent the continuously changing reaction rate over the interval.

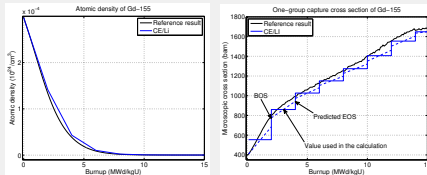


Figure 19 : Depletion with predictor-corrector method. Left: Atomic density of Gd-155 as function of burnup. Right: one-group capture cross section of Gd-155 as function of burnup.

The solution is divided in two parts: 1) Predictor calculation – reaction rates are calculated for the BOS composition. 2) Corrector calculation – the material is depleted over the interval, and new reaction rates calculated for the end-of-step (EOS) composition. The final burnup calculation is carried out from BOS to EOS, using the average of the predictor and corrector reaction rates, which corresponds to linear interpolation between the two points. The method usually results in improved accuracy, but since two transport solutions are required per step, also the running time is increased. In most cases the method is still superior to Euler with step length reduced in half.

Summary of main topics

Nuclear fuel is irradiated in the reactor core for an extended period of time, during which the physical characteristics change due to depletion of fissile uranium and build-up of plutonium and fission products. These changes cause various effects in reactivity and reactor safety parameters, which must be taken into account in reactor operation.

The physical changes are caused by radioactive decay and neutron-induced transmutation and fission reactions. The evolution of isotopic composition is characterized by the Bateman depletion equations – a system of coupled differential equations. The solution of depletion equations is a non-trivial problem due to the difficult numerical characteristics of the system, and the solution is typically based on the linear chains method or calculation of the matrix exponential.

The depletion problem is coupled to the transport problem by the changes in nuclide compositions and neutron flux. The coupled problem is non-linear, and the solution typically relies on operator splitting and consecutive solution of two linear problems. This introduces errors in the solution, which depend on the depletion algorithm and the step length used in the iteration.

Topics of next lecture

The topic of next lecture (29.3.2018) is heat transfer and coolant flow in nuclear reactors, and it is given by Ville Valtavirta from VTT.