2) Physics of breeding

Learning outcomes:

After working through this chapter, you will be able to:

1) evaluate the capacity of a nuclear reactor to breed fissile nuclear fuel from fertile nuclides;
2) assess the impact of power density, spectrum, fuel composition and choice of coolant on breeding;
3) develop unconventional approaches to breeding.

Background

The first condition for being defined as a Generation-IV reactor is that it should be sustainable and utilize its fuel effectively. Albeit not explicit in the above formulation, this should be interpreted as that the reactor should operate with a breeding ratio larger than unity, that its fuel should go to a reasonably high burn-up and that the spent fuel should be recycled. The requirements related to economic performance means that all of this should be possible while producing power at a competitive cost.

The long term operation of any fleet of light water reactors is limited by the availability of ^{235}U, which constitutes only 0.72% of uranium in nature. At some point, the cost of uranium ore may rise towards a point where competing means of producing power would make that fleet un-economic.

Already in 1944, Enrico Fermi pointed out the possibility of breeding fissile nuclear fuel from fertile ^{238}U [Westfall 2004], according to the reaction

$$n + {}^{238}U \rightarrow {}^{238}U + \gamma \rightarrow {}^{239}Pu + 2\beta^-$$

The neutrons would be provided by fission of a fissile nuclide, such as ^{235}U or ^{239}Pu. In the case that the production of ^{239}Pu through the above reaction is larger than the destruction of ^{235}U and/or ^{239}Pu by fission, it is common practice to state that the reactor has a conversion, or breeding ratio larger than 1.0. By recycling of the plutonium produced in the breeding process, a self-sustaining power system can be implemented, where the only material supplied to the fuel cycle is ^{238}U. This would extend currently reasonably assured uranium fuel resources from about a century [NEA 2018] to beyond 10 000 years.

Yet an even larger energy resource is available through the breeding of fissile ^{233}U from ^{232}Th [Chernick 1959]:

$$n + {}^{232}Th \rightarrow {}^{233}Th + \gamma \rightarrow {}^{233}U + 2\beta^-$$

The above processes are know as the uranium–plutonium and the thorium-uranium cycles. As we will see, the breeding potential and efficiency of fuel utilization of these cycles depend on neutron spectrum and power density. Moreover, the fuel composition selected will have an impact on the same, as well as on whether irradiated fuel can be recycled or not.

Breeding ratio

In order to assess the potential for breeding of a nuclear fuel, one introduces the η-value, which is the average number of neutrons produced in after absorption of a neutron in an actinide nucleus:

$$\eta = \nu \frac{\sigma_f}{\sigma_a} \simeq \nu \frac{\sigma_f}{\sigma_f + \sigma_c}$$

Where ν is the number of neutrons released in the fission event, σ_f is the spectrum averaged fission cross section, σ_a the cross section for absorption, and σ_c the cross section for capture through the (n,γ) reaction. For criticality to be possible ($k_{eff} = 1.0$), the η-value has to be larger than unity.

Now, let us define more precisely in words what we mean by a nuclear fuel having a breeding ratio larger than unity. Namely, we wish to ensure that it is possible to reload a nuclear reactor with actinides recovered from its own used fuel, topping up exclusively with a fertile nuclide. In terms of reactivity ρ of the fuel, this may be expressed as [Wallenius 2020]:

> *The sum of reactivity changes during burn-up and subsequent cooling, reprocessing and re-fabrication of the fuel should be larger than zero.*

Historically, this condition has been expressed in several different, less consistent ways. E.g. it is often stated that a reactor would have a breeding ratio larger than unity if the mass of fissile material in its fuel is larger at end-of-life than at beginning-of-life. However, as we will see, this condition is neither sufficient nor necessary for the ability to reload a reactor with its own spent fuel and a fertile top-up. Namely, since the average η-value of different fuels varies significantly with composition, neutron spectrum, power density and cooling time, we need to undertake a more sophisticated approach to characterizing the breeding potential of a nuclear fuel.

To this end, we define the reactivity based *instantaneous in-pile conversion ratio* as [Wallenius 2020]:

$$CR_{ip} = \frac{\sum_{A,m} \sigma_c(^m A) C(^m A) \eta(^{m+1} A')}{\sum_{A,m} \sigma_f(^m A) C(^m A) \eta(^m A)},$$

where the sums are taken over all actinides A with mass m, and A' is the actinide formed after capture of a neutron in nuclide ^{m}A. This formula is a measure of the reactivity produced in neutron captures divided by the reactivity destroyed in fission events.

I order to appreciate the utility of the above expression, we plot the η-value for ^{233}U and ^{239}Pu for the entire range of neutron energies that may be found in a nuclear reactor.

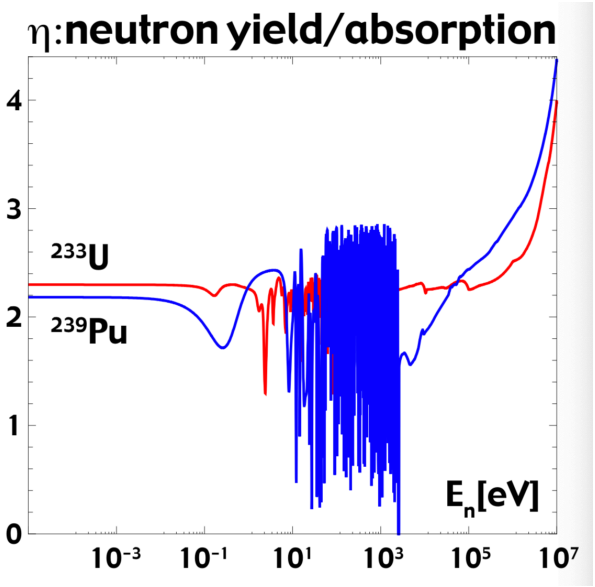

Figure 2.1: η-value for ^{233}U and ^{239}Pu

If we now assume that the fission cross section for all fertile nuclides is zero (which is approximately true in a thermal spectrum), we find for the binary systems discussed within the simplified context of the Th-U and U-Pu cycles that the in-pile conversion ratio at beginning-of-life would be equal to

$$CR_{ip} = \frac{\sigma_c(\text{fertile}) C(\text{fertile})}{\sigma_f(\text{fissile}) C(\text{fissile})}$$

Let us now study the case of a heavy water moderated CANDU reactor. If we fuel such a reactor with (^{232}Th,^{233}U)O_2 fuel, the cross section for neutron capture in ^{232}Th is 1.84 barn, and that for fission of ^{233}U is 118 barn. Solving for the conversion ratio to be larger to unity, we find that the fraction of the fissile isotope must be lower than 1.53 %. Figure 2.2 displays the reactivity evolution in an infinite fuel bundle lattice model of a CANDU reactor loaded with ($^{232}Th_{0.985}$,$^{233}U_{0.015}$)O_2 fuel, during an initial irradiation period of 200 days, followed by 365 days of cooling and finally removal of fission products.

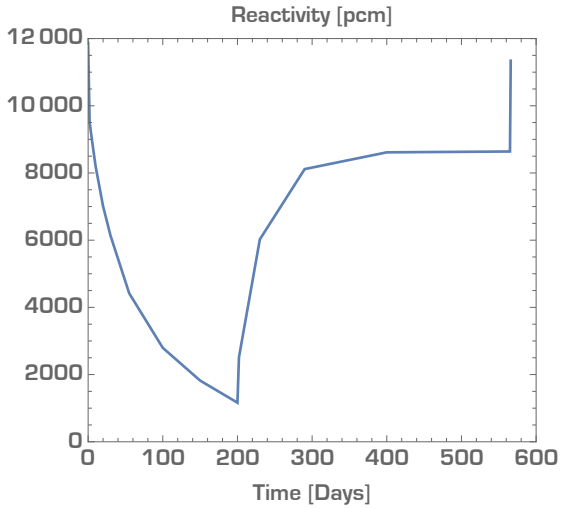

Figure 2.2: Reactivity evolution in a ($^{232}Th_{0.985}$,$^{233}U_{0.015}$)O_2 fuel bundle lattice of a CANDU reactor.

It is to be noted that in spite of having a conversion ratio of 1.03, the derivative of the reactivity with respect to burn-up is negative, i.e. we have a so called "negative reactivity swing". This phenomenon is due to fission product poisoning. Moreover, the second derivative of the reactivity evolution is positive. The reason is that the non-fissile isotope ^{233}Pa, which is formed by beta-decay of ^{233}Th, has a half-life of 27 days, prior to decaying into fissile ^{233}U. Hence, the build-up of reactivity is delayed. Indeed, after end-of-life, the reactivity of the fuel continues to increase by as much as 7000 pcm through the decay of ^{233}Pa. Finally, removal of the fission products increases reactivity to its initial value. This means that we may reload the reactor using its own spent fuel, and continue the fuel cycle as long as ^{232}Th is available for top-up. We may also note that in this case, the mass of ^{233}U obtained after reprocessing is equal to that in initial core load, i.e. the classical definition of breeding ratio may be applied.

A drawback of conducting breeding of nuclear fuel in thermal spectrum is the poorer neutron economy associated with such reactors. Fission product poisoning limits the burn-up of the fuel in the thorium-uranium cycle to less than 0.7%. The uranium-plutonium cycle cannot even be implemented in CANDU reactors with oxide fuel, since any composition having a conversion ratio larger than unity has a ^{239}Pu concentration of less than 0.3%, which renders the configuration sub-critical. Fast neutron reactors on the other hand, offer a much higher flexibility.

Neutron spectrum

In a fast neutron spectrum, fertile isotopes have a significant probability for undergoing fission. Moreover, the η-value of some fissile isotopes are slightly higher than in a thermal spectrum and fission product poisoning is less severe. Hence there are variety of fuel compositions that may be used for breeding. However, for a detailed analysis of the breeding potential of such fuels, our reactivity based definition of the in-pile conversion ratio must be used in its full form.

Let us first study the uranium-plutonium cycle. We take the case of a sodium-cooled hexagonal fuel rod lattice with (^{238}U-^{239}Pu)O_2 fuel. Adopting a rod outer diameter of 12 mm and a rod pitch of 14 mm, we find spectrum averaged cross sections for fission and capture of ^{238}U, ^{239}Pu and ^{240}Pu as listed in Table 2.1. Note in particular that the probability for fission of ^{240}Pu is 45%, and that the corresponding η-value is 1.38. Hence, a neutron capture in ^{239}Pu destroys reactivity to a much lesser extent than in a thermal spectrum.

Table 2.1: Spectrum averaged cross sections, fission probabilities and eta values for nuclides in the U-Pu cycle. The calculated values are for a sodium cooled rod lattice with oxide fuel.

Nuclide	σ_c [b]	σ_f [b]	$\sigma_f/(\sigma_f+\sigma_c)$	η
^{238}U	0.25	0.04	0.14	0.38
^{239}Pu	0.40	1.68	0.81	2.37
^{240}Pu	0.43	0.35	0.45	1.38

Inserting the values from Table 2.1 into the reactivity based formula for the in-pile conversion ratio we find that the fraction of ^{239}Pu in the fresh oxide fuel must be less than 14.3% to ensure $CR_{ip} > 1.0$. For lower fractions, the in-pile conversion ratio will be larger than unity.

Figure 2.3 displays the reactivity evolution in an infinite sodium-cooled rod lattice loaded with $(^{238}U_{0.895},^{239}Pu_{0.105})O_2$ fuel, operating at a linear power of 30 kW/m up to a burn-up of 100 GWd/ton. As can be noticed, the reactivity constraints to the achievable burn-up are much more relaxed in a fast spectrum breeder. Irradiation is followed by a cooling time of four years, after which fission products and minor actinides are removed, and ^{238}U is added to retain the initial density of actinides in the fuel.

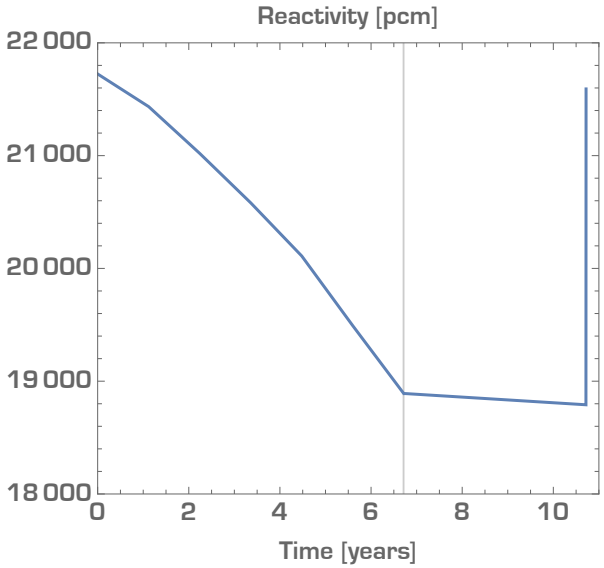

Figure 2.3: Reactivity evolution in a $(^{238}U_{0.895},^{239}Pu_{0.105})O_2$ rod lattice of a sodium-cooled reactor.

Here, we can see that the short half-life of the initial breeding product ^{239}Np (2.2 days) results in a reactivity evolution with a negative second derivative. Moreover, there is no significant reactivity increase during cooling. Rather, once the inventory of ^{239}Np has disappeared, the decay of ^{241}Pu into ^{241}Am, leads to a reduction in reactivity. We also note that, relative to the burn-up of the fuel, the reactivity change resulting from reprocessing is smaller than in a thermal spectrum, since fission product poisoning is of a smaller magnitude.

We may now have a look at the mass balance of the initial fissile nuclide. At end of irradiation, the concentration of ^{239}Pu is 95.5% of the initial value. According to the classical definition, this would correspond to a breeding ratio lower than unity. As we have seen, one must take into account the contributions to reactivity from all actinides destroyed and produced to properly characterize the breeding process.

The Th-U cycle may also be applied in a fast spectrum. Figure 2.4 illustrates the reactivity evolution for a $(^{232}Th_{0.908}$-$^{233}U_{0.092})O_2$ fuel irradiated to a burn-up of 100 GWd/ton at a linear rating of 30 kW/m. The ratio of cross sections and reactivity change during cooling here is such that a lower fraction of fissile is required to obtain identical reactivities at beginning-of-life and after reprocessing. Therefore, the initial reactivity margin is about 10 000 pcm smaller for this fuel than for the U-Pu cycle. This translates in lower flexibility in terms of core design, since only 10% of neutron leakage is permitted to maintain criticality for a Th-U breeder with oxide fuel reaching a burn-up of 100 GWd/ton.

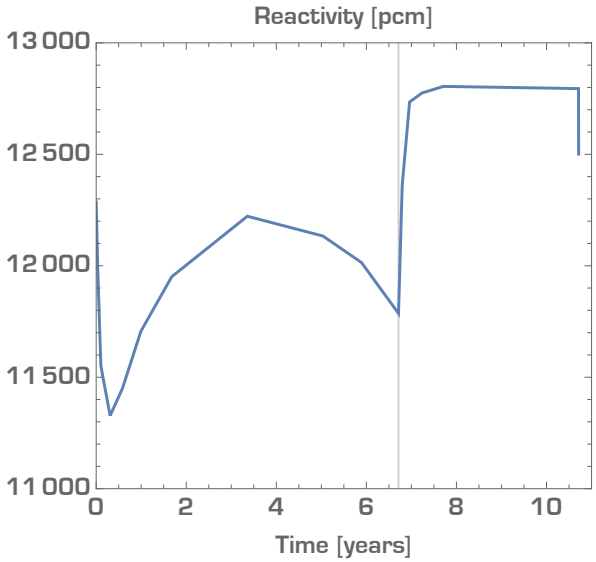

Figure 2.4: Reactivity evolution in a $(^{232}Th_{0.908},^{233}U_{0.092})O_2$ rod lattice of a sodium-cooled reactor.

We may also note the minimum in reactivity due to the delay in ^{233}U production from ^{233}Pa, as well as the negative reactive change resulting from reprocessing of the fuel. The latter is a result of ^{232}Th having a higher spectrum averaged cross section for capture than the fission product inventory after cooling.

In summary, we can conclude that the best opportunities for breeding of nuclear fuel is provided by a U-Pu fuel operating in a fast neutron spectrum. This option allows for a higher burn-up and a more flexible core design.

Equilibrium fuel cycle

In reality, there will never be streams of pure ^{233}U or ^{239}Pu for the initial feed of a breeder reactor. In practice, one would have to commence the breeding cycle using either reactor plutonium or enriched uranium. By repeated recycle of the spent fuel, one would in theory approach a so called equilibrium fuel composition, where the isotopic vectors of uranium and plutonium are identical at beginning-of-life, and after reprocessing and fertile top-up. This vector will depend on neutron spectrum, power density and time allowed for cooling prior to reprocessing. Since cycle averaged power density and cooling time in a real life industrial fuel cycle will not remain constant, the isotopic composition of the fuel will change over time, making the concept of equilibrium a hypothetical construction, and in best case an approximation of the actual composition.

Power density

To illustrate the impact of power density and cooling time, let us study the case of starting up a U-Pu oxide core with a plutonium vector derived from reprocessing of spent LWR fuel. This vector is listed in Table 2.2 and corresponds to the Pu composition of PWR fuel after 50 GWd/ton burn-up and 4 years of cooling.

Table 2.2: Pu vector of spent PWR fuel, after 50GWd/t burn-up and four years of cooling [NEA 1999].

Nuclide	Fraction
^{238}Pu	0,035
^{239}Pu	0,519
^{240}Pu	0,238
^{241}Pu	0,129
^{242}Pu	0,079

Let us now simulate the reactivity evolution in a sodium cooled rod lattice with (^{238}U,Pu)O_2 fuel having the Pu composition listed in Table 2.2. At a linear power of 30 kW/m this results in equal reactivity at beginning-of-life and after reprocessing when adopting a Pu fraction of 12.5%. The evolution of reactivity during irradiation is illustrated in Figure 2.5, compared with the cases of irradiation at linear ratings of 20 kW/m and 10 kW/m, respectively. In order to facilitate the comparison, the reactivity is plotted as function of burn-up, rather than of time.

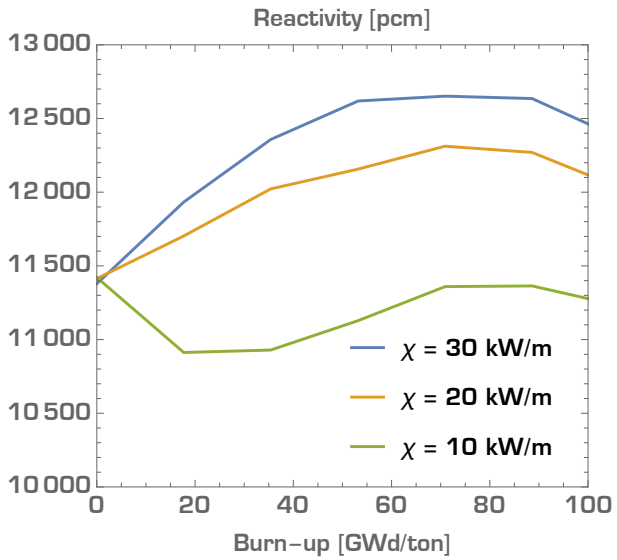

Figure 2.5: In-pile reactivity evolution of a ($^{238}U_{0.875}$,$Pu_{0.125}$)O_2 rod lattice cooled with sodium.

We note that the lower the linear rating, the larger reactivity loss in-pile occurs due to decay of ^{241}Pu. Hence, a fuel composition that provides a reactivity based conversion ratio of 1.0 for a higher power density features a conversion ratio much less than 1.0 for a lower. This introduces a considerable uncertainty when planning for fuel fabrication in a closed fuel cycle, since the cycle averaged power density depends on the frequency and duration of outages. A similar uncertainty is related to the actual cooling time applied between end-of-life, reprocessing, fuel fabrication and start of irradiation.

Fuel composition

Replacing the conventional oxide fuel with a so called high density fuel, one may improve the breeding capability of a given reactor configuration. The high density chemical compositions that have been applied as driver fuels in fast reactors are carbides, nitrides and metallic alloys. Such fuel have a higher density of actinides, allowing to increase the fraction of ^{232}Th or ^{238}U in the fuel, which raises the conversion ratio. This effect is partially counteracted by the hardening of the neutron spectrum arising from removing light atoms

from the fuel, which yields a larger relative reduction in capture cross section than in fission cross sections. Hence, for identical isotopic compositions, the use of high density fuels actually reduces the conversion ratio, as compared with the oxide reference. This fact has not always been well understood in the literature.

Table 2.3 compares the cross sections for capture in ^{238}U and fission in ^{239}Pu in high density fuels with those pertaining to oxides. The calculation was made for a sodium cooled rod lattice.

Table 2.3: High density fuel cross sections for capture in ^{238}U and fission in ^{239}Pu.

Fuel	Oxide	Carbide	Nitride	Metal alloy
σ_c [^{238}U]	0.29	0.26	0.25	0.20
σ_f [^{239}Pu]	1.80	1.73	1.68	1.61
σ_c [^{238}U]/σ_f [^{239}Pu]	0.16	0.15	0.15	0.12

Thus, from the purely spectral aspect, metal alloy fuel is expected to feature the lowest conversion ratio. Carbide and nitride fuels are expected to have a similar conversion ratio. This is reflected in the reactivity evolution of U-Pu fuels with 12.5% Pu fraction, as shown in figure 2.6. (Note that we need to plot reactivity as function of time when comparing fuels with different density, since the mass of actinides in a fuel rod is different.).

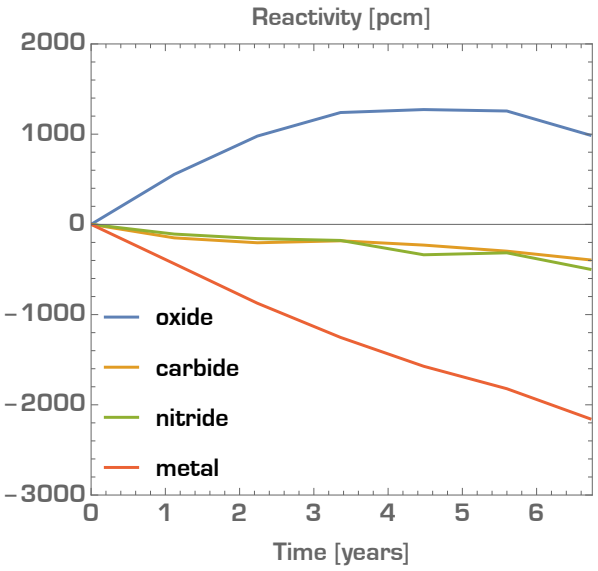

Figure 2.6: Reactivity evolution in ^{238}U-Pu fuels with a U:Pu ratio equal to 7/1. All reactivities are normalized to zero at beginning-of-life.

However, since the fission probability is highest for the metal alloy fuel, it also features the highest reactivity. Hence, we have a larger reactivity margin available for increasing the ^{238}U fraction in this fuel, the net effect of which is to maximize the conversion ratio.

Now, let us increase the fraction of ^{238}U in the high density fuels to obtain the same BoL reactivity as for the oxide fuel. This results in a Pu fraction of 10.9% for the carbide fuel, 10.6% for the nitride fuel and 9.2% for the metal alloy fuel. The corresponding evolution of reactivity is depicted in Figure 2.7.

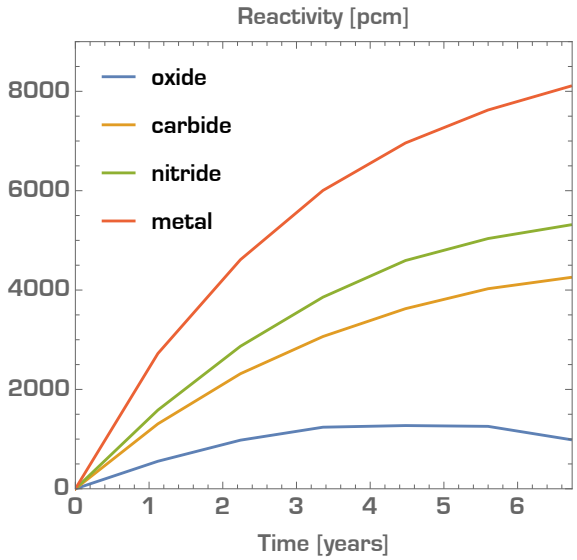

Figure 2.7: Reactivity evolution in U-Pu fuels with identical reactivity at BoL.

The metal alloy fuel now yields the highest conversion ratio. If one is interested in maximizing the breeding of nuclear fuel, this is the choice one would make. As we will discuss later in this text-book, this has implications on the reprocessing technology that can be applied.

Coolants

The choice of coolant might affect the neutron spectrum of a fast neutron reactor. Table 2.4 compares the cross sections for capture in ^{238}U and fission in ^{239}Pu for an oxide fuel rod lattice cooled with sodium, lead and helium at a density of 100 bar. The data are provided for the same rod pitch over diameter ratio (P/D = 1.17).

Table 2.3: High density fuel cross sections for capture in ^{238}U and fission in ^{239}Pu.

Fuel	Sodium	Lead	Helium
σ_c [^{238}U]	0.29	0.29	0.28
σ_f [^{239}Pu]	1.80	1.73	1.73
σ_c [^{238}U]/σ_f [^{239}Pu]	0.16	0.17	0.16

Possibly a bit contra-intuitive, the use of lead-coolant results in the highest ratio between neutron capture in ^{238}U and fission in ^{239}Pu. Hence the highest conversion ratio is expected to pertain to lead coolant. This turns out to be a result of in-elastic scattering of neutrons on lead nuclei at neutron energies between 0.5-1 MeV, which reduces the spectrum averaged cross section for fission. It is interesting to note that the helium coolant option incidentally yields similar cross sections and conversion ratios as in the case of sodium. This is reflected in the in-pile evolution of reactivity as illustrated in Figure 2.8.

In Figure 2.8, the reactivity evolution was simulated for identical rod pitch to diameter ratios. As we will discuss in Chapter seven, a lead-cooled reactor must be designed with a larger coolant volume fraction in the core, in order to remove the same amount of heat. As a result, the neutron spectrum in the lead-cooled core will soften further.

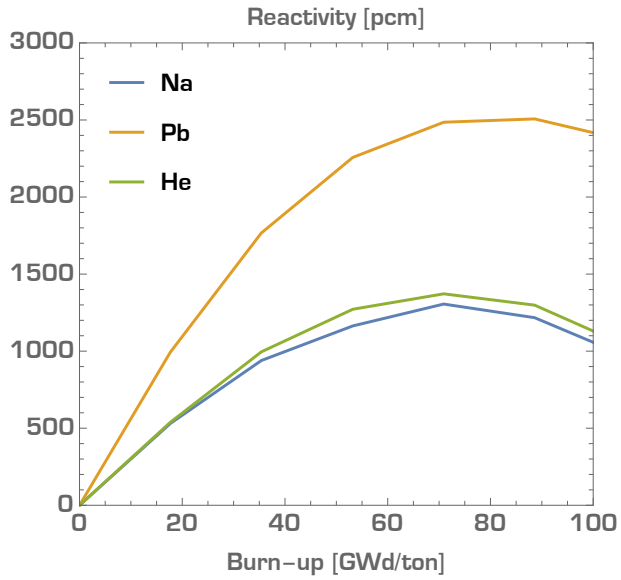

Figure 2.8: Reactivity evolution in ($^{238}U_{0.875}$,$Pu_{0.125}$)O_2 fuel rod lattices with sodium, lead and helium coolant.

In order to retain BoL reactivity, the ^{238}U:Pu ratio must be reduced, since a lead-cooled reactor with a large rod pitch features a much lower reactivity margin. This leads to a strong reduction in conversation ratio. Figure. 2.9 displays the reactivity evolution for (^{238}U,Pu)O_2 fuel rod lattices cooled by liquid lead with P/D = 1.17 and P/D = 1.50, where ^{238}U:Pu ratio has been adjusted to yield identical reactivities at BoL.

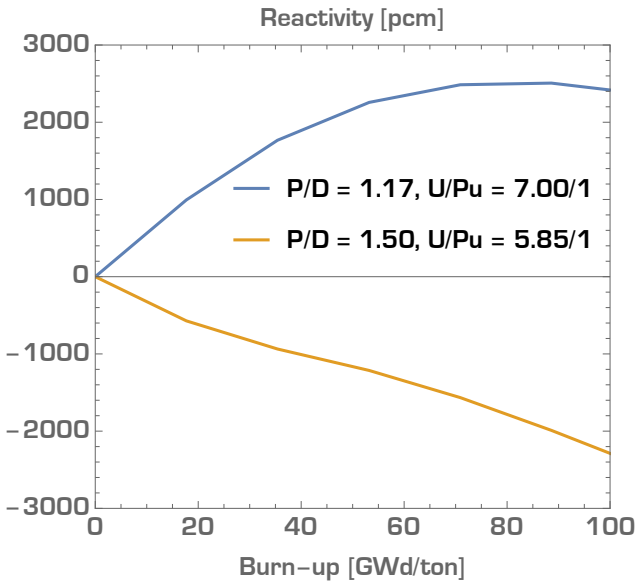

Figure 2.9: Reactivity evolution in (^{238}U,Pu)O_2 lead cooled fuel rod lattices having P/D = 1.17 and P/D = 1.50.

Consequently, in order to breed with a conversion ratio larger than unity using lead coolant, one needs to either operate an oxide fuel at a low power density, permitting to design the core with P/D ≈ 1.2, or one must apply a high density fuel solution. Concerning the latter option, metallic fuels have a propensity for dissolving into liquid lead, and are therefore not in general suggested as suitable for this environment. Nitride fuels, on the other hand, are compatible with lead and are currently being applied as the fuel for the lead-cooled BREST reactor concept, which is under construction in Russia. Figure 2.10 displays the reactivity evolution in a lead-cooled rod lattice with P/D = 1.5, using a (^{238}U,Pu)^{15}N fuel.

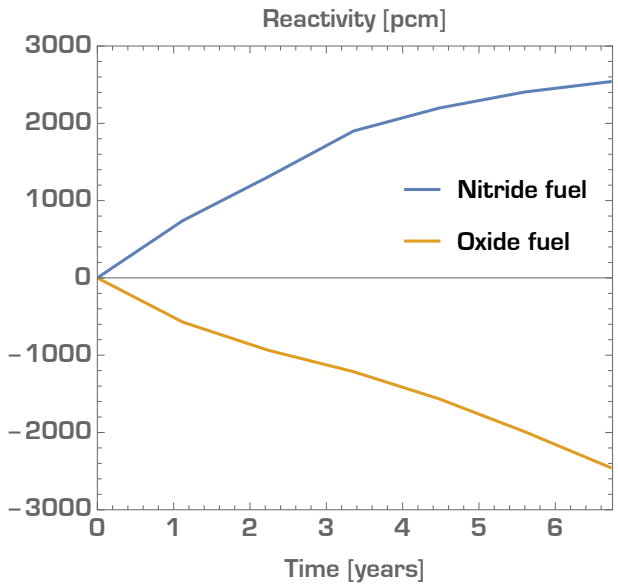

Figure 2.10: In-pile reactivity evolution in lead cooled nitride and oxide fuel rod lattices with P/D = 1.5. BoL reactivities are identical, as the nitride fuel features 12.1% Pu and the oxide 14.6%.

The reactivity loss during four years of cooling is about 1000 pcm. Thus, a conversion ratio larger than unity can be obtained in a lead-cooled reactor using nitride fuel, though the achievable breeding ratio is smaller than that in sodium-cooled reactors with dense fuels.

Unconventional breeding cycles

In nature, the only two fertile isotopes that can be used for breeding of nuclear fuel are ^{232}Th and ^{238}U. However, the use of nuclear power has produced other, relatively long lived even neutron number nuclides that could be separated from spent fuel for purposes of breeding. In particular, the minor actinides Np and Am are examples of such. Out of these, Np is considerably easier to separate and manage in a fuel fabrication facility. About 200 tons of neptunium has been produced in commercial nuclear reactors up to date, which is enough to fuel a number of power reactors. What is particularly interesting is that in difference to ^{232}Th and ^{238}U, ^{237}Np can be used by itself as a fuel in a reactor. Namely, the η-value for pure neptunium fuels is larger than unity in a fast spectrum [Wallenius 2019]. Table 2.4 displays the cross sections for capture and fission of ^{237}Np and its breeding product ^{238}Pu in a sodium cooled rod lattice (P/D = 1.17) with NpO_2 fuel.

Table 2.4: Spectrum averaged cross sections, fission probabilities and eta values for nuclides in the Np-Pu cycle. The calculated values are for a sodium cooled rod lattice with oxide fuel.

Nuclide	σ_c [b]	σ_f [b]	$\sigma_f/[\sigma_f+\sigma_c]$	η
^{237}Np	0.80	0.57	0.42	1.21
^{238}Pu	0.27	1.32	0.83	2.55

Figure 2.11 displayed the corresponding reactivity evolution. From this figure we can infer that the conversion ratio of the Np-^{238}Pu cycle is much larger than that of the ^{238}U-Pu cycle under similar conditions. One particularly puzzling aspect of this phenomenon is that the η-value or ^{237}Np, albeit being larger than unity, is much less than 2.0! According to conventional theories of breeding, the η-value of the fissioning nuclide should be larger than 2.0, in order for one of the produced neutrons to maintain criticality, and the

other to breed a new fissile nuclide to replace the one that was fissioned [Waltar 1981]. Yet, the conversion ratio of Np is clearly larger than unity! How can we explain this apparent paradox?

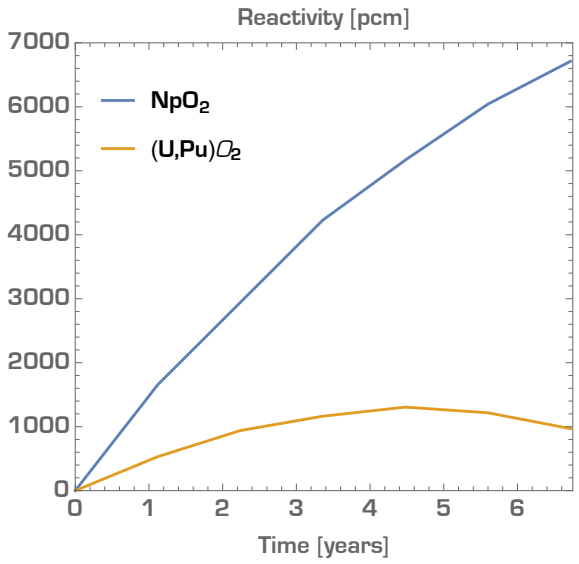

Figure 2.11: In-pile reactivity evolution in sodium cooled oxide fuel rod lattices with P/D = 1.17.

The key to understanding this puzzle is that neutrons that undergo capture in fissile component (^{237}Np) actually are the same as are captured in the fertile component (^{237}Np), and they are all breeding a new fertile/fissile nuclide (^{238}Pu) that has an η-value larger than the initial nuclide. Therefore, measuring only the η-value of the fissile component, incorrectly discounts for neutrons captured in the same. As a matter of fact, the conditions for breeding to be possible are $\eta > 1.0$, $\nu > 2.0$ and that *the sum of all reactivity changes during burn-up, cooling and reprocessing should be larger than zero*. In the case of the neptunium fueled reactor above, we have for the in-pile conversion ratio at BoL:

$$CR_{ip}(BoL) = \frac{\sigma_c(^{237}Np)\eta(^{238}Pu)}{\sigma_f(^{237}Np)\eta(^{237}Np)} = 2.96.$$

This is a remarkably large value.

Questions:

- What are the limitations for breeding of fissile fuel in a thermal spectrum reactor?
- Explain why a reactivity based criterion is required to assess the breeding potential in a fast spectrum.
- What determines the breeding efficiency of different fuel compositions?

Exercises:

- Calculate spectrum averaged cross sections for capture and fission in thermal and fast spectra using a Monte Carlo code, such as Serpent. Obtain the corresponding in-pile conversion ratios.
- Set up the Bateman equations for breeding of ^{239}Pu from ^{238}U and calculate the equilibrium composition assuming a sufficiently long irradiation time.
- Using a Monte-Carlo code, calculate the in-pile conversion ratio at BoL for reactors fueled with pure ^{234}U, ^{238}Pu and ^{241}Am. Discuss how pure inventories of such nuclides can be obtained without isotopic enrichment.

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