4) Minor actinide burning

Learning outcomes:

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

1) evaluate the contribution to radio-toxic inventory and heat emission of spent nuclear fuel pertaining to minor actinides.
2) assess the impact on safety of Generation IV reactors when introducing minor actinides into the fuel or blanket.
3) calculate the helium production and decay heat in fuels/blankets resulting from introduction of minor actinides

Background

In the previous chapter we have seen how plutonium is produced in nuclear reactors using uranium fuels. In addition to plutonium, spent fuel also contains so called "minor actinides", that is actinides found in minor amounts during plutonium production campaigns for the Manhattan project. These elements comprise of Neptunium (Np), Americium (Am) and Curium (Cm) and are formed along different production routes. Neptunium is the outcome of two subsequent neutron captures on ^{235}U:

$$n + {}^{235}U \rightarrow {}^{236}U + \gamma$$

$$n + {}^{236}U \rightarrow {}^{237}U^{*} \rightarrow {}^{237}Np + \beta$$

Americium appears in the system in two ways: Either through beta decay of ^{241}Pu, which has a half life of 14 years:

$$^{241}Pu \rightarrow {}^{241}Am + \beta$$

or through neutron capture on ^{242}Pu:

$$n + {}^{242}Pu \rightarrow {}^{243}Pu \rightarrow {}^{243}Am + \beta$$

where the half-life of ^{243}Pu is five hours. Curium is produced by neutron capture on americium. The relative fraction of plutonium, americium and curium in spent fuel depends on how long and under what flux the fuel has been residing in the reactor, as well as on how long the spent fuel has been residing in storage. Table 4.1 displays the typical composition of spent PWR (with a burn-up of 50 GWd/ton) fuel after four years of cooling.

Table 4.1: Composition of spent PWR fuel, after four years of cooling [OECD/NEA].

Nuclide	Fraction
Fission products	5,145 %
^{235}U	0.767%
^{236}U	0.552%
^{238}U	92.186%
^{237}Np	0.072%
^{238}Pu	0.042%
^{239}Pu	0.623%

Nuclide	Fraction
^{240}Pu	0.286%
^{241}Pu	0.155%
^{242}Pu	0.095%
^{241}Am	0.038%
^{243}Am	0.028%
^{244}Cm	0.010%
^{245}Cm	0.001%

From Table 4.1, it can be deduced that the minor actinides constitute less than 0.15% of the spent fuel mass. Comparing to the inventory of plutonium, we find that the MA/Pu ratio is about 1/10. The ratio will change slightly as function of storage time, due to the decay of ^{241}Pu into ^{241}Am and ^{244}Cm into ^{240}Pu.

However small, the inventory of minor actinides in spent nuclear fuel has a significant impact on the design and performance of geological repositories. Namely, americium contributes significantly both to long term radio-toxicity and heat load of the waste package .

The radio-toxicity relating to ingestion or inhalation of a nuclide must be distinguished from its radio-activity, due to variations in impact on biological functions. Here, we adopt a simplified approach and evaluate the effective dose E resulting from intake of a radio-active substance as

$$E = \sum_j \varepsilon_j A_j(t)$$

where the dose coefficient ε_j characterises the biological impact pertaining to the activity of the nuclide j. The dose coefficient has the unit of Sv/Bq and contains the entire information of absorbed physical dose (J/kg) and its corresponding biological impact. It is dependent on age, body-size and gender. A total dose of 1 Sv *is defined as* a 5% probability to acquire lethal cancer. The methods used for calculating the effective dose are continuously refined, and thus the dose coefficients change over time. The International Commission on Radiological Protection (ICRP) regularly publishes a recommended set of coefficients. In Table 1.3, values for ingestion by an average adult from ICRP 72 [ICRP 1996] are displayed for the actinides present in spent nuclear fuel. The corresponding values for inhalation are about a factor of 200 larger.

Table 4.2: Dose coefficients for ingestion by an average adult of nuclides present in spent nuclear fuel.

Nuclide	ε_{50}
^{235}U	47
^{236}U	47
^{238}U	44
^{237}Np	110
^{238}Pu	230
^{239}Pu	250
^{240}Pu	250
^{241}Pu	5

Nuclide	ε_{50}
^{242}Pu	240
^{241}Am	200
^{243}Am	200
^{244}Cm	120
^{245}Cm	210

Figure 4.1 displays the contribution of transuranium elements to the long-term radio-toxic inventory of spent PWR fuel.

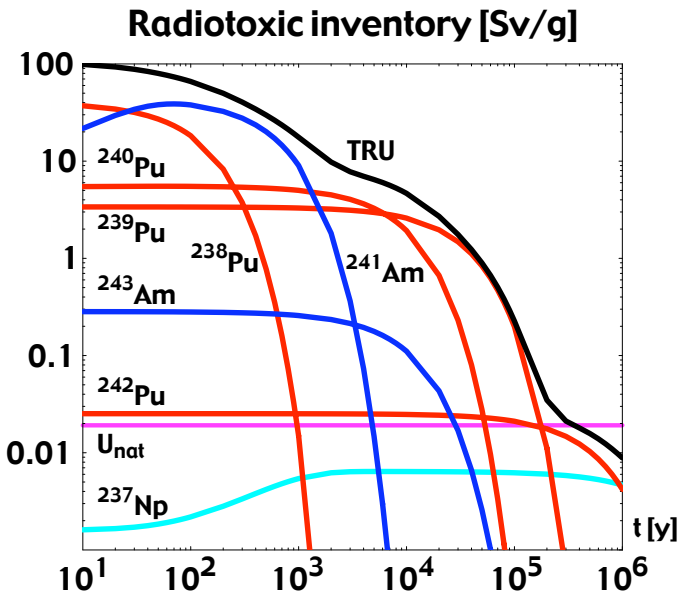

Figure 4.1: Contributions of individual transuranium nuclides to the specific long-term radio-toxic inventory of spent PWR fuel. Comparison is made with the specific inventory of uranium found in nature, which contains an equilibrium amount of radium and radon.

As can be seen, the contribution to the radio-toxic inventory of plutonium isotopes exceeds that of uranium in nature for about 300 000 years. Between 100 and 1000 years, the dominating nuclide is ^{241}Am. Here, we should emphasise that the very low mobility of transuranium elements in geological formations means that the radiological impact on the biosphere of a failing canister is dominated by fission and activation products. Only in the case of a human intrusion would americium and plutonium constitute a risk of radiological concern [Von Lenza 2007].

The radioactive decay of fission products and transuranium elements is accompanied by heat production. Alpha-particles emitted from Pu, Am and Cm nuclides have an energy of 5-6 MeV, which is deposited directly in the fuel. The Q-value for beta-decay is usually considerably smaller, but varies to a larger extent from nuclide to nuclide. Figure 4.2 displays the contributions to the total heating rate of one kg spent PWR fuel after a burnup of 50 GWd/ton. As can be seen, the major contributions to the shorter term beta-heating come from ^{137}Cs, ^{90}Sr and ^{241}Pu.

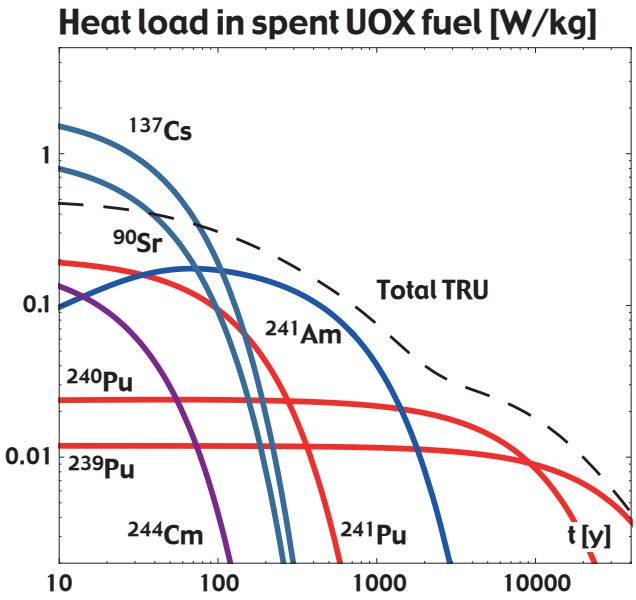

Figure 4.2: Contributions of fission products and transuranium nuclides to the heating rate of spent nuclear fuel.

During the active operation of a granite or clay repository, forced cooling is applied. Once the repository is closed, passive conduction has to ensure that the temperature of the water saturated bentonite, used as barrier for nuclide migration from failing canisters, does not exceed 100°C. This is the actual boundary condition determining the distance between two canisters, and hence the volume of the repository. From Figure 4.2 we may infer that after a relatively rapid decay of ^{137}Cs and ^{90}Sr, the heat load is dominated by ^{241}Am. Therefore, the design of the repository is made to passively manage the heat load from this nuclide, and the closure will be made 80 - 100 years after the last deposited fuel is unloaded from its reactor.

The second Generation-IV objective on waste minimisation and management may be translated into a requirement to recycle minor actinides for their transmutation to short-lived and/or fissionable nuclides. The potential for carrying out the latter task in fast neutron reactors was explored already in the 70's [Beaman 1979]. As we have seen above, the major benefit would result from recovering americium from the waste. Consequently, major efforts have been undertaken to identify and qualify possible means for making transmutation of americium possible in various reactor types, including LWRs [Youinou 2005], critical fast reactors [Buiron 2011, Kooyman 2017] and accelerator-driven systems [Tsujimoto 2004]. In this text-book, we focus on the liquid metal fast reactor option.

The transmutation of americium involves a large number of technical challenges, including its chemical separation, fabrication of americium bearing fuels and/or targets, the impact on reactor design, safety performance and spent fuel management. Aspects of the above will be treated in detail in Chapters 5, 6, 11 and 17.

Now, let us consider the paths for transmutation of americium in fast reactors. The fission probability of ^{241}Am and ^{243}Am in a typical fast reactor spectrum is about 10%. Hence, the vast majority of the americium inventory has be transmuted by neutron capture. Figure 4.3 illustrates how the neutron capture on ^{241}Am leads to the formation of ^{242}Am in its meta-stable state with a neutron energy dependent probability of 10-20%, otherwise forming the ground-state of this fissile nuclide. Whereas the meta-stable state has a half-life of 141 years, the ground state rapidly decays into ^{242}Cm with a branching ratio of 83%, and ^{242}Pu with a probability of 17%.

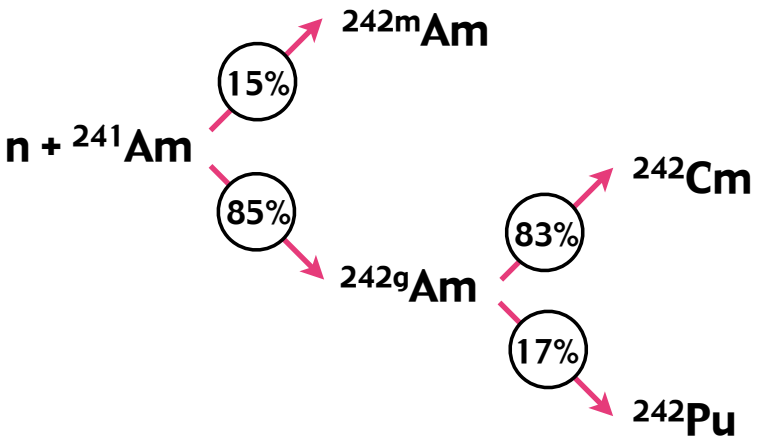

Figure 4.3: Transmutation of ^{241}Am following neutron capture.

^{242m}Am has the largest cross section for fission and the highest fission probability of all actinides with an appreciable life-time. Consequently, its formation contributes positively to reactivity and a large fraction is converted into fission products already in-pile. ^{242}Cm, although not being fissile, has a fission probability of 40% in a fast spectrum. However, its half-life of 162 days means that the major fraction of this nuclide will be converted to ^{238}Pu by alpha-decay. This particular decay turns out to be a major source of helium production as well as decay heat, with a direct consequence on fuel design and management. Finally, ^{242}Pu also has a fission probability of about 40%. In a closed fuel cycle, it will be transmuted to ^{243}Am with a 60% chance.

In summary, the majority of ^{241}Am transmutations leads to the formation of plutonium. The plutonium vector contains large fraction of ^{238}Pu, the decay heat of which contributes to improving proliferation resistance of the spent fuel.

The transmutation of ^{243}Am, on the other hand is conceptually more simple. Neutron capture will lead to the formation of ^{244}Cm. Revisiting Figure 4.2, we may note that the initial contribution from ^{244}Cm to the decay heat of spent LWR fuel is similar to that of ^{241}Am. However, its half-life is much shorter (18 years), which means that separation and storage of this nuclide could be an alternative to transmutation. However, the large decay heat (2.8 W/g) means that the if curium is separated, forced cooling would have to be applied to the storage, at a considerable cost [Salvatores??]. Other options include to recycle curium together with americium or to avoid its separation and keep it together with the lanthanide fission product stream [Bourge??]. In the former case, the impact on fuel cycle facilities will be considerable (see Chapter 5), and in the latter, innovative and complex separation chemistry has to be qualified for industrial use.

In order to meet the first Generation IV objective (efficiency of resource utilisation), the driver fuel of fast reactors would be manufactured from depleted uranium, which would should be converted to plutonium with an equivalent fissile conversion ratio equal to 1.0, as suggested in Chapter 2. Implementing recycle of minor actinides affects the equilibrium nuclide composition of the plutonium vector, due to the production of fertile Pu nuclides from the decay of ^{242}Cm and ^{244}Cm. Since the reactivity pertaining to plutonium depends on its composition, the relative ratio of uranium to plutonium would also be affected.

Once an "equilibrium" state of the fuel cycle has been achieved, the relative ratios of U/Pu/MA remain constant from one fuel load to the next. Prior to that, loading more MA than its equilibrium fraction would lead to a higher MA burning rate since the system strives to achieve equilibrium. On the other hand, loading less than the equilibrium fraction would result in a net production of minor actinides.

Prediction of the equilibrium fuel composition in a nuclear fleet containing Generation IV reactors is more complicated than the case of the classical breeder reactor, due to the delayed source of plutonium deriving from decay of curium. In the subsequent Chapter, you will learn how to calculate the equilibrium composition by solving the set of Bateman equations describing the connected transmutation paths of a closed fuel cycle.

In general, increasing the ratio of americium to plutonium in the fuel will lead to an improved specific minor actinide burning rate. This reduces the required fraction of power produced reactors operating with minor actinide bearing fuels in a nuclear fleet with a fully closed fuel cycle. However, there are a number of complications arising when increasing the inventory of americium in the fuel, including helium gas production, alpha decay heating and deterioration of important neutronic safety parameters.

Helium gas production

As described above, the alpha-decay of ^{242}Cm is a major source of helium gas. In conventional fuels, the noble gas inventory consists of the fission products xenon and krypton. On average, 0.23 xenon atoms and 0.02 krypton atoms are produced in a fission. The impact of the latter on swelling of the fuel during irradiation is discussed in Chapter 6. For each ^{241}Am nuclide transmuted by neutron capture, we expect more than 0.67 helium atoms to be produced in-pile or during cooling (see Figure 4.3). In contrast to Kr and Xe, He is expected to diffuse quickly through the fuel matrix, accumulating in the gas plenum of the fuel rod. In order to not exceed the permitted pressure load on the fuel cladding, the length of the plenum must be designed to accommodate this source of gas. This requirement will be applied for the core design approach implemented in Chapter 11.

Decay heat

In reactors operating on uranium based fuels, fission products are the main sources of decay heat, providing 6.5% of the total power during nominal operation. Using plutonium as fissile driver fuel, an additional contribution arises from the beta-decay of ^{241}Pu. When conducting transmutation americium, heat released by the alpha-decay of ^{241}Am, ^{242}Cm, ^{244}Cm and ^{238}Pu will increase the magnitude of the decay heat considerably. Taking proper account of this fact is important not only for assessment of loss of cooling accidents, but also fuel management procedures may be affected. Table 4.3 lists the specific decay heat production rate of the above listed actinides.

Table 4.3: Specific decay heating rates

Nuclide	Q_{decay} [W/g]
^{238}Pu	0.57
^{241}Pu	0.62
^{241}Am	0.12
^{242}Cm	123
^{244}Cm	2.84

Safety parameters

The presence of americium adversely influences several parameters essential for the safety performance of fast reactors, such as the fuel Doppler feedback, the coolant temperature coefficient and the effective delayed neutron fraction. The combination of these three phenomena leads to a reduced safety margin during transients, which must be compensated by reducing the power density of the fuel. In what follows, we will investigate the underlying mechanisms for the negative impact of americium on the aforementioned safety parameters.

Doppler feedback

As discussed in Chapter 3, Doppler broadening of capture resonances in even neutron number actinides is one of the most important safety mechanism in fast reactors. In order to operate, the Doppler effect requires the presence of neutrons in the resonance region. Figure 4.4 shows the relative contribution to Doppler feedback from neutron capture in ^{238}U resonances, as function of energy, when going from cold (T = 600 K) to

hot (T_{fuel} = 1800 K) state in a sodium cooled core with $(U_{0.8},Pu_{0.2})O_2$ fuel. The feedback deriving from captures taking place above 100 keV is negligible. Captures below 3 keV provide 60% of the feedback, in spite of constituting only 15% of the total capture rate.

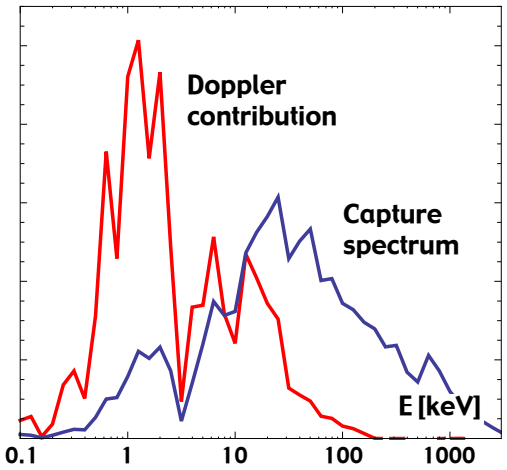

Figure 4.4: Relative contribution of neutrons to capture rate (blue line) and to the increase in capture rate by Doppler broadening (red line).

The soft tail of the neutron spectrum is significantly affected by americium's large cross section for capture of neutrons with high energy [Wallenius 2012]. Figure 4.5 compares the capture cross sections of ^{241}Am and ^{238}U. The capture cross section of americium is an order of magnitude larger than that of uranium in the displayed region. Therefore, in an americium bearing fuel, the probability for capture in uranium is reduced much more than the decrease in uranium concentration would indicate.

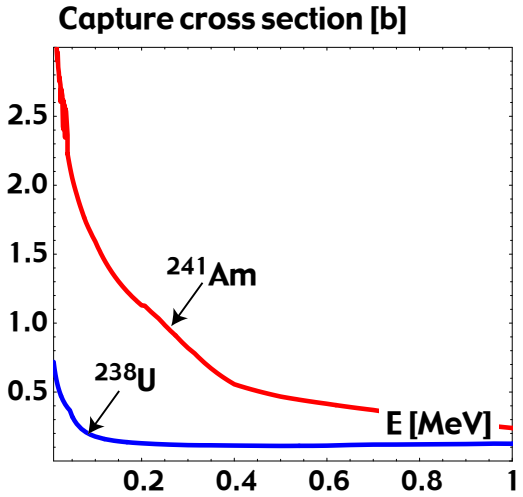

Figure 4.5: Capture cross sections of ^{241}Am and ^{238}U.

The outcome is a dramatic reduction in Doppler feedback when adding americium to the driver fuel of a Generation IV reactor. The effect is illustrated in Figure 4.6, showing how the Doppler constant (see Chapter 3) is reduced by a factor of seven when introducing 15% of americium into the MOX fuel of a sodium-cooled reactor.

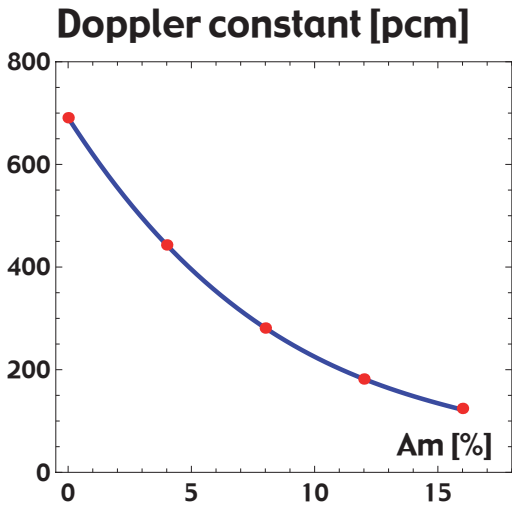

Figure 4.6: Doppler constant of a $(U_{0.8-x},Pu_{0.2},Am_x)O_2$ driver fuel in a sodium-cooled reactor.

Coolant temperature coefficient

As shown in Chapter 3, the spectrum hardening resulting from a coolant density decrease increases the probability for fission in even neutron number uranium and plutonium isotopes. This effect is accentuated when replacing ^{238}U with americium. Figure 4.7 illustrates the energy dependence of the fission probability for ^{238}U and ^{241}Am. As can be seen, americium is much more sensitive to changes in the magnitude of the neutron flux in the energy region between 0.2 and 0.8 MeV. Moreover, as fission of higher actinides produces more neutrons than fission of uranium and plutonium (see Table 4.4), a hardening of the spectrum will increase the average value of ν. The combination of these two effects lead to a higher coolant temperature coefficient in Generation IV reactors where the driver fuel contains americium.

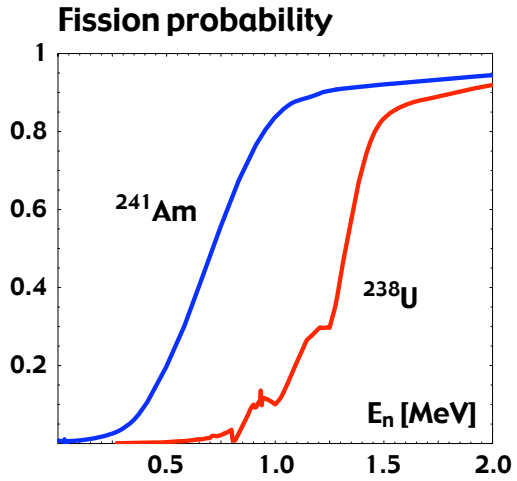

Figure 4.7: Fission probabilities of ^{241}Am and ^{238}Pu.

Table 4.4 compares sodium temperature coefficients calculated at T=690 K for different oxide fuels in a 1500 MW_{th} core with pin pitch to diameter ratio (P/D) equal to 1.24. The substitution of uranium with americium leads to an increase in the sodium temperature coefficient which is significant already for a few percent Am in the fuel.

Table 4.4: Sodium temperature coefficients in a 1500 MW_{th} core.

Fuel	α_{Na}
$(U_{0.8},Pu_{0.2})O_2$	+0.17pcm/K
$(U_{0.7},Pu_{0.2},Am_{0.1})O_2$	+0.34 pcm/K
$(U_{0.6},Pu_{0.2},Am_{0.2})O_2$	+0.47 pcm/K
$(U_{0.5},Pu_{0.2},Am_{0.3})O_2$	+0.58 pcm/K

Effective delayed neutron fraction

As shown in Chapter 3, the delayed neutrons have a lower median energy, and are less efficient in inducing fission in even neutron number nuclides than prompt neutrons. In fuels containing americium the high capture cross section in the range of the delayed neutron spectrum accentuates this effect, leading to a further reduction of β_{eff}. Moreover, since some fissions in Am and Cm takes place, more prompt neutrons are produced in each fission, and fewer delayed neutrons are produced. Table 4.5 lists the delayed neutron fraction for fission of americium and curium and Table 4.6 illustrates the reduction in β and β_{eff} as function of americium content in the fuel.

Table 4.5: Fission neutron yield and delayed neutron fraction for 1.0 MeV neutrons.

Nuclide	ν_{tot}	ν_d/ν_{tot}
^{238}U	2.53	1.89%
^{239}Pu	3.02	0.22%
^{241}Am	3.37	0.13%
^{244}Cm	3.42	0.13%

Table 4.6: Delayed and effective delayed neutron fraction (in units of pcm) for fuels with constant Pu fraction and varying Am content.

Fuel	β_{eff}/β
$(U_{0.8},Pu_{0.2})O_2$	390/460
$(U_{0.7},Pu_{0.2},Am_{0.1})O_2$	
$(U_{0.6},Pu_{0.2},Am_{0.2})O_2$	270/390
$(U_{0.5},Pu_{0.2},Am_{0.3})O_2$	220/350

Impact on transient behaviour

KTH has developed a consistent methodology to investigate the influence of americium on the behaviour of liquid metal cooled reactors during a set of representative design basis transients, such as the ones described in Chapter 3. The approach consists of designing a reference core with a uranium-plutonium fuel to have a fissile equivalent conversion ratio close to unity and a certain temperature margin to failure during the above set of transients. Then, by gradually increasing the americium fraction, the reduction in power density required to maintain the same margin to failure becomes a measure of the penalty resulting from the presence of americium in the fuel [Zhang 2010].

Figure 4.8 shows the maximum temperature in the MOX fuel of a medium sized sodium fast reactor, following a UTOP resulting from a reactivity insertion of one dollar with a rate of 0.05$ per second. For this accident, Doppler broadening is the most important negative feedback mechanism. Since the fuel temperature may increase by up to 1000 Kelvin during the transient, more than a dollar of negative feedback can be provided by MOX fuels with low content of americium. When raising the americium fraction, the Doppler constant is reduced and a higher asymptotic temperature is required to balance the positive reactivity insertion with negative temperature feedback from the fuel. For every additional percent americium, the maximum temperature during the transient increases with 75+15 K. In order to maintain a constant margin to melt, the nominal power density of the SFR would have to be reduced by six percent per percent americium.

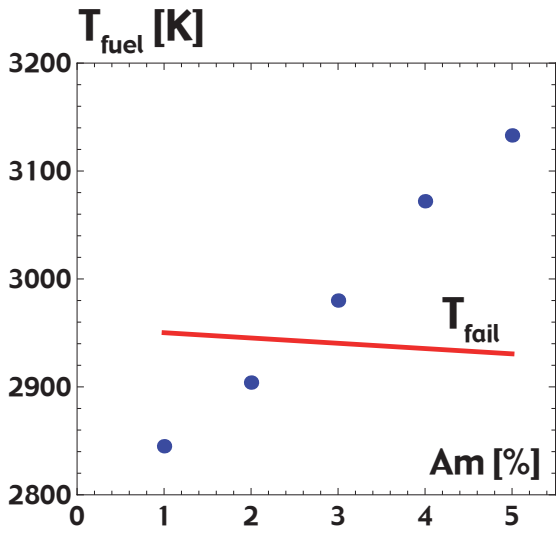

Figure 4.8: Fuel maximum temperature during a UTOP, as function of Am content in MOX fuel of a sodium fast reactor. The failure (melting) temperature is discussed in Chapter 6.

Heterogeneous recycling of americium in "minor actinide bearing blankets".

In order to mitigate the adverse impact on safety parameters of americium, one may introduce this element in the periphery of fast reactor cores, in the form of minor actinide burning blankets [Koch 1986]. Using neutrons leaking out from the driver fuel zone for the purpose of transmuting americium, the importance of neutrons produced in the blanket is low and up to 20% americium can be introduced without serious impact on the safety performance [Kooyman 2017]. The drawback of this approach is a relatively low consumption rate of less than 2 kg MA/TWh$_{th}$, and the large decay heat of the irradiated MABB assembly. Figure 4.9 compares the decay heat of MABB assemblies irradiated during 4100 full power days at the periphery of a 3600 MW$_{th}$ core, to that of a MOX fuel driver assembly.

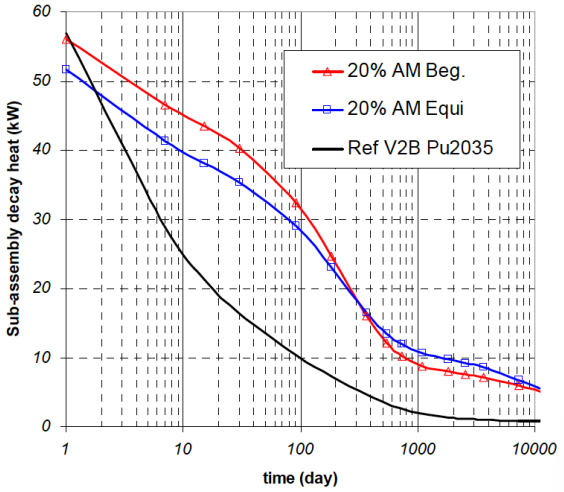

Figure 4.9: Evolution of MABB assembly decay heat [Buiron 2011].

Whereas the decay heat in the driver fuel assembly is reduced below the 7.5 kW limit for management in gas atmosphere within 180 days [Chabert 2015], it takes several thousand days for the MABB assembly to reach the same point. The main reason for this problem is the decay of ^{242}Cm, proceeding with a half-life of 162 days.

Questions:

1) What are the major contributors to radio-toxic inventory and heat emission of spent fuel?
2) What is the impact on safety when introducing minor actinides into the fuel of a fast reactor?
3) What are the mechanism for noble gas and decay heat production and that are unique for minor actinide bearing fuels?

Exercises:

1) Calculate cross sections for minor actinide transmutation using a Monte Carlo code, such as Serpent.
2) Using Serpent, calculate safety parameters of a fast reactor as function of MA content in the fuel.
3) Calculate the helium production in a driver fuel and MABB rods
4) Assess cooling time of driver fuel and MABB assemblies prior to carrying out fuel assembly management

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