

Design of a sub-critical reactor for transmutation of higher actinides

P. Mazgaj^{*,a}, D. Jimenez^b, D. Lopez^b

^a*Institute of Heat Engineering, Warsaw University of Technology
21/25 Nowowiejska Street, 00-665 Warsaw, Poland*

^b*Department of Reactor Physics
Royal Institute of Technology, Stockholm, Sweden*

Abstract

To reduce the storage time of spent fuel its radiotoxicity must be reduced through the transmutation of minor actinides. Generation IV reactors or sub-critical Accelerator Driven Systems (ADS) are options that appear technically feasible in view of the transmutation goal. The purpose of this work is to design a lead-cooled 400 MW_{th} sub-critical reactor which uses inert matrix nitride fuel. Many design issues are dealt with, such as the lifetime of the cladding material, the minimum reactivity swing fuel composition, the burning rates of MA and the safety coefficients. The results obtained show that ADS performs well in transmuted minor actinides and has good safety levels in respect of reactivity perturbations.

Keywords: Transmutation, Accelerator Driven Systems, Nitride fuel, Lead cooled ADS

1. Introduction

The aim of this assignment was to design a 400 MW_{th} sub-critical reactor with inert matrix nitride fuel and lead coolant, in order to maximize higher actinide burning rates under the constraint of a minimum reactivity swing. This report shows the methodology used to achieve the goal, the results obtained and the conclusions derived from them.

2. Methodology

The starting point of the design is to select a clad material that meets the objective of transmutation, while ensuring sufficient performance and safety, considering the given coolant (liquid lead) and fuel (nitride fuel with inert matrix). Due to the high dose

Table 1: Alloying elements of HT9 steel

C	0.2
Si	0.4
V	0.3
Cr	11.5
Mn	1
Fe	85.5
Mo	0.55
W	0.5

levels (up to 0.1 dpa per day) reached in this type of reactor and the significant corrosion rates in lead [1], standard stainless steel is discarded. In order to prevent swelling, nickel-free steel is selected [1]. This is also necessary to avoid He embrittlement, due to the reaction (n,α) in Ni. That means that high-Ni austenitic steels are also discarded. Therefore ferritic-martensitic steel is the best choice (with a temperature window of 673–823 K [1]). Embrittle-

*Corresponding author

Email address: piotr.mazgaj@itc.pw.edu.pl (P. Mazgaj*)

ment due to irradiation is a major problem in these types of steel. However, embrittlement can be minimized with a content of 9% Cr [1]. Regarding corrosion in lead, the selected type of steel performs better than the discarded austenitic steels, which confirms that the material was a good selected. This is because of the spontaneous formation of a protective Cr-oxide layer on the surface of the cladding [1]. The final choice was the ferritic-martensitic HT9, which also has higher thermal conductivity and a lower expansion coefficient than austenitic steels. The composition of the steel is shown in Table 1.

Since lead is used as a coolant, the selected cladding material will have a long term limit temperature of 820 K during normal operation, which must not be exceeded [1]. Since ferritic steels become brittle at low temperatures, the coolant inlet temperature should be high enough to avoid this problem (~700 K) [2]. The coolant mass flow must be calculated in light of the required increase of coolant temperature (less than 100 K) and the limited velocity of lead (< 2 m/s) due to the erosion of the protective oxide layer. Furthermore, the linear rating should be as large as possible in order to minimize fuel residence time and maximize the surface-to-volume ratio of the core. Taking into account these conditions, an iterative process in the MATLAB environment describing the thermal hydraulics in the hottest channel of the core is used in order to find the pin pitch and the core averaged linear power.

Once these parameters are defined, it is possible to define the dimension of the target. The radius of the spallation target is assumed to be the space occupied by 19 central sub-assemblies, whose geometry is known after the pin pitch is calculated.

Knowing the average linear power, the total thermal power and the number of fuel pins per assembly, the number of fuel assemblies in the core can be calculated, and thus the core radius can be defined.

All the given and calculated data at this point makes it possible to define a serpent input file (with an arbitrary fuel composition), which is used to calculate the neutron flux in the core. At issue here is the maximum flux received by the cladding, which opens the way to determining the irradiation time and thus the burn-up. Since serpent has no source mode calculation, an approximation of the real neutron flux

(which decreases exponentially with the core radius) with the fundamental mode flux is required. This approach considers that at the point of maximum real flux (where the dose to the cladding is at its maximum) the fundamental mode flux is about 10% smaller. This calculation is made with an estimated plenum height of 0.9 m, which shall be recalculated after the pin-cell calculations. Furthermore, the same Pu/MA ratio is maintained in the 2 fuel zones, while different fractions of inert matrix are used in both zones in order to get a value of 1.3 for the radial peaking factor.

As stated previously, since the burn-up will be given by the maximum acceptable dose to the selected cladding in the most exposed pin, which is assumed to be 200 dpa for ferritic steels (this dose is approximately equivalent to a neutron fluence of $3 \cdot 10^{23}$ n/s), the irradiation time is calculated using the fast neutron flux given by pin-cell calculations in serpent (defining a detector for neutron flux energies higher than 0.1 MeV).

Once the burn-up and irradiation time are defined, pin-cell calculations in serpent code are used again in order to find the Pu/MA ratio that minimizes the reactivity swing. Regarding the fuel composition used in the pin-cell calculations, the volume % of the inert matrix is calculated as the average from the 2 fuel zones in the whole core. The fuel should be uranium free and the ratio of Am to Cm may be fixed at 5:1.

Finally, the same pin-cell calculation gives the amount of He and Xe after the irradiation time. The gas plenum volume, and thus its height, is calculated by adopting the Ramses Criterion for the hoop stress resulting from the gas pressure. It is to be noted that the gas plenum is located in the lower part of the fuel pin.

With the serpent output files it is also possible to determine the burning rates of plutonium and minor actinides and the safety coefficients (Doppler, void worth, coolant temperature coefficient...) at BOC and EOC.

3. Results

The given design data are shown in Table 2.

The thermal hydraulics calculations specified in the methodology give axial temperature profiles for

Table 2: Design parameters

Clad inner diameter, mm	8.0
Clad outer diameter, mm	9.0
Fuel column height, m	1.0
Gas plenum height	TBD
Pins per sub-assembly	127
Hexcan wall thickness, mm	2.0
Space between hexcans, mm	2.0
Fuel zones	2.0
Radial steel reflector, rows	3.0
Radial peaking factor	1.3
Control elements	0
Proton beam energy, GeV	1.0
k-eff at BOL	0.97

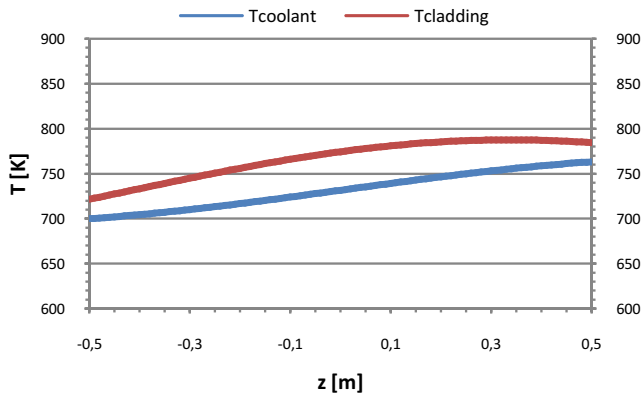


Figure 1: Coolant and cladding temperature axial profile

the coolant and cladding surface (Figure 1).

The obtained values for pitch, pitch over diameter and average linear rating are:

- $P = 14.7$ mm
- $P/D = 1.63$
- $X_{ave} = 26746$ W/m

The k-eff at BOL is 0.963 using two fuel zones with different volume % of inert matrix (60% for the inner zone & 50% for the outer zone) obtaining a radial peaking factor of 1.4, which seems to be acceptable.

From the serpent calculation representing the whole core is then obtained $\Phi_{clad} = 2.76 \cdot 10^{15}$ n/cm²s and so, assuming a maximum dose in the cladding of 200 dpa (equivalent to a neutron fluence of $3 \cdot$

Table 3: Reactivity swing for different Pu/MA ratios

	Pu/MA = 20/80	Pu/MA = 30/70
Reactivity swing, pcm	2,392	-1,199

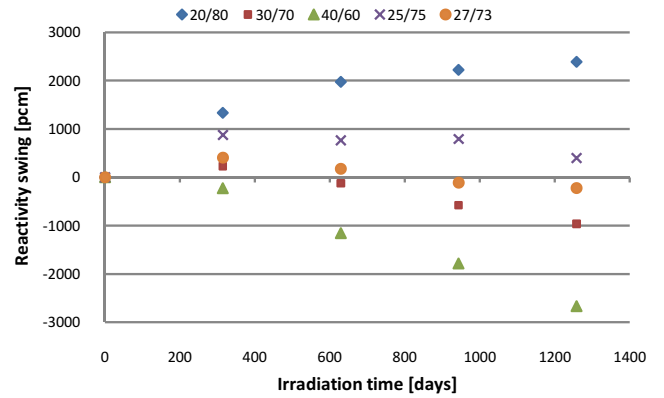


Figure 2: Reactivity swing as a function of Burn-up

10^{23} n/cm²), the total irradiation time can be calculated using the following relations:

$$\varphi \cdot t_{irradiation} \leq 3 \cdot 10^{23}$$

$$t_{irradiation} = 1256.5 \text{ days (3.44 yr)}$$

Once the irradiation time is calculated, the input file for pin-cell calculations is ready to be defined. Two calculations are performed with different Pu/MA ratios (20% and 30% of Pu) in order to define the optimum fuel composition to achieve the minimum reactivity swing. By extrapolation from the results obtained (see Table 3 and Figure 2), the required Pu/MA ratio is:

$$[Pu/MA]_{\text{minimum swing}} \approx 27/73$$

For this composition the reactivity swing is -629 pcm.

As can be seen in Figure 2, the change in reactivity is not linearly dependent on the Pu/MA ratio. That means that linear extrapolation gives a wrong value. Thus, some simulations with different ratios are performed in order to get the right swing.

Taking into account the requirements on fuel composition, the isotopic vector of Pu, Am & Cm is ob-

Table 4: Fuel composition in atomic fraction

Pu	1
Am	2.5
Cm	0.49
Zr	4
N	8

tained from spent UOx fuel and the ratio between Am & Cm fixed at 5:1. A sample of the fuel composition used in the pin-cell calculations (with 50% of inert matrix) is given in Table 4.

The previous composition is used in pin-cell calculations in order to find the amount of gases released. This amount is represented by the helium and xenon produced during the burn-up time, which is considered to be released to the fuel-clad gap (100% of release). The production of xenon during various steps of the irradiation time is reported in Table 5. Regarding the production of helium, it is evident that serpent gives a wrong value (it underestimates the production of helium). Therefore, helium must be estimated with the decay of Cm-242. Since Cm-242 is produced by the beta decay of Am-242 and is eliminated through capture, fission and alpha decay, the amount of helium at EOC can be calculated. The estimated amount of He at EOC is 0.28 g per pin.

From the amount of gas produced at the EOC it is possible to define the volume of the gas plenum (and thus its height) required to accommodate the He and Xe produced using the Ramses criterion for the hoop stress resulting from gas pressure [3]:

$$\sigma_{in} \leq \min \left[\frac{1}{3} R_m^{\min}(\theta_m); R_{po}^{\min}(\theta_m) \right]$$

$R_m^{\min} = 560$ MPa minimum tensile strength;
 $R_{po}^{\min} = 670$ MPa minimum yield stress (0.01% of plastic deformation) [4]. These values correspond to the selected cladding material at operational temperature.

According to the previous relation, $\sigma_{in} = 187$ MPa. Finally, using the ideal gas law and the hoop stress relation, it is possible to determine the plenum height:

$$V = \frac{nRT}{p_{gas}}; \sigma = p_{gas} \frac{r_{pin}}{t}$$

Table 6: Burning rates of actinides

Burning rate, kg/TWh	
Pu_{TOT}	19.79
Am_{TOT}	-41.16
Cm_{TOT}	-5.24
MA	-41.2

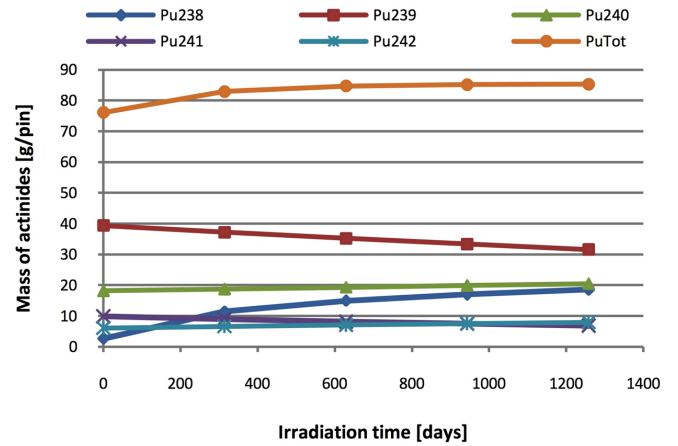


Figure 3: Isotopic evolution of plutonium with burn-up

where t is the cladding thickness.

$$H_{plenum} = 40 \text{ cm}$$

Once the plenum height is redefined, it is possible to determine the burning rates of actinides with pin-cell calculations. In the next figures (Fig. 3, Fig. 4 and Fig. 5) one can see the evolution of the actinides content in the fuel pin with the irradiation time. In particular, a burning rate of minor actinides of about 61 kg/TWh is obtained (see results in Table 6).

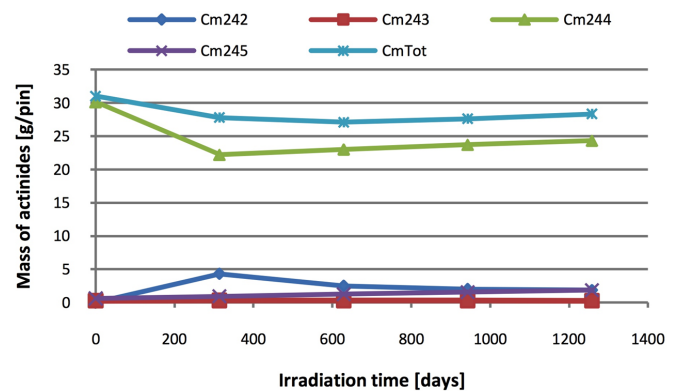


Figure 4: Isotopic evolution of curium with burn-up

Table 5: Production of xenon in fuel pin

g/pin	BOC	1	2	3	EOC
Xe-126	0	6.75E-09	6.33E-08	1.8E-07	3.74E-07
Xe-128	0	1.06E-04	4.82E-08	1.23E-03	2.37E-03
Xe-129	0	1.12E-04	1.12E-06	5.32E-06	1.48E-05
Xe-130	0	2.42E-04	9.6E-04	2.33E-03	4.37E-03
Xe-131	0	1.54E-0	3.12E-01	4.84E-01	6.56E-01
Xe-132	0	2.13E-01	4.3E-01	6.71E-01	9.14E-01
Xe-134	0	3.03E-01	6.05E-01	9.36E-01	1.27
Xe_{TOT}	0	0.67	1.35	2.1	2.85

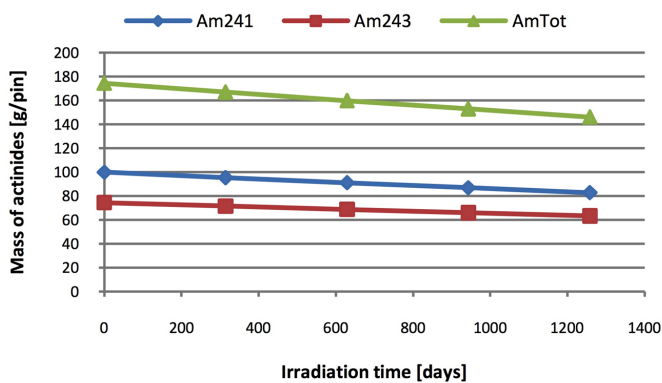


Figure 5: Isotopic evolution of americium with burn-up

In the previous figures one can notice a significant decrease in the americium content with burn-up (about 20% in 3.33 years). Also one can see that curium is burned during the cycle and tends to stabilize through the production of Cm-244.

However, after the cooling time of the fuel at the EOC (not taken into account here) the content of Cm-244 will decrease through beta decay.

The content of Pu increases with burn-up and at the end of the cycle stabilizes at a constant value, decreasing in quality over time. That seems reasonable, since the Pu for ADS fuel should come from spent ADS fuel after the first cycle, when a multi-recycling of minor actinides is desired [4].

Finally, the safety coefficients at the BOL and at the EOL were calculated, and are shown in Table 7.

As one can see from the calculated safety coefficients, an ADS is not intrinsically safe, unlike a LWR. The fuel temperature coefficient is less negative at the BOL, due to the high amount of Am in the fuel, but becomes more negative when americium is

Table 7: Safety coefficients reactivity at BOL and EOL [pcm]

TIME	α_M , pcm/K	α_F , pcm/K	void worth, pcm
BOL	0.2±0.05	-	983
EOL	0.08±0.01	-	411
		0.07±0.009	
		0.54±0.06	

burned. The void worth is calculated assuming total voiding in the core. As expected, it is positive, due to the inherent non safe characteristics of Am, and represents the weak point of the safety behavior of an ADS. Although the use of lead coolant eliminates the risk of coolant boiling, gas bubble introduction into the core remains a possibility (due to a broken steam generator or fission/helium gas release from a highly pressurized fuel pin at EOC [4]). In keeping with the fuel temperature coefficient, the void worth decreases when Am is burned, but always remains positive.

4. Conclusions

This report shows interesting results that seem to be in agreement with the ones from the available literature. The choice of cladding material is a critical point in the design of an ADS. Long fluence exposures of the cladding and heat deposition in the target are very restrictive conditions.

The thermal-hydraulics analysis confirmed liquid lead as a good coolant and a good spallation target material. The neutronic calculations made it possible to estimate an adequate system description, which is

required to achieve the desired transmutation objective.

Moreover, the final results of the burning rates and the safety coefficients are satisfying and they confirm the ADS as a good option for burning the minor actinides and closing the nuclear fuel cycle. Also sub-criticality ensures the safe operation of the reactor in spite of the use of fuels with a high content of americium.

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