MINOR ACTINIDE DESTRUCTION IN DEDICATED REACTORS

J. Tommasi, S. Massara, S. Pillon
DER/SIS/CAPRA, CEA-Cadarache, Bldg. 212
13108 Saint Paul lez Durance Cédex
France

M. Rome
EdF/SEPTEN, 12-14 avenue Dutrievoz
69628 Villeurbanne
France

Abstract

The neutronic performance of small size critical dedicated burners is investigated here according to the type of recycling performed (MA supply alone or together with some fissile element), the nature of the possible fissile supply (Pu, enriched U, U bred from Th), the coolant type (sodium or lead).

The main neutronic parameters are given, such as reactivity coefficients, together with mass balances, MA consumption and fuel composition. The trends for future studies are also indicated.

Introduction

Several reactor concepts enabling to burn minor actinides (MA) have been considered. A fast neutron spectrum has been shown to allow a positive neutron gain throughout the burning process [1], making it theoretically independent from an external neutron supply (e.g. by fuel enrichment in fissile nuclides, or a spallation source). However, MA burning has important consequences on the core safety, fuel cycle and economic related parameters, besides neutronic feasibility only.

The fraction of electrical power supplied to the grid by the MA burners present in the reactor fleet is a key parameter for any MA burning scenario. This fraction can range from a theoretical value as low as ≈6% (only MA loaded in the burners, and heat to electricity conversion of 40%, as today fast reactors) to a full 100% (MA are homogeneously "sprinkled" over all the reactors in the fleet).

The design of specific MA burners, called *dedicated reactors* hereafter, with important MA loads (ideally 100% of the reactor fuel) would allow to restrict specific cycle operations (fabrication, handling, reprocessing) and safety countermeasures to a limited fraction of the reactor fleet with respect to the homogeneous option.

Main assumptions

The main lines followed by our dedicated reactor studies have been first an important reduction in core size from conventional fast reactor designs, in order to enhance leakage effects in case of coolant voiding, as the presence of MA has a strong adverse effect on this parameter. The typical sizes retained for this study are in the range from 100 to 300 MW_{th}. Secondly a dense and "cold" fuel (i.e. with an important thermal conductivity) has been retained: the nitride fuel. Finally, a multiple recycling has been modelled in order to account for possible important changes between the first and equilibrium recyclings.

Within this frame, several parameters were varied, such as the type of recycling (i.e. whether MA alone or mixed with some fissile material are added to the spent fuel from a recycling to the next), the nature of this possible fissile supply (i.e. Plutonium, enriched Uranium, Uranium bred from Thorium), the coolant nature (sodium or lead), the possible use of a moderator within the core ($^{11}B_4C$, $ZrH_{1.8}$).

Tools and methods

The code system ERANOS1.2 has been used throughout this study to assess the neutronic parameters of dedicated cores [2]. Basic nuclear data are taken from the ERALIB1 library, containing data from JEF2.2 evaluations adjusted on a wide set of experimental configurations (≈350, but covering oxide fast reactor cores, standard or CAPRA-type ones, *not* dedicated small reactors with large amounts of minor actinides, non-oxide fuel, moderator within the core...).

For the first scoping studies, the calculation route was rather a crude one: homogeneous cell calculations with a fine (1968) group energy mesh providing by condensation the 33-group cross-sections used in whole core calculations, which were performed in diffusion theory. This preliminary range of calculations was only meant to provide trends in order to select a fewer number of cases on which to perform more refined calculations.

This second range of calculations has been performed with heterogeneous cell models, either two-zone (fuel in the inner zone ; steel, coolant and moderator mixed in the outer zone) or a full subassembly (S/A) model, the individual pins and wrapper being then explicitly taken into account. Then the whole core calculations were performed using transport theory using an S_n method (P_1, S_8). Once again, the cell calculations include a step in fine groups, and the core calculations use a condensed 33-group energy mesh.

A decay chain ranging from A=231 (^{231}Pa) to A=248 (^{248}Cm) has been used. The core burn-up is performed from beginning of life (fresh fuel) conditions to end of life (fully burnt fuel) conditions, i.e. no detailed batch management scheme has been investigated.

Two models of multiple recycling were used : the so-called *fertile* and *mixed* recyclings. A fertile recycling means that only MA are added as a topping material to the spent fuel of cycle n to form the loading of cycle n+1. Whereas in the mixed recycling MA are added together with some amount of fissile material (Pu, ^{235}U, ^{233}U).

The fertile recycling procedure can be detailed as follows : the spent fuel from cycle n is left to cool down for 5 years, then the fission products are removed and all the actinides recovered, with a 0.1% loss for Th, U and Pu, and a 1% loss for MA, going to the waste stream. Enough MA are added to obtain a given actinide mass (the same as the initial actinide mass of cycle 1). This homogeneous mixture, after a 2-year ageing, is then put in the core S/A to form the loading of cycle n+1. It is worth noting that in a fertile recycling, for a given fuel density in the S/A, the core size at equilibrium conditions is fixed by reactivity considerations : the reactivity must be closed to zero at the “end of cycle” conditions, i.e. at roughly half the fuel residence time with the simplified burn-up assumptions specified above.

The treatment of the mixed recycling is slightly more complex. The relative amounts of MA and fissile nuclides to be added to the spent fuel of cycle n are determined by three conditions: firstly, the total addition must restore a given initial actinide mass (that of the beginning of life conditions of cycle 1). Secondly, the ratio of the fissile (Pu, ^{235}U or ^{233}U) contents in the inner and outer core zones must be kept constant (for power flattening). Thirdly, the beginning of life reactivity of cycle n+1 must be the same as for cycle 1. The iterative algorithm used provides a unique solution to these requirements. In a mixed recycling the core size is no more a restrictive condition, provided that the necessary fissile topping required remains positive and lesser than the complement to the initial mass.

The coolant void reactivity is computed by fully voiding the core (fissile) zones, and keeping the reflector and shield regions full of their coolant. The Doppler effect is calculated by putting the actinide temperature from T_1 = 1500 to T_2 = 450 K, and the resulting *Doppler constant* is given by: $K_D = [k(T_1) - k(T_2)] / \ln(T_1/T_2)$.

Results

Results of the preliminary scoping studies

Here, core calculations were performed in diffusion theory, with a 33-group energy mesh, using data output by homogeneous cell calculations. A small core was used, modelled in a RZ geometry. The core height and radius were 60 and 66 cm respectively, with a thermal output of 250 MW. With a S/A pitch of 12.4 cm, this corresponds to 96 fissile S/A, divided in two fuel enrichment zones. The coolant is sodium, allowing for a "classical" S/A design.

A first result was that the reactivity loss in cores containing large amounts of MA is quite low, even for such small cores, allowing for large fuel residence times. A 1 500 equivalent full power days (efpd) residence time is assumed throughout the study.

The other main findings were that the use of oxide fuel made it difficult to perform an indefinite multiple recycling, while a denser fuel like nitride allowed it easily. Similarly the presence of an inert matrix mixed to the fuel impairs the recycling capability. Even with such a small core size, the coolant void reactivity remains quite important with a sodium cooling (and furthermore the delayed neutron fraction is lesser than in a standard fast reactor core). The Doppler constant remains quite low (hard spectrum) unless a very efficient (i.e. hydrogenated) moderator is added to the fuel in the form of separate pins. Finally the fuel composition at equilibrium is far from those of standard fast reactors. These points are illustrated in the following tables. Note that reactivities are measured in *pcm*, with *1 pcm = 10^{-5} Δk/kk'*. These tables are related to a mixed recycling with Pu coming from a UOX PWR spent fuel as a fissile material, and a recycling of Np+Am+Cm as MA. The moderator used is $ZrH_{1.8}$ representing a 5% volume fraction of the S/A.

Table 1. **Main parameters, 250 MW_{th} dedicated core, simplified scheme**

	First cycle	16^{th} cycle (quasi-equilibrium)
Average Pu content (w%)	46.2	63.2
Pu supply (kg/TWhe) MA supply (kg/TWhe)	– –	55.0 49.3
Reactivity loss (pcm/efpd) Sodium void worth (pcm) Doppler constant (pcm)	-7.63 1564 / 1675 -152 / -242	-7.80 1611 / 1587 -460 / -548
Delayed neutron fraction (pcm) Prompt neutron lifetime (10^{-7} s)	203 / 190 2.32 / 3.37	211 / 223 3.05 / 4.23
Max. Linear rating (W/cm) Max. Burn-up (%)	399 31.6	386

Values separated by a slash (/) are relative to beginning and end of life conditions. This convention will also hold for Tables 3, 4 and 5. Burn-up has here a small effect on the sodium void worth value because of a competition between the reduction in MA content and fission product build-up between beginning and end of life.

The fuel composition at the beginning of the equilibrium cycle is 4.4% U + 6.5% Np + 63.2% Pu + 17.0% Am + 8.9% Cm. The isotopic composition of Plutonium is:

Table 2. **Plutonium isotopic composition by mass, equilibrium cycle**

^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
17.2%	17.1%	38.9%	6.9%	19.9%

The fertile recycling scheme (i.e. supply of MA only) has been investigated, as it would allow the maximum MA consumption by fission (≈105 kg/TWhe).

In this case, the size of the core is determined by the need to have a well-balanced reactivity at equilibrium conditions. One is faced then to a dilemma : with no moderator within the core, the coolant void reactivity is small, but the Doppler feedback is vanishing, and to put some moderator within the core leads to increase the core size and so to reach large coolant void values. This is illustrated in table 3, with lead cooling instead of sodium (volume fraction in the fuel : 20-25% fuel, 5-0% moderator, 15% steel and 55% lead).

Table 3. **Core parameters, fertile recycling, with or without moderator**

Moderator	None	$ZrH_{1.8}$
Core power (MW_{th})	95	180
Core height/radius (cm)	45 / 47	55 / 66
Average burn-up (%)	18.9	17.9
Max. Damage (dpa)	164	128
Reactivity loss (pcm/efpd)	-4.59	-1.78
Coolant void (pcm)	-294 / -969	1548 / 659
Doppler constant (pcm)	-15 / -22	-157 / -208
Delayed neutron fraction (pcm)	143 / 156	154 / 172
MA feed (kg/TWhe)	107.1	106.9

Results of the refined scoping studies (RZ model)

These studies involve a more refined calculation scheme, i.e. transport calculation with heterogeneous cell model, see §3. This series of calculations involved a mixed recycling, with either a sodium or lead coolant, and different natures of the fissile supply (Pu from UOX PWR spent fuel, ^{235}U, ^{233}U). Results are summarised in tables 4 and 5 below. The core has a height of 50 cm and a radius of 53 cm, for a thermal output of 125 MW, and so an electrical output of 50 MWe. Twenty-one recyclings are performed to reach the equilibrium conditions, which are given in the tables.

Table 4. **Refined calculations, core parameters, sodium cooling**

Fissile composition	Pu	^{235}U	½ ^{235}U + ½ Pu	¼ ^{235}U + ¾ Pu	^{233}U
MA composition	MA	MA	MA	MA	0.9 MA + 0.1 Th
Average burn-up (%)	18.7	18.7	18.7	18.8	18.9
Max. Damage (dpa)	114	103	107	110	110
Reactivity loss (pcm/efpd)	-7.48	-14.61	-9.42	-8.30	-9.32
Coolant void (pcm)	1610/1698	-258/135	1089/1327	1394/1553	836/1049
Doppler constant (pcm)	-220/-270	-58/-50	-186/-233	-208/-255	-254/-339
Del. neutron fraction (pcm)	195/204	513/464	284/271	232/231	217/220
Prompt n. lifetime (10^{-7} s)	2.79/3.76	3.97/5.75	3.12/4.19	2.92/3.91	3.20/4.46
Fissile feed (kg/TWhe)	48.4	94.3	64.2	55.3	35.0
MA feed (kg/TWhe)	56.7	9.6	40.5	50.0	63.7

Sodium void values remain large with Pu as a fissile supply. Using enriched U (90% ^{235}U) instead of Pu leads to reduce the sodium void worth and to increase the delayed neutron fraction, but to the cost of a reduced Doppler feedback, of an increased reactivity loss, and of a quasi total loss of the MA burning capability (the U content in the core becomes very large).

Fissile supplies made of a mixture of U and Pu have thus been investigated, and allow to mitigate the drawbacks and advantages of pure U as fissile supply. However, the use of enriched U to operate dedicated cores in a long-term perspective can be questioned.

U bred from Th as fissile supply is also more attractive than Pu, but would require to start a Th cycle in conventional power reactors.

Table 5. **Refined calculations, core parameters, lead cooling**

Fissile composition	Pu	Pu	^{235}U	½ ^{235}U + ½ Pu	^{233}U
MA composition	MA	Am+Cm	MA	MA	0.9 MA + 0.1 Th
Average burn-up (%)	18.5	18.4	18.4	18.5	18.6
Max. Damage (dpa)	113	117	112	112	111
Reactivity loss (pcm/efpd)	-4.85	-4.71	-8.67	-5.83	-7.10
Coolant void (pcm)	1244/647	1485/829	-401/-356	869/421	70/-258
Doppler constant (pcm)	-203/-254	-328/-399	-134/-185	-188/-237	-273/-352
Del. neutron fraction (pcm)	179/191	163/182	318/291	222/219	204/210
Prompt n. lifetime (10^{-7} s)	2.76/3.76	3.01/4.11	3.31/4.56	2.93/3.95	3.40/4.67
Fissile feed (kg/TWhe)	26.9	21.9	49.3	34.1	24.0
MA feed (kg/TWhe)	78.8	83.8	55.7	71.4	73.4

It can be seen in table 5 that lead cooling allows reduced coolant void values and larger MA consumptions than sodium cooling, and that the use of ^{235}U as fissile supply has a lower negative impact on Doppler feedback and MA consumption.

Table 6 on next page gives the fuel isotopic compositions for the two cases in table 5 involving Plutonium as a fissile supply. Here the slash (/) separates compositions at the beginning of the first and equilibrium cycles. The complement to 100% is Uranium. This table shows the large Pu content in the fuel at equilibrium (≈55%) and its much degraded isotopic composition.

Always for a Pu fissile supply, the proportion of power coming from dedicated MA burners in the global reactor fleet power output ranges from 6% (Am+Cm burners) to 8% (Np+Am+Cm burners). This is quite low, but one must keep in mind the large number of small power individual burner reactors required to reach this proportion, that is roughly 2 dedicated 50 MWe burner every 1 large-size (1 500 MWe) power reactors. This motivates investigations for larger dedicated core sizes, even if several small dedicated cores can be grouped as modules in a same plant.

Table 6. **Refined calculations, lead cooling: fuel compositions (%)**

MA feed	Np+Am+Cm	Am+Cm
^{238}Pu	12.65 / 16.14	8.68 / 11.03
^{239}Pu	8.08 / 6.93	6.04 / 4.97
^{240}Pu	20.63 / 21.90	23.80 / 24.99
^{241}Pu	2.50 / 3.29	2.62 / 3.67
^{242}Pu	6.89 / 7.94	7.77 / 9.11
^{237}Np	10.77 / 7.81	0.56 / 0.57
^{241}Am	11.70 / 8.32	16.27 / 11.04
^{242m}Am	0.66 / 0.80	0.90 / 1.08
^{243}Am	9.94 / 8.00	14.27 / 11.00
^{243}Cm	0.09 / 0.11	0.14 / 0.16
^{244}Cm	7.26 / 9.11	10.90 / 13.53
^{245}Cm	1.85 / 1.98	2.69 / 2.85
^{246}Cm	0.83 / 1.03	1.25 / 1.56
^{247}Cm	0.12 / 0.14	0.18 / 0.22
^{248}Cm	0.04 / 0.05	0.06 / 0.07

Comparison with more detailed calculations

Table 7 compares the results for the first recycling between two different cell models: a 2-zone cylindrical model and a full representation of the S/A, with individual fuel and moderator pins being represented. In both cases, the core model is cylindrical (RZ), and flux calculations are performed in transport theory (S_n method). The fissile is a mixture of 50% Pu + 50% ^{235}U.

Table 7. **Comparison of calculational models**

Cell calculation	2-zone	Full S/A
Fissile content (%)	49.8	48.1
Reactivity loss (pcm/efpd)	-8.11	-7.92
Sodium void worth (pcm)	1 476/1 780	1 383/1 664
Doppler constant (pcm)	-113/-147	-107/-137
Delayed neutron fraction (pcm)	207/192	200/187
Prompt neutron lifetime (10^{-7}s)	2.42/3.28	2.35/3.16

As can be seen, the differences are small, less than 8% on all parameters, and allow to use a 2-zone cell model, involving much less calculation time than a full S/A representation.

Trends for future work

Future studies will cover several topics. Some investigations will be performed toward a size increase for the MA burners. A 3D (Hex,Z) core modelling, using nodal transport methods will be used to assess more accurately the retained designs. Transient studies will be carried out to assess the behaviour of such cores during incidental sequences. Finally, an uncertainty assessment will be necessary as MA are among the main contributors to reactivity in dedicated, with not so well known nuclear data for them. The uncertainty calculations will cover the fields of core reactivity, reactivity loss , actinide inventory during burn-up.

Conclusions

The dedicated core optimisation process involves contradictory trends. To increase the MA mass consumption rates and to enable an indefinite recycling, one has to turn to dense fuels, to replace sodium cooling by lead cooling, and to perform a fertile recycling (MA supply only). On the other hand, the use of an efficient (hydrogenated) moderator is necessary to recover a significant Doppler feedback, and small core sizes favour a low coolant void : these points favour a mixed recycling (MA + fissile material supply).

The scoping studies performed will allow to carry out transient studies, in order to check the kinetic behaviour during incidental sequences, and an uncertainty analysis, to assess the impact of the poor knowledge of MA cross-sections on reactivity, burn-up, reactivity coefficients.

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