The use of GFR dedicated assemblies in the framework of advanced symbiotic fuel cycles: An innovative way to minimise long-term spent fuel radiotoxicity

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Abstract

Transmutation is one of the most promising strategies to minimise minor actinide inventories. Particularly, during NPPs operation, Am and Cm isotopes are generated by capture and transmutation processes from U and Pu. High-neutron flux (typical of fast reactors) and high-neutron capture cross-section can help implement such a strategy. Fast reactors charged with some moderated dedicated assemblies (DAs) could supply such requirements for the transmutation process. DAs are filled with MA and are inserted in the active core with the aim of optimising and improving the reactor burning capability. Such optimisation of the layout and the analysis of the results obtained are the main topics of the work.

Introduction: The use of dedicated assemblies in GCFR for minor actinide disposal

From the radiotoxicity point of view, MA are dangerous for mid-term management, so starting from a destruction equation written for a generic nuclide in a generic spectrum, a destruction factor (*df*) can be defined[2][6] as follows:

$$\frac{dN}{dt} = -\sigma_a \cdot \varphi \cdot N(t) \Rightarrow N(t) = N(0) \cdot e^{-\sigma_a \cdot \varphi \cdot t}$$

$$N(t^*) = df \cdot N(0) \Rightarrow \varphi \cdot \sigma_a = \frac{\ln(\frac{1}{df})}{t^*} \Rightarrow \varphi = \frac{\ln(\frac{1}{df})}{\sigma_a \cdot t^*}$$

Where:

- *N(t)* is the concentration of the considered nuclide as a function of time;
- σ_a is its effective absorption cross-section;
- φ is the one-group flux;
- t^* is the time necessary to reduce the concentration of this nuclide by a factor *df*.

Using *df* as a reference figure of merit with the aim of maximising transmutation rate, high-fluence and high-MA neutron capture cross-section results are needed.

Fluence could be improved as follows:

- building the core with materials that reduce parasitical absorptions (strategy often followed for high flux thermal research reactors);
- choosing a hard spectrum (fast reactors).

Regarding the MA neutron capture cross-section, it is possible to improve such parameter by optimizing neutron spectrum as follows:

- choosing a neutron spectrum as soft as possible (since all the cross-sections show the 1/v trend in the low energy region);
- trying to "centre" the spectrum in suitable resonance regions (although this strategy seems very difficult to be rigorously applied in practice).

However, while fast reactors are characterised by high neutron flux, cross-sections are higher in thermal spectrum. So, in order to realise both conditions, it is possible to introduce particular dedicated ("over-moderated") elements in a fast reactor core, the so-called *Dedicated Assemblies* (DAs).

As already stated and described in previous works [1] [2] [4] [6] [7], gas-cooled fast reactor (GCFR) can be exploited for MA[1] burning through a transmutation process. In particular, the plate-type GCFR 2400 MW_{th} "E" proposed by CEA [3] [6] [7] is analysed: in this the DAs are inserted among standard assemblies.

DAs are filled with MA in a Coated Particle (CP) TRISO matrix. On the basis of previous calculations [2], it is possible to say that the CP TRISO with a kernel composed of inert matrix shows a good behaviour from the aspects of resistance to gaseous fission products leakage and of structural integrity at high-burn-up. Such a compound (mechanically stable) limits MA quantities chargeable for each DA in ranges from 2 to 9 kg. In the

[1] Np, Am and Cm are indicated as MA.

present work different DAs core layouts have been studied; indeed DAs can be placed in central and/or peripheral zones of the active core.

Calculations

The analysed configurations were selected taking into account the delayed neutrons fraction values: a configuration was considered acceptable if β is greater than 300 pcm (such threshold value is in agreement with previous GCFR designs [3]); this constraint implies loading an amount of around 6 kg of MA per DA. The evaluations of β were performed by MCNP5 code.

In the performed evaluations, besides the delayed neutrons fraction, the following trends were also considered: k_{eff} vs. burn-up, six-group neutron spectra vs. burn-up and MA masses trends vs. burn-up.

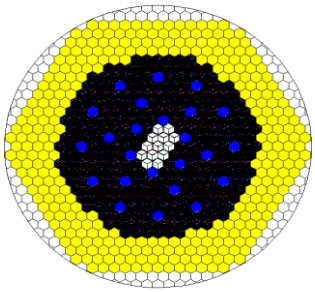
The burn-up calculations were performed by MONTEBURNS code.

Configuration analysis

Central geometry

The reference core layout [2] [4] was modified by inserting in a central position 13 DAs, each containing 6 kg of MA. In Figure 1 the X-Y section of GCFR core is shown; the core is composed of 13 DAs (white colour in the centre), reflector (yellow), standard assemblies (black), helium channels (blue) and the external "vacuum" (white colour on the external border). For this configuration the delayed neutron fraction is acceptable: β = 312 pcm. Hereafter this geometry will be called 13x6.

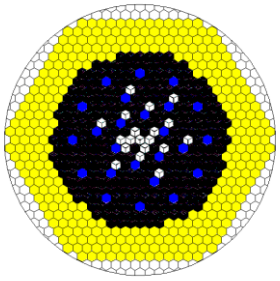
Figure 1: GCFR X-Y section for central geometry



Central and concentric geometry

The geometrical layout shown in Figure 2 presents DAs placed in the semi-peripheral zone of the core: 12 DAs are placed in two concentric zones near the centre and 7 DAs are in the centre of the core. Compared to the previous case, the total amount of MA contained in the core is increased significantly, however, this amount is more widely distributed; again the delayed neutrons fraction is acceptable: β = 308 pcm. This case will be indicated as 19x6.

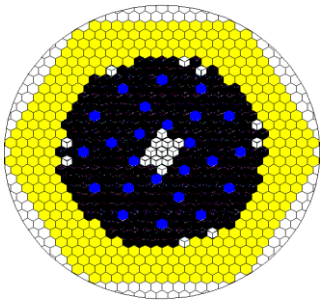
Figure 2: GCFR X-Y section for central and concentric geometry



Central and peripheral geometry

The third analysed configuration (Figure 3) presents 11 DAs in the centre (like the first case, 13x6) and 9 DAs in the peripheral zone (near radial reflector assemblies). In this geometry MA mass amount is greater than previous solutions, but, due to peripheral zone exploiting, an acceptable delayed neutrons fraction is again obtained: β = 313 pcm. This case will be indicated as 20x6.

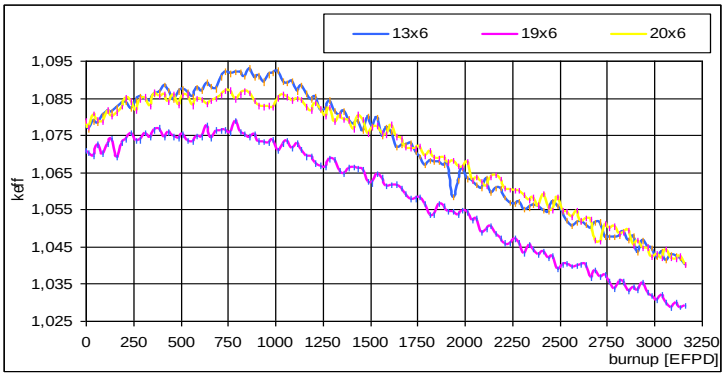
Figure 3: GCFR X-Y section for central and peripheral geometry



Comparison among obtained results

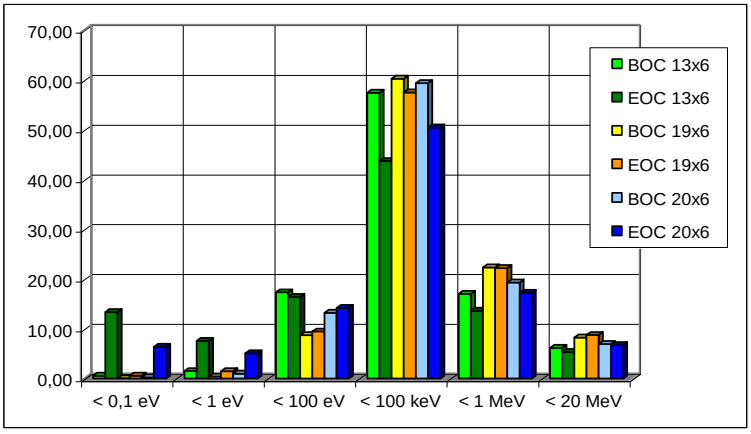
The comparison among results obtained from the different above-written configurations is explained below Trends of k_{eff} vs. burn-up parameters for the analysed cases are shown in Figure 4 (relative error is also reported); it is to note that k_{eff} trends are comparable for the 13x6 and 20x6 cases.

Figure 4: Comparison of k_{eff} vs. burn-up



In Figure 5 the six-group spectra at the Beginning of Cycle (BOC) and at the End of Cycle (EOC), i.e. at 3175 EFPD, are reported. The three different geometries imply different behaviours from neutronic (particularly neutron flux) point of view: in the range of interest for the transmutation process (energy values lower than 100 keV and higher than 100 eV), the 13x6 case presents the lowest spectrum values at BOC and at EOC conditions; moreover, this solution has the greatest spectrum variation between beginning and end of cycle; instead, solution 19x6 presents the lowest variation between BOC and EOC conditions for all considered energetic ranges. However, for all considered cases, the higher flux is in the range between 100 eV and 100 keV.

Figure 5: Comparison of six-group spectra



In Figures 6, 7 and 8 the comparison of MA masses trends vs. burn-up is reported; the material contained in DAs is indicated as M.2 while the material contained in standard assemblies is called M.1.

Figure 6: Trends of Am^{241} masses vs. burn-up for M.2 material

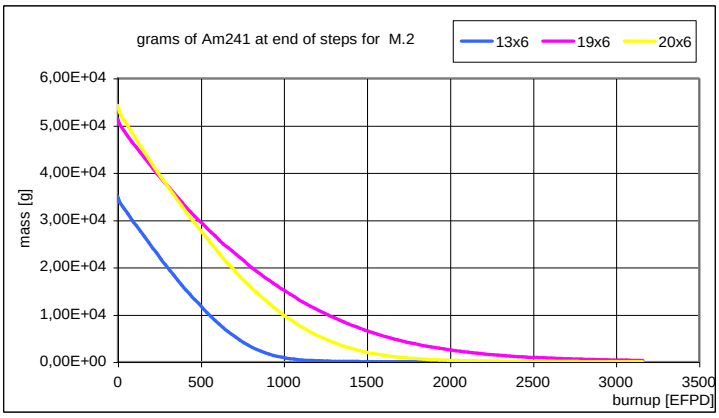


Figure 7: Trends of Am^{243} masses vs. burn-up for M.2 material

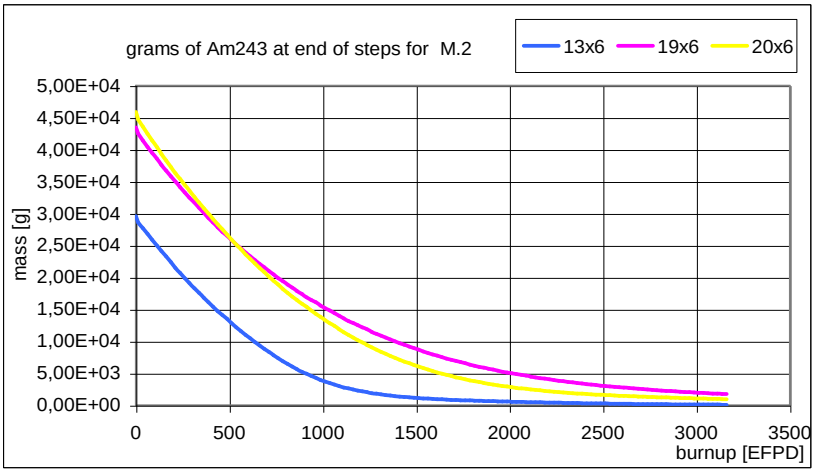
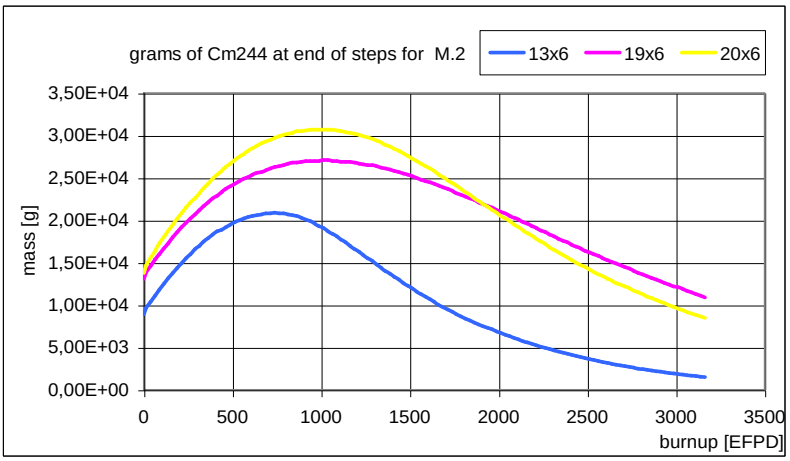


Figure 8: Trends of Cm^{244} masses vs. burn-up for M.2 material



The use of dedicated assemblies can considerably reduce Am mass. In contrast, Cm mass trends show an initial growth then followed by a decrease to arrive below the initial amount; in fact, while Am^{241} and Am^{243} vs. burn-up have monotonic decreasing trends (final masses are around zero), Cm^{244} vs. burn-up trends are not monotonic due to the initial increase arising from Pu^{242} and Am^{243} transmutations (heavy metal build-up).

As figures of merit, some relationships between masses at EOC and BOC were introduced. In particular, in Tables 1 and 2 two figures of merit are introduced; the ratio between M.2 masses at EOC and at BOC conditions, and the ratio between M.1+M.2 masses at EOC and at BOC conditions (indeed, in standard assemblies Am and Cm are generated by transmutation during operation, although they are not present at BOC).

Table 1: Masses ratios at EOC and BOC conditions for M.2

Nuclide		Am^{241}	Am^{243}	Cm^{244}	HM[2]
$\frac{M_{(M.2)EOC}}{M_{(M.2)BOC}} \cdot 10^2$	**13x6**	0.000	0.272	17.040	7.850
	19x6	0.525	4.023	83.206	36.018
	20x6	0.028	2.035	61.377	18.772

Table 1 shows that the 13x6 case presents the lowest values of the parameter for every MA analysed, while the 19x6 solution presents the greater values of the ratio. In Table 2 the second parameter is analysed (M.1 + M.2, i.e. the entire MA mass inside the whole core). Case 20x6 presents the lowest values of the mass ratio.

Table 2: Mass ratios at EOC and BOC conditions for M.1 + M.2

Nuclide		Am^{241}	Am^{243}	Cm^{244}	HM
$\frac{M_{(M.1+M.2)EOC}}{M_{(M.1+M.2)BOC}}$	**13x6**	4.686	3.618	4.374	0.893
	19x6	3.235	2.638	4.008	0.893
	20x6	3.045	2.395	3.433	0.892

The analysis of the parameters highlights the 13x6 and 20x6 configurations as the best suitable ones for setting up the core layout in order to improve the capability of MA burning. Looking at the first figure of merit (M.2 ratio), the best solution is 13x6, while for M.1 + M.2 ratio the best solution is 20x6 (obviously, the lower ratio value indicates better burning capacity). The MA mass charged in DAs and the mass of MA created in standard assemblies modify neutronic behaviour and influence the entire transmutation process; in the completed analysis of the burning process by transmutation way it could be more correct to consider all MA mass present in the active core (although it depends on the type of analysis carried out).

The decay process for minor actinide disposal

A further improvement in MA reduction could be obtained by means of radioactive decay; keeping MA in storage for a certain time (at least longer than a decade) before reprocessing it yields a significant reduction in Cm quantities compared to previous results at EOC.

Radioactive decay, due to Cm^{244} relatively short half-life, can significantly reduce the amount of this nuclide loaded in the core. Moreover, after the decay process, it is possible to handle a lower radiotoxic material inventory with evident advantages for reprocessing procedure and for health (mainly from the radioprotection point of view).

MA from NPPs SNF should be placed in an interim cooling pool for at least 1 year, due to the decay heat generated; then they should be placed in a storage pool for the decay process.

[2] Acronym HM (Heavy Metal) defines all the actinides (i.e. elements with Z>89).

In the further analysis it is assumed that total MA mass in each DA remains equal to previous analysis (i.e. without considering the disposal option). Obviously, after the disposal, a variation in the relative mass fraction values of Am and Cm is evident (due to their different half-lives). In relative terms, Am mass will increase while Curium mass will decrease in each DA. The amount of time assumed for the decay process in the present study is 30 years. The decay calculation has been performed by means of CARL code [5].

Comparison of analysed solutions for Am and Cm elimination (with "disposal" option)

Only 13x6 and 20x6 layouts are considered in this part of the study.

As for previous calculations, the same two figures of merit are taken into account. The mass preceding the decay process is considered as BOC mass (it is not equal to the mass loaded in the reactor after reprocessing); substantially, the decay process has been considered as an "inner" part of the cycle.

Tables 3 and 4 report the results for the two considered figures of merit in the "disposal" option (13x6d and 20x6d cases) and, for comparison purposes, in the "standard" option (called, as above, 13x6 and 20x6 cases).

Table 3: Mass ratios at EOC and BOC conditions for M.2

Nuclide		Am^{241}	Am^{243}	Cm^{244}
$\frac{M_{(M.2)EOC}}{M_{(M.2)BOC}} \cdot 10^2$	**13x6d**	0.000	0.288	16.8
	13x6	0.000	0.272	17.040
	20x6d	0.032	2.216	58.288
	20x6	0.028	2.035	61.377

It is remarkable that the 13x6d solution presents better values than the 20x6d solution. Focusing on the former case (13x6d case, disposal option), the ratio for Am^{243} is slightly greater than the one in the standard option 13x6 case, whilst for Cm, disposal option leads to a further reduction compared to the standard option.

Table 4: Mass ratios at EOC and BOC conditions for M.1 + M.2

Nuclide		Am^{241}	Am^{243}	Cm^{244}
$\frac{M_{(M.1+M.2)EOC}}{M_{(M.1+M.2)BOC}}$	**13x6d**	4.190	3.236	3.918
	13x6	4.686	3.618	4.374
	20x6d	2.721	2.390	3.093
	20x6	3.045	2.395	3.433

Analysing the results in Table 4, it is notable that the "disposal" option is a more suitable solution for MA reduction if all the fuel present in the active core is considered. Taking into account this parameter, the 20x6d case shows the lowest values.

Conclusions

The possibility of using dedicated assemblies in a GFR core could represent a good solution for MA reduction. Placing DAs in the central and peripheral zones allows an acceptable delayed neutron fraction. To increase the amount of DAs in the active core (maintaining an acceptable β value), placement in both the central and peripheral zones has been adopted. In the present study, different peripheral configurations were analysed to maximise reduction in MA amounts. Furthermore, two different reprocessing options were studied: the standard and the disposal options. In the former, MA are reprocessed immediately after the cooling pool while in the latter option, a longer storage time for spontaneous radioactive decay (about 30 years) is forecast before reprocessing. As expected, the disposal option is a preferable solution to reduce Cm^{244} mass.

In order to evaluate the best solution, two figures of merit have been used: the ratio between the masses at EOC and at BOC limited to the materials contained in DAs, and the ratio between the masses considering MA present in the whole core. The analysis of the results obtained leads to considering the 20x6d case as more suitable from the whole core burning capability point of view.

In future works, other burning strategies will be considered, e.g. different exploitation of active core in the peripheral zones rather than in the central zone.

The use of dedicated assemblies in the framework of advanced symbiotic fuel cycles could contribute to the closure of NPP fuel cycle with the aim of minimising long-term spent fuel radiotoxicity. Further studies are planned on the transmutation process with dedicated assemblies in fast reactors by the research group.

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