Fast reactors in minor actinide management

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Abstract

In the frame of Generation IV (Gen-IV) and Sustainable Nuclear Energy-Technology Platform (SNE-TP) Initiatives, the waste minimisation and the thermal load (on the geological repository) reduction constitute important issues. The effective way to manage MA is their transmutation in nuclear reactors: from the thermal to the fast critical and subcritical reactors. If both waste minimisation and thermal load reduction objectives are pursued then the TRU fuel multi-recycle is mandatory. This suggests the involving of the liquid-metal-cooled fast reactors (LMR), for which fuel recycling is a "natural" feature. Minor actinide transmutation is not a simple and easy issue. The loading of MA, from the core neutron design point of view, generally affects many of the physics parameters. In the present paper, two candidate solutions (cooled by heavy metal) will be compared from the viewpoint of some core design aspects. It will be shown that the MA effective management business requires a dedicated design approach. On the contrary, the introduction of MA into critical reactors with MOX-based fuels, classically (as usual) designed, deteriorates the safety margin of reactors, compromising the feasibility of the proposed solution itself.

Background

A reliable and economical nuclear fuel cycle, with a safe management of the high-level radioactive wastes (HLW), is mandatory in the pursuing of a sustainable development of the nuclear energy in both Generation IV and Sustainable Nuclear Energy-Technology Platform Initiatives [1]. If wastes minimisation and thermal load reduction are pursued, then minor actinides (MA), mainly neptunium (Np), americium (Am) and curium (Cm) should be appropriately managed. The effective way to manage MA is their transmutation in nuclear reactors. Several advanced critical and subcritical reactor concepts with enhanced performances, improved safety and environmental impact have been proposed.

Actinide recycle has long been an interesting and attractive concept associated with the closure the nuclear fuel cycle and the improvement of the nuclear waste management. The concept involves transmutation or fissioning of the high-lived actinides to shorter-lived fission products. Since the fuel recycle concept is essential for a "significant" waste reduction, the most promising option for any type of fuel recycle concept is the use of fast reactors. The primary incentives for transmutation of these higher actinide (TRU) isotopes is to eliminate as many as possible of them from the ultimate waste stream via processing spent fuel and to recycle the TRU as fast reactor fuel resource. The choice between fast subcritical and critical system will be a consequence of some constraints-choices concerning technological problems related to both reactor and associated fuel cycle, with important issues as the financial support and the industrial application time.

The exponential behaviour of the MA isotope concentration versus their equilibrium concentration:

$$C(t) = C_{eq}\,[1 - e^{-f(\sigma,\,\Phi,\,\lambda,\,\ldots)t}] \qquad (1)$$

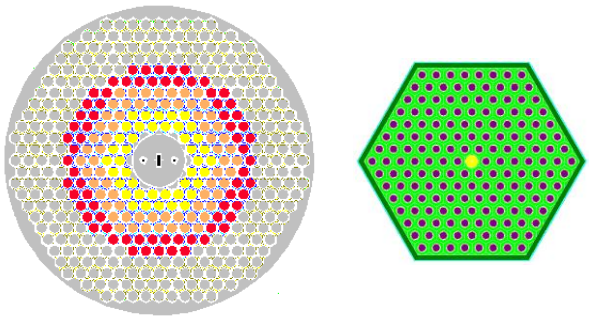
depends not only on the MA loaded amount in the core, but also on the cross-sections and the reactor neutron spectrum, with a time constant proportional to the reactor neutron flux. Now, in a critical reactor the maximum loaded MA amount in the core depends on the dynamic behaviour of the system; on the contrary, in a subcritical reactor there is no such limitation. Therefore, it is of interest the investigation of the transmutation capabilities of fast, heavy liquid metal-cooled, critical and subcritical systems.

Reactor core description (boundary conditions/introduction)

Two reactor concepts: the EFIT subcritical reactor and the ELSY critical reactor, developed respectively within the 5th and 6th EU Framework Programmes, will be compared in terms of: MA core loading, neutron performance and reactivity coefficients, transmutation capabilities, etc. Both reactors have common characteristics: same cladding material (ferritic-martensitic steel T91, with a maximum allowed temperature of 550°C taking into account the aluminisation process) and same coolant material (Pb which has a similar impact on the spectral properties) leading to common problems on the fuel assemblies (FA) radial power distribution which implies an optimisation process.

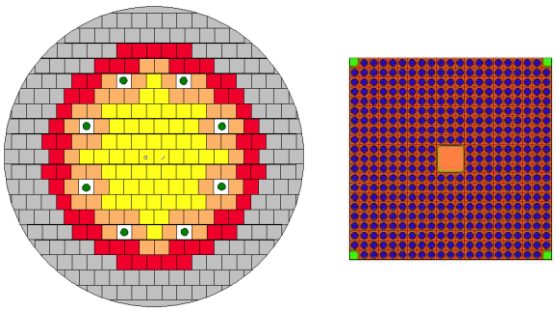
The objective of the Pb-EFIT core, Figure 1, is to burn nuclear waste producing electricity at an acceptable economical cost. The design [2] has been developed with the supplementary objective of achieving the maximum transmutation rate, so that only the MA are burned at the rate of about 42 kg/TWh_th (assuming an energy release of about 200 MeV per fission event). Using U-free fuel no Pu is produced through ^{238}U capture, while the Pu disappearance through fissions and captures should be partially balanced by Pu production through higher actinide transmutation and/or decay. In this way the Pu net balance, during the cycle, is almost null; in consequence the BU reactivity swing is around zero. The EFIT core is based on the hexagonal wrapped layout, Figure 1. The power flattening has been achieved through the use of different fuel volume fractions and/or by means of different pitch lengths and/or pin diameters.

Figure 1: EFIT core and FA layouts



ELSY is a 1 500 MWth (with an efficiency of 40%) MOX-fuelled and lead-cooled pool-type [3] fast reactor, with the purpose of electric energy production in a competitive and safe design. In the ELSY core, since the wrapperless FA geometry, Figure 2, does not allow to flatten the coolant outlet temperatures by flow rates regulation, which is one of the most important design aspect because of the Pb corrosion behaviour, the optimised temperatures distribution is obtained through the power distribution flattening carried out by the three radial zones enrichments.

Figure 2: ELSY core and FA layouts



The basic design constraints of the EFIT and ELSY reactors, assumed still valid in this study, are the following [2,3]:

- Cercer fuel type: $(Pu,MA)O_{1.88}$ + MgO; C(Pu) = $PuO_{1.88}/(Pu,MA)O_{1.88}$ = 0.457 for the EFIT reactor, distributed in three radial zones, in the ratio of 42/66/72 FA, with volume fraction: $AnO_{1.88}$/MgO = 0.43/0.57, 0.50/0.50 and 0.50/0.50.
- MOX fuel type: $(U,Pu)O_{1.97}$ for the ELSY reactor, distributed in three radial zones, in the ratio of 56/50/56 FA, with C (Pu) = $PuO_{1.97}/(U, Pu)O_{1.97}$ = 0.1454, 0.1767 and 0.2061.
- Pb inlet/outlet core temperature: 400/480°C.
- Pb velocity ≤ 2 m/sec, to limit corrosion and pressure drop.
- Maximum allowed peak linear rating: 220 and 320 Wcm^{-1} for EFIT and ELSY reactors, respectively.
- Maximum clad temperature: 550°C.
- Maximum fuel temperature: 1 330/2 100°C for EFIT and ELSY reactors, respectively.
- Fuel residence time: three years by three batches and five years by four batches for EFIT and ELSY reactors, respectively.
- Maximum burn-up rate: 100 MWd/kgHM.

- Peak clad damage: 100 dpa.
- Anti-reactivity of 10 $ (some 3 000 pcm) for each of the three independent control rod systems of the ELSY reactor.

Table 1 collects the fuel isotopic composition, with MA distribution (in wt.%) in the ratio of:

Np:Am:Cm = 3.8843:91.8184:4.2972

for both EFIT and ELSY cores.

Table 1: EFIT and ELSY fuel isotopic composition (wt.%)

	EFIT	ELSY		EFIT	ELSY		EFIT	ELSY
			^{238}Pu	3.7372	2.333	^{237}Np	100.0	100.0
^{234}U	/	0.003	^{239}Pu	46.4456	56.873			
^{235}U	/	0.404	^{240}Pu	34.1212	26.997	^{241}Am	82.2386	82.118
^{236}U	/	0.010	^{241}Pu	3.8448	6.104	^{242*}Am	0.2764	0.277
^{238}U	/	99.583	^{242}Pu	11.8500	7.693	^{243}Am	17.4850	17.605
			^{243}Pu	/				
			^{244}Pu	0.0012		^{243}Cm	1.5405	1.533
						^{244}Cm	69.8455	69.763
						^{245}Cm	26.5103	26.588
						^{246}Cm	2.0618	2.074
						^{247}Cm	0.0372	0.039
						^{248}Cm	0.0047	0.003

To develop this analysis the “heterogeneous” approach, with different MA contents, has been investigated assuming that for any investigated configuration the MOX + MA fuel is considered only in the high Pu-content zone (ELSY outer fuel zone) at the same geometry and operative constraints: the Pu content is adjusted only in that enrichment zone, in order to restore (if necessary) the initial steady-state reactivity level of the ELSY reactor. No other geometric or material or operating condition variation was introduced. The last assumption, generating the previous one, is a crucial point for this study.

For the neutron analysis, both steady-state and BU cycle evaluations, the MCNP5/MCNPX codes [5,6] and the JEFF-3.1 neutron cross-sections libraries [7] have been used.

ELSY core performances

It has been mentioned that no other geometric or material or operating condition variation have been introduced in this study, excepting the substitution of the third zone MOX fuel of the ELSY core with:

- MOX fuel at: MA/(U + TRU) = 10 and 20 wt.%.
- U-free inert matrix (Pu, MA) fuel at: MA/(U + TRU) = 12.83 wt.%.

Table 2 collects some of the main neutron analysis results, as well as the per cent variations with respect to the nominal configurations, by comparing the two options: MOX fuel at 20 wt.% in MA content and the U-free inert matrix fuel at 12.83 wt.% in MA content. With respect to the TRU the two options correspond to a MA content MA/(Pu + MA) of: 28.129 and 33.851 wt.%, respectively. The materials content is express in wt.%, the masses in kg and the burn-up in MWd/kg HM. Regarding the percentage variation of the $P_{Fract/zone}$ and $BU_{EoL/zone}$ items, the values shown in Table 2 are the maximum observed for all the three fuel-zones and the upper scripts in parenthesis are referred to the respective fuel-zone number. The observed variations of the neutron investigated parameters, with respect to the reference configurations, are not negligible; some of them (total average neutron flux, spectral parameters, etc.) could be considered as being acceptable. Of course for a better understanding and explanation of some of these discrepancies,

Table 2: ELSY core neutron performances with MA in the third fuel zone

	MOX 20% MA het.	Δ (%) ELSY	Δ (%) EFIT	U-free inert matrix	Δ (%) ELSY	Δ (%) EFIT
$C(Pu)_{aver.}$	17.829	+1.278	/	22.216	+26.20	/
MA/MA+Pu	28.129	/	-48.20	33.851	/	-37.66
MA_mass kg	2 457.48	/	-16.04	3 296.10	/	+32.74
k_{eff}	1.03018±0.00025	/	/	1.03215±0.00024	/	/
Aver. $\Phi_{n/cm2s}$	1.783E+15	-6.893	+48.34	1.796E+15	-6.214	+49.42
n-$source_{n/s}$	1.330E+20	+1.604	+275.7	1.343E+20	+2.597	+279.4
$P_{peak\ FA\ (MW)}$	13.919	+11.99	/	14.473	+16.45	/
$P_{Fract/Zone}$	0.248/0.315/0.436	+7.921(3)	/	0.247/0.324/0.430	+6.436(3)	/
$BU_{EoL/Zone}$	62.7/77.8/92.5	+21.39(3)	/	65.1/79.7/179.2	+135.2(3)	/
$\Delta k/k_{BU,Equil}$	-0.00627±0.00037	|45.43|	/	-0.00944±0.00037	|17.84|	/
ΔM_{MA_Equil}	-179.2	/	|11.37|	-274.7	/	|70.73|
ΔM_{MA_EoL}	-647.4	/	|45.92|	-999.0	/	|125.2|
$\Delta M_{(kg/TWh)}$	**-9.853**	**/**	**-77.59**	**-15.21**	/	**-65.40**
$Aver.E_{n,(MeV)}$	0.4218	+6.731	/	0.4320	+9.322	/
Aver. ν	2.9697	+1.715	-4.360	3.0033	+2.866	-2.196
Aver. α	1.3185	+8.859	-12.83	1.3305	+9.849	-12.03
Conv. factor	1.2018	+26.17	-26.57	1.2418	+30.37	-24.13
β_{eff}	0.00300±0.00035	-11.77	/	0.00314±0.00036	-7.647	/
$\Lambda_{fis\_lifespan\ sec}$	0.8521 E-6	-27.12	/	0.8375 E-6	-28.37	/
$A_{D\ 1200->1800\ K}$	-0.00393±0.00088	|48.90|	/	-0.00439±0.00095	|42.91|	/
$vk/k_{AC-Void}$	+0.04767±0.00037	+23.21	/	+0.05193±0.00041	+34.22	/

we must take into account the different sizes and architectures, as well as the operative conditions of the two systems. Concerning the safety parameters, the obtained results show a reduction of the reactivity feedback coefficients, reduction of delayed neutron fraction and faster kinetics. This situation imposes a verification of the feasibility of the proposed solution from safety viewpoint.

Figures 3 and 4 show the comparison between the two systems: ELSY (core loaded with MOX at 20% MA and U-free EFIT fuel) and EFIT, concerning the BU reactivity loss and the BU rate. It is interesting to observe that the BU reactivity loss per cycle of the ELSY core is about three times higher than that of the EFIT core. This is the direct consequence of the fact that this last core has been designed with the constraint of $\Delta K_{BU} \approx 0$, [2,4]. From design point of view the constraint of $\Delta K_{BU} \approx 0$ is equivalent to:

- nearly constant proton beam current (for the accelerator);
- nearly constant quantity of Pu in the core during the cycle – the system power is produced by burning MA.

Both conditions mentioned above mean a better neutron economy and a safe operating cycle.

Transmutation performances

Regarding the transmutation rate aspect, this study confirms the general statement that increasing the MA loaded in the core, with respect to their equilibrium concentration, increases the MA transmutation rate. This means that the MA transmutation performances of the ELSY core become appreciable when the MA amount, loaded in the core, is higher than the equilibrium concentration amount. The comparison of the MA masses variation is performed only with respect to the EFIT reactor, because the ELSY reference configuration at the BOL is MA-free configuration (ELSY produces about 120 kg of MA during five years of irradiation). It should be

Figure 3: ELSY and EFIT K_{eff} BU swing

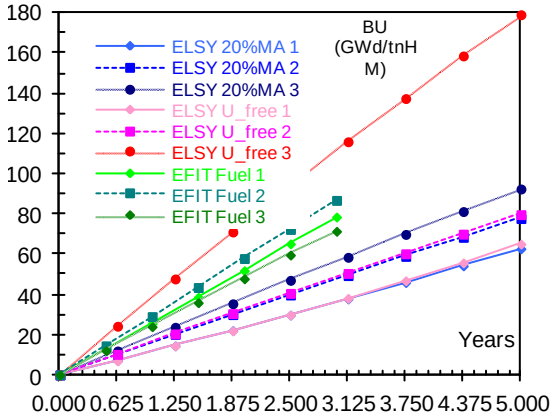
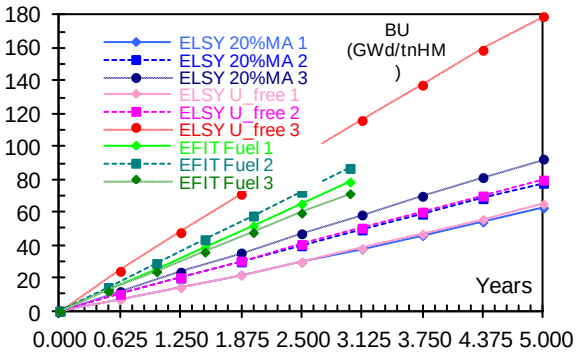


Figure 4: ELSY and EFIT BU rate



emphasised that the ELSY core loaded by EFIT fuel (having about 20% more MA than the MOX 20% MA fuel) is characterised by a MA transmutation rate per unit of energy produced of about 55% higher than the ELSY core loaded by MOX 20% MA fuel.

The transmutation capabilities of both systems have been investigated. Figure 5 shows the fuel inventory during five years irradiation cycle for the ELSY core loaded with MOX at 20% MA and U-free EFIT fuel, respectively, while Figure 6 shows the same fuel inventory for the EFIT core during its three-year irradiation cycle. The results in Figures 5 and 6 show a significant consumption of Am: between ~35 to ~39% of the initial amount in MOX at 20% Ma and U-free

Figure 5: ELSY core mass inventory

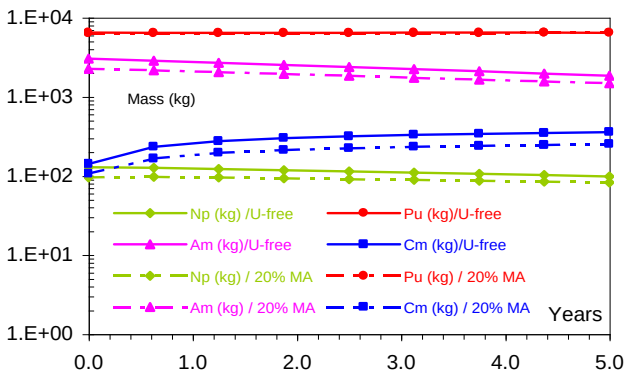


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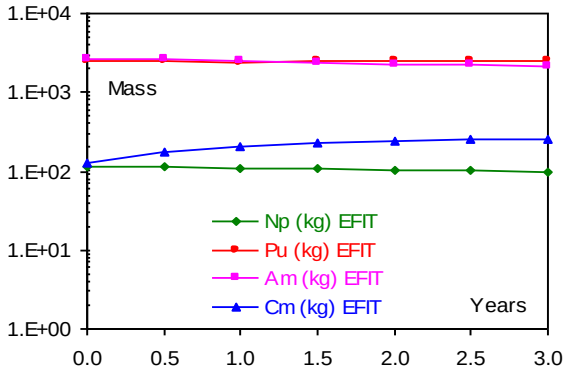
Figure 6: EFIT core mass inventory



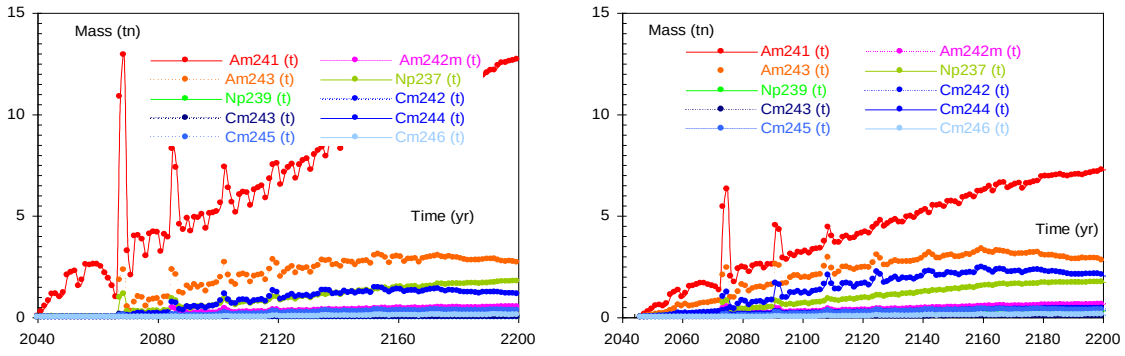
EFIT fuel, respectively, for the ELSY core. On the contrary an Am consumption of ~20% in the EFIT core is observed. In terms of mass reduction per unit of energy produced the results become between -11.87 to -18.01 kg/TWh for the ELSY core, instead of -55.78 kg/TWh for the EFIT core. Detailed results are collected in Table 3. These results show the MA transmutation efficiency of the EFIT core which has been in fact designed for this purpose. The same results show that MA recycling means firstly ^{241}Am transmutation. Figure 7 obtained from scenario studies results involving both thermal and fast reactors with MA recycling, clearly displays this situation. Another aspect of the transmutation of the MA is related to the fact that without particular provisions in the neutron design, e.g. in the choice of an appropriate enrichment, the MA transmutation is accompanied by Pu production whose impact on the fuel cycle should be evaluated.

Table 3: MOX 20% MA het and U-free inert matrix transmutation performances

ELSY and EFIT cores

	20% MA het. solution	U-free inert matrix fuel	EFIT core inert matrix fuel
MA mass (kg)	2 458	3 296	2 927
ΔM_{Np} (kg)	-12.953	-29.83	-13.81
ΔM_{Pu} (kg)	+117.72	-14.97	+0.240
ΔM_{Am} (kg)	-780.01	-1182.95	-562.92
ΔM_{Cm} (kg)	+145.54	+213.14	+133.04
ΔM_{Np} (kg/TWh)	-0.197	-0.454	-1.368
ΔM_{Pu} (kg/TWh)	+1.792	-0.228	+0.222
ΔM_{Am} (kg/TWh)	-11.87	-18.01	-55.78
ΔM_{Cm} (kg/TWh)	+2.215	+3.253	+13.18

Figure 7: MA isotopic composition; MA recycling in fast reactors

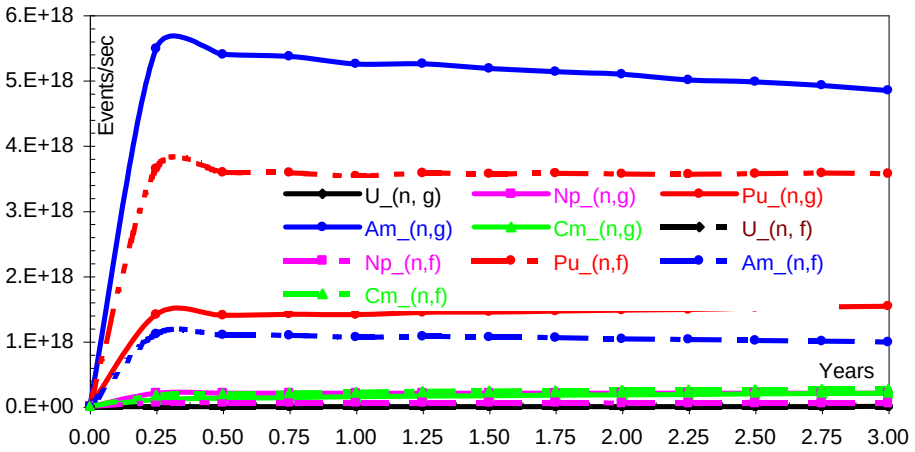


One of the most significant results in Table 3 concerns the Cm build-up. In terms of mass production per unit of energy produced, the following results have been observed for the ELSY core: +2.215 kg/TWh for MOX at 20% MA and 3.253 kg/TWh for U-free EFIT fuel. On the contrary, the EFIT core produces high amount of Cm: +13.184 kg/TWh. Nuclear reactions leading to the Cm production are the following:

$95Am^{241}(n,\gamma)95Am^{242}$ (83.8%) or $95Am^{241}(n,\gamma)95Am^{242m}$ (16.2%)	From EFIT evaluations
$95Am^{242} \rightarrow 96Cm^{242} + \beta^-$ (83.2%) or $94Pu^{242} + \beta^-$ (16.8%)	16.04 h
$95Am^{242}(n,\gamma)95Am^{243} \rightarrow \alpha + 93Np^{239} \rightarrow 94Pu^{239} + \beta^-$	7364.98 y and 2.355 d
$95Am^{243}(n,\gamma)\ 95Am^{244(m)} \rightarrow 96Cm^{244} + \beta^-$	10.1 h (26 min)
$95Am^{244}(n,\gamma)\ 95Am^{245} \rightarrow 96Cm^{245} + \beta^- \rightarrow \alpha + 94Pu^{241} \rightarrow 95Am^{241} + \beta^-$	2.05h & 8 500.19 y & 14.33 y

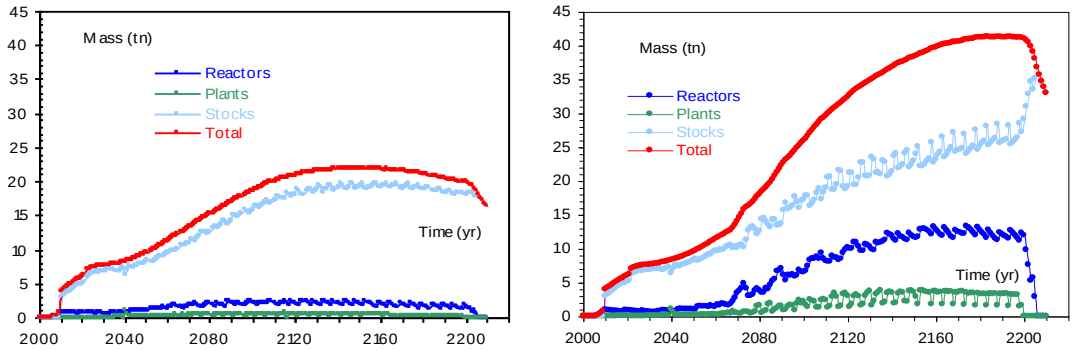
Figure 8 shows the capture (n,γ) and fission (n,f) reaction rate (events/sec) for the EFIT fuel in the outer zone, which has been used to feed the ELSY core.

Figure 8: MA capture and fission reactions rate (events/sec) EFIT third ring inert matrix fuel



Factors of about 3 and 0.2, between fissions and captures, have been observed for Pu and Am, respectively, putting in evidence that the MA transmutation is a complicated matter. Figure 8 results suggest that the Am consumption is due to the capture events rather than to the fission events, supporting the already mentioned reactions and through them the Pu and Cm production. Figure 9, deriving also from scenario studies involving both thermal and fast reactors, displays the Cm behaviour during the scenario period without and with MA recycling in fast reactors.

Figure 9: Cm inventory without and with MA recycling in fast reactors



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It should be noted that this result does not meet radiotoxicity and thermal load reduction expectations. On the other hand the EFIT mass balance (based on the maximum transmutation rate or "42-0" concept) is also achieved through the above Cm behaviour.

Conclusions

MA recycling capabilities and neutron performances on the square wrapper-less ELSY and on the hexagonal wrapped EFIT cores have been investigated.

The main result is that the MA management by their transmutation in fast reactors is a more complex problem. The choice between fast subcritical and critical system will be outcome of some considerations on the neutron performances, safety and technological aspects.

As expected, a significant MA transmutation rate has been observed if the loaded amount is significantly higher than its "equilibrium concentration", while increasing the MA loading in the core degrades the ELSY core neutron performances. The Pu production and mainly the Cm build-up due to the physics and its time constant (many times higher than the irradiation cycle length) become critical issues of the MA transmutation process. Safety analysis has shown a worsening of the ELSY core safety performances, with MA loading. In particular it is confirmed that the degradation of the safety characteristics becomes critical as the transmutation rate becomes significant.

On the other hand, it has been demonstrated that the EFIT core, designed to maximise the MA transmutation rate without Pu production, shows good performances regarding this aim accompanied by a safe behaviour. Of course a strong penalty is represented by the high Cm mass production.

Comparing the performances of a subcritical system specially designed for MA transmutation (EFIT type) and a power reactor (ELSY type), one could observe that the aim of an appropriate MA management, in safe way, is achieved only if the right solutions are adopted starting from the early design stage. This could allow and justify the "transition" from subcritical to critical systems for an enhanced MA management.

Acknowledgements

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