Potential of Fast Reactors for Transmutation of Actinides and Long-Lived Fission Products

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The paper will discuss some results of the work which has been performed within the frame of a contract between the Commission of the European Communities and the Siemens AG on "Transmutation of long-lived radionuclides by advanced converters".

The main items of the paper are:

- Impact of Minor Actinides (MA) recycling on the properties of MOX fueled fast reactor cores with different size.
- Comparison of homogeneous and heterogeneous recycling of MA in fast reactors.
- Comparison of oxide and metal fueled fast reactors with respect to their MA transmutation capabilities.
- Potential of fast reactors for long-lived fission product transmutation.

1 Introduction

The recycling of Minor Actinides (MA) and long-lived fission products in fast reactors with the final goal to reduce their contribution to the long term radiotoxicity of the waste has been studied within a contract between the Siemens AG and the Commission of the European Communities.

Since it is well known that introducing MA into the fuel of a fast reactor results in an increase of the positive sodium void effect (SVE) and a reduction of the Doppler constant, design measures were investigated to limit the deterioration of these important safety parameters to an acceptable amount. These design measures were core size reduction and heterogeneous instead of homogeneous recycling. Additionally, oxide and metal fuel cores were compared with respect to the influence of MA recycling on their core properties. Finally, the transmutation of the long-lived fission products Tc-99, I-129 and Cs-135 has been studied in positions at the core outer boundary and in in-core positions.

2 Boundary conditions and methods of calculations

The main boundary conditions for the definition of the different reactor cores under investigation were taken from the European Fast Reactor EFR. These were mainly

- the maximum fuel burnup of 20 % which is achieved after 6 years residence time with a load-factor of 80 % and 6-batch loading scheme,

- limits for the maximum linear rating of 500 W/cm at beginning of life (BOL) and 400 W/cm at end of life (EOL),

- a plutonium composition (in weight %) of

  Pu-238 : Pu-239 : Pu-240 : Pu-241 : Pu-242 : Am-241 = 2 : 54 : 26 : 10 : 8 : 2

  corresponding to a light water reactor (LWR) discharge burnup of 40 MWd/kg HM,

- a MA composition of Np-237 : Am-241 : Am-243 : Cm-244 = 49 : 37 : 11 : 3

consistent to the same LWR burnup with Uranium fuel.

The main methods of calculations are indicated in View Graph (VG) 1.

3 Investigation of fast reactor cores of different size with homogeneous and heterogeneous MA recycling

Starting from the European Fast Reactor EFR as a basis, three reduced core sizes were considered which were realized by, firstly, reducing the core height from 1.0 m to 0.7 m and 0.5 m with constant core diameter and, secondly, by reducing the core diameter from 4 m to 1.8 m for a constant core height of 0.7 m. The main design data of these cores are listed in the upper part of VG 2 and in VG 3.

The heterogeneous recycling was simulated by implementing 16 target subassemblies (S/A) into the large core no. 1 in VG 3 and 6 or 12 target S/A into the small core no.4. The smear density of the MA-oxide was varied from 100 % down to 66 % and 33 % in order to adapt the total MA content in the core.

In VG 4 the results for the mass balances of the small core and the large one with 0.5 % MA (just the Am-241 linked with the plutonium), 5.4 % homogeneous and 5.4 % heterogeneous MA content are gathered. The following trends can be stated:

- The MA transmutation efficiency in terms of % per cycle and of kg per TWh is very similar in both recycling strategies if for the heterogeneous version the MA production in the standard fuel S/A is also taken into account. This is also illustrated in VG 5 where all calculated values in kg per TWh are given as a function of the MA content. All points fit to a linear dependence without separate influence of core size or recycling strategy. For 5 % MA content a transmutation efficiency of 12 kg per TWh can be deduced which corresponds to the production of about 5.5 LWRs.

- For smaller MA contents the transmutation efficiency is reduced more than proportionally because of the MA production of the standard fuel of about 3 kg per TWh. For 2.5 % MA content only 4 kg per TWh or the production of 1.8 LWRs are transmuted.

- The efficiency in relative terms is larger in the larger cores because of their higher flux level. Again, the values are small for small MA contents and they reach 8 % per year in the large core versus 6 % in the small one. During the S/A life of 6 years total reductions of 36 % in the large cores and 30 % in the small cores can be reached. In the heterogeneous MA S/A the reductions are certainly larger: for both core types values of 10 % per year and 50 % per S/A lifetime can be stated. In any case multiple recycling of MA is necessary for a considerable reduction of the MA.

Some remarks have to be made concerning the build-up of Pu-238 in the MA containing fuel. Even in very hard neutron spectra the production by capture processes in Np-237 and Am-241 with their subsequent decays cannot be completely avoided. Therefore the amount of Pu-238 produced in standard fast reactor spectra can be significant. As an illustration VG 6 shows the Pu-238 fraction in the total Pu as a function of MA content at BOL for different Pu enrichments. The Pu-238 fraction goes up to 15 - 20 % for 15 % MA content with the lower value occuring in case of high Pu-enrichments, which is caused by the harder neutron spectra in the higher enriched cores. In the MA containing target S/As the Pu-238 content even reaches values of 80 %.

Presently a limit of 5 % is set for the Pu-238 fraction in the total Pu. This is caused by the fact that Pu-238 is emitting neutrons by spontaneous fission and α-particles, so that efficient measures for shielding and cooling (Pu-238 produces 560 W/kg) during refabrication and transport are necessary.

If it turns out that recycling strategies with larger EOL contents of Pu-238 are beneficial, a more accurate evaluation of the limiting value would be required. This would not only have to include the Pu-238 content itself, but generally the special isotopic composition of the fuel under consideration, the mass throughput of the plant, special aspects like the influence of subritical multiplication and possibilities to enhance the shielding and cooling efficiency.

Taking the 5 % limit as given for the time being, a BOL MA-content of 3 % should not be exceeded.

In the lower part of VG 4 the results for the sodium void effect and the Doppler constant are given. The fissile sodium void effect (SVE) is strongly increased in the homogeneous cases (30 % in the small core and 20 % in the large core for 5.4 % MA content) and the Doppler constant is reduced by 30 % in both cores. These disadvantages can be avoided to a large extent in case of heterogeneous recycling of MA: the fissile void effect is almost unchanged or slightly reduced compared to the reference case, the total core SVE (standard plus MA S/A) is increasing to a smaller amount (about 10 %) and the Doppler constant is less decreasing. This is an interesting advantage of the heterogeneous strategy.

4 Oxide and metal fueled cores with MA transmutation

For this comparison the EFR-like core no. 2 in chap 3 and VG 3 with 0.7 m core height and a thermal power of 2600 MW was taken. The main core and S/A data were identical for both fuel types with the exception of the fuel data itself with the related smear density (9.2 g/cm^3 for oxide, 11.9 g/cm^3 for metal) and the Pu-contents to adapt the reactivity to the right level (20.0/25.0 % for oxide, 16.4/19.4 % for metal for the inner /outer core zone). The cycle length has been increased from 292 to 352 efpd to keep the maximum burnup about constant.

The efficiency of the MA transmutation has been calculated for a MA content of 5 % in the total heavy metal of both fuel types and for both cases a reference case without additional MA was established. The results concerning the MA transmutation efficiency are gathered in the upper part of VG 7. They do not indicate a clear advantage for either fuel type:

- Due to the larger MA mass at BOL resulting from the higher metal fuel density, the transmuted mass per year is about 24 % higher for metal fuel than for oxide fuel.

- The relative changes per year are however slightly larger in the oxide fuel since differences with respect to microscopic cross-sections and flux level are largely compensating each other. Consequently the transmutation half-time is a little bit smaller in oxide cores.

Some important safety and operational parameters are gathered in the lower part of VG 7. Here the following aspects are important:

- Compared to the oxide fuel the metal fuel shows some specific advantages, which are based on the higher fuel density, i. e. lower Pu-enrichments, lower loss of reactivity per year due to fuel burnup and a longer fuel residence time. Assuming the same number of cycles per residence time for both cores, the absorber system requirements are reduced by about 45 % so that the control rod worth reduction by 13 % is more than compensated.

- The sodium void effect (SVE) deteriorates for metal fuel compared to oxide fuel by about 50 - 60 %. The Doppler constant is reduced by about 50 %.

A valuation of the changes of these important safety parameters is not easy because of their complex influence on the core safety behaviour.

For the sodium void effect the situation is quite clear since its increase is a disadvantage in all types of accidents. The role of the Doppler coefficient, however, depends upon the detailed accident conditions. In cases of unprotected loss of heat sink or loss of flow the smaller Doppler coefficient is an advantage since it reduces the reactivity to be provided by other negative reactivity effects (i. e. fuel expansion, core expansion, control rod drive line expansion) for reactor shutdown. In transient overpower conditions and transients with sodium voiding, however, smaller Doppler coefficients cause more severe accident conditions. Therefore, no clear tendency can be given for the influence of the MA on the core safety behaviour without detailed safety studies.

5 Transmutation of fission products in fast reactors

The fission products Tc-99, I-129 and Cs-135 with half-lifes of $2.1 \cdot 10^5$ years, $1.6 \cdot 10^7$ years and $2.3 \cdot 10^6$ years, respectively, belong to the long-lived nuclides in the high-level waste. Their risk contribution during storage is relatively high because of their pronounced mobility in geochemical environments around the waste repositories. Their transmutation by neutron capture into stable isotopes (Tc-99 $\rightarrow$ Ru-100, I-129 $\rightarrow$ Xe-130, Cs-135 $\rightarrow$ Ba-136) is therefore of interest.

The one-group capture cross-sections of these isotopes are rather small in a standard fast reactor neutron spectrum. They are below 0.5 barn so that in an average neutron flux of $3 \cdot 10^{15}$ cm^{-2} s^{-1} less than 25 % of the fission products are burnt during a fuel subassembly life of 6 years.

One possibility to increase the capture cross-section of the fission products is to place them in a moderated environment in order to soften the neutron spectrum. This effect was studied by introducing $ZrH_{1.7}$ in the target subassemblies containing Tc-99, I-129 and Cs-135 and by varying the $ZrH_{1.7}$ content. VG 8 shows the one-group cross-sections of these isotopes as a function of the $ZrH_{1.7}$ content. The strong influence of the moderator content can be seen, but the fission product content itself has a similar impact. The larger fission product content leads to a spectrum hardening in the target subassemblies so that the capture cross-section is decreasing.

VG 8 is also showing that the cross-section of Cs-135 is small even in case of a large moderator content. Therefore, there is no chance for a transmutation of this isotope in a fast reactor.

The following results are restricted to Tc-99 as the most frequent long-lived fission product. Its transmutation in a $ZrH_{1.7}$ moderated surrounding was studied first in periphal positions and then in in-core positions.

Basis for this study was the EFR core (core 1 in chap. 3, VG 3), in which the radial blanket row was completely replaced by 78 target subassemblies (S/A). For the design of these S/A it was assumed that 20 % of the volume consist of structural material (steel), 40 % are filled with sodium and that the remaining 40 % are left for fission products plus moderator. The fission product content was varied between 2 and 20 % and the moderator content between 5 and 20 %.

For a judgement about the transmutation efficiency the transmutation half-time, which should be as small as possible, and the transmutation rate in kg/a, which should be as large as possible, are two indicators. They are plotted in VG 9 as a function of the Tc-99 content with the $ZrH_{1.7}$ content as additional parameter. It is obvious that the Tc-99 content has a much stronger influence on both indicators than the $ZrH_{1.7}$ content.

If one considers the fact that during burnup of the reactor about 20 kg of Tc-99 are produced per year and that the corresponding value in a LWR is about 30 kg/a, it becomes evident that the transmutation rate should be beyond a value of about 70 kg/a so that the own production and that of two LWRs could be transmuted. This could be possible with about 20 % Tc-99 content and 10 - 20 % $ZrH_{1.7}$ content, but the corresponding transmutation half-time would be 40 - 50 years. This figure, however, is much too large since it would require the irradiation during some hundreds of years to reach a quantitative reduction of the Tc-99 waste. Consequently the use of in-core positions was investigated in the second step of this study.

The basis for this study was core 1 in chap. 3 and VG 3. In this core up to 84 fuel S/A have been replaced by Tc-99 containing S/As. Their position has been varied in order to optimize the radial power distribution.The final configuration with 84 Tc S/As is shown in VG 10. Since the number of fuel S/As is reduced by 84 out of originally 376, the total thermal power was reduced from 3600 MW to 2600 MW. In order to get some margins with respect to the maximum linear rating limits, the core height was increased in some cases from 1 m to 1.2 m. The $ZrH_{1.7}$ content was varied from 0 % to 10 % with a Tc-99 content of 10 % and 20 %.

The main results of these calculations were the following:

- The optimum $ZrH_{1.7}$ content is close to 3 % since larger values cause a stronger flux depression in the target S/As so that the capture rate decreases although the one-group cross-section is further increasing. This increase is, however, less pronounced than in the peripheral positions because the surrounding fuel S/As are mainly determining the spectrum.

- The Tc-99 content has again controversial influences on the transmutation half-time and the transmuted mass per year since both parameters are increasing with increasing Tc-99 content. Typical values for 3 % $ZrH_{1.7}$ are:

	$T_{1/2}$ [a]	ΔM [kg/a]
10 % Tc-99:	19	110
20 % Tc-99:	25	170

In view of the facts that the fast reactor itself produces about 20 kg/a and that one LWR produces about 30 kg/a one can conclude that with 10 - 20 % Tc-99 content the production of 3 - 5 LWRs could be transmuted. The related transmutation half-times are in the range of 20 - 25 years.

The total amount of Tc-99 which would be loaded into the reactor with 84 target S/As and 20 % Tc-99 volume fraction is roughly 6000 kg for a 1100 MW_e reactor.

In order to get an idea about the influence of the moderated Tc-99 subassemblies on the safety parameters of the core, the sodium void effect, the Doppler coefficient and the control rod worth have been calculated for a 1.2 m high core with 84 Tc-99 S/As with 20 % Tc-99 and 3 % $ZrH_{1.7}$ content. An end-of- equilibrium-cycle condition was simulated by adding appropriate fission product concentrations to the fresh fuel. The table below gives the results in comparison to the corresponding values of the 0.7 m high core 2 in chap 3:

	core with Tc-99 (1.2 m high)	core 2 (0.7 m high)
sodium void effect [$]	3.9	4.4
Doppler constant [$]	- 1.2	- 1.6
control rod worth [$]	28	23

Despite the relatively large core height of 1.2 m, the SVE is 12 % smaller compared to the flat homogeneous layout. This is a consequence of the higher Pu enrichment, the moderating effect of the $ZrH_{1.7}$ and of the strong heterogeneity of the core with 84 Tc-99 target S/As. On the other hand the Doppler constant is reduced by 27 % due to the higher Pu-enrichment in the core S/As and the harder neutron spectrum. But this should be acceptable in view of the smaller SVE.

The control rod worth is more than 20 % larger compared to the flatter core 2, which, however, is necessary because a larger burnup reactivity swing has to be compensated.

As a conclusion, one can state that the transmutation of the long-lived fission products Tc-99 or I-129 in special in-core positions seams to be in principal feasible since it is acceptable from the core design and safety point of view. A core with 2600 thermal MW could be loaded with about 6000 kg of Tc-99 and I-129 and it would transmute about 170 kg/a which corresponds to the production of 5 LWRs with 1300 MW_e each plus the production of the fast reactor itself. With respect to these absolute values the result is quite encouraging, but in relative terms it is very disappointing which is expressed by the transmutation half-times of 20 - 25 years for Tc-99 and 30 - 40 years for I-129. For a quantitative mass reduction of these isotopes one would need at least 5 half-times (giving a reduction to 3 % of the initial mass) and this would require an irradiation time of 100 - 200 years. Since the residence time of the target S/As will be not larger than 10 years the fission products have to be recycled more than 10 times. Each time the S/As have to be reprocessed and refabricated and the daughter product Ru-100 of Tc-99 has to be separated from the target material. Therefore, it is highly questionable whether such a recycling concept is technically feasible.

6 Conclusion

The investigations of the capabilities of fast reactors with respect to the transmutation of MA and long-lived fission products have lead to the following conclusions:

- The concepts of homogeneous and heterogeneous recycling of MA have similar transmutation efficiencies: 30 - 35 % of the initial MA content are transmuted during a S/A lifetime of 6 years. Therefore multiple recycling will be necessary for a quantitative reduction of the MA masses.

- Small cores are less efficient than larger cores because of their lower flux level. But they have the advantage to limit the positive sodium void effect, which is increasing with increasing MA content, to acceptable values.

- An upper limit of 5 % MA content is recommended for the homogeneous recycling which limits the deterioration of the sodium void effect and the Doppler effect to an acceptable amount.

- For 5 % MA content a transmutation efficiency of 12 kg/TWh can be deduced which corresponds to the production of about 5 Light Water Reactors.

- Special attention has to be paid to the build-up of Pu-238 in the fuel which reaches 7 % of the total Pu in case of 5 % MA, which requires efficient measures for shielding and cooling during refabrication.

- Heterogeneous recycling of MA is recommended for further studies, since it limits the number of MA containing S/As to about 5 % of all fuel S/As and since it avoids the deterioration of the sodium void effect and the Doppler effect. Measures with respect to the extremely high Pu-238 content have, however, to be taken in this concept.

- The comparison of oxide and metal fueled fast reactors with respect to their transmutation efficiency has shown no clear advantage for one of these concepts.

- The transmutation of fission products has been studied for the two cases where the target subassemblies were located either in peripheral or in in-core positions. It was shown that when using peripheral positions, the masses of Tc-99 and I-129, which can be transmuted per year, are in the range of the production of 2 - 5 LWRs, but the transmuting half-times are rather poor: 20 - 25 years for Tc-99 and 30 - 40 years for I-129 in in-core position whereas they are by a factor of 2 larger in peripheral positions. For a quantitative reduction 5 half-times would be necessary which mean an irradiation time of 100 - 200 years. This indicates that these recycling concepts seem not to make sense.

- Cross sections of ENDF/B–IV and –V, ENDL were used; some isotopes were taken from the ORIGEN2 library "Advanced oxide, LWR–Pu/U".
- Eigenvalue, flux and power calculations were performed in 3d–geometry using the diffusion code DEGEN.
- 26 group spectra were used to produce one–group cross–sections for burnup calculations with ORIGEN2. Burnt concentrations were then used again in flux and power calculations in DEGEN.
- Absorbers were inserted to keep k_{eff} between BOC and EOC almost constant. The relative insertion depths of the inner and outer control rods were adjusted to have almost equal power peaks in both core zones.
- Plutonium contents were adapted so that k_{eff} was about 1.01 at EOC with rods out and that the maximum linear ratings were similar in both core zones.
- Sodium void effects and Doppler effects were deduced via direct k_{eff} calculations.

VG 1 : Methods of calculations

parameter		unit	core 1	core 2	core 3	core 4
thermal power		MW	3600	2600	2000	450
core height		m	1.0	0.7	0.5	0.7
core diameter		m	4.0	4.0	4.0	1.7
no. of fuel S/As		–	375	375	375	84
no. of control rods		–	24	24	24	6
no. of pins per S/A		–	331	331	331	271
fissile	MA=0	$	5.9	4.4	3.0	2.5
void	MA=5%	$	6.5	5.1	–	3.1
effect	MA=15%	$	7.5	6.3	–	4.8
Doppler	MA=0	$	–1.7	–1.6	–1.4	–1.1
constant	MA=5%	$	–1.3	–1.2	–	–0.8
	MA=15%	$	–0.7	–0.6	–	–0.5
$\Delta\rho$ per	MA=0	$	–8.9	–10.4	–12.8	–12.3
full power	MA=5%	$	–4.6	–6.7	–	–8.9
year	MA=15%	$	+1.6	–1.5	–	–4.9
control	MA=0	$	25	23	19	31
rod	MA=5%	$	24	21	–	30
worth	MA=15%	$	20	17	–	27

VG 2 : Design and EOEC performance parameters of cores with different size

EFR – like :

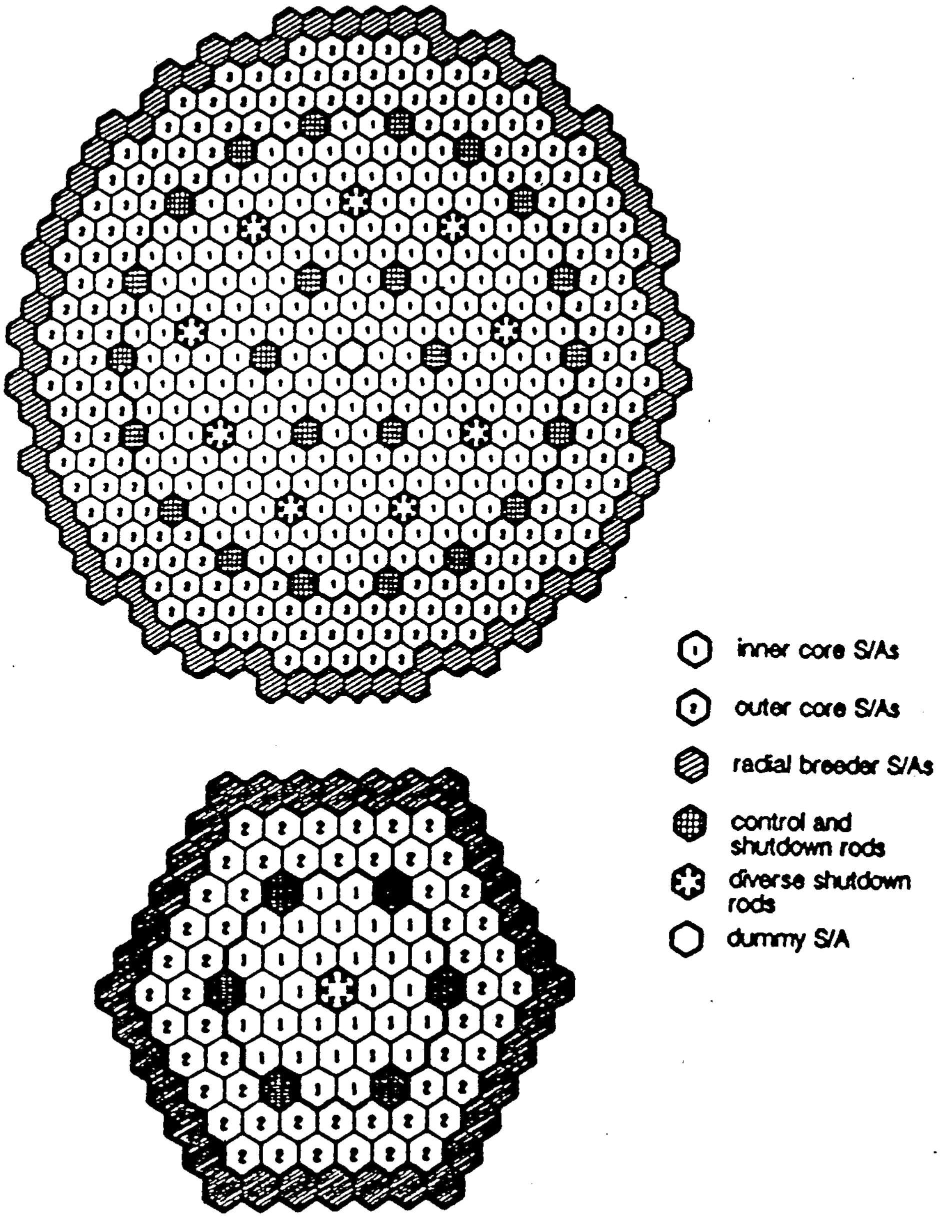
Core 1 :
1 m height,
3600 MW_{th}

Core 2 :
0.7 m height,
2600 MW_{th}

Core 3 :
0.5 m height,
2000 MW_{th}

Small Core :

Core 4 :
0.7 m height,
450 MW_{th}

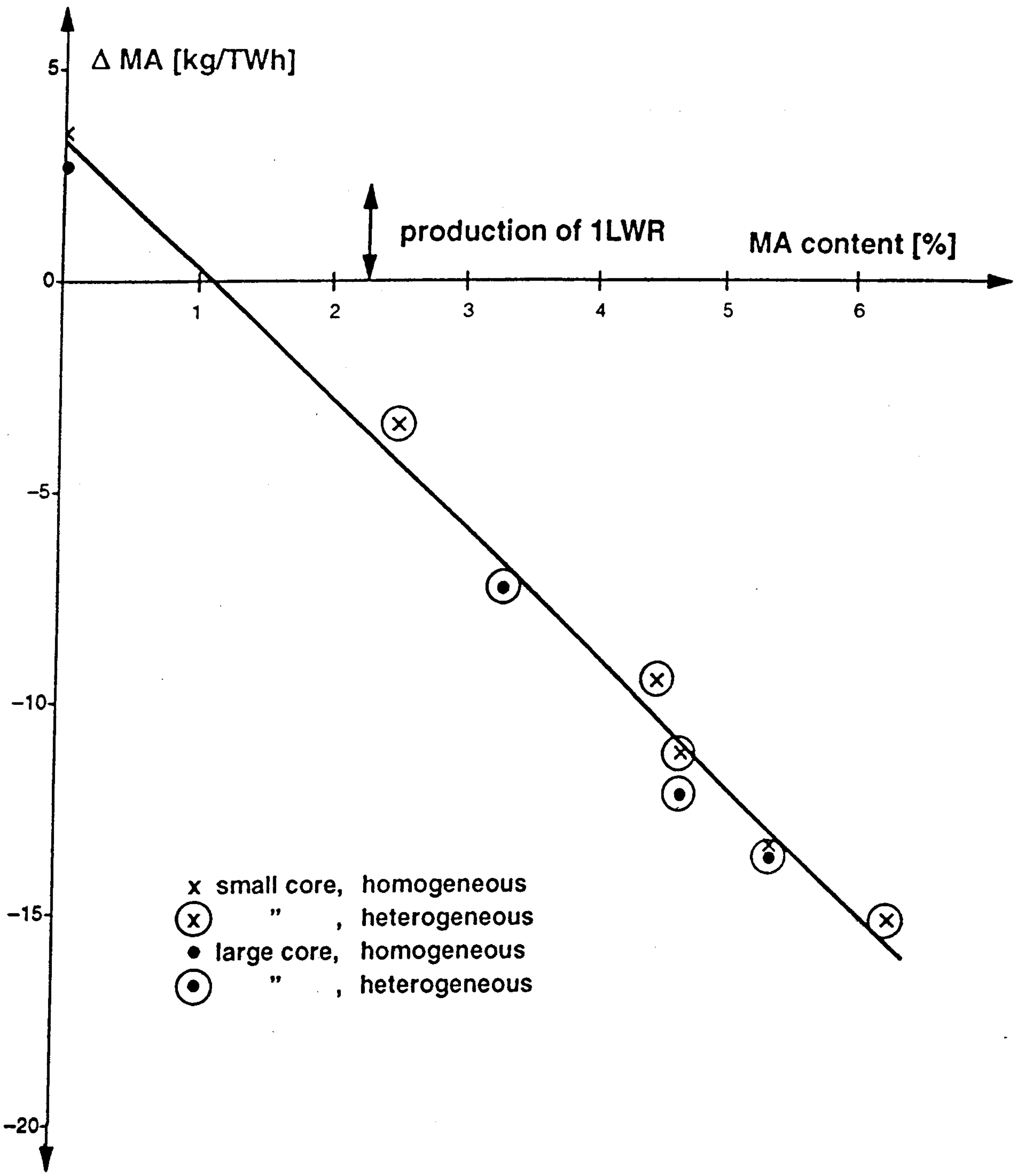


VG 3 : Core layouts with different core size

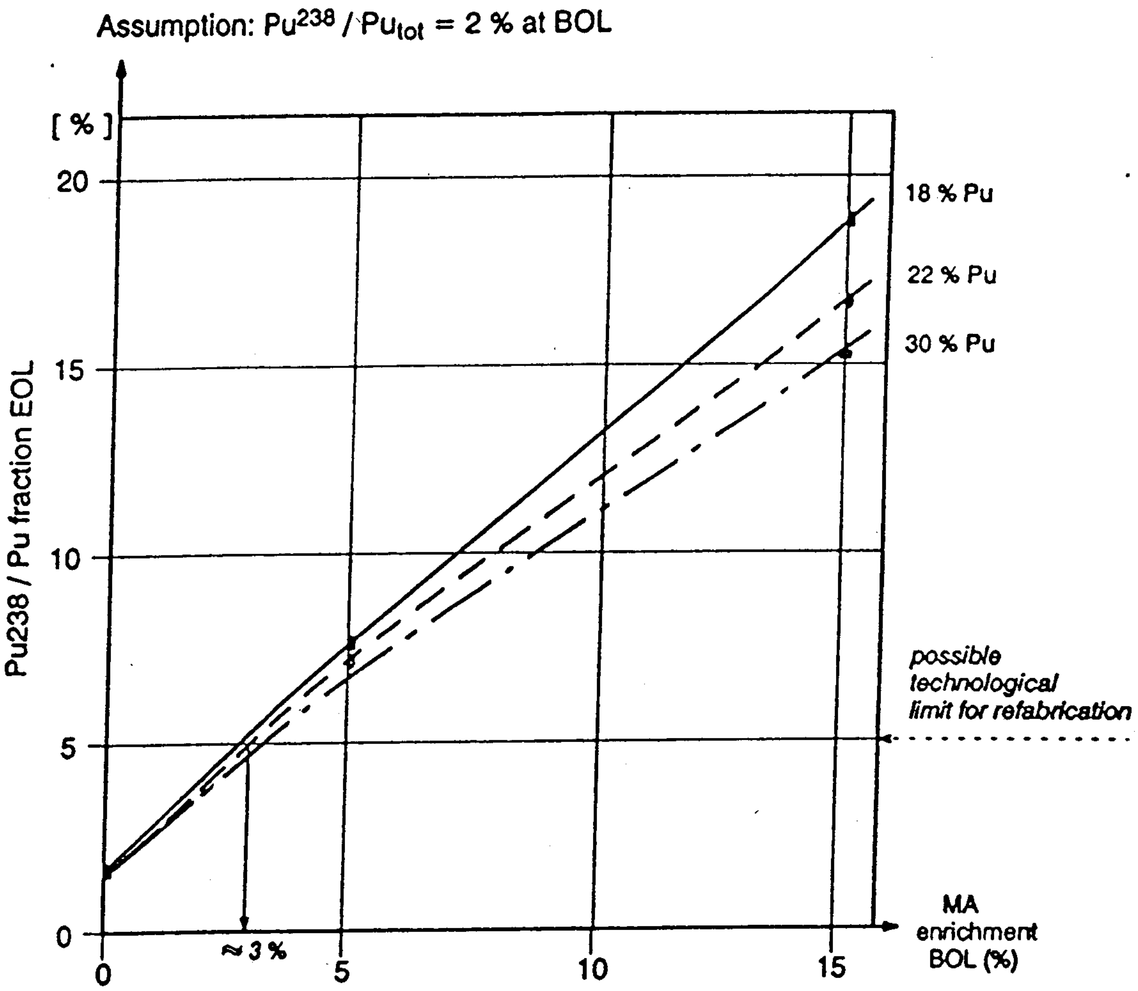
parameter		unit	600 MW core			2600 MW core		
			0.5% HOM	5.4% HOM	5.4% HET*	0.5% HOM	5.4% HOM	5.4% HET*
BOL inventory	Pu	kg	2044	2044	2030	6871	6871	6617
	MA	kg	41	448	460	138	1586	1590
ΔM per cycle	Pu	%	−2.7	−2.2	−2.3	−2.9	−2.2	−2.1
	MA	%	+9.8	−6.2	−5.6	+9.8	−8.0	−8.0
ΔM / TWh	Pu	kg	−31.2	−25.0	−26.8	−26.0	−20.0	−16.0
	MA	kg	+3.5	−13.4	−12.5	+2.7	−13.7	−12.6
$\Delta\varrho_{VOID}$ (total)		pcm	1150	1530	1300	1370	1670	1550
Doppler constant		pcm	−560	−380	−475	−600	−410	−450

* inter– or extrapolated from calculated values

VG 4 : Comparison of core properties of 600 MW_{th} and 2600 MW_{th} Minor Actinides transmuting fast reactors



VG 5 : MA transmutation capacity as a function of MA content



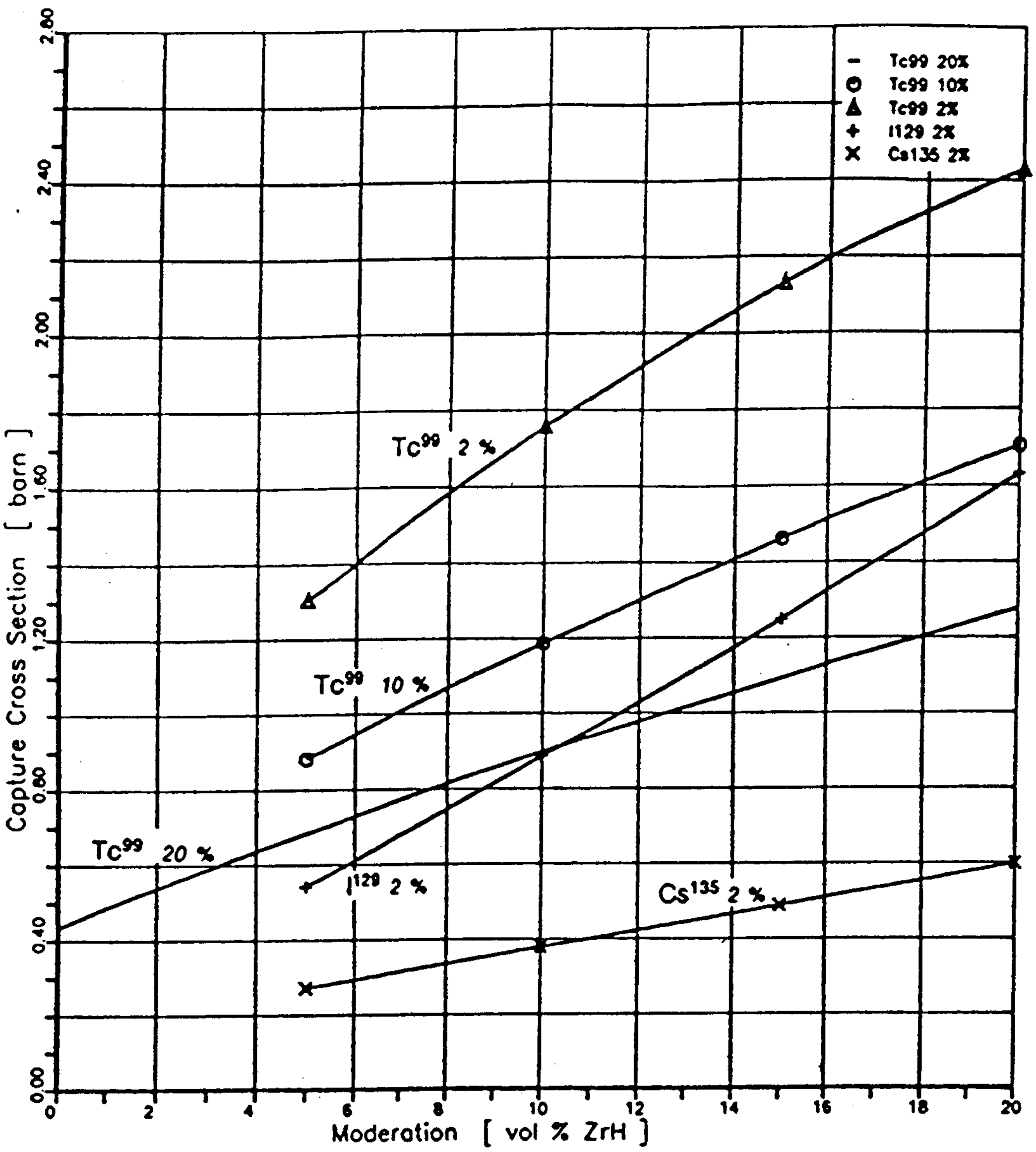
VG 6 : Pu238 generation in MA containing cores

parameter	unit	oxide	metal
MA inventory BOL	kg	1521	2004
MA inventory EOL	kg	934	1121
changes per year	kg	–98	–122
changes per TW_e h	kg	–12.8	–15.9
changes per year	%	–6.4	–6.1
transmutation half–life	a	10.5	11.0
Pu–238/Pu–tot EOL	–	0.078	0.076

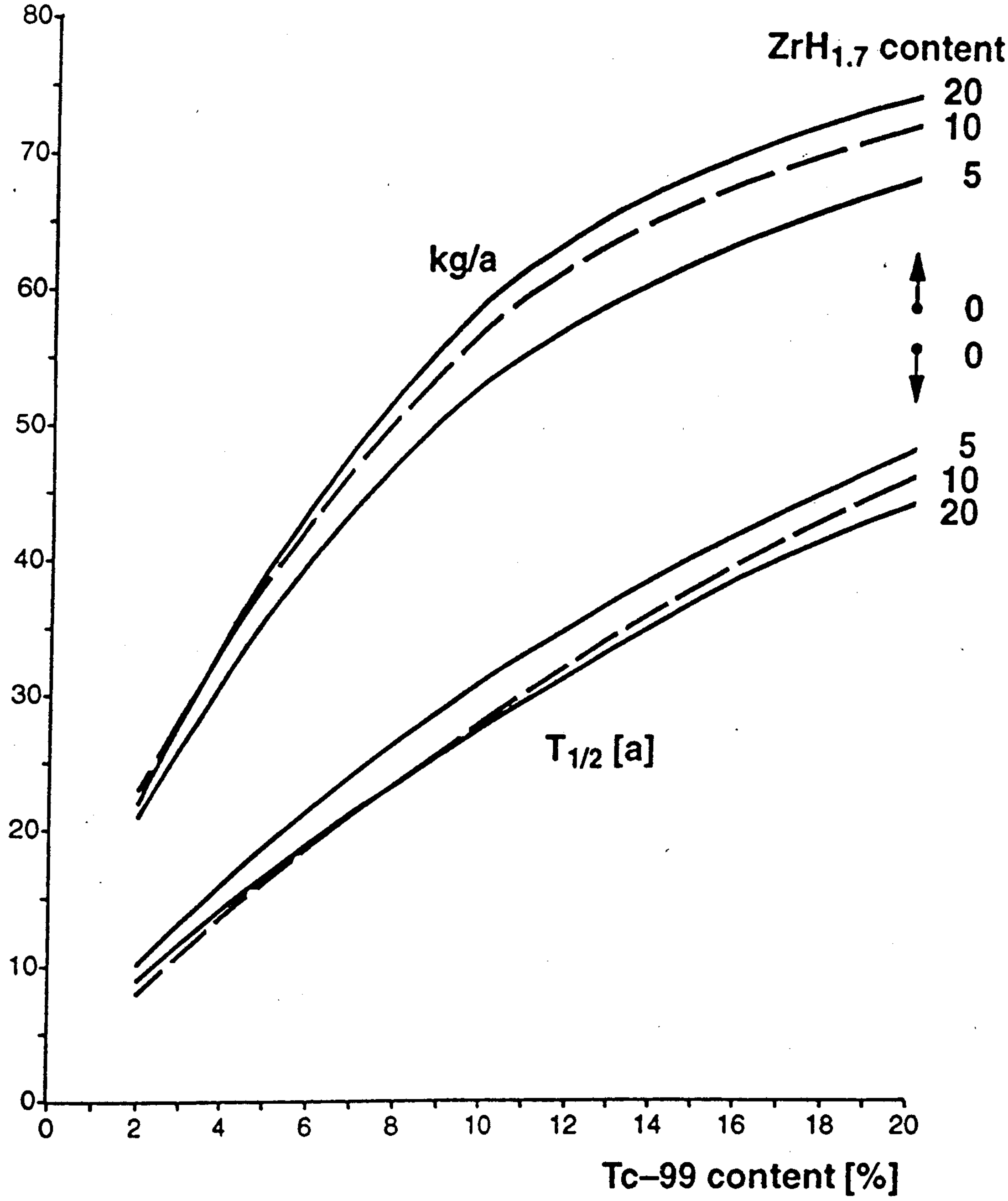
Mass balances in metal and oxide cores of 2600 MW_{th} with 5% MA at BOL

parameter	unit	oxide		metal	
		0% MA	5% MA	0% MA	5% MA
fissile void effect, EOC	$	4.4	5.1	7.4	7.9
Doppler constant, EOC	$	–1.6	–1.2	–0.8	–0.6
reactivity loss per cycle	$	–8.3	–5.4	–4.6	–2.7
control rod worth	$	23	21	20	18

VG 7 : Performance parameters of cores with metal and oxide fuel for 0 and 5% Minor Actinides

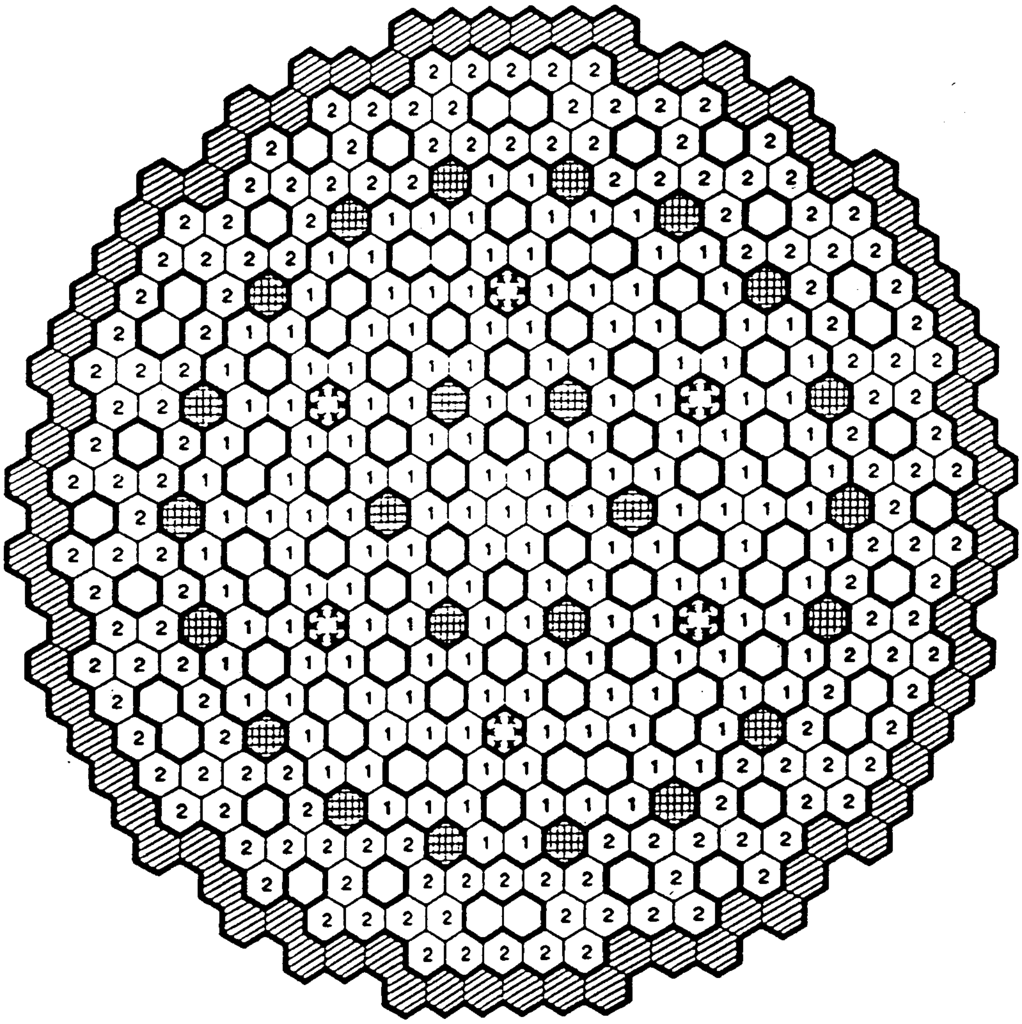


VG 8 : One–group capture cross–sections for Tc–99, I–129 and Cs–135 in dependence on moderator content



Half–life and rate of Tc–99 transmutation in peripheral positions

VG 9



Explanations

- inner core S/As
- outer core S/As
- reflector S/As
- control and shutdown rods
- diverse shutdown rods
- Tc-99 S/As

VG 10 : Cross-section of the core with 84 Tc-99 containing subassemblies