MINOR ACTINIDES TRANSMUTATION IN OXIDE FUELLED FAST REACTORS.

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ABSTRACT

The recycling of minor actinides in fast reactors is considered starting from the EFR type of the reactor, using oxide fuel in which these actinides are homogeneously distributed over the entire core. Burning capabilities of such a core are given, together with consequences on other core characterist its. The effect of the core size on the burning capabilities is detailed, firstly at constant core power, and secondly by reducing significantly the total core power, in order to check the neutron spectrum effect. Comparison of core performance parameters and material inventories are presented and discussed.

1- INTRODUCTION

Feasibility studies concerning a better management of the high level nuclear wastes have been launched in France, enhancing the interest for Minor Actinide (M. A) recycling in Nuclear Reactor. In this respect, Fast Reactors offer potential advantages, which have been studied starting from the European Fast Reactor (EFR) type of core.
The present communication aims at describing parametric studies performed on oxide fuelled cores, in which M. A. are homogeneously distributed all over the fuel. For the EFR type of core, the efficiency of the M. A. transmutation as a function of the initial content is discussed, together with the major consequences on the other core performances and safety characteristics. Then, the effect of the core size and the neutron spectrum hardening on the transmutation performances is developed. Comparisons are given as well for transmutation or burning efficiencies as for the evolution of heavy isotope compositions.
In these studies, only core parameters are discussed: the problems related with partitioning efficiency, fuel fabrication process or safety analysis are not considered.

2- CONTEXT OF THE STUDY

The studies developed hereafter are based on the French nuclear programme, as it stands presently and can be extrapolated to the next decade.
Today, the French nuclear electricity generation rate is somewhat higher than 300 TWh electrical per year, and could grow up to about 450 TWhe per year around 2010, according to some EdF expectations. This nuclear electricity is mainly produced by PWR types of reactors, fuelled with uranium oxide assemblies, the spent fuel being then reprocessed.
Such an electricity generation results in quite large amounts of plutonium and M. A.. With a production of 35 kg of plutonium and 2.5 kg of M. A. at the reactor output per each TWh electrical produced, there will be every year a spent fuel production containing about 10 tons of plutonium and about 1 ton of M.A.

These quantities correspond to uranium oxide fuelled assemblies, the isotopic composition of the M. A. when unloading the fuel from the reactor being approximately :

64 % Np237 ; 8 % Am241 % ; 20 % Am243 ; 6 % Cm244

Concerning plutonium, part of the available amount can be and is indeed already recycled in PWR with MOX fuel, which modifies the M. A. production both in quantity and in isotopic composition. Nevertheless, the quantities involved are still marginal and this effect will not be taken into account in the present studies.

3- PARAMETRIC STUDY OF MINOR ACTINIDES RECYCLING

3.1- **Calculation model**

The EFR core, as described for example in ref.(1), was taken as the reference for the present studies. The main characteristics of this core are :

- the total thermal power: 3600 MWth,
  the homogeneous core concept with two radial enrichments,
- 388 fuel subassemblies, distributed respectively into 208 and 180 between inner and outer zones,
  a fuel pin diameter of the order of 8.5 mm, allowing cycle length larger than one calendar year,
  a maximum fuel burn-up of 20 at. %, corresponding to 1600 efpd fuel residence time, and a 5 batches fuel management.

The fuel is made of mixed oxide UO2-PuO2, containing :

- depleted uranium :0.25 % U235, 99.75 % U238,
- plutonium coming out from PWR spent fuel: 2.8 % Pu238 ; 54.4 % Pu239 ; 22.8 % Pu240 ; 11.8 % Pu241 ; 7 % Pu242 ; 1.2 % Am241 corresponding to plutonium ageing after reprocessing

The corresponding fuel enrichments in plutonium oxide are respectively 16.8 % and 21.9 % for both radial zones, in the absence of any initial M. A..
When adding M. A. inside the core, these new isotopes are homogeneously distributed all over the fuel, replacing heavy atoms of both uranium and plutonium. The respective amounts of uranium and plutonium, or fuel enrichment, are adjusted in such a way to get the same service as for the reference fuel without M. A.: same burn-up, which means same fuel residence time, and same reactivity at end of equilibrium cycle.

The core calculations are performed in a 2 dimension model in RZ geometry, with the ERANOS code system and the JEF 1 cross sections data set. The multibatch fuel management is not explicitly represented: the whole core is burnt completely from beginning of life (all fresh fuel subassemblies) up to end of life (all fuel subassemblies completely burnt). In the following, the wording fuel cycle will represent the overall fuel lifetime, including the out of pile time.

3.2- **Variation of the minor actinide loading**

For the reference EFR core as defined previously, variation of the initial M.A. loading was studied first, in order to determine the burning capability and the consequences on other core characteristics. The corresponding results have already been reported *on* ref.(2), and are recalled here below.

Initial amounts of 2 %, 5 % and 10 % of respectively Np237 and Am241, homogeneously distributed all over the fuel, are loaded in the core. For each individual loading, the following procedure is applied :

the fuel is completely burnt up to 1600 efpd, with the correct fuel enrichment in the sense of fuel service,
the spent fuel is supposed to be reprocessed, with extraction of the fission products replaced by a mixture of uranium and plutonium, adjusted in such a way to get the same fuel service, without adding any new M. A. but only leaving the remaining ones,
the out of pile fuel evolution is simulated by taking 1 year for fuel fabrication and 1 year for fuel cooling and reprocessing,
the same process is repeated for each new fuel cycle, until an equilibrium has been reached.

This auto-recycling procedure allows to quantify the burning speed or burning rate for each of the two isotopes Np237 and Am241, which are two of the main contributors among the M.A. and the easiest to separate and to handle.

The results of this autorecycling show that almost half of the initial amount is burnt after one single cycle, whatever is the initial amount. Also, it is shown that an equilibrium state is obtained after about 5 to 6 cycles, the final isotopic composition being independent of the initial composition: after this recycling time, the isotopic composition is given by the core characteristics, and by the equilibrium between creation and destruction of each individual isotope.

The evolution of the M. A. amount as a function of the number of cycles is illustrated on figure 1, for the case of Np237 and for different initial amounts.

Concerning the other core characteristics, the introduction of M. A. inside the fuel has some other consequences, among which the most important are as follows :

reduction of the reactivity swing with burn-up, which corresponds to improved breeding characteristics,
reduction of the control rods efficiency,
increase of the sodium void reactivity of the fissile zone (about + 20 % on sodium void reactivity for a M. A. amount of 2 %), and corresponding y decrease of the Doppler coefficient.

The improvement of some of the core performances such as the reactivity swing are counterbalanced by the worthening of the safety related parameters, which imposes a limitation on the M.A. content in the core. For an EFR size of core, this limitation is very low, around 2 to 2.5 %. Moreover, the final content of Pu238, coming out from the initial Np237 amount, gives also a limitation on the initial M.A. amount.

For both these reasons, the initial M. A. amount in the fuel has been set to 2 % of the heavy atom mass for the following.

3.3- **Variation of the core size**

When defining the reference EFR core, hereafter called version O, different criteria were taken into account, concerning mainly safety and economics. The resulting core is characterized by some major parameters such as :

a large core volume (about 12.9 m3 for the fissile zone), allowing for the large core power (3600 MWth),
- a core cycle length larger than 1 calendar year (320 efpd), a mean fuel enrichment of about 19.2 %, associated with a fuel volume fraction of about 36 %.

All these parameters are more or less linked together, and will determine the burning capabilities of the core when adding a given amount of (M. A).
In order to check the influence of the core characteristics on the burning capabilities, two other core variants were studied :

variant 1, with the same core power (3600 MWth), but with a much smaller fuel pin diameter, allowing for a core volume reduction by about 40 %,
variant 2, with a reduced core power (560 MWth) together with a fuel pin diameter reduction, leading to a core volume of about 1.32 m3.

On table 1 are given the main core characteristics of the 3 variants. For the three cores, the main dimensionning conditions have been kept constant, especially :

same peak linear rating,
same maximum fuel burn-up, and same reloading frequency,
same reactivity margin at end of equilibrium cycle.

Going from variant O to variant 1 gives mainly the effect of increasing the fuel enrichment: the core volume reduction is coupled with a fuel lifetime reduction at constant fuel burn-up, which corresponds to almost the same annual flow of fuel through the reactor. For variant 2, there is also a very strong reduction of the core volume, which will enhance the effect of neutron leakage together with the effect of fuel enrichment. In terms of annual flow of materials, there is obviously the effect of the total core power.
For both core variants 1 and 2, other modifications will occur such as increased reactivity swing with burn-up, which could result in increased reactivity requirements: this was not taken into account in the present study.

For all three cores, the fuel evolution is calculated first with the standard fuel without M. A. at beginning of cycle, and then by inserting homogeneously the same percentage of 2 % of Np237 and then Am241.

The heavy atom mass evolutions are given on table 2, for the same energy produced and for the fuel at the reactor output. The main results are the followings :

- concerning the M. A., for the sum of the three main isotopes Np237, Am241 and Cm245, the transmutation rate is decreasing with the core size : this is illustrated on figure 2, as a function of the fuel enrichment,
  on the contrary, the burning rate for the plutonium is strongly enhanced when the core size diminishes (fig. 2): this is the well known effect of the internal breeding gain, which is decreasing when the fuel enrichment increases.

This shows that the transmutation rate of the total transuranium isotopes (Pu + Np + Am + Cm) is increasing with the fuel enrichment in plutonium: the breeding gain effect applies to this extended plutonium vector. When looking only to the transplutonium isotopes, or M. A., there is a competition between destruction and production of these isotopes: part of the plutonium is transmuted into M. A., which explains why the transmutation rate of M.A. is lowering when the plutonium content increases.
The enhanced burning capability of the core when increasing the fuel enrichment comes also from the neutron spectrum hardening. To illustrate this effect, the one group microscopic cross sections of some typical isotopes are reported on table 3, for the three core options and for the outer enrichment zone. The ratio between fission and capture is increasing by about 6 % for Pu239 and 12 % for Np237 and Am241 when going from the smaller enrichment (21.86 %) up to the higher one (34. 22 %). Nevertheless, this spectrum effect is quite small as compared to the previous one: the burning potential is multiplied by a factor of about 3, while the spectrum effect is of the order of some 10 %.

When reducing the core size, either by the fuel pin diameter at constant core power or by reducing both the core power and the pin diameter, many other characteristics will be modified. It is the case in particular for some safety related parameters, for example the sodium void reactivity coefficient whose value is very dependant on the core volume. Enhanced safety margins can occur from core volume reduction, thus allowing to increase the M. A. content inside the fuel ; this would improve the transmutation potential of a small core as compared to a large one, for a given energy production. Of course, such a statement would require to perform a complete safety analysis, not limited to the only sodium void coefficient.

4- EVOLUTION OF THE HEAVY ISOTOPES COMPOSITION

As shown in the first part of this study, a significant reduction on the M.A. inventory needs to recycle several times the different products coming out from the reprocessing, as well for the M. A. as for the plutonium. So, Fast Reactors must allow for multi-recycling of plutonium and M. A., especially for what concerns the isotopic composition of the fuel, but also with respect to the reactor itself.
In order to check the capability of the studied cores for the multi-recycling, a study was performed applying the following procedure for the three core variants :

the fresh fuel contains mixed oxide whose enrichment in Pu02 is adjusted with respect to the reactivity at end of equilibrium cycle, and with a given amount of M. A. (2 %),

- the spent fuel is supposed to be reprocessed, by extracting only fission products replaced by fresh fuel, the remaining heavy isotopes staying in the fuel.
- the added plutonium and M. A. isotopic compositions are typical from spent PWR fuel,

Such a procedure allows to follow the isotopic composition of the fuel in a closed Fast Reactor fuel cycle, while taking into account other constraints such as for example maximum burn-up and end of equilibrium cycle reactivity.

After 5 to 6 fuel cycles, and equilibrium is obtained for the isotopic composition, which depends on the core variant. This is shown on figure 3, where the plutonium compositions are expressed in terms of equivalent Pu239, for the three core variants. At equilibrium, the plutonium has the following composition :

- Variant O :

  3.7 % PU238 ; 48.7 % Pu239 ; 36.3 % Pu240 ; 5.5 % Pu241 ; 5.8 % Pu242

- Variant 1 :

  3.9 % PU238 ; 45.2 % Pu239 ; 37.6 % Pu240 ; 6.2 % Pu241 ; 7.1 % Pu242

- Variant 2:

  4.8 % Pu238 ; 37.9 % Pu239 ; 40.5 % Pu240 ; 6.8 % Pu241 ; 10.0 % Pu242

As already shown for the M. A., an equilibrium composition is obtained when, for each individual isotope, creation equals destruction, which depends only on the core characteristics: whatever is the initial fuel composition, with or without M. A., the equilibrium will be the same. This is one of the most important advantage of the Fast Reactors, which can accept any type of plutonium, all isotopes being burnt.
As the isotopic evolution of the fuel depends only on the core characteristics, it is possible to establish a simplified model giving this evolution for each type of reactor. Such a model, called “transfer matrix”, has been established on the basis of first order perturbations, with the following approach :

a reference fuel evolution is performed for given core characteristics, and with a given fresh fuel composition containing all heavy isotopes of U, Pu, Np, Am, Cm: the spent fuel has a composition Cref (EOL),
- for each heavy isotope individually, an elementary perturbation de is introduced in the fresh fuel, the fuel evolution giving a new spent fuel composition: C’ (EOL).

The perturbated spent fuel composition can be expressed in the following matrix form :

C’(EOL) = Cref (EOL) + (T)* de where (T) is the “transfer matrix”, containing as many lines and columns as there are isotopes in the fuel. Such a matrix being representative of a given core, i.e. of a given neutron spectrum, such a matrix has to be built for each type of core. This has been done for the three previous core variants.

In order to check the validity of such a model, comparisons have been performed between exact fuel evolution and evolution simulated with the "transfer matrices", for the three core variants and for different fresh fuel compositions. A very good agreement is obtained for the overall compositions, the discrepancies remaining below 1 % relative for each isotope.

Such a model, whose validity seems very satisfactory, will be used in a more general calculational tool, which simulates the evolution of the material inventories, uranium, plutonium, Minor Actinides, for an overall nuclear park and for different strategies. This model of "transfer matrix" allows to follow the mass balance of all heavy nuclei during the burn-up without any specific core calculation, whatever is the initial isotopic composition.

5- CONCLUSION

The management of high level wastes can be envisaged by several ways, among which the recycling inside Fast Reactors looks quite attractive. Indeed, Fast Reactors allow to fission almost all heavy nuclei, with large number of neutrons being available. Recycling of high level wastes, and particularly Minor Actinides, allows to reduce the corresponding inventories, which contributes in reducing the long term radiotoxicity y. Of course, improvements must be obtained also in the partitionning process and its efficiency, but this aspect was not considered here.

Fast Reactors allow to recycle any type of fuel composition, all isotopes being transmuted: the isotopic composition of the fuel, including Minor Actinides, tends to an equilibrium depending only on the reactor characteristics. It is shown that the burning potential of these reactors is very sensitive to the fuel enrichment in plutonium: a high plutonium enrichment favours burning of plutonium isotopes, but lowers the M.A. transmutation rate. This shows that small sized cores will enhance the plutonium burning capabilities per unit energy, while diminishing the M. A. transmutation rate. On the other hand, small sized cores have larger safety margins, which can allow to insert more easily large amounts of Minor Actinides. So, depending on the burning requirements, either plutonium, or only M. A., or both plutonium and M. A., the optimisation could lead to quite different core characteristics.

In the present studies, only homogeneous insertion of M.A. inside the fuel has been studied: M.A. are distributed all over the core, mixed within the UO2-PuO2 fuel. In a following step, heterogeneous distribution of M. A. will be studied: for example, target subassemblies containing M.A. can be implemented around the core, in the position of the previous breeder subassemblies, thus taking advantage of the large neutron leakage. In this heterogeneous case, there is a net differentiation between the plutonium considered as a fuel and M. A., which was less obvious in the previous homogeneous case.

References

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2- SZTARK, H., VAMBENEPE, G., VERGNES, J., ZAETTA, A., Minor Actinides recycling in an EFR type of Fast Neutron Reactor, Proceedings of a Specialists Meeting held in Obninsk, Russian Federation, 22-24 September 1992, IAEA-TECDOC-693

Figures

1- Evolution of the Np237 masses, for the three initial contents: 2 % 5 %, 10 %.

2- Mass evolution as a function of the mean core enrichment.

3- Plutonium evolution as a function of the number of cycles.

Tables

1- Main core characteristics of the 3 variants.

2- Heavy atom mass evolutions.

3- One group cross sections for the outer core zone.

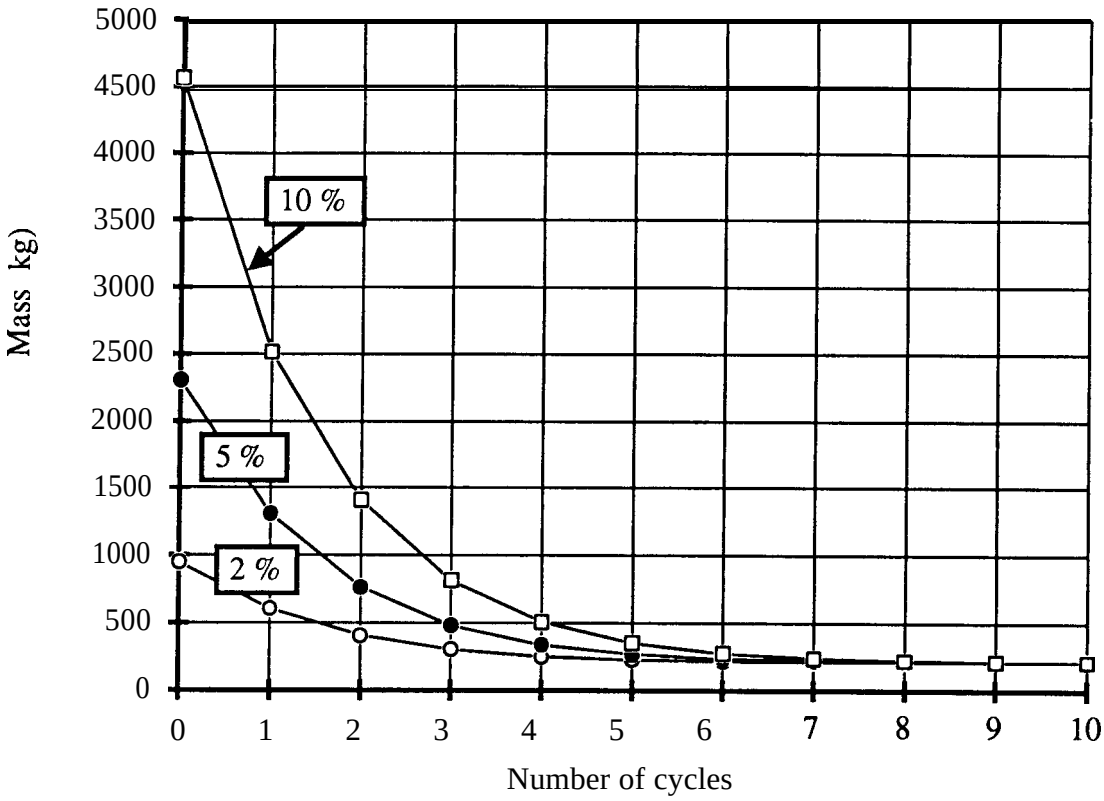

FIG. 1 Evolution of the Np237 masses, for the three initial contents: 2 %,5 %, 10%.

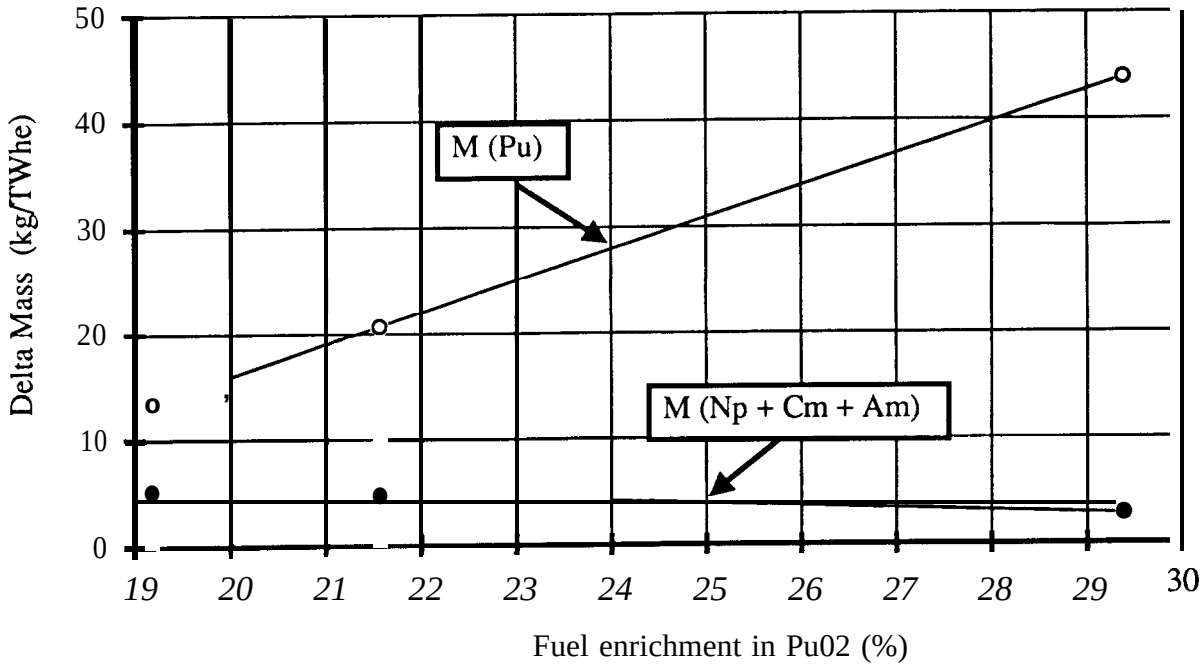

FIG. 2. Masse evolution as a function of the mean core enrichment.

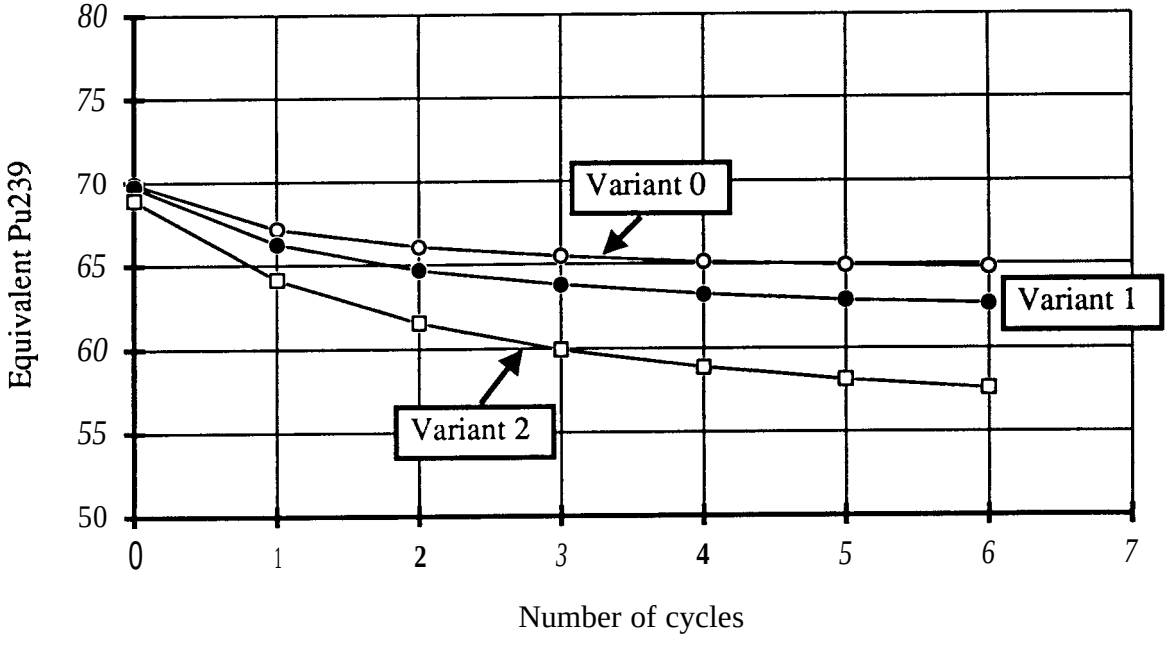

FIG. 3. Plutonium evolution as a function of the number of cycles

TABLE I. MAIN CORE CHARACTERISTICS OF THE 3 VARIANTS.

	VARIANT 0	VARIANT 1	VARIANT 2
Power (MWth)	3600	3600	560
Fuel pin diameter (mm)	8,5	6,5	6,5
Core radius (cm)	202,53	168,97	70,19
Fissile height (cm)	100	100	85
Core volume (m3)	12,89	8,97	1,32
Fuel volume fraction (%)	36	33	33
Mean enrichment (%)	19,17	21,56	29,38
Fuel lifetime (efpd)	1600	980	980
Core cycle length (efpd)	320	196	196

TABLE H. HEAVY ATOM MASS EVOLUTIONS.

	VARIANT O	VARIANT 1	VARIANT 2
Power (MWth)	3600	3600	560
Fuel pin diameter (mm)	8,5	6,5	6,5
Mean enrichment (%)	19,17	21,56	29,38
Pu burnt: core with 2 % Np (kg/TWh)	12,63	19,51	43,71
Pu burnt: core with 2 % Am (kg/TWh)	13,59	20,68	43,92
Total M.A. transmuted			
core with 2 % Np (kg/TWh)	4,98	4,84	2,78
core with 2 % Am (kg/TWh	5,21	4,75	2,83

TABLE III. ONE GROUP CROSS SECTIONS FOR THE OUTER CORE ZONE.

	VARIANT 0	VARIANT 1	VARIANT 2
Enrichment (%)	21,86	24,29	34,22
Pu239 - Capture	0,47	0,49	0,44
Fission	1,74	1,76	1,71
Fis / Capt	3,67	3,61	3,89
Np237 - Capture	1,40	1,42	1,31
Fission	0,37	0,37	0,38
Fis / Capt	0,26	0,26	0,29
Am241 - Capture	1,77	1,79	1,68
Fission	0,30	0,31	0,32
Fis / Capt	0,17	0,17	0,19