NEUTRONIC ANALYSIS OF A PLUTONIUM BURNER PWR PARTIALLY FED WITH INERT MATRIX FUEL

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ABSTRACT

The plutonium coming from dismantled warheads and that already stockpiled coming from spent fuel reprocessing have raised many concerns related to proliferation resistance, environmental safety and economy. The option of disposing of plutonium by fission has been discussed and many proposals for plutonium burning in a safe and economical manner have been put forward. The advantages of utilizing the pressurized water reactors (PWRs) for plutonium disposition are the well developed and reliable technology and their diffusion. The mixide-oxide (MOX) fuel form, that is used for plutonium recycling in power reactors, is well developed. Nevertheless, to eliminate the production of additional plutonium during irradiation, an improved design fuel was analysed, which replace the natural or depleted uranium with inert oxides.

This type of fuel offers the potential for annihilation of the major portion of the plutonium: commercial PWRs operating in a once-through cycle scheme could burn more than 98% of the loaded Pu-239 and more than 73% of the overall initially loaded reactor-grade plutonium. The plutonium still left in the spent fuel was quality-poor and then offered a better proliferation resistance. Power peaking problems could be faced with the adoption of burnable absorbers: zirconium diboride coating in the form of integral fuel burnable absorber (IFBA) appeared particularly suitable. In spite of a reduction of the overall plutonium loaded mass by a factor 3.7, there was not evidence of an increase of the Minor Actinides radiotoxicity after a time period of about 25 years.

INTRODUCTION

In the last years, the problem of disposing of excess plutonium has become more and more a topical and critical issue. As a matter of fact, large amounts of plutonium coming from spent fuel reprocessing have been already stockpiled and over the next decade, under already ratified agreements, significant amounts of weapons-grade plutonium (about 100 metric tonnes) are expected to be recovered from nuclear weapons dismantlement. The plutonium surplus raises concerns about proliferation, diversion and environmental damage. It may be noted that the reactor-grade plutonium could also be used to make nuclear weapons; the higher levels of plutonium isotopes, Pu-240 and Pu-242, only limit the efficiency and the capacity of the material for weapons use.

Therefore, in the present situation, an ever growing plutonium production appears to be no longer the goal and the already stored quantities, those continuously produced from operating reactors and those obtained from weapons dismantlement should be disposed satisfying at least four different goals: *i)* preclude reuse by the superpowers; *ii)* prevent environmental damage from plutonium contamination; *iii)* prevent proliferation from diversion to nonweapons states; *iv)* recover at least part of the economic efforts for plutonium production. In this framework, the option of annihilating plutonium by fission seems to be particularily suitable.

Several different solutions for burning plutonium have been so far proposed and discussed from the viewpoint of safeguards, proliferation resistance, environmental safety, technological background, economy and time schedule (Buckner, 1992), (Buckner and Parks, 1992), (Newman, 1993), (Walter and

Omberg, 1993), (Buckner and Biswas, 1995). The urgency in facing the plutonium disposition issue and the need to maintain the economic viability of the plutonium burning option suggest the utilisation of the already available and operating reactors: then, PWRs appear to be the main candidate for the incineration of plutonium. The fundamental advantage of the pressurized water reactor designs for plutonium disposition is that the technology is mature and has been demonstrated to be highly reliable as proven by performance trends over the last 30 yr.

In order to avoid any further plutonium production during irradiation, a new improved design oxide-type fuel has been recently proposed (Cerrai and Lombardi, 1992), (Lombardi and Mazzola, 1994), (Akie et al., 1994), (Paratte and Chawla, 1995). The present work aims to study a current technology PWR partially fed with inert matrix fuel, analysing some core effect and cycle parameters with 3-D calculations. The performances of a plutonium burner PWR (Burner Reactor) were compared to those of a standard uranium loaded PWR (Reference Reactor).

FUEL TECHNOLOGY

The inert matrix fuel is based on the dispersion of PuO_2 within a carrier matrix made of inert oxides. The matrix should have good chemical compatibility, suitable thermal conduc- tivity, low absorption cross section and good stability under irradiation. The candidates oxides would be: Al_2O_3, MgO, CaO, Al_2MgO_4, CeO_2, ZrO_2 and so forth.

Three different solutions appears to be particularly interesting: *(a)* solid pellets composed by a mixture of Al_2O_3, MgO and ZrO_2; *(b)* annular pellets made of stabilized zirconia; *(c)* solid or annular pellets made of spinel. The first solution would allow the spent fuel to achieve a stable rock-like form and not to be soluble into nitric acid (Akie et al., 1994), so that it could be buried in deep geological formations without need of vitrification or any other treatment; however, in-pile experience is not available so far. The second solution requires a hollow pellet in order to compensate the low thermal conductivity of the fuel matrix; in spite of some concerns about the Reactivity Initiated Accident, there is a positive experience on the behaviour of zirconium oxide under irradiation (Dastur et al., 1994). The third solution is being widely investigated in the frame of the research on actinides burning; the first irradiation experiments are under way but, to the Authors' knowledge, no results or performance data have been published yet.

In the frame of an agreement between ENEA and Polytechnic of Milan the first pellets have been manifactured based on solution *(a)*. The fabrication of sim-pellets of compositions *(b)* and *(c)* is under way. During this preliminary tests, aiming at the assessment of the inert matrix, the plutonium oxide was simulated by cerium oxide. The fabrication route was the Gel Supported Precipitation process, which has the main advantage to be a wet process, thus avoiding the fines contamination during manipulation. Two options are being presently investigated, i.e.: *i)* sol-gel of a solid solution of cerium oxide and inert oxides and then sintering; *ii)* sol-gel of cerium oxide dispersed within inert matrix powders and consequent sintering. This latter option would allow a microsphere coating for enhancing fission gas retention.

Figure 1 shows the SEM analysis of a sim-pellet fabricated following option *ii)*. The achieved density was 90% of T.D., being the goal 95%; cerium oxide (white spots) inside the inert matrix structure is well evident.

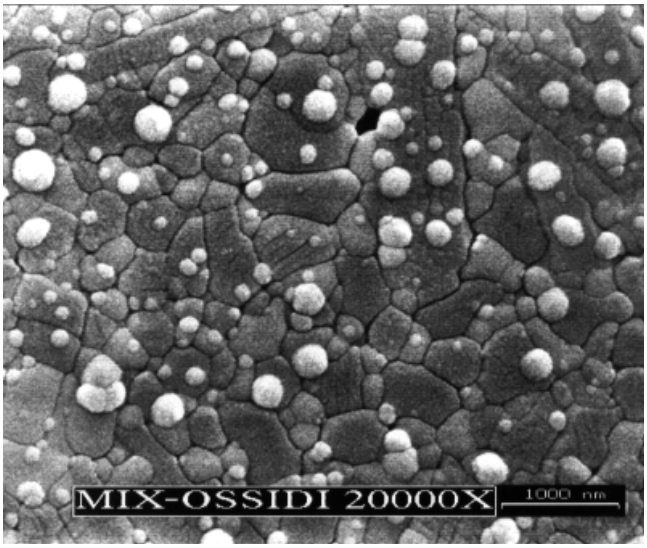

Figure 1. SEM analysis of a sim-pellet.

CORE AND ASSEMBLY CONFIGURATION

Neutronic calculations were performed for a carrier matrix composed by only $MgAl_2O_4$, considering this material as representative of a class of materials with similar neutronic properties such as MgO, CaO, ZrO_2, Al_2O_3. However, the identification of the matrix composition will depend on its thermo-mechanical characteristics and on its in-pile behaviour.

The plutonium considered in this analysis is of reactor-grade type: the initial plutonium vector (Pu-239/Pu-240/Pu-241/Pu-242) was 58%/24%/13%/5%, which is typical for a commercial, 34000 MWd/t irradiated, PWR spent fuel. The content of fissile plutonium was set equal to 0.294 g/cm^3; this figure ensured the fissile plutonium content to be equivalent to the U-235 mass contained in the 3.2% enriched Reference Fuel. Such amount guarantees the UO_2 rods inside the Burner Reactor assembly to achieve approximately the same burnup rate than those in the Reference Reactor (Lombardi and Mazzola, 1995b).

A full loading of inert matrix fuel would be attractive because of the large amount of plutonium that could be burnt. Nevertheless, due to the different neutronic features of plutonium with respect to uranium, such a reactor would have a dynamic behaviour dissimilar from the standard PWRs one, especially as far as reactivity coefficients and delayed neutron fraction are concerned. This fact might affect the reactor controllability, implying the need of additional investments for plant modifications. Possible countermeasures are the insertion of depleted uranium or the utilization of a thorium matrix. In this latter case, the insertion of small amounts of depleted uranium might also be necessary in order to reduce the proliferation potential of the built-up U-233; however, an increased plutonium inventory might be needed in order to achieve the same burnup.

In spite of a reduction of the loaded plutonium per batch, a partial loading solution does not present major problems concerning Doppler or void coefficients, being this concept close to the currently operating MOX-fuelled cores. In fact, previous

work demonstrated that the reactivity coefficients for a partial loading solution were only slightly different from those of a standard PWR (Lombardi and Mazzola, 1995a), (Puill and Bergeron, 1995).

The Reference Reactor for this analysis was an AP600 type, using 3.2% U-235 enriched fuel. This reactor is characterized by a lower power density than conventional PWRs. so that lower centre-line temperatures are achieved. The Burner Reactor was similar to the Reference Reactor, but in the fuel assembly, shown in Figure 2, 56 rods over a total of 264 (-21%) were replaced with inert matrix fuel pins. Fuel and Reactor main parameters are summarized in Table I.

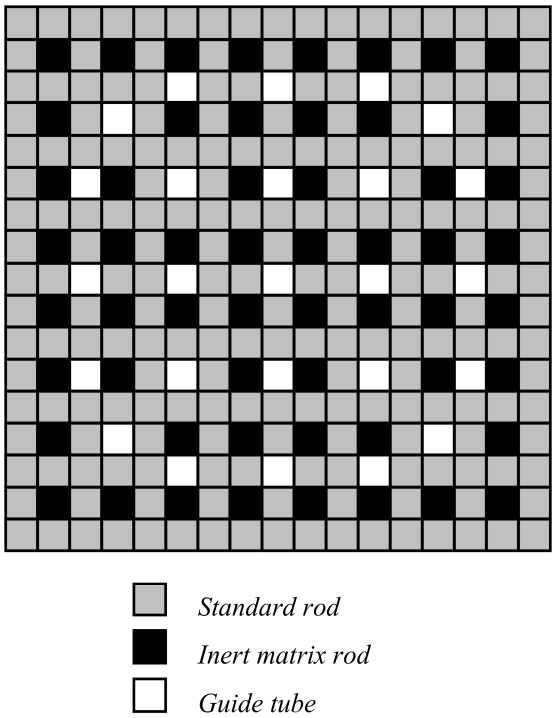

Figure 2. Fuel assembly layout for the Burner Reactor

Table I: Fuel and reactor parameters

Reactor power	2035 MWth
No. of assemblies	156
Assembly layout	17X17
Assembly pitch	215.04 mm
No. of rods per assembly	264
Average linear heat rate	13.5 kW/m
Pin pitch	12.598 mm
Pellet diameter	8.2 mm
Cladding outer diameter	8.356 mm
Moderator average temperature	305 ^{o}C

Due to the presence of a limited number of inert matrix fuel pins, both reactivity swings and reactivity coefficients were verified to be rather similar to those of the Reference Reactor (Lombardi and Mazzola, 1995a).

RESULTS

The CASMO-3 lattice program (Edenius and Forssen, 1992) was used to study the fuel assembly and to produce neutron parameters and cross sections for feeding the 3-D core simulator ABARTH (Facchini et al., 1995).

Because of the almost complete absence of fertile materials, the power generated from the inert matrix fuel steeply decreased during the fuel life. This caused an irregular distribution of the power between inert matrix fuel pins and UO_2 pins. Power peaks were evident in the inert fuel at Beginning Of Life (BOL) while, consequently, the power requirements from the UO_2 rods increased at End Of Life (EOL). Figure 3 shows the evolution of the power generated (normalized to the average one) in the two rods having the peak values at BOL (inert fuel rod) and EOL (UO_2 rod). Effective Full Power Days (EFPDs) were used to measure the irradiation time because of the different overall heavy metal content in the Burner and Reference Reactor fuel assemblies.

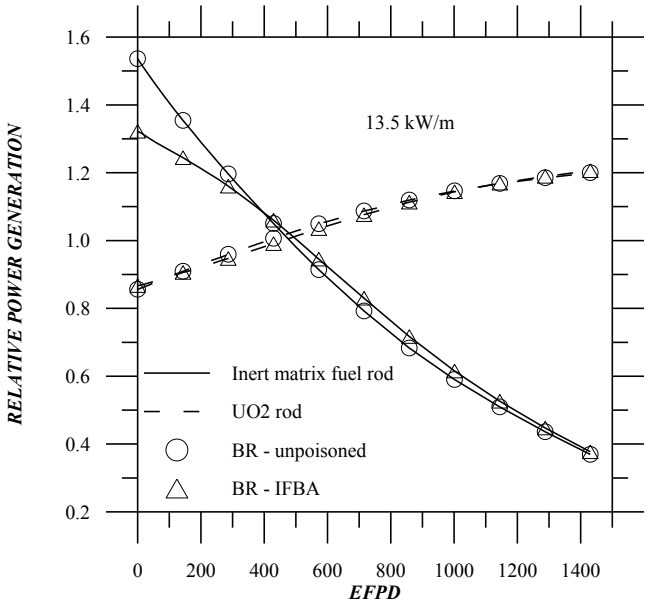

Figure 3. Relative power generation vs. EFPD

The normalized power went from 1.54 (BOL) to 0.47 (EOL) in the inert matrix fuel, while it varied from 0.86 to 1.18 in the uranium rods. Therefore, a high thermal stress would be produced in the inert fuel at BOL and in the standard fuel at EOL. The respect of thermal limits must be carefully verified in order to avoid concerns about fuel rod integrity, fission gas release and so forth.

By running ABARTH, each fuel cycle up to the equilibrium one was simulated. As far as the fuel management is concerned, a 3-batches out-in reloading strategy was adopted.

Objectives of the fuel cycle in the Burner Reactor were: *(a)* obtain an equilibrium cycle length comparable to that of the Reference Reactor, thus allowing the standard UO_2 rods in the Burner and Reference Reactor to reach the same burn-up rate; *(b)* keep the maximum linear heat rate as lower as possible; *(c)* achieve a high consumption rate of the plutonium loaded in the inert matrix fuel rods.

The obtained equilibrium cycle length was 409 EFPDs for the Reference Reactor and 393 EFPDs for the Burner Reactor. The soluble boron concentration in the moderator was, respectively, 945 and 1046 ppm of natural boron.

Due to the high reactivity of the inert matrix fuel rods, the radial core peaking factor of the Burner Reactor resulted to be 1.55, being that of the Reference Reactor 1.47; moreover, it moved from the centre of the core to the peripheral zone, where the fresh fuel assemblies were located, thus increasing the core leakage.

Since ABARTH is able to get a detailed 3-D description of the core, it is worthwhile considering the maximum rod linear heat rate in the whole core along the equilibrium cycle. The core was then discretized in 13 axial cells and the linear heat rate of each fuel rod segment was evaluated. The maximum values for inert matrix and standard fuel are shown in Figure 4.

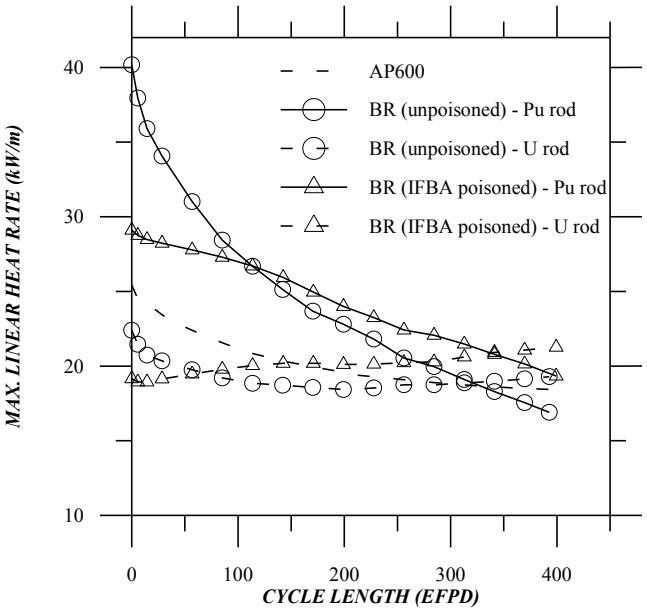

Figure 4. Max. linear heat rate through equilibrium cycle

The maximum values for the Burner Reactor were of 40 kW/m at BOC (inert matrix fuel) and 19 kW/m at End Of Cycle (UO_2 fuel). The corresponding figures for the Reference Reactor were 25 and 18 kW/m.

In order to dampen the peaking factors and then reduce the maximum rod linear heat rate, the utilization of burnable poisons (BPs) was considered, modifying accordingly the shuffling strategy.

The analysed BPs were erbia (Er_2O_3), gadolinia (Gd_2O_3) and zirconium diboride coating in the form of integral fuel burnable absorber (IFBA). The study requirements were: *i)* reduce the peaking factors both inside the assembly and across the core; *ii)* achieve the complete consumption of the BPs within the EOC in order to avoid any reduction of the fuel cycle length for the same initial fissile material inventory; *iii)* lower, when possible, the soluble boron content of the core for improving the reactor dynamic and safety.

Erbia did not appear to be a convenient solution in this particular case. When erbia was inserted only in the inert matrix fuel pins, a significant reduction of the rod peaking factors could be obtained only with high erbia loadings, because of its low absorption cross section; however, erbia was burnt very slowly through the fuel life, thus getting a penalty in the cycle length. On the other hand, the insertion of lower amounts of erbia in all of the rods did not dampen the power peaks across the assembly.

On the contrary, gadolinia was very efficient at BOC but its consumption was too fast, thus restoring the power peaks at EOC. Too high gadolinia loadings were not allowable for reactor start-up concerns.

The best performing BP was IFBA. In the chosen configuration, a layer of 0.06 mm thickness containing 0.223 g/cm^3 of B-10 was smeared on the fuel pellet. With this composition, the maximum power peak of the inert matrix fuel was reduced from 1.55 (unpoisoned case) to 1.32, as shown in Figure 3. As far as the radial power distribution is concerned, the assembly peak went from 1.55 to 1.41 and moved back to the central region of the core, thus reducing the core leakage. The cycle length was 399 EFPDs, slightly increased with respect to the unpoisoned case due to the reduced leakage, and the soluble boron concentration was lowered to 283 ppm of natural boron (Figure 5); let us outline the importance of this reduction for the reactor safety and operation.

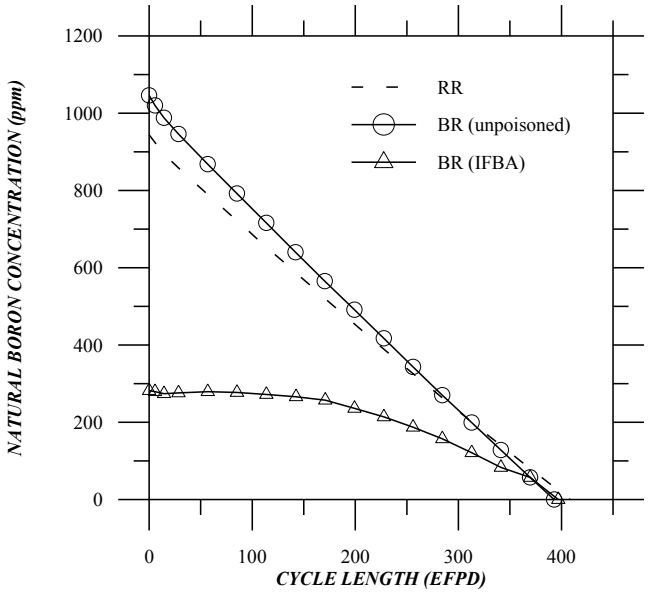

Figure 5. Boron concentration through equilibrium cycle

In spite of a slightly higher maximum linear heat rate at EOC in the uranium rods (21 kW/m against 19 kW/m for the unpoisoned case), the maximum value for the inert matrix fuel rods at BOC was reduced from 40 to 29 kW/m (see Figure 4).

PLUTONIUM DISPOSITION CAPABILITIES

The plutonium isotopic balance in the inert matrix fuel is shown in Table II at BOL and EOL.

Table II: Plutonium mass balance (wt%) at BOL and EOL

	BOL	*EOL*	*burnt*
Pu-239	58	1.06	98.17
Pu-240	24	10.26	57.25
Pu-241	13	6.17	52.56
Pu-242	5	9.27	
fissile Pu	71	7.23	89.82
fertile Pu	29	19.53	32.64
total	100	26.76	73.24
fiss./fert. Pu	2.45	0.37	

More than 98% of the loaded plutonium-239 was burnt at the end of the fuel life, while the burnt fraction of fissile plutonium was 89.8. Finally, 73.2% of the whole loaded plutonium mass was annihilated.

The plutonium still present in the discharged fuel was quality-poor, thus reducing its proliferation potential. In fact, the ratio of fissile over fertile plutonium isotopes was 2.45 at BOL and 0.37 at EOL. These low fissile plutonium fractions in the unloaded fuel should render the spent fuel not attractive for any attempt of plutonium recuperation. It is worthwhile remarking that the overall plutonium mass was reduced by a factor 3.7.

It is also interesting to compare the previous figures with those typical for the Mixed OXides (MOX) fuel (Wiese, 1993). The overall plutonium mass still present in MOX unloaded fuel after the first recycle ranges between 60 and 70% of the loaded quantity, depending on the attained burnup, which means the plutonium burnt fraction be between 30 and 40%. The fraction of fissile isotopes is still dominant: in fact the fissile plutonium percentage goes from about 71% in the loaded MOX fuel to values in the range between 52 and 56% in the discharged one. These figures show the validity of the proposed solution both in terms of plutonium overall reduction and proliferation resistance. However, the MOX fuel technology is well developed, while the present solution must undergo a long technological assessment.

RADIOTOXICITY ISSUES

Notwithstanding the large reduction of the overall plutonium mass which might be achieved by using inert matrix fuel, the issue of radiotoxicity should also be assessed. In the case of plutonium, the reduction of its mass and the increased proliferation resistance are key requirements but, nevertheless, an increase in the overall radiotoxicity might be a serious drawback in the proposed option.

An analysis was then performed to assess whether the option of burning plutonium within an inert matrix would produce any increase in the Minor Actinides (MAs) radiotoxicity. The calculation was performed by using the SAS2H module (Herman and Parks, 1991) of the SCALE integrated code system (SCALE 4.1, 1992).

The radiotoxicity of a mixture of different radionuclides was defined as its *Number of ALI* :

$$Number\ of\ ALI = \sum_i \frac{A_i(t)}{ALI_i}$$

where, for each *i*th radionuclide, $A_i(t)$ is the radioactivity at time t after irradiation and ALI_i is the Annual Limit of Intake through ingestion for the public (ICRP-30, 1984).

In order to have a reference figure, 1 t of 3.2% enriched uranium after irradiation for ~34400 MWd/tHM in the Reference Reactor was considered; the MAs contained in the spent fuel were labelled as *mix a*. In the plutonium burning option, the plutonium included in *mix a* was assumed to be recovered with 2% losses, fabricated as inert matrix fuel and irradiated in the Burner Reactor. The MAs contained in the inert matrix fuel after irradiation, together with those left in the 1 t of spent fuel after the plutonium recovery were indicated as *mix b*. In such a way it was possible to compare the MAs radiotoxicity in the case of the simple storage of the UO_2 spent fuel (*mix a*) with respect to that of the option of plutonium reirradiation as inert matrix fuel (*mix b*). In Figure 6 the decay of MAs radiotoxicity versus the cooling time are shown for *mix a* and *mix b*.

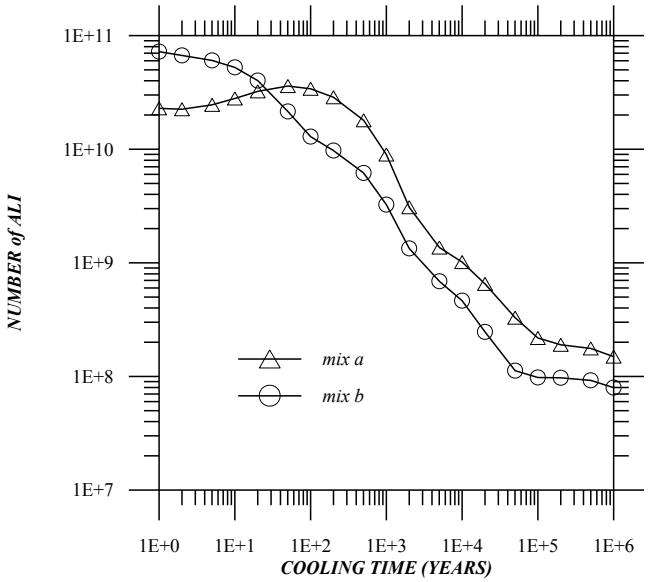

Figure 6. Radiotoxicity decay for *mix a* and *mix b*

The MAs radiotoxicity of *mix b* was larger than that of *mix a* for a period of about 25 years, because of the larger initial contributions of americium and especially curium. The plutonium direct contribution was clearly reduced. After this period, the radiotoxicity of *mix b* fell below that of *mix a*. The difference was about 20% and was due to the lowered direct contribution of plutonium and to the reduced americium build-up. A further reduction of about 20% in the difference of radiotoxicity of the two mixtures is obtained when referring to the produced energy.

As a matter of fact, in the Authors' opinion, such a low difference was probably not meaningful and it could be inside the uncertainties of the MAs cross sections. What is to be stressed is that the significant reduction of the plutonium overall mass and

the increase of its proliferation resistance did not involve any increase in the MAs radiotoxicity with respect to the MAs radiotoxicity of the UO_2 spent fuel. A significant reduction of MAs radiotoxicity is beyond the potentialities of this solution, which was addressed mainly to the plutonium issue.

FINAL REMARKS

The plutonium annihilation capabilities of a current technology PWR partially fed with inert matrix fuel was investigated. The inert matrix fuel was made of PuO_2 dispersed within a carrier matrix composed by inert oxides. Standard UO_2 rods and inert matrix fuel rods were inserted together in fuel assemblies which were uniformly loaded in the core.

An important characteristic of this solution was the irregular distribution of the assembly power between inert matrix fuel pins and UO_2 pins. Due to the fast decrease of the power generation inside the plutonium rods, power peaks were evident in the inert fuel at BOL while the power requirements from the standard fuel rods increased at EOL. The high reactivity of plutonium bearing fuel also caused peaking factors across the core.

The addition of burnable poisons might be envisaged for dampening these two effects. In this particular configuration, the best performing burnable poison was IFBA, which allowed to face the two problems getting at the same time a significant reduction of the soluble boron content of the core.

The inert matrix fuel showed very good plutonium annihilation capabilities: more than 98% of the loaded Pu-239 was burnt and 73% of the total loaded reactor-grade plutonium was consumed, thus reducing the initially loaded plutonium mass by a factor 3.7. The residual plutonium was quality-poor and then unattractive for any attempt of recuperation for misuse.

In spite of the large reduction of the plutonium mass, the MA radiotoxicity did not evidence any increase after some 25 years.

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The Authors worked in cooperation with equal contributions.

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