A ONCE THROUGH SCHEME FOR WEAPONS GRADE PLUTONIUM DISPOSITION IN LWRS: PROLIFERATION AND CRITICALITY ASPECTS

J. MAGILL, H.J. MATZKE, G. NICOLAOU,
P. PEERANI, J. VAN GEEL
Institute for Transuranium Elements,
Joint Research Centre,
European Commission,
Karlsruhe, Germany

Abstract

We investigate a novel scheme for burning weapons grade plutonium (W-Pu) in conventional light water reactors with a view to optimising the destruction rate of plutonium and increasing the proliferation resistance of the resulting spent fuel. The main feature of this scheme is the use of a fuel based on W-Pu and weapons grade uranium (W-U) in an inert matrix. The extended irradiation (þ2000 days) of this fuel leads to a high destruction rate (95%) of the Pu. Proliferation resistance of the spent matrix fuel is guaranteed by the very high levels of ^{238}Pu in the total plutonium (þ30%). The technicalities of how such inert matrix based fuel pins, with lifetime approximately 2000 days, may be used together with standard pins, with lifetime approximately 900 days in a PWR is described together with the criticality changes which may be expected through the use of fuel pins based on the inert matrix. On a timescale of a few hundred years the levels of ^{238}Pu, with half-life of 88.75 years, will decrease to around 5%. High proliferation resistance of the spent matrix based fuel can be recovered by re-irradiating the matrix for a further period of 1000 days. The process of re-irradiation and decay can be repeated, *without reprocessing*, for as long as the spent matrix is considered a proliferation risk.

1. INTRODUCTION

The relaxation in tension between the United States and the former Soviet Union, witnessed over the past few years, has given rise to tens of thousand of nuclear warheads on both sides which have been declared excess to current needs. According to the U. S. National Academy of Sciences report [1] on *Management and Disposition of Excess Weapons Plutonium* published recently, the excess amounts to 50 tonnes of weapons grade plutonium (W-Pu) in the U.S.A. At a recent NATO Advanced Research Workshop on *Managing the Plutonium Surplus: Applications and Options* [2] Russian papers consider the Russian excess of W-Pu to be 100 tonnes. At present the W-Pu and highly enriched uranium (W-U) are present in warheads and will have to be transferred for dismantling. In this process a major problem will arise with regard to proliferation of nuclear weapons to national powers or sub-national groups by theft or unauthorised diversion.

One option is to dispose of this material by ´burning´ it in nuclear reactors and this is the subject of the present paper. The problem with this approach is the fact the even the ´ashes´ resulting from burning of W-Pu (i.e. the spent fuel) can be used to make a nuclear explosive [3]. Following irradiation in the reactor, the spent fuel can be removed from the reactor, dissolved, and chemically processed to separate the plutonium which could then be used to construct a nuclear device. The compositions of different types of uranium and plutonium [3,4] are given in Tables I and II.

Although more difficult than with W-Pu, reactor plutonium (R-Pu) can be used to construct a nuclear explosive. It should be noted that although the critical mass of R-Pu is greater than that of W-Pu (see Table III), *the critical mass of R-Pu is less than that for weapons grade uranium (W-U).* In addition, heat generation in R-Pu is about a factor five higher than in W-Pu and the spontaneous emission of neutrons is about a factor 6 higher than in W-Pu. In Table III a summary of critical masses, heat production and neutron emission rates are given for various isotopes and isotope mixtures.

Table I: Composition (%) of different types of uranium.

	^{235}U	^{238}U
Natural Uranium	0.7	99.3
Weapons Grade Uranium (W-U)	93.5	6.5

Table II. Composition (%) of different types of plutonium.

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
MOX Grade Plutonium (R-Pu)*	1.34	62.77	23.48	8.31	4.1
Weapons Grade Plutonium (W-Pu)	0	94	5.3	0.7	0

* Plutonium recovered from uranium pressurised water reactor fuel with burnup 33GWd/tonne

Table III. Critical masses, spontaneous neutron emission and heat generation rates of various isotopes and mixtures [3,4].

Isotope	$t_{1/2}$ (y)	Bare critical mass (kg)	Spont. Neutron Emission Rate $(kg\ s)^{-1}$	Isotopic Power $(W\ kg^{-1})$
^{235}U	7.04×10^{8}	49	0.4	≅0
^{238}U	4.47×10^{9}	-	15	≅0
^{238}Pu	87.75	9/15*	2.32×10^{6}	560
^{239}Pu	2.41×10^{4}	10/15*	29	1.9
^{240}Pu	6.54×10^{3}	40/60*	9.24×10^{5}	6.8
^{241}Pu	14.4	12/15*	49	4.2
^{242}Pu	3.76×10^{5}	90/177*	1.85×10^{6}	0.1
^{241}Am	432	114	1.55×10^{3}	114
W-Pu	-	≅11	5×10^{4}	2.2
R-Pu	-	15	3.2×10^{5}	10.7
W-U	-	52	1.3	≅0

*α/δ phases of Pu

In this paper we propose a scheme to optimise the destruction rate of W-Pu and increase the proliferation resistance of spent fuel by increasing the relative amounts of the isotope ^{238}Pu present in the 'ash'. The destruction rate of Pu is maximised by embedding the W-Pu in an inert matrix, thereby avoiding the production of Pu from a fertile matrix. Methods for increasing the amounts of the isotopes of ^{238}Pu considered so far require reprocessing of the spent fuel to obtain ^{237}Np or ^{236}U which can then be added to the fresh fuel [5]. In the present approach, we avoid the reprocessing step by using weapons grade uranium as the additional source of ^{238}Pu through the reaction path shown in Fig.1 (the normal source from ^{239}Pu is also shown). This use of weapons grade U requires an extended burnup to avoid proliferation problems associated with the ^{235}U in the spent fuel. Only after about 2000 days irradiation in

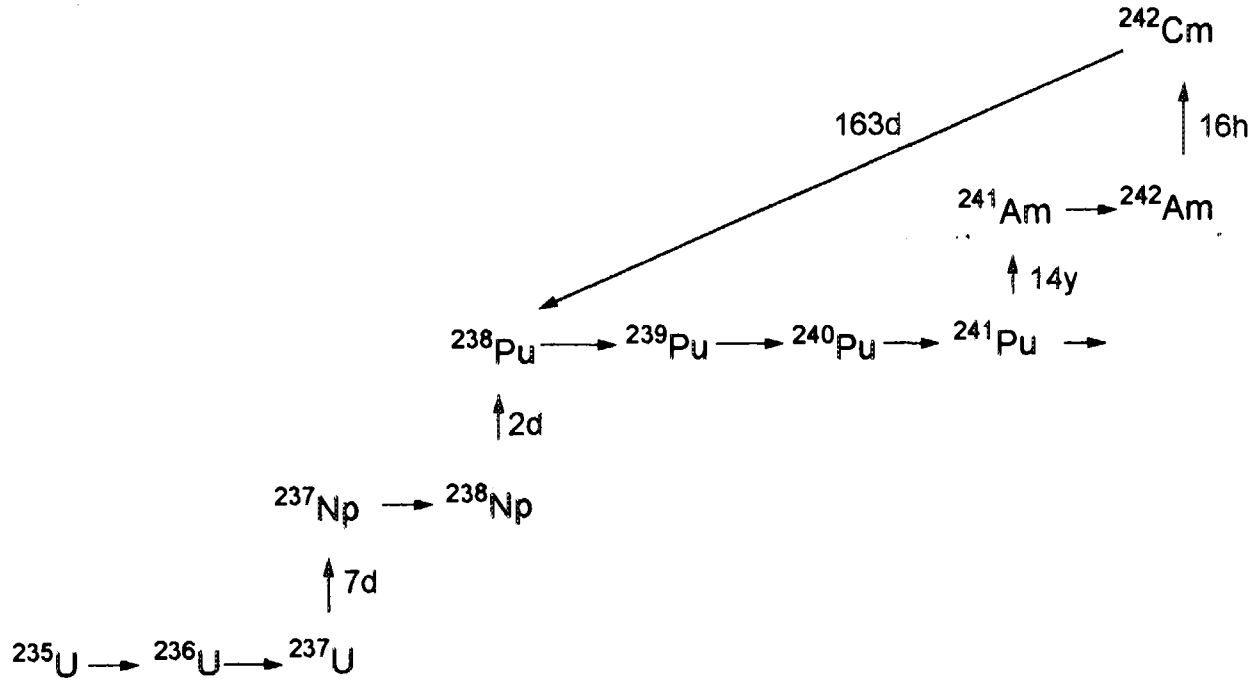

Fig.1. Main pathways for the production of ^{238}Pu from ^{239}Pu and ^{235}U. The sequence of (n,γ) capture processes is interrupted whenever a short lived nuclide is formed.

a PWR neutron spectrum (total flux = $3x10^{14}cm^{-2}s^{-1}$) is the percentage of ^{235}U in the uranium less than 20% i.e. the concentration below which enriched uranium is not considered as a proliferation risk (the bare critical mass of a 20% mixture of ^{235}U in ^{238}U is approximately one tonne [6]). This long irradiation, of course, then leads to a high level of Pu destruction. In the following sections the proliferation and criticality aspects of such an inert matrix containing mixed fissile material are considered.

2. CALCULATIONAL DETAILS

The calculations were made using the point depletion code ORIGEN2 [7] and checked independently with RADONN [8]. The one group cross section library used, PWRUS.lib [7], was that for a standard pressurised water reactor with 3.2% ^{235}U initial fuel enrichment, power level of 37.5 MW/tonne, and burnup of 33 GWd/tonne (achieved after 879 EFPD-effective full power days). To simulate the irradiation of a pin with a different fuel in this reactor, the following procedure was adopted: An initial calculation was made for the standard fuel configuration (i.e. 3.2% ^{235}U, 33 GWd/t) to determine the average neutron flux during irradiation.

Using this average neutron flux and the same one group cross sections (thereby ensuring that the neutron flux and spectrum correspond to a 3.2% ^{235}U enriched fuel with a burnup of 33 GWd/t over 879 EFPD), a new fuel configuration was defined e.g. inert matrix plus fissile material or as MOX, and the calculations were repeated for the irradiation period of 879 EFPD.

A difficulty which arises with the above procedure is that in the initial calculation burnup dependent cross sections are used to account for changes in the neutron energy spectrum brought about by the buildup of additional fissionable isotopes. Depending on the burnup, different cross sections are used especially for the plutonium isotopes. If a different fuel type such as the inert matrix-based fuel is used, the code assumes the core consists entirely of this material. The burnup of this material in the previously defined neutron energy spectrum is clearly different from the standard fuel but it is the inert matrix-based fuel which determines the burnup and therefore which cross sections are to be used.

This problem can be overcome by using burnup independent cross sections. In the present calculations, the burnup independent cross sections have been obtained by linearly averaging the burnup dependent cross sections as shown in Table IV.

Using these averaged cross sections then ensures that the material to be irradiated has no effect on the cross sections used. Clearly, there is no difficulty in extending the calculations to 1757 and 2000 days.

3. PROLIFERATION ASPECTS OF INERT MATRIX-BASED MIXED U-PU FISSILE MATERIAL

We consider the evolution during irradiation of an inert matrix based fuel pin containing 2.5% W-Pu and 2.5% W-U in a PWR. It is assumed, for simplicity, that the inert matrix has the same density as that of the standard fuel. The relevant details of the PWR have been described in the previous section. It is also assumed that the presence of the inert matrix plus fissile material does not disturb the neutron spectrum of the reactor. The results of such an irradiation are shown in Figs. 2 and 3 and summarised in Table V for the plutonium and uranium isotopes respectively. Here the masses of isotopes present per tonne of inert matrix are given at various irradiation times.

As can be seen from Fig. 3 and Table V, an irradiation time of approximately 2.3 cycles (2.3 × 879 days) of the W-U is required to ensure that less than 20% of the total uranium is ^{235}U. This consideration of the proliferation resistance of the W-U thereby fixes the irradiation time for the matrix. During this irradiation period, the plutonium isotope distribution shift almost entirely to ^{242}Pu. The large amounts of ^{238}Pu present originate from the irradiation of the W-U.

Table IV. One group burnup dependent and averaged cross sections for a PWR.

		Cross sections (barns) for fuel burnup (MWd/g-atom heavy metal) of:					Average
Nuclide	Reaction	0.0	2.0	4.0	6.0	8.0	
^{239}Pu	capture	69.09	64.31	59.93	55.85	52.05	60.3
^{239}Pu	fission	121.1	113.7	106.7	100.2	94.07	107.2
^{240}Pu	capture	222.8	176.2	139.4	110.2	87.17	147.2
^{241}Pu	capture	42.02	40.23	38.51	36.87	35.29	38.6
^{241}Pu	fission	125.9	121.4	117	112.8	108.7	117.2
^{242}Pu	capture	33.2	32.4	31.63	30.87	30.13	31.7
^{241}Am	capture	95.7	90.28	85.17	80.35	75.18	85.5

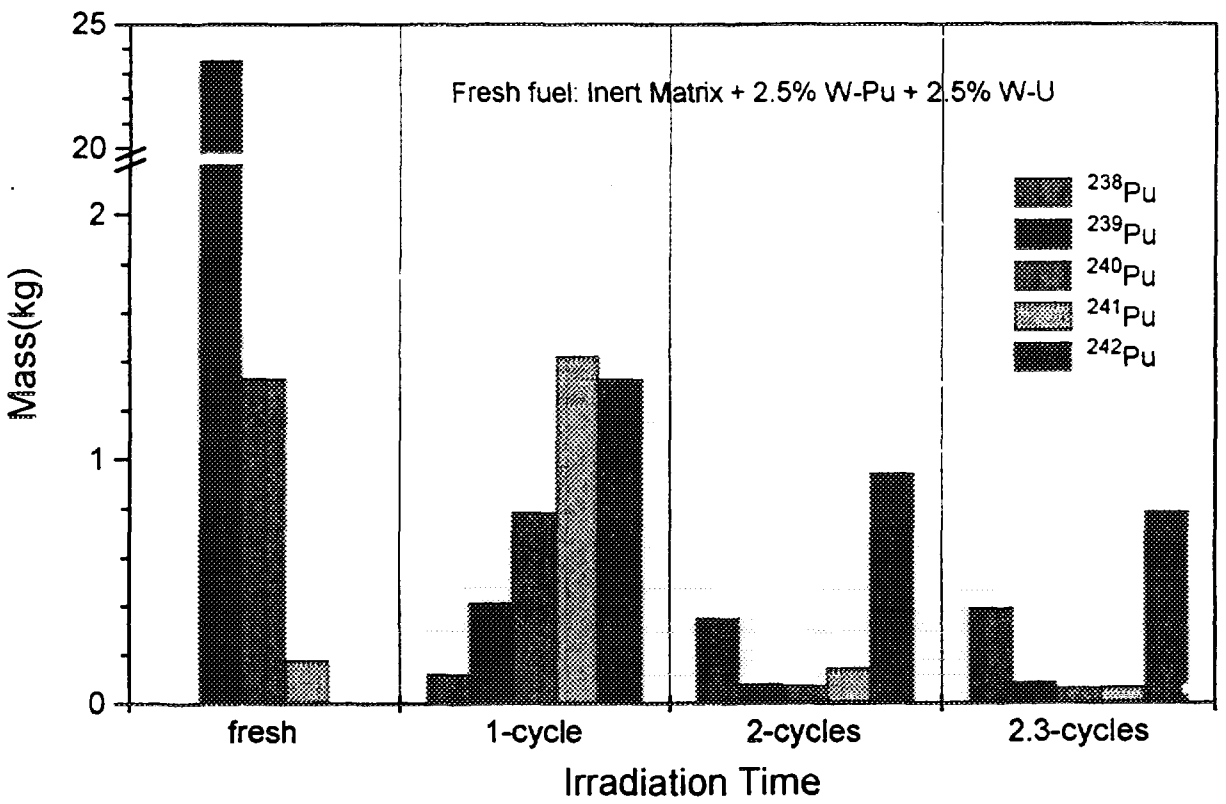

Fig. 2. Pu isotopic masses per tonne of matrix: in fresh fuel, and after 1, 2, and 2.3 cycles in a PWR (1cycle=879days).

Also from Table V, approximately 94% of the total plutonium has been destroyed over 2.3 cycles. What remains is mostly the isotope ^{242}Pu (which from Table III has a critical mass of 100 kg) with approximately 28% ^{238}Pu. If the spent fuel were to be reprocessed to separate out the plutonium, very high heating rates in excess of 150 Watt / kg of Pu metal are to be expected as can be seen in Fig. 4. Since the

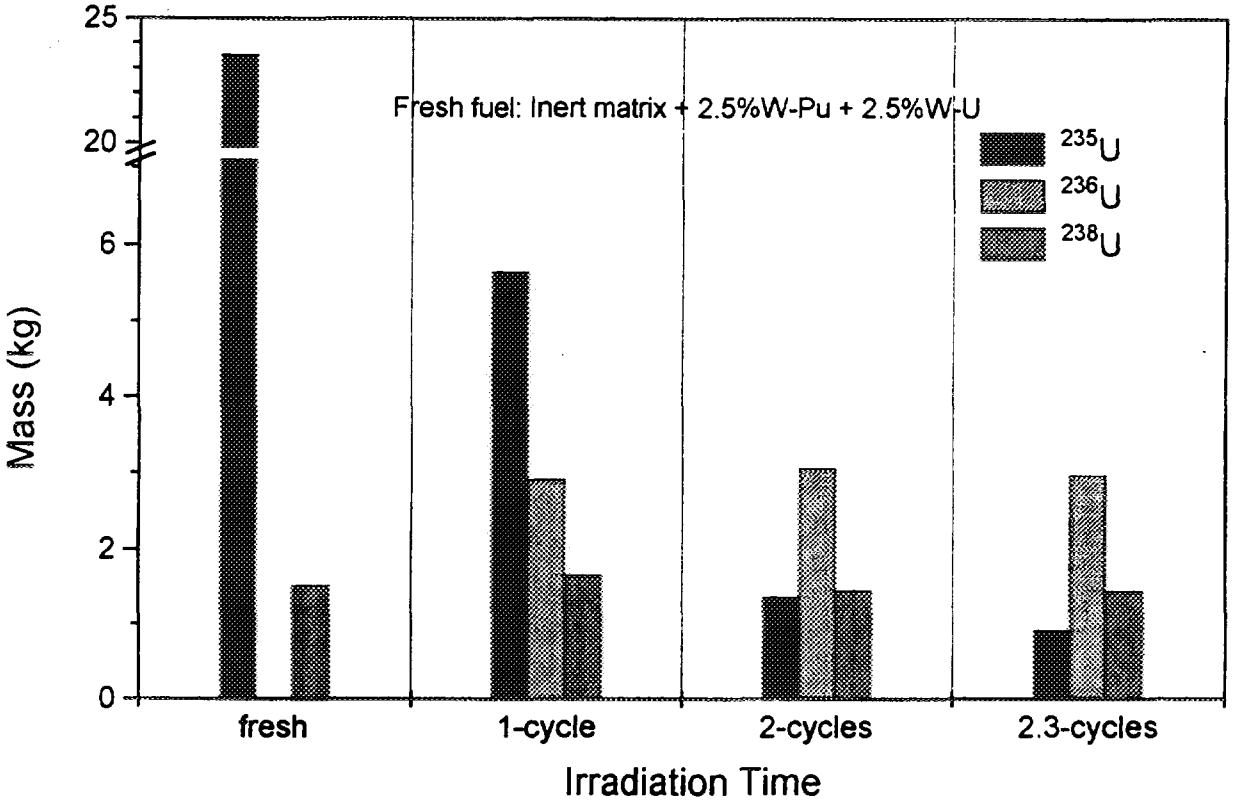

Fig.3. U isotopic masses per tonne of matrix: in fresh fuel, and after 1, 2, and 2.3 cycles in a PWR (1cycle=879days).

Table V. Evolution of an inert matrix containing 2.5% W-Pu+2.5%W-U in the neutron spectrum of a PWR (1cycle = 879 EFPD). Densities of matrix and standard fuel are assumed equal.

	fresh	1 cycle	2 cycles	2.3 cycles
total U	25kg	10.2kg	5.8kg	5.3kg
$^{235}U/U$	94%	55%	23%	17%
total Pu	25kg	4.1kg	1.6kg	1.4kg
$^{238}Pu/Pu$	0	2.9%	22%	28%

critical mass of this mixture is approximately 50 kg [8], heating rates of 7.5 kwatt per device will have to be contended with! Clearly, from this result the amounts of W-U present in the fresh matrix could be considerably reduced.

The neutron emission rates from such chemically separated plutonium are shown in Fig. 5. After 2.3 cycles the neutron emission rate is a factor 32 higher than from W-Pu but only a factor of 2 higher than from R-Pu which is obtained by reprocessing the fuel after 1 cycle.

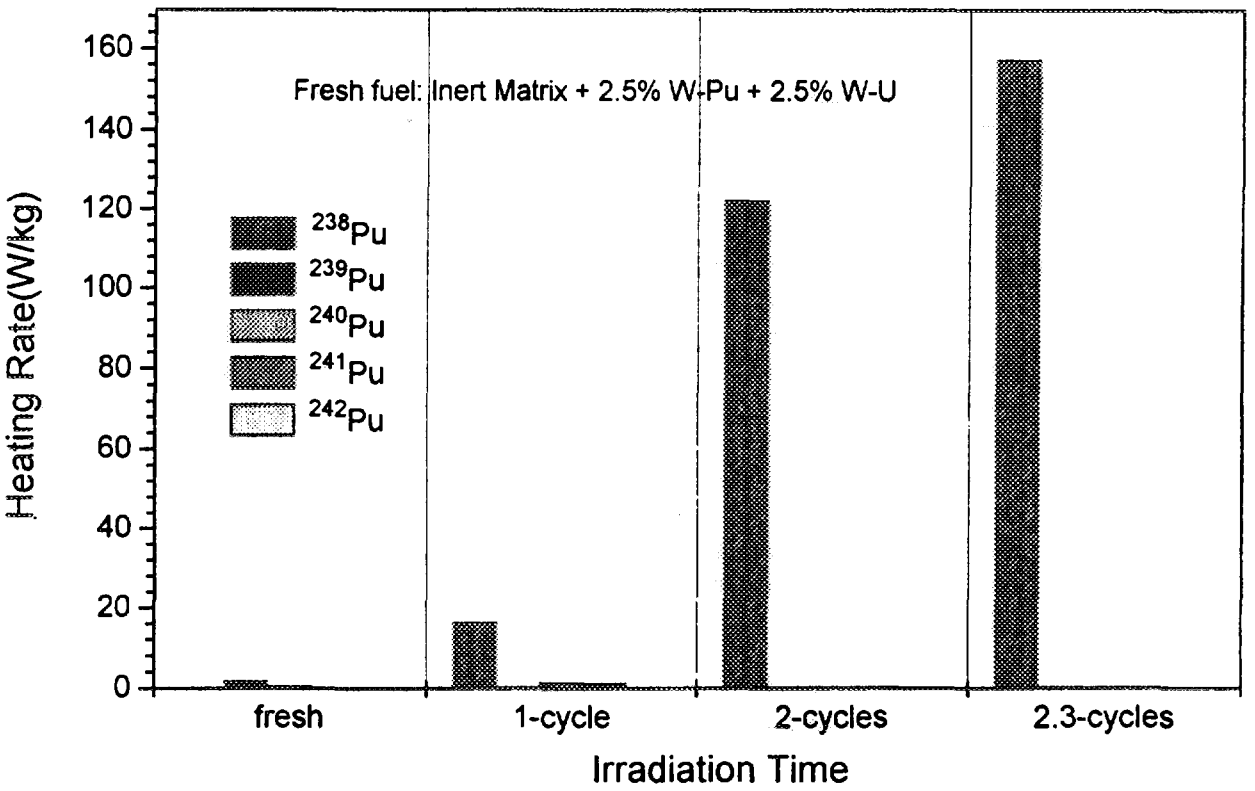

Fig. 4. Isotopic heating rates per unit mass of total Pu: for fresh fuel, and after 1, 2, and 2.3 cycles in a PWR.

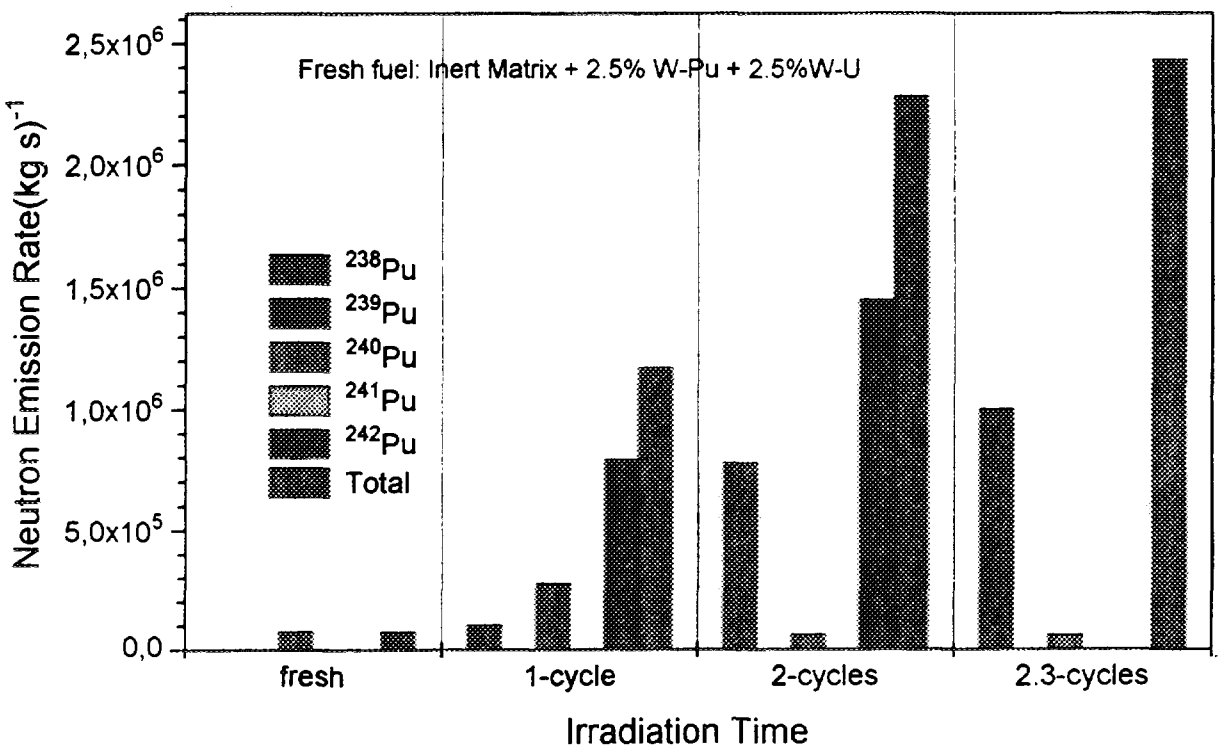

Fig. 5. Isotopic neutron emission rates per unit mass of total Pu: for fresh fuel, and after 1, 2, and 2.3 cycles in a PWR

4. CRITICALITY ASPECTS OF INERT MATRIX-BASED MIXED U-PU FISSILE MATERIAL

In the calculations it was assumed that the presence of inert matrix based fuel pins did not disturb the neutron flux in the reactor. This is only true provided that the variations for the standard and inert matrix based fuels are not too different. In this section we compare the k_∞ variations resulting from the irradiation of standard and inert matrix based fuels.

In Fig. 6 the variation of k_∞ of the standard fuel configuration (described in the previous section) of 3.2% ^{235}U in a natural U matrix is shown over one cycle (i.e. 879 days). Also shown is the average thermal power generated. The variation of k_∞ of an inert matrix containing 2.5% W-Pu and 2.5% W-U in a neutron flux of the LWR is shown in Fig 7. The irradiation time extends to 2000 days corresponding to 2.3 cycles of the LWR. Clearly, large variations in k_∞ can be seen. Indeed after about 900 days irradiation the configuration inert matrix plus fissile material becomes subcritical. It should be noted here that although $k_\infty < 1$ this material is still producing net energy with a multiplication factor $M \propto 1/(1-k_\infty)$.

How can such an irradiation be accomplished within a LWR with a standard fuel life of 879 days? One possible scheme is to consider the core composed of an inner core and an outer blanket with the following properties:

- the inner (critical) core contains standard and inert matrix fuel pins with $k_\infty > 1$.
- an outer (sub-critical) blanket of irradiated inert matrix fuel pins with $k_\infty < 1$.

In steady operation of the reactor, after each 293 days (one third of a standard cycle):

- one third of the standard pins (i.e. those which have been in the reactor for 879 days) are removed from the reactor core and replaced by fresh pins,
- one third on the inert matrix fuel pins irradiated in the central core for 879 days are removed to the blanket where they remain for another 1.3 cycle irradiation. These inert matrix pins are replaced by fresh inert matrix pins,

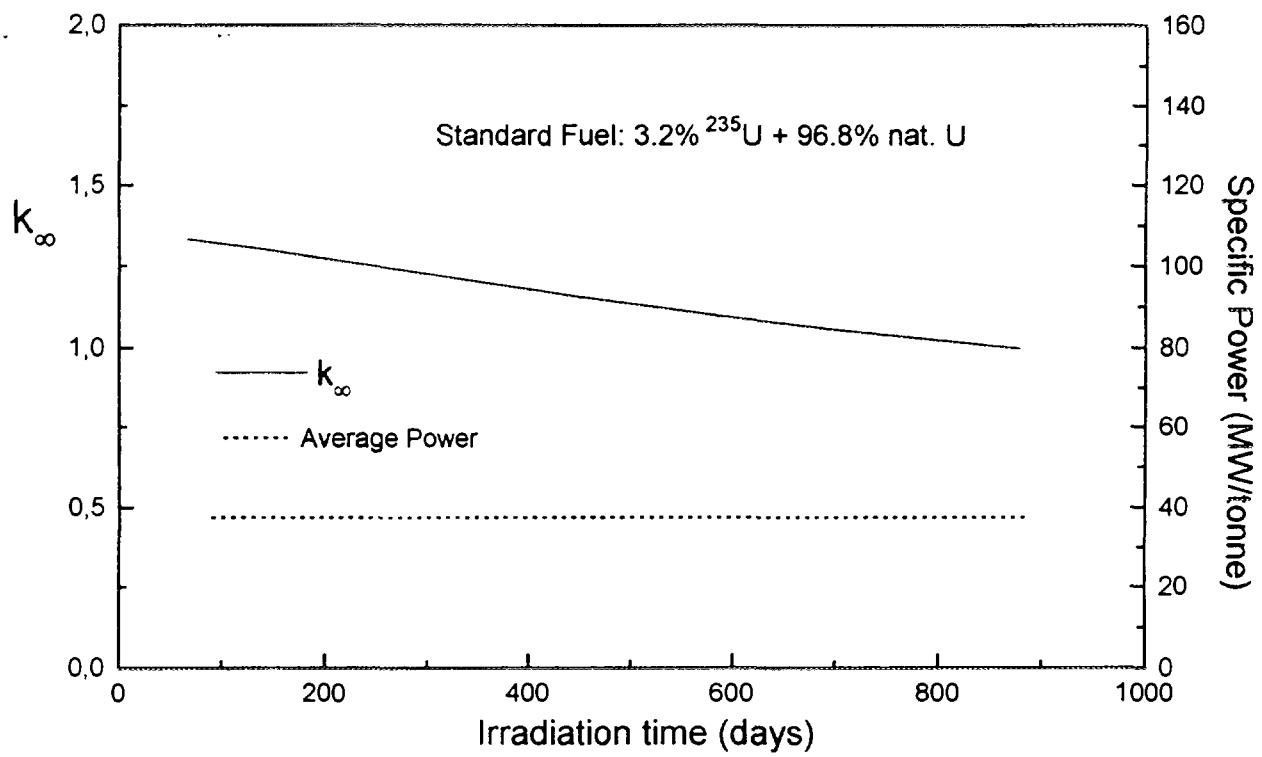

Fig. 6. Variation of k_∞ and average power with time for standard PWR fuel.

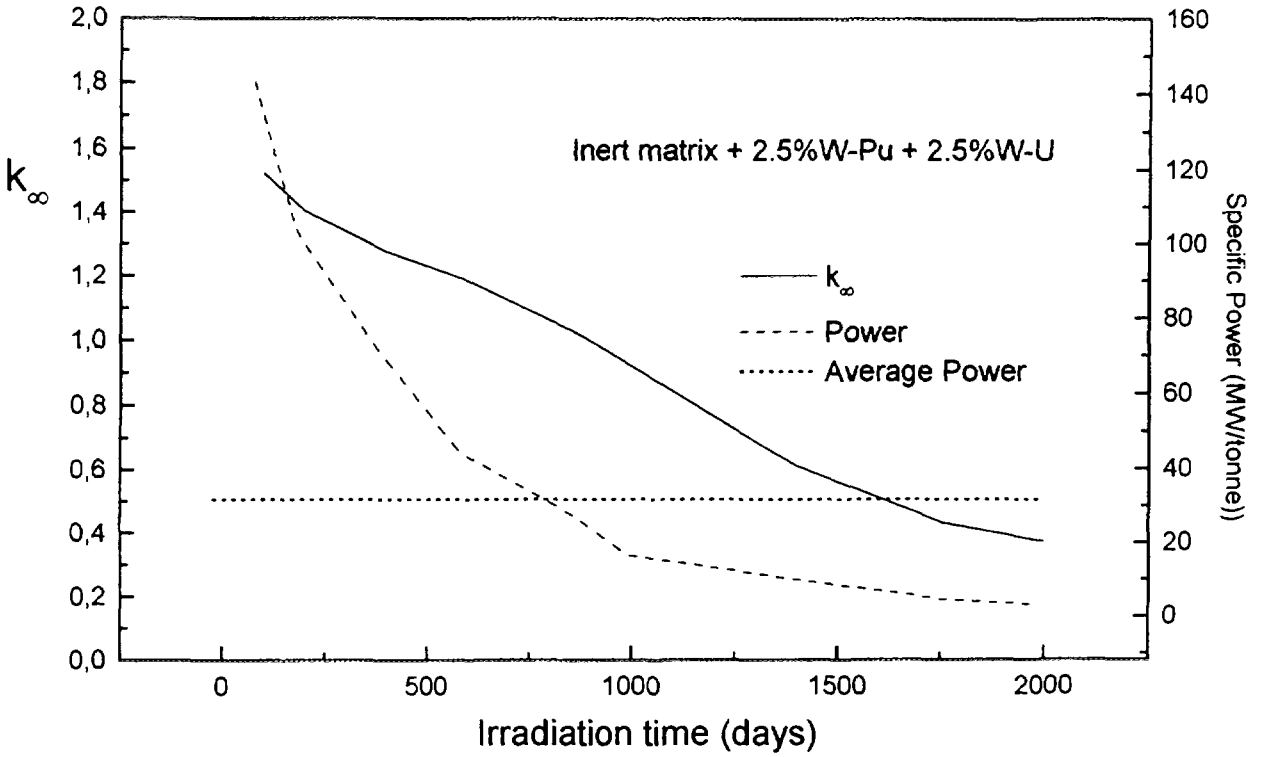

Fig. 7. Variation of k_∞ and power with time for the inert matrix based fuel in a PWR neutron spectrum.

- one quarter of the inert matrix pins in the outer blanket of the reactor are removed (these pins have been in the reactor for a period of 7 × 293 = 2050 days) from the reactor.

Such a scheme will ensure that the standard pins are in the reactor for 878 days whereas the inert matrix pins are in the reactor for the required 2050 days. It is not a requirement that the outer blanket should contain the sub-critical inert matrix based pins. Depending on the neutron distribution, this array of sub-critical pins could be placed in an annular cylinder in the core region or dispersed through the core.

The above heterogeneous core layout cannot be investigated with a zero dimensional code such as ORIGEN2. However, an approximate treatment of such a heterogeneous core can be made by considering a homogeneous core containing fuel at different stages of irradiation. Hence a standard core can be considered as containing fuel - one third of which is fresh, one third has been irradiated for 293 days and one third irradiated for 586 days. The variation of k_∞ over one third of a cycle i.e. 293 days is shown in Fig. 8. Similarly, one can consider a core consisting of inert matrix based fuel. Since the fuel here is in the reactor for seven periods of 293 days (≈2000 days), the homogeneous material contains a mixture - one seventh of which is fresh, one seventh irradiated to 293 days, one seventh irradiated to 586 days, etc. Notice that this core is being irradiated with the spectrum of the standard PWR. Provided that the variation of k_∞ with time is not too different from that of the standard PWR, this should be a reasonable approximation. The extent to which this is the case may be seen in Fig. 8. Here it can be seen that by varying the amounts of fissile material i.e. W-Pu and W-U, the ratio W-Pu:W-U, and through the use of a burnable neutron absorber [9], the variation of k_∞ can be approximated to that of the standard reactor.

5. SOME CONSIDERATIONS ON INERT MATRICES

In the previous sections the inert matrix has not been defined. In the calculations a given amount of weapons grade material is contained in a volume of inert matrix. If the density of the inert matrix were

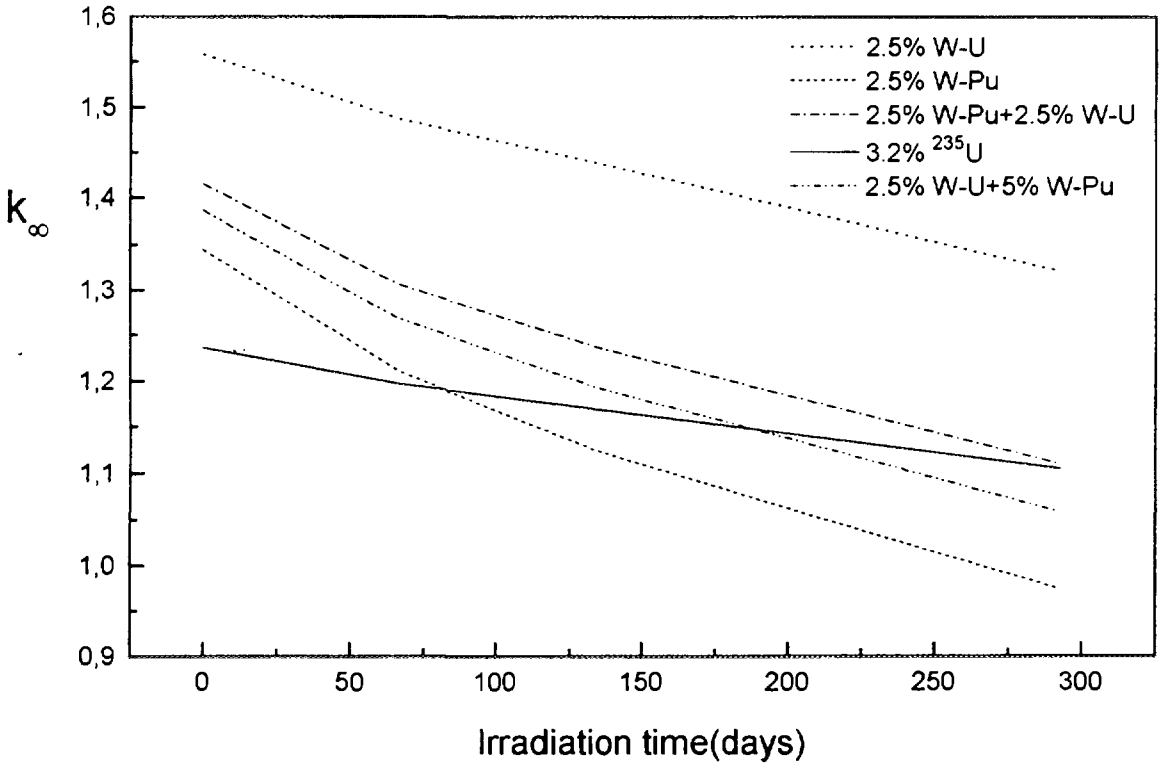

Fig. 8. Variation of k_∞ with time for different fuels and mixing ratios of W-Pu and W-U.

the same as that of the UO_2 fuel, then approximately 50 kg of W-Pu per tonne of matrix (i.e. 5% assuming equal densities) is required for equivalent power production.

The inert matrix is a material which replaces conventional UO_2. It should not form actinides by neutron capture, but otherwise have properties similar to or superior to those of UO_2. To avoid actinide formation, the atomic number of all components should be significantly less than 92. To have good reactor properties, the melting point T_m should be high (greater than 2000C; for UO_2 T_m = 2880C), the thermal conductivity and mechanical properties should be equivalent to those of UO_2, and the matrix should be compatible with the cladding and coolant. Finally, it should not have a high neutron capture cross section in order to obtain good neutron economy.

Two classes of ceramics can be selected which meet these criteria [10,11]. One class contains Al_2O_3, spinel ($MgAl_2O_4$), or MgO which have no solid solubility for actinides. In this case, a two phase heterogeneous fuel will result with UO_2 or PuO_2 particles in the inert matrix. The second class contains CeO_2, and zircon ($ZrSiO_4$), and shows solid solubilities high enough to allow fabrication of a homogeneous fuel. A major research activity is being pursued at ITU [12] to investigate fabrication routes and properties of the above candidate ceramics in order to define the most suitable matrix.

6. DECAY OF ^{238}Pu

Because the half life of ^{238}Pu is 87.75 y, the proliferation resistance of the spent inert matrix-based fuel will decrease with time. After approximately 200 years the level of ^{238}Pu in the Pu is less than 5% as can be seen in Fig. 9. If at this time the spent fuel is still considered a proliferation risk, one need only re-irradiate the material again without reprocessing. The result of doing this every 200 years up to 800 years is shown in Fig. 9. The neutron flux used here is that of the standard PWR. Finally, in Fig. 10 we show the variation of the masses of the Pu isotopes resulting from a series of irradiation and decay steps

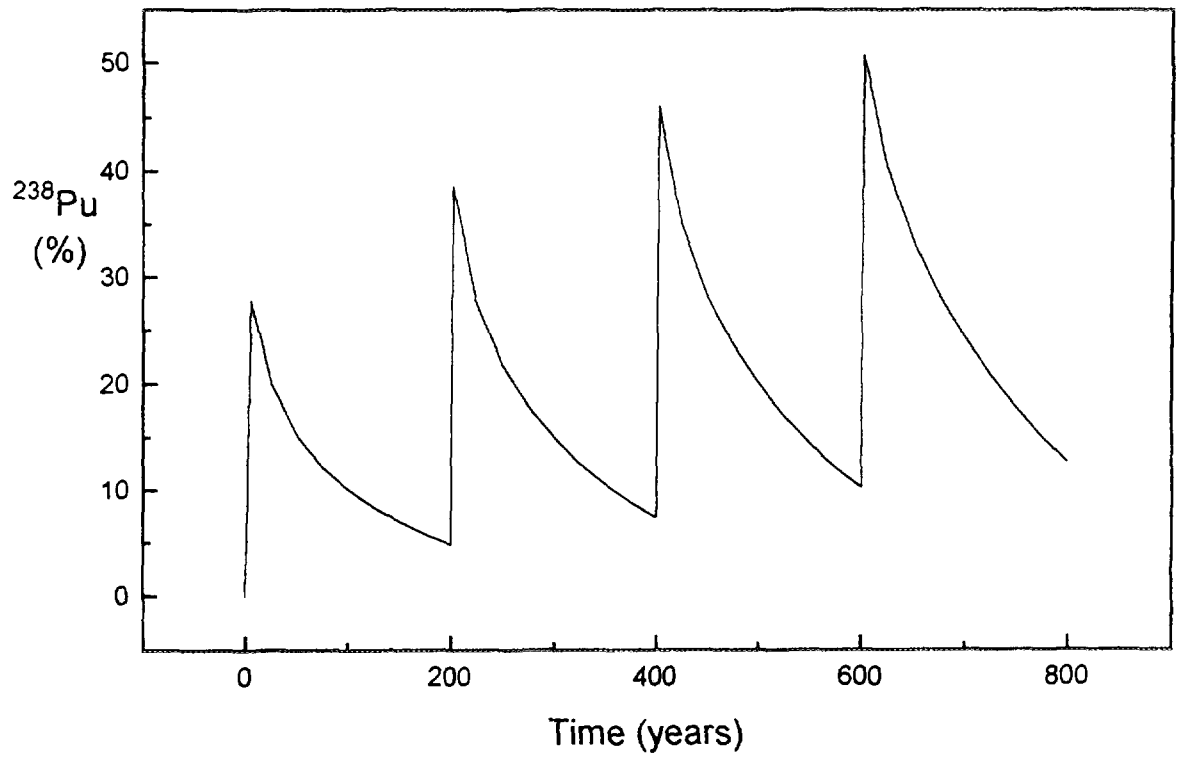

Fig. 9. The percentage of ^{238}Pu in total Pu resulting from irradiation and decay of an inert matrix containing 2.5%W-Pu+2.5%W-U. Initially the fresh matrix is irradiated for 2000 days. At 200, 400, and 600 years, the matrix is re-irradiated for 1000 days.

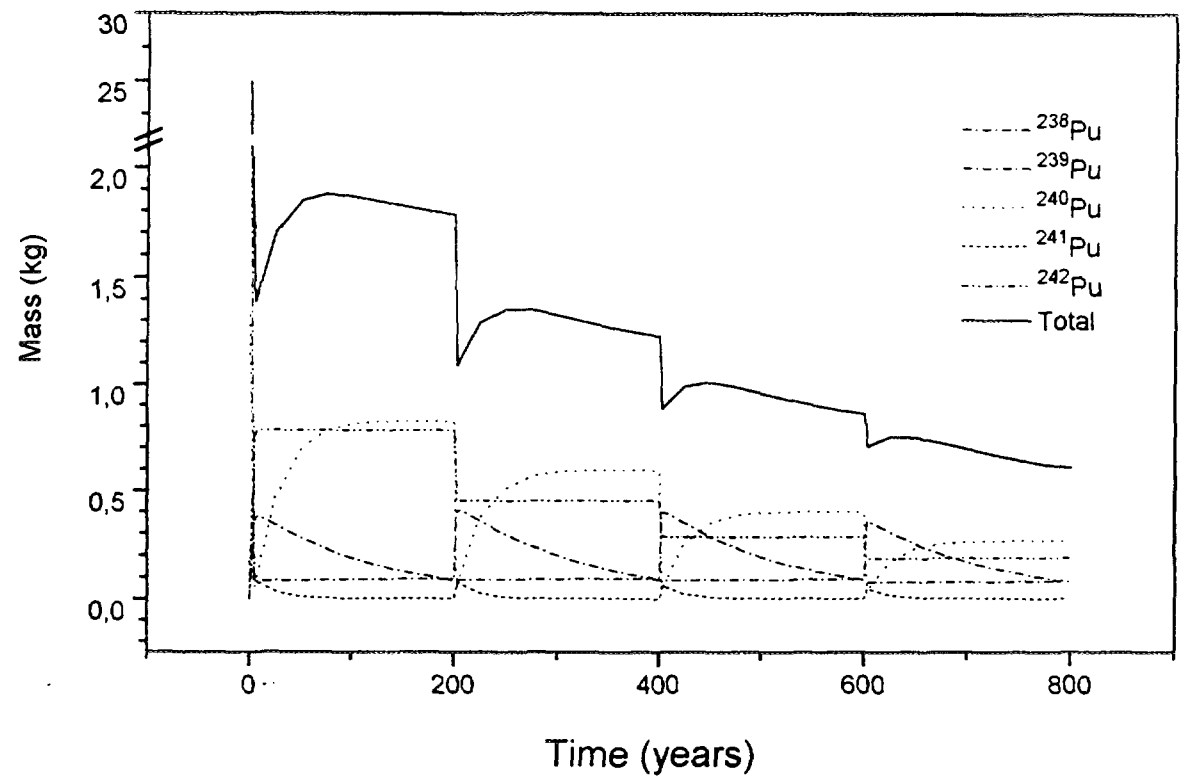

Fig. 10. The masses of Pu isotopes resulting from the irradiation and decay of an inert matrix containing 2.5%W-Pu+2.5%W-U. Initially the fresh matrix is irradiated for 2000 days. At 200, 400, and 600 years, the matrix is re-irradiated for 1000 days.

over a period of 800 years - starting with the fresh W-Pu in the inert matrix. The fresh matrix is irradiated for 2000 days. At 200, 400, and 600 years the matrix is re-irradiated for 1000 days. From Fig. 10 it can be seen that following an irradiation step, in which the total Pu decreases rapidly, the total Pu increases. This is due to the increasing amounts of ^{240}Pu which arise from ^{244}Cm formed during the irradiation step which then decays with a half-life of about 18 years.

7. CONCLUSIONS

An analysis of a novel scheme for W-Pu disposition by burning the plutonium in LWRs has been presented. The main characteristics of this scheme are that it provides a high destruction level of the W-Pu and a high proliferation resistance of the spent fuel ashes in a once through cycle. The high destruction level of the W-Pu is guaranteed by embedding the W-Pu in an inert matrix and, in addition, by irradiation in a standard PWR for 2000 days. After this time essentially only ^{242}Pu is present from the W-Pu irradiation. Proliferation resistance of the spent fuel is considerably increased by the use of W-U in the fresh fuel. The presence of this material leads to an additional source of ^{238}Pu which, after irradiation for 2000 days, forms approximately 30% of the total plutonium. In contrast to alternative methods for increasing the amounts of ^{238}Pu in the spent fuel, which are based on the use of ^{237}Np or ^{236}U obtained by reprocessing, the present method requires no reprocessing of the inert matrix-based fuel. The requirement of an irradiation time of approximately 2000 days in the reactor ensures that less that 20% of the total uranium remaining is ^{235}U, thereby overcoming a potential proliferation problem with the irradiated uranium (notice that the 20% level applies to separated uranium - in the present case, since this is mixed with alpha active waste, the 20% level could be significantly increased implying an irradiation time less than 2000 days). The materials problems of inert matrices and clad for such irradiations has not been considered but are under investigation [12].

The compatibility, with regard to neutronics and reactor operation, of mixing standard fuel pins with lifetime of 879 days with inert matrix based pins with lifetime of 2000 days in a standard PWR reactor is considered and a scheme for fuel pin exchange is proposed. An analysis of the criticality aspects of this scheme is made by assuming that the heterogeneous system can be approximated by a homogenous core consisting of fuel at different stages of irradiation. This analysis shows that the standard PWR variation in criticality, based on the variation of k_∞, can be closely approximated by varying the total amounts of fissile material in the inert matrix and by varying the mixing ratio of W-Pu/W-U. Hence, as far as neutronics and criticality are concerned there should be little difficulty in the use of inert matrix-based fuel pins in a standard PWR. The limitations of the use of such pins will be determined by other factors such as the requirement of a negative temperature coefficient, peak power limitations, fuel material behaviour, cladding resistance, and fuel pellet interactions.

Finally, since ^{238}Pu has a half-life of approximately 88 years, the proliferation risk of the spent matrix based fuel will decrease with time. Over a period of 200 years the level of ^{238}Pu in the plutonium will drop below 5% - the minimum value required for proliferation resistance. The advantage of the present scheme is that the high level of ^{238}Pu can be restored by re-irradiating the spent matrix for a further period of 1000 days. The process of re-irradiation and decay can be repeated, *without reprocessing*, for as long as the spent matrix is considered a proliferation risk.

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