

# **A STRATEGY FOR Pu DESTRUCTION IN PWRs**

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#### **Abstract**

We consider fuelling the core of a LWR with assemblies of standard and inert matrix based Pu fuels such that there is no net rate of Pu production. The Pu in the inert matrix fuel is obtained by recycling that from spent  $UO<sub>2</sub>$  fuel. The Pu produced in the standard  $UO<sub>2</sub>$  assemblies during reactor operation is then balanced by the Pu burnt in the inert matrix assemblies. This can be achieved by replacing approximately one fifth of the standard assemblies with inert matrix based Pu assemblies. Extended irradiation of the inert matrix assemblies is also considered.

Calculations are presented for a homogeneous core containing standard and inert matrix assemblies at various stages of irradiation. On this basis, it is suggested that the large variations in  $k_{\infty}$  associated with individual assemblies of inert matrix based Pu in a LWR neutron spectrum can be considerably reduced by a suitable choice of the cycle loading pattern for the assemblies.

## **Introduction**

In conventional PWR, fissile  $U^{235}$  is present to a level of about 3.2% in a  $U^{238}$  matrix. Although not directly fissionable, this fertile  $U^{238}$  is transmuted to Pu isotopes in the reactor neutron spectrum. During the lifetime of the fuel in the reactor some of this Pu is burnt. At the end of the fuel lifetime, however, a considerable amount of Pu remains and makes a major contribution to the radioactivity of the spent fuel.

Ideally, one would like to use a non-fertile, inert matrix instead of  $U^{238}$  and thereby avoid the main source of radiotoxicity<sup>1</sup>. In practice, however, problems arise when one attempts to do this. One such problem is associated with the Doppler effect in  $U^{238}$ . If for some reason the neutron flux increases in the reactor it results in an increase in temperature. This in rum leads to a thermal broadening of the absorption resonances at  $5eV$  in  $U^{238}$ , which results in neutrons being lost from the system and thereby acts as a self regulating mechanism to stabilise the reactor.

Various options are available by which one can reduce the net amount of Pu produced while at the same time having a significant amount of  $U^{238}$  present. In one such scheme the spent fuel could be reprocessed to remove the Pu isotopes and then mixed with fresh  $UO<sub>2</sub>$  fuel. This is the basis of Mixed Oxide (MOX) fuel fabrication.

In this paper, we consider an alternative to this approach, in which the recycled Pu is used in an inert matrix (Pu/IM). Assemblies of such Pu/IM pins would then be mixed with standard  $UO<sub>2</sub>$  assemblies.

In a LWR, plutonium is produced by neutron irradiation of the fertile matrix of  $U^{238}$ . In the case of the pressurised water reactor considered in the next section, plutonium is produced at a rate of approximately 9kg Pu per tonne of fuel during the lifetime (879 EFPD) of the fuel in the reactor (corresponding to approximately 300kg Pu per GWye, see ref. 2). An inert matrix having the same volume as one tonne of standard fuel and containing initially 50kg R-Pu (i.e. 10 w/o assuming the density of the inert matrix is half that of the standard fuel) will burn approximately 40kg Pu in the same neutron spectrum and over the same time, as shown in table 1. Hence, in a steady situation where no net Pu is produced, approximately 18% (i.e. 9/49) of the core should contain inert matrix assemblies.

In a second scenario, the inert matrix assemblies could be kept in the reactor for another 879 days i.e. twice as long as the standard assemblies. Over an irradiation period of 2x879 days approximately 46kg Pu are destroyed. On average, over 879 days 23kg Pu is destroyed. Using the same arguments as above, in a steady situation where no net Pu is produced, approximately 28% (i.e. 9/32) of the core contains Pu/IM assemblies.



Table 1. Composition (%) of different types of plutonium.

\*Plutonium recovered from pressurised water reactor fuel  $(3.2\% \text{ U}^{235})$  with burnup 33GWd/t

Notice in this latter scenario, keeping the inert matrix in the reactor for a further three cycles increases the amount of Pu burnt only from 40kg to 46kg. Clearly more Pu would be burnt if after three cycles irradiation, fresh Pu/IM assemblies were substituted since over the next three years irradiation another 40kg of Pu will be burnt instead of only  $46-40 = 6$ kg! This does mean, however, that larger quantities of Pu are present in the spent inert matrices i.e. 20 rather than 8kg Pu per ton of inert matrix.

# **Calculational Details**

The calculations were made using the point depletion code ORIGEN23 and checked independently with RADONN<sup>4</sup>. The one group cross section library used, PWRUS.lib<sup>3</sup> as that for a standard pressurised water reactor with  $3.2\%$   $235U$  initial fuel enrichment, power level of 37.5 MW/tonne, and burnup of 33 GWd/tonne (achieved after 879 EFPD-efFective full power days). Details of the calculation have been given elsewhere<sup>5</sup>.



Fig. 1. Pu isotopic masses in 0.5 tonne of matrix: in fresh fuel, and after 3, 6, and 7 cycles in a PWR (3 cycles = 878 EFPD). Fresh fuel contains 10 w/o R-Pu.

#### **Pu/lnert Matrix irradiation in PWR neutron Spectrum**

We consider the irradiation of an inert matrix based fuel containing 10 w/o reactor grade Pu  $(R-Pu)$  in a PWR. The relevant details of the PWR have been described<sup>5</sup>. It is also assumed that the presence of the inert matrix plus fissile material does not disturb the neutron spectrum of the reactor (in the next section the variation of  $k_{\infty}$  for this fuel is considered). The results of an irradiation over three cycles together with extended irradiation over six and seven cycles are shown in fig. 1. and summarised in table 2. for the plutonium isotopes. Here the masses of isotopes present per tonne of inert matrix are given at various irradiation times. In table 2, results of an irradiation using weapons grade Pu (W-Pu), with composition given in table 1, rather than R-Pu, are also given. Although the amount of Pu burnt increases with irradiation time (after 7 cycles 94% of the Pu has been destroyed), the rate at which it is being burnt is decreasing. Hence, over the first 3 cycles 40kg Pu is burnt. Over the next three cycles only 6kg is burnt. In an additional cycle only 0.7kg is burnt.

Table 2. Evolution of an inert matrix containing 10 w/o Pu in the neutron spectrum of a PWR (3 cycles  $= 879$  EFPD). Densities of the inert matrix is assumed to be half of that of the standard fuel.

	fresh	3 cycles	6 cycles	7 cycles
total W-Pu	50kg	7.9kg	2.4kg	1.9kg
total R-Pu	50kg	10.0kg	3.8kg	3.1kg

#### **Criticality Aspects of Pu/lnert Matrix Fuel**

In the above calculations it was assumed that the presence of inert matrix based fuel pins did not disturb the neutron flux in the reactor. A necessary condition for this to be the case is that the infinite neutron multiplication  $k_{\infty}$  for the standard and inert matrix based fuels are not too different. In this section we consider  $k_{\infty}$  variations from the irradiation of standard and inert matrix based fuels.



Fig. 2. Variation of  $k_{\infty}$  and average power with time for a standard PWR fuel.



Fig. 3. Variation of  $k_{\infty}$  and power with time for inert matrix based R-Pu fuel during extended irradiation in a PWR neutron spectrum.



Fig. 4. Equilibrium cycle loading pattern for standard  $UO<sub>2</sub>$  and Pu/IM fuel assemblies in a PWR. Regions of the core are denoted 1, 2, and 3 and contain fresh, once burnt, and twice burnt standard assemblies. Regions denoted 1\*, 2\*, and 3\* contain fresh, once burnt, and twice burnt Pu/IM assemblies.

In fig. 2. the variation of  $k_{\infty}$  of the standard fuel configuration (described in the previous section) of  $3.2\%$   $235U$  in a natural U matrix is shown over three cycle (i.e. 879 days). Also shown is the average thermal power generated. The variation of  $k_{\infty}$  of an inert matrix containing 10 w/o W-Pu and in a neutron flux of the LWR is shown in fig 3. The irradiation time extends to 2000 days corresponding to 7 cycles of the LWR.

During this time  $k_{\infty}$  from about 1.4 to 0.4 can be seen. After about 900 days irradiation the inert matrix plus fissile material becomes subcritical. Although  $k_{\infty}$  < 1, the Pu/IM assemblies still produce power since the 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 three regions as shown in fig.4.

In steady operation of the reactor, after each cycle (293 days), one third of the  $UO<sub>2</sub>$ fuel assemblies (i.e. those which have been in the reactor for 879 days) from positions 3 in fig. 4 are removed from the reactor core and replaced by fresh fuel in positions 1. Fuel assemblies in regions 1 and 2 are moved to regions 2 and 3 respectively. Similarly with the Pu/IM assemblies for a three cycle irradiation. For a six cycle irradiation, the Pu/IM assemblies stay in regions 1, 2, and 3 for two cycles each. For a seven cycle irradiation, the Pu/IM assemblies stay in regions 1 and 2 each for two cycle and in region 3 for three cycle.



Fig. 5. Comparison of  $k_{\infty}$  for standard and inert matrix based fuels in a PWR neutron spectrum accounting for periodic refuelling of fuel bundles.

The above heterogeneous core layout cannot be investigated with a zero dimensional code such as 0RIGEN2. 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 (or a Pu/IM core, three cycle irradiation) 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 values of  $k_{\infty}$  for the  $UO<sub>2</sub>$  and Pu/IM fuels over one cycle (293 days) given in fig. 5 show generally good agreement. Initially, the value of  $k_{\infty}$  for the Pu/IM fuel is about 10% higher than that for the standard fuel. This could be compensated for, however, by the use of a burnable poison.

For the extended irradiations, the Pu/IM fuel is considered to be in the reactor for seven cycles ( $\approx$  2000 days). As described above, the core can be considered as containing a homogeneous mixture - one seventh of which is fresh, one seventh irradiated to 293 days, one seventh irradiated to 586 days, etc. The results of seven cycle irradiations using R-Pu and W-Pu are shown in fig. 5. It can be seen that the variations in  $k_{\infty}$  are still relatively large indicating that the use of a PWR neutron spectrum is not a good approximation here.

## **Conclusions**

A strategy is described whereby standard  $UO<sub>2</sub>$  fuel assemblies are placed together with Pu/IM fuel assemblies in a PWR such that no net Pu is generated. The amount of Pu generated from the fertile  $U^{238}$  in the standard fuel is equal to the amount burnt in the inert matrix. Following discharge, spent  $UO<sub>2</sub>$  fuel is reprocessed to obtain the Pu which is then used to fabricate fresh inert matrix fuel. Spent inert matrix fuel is not reprocessed.

Two options are available for burning Pu in the inert matrix: either one can maximise the absolute amounts of Pu which are burnt or one can maximise the burning rates.

Maximising the amounts of Pu burnt requires an extended irradiation and results in minimum levels of the Pu in the spent inert matrix fuel. A zero net production of Pu requires that approximately 28% of the fuel assemblies to be inert matrix based. The spent inert matrix fuel contains approximately 3kg Pu in 0.5 tonne of inert matrix (which has the same volume as 1 tonne of standard fuel).

Maximising the burning rates occurs in a single 3-cycle irradiation, and results in higher levels of Pu in the spent matrix. A zero net production of Pu requires that only 18% of the fuel assemblies are inert matrix based. The spent inert matrix fuel in this case contains approximately 10kg Pu in 0.5 tonne of inert matrix.

An example of the equilibrium cycle loading pattern for a reactor core containing 180 fuel assemblies in which standard fuel is present in the reactor for 3 cycles whereas the Pu/IM fuel is present for up to 7 cycles is described. An analysis of the infinite neutron multiplication factor  $k_{\infty}$  for homogenous cores containing Pu/IM material at various stages of irradiation in the range of 1-7 cycles shows that extended irradiation results in large variations of reactivity. Over three cycles, however, the reactivity of both the standard and inert matrix based fuels are fairly similar. The initially higher values of for the Pu/IM fuel can be compensated by the use of a burnable poison. In addition, one may be able to accommodate such fuel within the core without significantly disturbing the Doppler effect since 82% of the core still consists of standard assemblies.

### **REFERENCES**

1. J. Magill, C. O'Carroll, P. Gerontopoulos, K. Richter, J. van Geel, *Advantages and Limitations of Thorium Fuelled Energy Amplifiers,* Proceedings of the technical committee meeting on Unconventional Options for Pu Disposition, Obninsk, Nov, 7- 11 1994, IAEA to be published.

2. Garwin, R. L., Grubb, M., and Mantale, E. (Editors), *Managing the Plutonium Surplus: Applications and Technical Options,* Kluwer Academic Publishers, Netherlands, 1994.

3. Croff, A. G., *ORIGEN2: A Versatile Computer Code for Calculating the Nuclide Composition and Characteristics of Nuclear Materials,* Nucl. Tech. 62 (1983) 335- 353.

4. Magill, J. and Pel, D., *RADONN: - A Software Package for Radionuclide Analysis,* in EUR 15741 EN (1994), Annual Report 1993, Institute for Transuranium Elements, Karlsruhe.

5. J. Magill, Hj. Matzke, G. Nicolaou, P. Peerani, J. van Geel, *A Once Through Scheme for Weapons Grade Pu Disposition in LWR's: Proliferation and Criticality Aspects,* Technical Committee Meeting on the Recycling of Plutonium and Uranium in Water Reactor Fuels, U.K. 3-7 July 1995, IAEA, to be published.

6. J. Magill, Hj. Matzke, G. Nicolaou, P. Peerani, J. van Geel, *A Method for the Destruction of Plutonium by Irradiation in a Light Water Reactor,* European Patent Application no. 95 110 397.7 (Euratom)