Radioactive Waste Management and the Nuclear Fuel Cycle
1990, Vol. 14(4), pp. 275–284
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

PLUTONIUM AND REACTOR TRANSMUTATION

A.G. ELAYI
Institut De Physique Nucléaire, B.P. N°1, 91406 ORSAY Cedex, France

(Received August 7, 1989)

Being a strategic material (atomic weapons) or a material used for the production of energy does not prevent plutonium from being potentially, directly or indirectly (through the radiotoxicity of the spent Mox fuel) a radioactive waste. Due to the large amounts of Pu available and produced, the transmutation of Np, Am and Cm is only meaningful if the plutonium problem can be assessed and solved. However we show that this leads to serious difficulties as the present technological options have been chosen in function of the production of energy and not in function of a strategy of transmutation.

INTRODUCTION

Hundreds of papers deal with the nuclear transmutation in reactors of the isotopes of neptunium, americium and curium with long half-lives produced during the fuel burn-up. As it is a material for the production of energy, (used in fast breeder reactors and partially instead of ^{235}U in thermal reactors), as well as a strategic material (weapons), plutonium is considered separately and is not included in these transmutation studies. The aim of this paper is to answer the following questions:

1. Can the total radiotoxicity of the plutonium produced in the different reactors be neglected with respect to the total radiotoxicity of neptunium, americium and curium?
2. If not, does the fact that it is either a strategic or an energy producing material prevent plutonium from being potentially, directly or indirectly (through the radiotoxicity of the Mox spent fuel) a radioactive waste?
3. If the plutonium is directly or potentially a radioactive waste, in order to be meaningful any policy to transmute Np, Am and Cm must also be able to solve the plutonium problem. Is this possible with present reactors and reprocessing options?

PLUTONIUM RADIOTOXICITY

Considering a certain amount of a radioisotope, its potential risk is defined as the risk directly associated with the possible ingestion, by a group of people, of the entire amount of the radioisotope, mainly through drinking water. The potential risk is related to the concepts of maximum permissible concentration (M.P.C.) and annual limit of intake by ingestion (A.L.I.) developed in the publications of the International Commission on Radiological Protection I.C.R.P.[1-5] They correspond to an annual equivalent dose limit of 50 mSv for the people working in the nuclear industry and they have a value 10 times lower for the public because of the corresponding annual equivalent dose limit of 5 mSv. The I.C.R.P. has regularly updated the (A.L.I.) values in order to take into account new metabolic data as well as new dose calculation procedures. The impact of the changes in the I.C.R.P. recommendations on the potential radiotoxicity of LWR, FBR and Candu fuels has been studied in reference 6. In the present paper, we use the (A.L.I.) values recommended by the I.C.R.P.-48 publication.[5]

The potential risk relative to a radioisotope is a function of three parameters: the amount of this radioisotope, its half-life and its radiological impact after ingestion, through the (A.L.I.) value. In Table I, we show the A.L.I. values for some of the Np, Pu, Am and Cm isotopes. Apart from those of ^{241}Pu and ^{242}Cm which have relatively short half-lives, all the other values are of the same order of magnitude. In contrast, the half-lives and the amounts of these isotopes produced during the fuel burn-up vary substantially from one isotope to the other. In Table II, we show the amount of neptunium, plutonium, americium and curium produced per ton of a 33,000 MWd/t light water reactor spent fuel. The amount of plutonium is 15 times larger in fast breeder reactors while it depends upon the composition of the Mox fuel recycled in LWRs. Its value is of about 40 kg for a 5.25% enriched Mox fuel recycled in a 33,000 MWd/t LWR and of about 50 kg for a 7.5% enriched

TABLE I
A.L.I. and Half-Lives from I.C.R.P.-48 Recommendations

	Np-237	Pu-238	Pu-239	Pu-240	Pu-241	Am-241	Am-243	Cm-242	Cm-244
Half-life (year)	2.14×10^{6}	87.7	2.41×10^{4}	6.55×10^{3}	14.4	432.6	7.38×10^{3}	0.446	18.11
A.L.I.	3×10^{3}	3×10^{3}	2×10^{3}	2×10^{3}	10^{5}	2.6×10^{3}	2.6×10^{3}	10^{5}	4.5×10^{3}

TABLE II
Amounts of Np, Pu, Am and Cm in Grammes Per Ton of Discharged Fuel

LWR	Np	Am	Cm	Pu
	450	300	30	10000

Mox fuel recycled in a 45,000 MWd/t LWR. In both cases, the plutonium used for these calculations originates from a fuel irradiated only once in the reactor as it is the case so far. The amount of Pu is significantly larger than that of Np, Am and Cm together.

As far as the radiotoxicity of these elements is concerned, Figure 1 shows the variation with time of the number of (A.L.I.) per GW(e) × year of Np, Pu, Am and Cm present in a 33,000 MWd/t LWR spent fuel and a 45,000 MWd/t LWR spent Mox fuel. Obviously, the radiotoxicity of Pu is far from being negligible as compared with the radiotoxicity of Np, Am and Cm. The transmutation of the long half-life isotopes of these elements is far from solving the problems of the radiotoxicity on the long range term because of the presence of the plutonium. This is particularly true for the period of time following a substantial decay of the americum and lasting until a substantial decay of the plutonium as it can be seen on the curves of Figure 1.

Since the total radiotoxicity of the plutonium is not negligible with respect to that of neptunium, americium and curium, what does the fact that it is both a strategic and an energy producing material imply? It seems that some contradiction exists, at a first glance at least, concerning the Pu problem: apart from being a by-product of the reactors using uranium fuel, it is intentionally produced for strategic purposes, in military graphite gas reactors, and for energetic purposes in FBR reactors; why could it be considered potentially as a radioactive waste when it is intentionally produced?

In the case of "good quality" strategic plutonium, the product itself plays both roles since it is, for a period of time (as long as it is used in a weapon) a strategic material and it will become afterwards (when the weapons will be decommissioned because of international treaties or new technological developments) a radioactive waste (the half-life of ^{239}Pu is of about 24,000 years). The fact that it may be used afterwards as an energetic material will not solve the previously mentioned waste problem as will be shown hereafter.

Being used as a material for the production of energy does not prevent plutonium from being indirectly a radioactive waste since it produces new radiotoxic materials including plutonium itself. The radiotoxicity of the spent fuel is far from being negligible with respect to that of the plutonium used for the Mox fuel. In order to show the relative importance of the radiotoxicity of the spent fuel at the different times of the future, we plot in Figure 2 the variation of the ratio of the radiotoxicity of the Mox part of the spent fuel irradiated in a 45,000 MWd/t LWR and the radiotoxicity of the same Mox part of the fuel when it is not irradiated. The total radiotoxicity increases up to several hundred years. Therefore, the use of Pu as an energetic material does not prevent it from being indirectly a radioactive waste with comparable radiotoxicity.

Since plutonium is potentially directly a radioactive waste, or is to be used as a material for the production of energy, let us assess further the consequences of its utilisation as an energetic material in LWRs or equivalent reactors.

As far as FBR are concerned, they are not considered here because their construction is postponed in all national programmes.

At present, plutonium is being recycled, in some countries, in LWRs. Present projects consist in replacing about one third of the uranium fuel by a Mox fuel in

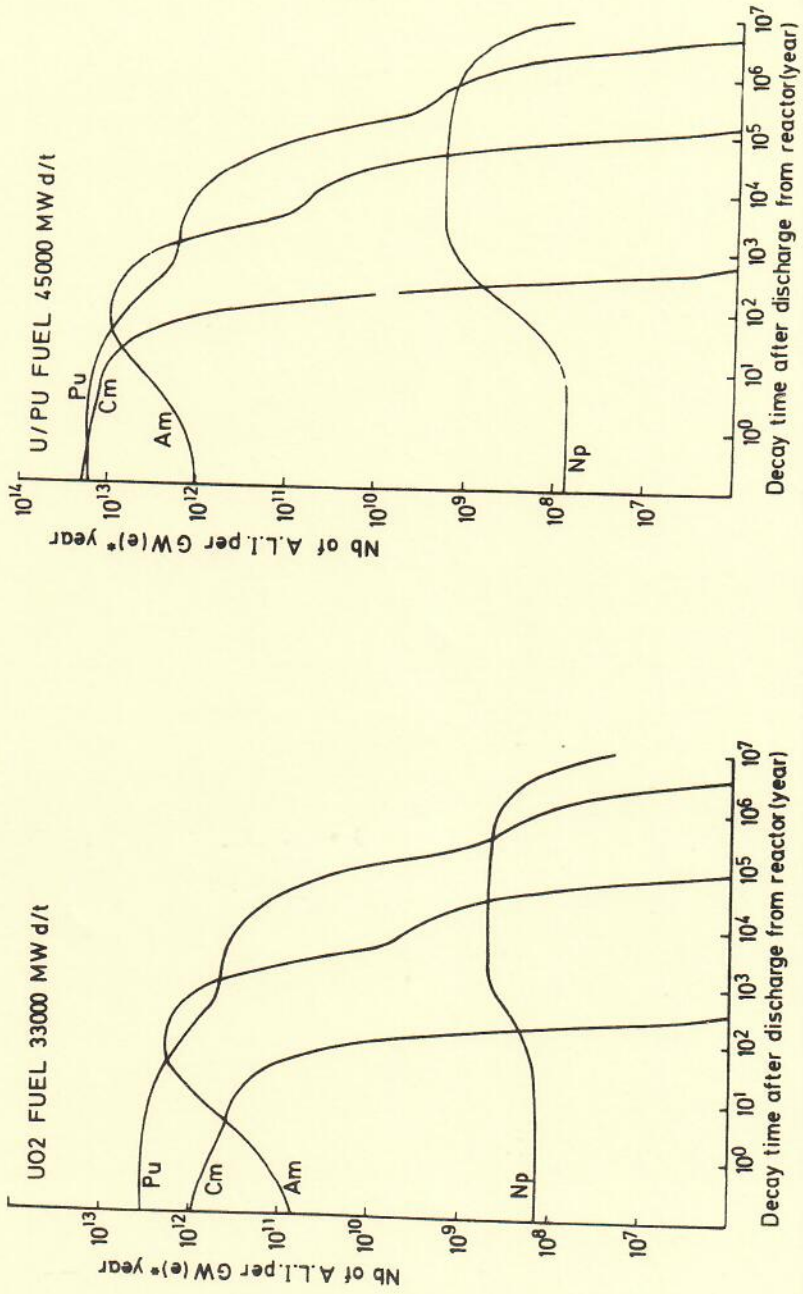

FIGURE 1. Normalized radiotoxicity of Np, Pu, Am and Cm in the standard spent fuel discharged from a 33,000 MWd/t LWR (1,a), in the Mox spent fuel discharged from a 45,000 MWd/t LWR (1,b). From ref. 6 and 7.

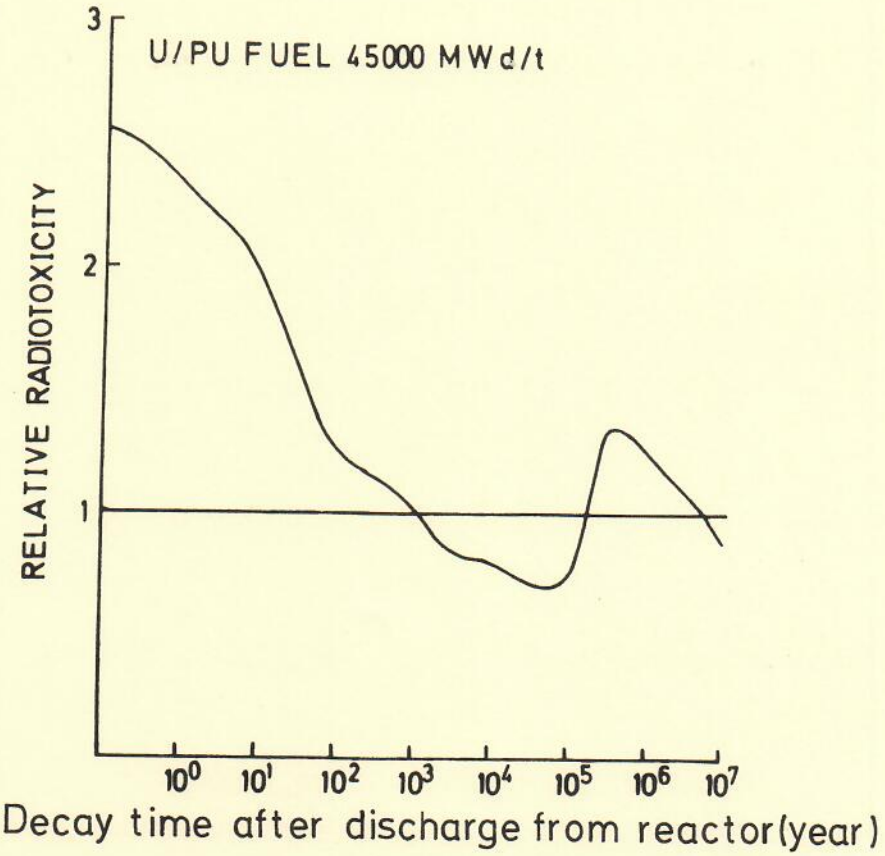

FIGURE 2. Ratio of normalized radiotoxicities of a Mox spent fuel at 45,000 MWd/t LWR and of the same non irradiated Mox fuel. This gives the simple transmutation effect.

LWRs. Considering the realistic following conditions: 5.25% PuO_2 enrichment of one third of the total fuel assemblies, we get an average value of about 17 kg of Pu per ton of fuel. In the LWR spent fuel, there is about 10 kg of Pu per ton of uranium fuel and about 40 kg of Pu per ton of Mox fuel. The total amount of plutonium is not reduced: plutonium recycling in LWR cannot lead to the suppression of Pu. Moreover, about 13 kg of this plutonium is degraded with regard to its further utilisation in LWRs, because it is issued from a second recycling as will be discussed hereafter.

Let us note that if the same amount of electricity had to be generated with uranium fuel alone, the amount of plutonium produced (10 kg/t) would have been larger than the increase in the amount of plutonium as calculated in the previous paragraph: there is some saving in long term potential radiotoxicity due to the use of Pu as discussed in ref. 7. In contrast, the absolute amount of Pu generated by the whole fuel increases as mentioned previously. This is also the case of the radiotoxicity of the whole fuel as will be shown in next paragraphs. Let us note that we have not taken into account the short lived fission products and the short term risks due to the reprocessing of the fuel.

QUANTITATIVE ASSESSMENT AND COMPARISON OF POTENTIAL RADIOTOXICITIES

In order to assess in a quantitative manner the potential radiotoxicity variation due to the recycling, let us call $R_1(t)$ the radiotoxicity of the Pu of a Mox fuel at a time t (assuming that it has not been irradiated) and $R_2(t)$ the radiotoxicity of the same Mox part of the fuel (assuming that it has been irradiated in the reactor). In Figure 2, we plot the variation of the ratio $R_2(t)/R_1(t)$ with time. Even though this ratio indicates whether the radiotoxicity increases or decreases at the different times of the future, it does not take into consideration the fact that the values of $R_1(t)$ and $R_2(t)$ change drastically over the range of time under consideration. The impact of the irradiation upon the radiotoxicity of the fuel can be evaluated numerically by introducing the parameter ΔR, the average change in the potential radiotoxicity of the Mox fuel over a period of time T between t_1 and $(t_1 + T)$:

$$\Delta R = \frac{1}{T}\int_{t_1}^{t_1+T} [R_2(t) - R_1(t)]\, dt$$

Having chosen a period of time T, this integral allows us to assess quantitatively the influence of any transmutation operation over the average potential radiotoxicity of any radioactive material. The sign of ΔR indicates whether there is an increase or a decrease in the average radiotoxicity taken over the period of time T. This complements the results of Figure 2 which indicates that some long term potential risks have been changed into a shorter term risk (up to several hundred years) and that there is some potential radiotoxicity increase at the very long range term because of the neptunium. In this study we shall consider a period of time equal to 10^7 years following the discharge of the fuel. From the value of $\Delta R/R \simeq -10\%$ corresponding to the conditions of Figure 2, we can conclude that the change in the average potential radiotoxicity due to plutonium recycling in the Mox part of the fuel is relatively small.

Considering the total fuel (one third of a Mox fuel and two thirds of a standard uranium fuel), let us call $R_3(t)$ and $R_3'(t)$ the radiotoxicities of the Mox and the standard fractions of the spent fuel discharged from a LWR. $R' = [R_3(t) + R_3'(t)]/R_1(t)$ is the ratio of the radiotoxicity of the spent fuel and the radiotoxicity of the plutonium in the non-irradiated fuel. Assuming for example that the recycling takes place in a 45,000 MWd/t, LWR, the radiotoxicity increases significantly for all the periods of the future as shown from the plot of R' in Figure 3. A quantitative assessment of the change in the average fuel radiotoxicity over the period T can be obtained from the ratio $\Delta R''/R''$ where:

$$\Delta R'' = \frac{1}{T}\int_{t_1}^{t_1+T} [R_1(t) - R_3(t) - R_3'(t)]\, dt \quad \text{and} \quad R'' = \frac{1}{T}\int_{t_1}^{t_1+T} R_1(t)dt$$

We find a value $\Delta R''/R'' = 180\%$, showing that the average radiotoxicity of the spent fuel taken over a period of time equal to 10^7 years is about two times larger than that of plutonium in the non-irradiated fuel.

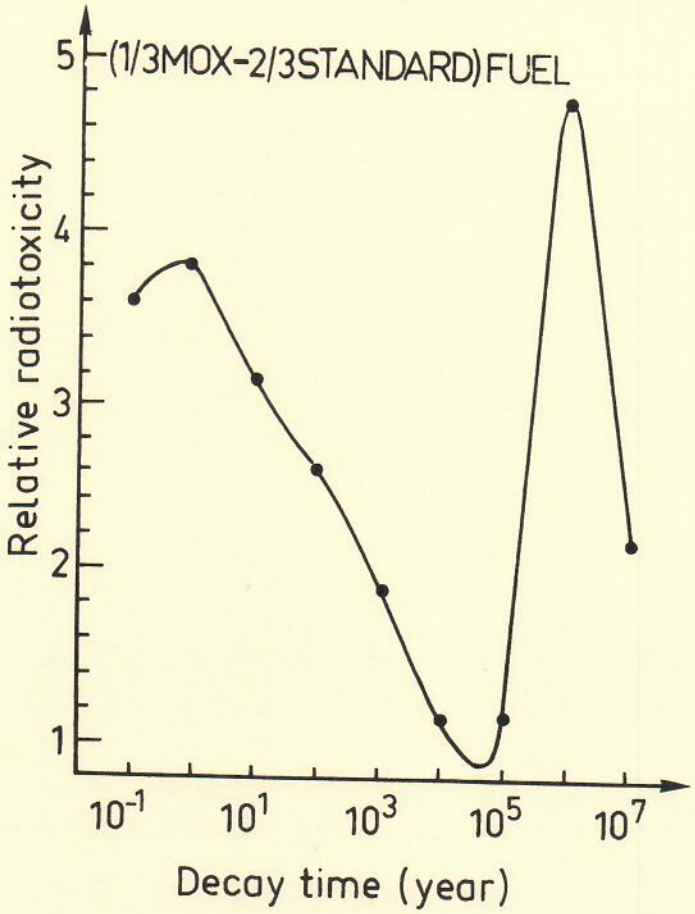

FIGURE 3. Same ratio as in Figure 2 for the total fuel cycle make up (⅓ Mox ⅔ standard fuel).

SUCCESSIVE RECYCLINGS OF PLUTONIUM

What is the effect of successive recyclings upon plutonium? It is well known that more and more heavy isotopes will be produced as a function of the irradiation time in LWRs because of the relatively high absorption cross-section values for thermal neutrons. In particular, the amount of non fissile ^{242}Pu will degrade significantly the quality of the plutonium for its further utilisation as a fuel in these reactors. For example, Table III shows the percentage of Pu necessary to produce a fuel equivalent to a 3% U enriched fuel as a function of the number of recyclings in the reactor.[8]

Even though successive recyclings of such amounts of plutonium are not feasible from the technical point of view, Table III is nevertheless interesting because it shows to what extent the quality of the plutonium would be degraded from one recycling to the other. This degradation is so important that even plutonium from

TABLE III
Percentage of Plutonium from the *n*th Recycling to be Mixed with Natural Uranium to Obtain a Fuel Equivalent to a 3% U Enriched Fuel

n	1	2	3	4	5	6	7	8
%Pu	4.5	15.5	25.7	30.9	35.5	40.3	45.5	51.3

a second recycling has too poor a quality to meet the fuel quality requirements of light water reactors. In order to overcome this difficulty, mixing fuel from different origins is under consideration for the future in different countries and in particular by COGEMA, the French fuel fabrication and reprocessing company. Thus the UP2-800 reprocessing plant has been designed with two shearing and dissolution lines to operate parallel reprocessing of Mox and standard LWR fuel, the dissolution being mixed before the solvent-extraction stage. However, if this mixing changes the relative amounts of the different isotopes of plutonium in the final fuel, it does not change of course the quality of the fraction X extracted from a fuel after a second recycling; this fraction should be quantitatively 3 times larger than an equivalent fraction of plutonium originating from a fuel irradiated once only. This will limit this fraction X to a relatively low value because of technical reasons related to LWRs. As far as the amounts of Mox fuel to be reprocessed in the available and under construction facilities which cannot reprocess pure spent Mox fuels, it is proposed that one sixth only of the reprocessing capacity can be used for spent Mox fuel, this ⅙th fraction must be mixed with another ⅚th fraction of standard fuels. Successive recyclings of a significant fraction of plutonium are far from being possible at present.

Even though not enough data are available to us about second recyclings in the actual LWRs, what can be concluded at present is that a part of the fuel (two thirds for the time being, maybe one half in the future?) will consist of enriched uranium. This will produce in all cases a certain amount of Np, Pu, Am and Cm. One third of the fuel will contain Pu originating from a fuel irradiated once (first recycling); however, a smaller fraction of it may be issued from a second recycling. . . . In these conditions, the amount of plutonium is larger in the spent fuel than in the fresh one. If some improvements concerning the amount of recycled plutonium can be made, this does not seem to be significant under present day conditions and options.

PLUTONIUM RECYCLING IN AN INERT MATRIX

Since the recycling of plutonium in a uranium matrix cannot lead to the progressive suppression of this element, does the fact that we use an inert matrix can lead to this suppression, keeping in mind that the recycling must take place within present options, that is in light water or equivalent reactors? M. P. Reuss (CEA-Saclay) kindly performed a simplified study to assess the feasibility of such a recycling.[9]

We summarize in the following paragraphs the main results; the main assumptions used being the following ones:

a): One group theory and homogeneous lattice. The parameters for the neutron balance and the evolution equations are averaged over the neutron spectrum and over the total volume of the assemblies under consideration.

b): Constant cross-sections: since the percentages of the different isotopes of plutonium do not change much, the neutron spectrum does not change either, and the average cross-sections do not vary much with time. Their values correspond to a mid-time irradiation situation of a Mox fuel in a depleted uranium matrix recycled in a light water reactor (see Table IV).

c): Constant power: the power is considered to be the same in the different types of fuel assemblies.

Replacing the only Mox part of the fuel by plutonium in an inert matrix and taking the amount of plutonium in the Mox fuel (Ro) as a reference, we get the following results: the amount of plutonium necessary to have in an inert matrix a 3 batch core with a 3-year fuel residence time is approximately 1.2 Ro; however the total absorption cross-section of the fuel is too large making this option unrealistic. If the number of cycles could be reduced from 3 to 2, the amount of plutonium necessary for the fuel becomes equal to 0.94 Ro and the total macroscopic absorption cross-section becomes slightly smaller than that of the Mox fuel. This, however increases the cost of the fuel (smaller burn-up) besides the R & D programme to study the feasibility in a real situation. If the number of cycles could be reduced to one, a larger amount of the total fuel (may be ½ or ⅔) may be replaced by plutonium in an inert matrix; however this increases even more the fuel cost. Since this model does not give the amounts of Np, Am and Cm produced during the burn-up, we have not made a detailed radiotoxicity study.

Nevertheless, whatever the number of cycles could be, the quality of the plutonium is so much degraded during the irradiation (Table V) that it cannot be recycled again as a fuel, even after mixing it with another fresh plutonium part drawn from a fuel irradiated once only.

In short we can say that since plutonium is not regenerated within the matrix, a larger amount of plutonium is "burned" in the fuel with the inert matrix. The plutonium left after burn-up is equal to 0.68 Ro and 0.55 Ro in the cases of a one and a two cycle options instead of 0.79 Ro in the case of a Mox fuel. However, the use of an inert matrix does not lead to the progressive suppression of plutonium as any transmutation programme should do.

TABLE IV

Cross-Sections in Barns at a Mid-Time Irradiation Situation of a Mox Fuel in a Depleted Uranium Matrix Recycled in a LWR Reactor

Isotope	238	239	240	241	242
Production	0.336	105.1	1.8	123.9	1.5
Absorption	0.939	56.8	41.0	56.6	25.6
Capture	0.820	20.2	40.4	14.8	25.1

TABLE V

Plutonium Isotopic Composition in Parts Per Thousand at the End of the Irradiation of Plutonium Fuel in an Inert Matrix Recycled in a LWR. One Cycle Corresponds to a One Year Exposure

Conditions	Concentrations	239	240	241	242
Initial (UO_2)	1	598	231	113	57
Mox/3 cy.	0.788	444	277	192	86
Pu-inert/1 cy.	0.681	386	314	207	92
Pu-inert/2 cy.	0.551	295	329	255	121
Pu-inert/3 cy.	0.470	240	329	285	146

CONCLUSION

Reactor transmutation, considered as a procedure for progressive elimination of all the long half-life radioisotopes produced during the fuel burn-up is not feasible with present technological options. Figure 2 shows that a part of the mid-term radiotoxicity is transformed into a shorter term one in the Mox part of the fuel, producing a 10% reduction in the average radiotoxicity of this part; (taken over a period of 10^7 years) but this can only be obtained if a larger amount of radiotoxicity is generated in the total fuel. Besides, the handling of the highly neutron and gamma-ray active fuel during the reprocessing and the presence of isotopes with high absorption cross-sections in the reactor increase the short term risks. Inert matrix does not bring a satisfactory solution either.

Insight to plutonium recycling cannot be found in serious transmutation effects but rather in energy production and uranium saving. Unless new technological options appear, long half-life isotopes will accumulate for quite a long time in the different reactor fuels.

REFERENCES

1. I.C.R.P. Publication 2, *Report of Committee II on Permissible Dose for Internal Radiation*, Pergamon Press, New York, 1959.
2. I.C.R.P. Publication 6, *Recommendations of the International Commission on Radiological Protection*, Pergamon Press, New York, 1964.
3. I.C.R.P. Publication 30, *Limits for Intakes of Radionuclides by Workers*, Pergamon Press, New York, 1979, 1981.
4. I.C.R.P. Publication 26 (1977), "Recommandations de la C.I.R.P.," Pergamon Press, Paris, 1980.
5. I.C.R.P. Publication 48, *The Metabolism of Plutonium and Related Elements*, Pergamon Press, New York, 1986.
6. A.G. Elayi and J.P. Schapira, *Radioactive Waste Management and the Nuclear Fuel Cycle, 8*, 327 (1987).
7. A.G. Elayi and J.P. Schapira, IPNO-DRE-87-07.
8. P. Reuss, "Modèle pour les études de cycles de combustible nucléaire code cyclone," Rapport SERMA/SPM n°330"S".
9. P. Reuss, Etude du recyclage du plutonium sur support inerte dans les réacteurs à eau sans pression, Internal Report CEA/SERMA/S/1047.