institut de physique nucléaire
LABORATOIRE ASSOCIÉ A L'IN2P3

LONG-TERM RADIOTOXICITY; IN THE FRAMEWORK OF THE ICRP-48, OF HIGH LEVEL WASTES AND SPENT FUELS PRODUCED BY LIGHT WATER REACTORS : IMPACT OF BURN-UP EXTENSION AND OF THE USE OF MIXED OXIDE FUELS.

A.G. ELAYI and J.P. SCHAPIRA
Institut de Physique Nucléaire, P.Box 1, 91406 Orsay (France)

IPNO-DRE 87-07

UNIVERSITÉ PARIS-SUD
I.P.N. BP n° 1 - 91406 ORSAY

LONG-TERM RADIOTOXICITY, IN THE FRAMEWORK OF THE ICRP-48, OF HIGH LEVEL WASTES AND SPENT FUELS PRODUCED BY LIGHT WATER REACTORS: IMPACT OF BURN-UP EXTENSION AND OF THE USE OF MIXED OXIDE FUELS.

A.G.Elayi and J.P.Schapira

Institut de Physique Nucléaire, P.Box 1, 91406-Orsay (France)

ABSTRACT

The time evolution of radiotoxicities of spent fuels and high-level wastes have been calculated up to 10^7 years, in the framework of the recent I.C.R.P.-48 guideline, in which the ratio dose/ingested-activity has been divided by 10 for Np, multiplied by 10 for Pu and by 2 for Am, Cm and Cf, with respect to the previous I.C.R.P.-30 values. In the case of burn-up extension of the standard 33,000 MWd/t enriched uranium fuel, and in the case of plutonium recycle in light-water reactors, one shows that the spent fuel radiotoxicity is now dominated by its plutonium content, most of the time up to about 10^5 years. Possible incineration effects are discussed within these two fuel cycle options

1. Introduction

For many years, extensive work has been done on radioactive materials decay characteristics, such as isotopes and chemical inventories, heat emission and radiotoxicity. Most studies refer to milling wastes, spent irradiated fuels and reprocessing wastes related to the use of light water reactors with no recycle of uranium and plutonium, and to a less extent, with recycle, as well as to those wastes coming from other types of reactor (fast neutron breeder, Magnox and Candu) (see e.g ref.1-4). Moreover, most of the radiotoxicities were calculated in the framework of the first I.C.R.P.-2/6 guideline (refs.5,6), published by the International Commission on Radiological Protection, and in a few cases (refs.7-9) in the framework of the recent I.C.R.P.-30 guideline (ref.10). In a near future, the latter one will be taken as the basis for nuclear safety and radioprotection regulations.

The present study has been initiated during the work of the so-called "Castaing Commission" (refs.11,12), an official panel of experts of various origins, chaired by Professor R. Castaing, which acted during the period 1981-1984 as an advisory committee for the "Conseil Supérieur de Sûreté Nucléaire" (C.S.S.N) on the subject of irradiated fuels and radioactive wastes management. At that time the study was conducted in order to assess the impact of various schemes of minor actinides extraction (neptunium, americium and curium), on the long-term

radiotoxicity of the resulting high level wastes (ref.12). A comparison was made between the standard pressurized water and the fast neutron reactors at an average burn-up of 33,000 and 75,000 MWd/t* respectively. The present work** deals now with the characterization of the radiotoxicity of high level wastes (HLW) and spent fuels produced:

- first in the case of the use of fresh enriched uranium in a pressurized light water reactor at an extended average burn-up of 45,000 MWd/t.
- secondly in the case of the first recycle of plutonium, that is to say the use of mixed oxide fuels (MOX fuel: UO_2 - PuO_2), in the same type of reactor and at the same average burn-up.

As a matter of fact, since the U.S. decision in 1976 to defer reprocessing indefinitely, there has been a continuous incentive to extend the average discharge burn-up to values higher than the current one of 33,000 MWd/t. Whereas at that time, saving uranium was probably

(*) : MWd/t (Megawatt(thermal)*day per metric ton of heavy metal) is the usual unit of burn-up of a discharged fuel.

(**) : This work is part of a scientific program on reprocessing and radioactive waste management, supported by the Centre National de la Recherche Scientifique (C.N.R.S.).

the basic reason for such an extension, it appears now, with the decrease of uranium price since 1981, that the major incentives are reactor availability and operationnal cost as well as personnal exposure decrease, through larger reactor cycle lengths (see e.g. refs.13,14). With an average burn-up of 45,000 MWd/t, contemplated now by the French electrical utility, Electricité de France (E.D.F), for some of its reactors (ref.15), the cycle length could in principle be increased from 12 to 18 months.

On the other hand, the use of plutonium as fissile material in thermal reactors has been considered since the 1950's, i.e since the launching of this reactor line. Widely and openly discussed during the 70's in the U.S. with the G.E.S.M.O. exercise (ref.16), plutonium recycle remained since at an experimental and marginal stage, due, in particular, to lack of reprocessing facilities and also because of the generally accepted view that plutonium should be used in fast neutron reactors, in a much more efficient way. It appears that this situation might now evolve (see e.g refs.14,17), at least outside the U.S., with the recent success, as far plutonium production is concerned, of the reprocessing plant UP2-400 at La Hague (France) and the prospect of large facilities in various countries such as the U.K. (Thorp), France (UP3), the F.R.G. (Wackersdorf) and Japan (Aomori). With the expected availability of significant quantities of plutonium at a time when there is a general slowing-down of fast breeder programs (see e.g. ref.17), including in France, some

electrical utilities, in Europe and Japan, consider now using mixed oxide fuels, to a limited extent, as partial substitute to U-235 in their actual fuel assemblies.

The present study aims at evaluating the long-term radiological impact associated with these fuel cycle options, which very likely will reach an industrial stage in a near future. The waste products of high activity, generated in these two options, are of two types: spent fuels in the case of an open cycle, and high level wastes (ultimately in the form of vitrified glasses) in the case of reprocessing, the difference being that, in the latter option, practically all the uranium and plutonium have been removed. The amount of waste products of each type will depend on many factors: decision to reprocess high burn-up fuels in order to recover the plutonium content, decision to proceed with further plutonium recycle in light-water reactors (use of the second generation plutonium) or to use it for a fast breeder program. Although for the time being, it appears that spent MOX fuels will be stored after one recycle without reprocessing, radiotoxicity decays of both spent fuels and high level wastes will be presented in this work.

Whereas most previous calculations used earlier radiological data (refs.5,6), this work takes into account the last I.C.R.P.-48 values (ref.18), which include substantial changes ,with respect to the I.C.R.P.-30 (ref.10), for every transuranians radiotoxicity.

2. Nuclear waste radiotoxicity and the I.C.R.P. recommendations

The long-term radiological risk is associated with the possible release into the biosphere of radioactive substances which have been dumped in a final disposal site. The world-wide adopted solution for the nuclear wastes generated during post-fission operations, lies in the final subsurface and deep geological disposal of either the spent fuels or all the wastes produced first during the reprocessing of these spent fuels (vitrified glasses, low and medium active wastes) and secondly during the possible subsequent use of the extracted plutonium and uranium.

Two types of risk are usually considered:

- the potential or maximum risk directly associated with the possible ingestion, by a group of persons, of the entire content of the wastes, mainly through drinking water.
- the real or residual risk which takes into account all the artificial and geological barriers which, to a large extent, can reduce the amount of radionucleides reaching human beings at various times of the future.

The potential risk is a useful concept, when one wants to compare long-term radiological risks for different reprocessing parameters (e.g. cooling times before reprocessing, extraction performances of certain radionucleides such as long-lived alpha-emitters), or for different fuel cycle options, as in the case of the

present work. We therefore have calculated the potential risk of the two types of wastes produced in the two options mentioned above, as well as in the standard case at 33,000 MWd/t.

Whereas real risk estimations finally depend on many geological, geophysical and geochemical parameters, as well as on transfer models, the potential risk results from the knowledge of the waste inventories and of the radiological impact of each radionucleide after ingestion.

The latter can be easly deduced from the annual limit of intake by ingestion (A.L.I.) of each radionucleide. These limits, published in the recent I.C.R.P.-30 guideline (ref.10), concern the people working in the nuclear industry and therefore correspond to an annual equivalent dose limit of 50 mSv (1 Sievert= 100 rems). With respect to the previous A.L.I. given in the I.C.R.P.-30 (ref.10), important modifications concerning all the transuranians have now been introduced. From recent metabolic studies, it has been proposed, in the I.C.R.P.-48, that the fractional absorption from the gastrointestinal tract for population exposure to all compounds, should be taken equal to $f_1 = 10^{-3}$ for Np, Pu, Am, Cm and Cf. This follows from the fact that the radiological impact of neptunium was largely overestimated, in the previous I.C.R.P.-30, by roughly a factor of 10. The inverse situation was found for plutonium for which an increase of severity by a factor of 10 has been adopted. On the other hand, the radiotoxicity

increases by a factor 2, for americium, curium and californium.

Because one is dealing here with long-term risks, one has to divide by 10 these A.L.I. values, in order to take into account the 5 mSv annual limit for the public. Some values of A.L.I., together with half-lives, are reported in table 1.

The radiotoxicity T_i of a radionucleide i of activity A_i can then be expressed by $T_i = A_i/(A.L.I.)_i$. The total radiotoxicity T(t) of a waste containing various radionucleides i of activity $A_i(t)$ at time t, is therefore obtained by summing over all the isotopes i:

$$T(t) = \Sigma_i \, A_i(t)/(A.L.I.)_i$$

where the A.L.I. coefficients refer to the public.

Since the final goal of power reactors is to produce electricity, all these quantities have been divided by the amount Q of electricity produced (in units of GW(e)*year, which corresponds to the annual production of a 1000 MW(e) reactor operating at 100 %). The energy conversion efficiency of the reactor has been taken equal to 1/3. Moreover, this normalization seems a proper way to compare different fuel cycles or different parameters within a certain fuel cycle, with respect to long-term radiotoxicity. We have given the total radiotoxicity R=T/Q, normalized in this way, together with the partial radiotoxicity of the chemical elements which are the major contributors through one or more of their isotopes. As pointed out by W.Bocola (ref.19), this approach is justified by the fact that transfer properties through

artificial and geological barriers depend directly on the chemical elements inventory.

3. The computer program for decay studies of radioactivity and radiotoxicity, and the input data

A computer program called ISODEC has been written for the Univac-1100/92 computer of the University of Paris-Sud. It solves the Bateman's equations (ref.20), which govern the radioactive decay of a set of nuclei coupled by radioactive decays. The exact solution of these equations can easly be obtained using the Laplace transform, a well-known method which turns out to be well suited for computer writing. Because one is interested in the long-term radiotoxicity, all the nuclei from californium to bismuth have been included, as well as those fission products which have a long lifetime (Tc-99, I-129 and Cs-135). Moreover, the isotopes Sr-90 and Cs-137 have also been considered because of their important contribution to the short term radiotoxicity. A special subroutine determines all the decay chains involving each of the transuranic nuclei, by first selecting those which are the beginning of one of these chains. For each isotope and chemical element, constituting the waste, the program calculates the activity, the number of nuclei, and the radiotoxicity as function of time, up to 10^7 years. The program takes also into account the reprocessing that may

take place at a certain date, and therefore it can handle various types of wastes (spent fuels, HLW, low and medium active wastes). Finally, a few other features have been built in the program, such as the possibility of making direct comparisons of the studied type of waste with a standard type (e.g. the 33,000 MWd/t fuel cycle).

The input data, i.e. the isotopic composition of the discharged fuel, have been taken from internal reports of the Commissariat à l'Energie Atomique (C.E.A.) (refs.21-25). In these reports, the evolution of the isotopic composition of a fuel during its irradiation in the reactor is calculated with the computer codes APOLLO (for the transuranians) and PEPIN (for the fission products). The fuel parameters taken in these reports are those of realistic fuels that E.D.F. plan to use in the near future. These codes have been developed by the C.E.A. (refs.26,27). The other data (half-lives and radiotoxicities) have been taken from refs.10,18 and 28.

4. General results and remarks

The three types of irradiated fuels, studied in this work under the label A,B,C, are or will be discharged from the pressurized water reactors currently operating in France. Fuels A and B use enriched fresh uranium, and correspond respectively to the standard average burn-up of 33,000 MWd/t, and to an extended average burn-up at 45,000 MWd/t. Fuel C uses mixed oxide at an average burn-up of

45,000 MWd/t. The plutonium loaded in the latter is extracted during the reprocessing of fuel A. The reprocessing parameters (a cooling time of 3 years and a removal efficiency of 99.9, 2.3 and 99.6 % for uranium, neptunium and plutonium respectively) are the expected one for the future plant UP-3 at La Hague (France) (ref.12). No further recycle of the plutonium has been taken into account, since one recycle only is contemplated at the present time in the French program. The isotopic composition of these three types of fuel, when they are being loaded in the reactor, is given in table 2.

Concerning the radiotoxicity decay of the 3 types of spent fuels A,B and C, three different time periods can be considered on figs.1a to 3a: the first one, the short range period lasts about 500 years; the second one, the medium range period, lasts from about 500 years to 500,000 years; the third one, the long range period is beyond 500,000 years.

Up to 500,000 years, and for the three types of fuels, the radiotoxicity is dominated by the various plutonium isotopes (Pu-241 and Pu-238 for the first few hundred years and then Pu-239 and Pu-240). The relatively long lifetime fission products do not contribute significantly to the radiotoxicity of the spent fuels.

Beside the major contribution of plutonium, the radiotoxicity, during the first short term range, is dominated by Sr-90 and then by Am-241 in the case of fuels A and B. In contrast, the radiotoxicity of fuel C is dominated by curium and americium, because of much larger

capture effects on the initial plutonium content of the fuel and of a lower strontium thermal neutron fission yield with Pu-239/241 as compared with U-235.

During the medium term range and after a certain period of time over which americium dominates or is equivalent to plutonium, this latter element dominates completly the radiotoxiciy of the fuel.

Afterwards, one observes the increasing contribution of neptunium (produced essentially by Am-241 α-decay) and of the various other decay products (Pb, Ra, Th and Pa) of the transuranic nuclei.

The picture is of course completly different for the HLW (figs 1b to 3b), for which four periods of time can be observed: the first one, up to few hundred years, is dominated by strontium, the next ones by americium (few 10^2 - 10^3 years), plutonium (few 10^3 - 10^5 years) and neptunium (after few 10^5 years)

5. Discussion on the radiological impact of the two fuel cycle options B and C

5.1 The case of extended burn-up

The impact on the radiotoxicity of the burn-up extension can be seen on fig. 4, which represents the time variation of the ratio:

$$r_{b,a}(t) = R_b(t)/R_a(t)$$

of the normalized radiotoxicities $R_b(t)$ and $R_a(t)$ in the case of an open cycle (spent fuels) and in the case of reprocessing (vitrified glasses). One sees (fig.4) that at short term, the radiotoxicity of the discharged spent fuel of type B is approximately 20% higher than that of type A. Afterwards the difference decreases, vanishing after about 300 years; the radiotoxicity of type A then becomes slightly larger. This can be explained in the following manner: the high burn-up produces larger quantities of heavier isotopes (Pu-241, Cm-242, Cm-244) of relatively shorter half-lives. Moreover, there is an effect on the increase of radiotoxicity, due to the α-decay of Cm-242 to Pu-238 (see the A.L.I. values in tab.1). Beside, the presence of curium has a short term effect on occupationnal exposure (see e.g. ref.29). When these effects disappear, the radiotoxicity is due to the isotopes Pu-239/240, which are slightly higher under low burn-up conditions. After a long period of time, the radiotoxicity of fuels A and B fluctuates according to the presence of the lighter elements in the different chains.

In the case of vitrified glasses (fig. 4), the conclusions are different. Because plutonium is now removed, the radiotoxicity of fuel B remains always higher, due to larger quantites of americium and curium at the time of discharge. These relatively short-life nuclei are responsible for the long-term and very long-term radiotoxicity through their decay products (essentially neptunium and plutonium).

5.2 The case of the use of mixed oxide

There are two approaches in assessing the long-term radiological impact of the utilization of plutonium oxide as a fuel instead of uranium oxide in light-water reactors. The first one considers an open fuel cycle option, without reprocessing, for the fuel A, and therefore the C option as independent of A. In the second one, the two options are treated together in the sense that reprocessing of the spent fuels A produces the plutonium partly consumed in the fuel cycle C.

In the first approach, the comparison between the two fuel options A or B without recycle is represented by the time variation of the ratio:

$$\Gamma_{c,b}(t) = R_c(t)/R_b(t)$$

where one has taken the fuel B as reference in order to make the comparison at the same average burn-up of 45,000 MWd/t. This ratio is represented on fig.5. Because of a large plutonium content in the fuel C when loaded in the reactor, the radiotoxicity of the MOX spent fuel remains larger by a factor varying between 3.3 and 4.6, up to 10^6

years. This enhancement is even more important if one compares HLW in the same way. As in the previous case of §5.1, this is directly related, through their decay products, to the larger quantities of curium and americium which are not removed during reprocessing.

In the second approach, one takes into consideration the incineration effect introduced by the use of mixed oxide.

One can first wonder wether recycling plutonium in a light-water reactor has a direct incineration effect, as far as radiotoxicity is concerned. For this purpose, one considers the time variation of the ratio of the radiotoxicity of the spent fuel C to the radiotoxicity of the same non irradiated fuel C_o, radiotoxicity which is largely dominated by the initial plutonium content (fig. 6):

$$r_{c,co}(t) = R_c(t)/R_{co}(t)$$

One sees the influence of short lifetime isotopes (Cm-244/242, Pu-241/238) during the first short term period, responsible for an increase of the radiotoxicity of the spent fuel C with respect to that of the non irradiated fuel C_o. The incineration effect becomes slightly beneficial between few 100 years and about 10^5. This is due of course to the long-period plutonium isotopes incineration. After 10^5, the relative radiotoxicity increase is due to neptunium and the end chain products, which are more important in the spent fuel. However at this time of the future, the radiotoxicity has been reduced by some order of magnitudes. One can hardly speak

of an important incineration effect. This is not surprising, considering the short irradiation time (3 years). As it is known, much longer irradiation times are needed to observe a significant incineration effect, which means in practice the need for more than one recycle.

Secondly one can even consider the whole operation of fuel cycles A and C. Indeed, one notices that the amount of plutonium needed for one metric ton (Mt) of fuel C comes from the reprocessing of approximately 6.8 Mt of spent fuel A. One has neglected the elapsed time, of the order of 2 years (refs.24,25), between the separation of that plutonium and its utilization, as well as the time of irradiation in the reactor (3 years), because one is here concerned with long term effects. By coupling both fuel cycles, the normalized radiotoxicity $R_{ca}(t)$ can be expressed by the following relationship:

$$R_{ca}(t) = T_{ca}(t)/Q_{ca}$$

where $T_{ca}(t)$ represents the net "production" of radiotoxicity during the operation of 6.8 cycles A to produce the needed plutonium and one cycle C, and Q_{ca} the total amount of electricity produced.

$$T_{ca}(t) = 6.8*T_a(t)+[T_c(t)-T_{co}(t)]$$

$$Q_{ca} = 6.8*Q_a+Q_c$$

As in the case of extended burn-up, one can now compare this closed fuel cycle (A+C), with only one recycle, to the open fuel cycle (A), using the time variation of the ratio

$$\Gamma_{ca,a}(t) = R_{ca}(t)/R_a(t)$$

This ratio (fig.7) evolves over time in a way similar to the one of the extended burn-up (fig.4). The incineration effect observed after 100 years can be explained in the same way.

6. Conclusion

Taking only into account potential risks expressed by the number of A.L.I. content of a waste, associated with the production of a certain quantity of electricity, some general conclusions can be drawn.

The radiotoxicity of the three types of spent fuels considered in this work, is now completly dominated by the plutonium up to some 10^5 years. This, of course, is due to the recent enhancement by a factor 10 of the plutonium A.L.I.. As a matter of fact there has been a continuous increase of severity for plutonium from the first I.C.R.P.-2/6 to the recent I.C.R.P.-48. On the other hand, neptunium, whose impact was largely overestimated in the I.C.R.P.-30, has a significant contribution to the total radiotoxicity only after 10^6 years, and at that time it becomes equivalent to that of the end chain products.

Burn-up extension without reprocessing, shows a slight increase of radiotoxicity up to about 300 years and then a beneficial effect on long-term radiotoxicity.

As far as MOX fuel is concerned, the radiotoxicity of spent fuels (per ton or per GW(e)*year) produced with plutonium first recycle is higher than the corresponding values of the 45,000 MWd/t UO_2 fuel cycle, by a substantial factor of the order of 4. A factor even higher, of the order of 10, is obtained if one compares HLW in the same way. This points out the interest of removing minor actinides in the reprocessing of MOX spent fuels (see e.g. ref.12). Nevertheless one can consider

that, using plutonium in MOX fuel, has an effect on reducing the radiotoxicity. With respect to this point two scenarios can be examined:

In the first scenario, one considers that large amounts of plutonium are available as nuclear wastes of high radiotoxicity and that the aim of using them is, among others, to destroy a part of their radiotoxicity. If a large amount of radiotoxicity is effectively present in MOX spent fuels, this is partially due to the initially loaded plutonium in the reactor. The global effect of the irradiation has been shown to be beneficial after a relatively short period of time, as far as radiotoxicity is concerned, especially if one keeps in mind that electricity has been simultaneously produced.

In the second scenario, one takes into account the fact that the MOX fuel needed for one light-water reactor cycle, will practically be obtained if plutonium is extracted from a certain number of reactor cycles at 33,000 MWd/t. The global radiotoxicity, resulting from this plutonium production in these light-water reactors and from the MOX spent fuels, can be directly related to the total amount of electricity produced. This is the case if the whole operation was planned in a global manner. It has then been shown that the global effect is similar to the one obtained by just increasing the burn-up from 33,000 MWd/t to 45,000 MWd/t without reprocessing.

One must however emphasize that in comparing long-term radiological effects of different fuel cycle options, one has to take into account all the wastes (and their

management) produced in the possible post-fission industrial operations. In the case of plutonium recycle, these operations (reprocessing, mixed-oxide fuel fabrication) are known to generate various other types of solid wastes of low and medium activity, which contain some plutonium at a usually very low concentration, depending on the performances of these operations. A more complete comparison should also take this into consideration. Finally, one must be aware of possible short term radiological impacts, because of the use of large amounts of plutonium in reactors, since in particularly, the radiotoxicity of the corresponding fuels is higher than the UO_2 fuel before, during and after irradiation in the reactor.

7. Acknowledgements

We are grateful to Messrs Schwartz J.P. and Nimal J.C. of the Service d'Etudes de Réacteurs et de Mathématiques Appliquées of the Commissariat à l'Energie Atomique (C.E.N-Saclay) for having supplied us with their calculated spent fuel data. Finally, we thank the colleagues of one of the author, during the "Commission Castaing", Dr Lafuma J. and Mr Zerbib J.C., for the last I.C.R.P. data they kindly conveyed to us.

References

1/ Clegg L.J. and Coady J.R., *Radioactive Decay Properties of CANDU Fuel. vol. 1: the Natural Uranium Fuel Cycle*, AECL-4436/1 and 4436/2, January, 1977.

2/ Ewart F.T., *Fuel Inventories and Derived Parameters for the Reactor Systems CDFR, LWR, AGR and Magnox*, AERE/R-10037, April, 1981.

3/ Haugh H.O., *Decay Calculations on Medium-Level and Actinide-Containing Wastes from the LWR Fuel Cycle. Pt. 2. Comparison of Radiotoxicity*, KFK-3222, December, 1981.

4/ Croff A.G., Liberman M.S. and Morrison G.W., *Graphical and Tabular Summaries of Decay Characteristics for Once-Through PWR, LMFBR and FFTF Fuel Cycle Materials. (Spent Fuel, High-Level Waste Fuel Can Scrap.)*, ORNL/TM-8061, January, 1982.

5/ *Report of Committee II on Permissible Dose for Internal Radiation*, I.C.R.P. Publication 2. New York: Pergamon Press, 1959.

6/ *Recommendations of the International Commission on Radiological Protection*, I.C.R.P. Publication 6. New York: Pergamon Press, 1964.

7/ Cohen B.L., *Effects of I.C.R.P. Publication 30, the 1980 B.E.I.R. Report, and ORIGEN-2 on Hazard Assessments of High-Level Wastes*, Trans. Am. Nucl. Soc., vol. 38, p.84, 1981.

8/ Croff A.G., *Potential Impact of I.C.R.P.-30 on the Calculated Risk from Waste Repositories*, Trans. Am. Nucl. Soc., vol. 39, p.74, 1981.

9/ Choi Jor-shan and Pigford T.H., *Water Dilution Volumes for High-Level Wastes*, Trans. Am. Nucl. Soc., vol. 38, p.176, 1981.

10/ *Limits for Intakes of Radionucleides by Workers*, I.C.R.P. Publication 30. New York: Pergamon Press, part 1 1979, part 2 1980, part 3 1981.

11/ *Rapport du Groupe de Travail sur la Gestion des Combustibles Irradiés*, Ministère de la Recherche et de l'Industrie, December, 1981 - November, 1982.

12/ *Rapport du Groupe de Travail sur les Recherches et Developpements en Matière de Gestion des Déchets Radioactifs*, Ministère du Redéploiement Industriel et du Commerce Extérieur, October, 1983 - October 1984.

13/ Bairiot H., *High Burn-up - Status and Expectations*, I.A.E.A.-CN-42/49, vol. 3, Nuclear Power Experience. Vienna, September, 1982.

14/ *Proceedings of a Specialists Meeting on Improved Utilization of Water Reactor Fuel with Special Emphasis on Extended Burn-up and Plutonium Recycling*, I.A.E.A.-IWGFPT-20. Mol (Belgium), 7-11 May, 1984.

15/ Lavoine O., Houdaille B. and Cambon J.L., *High Burn-up Program in the French Power Plants*, in reference 14/, p.-145.

16/ *Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors*, NUREG-0002/ES, NUREG-0002/Vol.1, NUREG-0002/Vol.2, and NUREG-0002/Vol.3, August, 1976.

17/ Bairiot H., *Laying the Foundations for Plutonium Recycle in Light Water Reactors*, Nuclear Engineering International, p. 27, January, 1984.

18/ I.C.R.P. Publication 48 *The Metabolism of Plutonium and Related Elements*, (Pergamon Press, New York, 1986)

19/ Bateman H., *Solution of a System of Differential Equations Occuring in the Theory of Radioactive Transformations*, Proc. Cambridge Philos. Soc. vol.15, p. 423, 1910.

20/ Bocola W., *Characterization of Long-Lived Radioactive Wastes to be Disposed in Geological Formations*, Part I and II, RT/PROT-18, 1978 and RT/PROT-19, 1979.

21/ Devillers C., Payan J.P. and Manesse D., *Activité β, α, Neutronique, Spectre d'Emission γ et Puissance Résiduelle d'un Combustible P.W.R. Irradié*, internal report S.E.R.M.A./S/314, June, 1977.

22/ Ermuncu G., Nimal B. and Nimal J.C., *Puissance Résiduelle et Spectre γ Emis par les Produits de Fission d'un Combustible Enrichi à 4.5 % et Porté à 45,000 MWj/t*, internal report S.E.R.M.A./T/439, July, 1980.

23/ Ermuncu G., *Emissions Neutroniques et α et Concentrations en Transuraniens d'un Combustible de Type P.W.R. Irradié à 45,000 MWj/t et Enrichi initialement à 4.5 %*, internal report S.E.R.M.A./T/471, June, 1981.

24/ Ermuncu G., *Emissions β et γ dues aux Produits de Fission d'un Combustible UO_2 - PuO_2 Porté à 45,000 MWj/t*, internal report S.E.R.M.A./T/468, June, 1981.

25/ Ermuncu G., *Emissions Neutronique et α d'un Combustible UO_2 - PuO_2 Porté à 45,000 MWj/t*, internal report S.E.R.M.A./T/472, June, 1981.

26/ Nimal J.C., *Note de Présentation du Code PEPIN*, internal report D.E.M.T. 86/050, S.E.R.M.A./LEPF/86/760, January, 1986.

27/ Hoffman A., Jeanpierre F., Kavenoky A., Livolant M. and Lorain H., *APOLLO, Code Multigroupe de Résolution de*

l'Equation de Transport pour les Neutrons Thermiques et Rapides, Note C.E.A. - N - 1610, 1972.

28/ *"Radionucleide Transformations" : Energy and Intensity of Emissions*, I.C.R.P. Publication 38. New York: Pergamon Press, 1983

29/ Mauro J.J, Eng R., Marschke S., Chang W. and Coleman A.C., *The Environmental Consequences of Higher Fuel Burn-up*, A.I.F./N.E.S.P. - 032, June, 1985.

Table -1-

Isotope	Half-life (year)	M.P.C. (Bq/m³) (refs.5,6)	A.L.I. (Bq) (ref.10)	A.L.I. (Bq) (ref.18)
Sr-90	28.15	$1.5\ 10^{4}$	10^{6}	/
Tc-99	$2.14\ 10^{5}$	$1.1\ 10^{6}$	10^{7}	/
I-129	$1.57\ 10^{7}$	$7.4\ 10^{3}$	$2\ 10^{4}$	/
Cs-135	$2.95\ 10^{6}$	$3.7\ 10^{6}$	$3\ 10^{6}$	/
Cs-137	30.154	$7.4\ 10^{5}$	$4\ 10^{5}$	/
Np-237	$2.14\ 10^{6}$	$1.1\ 10^{5}$	$3\ 10^{2}$	$3\ 10^{3}$
Pu-238	87.7	$1.8\ 10^{5}$	$3\ 10^{4}$	$3\ 10^{3}$
Pu-239	$2.41\ 10^{4}$	$1.8\ 10^{5}$	$2\ 10^{4}$	$2\ 10^{3}$
Pu-240	$6.55\ 10^{3}$	$1.8\ 10^{5}$	$2\ 10^{4}$	$2\ 10^{3}$
Pu-241	14.4	$7.4\ 10^{6}$	10^{6}	10^{5}
Am-241	432.6	$1.5\ 10^{5}$	$5.2\ 10^{3}$	$2.6\ 10^{3}$
Am-243	$7.38\ 10^{3}$	$1.5\ 10^{5}$	$5.2\ 10^{3}$	$2.6\ 10^{3}$
Cm-242	.446	$7.4\ 10^{5}$	$2\ 10^{5}$	10^{5}
Cm-244	18.11	$2.6\ 10^{5}$	$9\ 10^{3}$	$4.5\ 10^{3}$

Half-life, Maximum Permissible Concentration[6] and Annual Limit of Intake[10,18], for adults of the public, of the major transuranians and of the fission products contributing significantly to the radiotoxicity. These limits are the most restrictive one with respect to the chemical form.

Table -2-

Isotope	Fuel-A 33,000 MWd/t	Fuel-B 45,000 MWd/t	Fuel-C 45,000 MWd/t
U-235	3.25	4.5	0.711
U-238	96.75	95.5	99.284
U-total	100.	100.	94.
Pu-238			1.7
Pu-239			58.1
Pu-240			22.3
Pu-241			11.3
Pu-242			5.4
Am-241			1.2
Pu/Am-total	0.	0.	6.

Isotopic and chemical composition of the three types of fuels at the time of loading in the reactor. The other uranium isotopes have been neglected. The presence of Am-241 in fuel-C is due to the decay of Pu-241 during the time which elapses between the plutonium extraction from the spent fuel-A and the reactor loading. This time span is of the order 2 years.

List of Figures

Fig.6: Ratio of normalized radiotoxicities of a MOX spent fuel at 45,000 MWd/t and of the same MOX fuel non irradiated. This gives the simple incineration effects (see text).

Fig.7: Ratio of average normalized radiotoxicities of a MOX spent fuel at 45,000 MWd/t + of the corresponding UO_2 HLW and of the UO_2 spent fuels light-water reactors at 33,000 MWd/t. This gives the global incineration effects (see text).

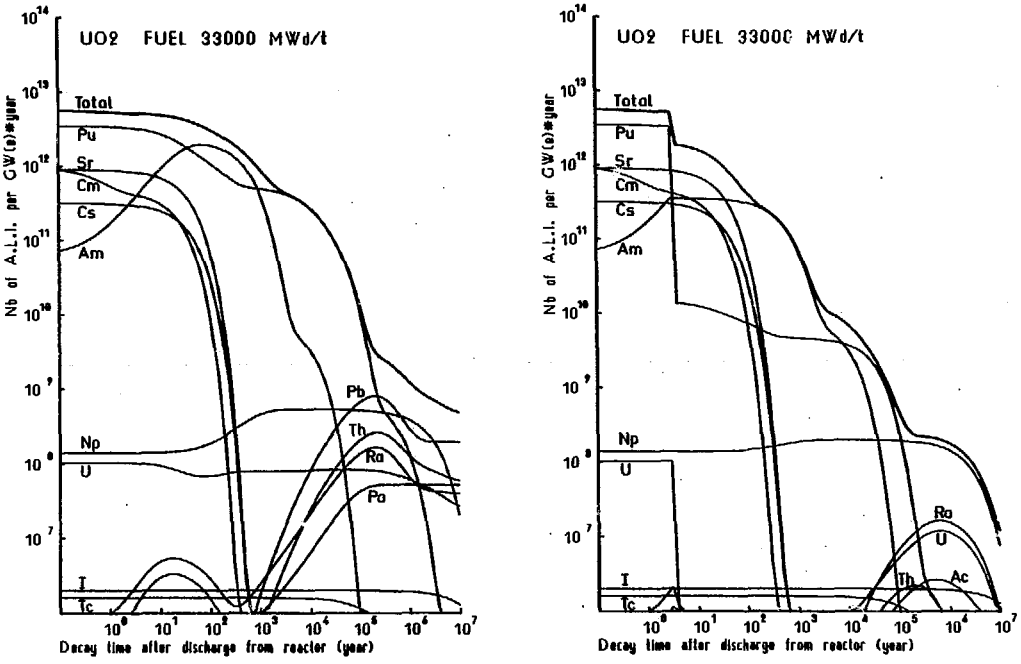

Fig.1: Normalized radiotoxicity of spent fuels (1,a) discharged from a 33,000 MWd/t light-water reactor,and of the corresponding HLW (1,b), with that of some chemical elements. The reprocessing parameters are the following: cooling time 3 years, removing efficiency U: 99,9 %; Np: 2,3 %; Pu: 99,6 %.

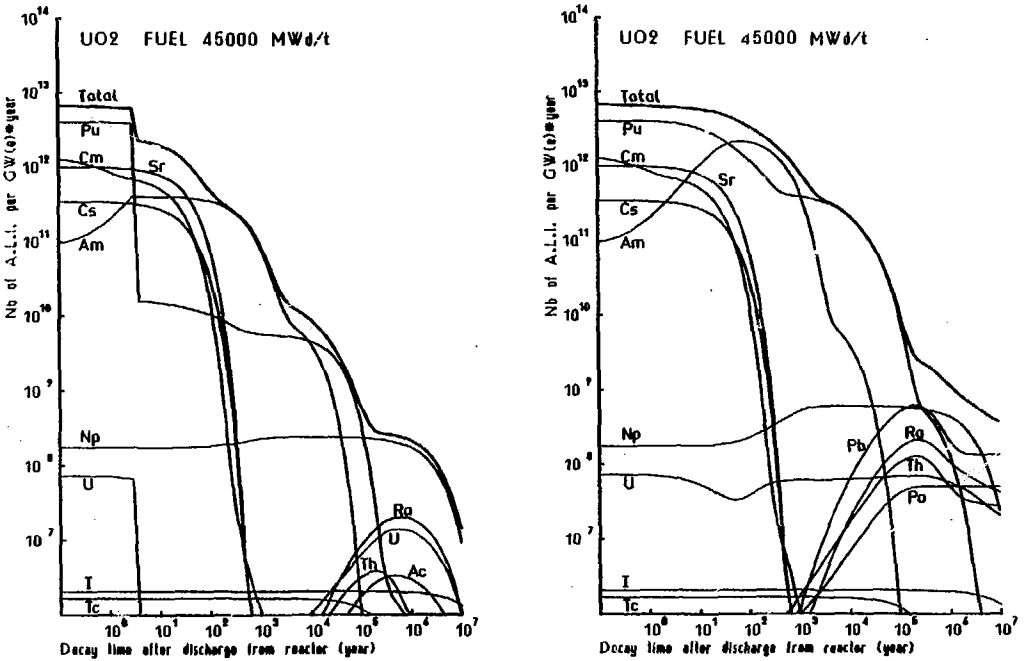

Fig.2: Normalized radiotoxicity of spent fuels (1,a) discharged from a 45,000 MWd/t light-water reactor, and of the corresponding HLW (1,b), with that of some chemical elements. Same reprocessing parameters as in fig.1.

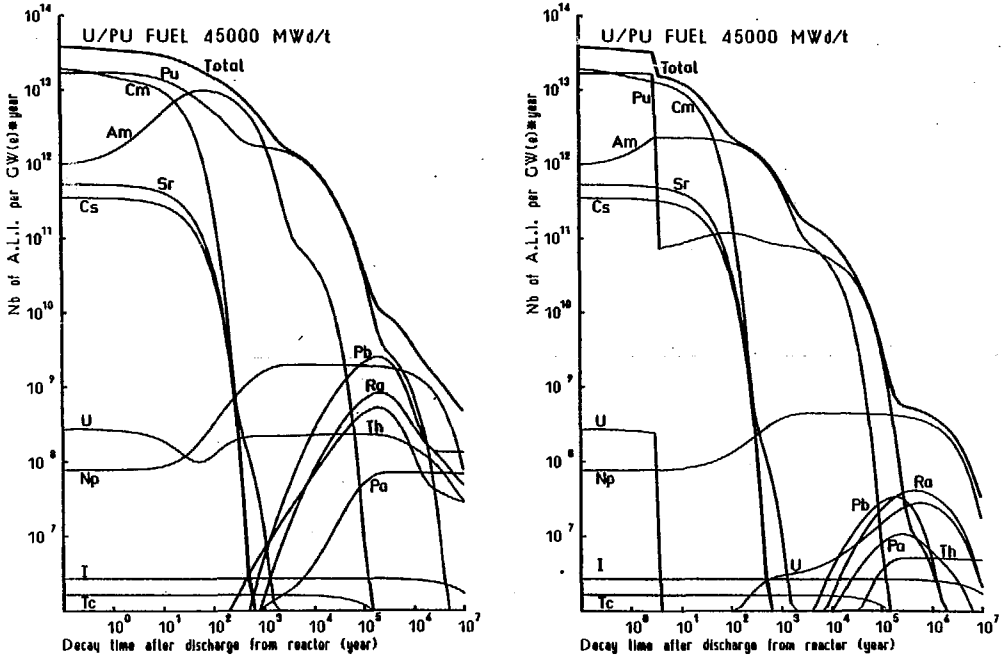

Fig.3: Normalized radiotoxicity of MOX spent fuels (1,a) discharged from a 45,000 MWd/t light-water reactor, and of the corresponding HLW (1,b), with that of some chemical elements. Same reprocessing parameters as in fig.1.

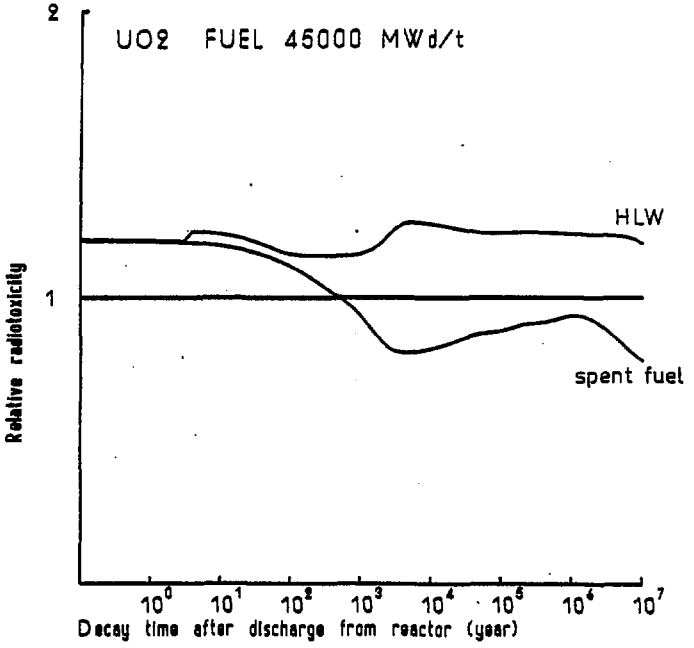

Fig.4: Ratio of normalized radiotoxicities of spent fuels at 45,000 MWd/t and at 33,000 MWd/t, discharged from a light-water reactor. The same ratio is also given for HLW.

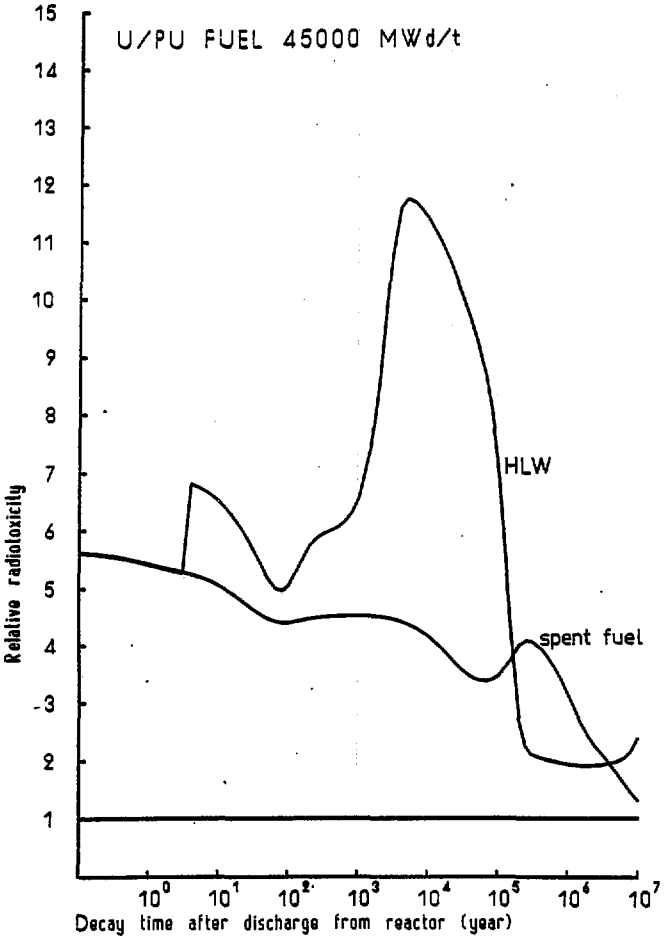

Fig.5: Ratio of normalized radiotoxicities of MOX spent fuels at 45,000 MWd/t and UO_2 spent fuels at 45,000 MWd/t, discharged from a light-water reactor. The same ratio is also given for HLW.

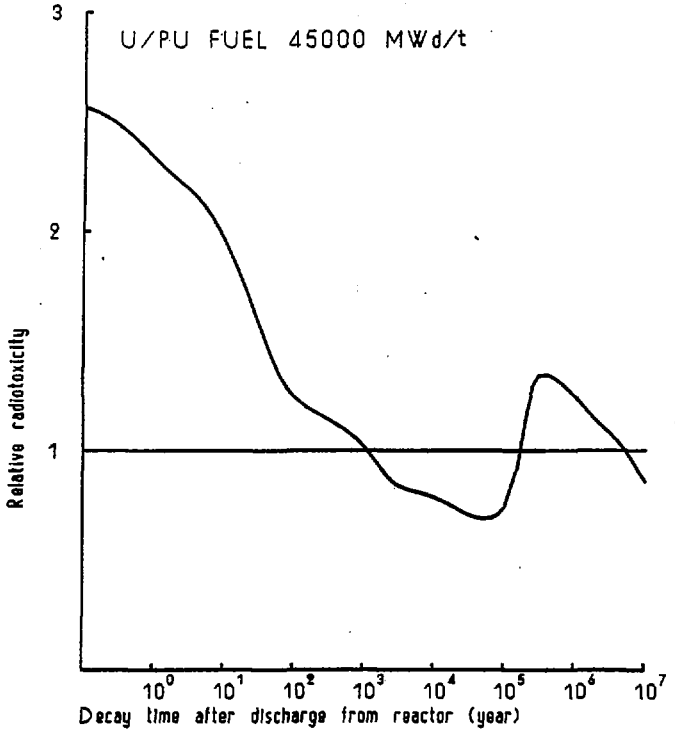

Fig.6: Ratio of normalized radiotoxicities of a MOX spent fuel at 45,000 MWd/t and of the same MOX fuel non irradiated. This gives the simple incineration effects (see text).

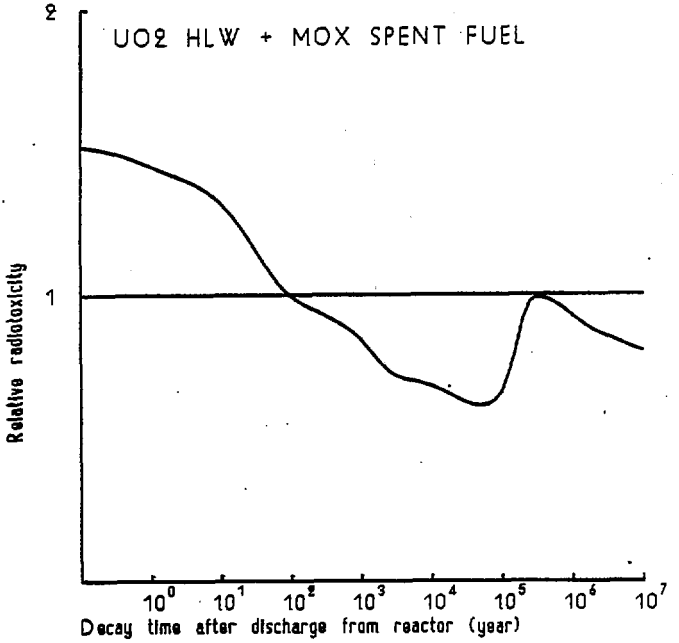

Fig.7: Ratio of average normalized radiotoxicities of a MOX spent fuel at 45,000 MWd/t + of the corresponding UO_2 HLW and of the UO_2 spent fuels light-water reactors at 33,000 MWd/t. This gives the global incineration effects (see text).