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IMPACT OF THE CHANGES FROM I.C.R.P. -26 TO I.C.R.P. -48 RECOMMENDATIONS ON THE POTENTIAL RADIOTOXICITY OF THE DISCHARGED LWR, FBR AND CANDU FUELS

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ABSTRACT

In order to take into account new metabolic data, the International Commission for Radiological Protection (I.C.R.P.) had to regularly update the recommended values used for radiotoxicity definition and calculation. In the present work we compare the fuel radiotoxicity from different reactors and we study the impact of the changes in the I.C.R.P. recommendations on these radiotoxicities.

INTRODUCTION

In the study of the possible release into the biosphere of radioactive substances from a discharged nuclear fuel, two types of risks are usually considered:

- the potential or maximum risk which is associated with the whole radiotoxicity content of the fuel; as far as long-term hazard is concerned, this is usually evaluated by the possible ingestion by a group of persons, mainly through drinking water, of the total content of the fuel;
- the real or residual risk which takes into account (in a final geological disposal) the reduction, thanks to artificial and geological barriers, of the hazard due to possible release of radioactive substances into the environment.

The potential risk is evaluated, particularly, through the concepts of maximum permissible concentration in water (M.P.C.) and of annual limit of intake (A.L.I.) developed in the publications of the International Commission on Radiological Protection (I.C.R.P)[1-3]. The present work*

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deals with the impact on the potential radiotoxicity of discharged nuclear fuels of the various changes which have been taking place, for the last decade, in the I.C.R.P. recommendations concerning the dose effect of the ingestion of certain radionuclides,

. Since the publication of the I.C.R.P-26 guideline[4] in 1977, two modifications (the first one[3] took place between 1979 and 1981 and the second one[5] in 1986) have been made concerning the radiotoxicities of various radionuclides (see Tab. I). These modifications updated the parameters used to define the radiotoxicity, taking into account new metabolic data, as well as new dose calculation procedures. The A.L.I. values obtained with these two modifications have been published respectively in the I.C.R.P.-30 and I.C.R.P.-48 recommandations. It has been proposed in the last one 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 implies that, with respect to the I.C.R.P.-30 values, the radiotoxicity has been divided by 10 for Np, multiplied by 10 for Pu and by 2 for Am, Cm and Cf. This changes significantly the relative weight in the high-level wastes (HLW) and in the spent fuels of the radiotoxicity of Pu and results in a substantial change of the total radiotoxicity of the fuel.

The impact of the I.C.R.P. modifications on the radiotoxicity of spent fuels discharged from the following reactors (see Tab. II) is studied in the present work:

1. LWR (Light Water Reactor) at an average burn-up of 33,000 MWd/t[**,6] and 45,000 MWd/t[7,8], fueled with enriched uranium;
2. LWR at an average burn-up of 45,000 MWd/t[9,10], fueled with natural uranium and plutonium (mixed oxide fuel, MOX);
3. CANDU (Canadian Uranium fuel moderated with heavy water) at an average burn-up of 7,500 MWd/t[11], fueled with natural uranium.
4. FBR (Fast Breeder Reactor) at an average burn-up of 48,600 MWd/t[12], fueled with natural uranium and plutonium (mixed oxide fuel, MOX).

For the sake of comparison, all the radiotoxicities, calculated in this work, are normalized to the quantity of electricity produced in GW(e)*year. The energy conversion efficiency has been taken equal to 1/3 for all reactors..

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

RADIOTOXICITY OF CANDU AND FBR FUELS IN THE FRAMEWORK OF THE I.C.R.P.-48 RECOMMENDATIONS

In a recent publication[13], we studied the long-term radiotoxicity of high level wastes and spent fuels produced in above mentioned cases 1 and 2, which refer to the French nuclear power stations operated or under investigation by the electrical utility E.D.F.. In order to complete and update this study, we show in the frame of the I.C.R.P.-48 recommendations similar curves for FBR and CANDU (based respectively on data for Superphenix[6] and Pickering[11] reactors) on figs.1a;b and 2a;b. The parameters, which define the high-level waste characteristics, are those foreseen in the future UP-3 reprocessing plant at La Hague (France)[14]. These curves represent, up to 10^7 years, the time variation of the radiotoxicities for spent fuels and high level wastes. We also show on fig.3 the variation up to 10^7 years of the radiotoxicities, normalized to the GW(e)*year, of the five fuels mentioned above based on the I.C.R.P.-48 recommendations . The figure gives a direct comparison between the five fuels. However we must keep in mind that in the different cases the order of magnitude of the amount of radioactive substances present in the reactor after a fixed time of irradiation is not at all the same, even though the radiotoxicity discharged from the reactors, normalized to the GW(e)*year, are of the same order of magnitude.

IMPACT OF THE CHANGES FROM I.C.R.P.-26 TO I.C.R.P.-30 RECOMMENDATIONS

We have calculated the ratios of the total radiotoxicities of the spent fuels computed from the I.C.R.P-30 guideline to those computed from the I.C.R.P-2/6 guideline. Curves representing the variations of these ratios up to 10^7 years are shown on fig.4. The fuels defined above can be classified into group 1 for uranium fuels, and group 2 for MOX fuels. The following remarks concerning the impact of the I.C.R.P change can be made: as far as group 1 is concerned, the effect of the change of strontium A.L.I. values results in a reduction by a factor of about 8 of the total radiotoxicity of the fuel, since strontium is the major contributor to the total radiotoxicity of the fuels belonging to this group. This reduction effect lasts some decades while in group 2 this effect is completely counteracted by the importance of plutonium and results in an increase of the radiotoxicity of the spent fuel. As strontium decreases, the relative radiotoxicity plotted in fig.4 increases significantly in all cases, because of the decrease of the A.L.I. values for plutonium and americium, and reaches a maximum at about 10^3 years. At about 10^5 years, the effect of these radionuclides decreases and again at about 10^6 years the decrease of neptunium A.L.I. values is responsible for the increase of the fuel radiotoxicity.

IMPACT OF THE CHANGES FROM I.C.R.P.-30 TO I.C.R.P.-48 VALUES

We first study as an example the impact of the changes produced by I.C.R.P.-48 recommendations on a 33,000 MWd/t L.W.R. fuel. For this purpose, we show on figure 5 the variation up to 10^7 years of the radiotoxicities of neptunium, plutonium and total fuel before and after these changes; we do not represent Am and Cm in order to keep the figures clear. We represent on fig. 6 the variation with time of the ratios of the total radiotoxicities of the five types of fuel calculated from the I.C.R.P.-48 guideline to those calculated from I.C.R.P.-30 guideline.

The following remarks can be made. Up to about a hundred year, Pu, Am, Cm and the fission products contribute significantly to the radiotoxicity of the spent fuel. The .increase of the radiotoxicity of Pu and Am results in a 3-fold increase of that of the spent fuels except for F.B.R. fuel where a 4 to 5-fold increase is observed because the relative amount of plutonium is more important in this case. From a few hundred years to about 10^4 years, the relative weight of the radiotoxicity of plutonium in the fuel is a function of the decrease of the activities of curium and fission products, plutonium- 238 and 241. This results first in a slight decrease followed by a significant increase (by a factor 7 to 10) of the relative total radiotoxicity of the fuel. This 7 to 10 fold increase is due to the fact that the radiotoxicity of plutonium is during this period the major contributor to the total radiotoxicity of the fuel. Beyond 10^4 years, the relative radiotoxicity (computed from I.C.R.P.-48 to the one computed from I.C.R.P.-30) of the fuel decreases because of the decay of plutonium- 239 and 240. Beyond 10^5 years the contribution of neptunium-237 becomes important and the relative radiotoxicity of the fuel becomes smaller than one.

To summarize, the I.C.R.P.-48 recommendations result first in a general increase of the relative radiotoxicity of the total fuel up to about 10^5 years; this is mainly due to plutonium isotopes the relative weight of which changes however significantly over this period; second in a decrease of the relative radiotoxicity between 10^5 and 10^7 years because of the neptunium contribution.

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Table -I-

Half-life, Maximum Permissible Concentration[4] and Annual Limit of Intake[3,5], 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. Typing errors in the third column, which appeared in the same table of ref.13, have been corrected.

Isotope	Half-life (year)	M.P.C. (Bq/m^3) (refs.1,2)	A.L.I. (Bq) (ref.3)	A.L.I. (Bq) (ref.5)
Sr-90	28.15	1.5 10^{4}	10^{5}	/
Tc-99	2.14 10^{5}	1.1 10^{6}	10^{7}	/
I-129	1.57 10^{7}	7.4 10^{2}	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^{6}	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}

Table -II-

Isotopic and chemical composition of the five types of fuels at the time of loading in the reactor. The other uranium isotopes have been neglected. The presence of Am-241 in MOX fuels is due to the decay of Pu-241 during the time which elapses between the plutonium extraction from the 33,000 MWd/t spent fuel and the reactor loading. This time span is of the order of 2 years.

Isotope	LWR 33,000 MWd/t	LWR 45,000 MWd/t	LWR 45,000 MWd/t	CANDU 7,500 MWd/t	FBR 48,600 MWd/t
U-235	3.25	4.5	0.711	0.711	0.4
U-238	96.75	95.5	99.284	99.824	99.6
U-total	100.	100.	94.	100.	84.7
Pu-238			1.7		1.7
Pu-239			58.1		56.7
Pu-240			22.3		23.5
Pu-241			11.3		10.8
Pu-242			5.4		5.6
Am-241			1.2		1.6
Pu/Am-total	0.	0.	6.	0.	15.3

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Fig.2: Radiotoxicity per GW(e)*y of spent fuels (1.a) discharged from a 7,500 MWd/t Candu reactor, and of the corresponding HLW (1.b), with that of some chemical elements. Same reprocessing parameters as in fig.1. The I.C.R.P.-48 A.L.I. values[8] have been used in these calculations.

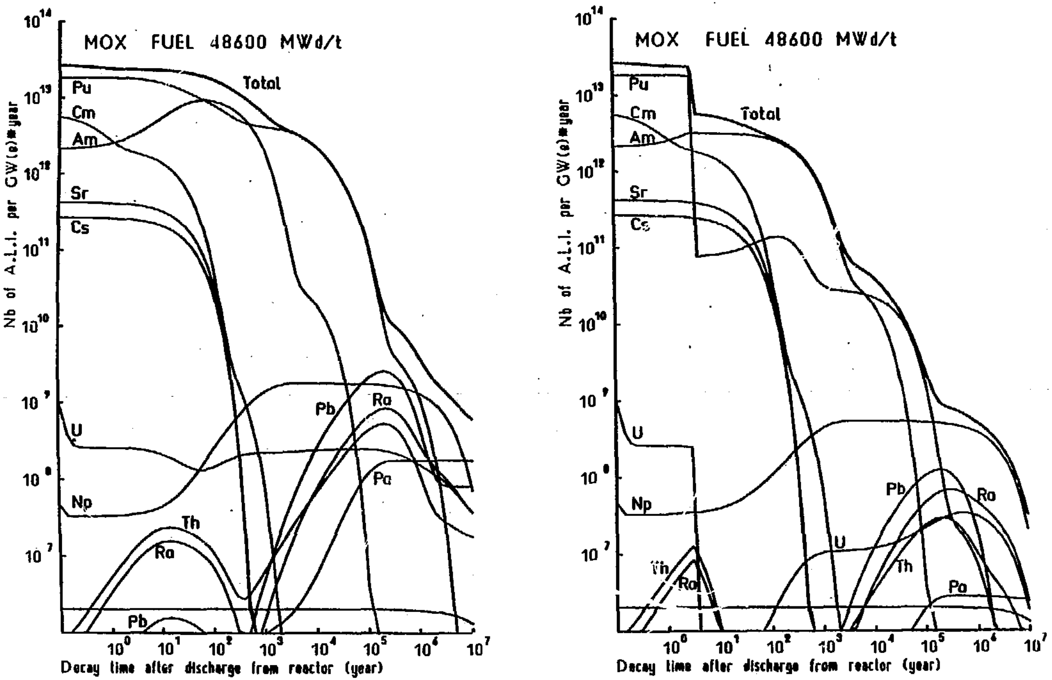
Fig.3: Total radiotoxicity per GW(e)*y computed for five different types of discharged fuels. The MOX fuels at 45,000 MWd/t and 48,600 MWd/t correspond to LWR and FBR respectively. The I.C.R.P.-48 A.L.I. values[8] have been used in these calculations.

Fig.4: Total radiotoxicity computed from the I.C.R.P.-30 guideline[3] normalized to the corresponding radiotoxicity computed from the I.C.R.P.-26 guideline for five different types of discharged fuels. The MOX fuels at 45,000 MWd/t and 48,600 MWd/t correspond to LWR and FBR respectively. The correspondence between M.P.C and L.A.I. is

calculated by assuming that the reference man typically drinks one m^3 of water per year.

Fig.5: Total, plutonium and neptunium radiotoxicity per GW(e)*y, computed from the I.C.R.P.-48 and I.C.R.P.-30 guidelines[5,3].

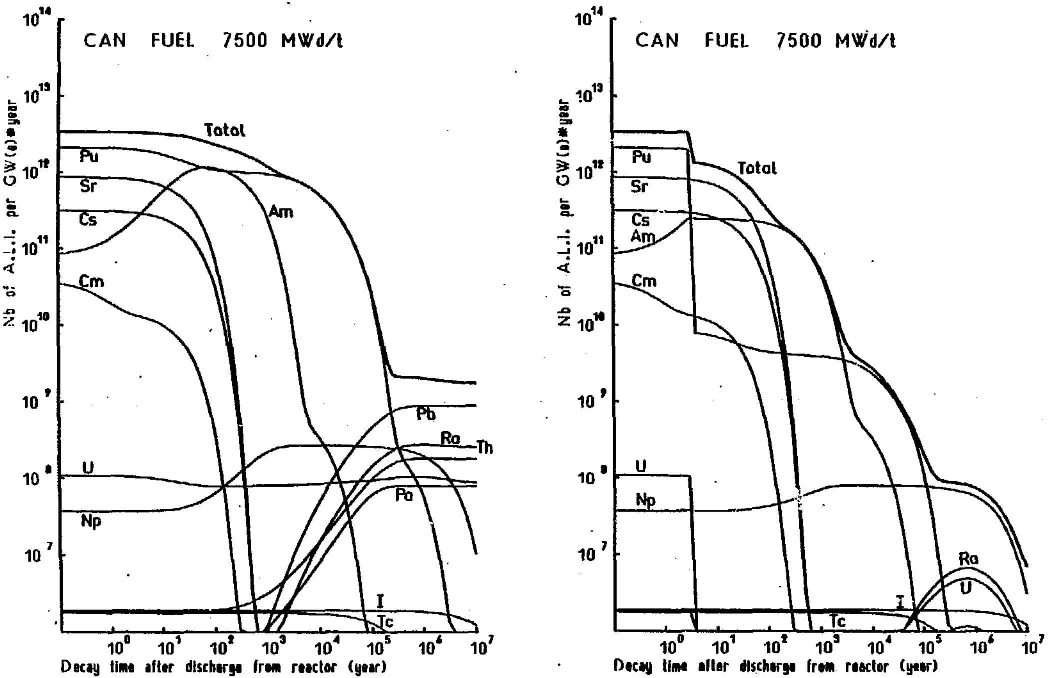
Fig.6: Total radiotoxicity computed from the I.C.R.P.-48 A.L.I. values[5] normalized to the corresponding radiotoxicity computed from the I.C.R.P.-30 values[3] for five different types of discharged fuels. The MOX fuels at 45,000 MWd/t and 48,600 MWd/t correspond to LWR and FBR respectively.



1.a

1.b

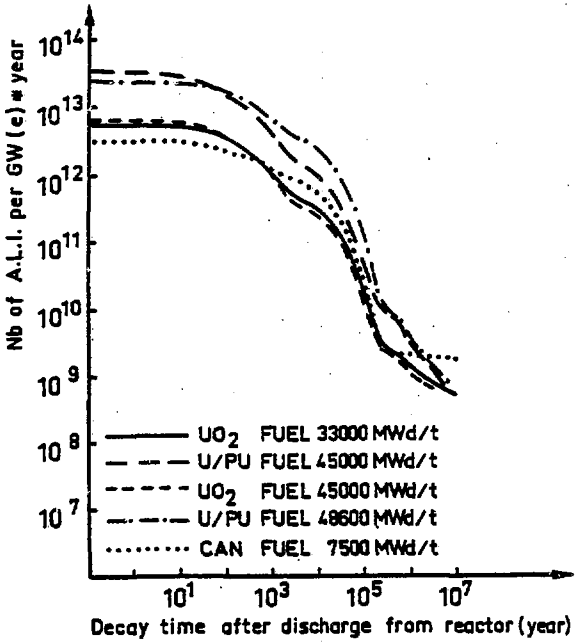
Fig.1: Radiotoxicity per GW(e)*y of spent fuels (1,a) discharged from a 48,600 MWd/t fast breeder reactor, and of the corresponding HLW (1,b), with that of some chemical elements. The reprocessing parameters are the following: cooling time 3 years, removing efficiency U: 99,9 %; Np: 2,3 %; Pu: 99,6 %. In 1,c, the contribution of the main isotopes is shown in the case of spent fuels. The I.C.R.P.-48 A.L.I. values[5] have been used in these calculations.



2.a

2.b

Fig.2: Radiotoxicity per GW(e)*y of spent fuels (1,a) discharged from a 7,500 MWd/t Candu reactor, and of the corresponding HLW (1,b), with that of some chemical elements. Same reprocessing parameters as in fig.1. The I.C.R.P.-48 A.L.I. values* have been used in these calculations.



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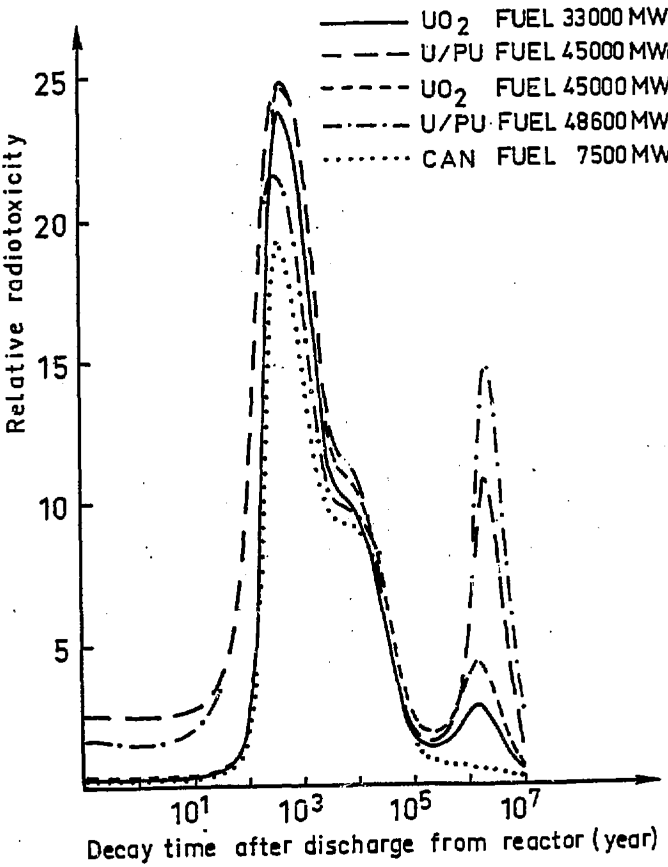

Fig.4: Total radiotoxicity computed from the I.C.R.P.-30 guideline[2] normalized to the corresponding radiotoxicity computed from the I.C.R.P.-26 guideline for five different types of discharged fuels. The MOX fuels at 45,000 MWd/t and 48,600 MWd/t correspond to LWR and FBR respectively. The correspondence between M.P.C and L.A.I. is calculated by assuming that the reference man typically drinks one m^3 of water per year.

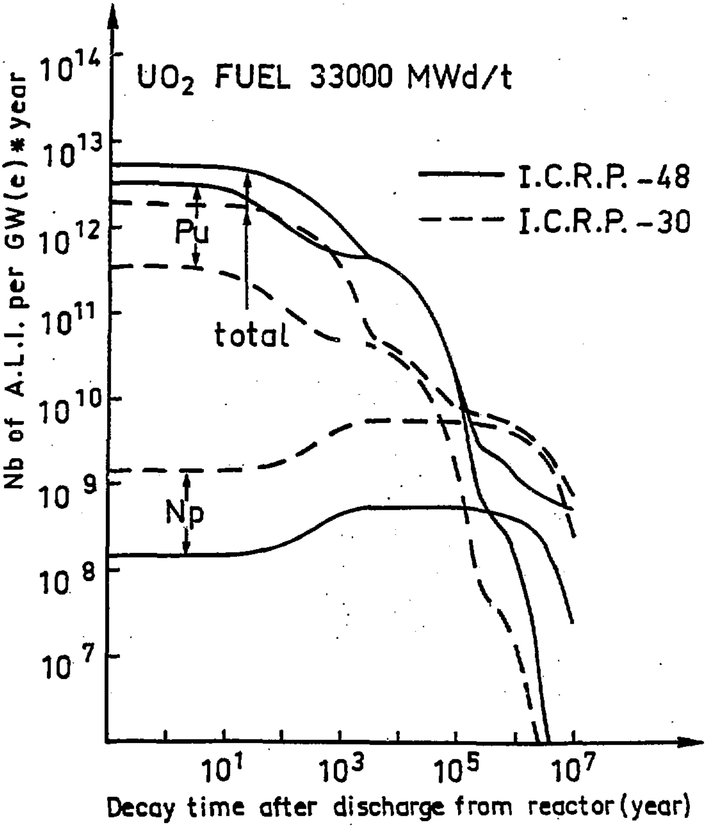

Fig.5: Total, plutonium and neptunium radiotoxicity per GW(e)*y, computed from the I.C.R.P.-48 and I.C.R.P.-30 guidelines[6,3].

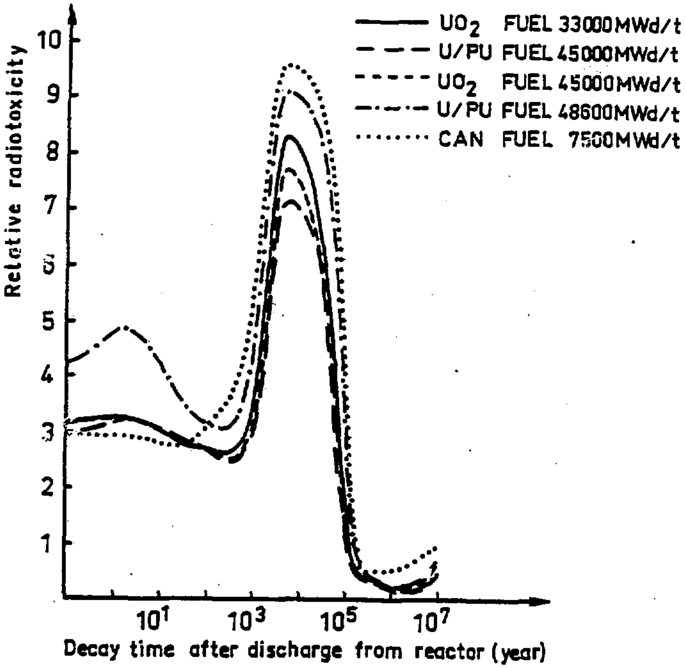

Fig.6: Total radiotoxicity computed from the I.C.R.P.-48 A.L.I. values[4] normalized to the corresponding radiotoxicity computed from the I.C.R.P.-30 values[3] for five different types of discharged fuels. The MOX fuels at 45,000 MWd/t and 48,600 MWd/t correspond to LWR and FBR respectively.