REDUCTION OF MINOR ACTINIDES IN NUCLEAR WASTE VIA MULTIPLE RECYCLING IN FAST REACTORS

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ABSTRACT

Starting from an initial fuel irradiation in a PWR, the long-term potential radio-toxicity of the spent fuel, which becomes waste in the once-through option, has been compared to the toxicity of residual waste from multiple recycling in a fast reactor. Either the plutonium alone is recycled, or the plutonium and the minor actinides (Np, Am, Cm), all supposed to be recovered by reprocessing with a 99.5 % yield.

Seven successive re-irradiations in a fast reactor of the EFR type have been explicitely represented, with realistic out-of-pile times. This covers a period of time of about one century. Minor Actinides are assumed to be homogeneously recycled, i.e. mixed with the (U,Pu) oxide fuel.

The advantage of the Pu + M.A. recycling strategy is to reduce the radio-toxicity of the actinides by the following factors with respect to the PWR once-through case :

- at 1000 years, by a factor 40,
- at 10,000 years, by a factor 25,
- at 100,000 years, by a factor 30.

In addition, relating the actinide toxicity to the energy production scales down by a factor (e.g. 2.5) the curves for the actinides in the recycle cases.

The actinide isotope whose contribution is dominant from 200 to 1000 years is Am241.

Halving the out-of-pile times would reduce the quantities of Am241 effectively recycled, what is favourable to refabrication ; the toxicity of the waste itself is hardly influenced.

With respect to waste toxicity, recycling Pu + A.M. in fast reactors will be a success, provided that the recovery yield at separation can be brought high enough, as well for Pu isotopes as for M.A. isotopes and first of all for Americium : respective targets of 99.9% (Pu) and 99.5% (Am) should be aimed at in research work.

Under such conditions indeed, the actinide toxicity will be reduced to the level of toxicity which corresponds to the natural uranium ore initially used, after about 1,000 years ; in contrast to the PWR, once-through strategy (for which 300,000 years are needed) and also to recycling Pu alone (15,000 years are still required), this period of time makes sensible a surveillance programme for waste repositories.

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1. INTRODUCTION

The problem of long-ranging toxicity in nuclear waste due to alpha-emitting actinides is now clearly identified. While bêta- and gamma-emitting fission products dominate the activity of nuclear waste over some hundred years, beyond that time the residual activity is nearly entirely due to the actinides.

The idea has thus emerged to recycle the quantities of generated actinides in nuclear reactors : one would recycle not only the plutonium, as in the present MOX recycling programmes, but also neptunium, americium and curium, the so-called minor actinides. Many recent studies, like [1] and [2] for example, have considered as well thermal as fast neutron reactors to that aim.

In the studies presented below, fast reactors are considered for recycling. Realistic schemes are assumed for the in-pile and out-of-pile times, and successive recycling operations are explicitly represented over a total period of about 100 years.

Minor actinides are assumed to be mixed homogeneously with the usual mixed oxide (U,PuO2), in the same stoechiometric fraction as at the preceding discharge.

Reactors and schemes are defined in section 2.
The method of calculation is mentioned under section 3.
The impact on the toxicity of nuclear waste from recycling, either plutonium only, or plutonium and minor actinides, is compared and discussed under section 4.
Section 5 draws conclusions.

2. REACTORS AND SCHEMES

The initial fuel irradiation is assumed to take place in a 900-MWe, UO2-fuelled PWR, analogous or close to 34 reactors running in France and 5 in Belgium. The U235 fuel enrichment is 3.7 %, so that a burnup of about 43,000 MWd/tonne can be achieved over an operation time of 4 years.

While the total PWR fuel inventory amounts to 72 tonnes of heavy metal, the fate of one tonne of uranium initially loaded is followed in a multiple recycling scheme.

The reactor used for recycling is a 1450-MWe fast reactor, similar to the first consistent design of the European Fast Reactor (EFR), already considered for such studies in [2]. Its average plutonium enrichment is close to 21 %, and one will associate one twelfth of such a reactor to the LWR above, as far as fuel quantities are concerned. In other terms, the actinides (either Pu alone, or Pu, Np, Am, Cm together, according to the case), produced from 4 years of operation of 12 PWRS can be recycled in one core load of such a fast reactor (FR).

Seven successive recycles of the Pu and minor actinide (MA) flows are explicitly represented, covering nearly a period of 100 years. The detailed time schemes are given in Table I below.

As the fissile zones alone of the FR reduce progressively the Pu quantities, because they have a negative breeding gain, use is made of part of the Pu generated in blanket to restore criticality at reloading ; the hypothesis of a FR just self-sufficient in Pu is thus made.

Table I
Recycling time schemes

steps	*Duration (years)*
Initial PWR irradiation	4
Storage+cooling	3
Reprocessing+fabrication	2
FR irradiation n°1	5.5
Subsequent Storage+cooling	5
Reprocessing+fabrication	2
FR irradiations n°2 to n"7	5.5

In view to establish the benefits of FR recycling on waste toxicity, the comparison is made with respect to a reference case without recycling, corresponding to the once-through strategy : after the PWR irradiation, all spent fuel assemblies are stored and constitute waste.
A first recycling strategy consists of re-irradiating successively in FR the Pu alone, supposed to be recovered with 99.5 % efficiency at reprocessing ; minor actinides are rejected as waste.
In the second recycling strategy, both the Pu and the M.A. are recovered, with 99.5 % efficiency at reprocess ing, and re-irradiated successively in FR ; only the 0.5 % residues go to waste.

Sensitivity studies have been added on the influence of the separation yields of Pu and of the M.A., and of the out-of-pile times.

3. METHODS OF CALCULATION

The computer programme used for this multiple recycle scheme is ORIGEN-2 [3]. It treats neutron flux irradiation periods as well as natural decay. It allows to calculate the evolution of activity for a very large variety of actinides (130 isotopes) and of fission products (850 isotopes). Among the actinides, one follows in particular all isotopes of interest for uranium, neptunium, plutonium, americium (10 isotopes) and curium (11 isotopes).
The version of the programme used was released in 1990.

The cross-section library retained for thermal reactor irradiation is that referred to as '50,000 MWd/t' ; the fast reactor library corresponds to the 'advanced' one. Parallel calculations with more detailed methods have served to check the validity of the burnup results. The LWR calculations were backed by a comparison with results produced in an OECD expert group [4], while the FR burnup calculations had been checked with respect to EFR design methods in [2].

The activities have been converted in relative toxicities, by dividing them by the values of Annual Limits of Intake (ALI), for an ingestion, recommended for the workers by the International Committee of Radiological Protection (ICRP) in 1990 [5]. These limits, defined in Becquerels, take the biological effect of the radiations into account.

It is worth mentioning that the 1990 ICRP recommendation has modified the ALI values for Pu, NP, Am, Cm with respect to the former 1986 recommendation : the ALI are now lower (thus more limitative) for Pu (by a factor 10), and for Am241, Am243 and Cm (by a factor 2), while they have been relaxed by a factor 10 for Np237 .

A potential toxicity value can be calculated in the same way for the quantity of natural uranium ore (7.6 t) which is used to prepare one tome of enriched uranium feeding the PWR.

Dividing the toxicity of the waste by the toxicity of the initial uranium ore allows to know after how long a time waste will become as harmless as the uranium ore is ; this is analogous to the method of the 'risk factor', as used in [1].

Maximum potential hazards only are considered here : no attempt is made to evaluate the possible release rates of fission products and actinides out of waste repositories.

4. RESULTS

4.1 Variation of M.A. quantities over 100 years of recycling

For the case of the recycle of Pu and M.A., the evolution of M.A. quantities over the 100-year period explicitly considered is given in Table II. Masses are showed in g per tonne of u initially loaded for four important M.A. isotopes : Np237, Am241, Am243 and Cm245. One observes the following :

Np237 is reduced by about a factor 2 after each of the initial FR irradiations ; afterwards the reduction factor becomes progressively smaller, leading to a minimum fraction of about l/15th of the initial amount ;

Am241 is growing in the two first recycles, because of the natural decay of Pu241 during the out-of-pile times ; then comes a slow, progressive decrease, which derives from the reduction of its predecessor Pu241 ;

Am243 too is first growing, and then slowly decreasing ; this comes from the reduction by irradiation of its predecessor Pu242

- Cm245 requires some 7 cycles to become under control ; this isotope gives a small, long-term contribution to the build-up of Pu241 and Am241.

Table III gives for the 3 cases : no recycle, Pu recycle, Pu and M.A. recycle, the quantities of the same 4 isotopes which accumulate in waste after 100 years of recycling.

It appears from this comparison that; as far as waste is concerned, there is no advantage to recycle PU only (except of the energy production). There is a large incentive to recycle M.A. in addition to the Pu, so that masses after 100 years can be reduced, with respect to the PWR case, by the following factors :

90 for Np237
60 for Am241
20 for Am243
4 for Cm245.

It should be stressed that the recycling programme cannot be stopped after this period, otherwise the reduction factors above would fall to about 14 for Np237 and 3 for Am241 ; there would be no reduction at all for the 2 last isotopes.

Such a recycling programme needs to be pursued.
On the other hand, the M.A. can obviously be concentrated prior to recycling.

4.2 *Long-term evolution of waste toxicity*

In terms of toxicity, it is first useful to note what are the actinide isotopes contributing mostly to the total toxicity. This is done in Fig.1 for the waste from the initial PWR irradiation. The major contributors are, respectively :

Am241 between 100 and 1000 years ;
PU239 and PU240 between 1000 and 10,000 years ;
Pu239 around 100,000 years ;
Np237 and its successor Th229 beyond 100,000 years.

One notes that the importance of Np237 is smaller than in earlier work, like [1]. This is essentially a result of the re-evaluation of the ALI values for Np in the last ICRP recommendation. Now, successive re-evaluations in either sense let suggest that the results should be given a relatively large uncertainty.

Fig. 2 compares the time evolution of waste toxicity for the different cases considered : PWR alone, PWR plus 7 cycles in fast reactor, recycling Pu only, or M.A. in addition to Pu.

The total toxicity contained in waste is their maximum potential hazard, remaining trapped in packaged waste, should anyone suddenly ingest them. The effect of possible migration is not accounted for.

Fission products dominate first the waste toxicity. After a few hundred years their contribution is rapidly falling to a very low level, corresponding to the presence of Tc99 and I129. From that time on, the actinides dominate waste toxicity. Recycling M.A. in addition to Pu decreases their toxicity so that, with respect to the PWR case, the reduction is as follows :

- at 1000 years, by a factor 40,
  at 10,000 years, by a factor 25,
  at 100,000 years, by a factor 30.

These reduction factors are significant, although they do not reach the factor 200, theoretically associated with residues of 0.5 %.

4.3 *First results in terms of acceptable toxicity levels*

On Fig. 2, all toxicities are related to that of the quantity of uranium ore needed for the fabrication of the initial tonne of enriched uranium feeding the whole scheme. A first, simple criterion would thus be to declare acceptable (i.e. requiring no surveillance anymore) nuclear waste when their toxicity by ingestion has come down to this level.

According to this, fission products are acceptable after about 300 years. In contrast, actinides would need much longer times :

300,000 years (PWR, once-through), nearly 100,000 years (Pu recycle), or 20,000 years (Pu and M.A. recycle).

4.4 *Toxicity versus energy production*

In the FR recycling schemes considered here over a century, the total production of electrical energy is about 2.5 times higher than in the PWR irradiation alone. It appears thus justified to scale down by such a factor 2.5 the curves giving the toxicity of the actinides with recycles on Fig. 2.

4.5 *Sensitivity studies*

Two types of variants have been considered. In the first one the out-of-pile times were changed. In the second one, the separation yields of Pu and M.A. at reprocessing were varied.

If the out-of-pile times can be halved, this favorably affects the quantities of Am241 during the cycling operations themselves. For example, they are reduced by one third for the first two refabrication campaigns. On the other hand, the activity of Am241 in waste after 100, 200 or 1000 years is hardly influenced.

Assuming 80" % yield for the M.A. (what is already a hard challenge for Am) results in an increase of the actinide toxicity, given above for the 99.5 % assumption, by a factor 10 in the time range from 100 to 2,000 years. This stresses how important it is to develop high purification separation methods for the M.A., and first of all for Americium.

Improving the separation yield for the Pu isotopes from the 99.5% assumed above to 99.9% Is beneficial, as it further reduces the actinide toxicity by a factor 3 around 1000 years and 4 between 10,000 and 1000,000 years.

4.6 *Acceptable toxicity levels: recapitulation*

When one combines the findings of par. 4.4 and 4.5, assuming in particular recovery yields of 99.9% for Pu and 99.5% for M.A., the specific actinide toxicity is reduced by recycle down to the level of the natural uranium ore toxicity after, respectively, 15,000 years (Pu recycle) or about 1,000 years (Pu + M.A. recycle).

The latter time range can still be covered by human memory, so that a surveillance programme makes sense ; in contrast the former time ranges, much longer, escape from this scope.

(Simple criteria based on maximum potential toxicities are not meant here to be preferable to more elaborate criteria covering possible geological transfers by migration up to the groundwater. The point is rather to deduce orientations and priorities for further activities, based on parametric studies.)

5. CONCLUSIONS

Starting from an initial fuel irradiation in a PWR, the long-term potential radio-toxicity of the spent fuel, which becomes waste in the once-through option, has been compared to the toxicity of residual waste from multiple recycling in a fast reactor. Either the **plutonium alone is recycled, or the plutonium and the minor actinides (Np, Am, Cm), all supposed to be recovered by reprocessing with a 99.5 % yield.**

Seven successive re-irradiations in a fast reactor of the EFR type have been explicitly represented, with realistic out-of-pile times. This covers a period of time of about one century. Minor Actinides are assumed to be homogeneously recycled, i.e. mixed with the (U,Pu) oxide fuel.

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Halving the out-of-pile times would reduce the quantities of Am241 effectively recycled, what is favorable to refabrication ; the toxicity of the waste itself is hardly influenced.

With respect to waste toxicity, recycling Pu + A.M. in fast reactors will be a success, provided that the recovery yield at separation can be brought high enough, as well for Pu isotopes as for M.A. isotopes and first of all for Americium : respective targets of 99.9% (Pu) and 99.5% (Am) should be aimed at in research work.

Under such conditions indeed, the actinide toxicity will be reduced to the level of toxicity which corresponds to the natural uranium ore initially used, after about 1,000 years ; in contrast to the PWR, once-through strategy (for which 300,000 years are needed) and also to recycling Pu alone (15,000 years are still required), this period of time makes sensible a surveillance programme for waste repositories.

Note

The present results and orientations should be checked with regard to the long-term waste disposal assessments.

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Table 11

Evolution of M.A. Quantities over one Century of Recycling Operations (Pu + M.A.) (in g/t of uranium initially loaded in PWR)

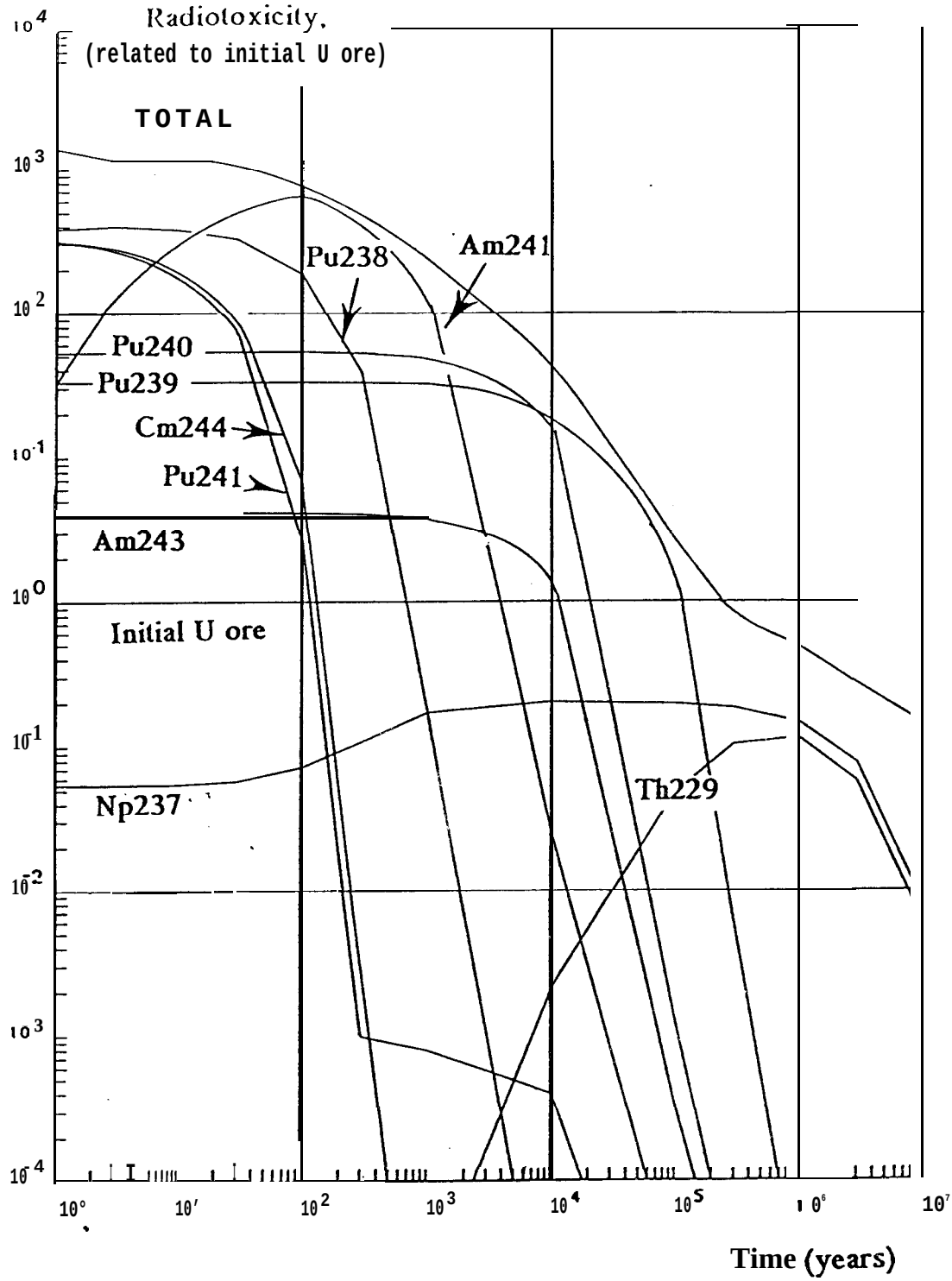
The first line always *refers* to the reactor loading (begin/end of irradiation), the second one to the waste (underlined figures)

	PWR	FR n°1	FR n°2	FR n°3	FR n°FR n°7		Sum of Waste
Np^{237}	627 3	627/297 1,5	300/155 0,8	158/93 0,5		51/46 0,24	9
Am^{241}	77 1,5	429/31 2	465/258 1,7	370/214 1,5		318/199 1,46	26
Am^{243}	169 0,8	168/229 1,1	228/243 1,2	241/230 1,15		169/156 0,8	8
(Cm^{244})	62 0,3	51/89 0,37	68/107 0,44	81/112 0,46		69/82 0,34	1
Cm^{245}	2,8 0,01	2,8/16 0,1	16/23 0,1	23/27 0,1		24/22 0,1	0,7

Table III

Quantities of Minor Actinides Accumulating in the Waste after one Century.
(in g/t of Uranium initially loaded in PWR)

	PWR once-through	PWR +7 Recycles in FR (Pu)	PWR +7 Recycles in FR (Pu + M.A.)
Np^{237} PWR	629	629	3
FR 1 to 7	-	169	4
Transmutations	170	125	2
Total	800	922	9
Am^{241} PWR	299	299	1,5
FR 1 to 7	-	1340	11
Transmutations	1200	-112	14
Total	1500	1527	26
Am^{243} PWR	169	169	0,85
FR 1 to 7	-	572	7.15
Transmutations	-1	-3	—
Total	168	738	8
Cm^{245} PWR	2,8	2,8	0,014
FR 1107	-	10	0,7



Toxicity vs time
for the Actinides contained in Wastes
(PWR, once-through)

FIG. 1

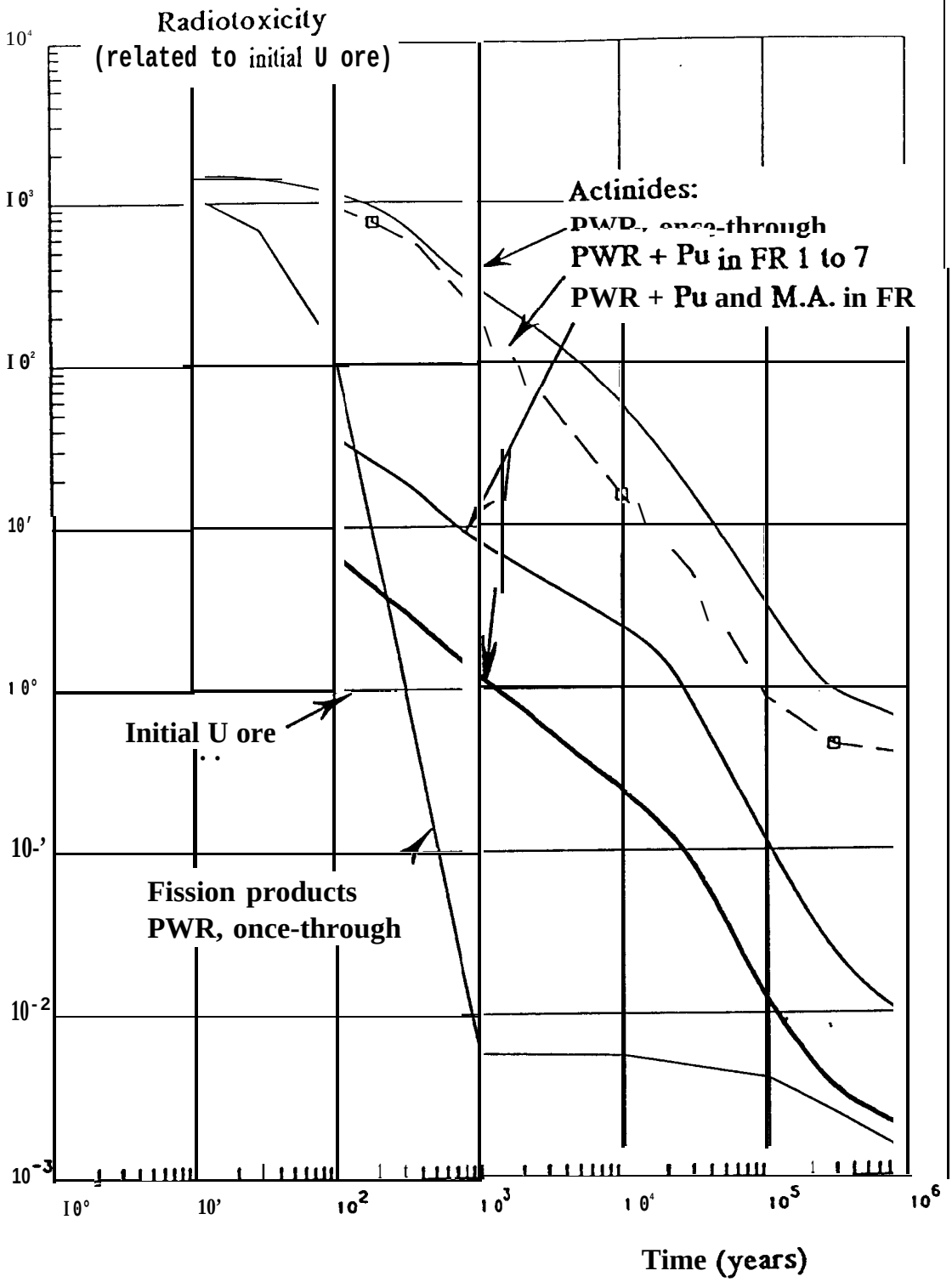


FIG. 2

Toxicity vs time
for the Actinides and Fission Products in Wastes