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**LONG-TERM α -HAZARD OF HIGH ACTIVITY WASTE
FROM NUCLEAR FUEL REPROCESSING**

by

F. GIRARDI, G. BERTOZZI

1974



**Joint Nuclear Research Centre
Ispra Establishment - Italy**

Chemistry Division

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Luxembourg, December 1974 — 30 pages — 4 Figures — B.Fr. 110.—

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Four barriers were identified, which could be evaluated in a probabilistic way by taking into account the great uncertainties present in each of them. The barriers are:

- 1) quality of the segregation afforded by deep geological formations,
- 2) stability of conditioned waste (chemical and physical),
- 3) retention by immediate surroundings,
- 4) distribution pattern of actinides in the environment.

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The analysis of a fictional accident showed that the uncertainties connected with the evaluation of the barriers' value are rather large. Additional studies, particularly on the stability of conditioned waste and ecological properties of the environment towards actinides, would considerably improve our knowledge of the value of the barrier system.

Chemical separation of actinides from high activity waste would be an additional option of undoubted value for the disposal of high activity waste. Its value for the overall safety of the entire waste inventory depends on many factors which need better evaluation, such as safety of the disposal of the separated actinides and the amount and quality of the additional waste generated by the separation process.

An analysis of various levels of possible separations suggests that a reasonable target might be: Pu, Am and Cm, decontamination factor 10^3 ; Np, coextraction with U and Pu with a 90% yield.

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1. INTRODUCTION

There is much debate among scientists and the general public concerning the long-term hazard of highly active wastes from nuclear fuels reprocessing, mainly due to the fact that they contain a few α -emitting nuclides, which maintain their radiological hazard for periods of thousands of years.

With the approval of the pluriannual program of the Joint Research Centre, an experimental activity on the feasibility of the chemical separation of long-lived α -emitters (or actinides) and their successive burn-out in a nuclear reactor has been initiated.

In order to set appropriate goals for the chemical separation, it was decided to carry out a conceptual study with the attempt of evaluating the benefits that the chemical separation of actinides would bring to the disposal concepts which are currently being studied in various countries.

During the study we realized that although an appreciation of the relative merits of various decontamination processes was a relatively simple task, it was practically impossible to reach any type of conclusion on the absolute values of decontamination required.

It also became evident that while the hazard diminution which could be achieved by various decontamination processes was in the range of $10^3 - 10^4$, the uncertainties existing in tentative evaluations of absolute hazards were in the range of 10^6 or higher. Some effort was therefore also devoted to the identification of factors responsible for such uncertainties and which should receive an appropriate amount of research effort.

2. BUILD-UP OF α -EMITTERS IN FUELS FROM LIGHT WATER REACTORS (LWR) AND FAST BREEDER REACTORS (FBR)

The α -emitter content and decay characteristics of spent fuel elements from LWR and FBR have been calculated from reference literature data. These data are relatively abundant for light water reactors⁽¹⁻⁸⁾. Data on fast breeder reactors are less abundant^(1,6-10) and, moreover, differ from source to source, since the actinide content of spent fuels largely depends on both the initial fuel composition, and the isotopic composition of the Pu used and on the burn-up obtained.

The following reference data have been assumed for our calculations:

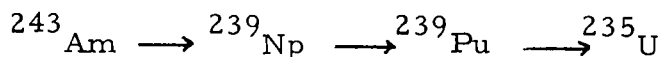
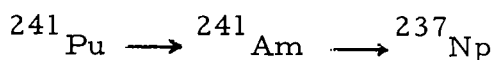
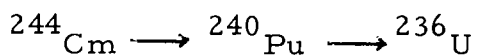
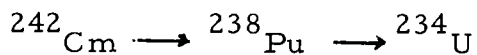
light water reactors:

3.3% enriched uranium, burn-up 33,000 MWd/t, mean specific power 30 MW/t, recycling of 1/3 of the fuel load every year.

fast breeder reactors⁽⁹⁾:

^{238}U - 10% ^{239}Pu fuel, burn-up 80,000 MWd/t, mean specific power 148 MW/t, recycling of 1/3 of the fuel load every 153 days.

Table I shows the data assumed for the actinides concentration in both types of fuel. The decay of the α -activity of individual nuclides has been calculated down to 10^6 years by applying the Bateman equation to the following decay chains:



The total α -activity has been plotted vs. decay time in figure 1. It should be noted that the same curve is also fairly representative of the decay of the biological hazard, since all the nuclides involved have similar RCG (Radiation Concentration Guides).

For both LWR and FBR fuels the α -activity of the fuel does not decay by more than a factor 10 over 1,000 years. FBR fuels are 15-20 times more α -active than LWR fuels. If the production of α -activity of LWR and FBR is compared for equal power production, the larger activity of FBR is partly compensated by the smaller amount of spent fuel which is yearly sent to reprocessing. The production of α -emitters by FBR remains, however, substantially higher (7-10 times) than the one of LWR. The α -activity from FBR fuels depends almost exclusively on the initial Pu load, while the contribution of the U and Np isotopes is less than 1%.

The activity of the various α -emitting nuclides after 1,000 years is reported in Table II, which shows that 99% of the activity is due to only three nuclides, ^{239}Pu , ^{240}Pu , and ^{241}Am , the last one being formed mainly by the decay of ^{241}Pu .

3. ACTINIDE CONTENT OF SOLIDIFIED HIGH ACTIVITY WASTES FROM SPENT FUEL REPROCESSING (Purex Process)

In order to calculate the α -activity of high activity waste from reprocessing of the mentioned spent fuels, we have assumed that:

- 99.5% of the U and Pu are extracted in the first extraction cycle,
- Am and Cm remain in the aqueous waste,
- Np also remains in the aqueous waste. This last assumption is clearly fictional, since a partial to almost complete extraction can be obtained by proper adjustment of the Np valence state⁽¹¹⁻¹⁴⁾, but we shall discuss below the implications of a more or less complete extraction of Np.

We have also assumed that after a suitable interim storage in a concentrated liquid form, the waste is converted into a vitrified product containing 16% of non-volatile fission products. The glass quantity produced by the reprocessing of 1 ton of LWR fuel would be approximately 150 kg. Since the fission product content depends upon design parameters (basically heat conduction in the glass block), these figures are only approximate.

We also assume that during these processes no segregation of actinides occurs, and they are homogeneously distributed in the glass blocks at the end of the process.

In Table III and IV the α -activity of LWR and FBR solidified wastes is tabulated for different decay times down to 10^6 years, while figure 2 shows the decay of the total α -activity of both types of waste.

It may be seen that the α -activity of LWR waste for the first thousand years is essentially due to curium and americium isotopes and their daughters. After 10^3 years the ^{239}Pu contribution is only 3%, while ^{237}Np (even in the pessimistic hypothesis of no extraction) is only 0.5%.

After 10^4 years the activity is equally divided between ^{243}Am and the ^{239}Pu - ^{240}Pu couple, while after a longer period ^{239}Pu and ^{237}Np prevail.

The total α -activity when the fission products have practically decayed (1,000 years, at a conservatively rounded off figure) will be about 0.5 mCi/g. A 150 kg glass block, obtained from the reprocessing of 1 ton of fuel, will therefore hold about 75 Ci of α -active isotopes.

In the case of FBR the higher burn-up obtained, and the correspondingly higher fission product concentration, makes it necessary to prepare, for each ton of reprocessed fuel, a three-fold higher quantity of vitrified waste. The amount of energy produced is also of course pro-

portionally higher.

If the actinides concentration is compared with that of LWR vitrified wastes, a few differences can be observed:

A much higher concentration of all Pu and Am isotopes (almost 10-fold) is only partially compensated by a lower ^{244}Cm concentration; as a result the total α -activity is higher, and remains substantially higher, particularly for the first few thousands of years. After 1,000 years the total α -activity would be about 3 mCi/g of glass, almost seven times higher than the corresponding activity from LWR glasses. The prevailing isotopes are the same, with a dominant contribution of Am isotopes (92.6% of the total).

It must be stressed, however, that the input data come from an arbitrarily taken reference design and that the expected actinide content of spent fuels from future commercial FBR is still uncertain.

4. LONG-TERM α -HAZARD OF DISPOSED HIGH ACTIVITY WASTE

As previously stated, reprocessing of LWR fuels will yield, for each ton of spent fuel, about 150 kg of vitrified waste, containing after 1,000 year storage, 0.5 mCi of α -emitters per gram of glass, or 75 Ci/block of glass.

A reprocessing plant with a capacity of 1,000 ton/year (serving about thirty 1,000 MWe power plants) will produce about one thousand such blocks per year.

The assessment of the long-term biological hazard of such waste is a rather difficult task.

Various units of measurement of the biological hazard have recently been proposed for long-term safety analysis, such as the hazard measure^(6,15) and the relative radiotoxic risk⁽¹⁶⁾, with the purpose of comparing relative biological hazards under similar circumstances.

These units were used to quantify the hazard of disposed waste by direct comparison with the hazard of uranium ores or mill tailings produced by uranium mining. Exhaustive ecological studies will undoubtedly be necessary to prove the validity of a direct comparison, since the equilibrium concentrations in the various ecological compartments and the transfer routes to man of α -emitters from either waste or uranium ores or mill tailings might be rather different.

Another unit of measurement, the potential hazard index, has been proposed by F. Gera and J. Jacobs⁽²⁾, and also includes a weighing factor to account for the probability that the radionuclide reaches man. This is certainly a better definition of the problem, but not yet a step forward towards its solution, since all the uncertainties remain in the evaluation of the weighing factor.

With the existing uncertainties, it would seem fair to state that the activity of each block and the number of blocks produced by a multi-annual operation of the reprocessing plant justify concern that unpredictable changes of whatsoever nature might bring the actinide content of the solidified waste back to the biosphere and to man in less than hundreds of thousands of years.

There is also no doubt that any statement about the multimillenary safety of a disposal scheme must have convincing evidence and must be based on real knowledge, and not simply on an educated guess.

We are attempting to evaluate the hazard by applying a safety analysis approach which has emerged in the analysis of nuclear reactor safety. The approach is based on the continuous evaluation of the availability and potential accidental behaviour of "barriers" provided between the dangerous product (the waste in our case) and the public⁽¹⁷⁾. The availability of each barrier can be quantitatively assessed by probabilistic methods which can take into account in a logical way the considerable uncertainties connected with each barrier.

The application deals with one of the most studied disposal philosophies, permanent segregation of vitrified high activity waste in a deep geological formation. The same approach could of course be applied to other disposal philosophies such as retrievable storage in a geological formation, engineered storage, etc.

Barrier identification is a first step in this approach. We have identified in this preliminary analysis four barriers which should be further analyzed.

a) Segregation afforded by deep geological formations

The value of this barrier can of course only be analyzed when referring to specific disposal sites. In a general way the uncertainties connected with such long-term extrapolation as those required by actinides decay must be stressed. As F. Gera and D. G. Jacobs have pointed out⁽²⁾ geology up to the present time has been a science with a limited predictive capability. So far no consistent comprehensive theory capable of explaining the major geological features of the earth has been available, although developments of the last few years (plate tectonic theory) may eventually provide earth scientists with a logical model capable of explaining the major features of the earth.

It seems reasonable therefore, to devote, at the present state of knowledge, a high proportion of the research potential to the study of possible additional barriers against the most probable accidents.

Water invasion and transport of the radioactive materials by diffusion or actual flow through the formation has been considered as the most probable accident in almost all analyses of permanent disposal formations. In order to minimize the consequences of this type of accident, there are other barriers which can be interposed between the waste and the biosphere, and which perhaps are under better control than the first one for the present technology.

b) Good chemical and physical stability of conditioned waste against flooding water

Glassy matrices have very low solubilities, and leaching rates for the most important fission products have been measured experimentally for many types of glasses. The effects of the presence of α -emitters, however, have been considered only marginally up to the present time. No data, for example, have been reported on the leaching rate of actinides, to the best of our knowledge.

The physical stability of the conditioned waste is also extremely important, since the leaching rate is proportional to the surface exposed to the water. A waste which maintains its physical integrity under severe conditions of radiation damage and physical stresses is undoubtedly much better than a waste which in the long term becomes pulverized.

Despite the importance of this parameter, very little is known about the long-term effects of phenomena such as chemical change following nuclear decay and radiation damage induced by α -particles and recoil nuclei.

The effects of these phenomena, which take place in time periods which are orders of magnitude higher than those obtained in practice in laboratory experiments, need to be studied with special care.

For reasons mentioned, while glassy matrices constitute a most effective barrier against the effects of water invasion in medium terms, additional knowledge is required in order to demonstrate the validity of this barrier for multimillenary storage.

c) Good retention properties of the immediate surrounding environment

A third barrier is given by the retention capacity of the immediate surroundings of the disposal site, or, in general, by the geochemical nature of the soil which is interposed between the geological formation

selected for disposal and possible receiving aquifers.

Although the knowledge of parameters which affect the retention and release of actinides by different soil types is still limited, it may be estimated that for near neutral pH the actinides will tend to be absorbed by most soils with fairly high distribution coefficients, although perhaps with a fairly low retention capacity. The result may be the diminution of the actinide transfer rate, perhaps to values which are low enough to allow for their decay before they reach the biosphere. Denham et al.⁽¹⁸⁾ in a theoretical study have estimated that a soil column with a length of 16 km (from disposal site to a river, sandy soil with a moderate exchange capacity) and an average ground water velocity of 30 cm per day, would induce retardation effects which might be as high as $10^5 - 10^6$ years.

Should all three barriers mentioned fail, then the α -emitters would reach the ground-water and become accessible to the biosphere. They would tend to distribute throughout the environment according to their ecological distribution patterns.

Although our knowledge of them is very limited and fragmentary, there is at least a hope that the environment itself might be a fourth barrier.

d) Distribution pattern of actinides in the environment

The information currently available on the ecological aspects of the dissemination of α -emitters in terrestrial and aquatic environments was obtained from various sources, such as the small systematic releases of a few nuclear plants (Windscale sea-discharge, Hanford discharge to the Columbia river etc.), surface and underwater burst tests (Eniwetok and Bikini atolls, Redwig tests, etc.), fall-out studies, and accidents involving nuclear devices, such as the SNAP 9A burn-up in 1964 (^{238}Pu release) and military aircraft disasters (Palomares,

Thule)⁽¹⁹⁻²¹⁾.

How well this information may apply to an accidental release from disposal sites is a matter for argument, but the following trends are evident (at least for plutonium, to which most of the literature data refer):

- Pu is rapidly depleted from both fresh-water and sea-water. The mechanisms involved are still debated. Physico-chemical phenomena such as adsorption on settling suspended matter probably plays a major role, but biological sedimentation via zoo- and phytoplankton may also play an important role.
- The sedimentary Pu is redistributed both in the depth of the sediment and on the bottom surface, again by physico-chemical or biological processes, probably with rather high rates of redistribution⁽¹⁹⁾.
- Pu tends to concentrate in parts which do not enter the food chain, such as shells and bones. Each ring of the food chain induces therefore a substantial decrease of the Pu level.

Clearly from this information it is not yet possible to draw any reasonable quantitative long-term picture of the ecological distribution of the α -emitters.

However, the fact that fall-out ²³⁹Pu has not concentrated to any considerable extent in any compartment of the various ecological systems on which data are available, at least in the relatively short time span of about 20 years, justifies the hope that ecological processes tend to dilute the sedimented Pu more than to concentrate it.

Most of the information available, which is already scarce, refers to Pu, while the hazard after 1,000 years is mostly due to Am, on which there is even less information.

The chemical properties of the two elements are such that the mobility of Am in its geochemical processes should be higher than that of Pu, while little can be said concerning the fate of Am in the biological pro-

cesses. Again, however, an educated guess may be that the behaviour of Am should not differ much from that of Pu, and presumably any difference should be towards a greater dispersion rate in the environment than that of Pu.

5. EFFECTS OF A HYPOTHETICAL ACCIDENT AND UNCERTAINTIES CONNECTED TO THEIR EVALUATION

It is still premature to estimate the value of the whole system of barriers in quantitative terms.

In order to have at least a rudimentary idea of the amounts of α -emitters which could be found in the environment following an accidental release, we have hypothesized the following accident:

- 1) after 1,000 years the first barrier fails, and the geological formation is flooded with water.
- 2) the container of a glass block has corroded to such an extent that the glass is exposed to the leaching action of the water. The glass block, however, maintains its integrity.
- 3) the water flows through the formation and finally arrives at a large water body. The water flow-rate and the ion exchange behaviour of the soil through which the water is flowing are such that the actinide content of the water is entirely transferred to the water body.
- 4) the actinides are rapidly depleted from the water, and accumulate in the sediment in the vicinity of the water inlet.

The calculations have been limited to the fate of ^{241}Am contained in a glass block from reprocessing of LWR fuels ($286 \mu\text{Ci/g}$, 62.8% of the total α -activity).

If a leach factor of 10^8 week/cm is assumed for ^{241}Am , according to the definition by Elliot and Auty⁽²²⁾, the leaching rate from the sur-

face of one glass block (about 2 m^2), would be approximately 0.6×10^{-4} g/day, or about $13 \mu\text{Ci}/\text{year}$ of total α -activity (assuming an identical behaviour for all the actinides).

If we hypothesize that the actinides will be uniformly distributed over a surface of 1 km^2 , without redistribution in time, a period of about 150 years would be required to obtain in the sediment the concentration reached by the fall-out ^{239}Pu as measured in a few typical samples from different continents ($2 \text{ mCi}/\text{km}^2$)⁽¹⁹⁾.

The α -activity per unit volume of sediment would be about 1.3×10^{-11} Ci/litre, which is about 60 times higher than the natural radioactivity of sea water⁽²³⁾. The redistribution of actinides by geochemical mechanisms and food chain transfer cannot be predicted with any degree of accuracy, as too much data is missing.

The build-up of actinides in the sediments at a rate of $13 \mu\text{Ci}/\text{km}^2/\text{yr}$ can be considered, as previously stated, only as a preliminary reference figure, which simplifies the evaluation of how much better or worse the real conditions could be.

A few remarks on the effects of the existing uncertainties are given below:

- 1) A leach factor of 10^8 week/cm was used in our calculations, with the assumption that the leaching rate of actinides should not differ much from that of the rare earth elements, to which the 10^8 figure applies⁽²²⁾. Uncertainties of an order of magnitude could easily exist.
- 2) The assumption that the glass block has conserved its integrity after 1,000 years is purely fictional. Should the glass become pulverized, then the exposed surface could easily be increased by a factor $10^3 - 10^4$.
- 3) The hypothesis that just one glass block is damaged, is also purely fictional. It is perhaps more probable that, if damage occurs to one, it would also occur to others, involving the entire stock of $10^3 - 10^4$ glass

blocks which might be present at the disposal site.

- 4) Due to the ion exchange properties of the geological strata through which water percolates the actinides will probably move at a much slower rate than the water. The hold-up time, as previously indicated, could reach values as high as 10^5 - 10^6 years. On the other hand the actinide concentration in the soil during percolation would be three orders of magnitude higher, and the hazard correspondingly will be higher, should the soil be brought for any reason directly to the earth surface.

In conclusion, it would seem that, if the hypothetical accident previously described is taken as reference, there are many unknown quantities, which could increase or decrease the hazard by factors as high as 10^6 .

As previously stated, the safety of the entire barrier system could be viewed in a probabilistic way, each barrier contributing with a certain probability distribution and at a certain confidence level. It is clear that, with such great uncertainties, the requirements set for the first barrier (segregation afforded by geological formation) must be very stringent. This does not mean, however, that the value of the barrier which follows is low. On the contrary, it would seem that if the range of the existing uncertainties could be somewhat narrowed, there is the probability that a safety system made up by a balanced combination of all the barriers would be the most valid.

The greatest effort should therefore be devoted to studies related to

- long-term physical stability of solidified waste,
- mechanism and rate of actinides leaching by ground-water,
- mechanisms and rates of transfer of actinides into the environment.

6. CHEMICAL SEPARATION OF ACTINIDES FROM HIGH ACTIVITY WASTES

Chemical separation of actinides could be considered as a possible additional option, which has a great advantage over the others mentioned in that it is not of a probabilistic nature.

Apart from the obvious advantage of having to deal with a decreased quantity of actinides, their chemical separation considerably increases the value of the second barrier, - the physical stability of the glass blocks. The energy deposited in the glass by α -decay would, in fact, have largely diminished, and the probability that the glass maintains its physical integrity would be correspondingly increased, also taking into account the increased importance of spontaneous annealing of radiation damage.

The benefits of chemical separation probably extend also to the third barrier, the retention of actinides by the immediate surrounding environment, since absorption takes place with a high distribution coefficient but a low retention capacity.

The obvious disadvantages are a greater complexity and cost in the reprocessing operations, with the necessity of a separate disposal system for the separated actinides, which must, of course, be intrinsically safer.

The data reported in Table III and IV for LWR and FBR wastes clearly show that improvements of the extraction of Pu and U in the first extraction cycle can bring only minor advantages to the long-term α -activity of the waste, since the contribution of uranium isotopes is irrelevant and the ^{239}Pu and ^{240}Pu concentration derives mostly, in the long-term, from the decay chains of ^{243}Am and ^{244}Cm . A secondary chemical separation process of the aqueous waste is therefore required.

A calculation of the α -activity after a 1,000 years storage time was made for waste which has undergone various levels of decontamination. The waste considered was firstly a LWR waste, obtained after reprocessing of the fuel, with a recovery of 99.5% of Pu and U. It was assumed that the decontamination is carried out after a few months' storage, when only a minor amount of ^{244}Cm had decayed to ^{240}Pu .

A few years of interim storage in the liquid form, which might be necessary for technological reasons will, however, only have a minor effect on the $^{244}\text{Cm}/^{240}\text{Pu}$ ratio.

The results are shown in Table V as the total residual α -activity of glasses formed from the decontaminated waste as a function of the decontamination factor obtained for various actinides. The decontamination factor (D.F.) was defined as the ratio of the α -activity in the liquid waste before and after the removal of the various actinides.

A D.F. of at least 100 was taken into account as the very minimum required to pay for the additional complexity of reprocessing. In this case the elements which should be taken into consideration are Am, Cm, and Pu. The residual radioactivity ($6.7 \mu\text{Ci/g}$) would be divided among $^{241}\text{Am} + ^{243}\text{Am}, ^{240}\text{Pu}$ (mainly formed by ^{244}Cm decay) and ^{237}Np . It would be almost useless to improve their removal for a D.F. of higher than 10^3 (residual radioactivity $2.6 \mu\text{Ci/g}$) since the prevailing α -activity would be due to ^{237}Np .

In order to decrease still further the residual α -activity, it is necessary to anticipate the ^{237}Np separation. It would be sufficient to obtain a 90% extraction of ^{237}Np in the first extraction cycle to lower the residual radioactivity to $0.7 \mu\text{Ci/g}$, and only a slight advantage would be obtained by increasing the extraction of Np to 99% ($0.5 \mu\text{Ci/g}$ instead of $0.7 \mu\text{Ci/g}$).

Lower residual activities would require a process capable of giving a D.F. of 10^2 for Np and 10^4 for Pu, Am and Cm (residual α -activity $\sim 0.1 \mu\text{Ci/g}$).

A residual α -activity which is lower by a factor 10 would require a better extraction of U (at least 99.9% instead of the 99.5% assumed), a D.F. = 10^3 for Np and a D.F. = 10^5 for Pu, Am, and Cm. The residual radioactivity ($\sim 0.01 \mu\text{Ci/g}$) would be comparable with that of 0.4% U ores.

A reasonable target for the decontamination of high activity wastes seems to be a D.F. of 10^3 for Pu, Am and Cm (obtained by a specific process on the aqueous waste) and a D.F. = 10 for Np (preferably obtained by co-extracting Np during the recovery of U and Pu values with a 90% yield). The residual α -activity of the glass ($0.7 \mu\text{Ci/g}$ after 1,000 years storage) would seem low enough to greatly decrease concern about the possible failure of the entire barrier system, although more quantitative evaluations are still required. Figure 3 shows the decay curve of high activity waste which was subjected to such a decontamination procedure and then vitrified.

The results of similar calculations are shown on Table VI for FBR waste for three decontamination levels.

The effects of increasing D.F. are very similar to those obtained on LWR fuels, but, as previously mentioned, the uncertainties on the spent fuel composition are probably too high for a conclusion to be drawn on the decontamination required. It might seem reasonable however, to state that a separation process designed for a balanced reduction of Pu, Am, Cm and Np from LWR waste should be applicable without prohibitive changes to FBR fuels also.

7. CONCLUSIONS

The concentration of the long-lived α -emitters in vitrified high ac-

tivity wastes arising from nuclear fuel reprocessing was calculated under specified reference conditions.

The assessment of the long-term hazard to man of such waste, when permanently disposed of in a deep geological formation, seems rather difficult at present. Fundamental knowledge of the many factors which should be taken into account in case of failure of the segregation is rather limited, and a direct comparison with existing α -bearing sources such as uranium ores or mill tailings from uranium mining does not account for the different pathways by which the α -emitters could reach man in different cases.

An attempt to evaluate the long-term hazard to man is made by using the "barrier" approach derived from the analysis of nuclear reactor safety. The results of a first analysis of the problem are briefly sketched in Figure 3.

Four barriers were identified:

- quality of the segregation by the deep geological formation,
- stability of conditioned waste (chemical and physical),
- retention properties of the immediate surrounding environment,
- distribution patterns of actinides in the environment.

The resulting barrier system can be evaluated in a probabilistic way, by taking into account the considerable uncertainties existing in each barrier, and work is in progress in that direction.

The analysis of a fictional accident, with all the reservations which are imposed by the large number of hypotheses carried out, suggests that the barrier system would probably supply good long-term safety. Additional studies, particularly on the stability of the conditioned waste and the ecological properties of the environment towards the actinides, could considerably improve the value of the barrier system.

Chemical separation of actinides from high activity waste would be an additional option of undoubted value for the disposal of high activity waste. Its value for the overall safety of the entire waste inventory depends on many factors which need better evaluation, such as safe disposal of the separated actinides and the amount and quality of the additional waste generated by the separation process.

Work is in progress to assess the possibility of burning the actinides in nuclear reactors (thermal reactors or fast breeders). The burn-out in a thermonuclear reactor may also become a most effective solution of the problem in the long terms⁽²⁴⁾.

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TABLE I : Actinides concentration (g/ton) assumed for spent fuels of light water (LWR) and fast breeder reactors (FBR).

	LWR Burn-up 33.000 MWd/t	FBR Burn-up 80.000 MWd/t
^{242}Cm	8	42.3
^{244}Cm	31	41.9
^{241}Am	50	1460
^{243}Am	92	711
^{237}Np	450	180
^{238}Pu	168	1840 (2590 *)
^{239}Pu	5300	117000 (130000 *)
^{240}Pu	2140	52400 (51800 *)
^{241}Pu	1000	14400 (26000 *)
^{242}Pu	340	9020 (8760)
^{235}U	8000	7
^{236}U	4520	10
^{238}U	940000	719000 (782000)

* assumed concentration in the fresh fuel (9).

TABLE II : Calculated residual α -activity of LWR and FBR spent fuels after 1000 years' storage (curie/ton)

	LWR	FBR
^{242}Cm	-	-
^{244}Cm	-	-
^{241}Am	755.6 (49%)	11379 (39%)
^{243}Am	16	123.5
^{238}Pu	1	11
^{239}Pu	319.9 (21%)	7054.7 (24%)
^{240}Pu	445.4 (29%)	10764.7 (36%)
^{242}Pu	1.3	35
^{237}Np	0.9	9
^{238}U	0.3	0.2
^{235}U	0.02	-
total	1540.7	29377

TABLE III : α - activity of solidified LWR waste ($\mu\text{Ci/g}$)
vs. decay time

Years	10^1	10^2	10^3	10^4	10^5	10^6
^{244}Cm	11000	325	-	-	-	-
^{241}Am	1207	1108	286	-	-	-
^{243}Am	106.7	105.6	97.7	44.9	0.018	-
^{238}Pu	1091.4	546	0.52	-	-	-
^{239}Pu	11	11.4	13.6	23.4	2.58	-
^{240}Pu	30.5	60.1	54.4	21.2	0.0015	-
^{242}Pu	0.046	0.046	0.046	0.046	0.039	0.007
^{237}Np	2.13	2.17	2.35	2.40	2.34	1.70
^{234}U	0.03	0.23	0.42	0.41	0.32	0.026
^{235}U	0.0006	0.0006	0.0006	0.0008	0.002	0.002
^{236}U	0.009	0.009	0.011	0.020	0.026	0.025
^{238}U	0.0104	0.0104	0.0104	0.0104	0.0104	0.0104
Total	13450	2159	455.2 ^(*)	92.38	5.337	1.774

(*) per cent contribution of the various isotopes : ^{241}Am 62.8%, ^{243}Am 21.5%,
 ^{240}Pu 12%, ^{239}Pu 3%, ^{237}Np 0.5%, U total 0.1%.

TABLE IV : α - activity of solidified FBR waste ($\mu\text{Ci/g}$)
vs. decay time

Years	10^1	10^2	10^3	10^4	10^5	10^6
^{244}Cm	5250	157.8	-	-	-	-
^{241}Am	12216	10866.3	2582.7	0.001	-	-
^{243}Am	300	297.5	274.4	122	0.04	-
^{238}Pu	1519.5	738.2	0.54	-	-	-
^{239}Pu	80.7	81.2	86.6	110.2	13.05	-
^{240}Pu	139.4	152	138.7	53.9	0.004	-
^{242}Pu	0.4	0.4	0.4	0.384	0.325	0.063
^{237}Np	0.284	0.794	2.372	2.894	0.809	2.09
^{238}U	0.003	0.003	0.003	0.003	0.003	0.003
^{236}U	-	-	-	0.028	0.042	0.042
^{235}U	-	-	-	-	0.006	0.006
^{234}U	0.040	0.31	0.57	0.56	0.43	0.036
^{233}U	-	-	0.006	0.119	1.025	2.276
Total	19506	12294	3086(*)	290	18	4.5

(*) per cent contribution of the various isotopes :

^{241}Am 83.7%, ^{243}Am 8.9%, ^{240}Pu 4.5%, ^{239}Pu 2.8%.

TABLE V : Residual α -activity of solidified LWR waste, obtained after various chemical treatments on the aqueous waste to remove the actinides

Decontamination obtained				activity of vitrified waste after 1000 years residual α -emitters (%)			
extraction in the first cycle			decontamination factor from high activity waste			/BC/g	
U	Pu	Np	Pu	Am	Cm		
99.5	99.5	0	0	0	0	455	Am + Pu 99.3
99.5	99.5	0	10 ²	10 ²	10 ²	6.7	Am + Pu 67 , Np 32
99.5	99.5	0	10 ³	10 ³	10 ³	2.6	Am + Pu 17.3 Np 81.6
99.5	99.5	0	10 ⁴	10 ⁴	10 ⁴	2.2	Am + Pu 2.1 Np 96.5
99.5	99.5	90	10 ²	10 ²	10 ²	4.8	Am + Pu 94 Np 4.4
99.5	99.5	90	10 ³	10 ³	10 ³	0.7	Am + Pu 65 Np 31
99.5	99.5	90	10 ⁴	10 ⁴	10 ⁴	0.3	Am + Pu 15.6 Np 74 U 10,4
99.5	99.5	99	10 ²	10 ²	10 ²	4.6	Am + Pu 99
99.5	99.5	99	10 ³	10 ³	10 ³	0.5	Am + Pu 92 Np 2 U 6
99.5	99.5	99	10 ⁴	10 ⁴	10 ⁴	0.096	Am + Pu 47 Np 22 U 31
99.5	99.5	99	10 ⁵	10 ⁵	10 ⁵	0.057	Am + Pu 8 Np 37 U 55
99.9	99.5	99.9	10 ⁵	10 ⁵	10 ⁵	0.012	Am 33, Np 17, U 50

TABLE VI : Residual α -activity of solidified FBR waste, obtained after various chemical treatments on the aqueous waste to remove the actinides

Treatment	Pu, Am, Cm decontamination factor : 10^3		Np extraction 90 % Pu, Am, Cm decontamination factor : 10^4		Np extraction 99 % Pu, Am, Cm decontamination factor : 10^5	
	$\mu\text{Ci/g}$	%	$\mu\text{Ci/g}$	%	$\mu\text{Ci/g}$	%
^{241}Am	2.583	76.7	0.258	76	0.026	70
^{243}Am	0.274	8.1	0.0274	8.1	0.003	
^{239}Pu	0.086	2.5	0.0086	2.5	0.001	
^{240}Pu	0.139	4.1	0.0139	4.1	0.001	
^{237}Np	0.283	8.4	0.0283	8.3	0.003	
^{238}U	0.003	0.09	0.003	0.9	0.003	
total	3.37		0.339		0.037	

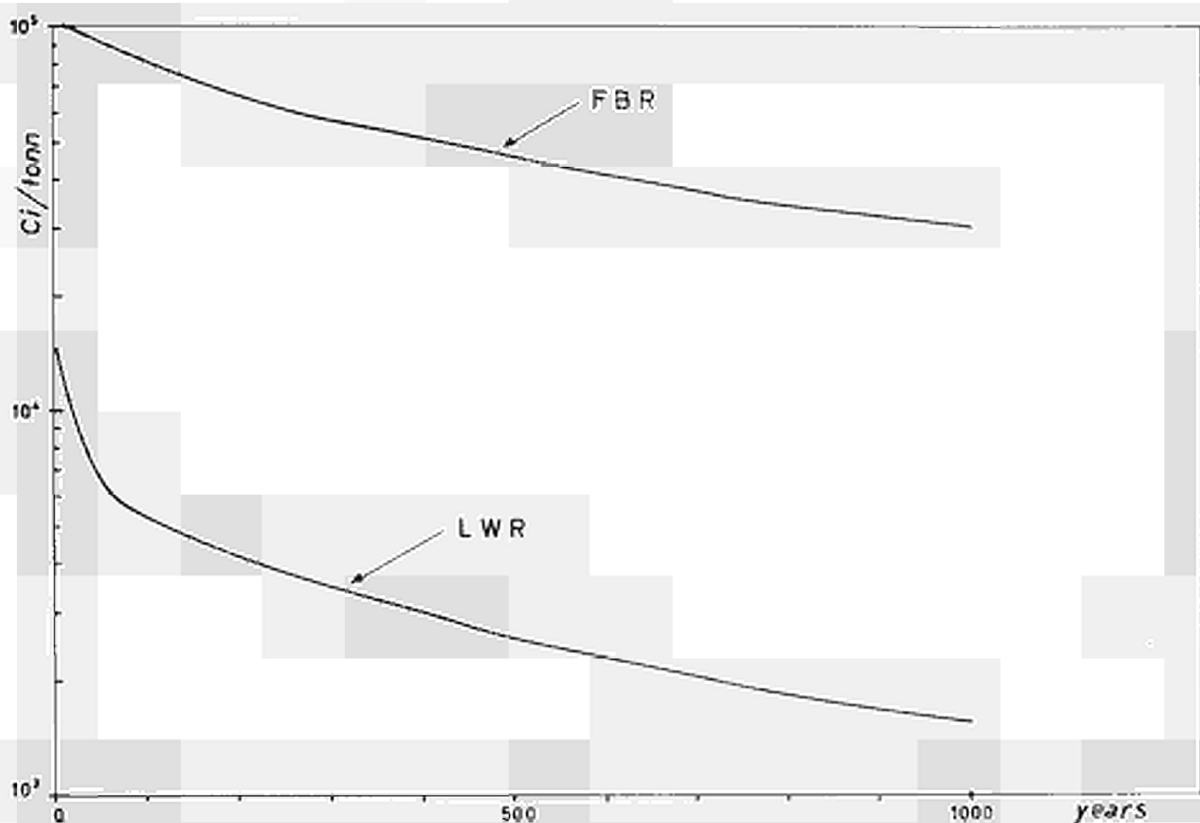


FIGURE 1. Calculated total α -activity of spent LWR and FBR fuels vs decay time.

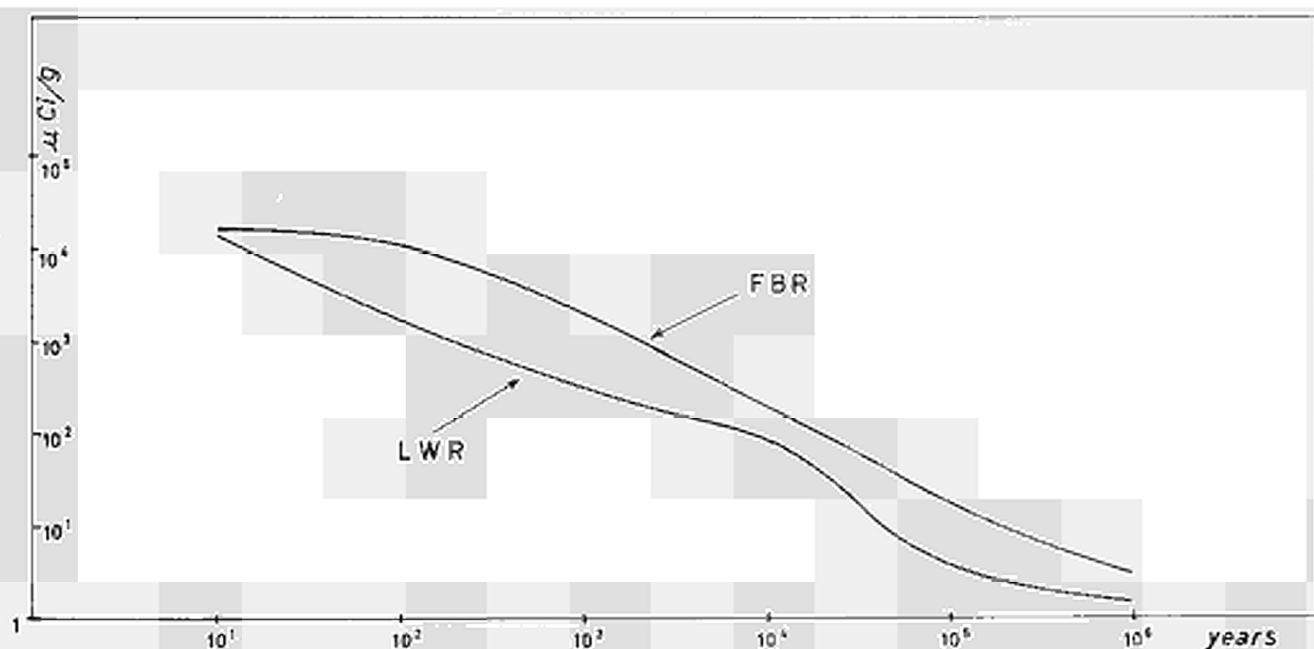


FIGURE 2. Calculated total α -activity of solidified waste from spent LWR and FBR fuels vs decay time.

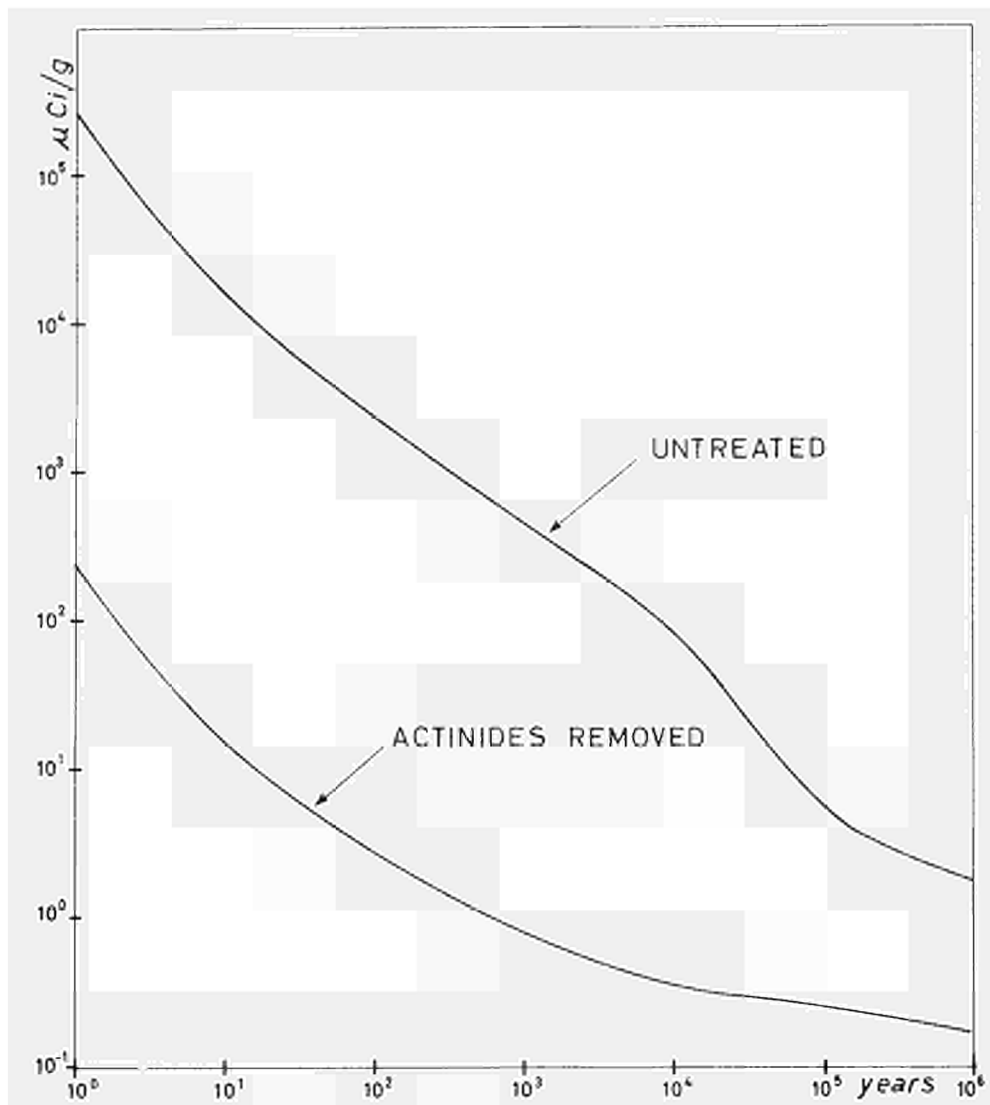


FIGURE 3. Calculated α -activity of solidified waste from spent LWR fuels, obtained after chemical separation of actinides, vs decay time.
 Process: Coextraction of 90% Np with U and Pu values and Separation of 99.9% Pu, Am, Cm from high activity aqueous waste.

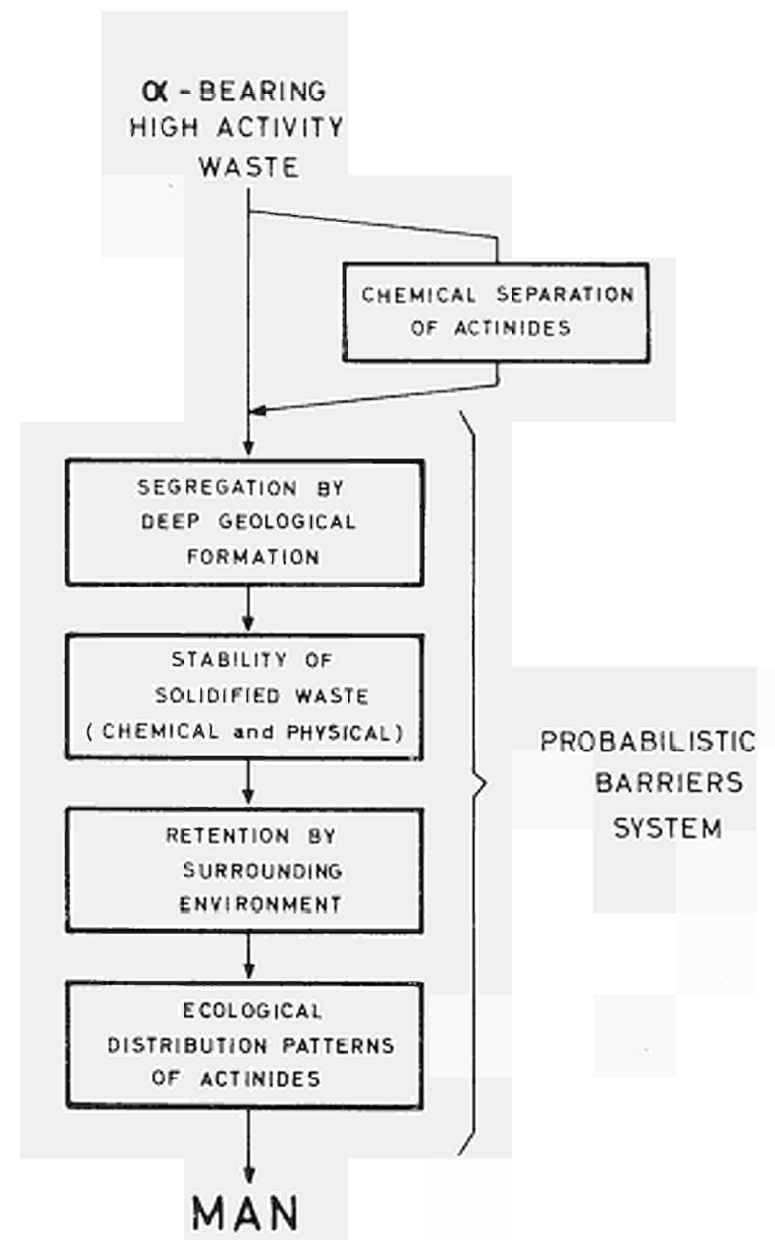
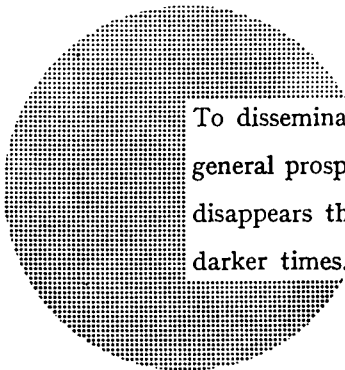


FIGURE 4. The "Barriers" approach applied to hazard analysis of high activity waste permanently disposed in a deep geological formation.

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