CRITICALITY SAFETY ANALYSIS OF THE ACCELERATOR TRANSMUTATION WASTE SYSTEM

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ABSTRACT

The Accelerator Transmutation Waste system (ATW) is under development at the Los Alamos National Laboratory, it consists of a particle accelerator producing a proton beam having an energy of 1.5 GeV, these particles are introduced in the upper part of a molten Pb-Bi column and they produce by spallation reaction a high strength neutron flux ($1.0X10^{16}$ n/(cm^2 sec). The neutrons enter on a heavy water blanket where the Actinides and long-lived Fission Products circulate in vertical tubes.

Our goal is to perform an independent verification of the feasibility of actinide burning in the ATW system.

Our work is divided in four tasks:

- a) production of an actinide and long-lived fission product cross section library from JEF 2.2;

- b) simulation, using MCNP and KENO IV Monte Carlo codes, of the ATW configurations existing in literature;

- c) validation of the cross sections by comparison of Keff and reaction rate results, calculated with MCNP and KENO IV, with experimental benchmarks and intercomparison between our calculations of a PWR unit cell and the computations carried out with various codes and cross section libraries (NEACRP criticality working group data);

- d) simulation of the ATW configuration.

The two first tasks are almost completed with excellent agreement between our results and Los Alamos one.

SINTESI DELLA RELAZIONE ALLEGATA

Questo lavoro è diviso in tre sezioni: l'impostazione generale del problema del bruciamento degli attinidi, la valutazione delle potenziali prestazioni dei più importanti progetti presentati negli ultimi dieci anni e gli aspetti di sicurezza connessi ad ognuno di essi per considerare la loro autorizzabilità.

L'impostazione generale presenta i parametri fisici che determinano la trasmutazione degli attinidi. Cinque variabili sono state considerate: la matrice contenente gli attinidi, il tempo d'irraggiamento, l'intensità del flusso neutronico, e i fattori di decontaminazione raggiunti nella fase di ritrattamento del ciclo del combustibile.

Quattro sistemi di bruciamento di attinidi sono stati considerati: il sistema di trasmutazione di scorie radioattive di Los Alamos (ATW, soltanto come bruciatore di rifiuti), il proggetto OMEGA (sviluppato a JAERI, Giappone), l'idea SIEMENS e il Reattore Veloce Integrale (IFR, dell'Argonne National Laboratory, Stati Uniti d'America).

Per considerare gli attinidi bruciati, l'obiettivo da raggiungere è che dopo l'irraggiamento e 500 anni di decadimento delle strutture che li contengono, il rischio ad essi associato sia minore di quello associato al minerale d'Uranio naturale impiegato nella fabricazione del combustibile fresco, pertanto recuperando la situazione naturale.

Le variabili che determinano il bruciamento degli attinidi sono:

il flusso neutronico e i fattori di decontaminazione.

I quattro sistemi analizzati raggiungono lo scopo, ma a differenti velocità. In effetti il rapporto tra i tassi di trasmutazione e produzione nel sistema di Los Alamos è il più conveniente.

Il sistema di Los Alamos può bruciare i prodotti di fissione a lunga vita.

Il progetto IFR ha un ciclo del combustibile molto pulito (può bruciare i propri attinidi più quelli scaricati degli LWR) ma, per distruggere gli attinidi prodotti dagli LWR, occorrerebbe una popolazione di IFR tre volte quella degli LWR.

Ogni opzione del progetto OMEGA potrebbe bruciare più della produzione di dieci LWR, ma presenta seri problemi del punto di vista della autorizzabilità.

ANALYSIS OF MINOR ACTINIDE BURNING PROBLEMS

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1. INTRODUCTION

Deep burial was been considered the most promising arrangement for high level nuclear waste. Today, the need of public consent imposes to analyze new solutions such as the transmutation of actinides. Those are the nuclides responsible of the long term radiological risk. The aim is that the storage time for nuclear waste in the final repository could be eliminated or at least reduced.

In the last years, several systems for actinide burning were proposed to achieve such goal.

The fuel cycle associated to commercial nuclear reactor, would be modified by adding two more steps to the normal fuel cycle: partitioning and transmutation. In the partitioning step, actinides are extracted from the HLW (High Level Waste) produced in reprocessing plants, while, in the transmutation step they are transmuted in nuclides with shorter mean life or smaller activity.

Our work aims to give a contribution in evaluating the intrinsic capacity of the proposed burning systems comparing the potential risk associated to the transmuted material with the one associated to the original mineral coming from the environment.

In the frame of the previous consideration, the main parameters effecting the transmutation were investigated. To compare the potential risk due to the waste produced by each actinide burning system, among themselves and with those of radioactive materials existing in nature, a unit of measure for the risk has been defined.

The most of actinides mass present in spent fuel is Pu (about 94%). Due to the diversity of solutions and of the physical implications, it was

found convenient to split the actinide burning problem in two areas: minor actinide and Pu transmutation. In the present paper, only minor actinides are considered.

2. THE RISK INDEX

To compare the potential risks due to the wastes produced by each actinide burning system, among themselves and with those of radioactive materials existing in nature (assumed as reference value to decide when the actinides are successfully burned), a unit of measure for the risk has to be defined.

The potential biological hazard may be measured by the quantity of water required to dilute each individual nuclide to its Radiation Concentration Guide (RCG) value for the unrestricted use of water.

To evaluate the importance of the parameters acting on the biological hazard, a sensibility analysis using ORIGEN2 /1/ code were carried out. The actinide isotopic composition of a standard PWR spent fuel is used in all calculations. The PWR is considered using 3.3% enriched fuel having a specific power of 30 MW/tU and at a burnup of 33 GWd/tU. The envisaged fuel cycle can be summarized as follows: one tonne of standard PWR spent fuel is treated in a reprocessing plant; subsequently actinides are stripped from HLW in a partitioning plant; finally they are dispersed in a matrix (inert or nuclear fuel) and irradiated in a transmutation system. The initial actinide inventory is assumed equal to 100 times the one coming from a PWR.

In the sensibility analysis, the effect of the following parameters was evaluated: the nature of the matrix; the irradiation time; the neutron flux level; the neutron spectrum; the decontamination factor; the spent fuel cooling time between discharge and transmutation.

Calculation were carried out for both thermal /2/ and fast /3/.

3. EVALUATION OF PARAMETER IMPORTANCE

For the transmutation systems here considered, we have an actinide concentration within the range $5.0\ 10^{-3}$-$8.0\ 10^{-3}$ g/cm^3 for ATW /4/ and 2.16 g/cm^3 in the OMEGA project; that means from 1 to 300 times the PWR spent fuel.

Our calculations were carried out considering an actinide concentration of 100. In any case, it is possible to demonstrate that actinide and fission product hazard is proportional to the actinide initial concentration /5/. Then all results can be extended to any actinide concentration using an appropriate scaling factor.

3.1. Influence of the matrix on the environmental impact

To analyze the dependence of the environmental impact on the matrix containing actinides, four matrices were considered: ZrO_2, CW_2, UO_2 and ThO_2. For such cases, the actinide concentration assumed is 10 times that of a PWR spent fuel and the irradiation time is 1100 days. The flux values used in both ORIGEN and ORIGEN2 /6/ were modified to take into account the different absorption properties of each matrix.

The minimum amount of unburned actinides is obtained using a ZrO_2 matrix: it is about one fifth that for a UO_2 matrix. For, geological times (more than 5000 years) the risk of the fission products is negligible compared to the actinide hazard.

From the burning point of view, the most efficient behavior corresponds to the inert matrices (ZrO_2 and CW_2), the worse to ThO_2.

The absence of ^{237}Np reduces by a factor of 4 the total mass of actinides remaining in the ZrO_2, matrix; for the same matrix the ratio between hazards at the reactor discharge with and without ^{237}Np is about 1.5. This value remains the same after 5000 years. This means that the absence of ^{237}Np reduces the environmental impact in an important manner if the index considered is the total mass of actinides; on the other hand such reduction is less important if the index is the radioactive hazard. The ZrO_2 matrix was selected for the next calculations.

3.2. Environmental risk versus irradiation time

In evaluating the influence of the irradiation time on the environmental risk, the following values are considered: 0, 1100, 3300, 9900, 11000 and 22000 days. The neutron flux is 2.81 10^{13} n/cm^2 s.

From the result, ref /7/, it is possible to state that for irradiation times below 9900 days, it is more convenient to bury actinides because the hazard level obtained by irradiating is higher than that without irradiation in geological times.

The agreement between ORIGEN and ORIGEN2 gets worse increasing irradiation time: the relative difference between the total mass of actinides raises to 29.92% from 5.14%. Calculations with ORIGEN2 are the most conservative ones; they always lead to a greater amount of unburned actinides. ORIGEN2 is the most conservative code; for this reason, it was selected for the next calculations.

The differences in the total mass of actinides and the risk ratios obtained by calculations performed with or without ^{237}Np significantly decreased by longer irradiation times.

As irradiating for the whole reactor life reduces the hazard only two orders of magnitude, the irradiation time parameter does not solve the actinide burning problem.

3.3 Environmental risk versus neutron flux

In evaluating the environmental risk versus neutron flux, the irradiation time was 11000 days and the flux values were in the LWR range: from 2.5 10^{13} to 8.43 10^{13} n/cm²/.

The results show that, after few hundred years, the hazard of actinides is only two orders of magnitude lower than natural decay, the neutron flux, within the selected range, does not solve the actinide burning problem.

Considering that there are in literature new actinide burning systems having thermal fluxes higher than LWRs, such as the Siemens idea /8/ and the Los Alamos Spallation System /4/, higher thermal fluxes were explored ranging from 1.0 10^{14} to 5.0 10^{15} n/cm² s.

The resulting behavior of the concentration of the most important long-lived fission products as a function of the neutron flux is:

^{99}Tc: concentration is inversely proportional to the neutron flux (the concentration decreases by one order of magnitude when the flux increases by one order of magnitude);
^{129}I: concentration decreases sharply when the flux increases from 2.81 10^{13} to 5.0 10^{14} n/cm² s, afterwards it decreases slowly;
^{135}Cs: concentration has a behavior similar to ^{129}I, but less marked.

Results show that the ratio between the actinide hazard at the beginning and at the end of the irradiation in the transmutation system has a dramatic drop for flux levels greater than 5.0 10^{14} n/cm² s.

The flux results one of the most critical parameters in determining the actinide burning because, for fluxes greater than 5.0 10^{14} n/cm² s, the hazard can be reduced by more than six orders of magnitude, for any cooling time.

It should be noted that at high flux level, the hazard due to the inert matrix is comparable with the fission product one. For the above reason the matrix is another important physical characteristic in determining actinide burning: it is necessary to use a matrix of low atomic weight.

The fission product hazard increases with flux up to 1.0 10^{15} n/cm² s. With flux at 5.0 10^{15} n/cm² s the fission products hazard begins to decrease. It demonstrates that long lived fission product burning is possible. It is a very important achievement, because to solve the problem of long-lived radioactive waste transmutation, it is necessary to burn long-lived fission products too.

3.4. Neutron spectrum

Fast flux values in the range from 5.0 10^{15} to 1.0 10^{17} n/cm^2 s were considered; the selected irradiation time was 11000 days.

It results that for a flux of 5.0 10^{16} n/cm^2 s the actinide hazard has a step wise decrease.

The total radioactive ingestion hazard (actinides + fission products + activation products) increases, /3/, with the neutron flux level due to the matrix activation product contribution.

3.5. Influence of the decontamination factor on the environmental impact

For our purpose, decontamination factors signify the percent of uranium and plutonium removed in the reprocessing phase. Three decontamination factors were considered: 99% (corresponding to an industrial reprocessing plant), 99.9% and 99.99% (reached in research laboratory); and two values of the thermal neutron flux: 5.0 10^{14} n/cm^2/s and 5.0 10^{15} n/cm^2/s. The selected irradiation time is 11000 days.

By examining the result reported in /2/ we can say:

- for a low flux the decontamination factor is a very important parameter in determining the hazard even though irradiation has a useful effect: for realistic values (99%) the hazard after 11000 irradiation days results still comparable with the one relative to the actinide natural decay;

- for a high flux level the influence of the decontamination factor on the resulting hazard is weak: the curves relative to a decontamination factor of 99.99% almost coincide with the ideal one (decontamination factor equal to 100%) while for a value of 99% the hazard rises by less than one order of magnitude.

These results demonstrate that the decontamination factor has the same influence of the neutron flux on the environmental impact.

3.6. Influence of cooling time

The following Tables I and II show the minor actinide hazard as function of the cooling time. The resulting hazard is almost independent by the parameter. It is possible to see the "threshold" effect of the flux for both thermal and fast spectra.

Table I.

Flux (n/cm2 s)	160 days	1100 days	3300 days	11000 days

$5.0\ 10^{13}$	0.027	0.039	0.035	0.033
$1.0\ 10^{14}$	0.005	0.007	0.006	0.006
$5.0\ 10^{14}$	$6.44\ 10^{-7}$	$9.70\ 10^{-7}$	$9.29\ 10^{-7}$	$9.15\ 10^{-7}$
$1.0\ 10^{15}$	$7.82\ 10^{-8}$	$1.11\ 10^{-7}$	$9.87\ 10^{-8}$	$9.17\ 10^{-8}$

Ratio between the minor actinide hazard at discharge and charge to the transmutation machine, thermal fluxes , irradiation time 11000 days.

Table II.

Flux (n/cm2 s)	160 days	1100 days	3300 days	11000 days
$5.0\ 10^{15}$	0.035	0.063	0.035	0.033
$1.0\ 10^{16}$	0.002	0.003	0.006	0.006
$5.0\ 10^{16}$	$1.12\ 10^{-10}$	$1.45\ 10^{-10}$	$1.11\ 10^{-10}$	$9.00\ 10^{-11}$
$1.0\ 10^{17}$	$1.09\ 10^{-10}$	$1.40\ 10^{-10}$	$1.06\ 10^{-10}$	$8.62\ 10^{-11}$

Ratio between the minor actinide hazard at discharge and charge to the transmutation machine, fast fluxes , irradiation time 11000 days.

4. THE TARGET

After irradiation and a 500 years decay of the structures containing the actinides, the value for actinide risk must be lower than that of the natural uranium ore used to manufacture the fresh fuel, in order to restore the natural situation.

The above decay time was selected so that the final storage deposit could be designed for a long period of time but not for millions of years. On the other hand 500 years is the age of many old buildings made by the man and still existing today.

The quantitative definition of this criteria presents two problems:

- the file of specific hazard data of ORIGEN2 was taken fror ICRP-2 /9/ (in the subsequent ICRP documents the actinide radiotoxicity was increased);
- the actinide hazard should be compared with the hazard corresponding to the mass of uranium ore needed to produce the fuel.

For the above reasons, before carrying out the calculations to simulate the actinide burning systems, the ORIGEN2 specific hazard file was updated using the data from /10/.

4.1. Calculation method

The hazard due to all nuclides included in the uranium ore except uranium and its daughters in secular equilibrium was considered negligible. Therefore, the hazard corresponding to the fresh fuel plus that of the uranium daughters in secular equilibrium becomes the same as that of the natural uranium ore. This is reasonable for LWR fuel, while for IFR fuel, the hazard was considered due to the amount of PWR fresh fuel needed to produce the plutonium mass present in the IFR.

The radiotoxicity data coming from ORIGEN2 calculations were divided by the decay data corresponding to PWR fresh fuel or, in the IFR case, by the decay data corresponding to the PWR fresh fuel needed for the plutonium production, which is the corresponding reference value.

5. THE BURNING SYSTEMS

In the last ten years, six new designs suitable for actinide burning were presented:

- three fast reactors, under the OMEGA Project name, developed by Japan Atomic Energy Research Institute (JAERI): a sodium cooled reactor with metallic fuel, an helium cooled reactor with particle fuel incorporating actinides and a subcritical reactor with metallic fuel driven by a spallation source;

- Integral Fast Reactor (IFR) /11/ project developed by Argonne National Laboratory, it is a fast sodium cooled reactor using metallic fuel with pyrometallurgic reprocessing remote refabrication;

- the "Siemens Idea" that consists in using a minor actinide layer on the inner side of a PWR cladding;

- the Los Alamos National Laboratory project "Actinide Transmutation Waste" (ATW) that is a subcritical thermal reactor driven by a spallation source /12/.

5.1. ORIGEN2 Simulation of the Los Alamos system

Calculations were performed considering a thermal neutron flux of 1.0 10^{16} n/cm^2 s and with the actinides contained in a FNa matrix at the same concentration as that in PWR spent fuel.

The simulation shows that the actinides are burned after an irradiation time of 25 days. In fact, already before 500 years, the actinide and fission product hazards become lower than the hazard associated with natural uranium ore. Furthermore, for 100 irradiation days, the actinide and fission product hazard becomes lower than the hazard associated with natural uranium ore just at the end of the transmutation phase.

At the reactor discharge, the radioactivity of ^{99}Tc and ^{129}I are 1.108 10^{-2} Ci and 5.766 10^{-5} Ci respectively. It can be useful to remember that the radioactivities for the above mentuoned two longlived fission products are 13.09 Ci and 3.134 10^{-2} Ci, for each tonne of standard PWR spent fuel.

Then, we can conclude that a system having these characteristics can burn within 25 days the actinides produced by a PWR during 1100 days, generating about one thousand times less longlived fission products than a standard PWR.

5.1.1. The importance of the decontamination factor

From the results for the Los Alamos System performances, it is possible to state that: for a decontamination factor of 99.99% this system can burn actinides at a rate 44 times faster than the production rate, while this ratio drops to 11 for a decontamination factor of 99.9%. In addition, for a decontamination factor of 99%, calculations show that to reach the target, an irradiation period of 300 days is necessary; therefore the ratio between burning and production rates is 3.6.

The ratio between the value of ^{99}Tc radioactivity in PWR spent fuel and after burning changes from 1181 for a decontamination factor of 100% to 34.72 for 99%, while the ratio between the value of ^{129}I radioactivity in PWR spent fuel and after burning changes from 544 for a decontamination factor of 100% to 19.53 for 99%.

5.1.2. The Los Alamos system as a fission products burner

This simulation was performed considering a thermal neutron flux of 1.0 10^{16} n/cm^2 s and with the most important, from the hazard point of view, fission products contained in a FNa matrix at the same concentration as in a PWR spent fuel. Two irradiation periods were considered: 25 and 100 days.

From the resulting behavior of the fission product hazard as a function of the decay time, it is clear that the irradiation at this high flux cannot burn

the short lived fission product (^{90}Sr and ^{137}Cs), but can reduce by more than one order of magnitude the long lived fission product hazard.

The ratio between ^{99}Tc radioactivity in PWR spent fuel and in the waste produced by Los Alamos system after 100 irradiation days is about 525, while the same ratio for ^{129}I becomes 48.

The Los Alamos APT system can burn the long lived fission products at a rate of 11 with respect to the production rate in a PWR.

5.2. Analysis of Siemens' idea

Calculations were performed considering actinides at the concentration in PWR spent fuel and a thermal neutron flux of $2.5\ 10^{14}$ n/cm^2 s.

The result is that actinides are burned after an irradiation time of 1100 days. For this irradiation period the actinide hazard becomes lower than that associated with natural uranium ore feed and the fission product hazard remains negligible.

This system could burn the actinides at the same rate as that of production.

5.3. Study of the IFR project

The ORIGEN2 calculations lead to a spent fuel composition which is in good agreement with that found in literature. The relative variation is within 30% for all nuclides except for some curium isotopes; this value is comparable with the errors introduced by the propagation of cross section uncertainties.

It can be stated that within the accuracy of ORIGEN2 calculations, the target is reached. The ratio between the value of ^{99}Tc radioactivity in PWR spent fuel and the corresponding IFR one is 0.32, while the ratio between the value of radioactivity due to ^{129}I in PWR spent fuel and IFR spent fuel one is 0.22. Therefore the production of ^{99}Tc and ^{129}I in IFR is 3 and 4.5 time that in PWR fuel respectively.

5.4. Simulation of the OMEGA project

OMEGA is the acronym derived from Options in Making Extra Gains from actinides and fission products. This project includes three options:

- M-ABR, Na cooled, minor actinide metal fuel burner reactor /13/,
- P-ABR, He cooled, minor actinide particle fuel burner reactor /13/

- Accelerator-driven actinide transmutation reactor, which consists of a Na cooled subcritical core with actinide metal fuel and a tungsten spallation target /14/.

The accelerator driven actinide transmutation reactor has the same type of metallic fuel and a neutron flux in the range of that of the M-ABR inner and outer cores. Furthermore, the flux value is the half that of the P-ABR, but the actinide concentration is approximately twice since the P-ABR fuel is made of particles of actinide nitrates. The decrease of the flux is compensated by the increase of concentration.

It means that the three options of the OMEGA project have approximately the same rate of actinide burning; for this reason, the accelerator-driven actinide transmutation reactor was chosen for the simulation.

The initial composition considered in the simulation, corresponding to 1 tonne of actinides, were taken from /14/, taking into account the actinide concentration ratios from /13/. A fast flux of 4.0 10^{15} n/cm^2 s was considered in the calculations and three irradiation intervals: 365 days, 500 days and 5500 days.

Taking into account that of the 3,160 kg of actinides present in the reactor core, 266 kg are burned within 365 days, the results are in good agreement with the 250 kg/yr of /14/.

The reference hazard was calculated from the amount of natural uranium ore used for the fabrication of the PWR fresh fuel from which, after irradiation, the americium mass present in the OMEGA accelerator-driven actinide transmutation reactor was stripped.

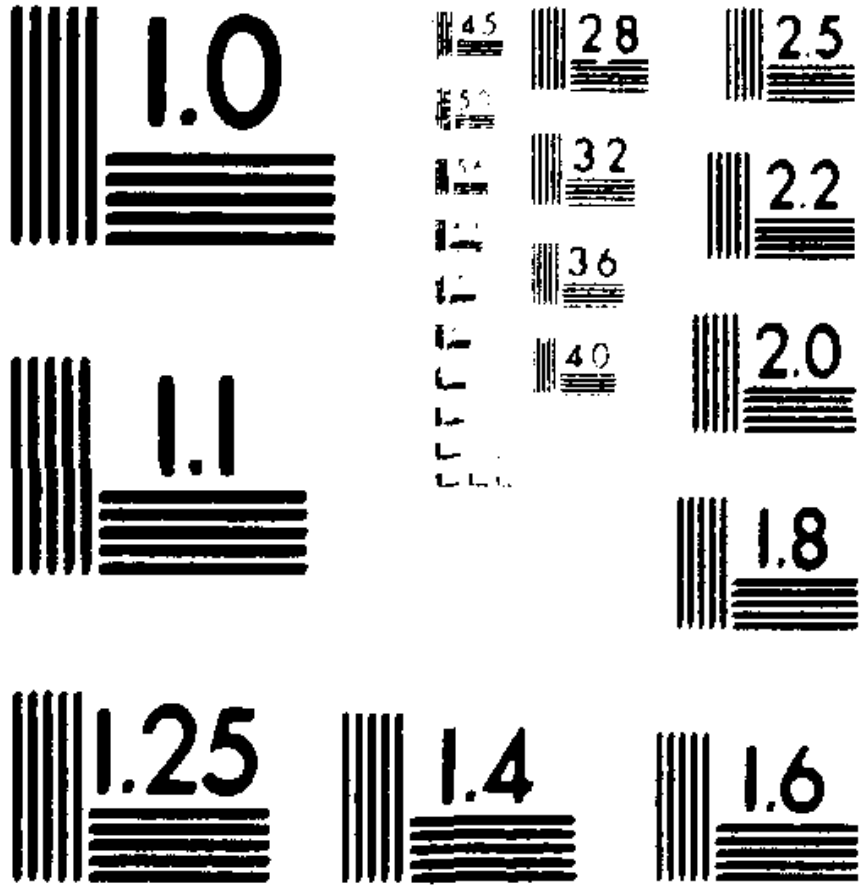
The results of this last calculation step show that after 5500 irradiation days and a decay time of less than 500 years, the actinide and fission product hazards are lower than the natural uranium ore hazard. This means that 3160 kg of the minor actinide inventory are burned within 5500 days (15 years).

Assuming a reactor life of 30 years and that the OMEGA accelerator-driven system is not refuelled in the last 15 years, it could burn 7150 kg of actinides that is more than the production of 10 LWRs, /14/.

6. CONCLUSIONS

The existing LWRs are not able to solve the minor actinide burning problem efficiently.

All four actinide burning systems analyzed reach the target but with different irradiation times. In fact, with respect to the ratio between



transmutation and production rates, the Los Alamos system is the most convenient one.

As regards the longlived fission product the Los Alamos system can burn the longlived fission products generated during the transmutation process.

Both the long-lived fission products production and the actinide transmutation-production ratio are strongly influenced by the decontamination factors in the reprocessing and partitioning phases.

As Siemens' idea, even with an ideal decontamination factor of 100%, burns actinides only at the production rate, it can not be considered a definitive solution, but it could be used in a program for the reduction of the HLW radiotoxicity.

IFRs have a very clean fuel cycle (they burn their own actinides plus those discharged from a PWR) but, to destroy the actinides produced by LWR reactors, an IFR population greater than the LWR one (about 3 times /15/) would be necessary. On the other hand the amount of long-lived fission products generated during the burning of the actinides is comparable to that in PWR spent fuel.

Each option of the OMEGA project could burn more than the actinide production of ten LWRs, but the could presents serious safety related problems because each option contains several critical masses of minor actinides which cannot exclude the recriticality of the molten core in case of severe accident. Moreover, the M-ABR and the P-ABR, due to the reduction of the delayed neutron fraction, give an energy release, in a reactivity accident, 4 times bigger than the PWR one.

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