Feasibility Analysis of Minor Actinide Burning in Thermal Reactor Systems

A. Buccafurni, ENEA DISPP.A. Landeyro, ENEA DRIA. Orazi, ENEA DISP

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1.- INTRODUCTION

Our work is devoted to analyze the different actinide burning systems with the purpose to evaluate their performances and verify the possibility to eliminate, or at least reduce, the storage time for the nuclear wastes in final deposit.

To achieve such goal two steps are added to the normal fuel cycle: partitioning and transmutation. In the partitioning step the actinides are extracted from the High Level Wastes (HLW) produced in reprocessing plants while, during the transmutation step they are burned.

To evaluate the potential performances of the different systems which could be suitable for actinide transmutation, it is necessary to. find and analyze the parameters acting on actinide burning as well as to compare the potential risk due to the wastes produced by each system with the one associated to radioactive material existing in nature.

Other important parameter to be considered is the ratio between the transmutation and generation speed which highlights how fast is the system in burning actinides.

For the above considerations, a preliminary analysis of the parameters which influence the actinide burning will be followed by the evaluation, as accurate as possible by using ORIGEN2 generation and depletion code, of some of the most important transmutation systems, operating with thermal neutron spectra, presented during the last ten years.

The most important actinide is plutonium because it represents the most of the actinide mass present in LWR spent fuel

(about **94%).** For the diversity of solution and physical implication, it is convenient to divide the actinide burning problem into two parts: plutonium burning and minor **actinide** burning. In the present report only the minor **actinide** burning problem is analyzed.

2 - TRANSMUTATION

A general form for the generation and disappearance of a **nuclide** nuclear transmutation and radioactive decay may be written /1/ as follows:

$$\underline{dN}_{i} = \sum I_{ij} \Lambda_{j} N_{j} + \Phi \sum f_{ik} \langle \sigma_{k} \rangle N_{k} - (\Lambda_{i} + \Phi \langle \sigma_{i} \rangle) N_{i} \quad (i = 1, ..., N) \quad (1)$$

$$dt$$

where: N_i is the atom density of nuclide i-th, $\langle \sigma_i \rangle$ is the spectrum-averaged neutron absorption cross section for nuclide i-th, A_i is the radioactive disintegration constant for nuclide i-th, lii and f_i are the fractions of radioactive disintegration and neutron absorption by other nuclides which lead to the formation of species i and Φ is position- and energy-averaged neutron flux, which is assumed to be constant over short intervals of time.

Rigorously, the system of equations (1) is non linear since the neutron flux will vary with changes in the composition of the fuel. However, the variation with time is slow and, if the neutron flux is considered constant over short time intervals, the system of equations (1) is an homogeneous set of simultaneous first-order ordinary differential equations with constant coefficients, which may be written in matrix notation:

~
$$(N) =$$
 $(N N) \cdot X (N) (2)$

Equation (2) has the known solution:

$$(\mathfrak{A} t)$$

$$\mathfrak{X} = \mathfrak{X} (0) \mathbf{e}$$
(3)

Where $\mathfrak{X}(\mathbf{O})$ is a vector of initial atom densities and \mathfrak{A} is a transition matrix containing the rate coefficients for radioactive decay and neutron capture

If the nuclide concentration relative to the total actinide mass is assumed to be constant and equal to the one existing in PWR spent fuel, the equation (3) can be rewritten as:

with the hypothesis to keep constant the volume of matrix in which the minor actinides are dispersed.

In equation (4) $\mathbb{Z}(O)$ is the vector of actinide atom densities of PWR spent fuel and C is an arbitrary multiplier factor.

3 - THE RISK INDEX

To compare the potential risks due to the wastes produced by each actinide burning systems among them and with the one relative to radioactive materials existing in nature (assumed as reference value to decide when the actinides are successful burned), a measure unit for the risk have to be defined.

The total risk associated with actinides plus fission and activation products generated during the transmutation process, may be described by:

$H = \sum h_{j}m_{j} + \sum h_{fp}m_{k} + \sum h_{ap}m_{i}(5)$

where:

H is the total hazard (risk),

h; the specific hazard of j-th nuclide actinide,

h_{fp} the specific hazard of k-th fission product.

n_{ap}the specific hazard of i-th activation product.

M n is the mass of the n-th nuclide.

The potential biological hazard may be measured by the volume of water that should nationality be added to the waste in order to render the resultant diluited water fit enough for drinking purpose by people. The hazard measure unit defined above is referred in literature /2/ with several names: Water Dilution Volume, Untreated Dilution Index, Toxicity Index etc..

The specific hazard of each nuclide can be written as follows:

h = S.A. / MPC (6)

where **S.A**, is the specific activity (Bq/g) and **MPC**, the Maximum Permissible Concentration (Bq/ems).

Using the last releases of ICRP(ICRP90/3/) **MPC** can be calculated from **ALI** (Annual Limit of Intake) and the average daily intake of water established for the standard man.

Considering the initial actinide concentrations proportional to the one in the standard PWR spent fuel and a constant volume for the matrix in which the actinides are dispersed, the total risk can be written as:

 $H = C \sum h_j m_j (Z_{0j}) + C \sum h_{fp} m_k (Z_{0j}) + \sum h_{ap} m_i (7)$

where the arbitrary multiplying factor C (see equation (4)) represents the ratio between the considered total **actinide** mass and that in PWR spent fuel while Z_{0j} is the atomic density of the j-th actinide nuclide in PWR spent fuel.

Equation (1) shows that **H** is a function of Φ , **t** (the irradiation time) and **Sp** (the spectrum) which determines the values of C σ >. From (7) it is clear that **H** is also a function of $\sum h_{ap}m$ i which is the expression of the hazard due to the activation of the matrix containing actinides. All these variables are characteristic parameters of the transmutation systems.

If the matrix activation product contribution can be neglected, \mathbf{H} is proportional to the initial concentration of the actinides in the transmutation machine. This concentrations depends on the reactor type as well as on the decontamination factors (**Df**), which represents the percent of uranium and plutonium removed in reprocessing and partitioning steps of the fuel cycle.

Using the functional relationship for the total hazard /4/, equation (7) becomes:

$H = C F(\Phi, t, Sp, Df) + H (matrix)$ (8)

As this document is expressly addressed toward the transmutation system operating with thermal neutron spectra, the importance of the above parameters, but the spectra, will be analyzed ahead.

4- THE STANDARD PWR SPENT FUEL

Standard PWR means a pressurized reactor using UO_2 fuel, uranium enriched 3.3 weight percent, and operating with a specific power of 30 MW/TU and a burnup of 33,000 MWD/TU.

The isotopic compositions of uranium and plutonium present in the standard PWR spent fuel above defined are listed on table 1.

5- THE FUEL CYCLE

The hypothesized fuel cycle can be summarized as follows: one ton of standard PWR spent fuel is treated in a reprocessing plant and successively actinides are stripped from the produced HLW in a partitioning plant; finally they are dispersed in some matrix (inert or nuclear fuel) and irradiated in the transmutation system.

6 - THE INITIAL ACTINIDE CONCENTRATION

In our calculations, the volume of PWR standard fuel containing a ton of uranium (about 9.615 10⁴ cm³) is filled with the matrix containing the actinides.

If the actinides are concentrated 100 times more than in PWR spent fuel, in this volume there are 65 kg of actinides with a density of 6.86 10⁻¹ gr/cm³ and composition listed on table II.

The presentation of the Los Alamos concept at the Summer Meeting of American Nuclear Society /5/ reports an actinide concentration within the range from 5.0 10-³ gr/cm³ to 8.0 10-³ gr/cm³ (mean value 6.5 10-³ gr/ems): that is the actinides initial concentration is practically the one existing in standard PWR spent fuel.

On the other hand, in the last version of the OMEGA project /6/ which is driven by a spallation source, the actinide density is about 2.16 gr/ems: the actinide initial concentration is practically 300 times that in standard PWR spent fuel.

Some of our calculations have been performed considering, at the starting of the transmutation step, an **actinide** concentration 100 times the Standard PWR spent fuel one. This value is within the range of the transmutation projects presented in the last 10 years.

in any case, if the actinide initial concentrations are proportional to those existing in standard PWR spent fuel, it is possible to demonstrate that the actinide and fission product hazard is proportional to the actinide initial concentration (see formula (7)). Then, all the results of this report can be extended to any actinide concentration simply by using an appropriate scaling factor.

7. " PARAMETER IMPORTANCE EVALUATION PROCEDURE

The Radioactive Ingestion Hazard measured by the quantity of water required for the dilution of each individual nuclide to its Radiation Concentration Guide (R. C. G.) value for unrestricted use of water was adopted as risk index by Claiborne in one of his first reports concerning the actinides transmutation (1 972) /7/, in such report the risks associated with pitch blende and carnotite were proposed as reference risk values.

This index was one of the most used during the seventies for all the analysis concerning actinides burning.

For the above reasons and because it is calculated directly by ORIGEN and ORIGEN2 codes /8/, which have been used to carry out the analysis, this index have been adopted for sensibility analysis in order to estimate qualitatively the relative importance of the several independent variables of formula (8).

The hazard due to the decay of the matrix containing the Minor actinides is drawn in all the figures relative to this qualitative analysis with the purpose to be used as reference in evaluating the gain obtained by irradiating the matrix.

8- INFLUENCE OF THE MATRIX ON THE ENVIRONMENTAL IMPACT

To analyze the dependence of the environmental impact of the matrix containing the minor **actinides**, four matrices were considered: ZrO_2 , CW_2 , UO_2 and ThO_2 .

Calculation were performed considering a PWR standard fuel element. in the ThO_2 and inert matrix cases, the fuel (UO_2) volume was replaced by the same volume of ZrO_2 , CW_2 or ThO_2 .

The actinide concentration is 10 times that of PWR spent fuel and the irradiation time 1,100 days.

The flux values to be introduced in both ORIGEN and ORIGEN2 codes were modified to take into account the different absorption properties of matrices.

The compositions of the considered matrices are listed on table III.

The minimum amount of unburned actinides is obtained by using a ZrO_2 matrix: about one fifth of that for UO_2 matrix.

Fig. 1 shows actinide and fission product hazard versus decay time: for geological times (more of 5,000 years) the fission product risk is negligible if compared with the actinide one.

The most efficient behavior, from burning point of view, corresponds to the inert matrices $(ZrO_2 \text{ and } CW_2)$, 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 such matrix the ratio between the reactor discharge hazards is about 1.5 considering and excluding ²³⁷ Np and it is almost unchanged after 5,000 years. This means that, if the risk index is the total mass of actinides, the absence of ²³⁷Np reduces the environmental impact in an important manner but, if the index is the hazard ratio, the reduction becomes less important.

The ZrO₂ matrix was selected for the next calculations.

9 - ENVIRONMENTAL RISK VERSUS IRRADIATION TIME

In evaluating the irradiation time influence on the environmental risk, the actinide were concentrated 100 times with respect to the amount existing in PWR spent fuel and dispersed in a ZrO_2 matrix. The considered irradiation times are: 0.0 - 1,100 - 3,300 - 9,900 - 11,000 and 22,000 days; the neutron flux is 2.81x10¹³ n/(cm² sec).

Calculations with ORIGEN2 are the most conservatives ones; they always lead to a greater amount of unburned actinides: the ratio between the total amount of actinides for 11,000 irradiation days and 22,000 days is 67.85 for ORIGEN and 64.31 for ORIGEN2. In the calculation of reference /9/ the ratio between the total amount of actinides without recycling and with recycling is 17, this demonstrates the higher efficiency of the ZrO_2 matrix.

Fig. 2 displays the behavior of the actinide hazard versus decay time for six irradiation periods plus the curve corresponding to 0.0 irradiation days (natural decay). From this picture it is possible to state that for irradiation time below 9,900 days it is more convenient to bury actinides (not irradiating) because the

hazard level obtained by irradiating for geological times is still higher than the not irradiated one.

The agreement between ORIGEN and ORIGEN2 became worse with the increase of the irradiation time: the relative difference between the total mass of actinides increase from 5.14% to 29.92%.

As ORIGEN2 results to be the most conservative code, it was selected to perform the next calculations.

The differences in the total mass of actinides or in the risk ratio between the calculations performed including or excluding ²³⁷Np are significantly decreased by using longer irradiation times.

Up to 9,900 irradiation days, the burial of the minor actinides results the most convenient solution for final waste disposal.

As irradiating for the whole reactor life the hazard can be reduced only of two order of magnitude, the irradiation time parameter does not solve the actinide burning problem.

10 - ENVIRONMENTAL RISK VERSUS NEUTRON FLUX LEVEL

in evaluating the environmental risk versus neutron flux, actinides were concentrated 100 times with respect to the amount in a PWR spent fuel and dispersed in a ZrO_2 matrix, the irradiation time is 11,000 days and the flux levels were chosen in the LWR range: from 2.5x10¹³ to 8.43x10¹³ n/(cm² see).

In fig. 3 the radioactive ingestion hazards for actinides and fission products versus decay time are plotted for several neutron fluxes.

Considering that there are in literature some new actinide burning systems operating with thermal fluxes higher than LWRS's, such as the Siemens idea /10/ and Los Alamos Spallation System /5/, it was found interesting to study higher thermal fluxes in the range from 1 $.0x10^{14}$ to $5.0x10^{15}$ n/(cm² see).

Table IV presents the behavior of the concentration of the most important long-lived fission products as a function of the neutron flux:

⁹⁹Tc 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 concentration decreases sharply when the flux increases from 2.81 xl 013 to 5.0x1014 n/(cm² see), then it starts to increase slowly;

¹³⁵Cs concentration has a behavior similar to 1291, but less marked.

By increasing the flux level ¹²⁹ becomes the main component of long term risk.

Table V shows the influence of the flux level and ²³⁷ Np on long-lived fission product concentrations: the behavior of the three nuclides is quite similar to that described for table IV, but ⁹⁹ Tc, ¹²⁹ I and ¹³⁵Cs concentrations are respectively 5, 4.5 and 4 times smaller than the corresponding ones considering the ²³⁷ Np.

In fig. 4 the values of **actinide** hazard are plotted versus decay time for flux levels higher than the LWR ones.

Table VI shows that the ratio between the **actinide** hazard at the end of irradiation in the transmutation system with the initial one has a dramatic drop for flux levels greater than 1.Oxl 0^{14} n/(cm² see).

In fig. 5 the Total Radioactive Ingestion hazard (actinides + fission products + activation products) is drawn versus decay time: the hazard increase at the highest flux $(5.0x10^{15} n/(cm^{2} see))$ is due to the matrix activation contribution.

With the present LWR operating flux levels the actinide burning problem can not be solved.

The flux results one of the most critical parameters in determining the actinide burning.

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

The fission product hazard increases with flux up to $1.0x1 \ 0^{15}$ n/(cm² see), while from $5.0x10^{15}$ n/(cm² see) it begins to decrease. That demonstrates that long lived fission product burning is possible.

This is a very important achievement because, for solving the problem of long lived radioactive waste transmutation, it is necessary to burn long lived fission products too.

The environmental impact could be worse by burning **actinides** and not fission products /1 1/ because same long-lived fission products, such as ⁹⁹ Tc, ¹²⁹ I and ¹³⁵Cs, have infinite volubility and high mobility in water.

11 - ABSENCE OF ²³⁷Np

In the calculations performed at high flux, long irradiation time and excluding ²³⁷Np, the **actinide** hazard is reduced to one half, while the concentration of long-lived fission products ⁹⁹Tc, ¹²⁹I and ¹³⁵Cs decrease by a factor ranging from 4 to 5 (see fig. 6).

These reductions does not change the problem dimensions; this means that, as the absence of ²³⁷Np is an important but not decisive parameter, it should be more convenient to burn ²³⁷Np together the other minor actinides.

12 - INFLUENCE OF THE DECONTAMINATION FACTORS ON THE ENVIRONMENTAL IMPACT

For our purpose, decontamination factors have the same mean as the percent of uranium and plutonium removed in reprocessing phase.

Three values of decontamination factors were considered: 99% (corresponding to an industrial reprocessing plant), 99.9% and 99.99% (reached in research laboratory) with two values of thermal neutron flux: 5.0 X 10¹⁴ n/(cm² see) and 5.0 X 10¹⁵ n/(cm² see). The U and Pu isotopic compositions are derived from the values of Table I by taking into account the decontamination factors and that the **actinide** concentrations is 100 times that of PWR spent fuel.

The selected irradiation time is 11,000 days and ZrO_2 is the containing matrix.

Figures 7 and 8 show the behavior of the **actinide** hazard with the decay time.

By examining the above mentioned figures we can say:

for low flux (fig. 7) the decontamination factor is a very important parameter in determining even if the irradiation is useful: for realistic values (99%) the hazard after 11,000 irradiation days results not enought lower than the one relative to the actinide natural decay;

- for high flux level (fig. 8) the influence of the decontamination factor on the resulting hazard is weak: the curves relative to a decontamination factor of 99.99% is almost coincident with the ideal one (decontamination factor equal to 100%) while for a value of 99% the hazard rises less than one order of magnitude.

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

13 - THE TARGET

After irradiation and 500 year of natural decay, the risk relative to the structures containing the actinides must be lower than the one relative to natural uranium ore used to manufacture the fresh fuel; that is with the purpose to restore the nature. 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 a lot of old building made by the man and still existing today.

The quantitative definition of this criteria presents three problems:

the file of specific hazard data of ORIGEN2 were taken from ICRP-2 (1960)/12/ (in the successive ICRP releases the actinide radiotoxicity was increased);

- the uranium hazard increases with the time (see Fig.9);

the actinide hazard should be compared with the hazard corresponding to the mass of Uranium ore need to produce the fuel.

For the above reasons, before the calculation to simulate the actinide burning systems, the 0RIGEN2 specific hazard file was updated using the data from /3/.

13.1 - CALCULATION METHOD

The hazard due to all nuclides, but uranium and its daughters in secular equilibrium, included in the uranium ore was considered negligible. Therefore the hazard corresponding to the fresh fuel plus uranium daughters in secular equilibrium becomes the same of the natural Uranium ore one.

The hazard calculated as described before represents the new reference for thermal actinide burning systems.

Fig. 10 shows the comparison of the new criteria with the data of /13/. The agreement is good except for very long decay

time: this is due to the decreasing, in the last ICRP releases, of the hazard index of ¹²⁹ and ²²⁶Ra for fission products and actinides respectively.

14 - ORIGEN2 SIMULATION OF THE LOS ALAMOS SYSTEM

The calculations were performed considering a thermal neutron flux of 1.OXI 0¹⁶n/(cm² see) and with actinides contained in FNa matrix at the same concentration as that in PWR spent fuel.

The simulation shows that actinides are burned after an irradiation time of 25 days. Furthermore, for 100 irradiation days, the actinide and fission product hazards become lower than the hazard associated with natural uranium ore just at the end of transmutation phase (see figure 11).

At the reactor discharge after 100 irradiation days, the radioactivity of ⁹⁹Tc and ¹²⁹I are 1.1 08x1 0⁻²Ci and 5.766 x1 O-S Ci respectively.

Could be useful to remember that the radioactivities for the above mentioned two long-lived fission products are 1.309X1O+ICi and 3.1 34X1 0⁻² Ci for each ton 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 operating 1,100 days with a long-lived fission product generation which is about one thousandth lower than that of a standard PWR (Fig. 12).

14.1 - THE DECONTAMINATION FACTOR IMPORTANCE

Figs. 13 and 14 show the influence of the decontamination factor on the Los Alamos System performances. From fig. 13 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 one, while this ratio drop to 11/1 for a decontamination factor of 99.9%. In addition, for a decontamination factor of 99.9%, calculations show that to reach the target, an irradiation period of 500 days is necessary; therefore the ratio between burning and production rates is 3/1.

Fig. 14 shows the radioactivity values corresponding to ⁹⁹Tc and ¹²⁹I produced by Minor actinides burning. The ratio between the ⁹⁹Tc radioactivity in PWR spent fuel and these values changes from 34.72 up to 1,181 for a decontamination factor of 99% and 100% respectively while, for ¹²⁹I, the change of the above defined ratio, with the same decontamination factors, is from 19.53 to 544.

15 - ANALYSIS OF SIEMENS IDEA

The SIEMENS idea /10/ consists in coating the actinides in a PWR cladding inner surface .

Calculations were performed considering a Zircalloy2 matrix, actinides at the same concentration in PWR spent fuel and thermal neutron flux of $2.5 \times 1014 \text{ n/(cm}^2 \text{sec})$.

The result is that actinides are burned after an irradiation time of 1,100 days (see fig.15). For this irradiation interval 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 just at the production rate.

16 - CONCLUSIONS

The existing LWRS are not able to solve effectively the actinide burning problem.

As Siemens idea could, even with an ideal decontamination factor of 100%, burn actinides only at the production rate, it can not be presented as definitive solution but, it could be used in the framework of an HLW radiotoxicity reduction program.

From the ratio transmutation-production rate and long-lived fission product production point of view, Los Alamos system is, theoretically, the most promising one. It should be noted that the decontamination factor, in reprocessing and partitioning phases, is one of the most important technological aspect affecting the system performances. In fact, the above defined ratio drop from 44/1 to 3/1 when the decontamination factor decreases from 99.99% to 99% which represents a more reasonable but still optimistic estimate of the value reached in an industrial reprocessing plant.

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| Isotope | Mass (Gram - Atoms) | Mass (Grams) |
|---------|------------------------|-----------------|
| U-234 | 7.102E-1 | 1.662E+2 |
| U-235 | 3.591E+1 | 8.438E+3 |
| U-236 | 1.718E+1 | 4.059E+3 |
| U-238 | 3.965E+3 | 9.437E+5 |
| Pu-238 | 5.738E-1 | 1.366E+2 |
| Pu-239 | 2.098E+1 | 5.015E+3 |
| Pu-240 | 9.532 | 2.288E+3 |
| Pu-241 | 4.842 - | 1.167E+3 |
| Pu-242 | 1.784 | 4.316E+2 |

Table I-- Standard PWR spent fuelUranium and Plutonium isotopic composition

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Table IJ-Minor actinides composition concentrated 100 times

| PWR sp | ent fuel. | |
|----------|---------------------|----------|
| Nucli de | mass (gram-atom) | mass |
| Np-237 | 2.040E+2 | 4.835E+4 |
| Am-241 | 1.920E+1 | 4.627E+3 |
| Am-242m | 3.840E-1 | 9.293E+1 |
| Am-243 | 3.890E+1 | 9.453E+3 |
| Cm-242 | 2.120 | 5.130E+2 |
| Cm-243 | 3.I10E-2 | 7.557 |
| Cm-244 | 1.100E+1 | 2.684E+3 |
| Cm-245 | 7.890E-1 | 1.933E+2 |
| Cm-246 | 9.000E-2 | 2.214E+1 |
| Cm-247 | 1.160E-3 | 2.865E-1 |
| Cm-248 | 7.790E-5 | 1.932E-2 |

| Matri x | densi ty (g/cm) | nucl i de | mass (atom-gram) | mass (g) |
|------------------|----------------------------|--|--|--|
| | 10.5 | U-237 U-235 U-238 | 1. 130 1.404E+2 4. 034E+3 | 2.644E+2 3.299E+4 9.601E+5 |
| Zr0 ₂ | 5. 89 | C 238 C Al Si -28 Si -29 Ti -46 Ti -47 Ti -48 Ti -49 Ti -50 Cr-50 Cr-50 Cr-52 Cr-53 Cr-54 Mn-55 Fe-54 Fe-56 Fe-57 Fe-58 co-59 Ni -58 | 4. 034E+3 1. 5 4. 0 0. 607 0. 034 0. 304 0. 277 2. 771 0. 204 0. 200 5. 04 57. 42 6. 415 1. 574 0. 327 4. 037 61. 018 1. 439 0. 31 0. 915 111. 86 | 9.601E+5 18 108 16.99 0.986 13.98 13.01 133.0 9.99 10.0 252.0 2986.84 339.99 84.99 17.99 217.99 3417.0 82.02 17.98 53.98 6487.88 |
| | | Ni -58 Ni -60 Ni -61 Ni -62 Ni -64 Zr-90 Zr-91 Zr-92 Zr-94 Zr-96 Nb-93 Mo-92 Mo-92 Mo-94 Mo-95 Mo-96 Mo-97 | 111. 86 41. 783 1. 869 5. 645 1. 609 4513. 54 999. 15 1502. 3 1483. 34 232. 53 10. 258 0. 957 0. 532 0. 926 0. 958 0. 546 | 6487.88 2506.98 114.0 349.99 102.98 406218.60 90922.65 138211.60 139462.16 22322.88 953.99 88.04 50.00 87.97 91.97 52.96 |

Table III- Matrix compositions

| Matri x | densi ţ y (g/cm) | nuclide | mass (atom-gram) | mass (g) |
|------------------|----------------------------|------------|---------------------|-------------|
| Zr0 | 5.89 | Mo-98 | 1.357 | 132.99 |
| ۷ | | Mo-100 | 0.54 | 54.00 |
| | | Sn-112 | 0. 321 | 35.95 |
| | | Sn-114 | 0. 219 | 24.97 |
| | | Sn-115 | 0. 113 | 12.99 |
| | | Sn-116 | 4. 681 | 542.99 |
| | | Sn-117 | 2.47 | 288.99 |
| | | Sn-119 | 2.739 | 325.94 |
| | | Sri - I 20 | 10. 3 | 1236.0 |
| | | Sn-122 | 1.467 | 178.97 |
| | | Sn-124 | 1.823 | 226.05 |
| CW2 | 17.15 | C | 4989. 73 | 59876.76 |
| 2 | | W-180 | 1171.33 | 210839.40 |
| | | W-182 | 2343.64 | 426542.48 |
| | | W-183 | 1267.34 | 231923.22 |
| | | W-184 | 2703.36 | 497418.24 |
| | | W-186 | 2493.79 | 463844.94 |
| Th0 ₂ | 9.86 | Th-232 | 4019. 13 | 932438.16 |
| FNa | 2.558 | F-19 | 5569.02 | 105811.29 |
| | | Na-23 | 5569.02 | 128087.33 |

Table 111- Matrix compositions (continuation)

| Table İV | : Neutronic fl concentrati matrix, ORIG | ux influence on (g) at ¹ EN 2 calcula | e on long li 11000 days o tions | ved fission f irradiatic | products on in Zr02 |
|-----------------|---|--|--|------------------------------------|----------------------------------|
| l sotope | flux 2.81E+13 n/cm2s | flux 8.43E+13 n/cm2s | flux 5.0E+14 n/cm2s | flUx 1.0E+15 n/cm2s | flux 5.0E+15 n/cm2s |
| Tc-99 | 3.987E+2 | 5.460E+1 | 5.062E+1 | 3.218E+1 | 4. 271 |
| 1-129 | 1.674E+2 | 2.068E+1 | 2970 | 1.852 | 8.446 |
| Cs-135 | 1.006E+3 | 6. 154E+2 | 9.208E+1 | 6.629E+1 | 8.496E+1 |

Table V : Neutronic flux influence on long lived fission products concentration (g) at 11000 days of irradiation in ZrO2 matrix without Np-237, ORIGEN 2 calculations

| lsotope | flux 2.81E+13 n/cm2s | flux 8.43E+13 n/cm2s | fl ux 5.0E+14 n/cm2s | flux 1.0E+1 5 n/cm2s | flux 5.0E+15 n/cm2s |
|---------|----------------------------|-----------------------------------|----------------------------|-----------------------------------|---------------------------|
| Te-99 | 8.015E+1 | 1.024E+1 | 9.947 | 8. 137 | 8.913 E-1 |
| 1-129 | 2. 478E+1 | 3. 106 | 4.173 E-1 | 3.461 E-1 | 2.304 |
| Cs-135 | 1.919E+2 | 1. 132E+2 | 1.753E+1 | 1.590E+1 | 2.333E+1 |

Table VI - Radiotoxicity ratio between the values at discharge and at charge, Actinides concentrated 100 times PWR spent fuel at several thermal fluxes in ZrO₂ matrix, ORIGEN2 calculations

| Flux | Ratio at |
|------------|-------------|
| (n/cm2/s) | di scharge |
| 2.81F+13 | 0 089 |
| 4.50E+13 | 0. 01819 |
| 5.62E+13 | 0. 00688 |
| 8.43E+13 | 0. 00208 |
| 1.0E+14 | 0.00196 |
| 2.5E+14 | 9.40348E-5 |
| 5.0E+14 | 6.25186E-7 |
| 1.0E+1 5 | 1.37448E-7 |
| 5 .0E+1 5 | 7.48633E-10 |
| | |





Fig. 3. Radioactive Ingestion Hazard m**3 water at RCG, ZrO2 matrix 100 PWR spent fuel, 11,000 irradiation days



24.2.\$2 FIGB03



24.2.92 FIGB04

Fig. 5. Total Radioactive Ingestion Hazard m**3 water at RCG Zr02 matrix, 1 00 PWR spent fuel, 1 1,000 irradiation days



24.2.92 FIGB05



^{₩54}





Fig^{*} 9. Radioactive Ingestion Hazard m**3 water at RCG for typical fresh fuels











24.2.52 FIGB13

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