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## Transmutation of Long-Lived Minor Actinides and Fission Products by Accelerated Protons

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## KERNFORSCHUNGSZENTRUM KARLSRUHE

Institut für Neutronenphysik und Reaktortechnik Projekt Nukleare Sicherheitsforschung

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Transmutation of Long-Lived Minor Actinides and Fission Products by Accelerated Protons

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## Abstract

In this report a methodology is developed to describe theoretically the transmutation of minor actinides (MA) with the help of an accelerator driven subcritical fuel region with high energetic protons of 1.6 GeV and a current of about 100 mA. This methodology is applied to a PHOENIX-type arrangement of fuel containing minor actinides as Am, Np and also long-lived fission products as <sup>129</sup>I and <sup>99</sup>Tc. It is shown that the MA are burnt effectively in such an arrangement. A balance of the needed energy is made including the energy, given to the net.

## Transmutation von langlebigen minoren Aktiniden und von Spaltprodukten durch beschleunigte Protonen

## Kurzfassung

In diesem Bericht wird eine Methodologie entwickelt, die es gestattet, die Transmutation von minoren Aktiniden (MA) wie Am, Np, Cm und einige langlebige Spaltprodukte wie <sup>129</sup>I und <sup>99</sup>Tc mit Hilfe von hochenergetischen Protonen von 1,6 GeV Energie und einer Stromstärke von ungefähr 100 mA effektiv durchzuführen. Angewendet wird die Methodologie auf eine unterkritische Anordnung, die minore Aktiniden enthält, nach Art der PHOENIX-Anordnung im BNL. Eine Energiebilanz wird vorgenommen, die die Energie, die ans Netz gegeben werden kann, enthält. Der Abfall von 45 bis 70 Leichtwasserreaktoren kann mit verschiedenen Anordnungen auf diese Weise vernichtet werden.

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## I. A METHODOLOGY FOR THE NEUTRONIC ANALYSIS OF FISSION CORES DRIVEN BY ACCELERATED PROTONS

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## Abstract

The methodology is based on three key codes, namely HERMES, the KFA adaptation of HETC, MCNP (3), and KORIGEN, the KfK adaptation of ORIGEN. Two main links between these codes were established, namely a neutrondistribution source for energies below 20 MeV, derived from the HERMES execution and fed into the MCNP run, and effective one group microscopic cross sections and target integrated yields, derived from the combined HERMES-MCNP execution and used to update the KORIGEN libraries prior to a KORIGEN time step run. Data from the KfK NUKLIDKARTE, supplemented with data from the TABLE OF ISOTOPES, were used to expand the reactor-oriented KORIGEN libraries to include the decay properties of some 2500 ground and meta-stable levels.

The use of MCNP tallies to derive the effective 1G cross sections limits the number of elements in the MCNP input to about 20 - 25. This number is generally adequate, except as concerns the absorption effect of fission product accumulation. An approximation procedure for this effect was developed. The use of MCNP tallies is also time consuming, therefore a burnup study may be conducted with KORIGEN libraries update only at BOC, neglecting the variation of the effective 1G cross section with burnup, often of a very small magnitude.

Besides the KORIGEN libraries update, the combined HERMES-MCNP run generates a number of integral results of interest, such as the core criticality, the power, the flux, a flux spectrum, a list of 1G cross sections, isotopic weights in the core. The methodology has been partially applied in calculating the 'thick target' benchmark for the outcome of proton bombardment of Pb and W targets. It has been applied in full in the analysis of proton-driven fission cores for the transmutation of americium, neptunium, tecnitium 99 and iodine 129.

## **1. INTRODUCTION AND BACKGROUND**

Participation in the 'thick target' benchmark for the outcome of proton bombardment of targets has brought a realization that there is a need for methodology development. In order to come up with a complete benchmark result, as requested, four codes had to be invoked, namely HERMES, MCNP, NJOY-JOYFOR, and KORIGEN. The KORIGEN data base was found lacking for the purpose at hand, and the KfK NUKLIDKARTE, as well as the TABLE OF ISOTOPES, had to be used in order to expand it to 2500 ground and meta-stable levels. It all amounted to a complex operation, a good deal of the time consumption being due to the fact that the elements brought into the computations were not integrated in a single capacity.

For burnup analysis of targets, a non integrated capacity will pose even greater management loads. A statement of methodology is also advantageous from the point of view of accuracy. The texts available on proton-driven transmutation are often not specific about the methods employed, leaving open such questions as appropriateness, approximative measures, and consistency.

The codes available for the calculation of the proton interaction with matter frequently are adaptations of the HETC code. In it the production and transport of particles is calculated, based on the so-called Bertini tape, and on models of direct reactions, evaporation, and fission. The treatment of neutron interactions, as of other particles, is based on simple cross sections models. Below 20 MeV of neutron energy it is advantagous to utilize the extensive evaluated files capacities for neutron interactions, compiled throughout years to suit the needs of the reactor analysts. Here the choice is between deterministic codes such as 2DB or TWODANT, and monte-carlo codes such as MORSE or MCNP. One outcome of the calculation is a neutron spectrum, encompassing an energy range from the proton-beam few hundreds of MeV to zero. This is to be input to ensuing burnup calculations either in the form of group fluxes coupled with a fixed library of group cross sections, as e.g. in the 2DB code, or in the form of an integrated flux coupled with single-group, spectrum averaged, cross sections, as e.g. in the KORIGEN code.

The HERMES code{1}, a KFA version of HETC, has been in use for some time in KfK, succesfully utilized to obtain results required in the 'thin-target' and 'thick-target' benchmarks{2}. This KFA adaptation enables the nuclear engineer ready access to various neutron tallies as well as to a selected list of neutron and nuclei

events. The addition of some capacities is described in the text below. The neutron distribution in space and in energy below 20 MeV is tailored as input to the MCNP3 code{3}. This latter code has been in use for many years in KfK, mainly in fusion blanket research. The geometry description for the MCNP is the socalled 'combinatorial', much the same as in the HERMES code; the energy description is continous. The code is most suitable for hard neutron spectra, a lack of self shielding in the range of unresolved actinide resonances making its performance for thermal systems somewhat inferior. An HERMES-MCNP execution is used to obtain integral parameters for the target, as well as data needed for ensuing burnup calculations. These data are in the form of a nuclei yield distribution and of one-group effective microscopic cross sections, used subsequently to update KORIGEN libraries. The latter code is the KfK adaptation of ORIGEN and long in use in KfK{4}.

The HERMES-MCNP chain of calculations is described in section 2. The necessity and form of the KORIGEN libraries update is explained in section 3. Section 4 deals with some approximations and constraints. Application issues are discussed in brief in section 5.

## 2. THE HERMES-MCNP CHAIN

The input to the HERMES code consists of a combinatorial-geometry description of the core-target, beam parameters, and a nuclei list of densities for the target. The capacity of HERMES input was expanded to enable a mixture list of up to 99 nuclei, with a mass range between 1 and 260. The code is run with a lower limit of 20 MeV for neutron interactions. Certain output items, either directly availabe, or derived, are collected for general design interest and for specific subsequent uses. They include

- a. the total fission rate (per beam proton)
- b. the total spallation rate (per beam proton)
- c. spallation rates per nuclide (per beam proton)
- d. the total yield of nuclei (per beam proton)
- e. the total no. of neutrons falling below 20 MeV (per beam proton)
- f. the total neutron flux (above 20 MeV, per beam proton)
- The 'submission' output file of HERMES, condensed from the HETC 'history' tape to a list of nuclei and neutron events, is used to generate
- g. a nuclide spallation yield distribution list
- h. a list of neutrons below 20 MeV, each with its phase-space coordinates, in a

format appropriate as source for MCNP.

The input to the MCNP code consists of the combinatorial-geometry description of the core-target, the nuclei list of densities for the target, and the source produced with the preceding HERMES execution. Included in the input is also a list of requested tallies for the target mixture nuclei, for fission, capture, n,2n, and n,3n reactions. The code will run to a successful conclusion provided the target, as loaded, is subcritical. Certain output items are collected for general study purposes and for specific applications subsequently. These include a. the total multiplicatin M (per source neutron) hence the criticality k =M/(M+1)

- a. the flux below 20 MeV (per source neutron)
- b. the total fission, capture, nxn, rates (per source neutron)
- c. the nuclei fission, capture, nxn, rates (per source neutron)
- d. the % neutrons, out of total absorption and leakage, leaking
- e. the effective no. of neutrons per fission for the target
- f. the neutron energy spectrum below 20 MeV
- g. a 'fission yield' list of nuclei. This is not a proper MCNP output item; it is approximated by applying the relative nuclide fission rates (item c above) to a list of fast fission yields for 12 actinides, especially compiled from a KORIGEN data base{5}.

For a specified proton current HERMES and MCNP output items are combined to yield integrated target data. They include

- a. the proton beam power, MWe
- b. target average flux, n/cm\*\*2/sec
- c. target total power, MWth
- d. target average power density, MWth/Liter

Other integrated target data, which however do not depend on the beam current, are

a. the overall average neutron energy (measure of spectrum hardness)

- b. a 'unified' fission nuclei yield list for the target
- c. a list of effective one-group microscopic cross sections for fission, capture, (n,2n), (n,3n), for the target nuclei.

The rationale, definition, and application of data items b and c above are described in the next section: they derive from the terminology and organization of the KORIGEN data bases.

#### 3. KORIGEN, AND KORIGEN LIBRARIES UPDATE.

The Bateman equation for the time dependence of a nuclide, say nuclide 0, can be written as

$$\frac{dN_{o}}{dt} = \sum_{j} N_{j} \lambda^{j} \ell^{j} \rightarrow \circ - \lambda \circ N_{o} \qquad (decays) \qquad (1)$$

$$+ \sum_{j} N_{j} \varphi \sigma_{j} t^{j} \rightarrow \circ - \sigma \circ \varphi N_{o} \qquad (neutron reactions)$$

In this equation

$$\begin{array}{lll} N &= & nuclide \ density \\ \varphi &= & flux \left( n/cm^{**}2/sec \right) \\ \sigma &= & microscopic \ absorption \ cross \ section \left( cm^{**}2/10^{**}24 \right) \\ \lambda &= & decay \ constant \ (1/sec) \\ \ell j \rightarrow 0 &= & the \ probability \ that \ nuclide \ j \ decays \ to \ nuclide \ 0 \\ t j \rightarrow 0 &= & the \ probability \ that \ neutron \ absorption \ creates \ 0 \ out \ of \ j \end{array}$$

The KORIGEN data stream into equation 1 is organized in concert with the distinction between fission reactions and other, capture, (n,2n) and (n,3n), reactions. Thus the transfer term in the neutron reactions part of the equation is split up:

$$\sum_{j} N_{j} \sigma^{j} t^{j \to o} = \sum_{j} N_{j} \sigma_{f^{j}} t^{j \to o} + N_{-1} \sigma_{c}^{-1} + N_{+1}, \sigma_{2n} + +$$
(2)

where

 $\sigma_f$  = fission cross section (1G, MCNP)  $\sigma_c$  = capture cross section (1G, MCNP)  $\sigma_{2n} = (n,2n)$  cross section (1G, MCNP)  $f_{j\to 0} = probability$  that when 'j' fissions, '0' is produced '-1' indicates the nuclide which produced '0' by capture '+1' indicates the nuclide which produces '0' by (n,2n)

The fission term, namely,

$$\phi \sum_{j} N_{j} \sigma_{\neq} t^{j \to o}$$
 (3)

becomes, in the presence of spallation reactions,

$$\phi_L \sum_j N_j \sigma_f^j t^{j \to o} + \phi_H \sum_j N_j \sigma_s^j j_s^{j \to o}$$
(4)

where

 $\sigma_s = spallation cross section (HERMES)$   $s^{j \rightarrow 0} = probability that when 'j' is spallated, '0' is produced$  $<math>\phi_H = neutron flux above 20 MEV (the 'HERMES' flux)$  $\phi_L = neutron flux below 20 MEV (the 'MCNP' flux)$ 

Define  $F_{j\rightarrow 0}$ , an 'extended' fission yield for nuclide j, by

$$F_{j\to 0} = [\phi_L \sigma_{f^j} t^{j\to 0} + \phi_H \sigma_{s^j} s^{j\to 0}] / [\phi_L \sigma_{f^j} + \phi_H \phi \sigma_{s^j}]$$
(5)

Define  $\sigma_{f^j}$  , an 'extended' fission cross section for nuclide j, by

$$\sigma_{F^{j}} = [\phi_{L} \sigma_{f^{j}} + \phi_{H} \sigma_{s^{j}}] / [\phi_{L} + \phi_{H}]$$

Define  $\varphi$  , a total neutron flux for the target, by

$$\Phi = \Phi_{\mathsf{L}} + \Phi_{\mathsf{H}} \tag{7}$$

Then the 'extended' fission term for the Bateman equation can be cast as

$$\sum_{j} N_{j} (\phi_{L} \sigma_{fj} t^{j \to o} + \phi_{H} \sigma_{sj} s^{j \to o}) = \phi \sum_{j} N_{j} \sigma_{F}^{j} F^{j \to o}$$
(8)

This latter form for the fission term already enables a utilization of KORIGEN in a scheme much as in reactor applications. However, it is useful to further introduce Y<sup>o</sup>, an overall single yield for nuclide '0' for the given time step, by

$$\phi \sum_{j} N_{j} \sigma_{F} j_{F} j \rightarrow \circ = \left[ \phi \sum_{j} N_{j} \sigma_{F} j \right] Y \circ$$
(9)

As long as the flux level is assumed constant for a burnup step, also the total yield is relatively unchanged in a burnup time step, or even in a full cycle, as long as only small changes in the relative percentages of the fissioning nuclides are envisaged. Thus the RHS of the latter expression is the 'fission' term in the current KORIGEN application.

The original 'Fission Product' library of KORIGEN contains, in its neutron data field for each nuclide, a list of 12 yields for the nuclide, as produced, respectively, by a list of 12 actinides. In the current application there is only one (total) yield which, by Eq. 9, may be attributed to each of the target nuclei. The KORIGEN 'Fission Product' library is thus updated with this single yield list, and the KORIGEN program is accordingly modified to refer, for every fissionable nuclide (i.e. every target nuclide, since every nuclide undergoes spallation, and currently ' fission ' was made to include spallation) to this single yield.

In parallel to the 'extended' fission cross section definition, Eq 6., the effective one-group microscopic capture and 2n cross sections are

$$\sigma_{C} j = \frac{\phi_{L}}{\phi_{H} + \phi_{L}} \sigma_{c} j \text{ (MCNP)}; \ \sigma_{2n} j = \frac{\phi_{L}}{\phi_{H} + \phi_{L}} \sigma_{2n} j \text{ (MCNP)}$$
(10)

The original 'Actinides' library of KORIGEN contains nuclides which actually undergo fission. Currently, 'fission' including spallation, such a library should contain any nuclide which may be present in the target mix, hence an extensive list of nuclides. In the present methodology the 'Actinides' and 'Fission Product' libraries both extend over 2500 nuclei (ground and meta-stable states). The microscopic cross sections for fission ,capture, (n,2n), (n,3n), derived via Eqs. 6 and 10, are used for an 'Actinide' library update for every burnup step, or once at BOC, much as the 'Fission Product' library is.

Compared with the original KORIGEN libraries, the libraries for the present application must include many more nuclei. This, to accomodate the vast range of nuclei emerging from the spallation reactions, namely neutron knockouts, precompound reactions, evaporations and fissions, as well as a host of emissions of particles other than neutrons. The basis for this extension were the KfK NUKLIDKARTE{6} and the TABLE OF ISOTOPES {7}. Data from these two sources were used to construct the decay entries for some 2500 levels, ground and metastable. The neutron interaction data for the 'AC' library, and the nuclei yields for the 'FP' library, are enterd, as described above, in a 'KORIGEN update' step.

A few points deserve mentioning, as concerns the application of formulae above. Define

 $R_s o = spallation yield per proton of nuclide '0'$ 

R<sub>s</sub> = total spallation nuclei yield per proton

By definitions above

$$R_{s} \circ = \sum_{j} N_{j} \phi_{H} \sigma_{s} j s^{j \to o}$$

$$R_{s} = \sum_{j} N_{j} \phi_{H} \sigma_{s} j$$

$$(11)$$

Next, by equations 5, 6, 9, and 11,

$$Yo = \frac{\sum_{j} N_{j} \phi_{L} \sigma_{f^{j}} t^{j \to o} + R_{s}^{o}}{\sum_{j} N_{j} \phi_{L} \sigma_{f^{j}} + R_{s}}$$

Since both Ros and Rs are HERMES outputs, Eq 12 is immedetially applicable

without a need to extract  $\sigma_{s^j}$ . However  $\sigma_{s^j}$  needs be explicitely determined for application in Eq 6. Let

$$\Sigma_s$$
 = macroscopic spallation cross section

then

$$\Sigma_{\rm s} = R_{\rm s} / \phi_{\rm H} \tag{13}$$

and the microscopic spallation cross section is determined from

$$\sigma_{s}^{i} = \sigma_{inl}^{j} \frac{\sum s}{\sum_{j}^{N_{j}} \sigma_{j}^{j} \sigma_{inl}}$$
(14)

The  $\sigma j_{ini}$  'inelastic' cross sections in the latter expression are readily derived from ref.8.

A summarized vesion of the HERMES-MCNP-KORIGEN calculational flow is shown as Table 1.

TABLE 1. THE HERMES-MCNP-KORIGEN CALCULATIONAL CHAIN

```
1. Input
New core nuclei densities
```

\_\_\_\_\_

\_\_\_\_\_

2. Run

HERMES

obtain

total spallation nuclei yield (per source proton) nuclei yields (per source proton) neutron flux above 20 MeV (per source proton) total no. of neutrons below 20 MeV (per source proton) neutron source distribution below 20 MeV (for MCNP)

-----

3. Run

MCNP

obtain

source multiplication

neutron flux below 20 MeV (per source neutron)

fission, capture, (n,xn) rates (per source neutron)

-----

4. Combine

**HERMES and MCNP** 

obtain

core average flux (n/sec/cm\*\*2)

core power (MW)

core criticality

effective 1G fission ,capture, (n,xn) cross sections

('fission' = spallations above 20 MeV + fissions below 20 MeV)

effective unified nuclei yield distribution

(spallation yields above 20 MeV + fission products below 20 MeV)

-----

5. Update

KORIGEN LIBRARIES

the 'AC' library with the 1G cross sections

the 'FP' library with the nuclei yield distribution

-

6. Run

KORIGEN ( for a specified time period, on the core average flux) obtain

new core nuclei densities

-----

7. go to 1

\*\*\*\*\*

#### 4. APPROXIMATIONS AND CONSTRAINTS

a. It was mentioned in section 2 that the fission product yields of the core actinides, for fissions below 20 MeV, and the spallation nuclei yield above 20 MeV, were combined in a single yield distribution for the target, for a given time step. MCNP tallies provide for fission rates for the core actinides, but not for their fission product yields. The needed yield lists are drawn, or approximated, from a compilation of yield distributions for fast fissions in 12 actinides{5}, namely Th232, U233,235,236,238, Pu238,239,240,241,242, Np237, Am241. If a core nuclide is one of these 12 nuclei, then its FP list is directly read from the compilation. If a core nuclide is not in the list of 12 nuclei, then its FP list is assumed to be one of the 12 available lists, the assignments being

the Th232 FP list is assigned to nuclei with mass numbers 232 or lower the U 233 FP list is assigned to nuclei with mass numbers 233 the U 233 FP list is assigned to nuclei with mass numbers 234 the U 235 FP list is assigned to nuclei with mass numbers 235 the U 236 FP list is assigned to nuclei with mass numbers 236 the Np237 FP list is assigned to nuclei with mass numbers 237 the U 238 FP list is assigned to nuclei with mass numbers 238 the Pu239 FP list is assigned to nuclei with mass numbers 239 the Pu240 FP list is assigned to nuclei with mass numbers 240 the Pu241 FP list is assigned to nuclei with mass numbers 241 the Pu242 FP list is assigned to nuclei with mass numbers 242 or higher

b. MCNP tallies are used to generate the effective 1G microscopic cross sections for the core nuclei. As a consequence of MCNP input limitations, only 20 to 25 nuclei may have their tallies requested in a single MCNP run. Most of these tallies have primarily to be designated for 1G cross sections of the core mix nuclei, therefore it is impossible to generate effective 1G cross sections for any relevant (large) number of fission products, as would be required for an accurate assessment of their effect on the flux and the absorption rate in the core. An alternative would be to conduct a study aimed at establishing a 'lumped' single 'fission product' with a (problem, or spectrum, dependent) cross section tailored to approximate the combined absorption effect of all fission products.

Currently a more expedient 'lumping' procedure is adopted, namely a certain

nuclide is chosen to represent the total FP effect. In making, and justifying, the choice of such a representative 'lump-nuclide' a few nuclides were first identified, whose 1G cross sections for a typical large fast-spectrum reactor were close to the 1G cross section average for all FP for such a specrtum. These were Tc99, Ru100, I129, and Xe129, with 1G cross sections of, respecively, .246, .200, .144, .157 barns, compared with .160 barns for the average over all FPs. Next it was observed that for relatively large variations in the neutron spectrum the 1G cross sections of the four nuclei varied in a close proportion to each other (excluding the I129 cross section for the softest spectrum), suggesting similarities in the energy dependent cross sections. The variations are shown as Table 2.

# TABLE 2. VARIATION OF SOME EFFECTIVE FISSION PRODUCT CROSS SECTIONSWITH THE NEUTRON SPECTRUM

average neutron energy   950 keV   600 keV   370 keV   200 keV   50 Kev					
	σ/σ <sub>0</sub>	σ/σ <sub>o</sub>	σ <sub>o</sub>	σ/σ <sub>0</sub>	σ/σ <sub>o</sub>
nuclide					
Tc 99	.50	.71	.246 barns	2.3	12
Ru 99	.47	.68	.200 barns	2.5	14
129	.53	.73	.144 barns	2.2	5
Xe129	.54	.73	.157 barns	2.7	13

It is thus plausible that this variation of 1G cross sections with spectrum approximates the respective variation the FP average cross section, hence any of these four candidates could (with a small adjustment of density, to equalize macroscopic cross sections) serve as a 'lump-nuclide'.

One can also quantity, to some extenet, the level of inaccuracy that may result of this expedient measure. In a study, based on the current methodology, three core types were examined, with spectrum hardnesses matching those of the three last columns of Table 2 above. The total EOC reactivity effect of the FPs, as currently modelled, was 2.0%, 3.2%, and 12%, respectively from the hardest to the softest spectrum. Even if one assumes an error of 30% in the estimated reactivity reduction due to FP presence, the errors by the current FP modelling are tolerable for relatively hard spectra. This may not be the case for very soft spectra.

c. The use of tallies in MCNP for the nuclei reaction rates also limits the efficiency of the methodology. It is found that the computing time per neutron history is up to an order of magnitude longer, compared with an MCNP execution lacking these tallies request. This may become a real-time difficulty when a parametric survey is needed, calling for a large number of MCNP executions. The difficulty may be circumvented if one assumes that during a cycle of a given core the spectrum change is small so that the effective 1G cross sections of BOC approximately hold for the duration of the cycle. Then the KORIGEN 'AC' and 'FP' libraries are generated only once, namely at BOC, and the MCNP executions for time steps subsequent to BOC become more efficient, since they aim at power and flux updates only. The ansatz of negligible changes in 1G cross sections during burnup can, of course, be tested at EOC.

## **5. APPLICATION**

a. PROSDOR (PROton-Spallation Driven cORes) is a program written for a semiautomated execution of the HERMES-MCNP-KORIGEN calculational chain. This driver program utilizes two permanent files, namely

ORCOMLIB (Origen COMprehansive LIBrary) - a KORIGEN formated library containing the decay data for all KfK NUKLIDKARTE entries from H1 to Es256; YIELDLIB - a list of fission yields for 12 actinides, and contains the

pieces of programming which generate the neutron source for the MCNP run and the updates for the KORIGEN libraries. It generates also a printed output containing integral entities of interest, such as target flux, power, spectrum. An example of this printed output is given as Table 3. It also generates three temporary files for internal usage, which may however be of use otherwise. These are

NSORS - a neutron by neutron list of phase space coordinates for spallation neurons of energies below 20 MeV.

PRODC - a detailed list of the yields of nuclei emanating from the spallation reactions, each nuclide identified by Z and A.

YLDLIST - a list detailing the combined, spalltion + fission, yield distribution for the core.

The KORIGEN execution is not automated into the driver program : it is felt that there is a considerable amount of judgment to be exercized by the researcher inbetween burnup steps. Currently an auxilliary program extracts nuclei densities from a KORIGEN calculation and arranges them in the required HERMES and MCNP formats for the subsequent time point calculation. b. The methodology has been applied, in part, in computing the 'thick target' of the recent international benchmark problem {2}. In this benchmark problem the target was a cylinder of either lead, or tungsten, entered at center base with 800 MeV protons. The methodology was invoked to calculate (i) the net neutron yield, and its spectrum, (ii) the neutron leakage spectrum, (iii) the distributions over Z and A of the spallation yield nuclei, at time zero, and also after decay periods of a year and one hundred years.

c. The methodology, embodied in the PROSDOR driver, was instrumental in a preliminary study of possible incineration concepts for the minor actinides, and the tecnitium 99 and iodine 129 of LWR waste. A companion article {9} describes three proton-core machines which seem to have the capacity of performing these incinerations.

d. Mentioned above was the efficiency limitation imposed by the use of MCNP tallies in generating effecive 1G cross sections. In the reaserch leading to the above waste burner concepts it was found indeed useful to use two PROSDOR drivers: one at BOC, with a tallies request, and the other for all subsequent burnup steps, omitting the tallies request and using the BOC KORIGEN libraries throughout the cycle. A KORIGEN library update at EOC was used to check back on KORIGEN data cycle invariabilty.

## TABLE 3. EXAMPLE OF PRINTED OUTPUT FOR A PROTON-DRIVEN FISSION CORE

Target input		
target radius	s (cm) =	73.0
target heigh	t (cm) =	146.0
target volum	ne (m**3) =	2.44
proton curre	nt (milliamps)  =	= 102.0
protons ener	rgy (Mev) =	= 1600.0
watts*sec pe	er fission $= 3.0$	)/10**11
element	number density	total tons in target
1 60120	0.1298e+00	6.32160
2 80160	0.2364e-02	0.15351
3 110230	0.4933e-02	0.46048
4 260560	0.1076e-01	2.44552
5 430990	0.1248e-05	0.00050
6 441000	0.7214e-03	0.29278
7 441010	0.6926e-05	0.00284
8 531290	0.3301e-02	1.72825
9 541290	0.3943e-03	0.20644
10 541300	0.7493e-04	0.03953
11 942380	0.1740e-04	0.01681
12 942390	0.5395e-03	0.52331
13 942400	0.3143e-03	0.30614
14 942410	0.1216e-03	0.11894
15 942420	0.6679e-04	0.06560
16 952410	0.7152e-05	0.00700
17 952420	0.6011e-07	0.00006
18 952430	0.2783e-05	0.00274
19 962440	0.2299e-06	0.00023
20 962450	0.4074e-08	0.00000

extractions from the hermes run	
total nuclei yield per proton	= 7.34
transmutation events per proton	= 7.03
total flux (cm <sup>-1</sup> ) per proton	= 0.1155e + 03
neutrons below 20 mev per proton	= 17.3

Extractions from the I	MCNP	run
------------------------	------	-----

•

multiplication	=	11.04
k-effective	=	0.917
total flux (cm) per neutron	=	0.1326e+04
fission rate per neutron	=	0.5240e+01
capture rate per neutron		0.9893e+01
(n,xn) rate per neutron	Ξ	0.1692e-01
percent neutron leakage	=	7.06
No. of neutrons per fission	=	2.91

Target	entities
--------	----------

beam current	= 102.0 milliamps
beam power	= 163.2 MW
target power	= 1733 MW
target power density	= 0.709 MW/Liter
target flux	$= 0.599e + 16 n/s/cm^2$
flux-averaged neutron energy	= 0.035 MeV
flux fractions below 20 Mev	

0.1 ev -	1 ev	0.00007
1 ev -	10 ev	0.00706
10 ev -	100 ev	0.04339
100 ev -	1 kev	0.12764
1 kev -	10 kev	0.17869
10 kev -	100 kev	0.23296
100 kev -	1 mev	0.27751
1 mev -	10 mev	0.13191
10 mev -	20 mev	0.00077

Korigen library update

'ac' library is updated

'fp' library is updated

#### 6. SUMMARY

A methodology was put together for a consistenet burnup analysis of subcritical cores, driven by accelerated protons. Interactions above 20 MeV of neutron energy are handeled by the HERMES (KFA version of HETC) code; neutron interactions below 20 MeV are calculated with the MCNP code. The outcome of the combined HERMES/MCNP execution is transformed into unified lists of nuclei yields and effective 1-group microscopic cross sections. The KORIGEN (KfK ORIGEN adaptation) 'Fission Products' and 'Actinides' libraries are updated, respectively, with these lists.

The KORIGEN code is used to advance the burnup steps. Modifications were introduced in both HERMES and KORIGEN to enable the methodology. The HERMES capacity was enlarged to cope with 100 elements in a target mix, and with mass numbers up to 260. The KORIGEN libraries were re-based with decay data for 2500 ground and meta-stable states.

The KORIGN code was adapted to the notion of a single, target and burnup dependent, yield list for nuclei emanating from either spallation or fission events.

The methodology is embodied in a semi-automated link between HERMES, MCNP, and KORIGEN. HERMES/MCNP are run as one unite, generating target integral entities, updated KORIGEN libraries, and KORIGEN input. User judgment is left for a separate execution of the KORIGEN burnup step. An auxiliary program uses the outcome of the KORIGEN burnup step to produce the next time-point HERMES/MCNP input.

This methodology was instrumental in establishing feasibility and attractivness of some subcritical core cycles capable of transmuting the long-lived Am241, Np237, Tc99,I129, to short-lived elements.

#### REFERENCES

1. P. Cloth et al.,

HERMES - A Monte Carlo Program System for Beam-Material Interaction Studies KFA Report JUL-2230, May 1988

#### 2. D. Filges et al.,

Benchmark Specifictions for Intercomparioson of Thick-Target Calculations for Transmutation Purposes Letter distributed to participants, Forschungsanlage Julich Oct 1992

#### 3. J. Briesmeister et al.,

MCNP(3B) Monte Carlo Neutron and Photon Transport Code NEA Data Bank Aug. 1989

## 4. U. Fischer, H.W. Wiese,

Verbesserte Konsistenete Berechnung des Nuklearen Inventars Abgebrannter DWR-Brennstoffe auf der Basis von Zell-Abbrand-Verfahren mit KORIGEN KfK 3014, Jan 1983

#### 5. H. W. Wiese,

Private communication KfK 1993

#### 6. H. Klewe-Nebenius,

KfK NUKLIDKARTE Source File Private communication, Dec 1992

#### 7. C. M. Lederer, V.S. Shirley,

TABLE OF ISOTOPES. 7th Edition John wiely & Sons

#### 8. D. Filges, T.W. Armstrong, P. Cloth,

Validation of Spallation Models for Particle Production Proc. Int. Collaboration on Advanced Neutron Sources ICANS -VIII, Sep 1983, p.194

## 9. M. Segev,

Transmutation of Long Lived Minor Actinides and Fission Products in Cores driven by Accelerated Protons Submitted Nucl. Sci. Eng. 1993

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## II. TRANSMUTATION OF LONG LIVED MINOR ACTINIDES AND FISSION PRODUCTS IN CORES DRIVEN BY ACCELERATED PROTONS

M. Segev, H. Küsters, S. Pelloni

#### ABSTRACT

A preliminary study shows that the Minor Actinides and the Tc99 and I129 of LWR waste may be transmuted in subcritical cores, driven by the spallation neutrons which emanate from the bombardment of these cores with 1600 MeV protons. Three core types are required. Core type I is fueled by the M.A. waste in oxide form, cooled by Na, and its neutron energy spectrum has an average of about 350 keV. Driven by a proton current of 20 mA, the core incinerates the M.A. waste of 14 1000MW LWRs. Core type II is fueled by Pu waste and is loaded with Tc99 and I129 waste. The fuel is oxide, cooling is by Na, the neutron specrtum is softened to an average energy of about 200 keV by graphite. The ratio of carbon to fuel is chosen to optimize the transmutation of the Tc. Driven by a proton beam of 130 mA, and loaded with the Pu waste of 19 1000MW LWRs, the core incinerates the Technetium waste and 60% of the lodine waste of these 19 LWRs. A fraction of the Plutonium coming out of the core II operation, and the remainer 40% of 1129, are loaded into core type III. In order to optimize the 1129 transmutation this core has a high carbon to fuel ratio, softening the neutron spectrum to an average energy of about 50keV. The core is driven by a proton current of 25 mA to burn out the 1129.

Core I necessitates an energy extraction of 100,000 MWd/T(initial HM). A full incineration in cores II and III necessitates an extraction of 125,000 MWd/T. 17% of the waste Pu is transmuted to fission products through the operation of cores II and III. The isotopics of the 83% Pu remaining is about 2/51/30/11/6 (238/139/140/141/242), as compared with 2/55/26/12/5, the isotopics of the LWR Pu waste.

All Three core types are net producers of grid electricity. All three core types remain subcritical with the loss of Na.

#### Kurzfassung

Eine vorläufige Studie zeigt, daß minore Aktiniden (MA) und <sup>99</sup>Tc wie <sup>129</sup> aus dem LWR-Abfall in unterkritischen Cores mit Spallationsneutronen transmutiert werden können. Diese Neutronen stammen aus dem Beschuß dieser Cores mit 1600 MeV Protonen. Drei verschiedene Core-Varianten werden untersucht: Core I ist beladen mit MA in Oxide Form, die Kühlung wird mit Natrium gewährleistet. Die mittlere Energie des Neutronenspektrums in einer derartigen Anlage liegt bei 350 KeV. Mit einem Protonenstrom von 20 mA kann dieses Core den Abfall von 14 Eintausend MW LWRs vernichten. Das Core II enthält Plutonium und wird mit 99Tc und <sup>129</sup> Abfall beladen. Der Brennstoff ist Oxid, die Kühlung ist Na; und das Neutronenspektrum wird durch die Moderation an Graphit zu einer mittleren Neutronenenergie von ca. 200 KeV führen. Der Protonenstrahl dieser Anlage hat eine Intensität von 130 mA, und das Core wird mit Pu-Abfall aus 15 Eintausend MW LWR Reaktoren beladen. Dieses Core vernichtet den Technetium-Abfall und 60% des Jod-Abfalls dieser 15 Reaktoren. Core III hat ein hohes C/Pu Verhältnis, um in dem weicheren Neutronenspektrum ( $\bar{E} \sim 50$  keV) die Vernichtung von <sup>125</sup> zu gewährleisten. Dieses 3. Core wird mit einem Protonenstrahl von 25 mA beschossen.

Core I verlangt eine Energie-Extraktion von 100.000 MWd/THM. Die völlige Vernichtung des Abfalls in Core II und Core III verlangt eine Energie von 125.000 MWd/T. 17% des in den Cores II und III vorhandenen Abfall-Plutoniums wird transmutiert zu Spaltprodukten. Das Isotopenverhältnis des verbleibenden Plutoniums (83%) ist etwa 2/51/30/11/6/ (Pu238/Pu239/Pu240/Pu241/Pu242).

Alle 3 Core-Typen sind Nettoerzeuger elektrischer Energie. Alle 3 Varianten verbleiben unterkritisch bei Natriumverlust.

#### **1. INTRODUCTION**

Accelerator- based transmutation studies usually assume transmutation to take place in a site surrounding the spallation target. In a recent proposal, however, for the transmutation of LWR waste minor actinides, the spallation target is also the transmutation core {5}. The originators of this PHOENIX concept claim that a 8-module system, loaded with 26 tonnes of M.A. and driven with a current of 102 mA-1600 MeVprotons, will transmute into Pu238 and fission products the M.A. waste of 75 1000 MWe LWRs. this claim cannot be substantiated. The PHOENIX module contains mostly light and intermediate mass materials (e.g. iron, sodium, oxygen) and was tailored for high (40%) leakage for a possible further 1129 transmutation. Consequently it will generate only 20 spallation neutrons per beam proton, not 50, as factored into the assessed performance of PHOENIX.

The idea of a transmutation core being also the spallation target is appealing, nonetheless, since the neutron spallation flux will not diffuse and diminish prior to arriving at the fission site. In pursuing a modified PHOENIX concept, aiming at a commercially meaninigful performance, the incinerator module should rather be of a relatively low leakage. This means a larger volume and, for a cylindrical core model, equal height and diameter. one can approach the performance claim of the PHOENIX originators with a 1.75 cubic meter, 24% leakage, core (compared with the original 1.15 cubic meter, 40% leakage, module).

The idea of transmuting waste elements by the waste itself is further explored as concerns the incineration of the long lived Tc99 and I129. In this case waste plutonium forms fuel, and waste Tc/I poison, in a subcritical core designed to burn out the Tc/I. The neutron spectrum is softened by the use of graphite in order to augment the effective Tc/I cross sections, opting for a C/Pu ratio such that the depletion rate (flux times cross section) is the highest. The Tc/I waste of 18 years operation of 19 LWRs is accumulated to load the 5 cubic meter core with 8.8 tonnes Tc, 2.1 tonnes I, and 5.7 tonnes Pu (the latter figure being the yearly Pu waste output of 19 LWRs). A current of 130 mA - 1600 MeV protons applied to this core will transmute the yearly Tc99 output of 19 LWRs and the yearly 1129 output of 11 LWRs. The remainder of 1129 is allowed to accumulate before another core, with a higher C/Pu ratio, isloaded and operated to incinearte the yearly I129 output of 8 LWRs.

In what follows section 2 is a brief outline of the calculational methodology used to arrive at the conclusions stated above. The role of basic neutron interaction data is discussed in

section 3. Section 4 expends on some basic data, principles, constraints, and relashionships specific for the transmutation study at hand. Section 5 details the makeup and operation of the three incinerator types. Summary and conclusions are given in section 6.

## 2. THE CALCULATIONAL METHODOLOGY

A detailed exposition of the calculational scheme is brought in a separate publication {6}. The spallation reactions and the transport of neutrons of energies above 20 MeV are calculated with the HERMES code {7}. HERMES is a KFA adaptation of HETC, user friendly by its "detector" tallies and its " submission file". The former enables ready access to neutron currents, fluxes, and spectra, and to yield distributions of the product elements of the spallations; the latter is a handy account of neutrons and nuclei emissions

Neutron transport below 20 MeV is carried out with the MCNP (3A) code {8}. Data from the HERMES submission file is formed into an input source for the MCNP calculation. The combined results from the HERMES/MCNP calculation provide such core data as criticality, per-proton power, flux, spallation rates, fission/capture/(n,xn) rates, and a yield distribution for elements. The spallation + fission rates are combined in "generalized-fission" rates (for later compliance with terminology and practice in the KORIGEN code); elemental rates combine with the flux to produce effective microscopoic one-group cross sections (also for subsequent use in KORIGEN) for fission, capture, and n,xn; HERMES spallation product yields and MCNP fission yields are combined in a single target yield distribution (again for application in KORIGEN).

KORIGEN {9}, the KfK adaptation of the ORIGEN code, is used to advance the core nuclei densities with time. Two (of three) KORIGEN libraries, namely the "fission product" and "actinides" libraries are updated, respectively, with the target yield distribution and with the one-group cross sections from a preceding HERMES/MCNP calculation. The relevance of these HERMES/MCNP derivatives to the KORIGEN concepts of "fission products" and "actinides" and "actinides" and "actinides" and "actinides" and "actinides" the KORIGEN concepts of "fission products" and "actinides" and "actinides" and "actinides" and "actinides" to the KORIGEN concepts of "fission products" and "actinides" actinides and "actinides" actinides actin

the KORIGEN concepts of "fission products" and "actinides" is clarified in section I above. The customary, reactor-oriented, KORIGEN data bases, too narrow for spallation-fission applications, were expanded to include decay data from some 2500 ground and meta-stable states. This, coupled with the updating of the KORIGEN data bases with a new yield distribution and new 1G xsections following every HERMES/MCNP step, enables one to correctly account for the effect of spectral changes on the burnup. The integral flux, needed as KORIGEN input, is too a HERMES/MCNP output item.

## **3. BASIC NEUTRONIC DATA**

The status of basic interaction data is not satisfactory. It is beyond the expertise of the authors to comment upon the data utilized in HERMES for the spallation calculation. However, it has been noted that HETC calculations, based on the so called 'Bertini-tapes' and models for evaporation and fission, are sometimes lacking in comparison with experimental data {10}. In particular the modelling of the fission- evaporation competition is far from being settled {11}. This has direct influence on the neutron yield, and yield spectrum, below 20 MEV, hence important implications on the performance of a given core.

Comparing neutron cross section data from various evaluated files often displays a vast disparity in the range of 0.1 to 10 MeV. For example, the ENDF/B-IV and ENDL Am241 capture cross section evaluations are sharply lower than the other evaluations{12}. Such discrepancies have considerable implications on the outcome of Am241 transmutation, but more so on the criticality of cores made of M.A. fuels, and on the power generated in such cores.

Currently we have based our calculations on MCNP(3A) tables derived from JEF2 evaluated files. Our attempts to generate ENDF/B-VI based data for MCNP were only partly successful, due to problems associated with the execution of the ACER module in NJOY, coupled with the fact that MCNP(3A) cannot cope with double-differential data, a feature of cosiderable frequency in ENDF/B -VI evaluated files.

Given the magnitude of uncertainty in cross sections, in particular as concerns

M.A. burners cannot be considered reliable to better than 30% to 40%.

#### 4. SOME TRANSMUTATION FUNDAMENTALS

(i) Of concern are the long lived minor actinides, mainly neptunium and americium, and the long lived fission products, mainly Tc99 and I129. It is assumed that LWR Pu waste has uses. Since the incineration of technetium and iodine implies the use, and degradation, of LWR waste Pu, one has to assume that envisaged uses for this Pu may be revitalized by blending it with 'clean' Pu, available in an assumed future, from fast breeders.

(ii) The yearly waste of a 1000 MWe LWR, disposing of fuel burned to an average 33,000 MWD/T, consists of

1125 Kg Fission Products of which : 25.1 Kg Tc99 5.9 Kg I129

300 Kg Plutonium of which : 167 Kg Pu239

35 Kg Minor Actinides of which: 98% Np+ Am

(iii) The incineratrion of either M.A. or Tc/l is based on a strategy as follows. A proton beam plus a core, together an 'incinerator', serve a given number of LWRs, a 'LWR park'. The core of the incinerator contains a large amount of the element(s) to be transmuted - the 'transmutable'. During an 'incineration period', which may be a year or a number of years, the no. of kilogrmas transmutable incinerated has to equal the no. of kilograms transmutable newly accumulated in the waste of the LWR park; at the end of an incineration period that which was incinerated is resupplied from the park to the incinerator.

(iv) A basic relation holds between the amount of energy extracted from the core, expressed e.g. as MWd/T, and the weight transmuted. Introduce the following definitions

Ρ	=	the core power in MW
n <sub>f</sub>	=	the number density (in 10**24) of the fuel
σf	Ξ	the mic. xsection, 1G effective, of the fuel (in barns)
φ	=	the flux (in 10**15 n/sec/cm**2)
V	==	the core voulme (in cubic meters)
σ <sub>a</sub>	=	the mic. xsection, 1G effective, of the transmutable (barns)
LHM		= The heavy metal loaded weight
Lt	=	The transmutable loaded weight

In deriving below a basic relationship we assume:

- a. the energy per fission is 3\*10\*\*(-11) watts\*second
- b. 239 is a good representative mass number for Pu or M.A.

Р	=	30.000	Nf	σf	ΦV
•		~ ~ . ~ ~ ~			

 $= (239/.6023) N_{f} V$ LHM

P/L<sub>HM</sub>  $= 75.6 \sigma_{f} \phi$ 

Years to reach 100000 MWd/T	$= 3.62/(\sigma_{f} \phi)$

Weight transmuted in 1 year  $= 0.03125 \sigma_{a} \phi L_{t}$ 

(1)

WEIGHT TRANSMUTED WITH 100000 MWD/T = 0.113 ( $\sigma_a/\sigma_f$ ) L<sub>t</sub>

Some specific applications of this last relationship will be made in the disscussion section.

(v) assume that the k-infinity of the core is below 1, namely that the core is subcritical even if very large, then an expression for the flux can be written on the basis of a simple one-group balance:

$$\phi = \frac{S}{N_{Tc/l} \sigma_{Tc/l} + N_{fuel} (\sigma_a - v\sigma_f)_{fuel}}$$
(2)

where

- S = neutron source
- N = number density
- $\sigma$  = mic., 1G effective, cross section

The denominator in Eq 1. is positive. In an M.A. incineratoer k- infinity is < 1, and in a Tc/l incinerator k-infinity of the Pu fuel is > 1, but the Tc/l term keep the denominator positive. Thus the core flux and the effective 1G transmutable cross section are related by a reciprocal relation. Since the transmutation rate is determined by the product of flux and cross section, neither a very high, nor a very low, cross section will do. In trying to tune the transmutation rate through spectracl shifts (using the energy dependence of the cross section), it is rather an intermediate cross section which is sought.

(vi) A few constraints are imposed by technological end economical considerations.

a. There is a limit to the exposure that the fuel cladding (or other components of the core, for that matter) can endure. It is customary to assume that future oxide fuel will withstand an exposure to an energyextraction of 100000 MWd/T. As will be shown, this limit is respected in the present concept for an M.A. incinerator (as it is in the original PHOENIX concept). Our proposal for the incineration of Tc99 and I129, however, requires an extraction of 125000 MWd/T. If such a limit cannot be attained, then the transmutation of these isotopes will fall short of 100%.

b. The flux level in the core is constrained by the necessity to stay below certain limits in the core power density. We have not attempted any heat transfer analysis since our study is of a preliminary nature, but we stay below 1 MW/Liter, a limit well acceptable for Na cooled, pin structured, lattices.

c. The economical limits are more difficult to spell out. However it is obvious that

an incinerator, consisting of a proton machine plus a fission core, is expected to serve the transmutation needs of a few dozens of 1000 MWe LWRs, if there is to be commercial interest in it. To that goal we found it necessary to assume that proton currents of the order of 10 - 200 mA can be achieved {5}.

d. Of economical benefit is also to have the incinerator generate net electricity to the grid, rather than be a net consumer of grid electricity. In tailoring our concepts to net electricity productions we have assumed that the amount of core MWth needed for the proton beam operation is 8 fold the beam MWe. This we found to be the usual overall effiency assumed by other researchers.

## **5. THE INCINERATORS**

## I. INCINERATION OF MINOR ACTINIDES

The incinerator design follows the basic PHOENIX approach, namely of a lattice of oxide M.A. as fuel, loaded into an FFTF type lattice.

BEAM - protons of 1600 MeV, 20mA.

CORE - cylinder of 65 cm radius by 130 cm height (1.72 cubic meters)

LATTICE - fuel : (MA)O<sub>2</sub> , radius = .254 cm clad : SS , radius = .292 cm coolant: Na

SPECTRUM- average neutron energy 350 keV

effective 1G fuel fission cross section 0.56 barns

effective 1G fuel capture cross section 0.90 barns OPERATION -

LWR park serviced :	14 LWRs of 1000 MWe				
initial core load :	5000 Kg minor actinides				
incineration period:	2.5 years				
energy extraction:	100000 MWd/T (initial MA)				
core criticality :	.90 (BOC)95 (EOC)				
power density :	.25 (BOC)60 (EOC) MW/Liter				

2.2 (BOC) - 5.1 (EOC) 10\*\*15 n/cm\*\*2/sec average flux . net power to grid : 200 (BOC) - 800 (EOC) MW th Na void reactivity : + 5.0% weight incinerated : 1210 Kg minor actinides minor actinides in core: BOC EOC Np237 - .413 .416 Am241 - .482 .453 Am242 -.010 .089 Am243 - .087 Cm242 -.001 Cm244 - .017 . 029 Cm245 - .001 .002 discharge materials: 1210 Kg Pu238 -.429 239 .009 -

.015

.491

242 .056 -Fission Products -

-

240

A few remarks are in order:

a. The amount of Pu239 produced via the incineration of the minor actinides is 0.2% of the LWR Pu239 waste.

b. The amout of fission products produced via the incineration of the minor actinides is 1.5% of the LWR F.P. waste.

c. The isotopic M.A. percentages in the core are quite close at BOC and at EOC to assume that within a few cycles an equilibrium operation can be established. This point has been demonstrated in the PHOENIX report {5}.

## II. INCINERATION OF TC99 AND (PART OF) I129

The incinerator design is based on the idea that the plutonium of the LWR waste can be used to burn out the Tc99 and I129. In an FFTF type lattice the effective 1G cross sections of Tc and I are too low for a commercially acceptable incineration rate, therefore the incinerator core is designed to contain also graphite, with the purpose of spectrum softening that would raise the Tc and I cross sections.

BEAM - protons of 1600 MeV, 128 mA.

CORE - cylinder of 89 cm radius by 201 cm height (5 cubic meters)

LATTICE-  $PuO_2$ ,  $Bel_2$ , and Tc pins uniformly interspaced in a graphite matrix. The  $PuO_2$  pins are cooled by annular Na channels.

Number of pins in core - 43260 (43260/3 for each pin type)

- .761 cm

pin pitch

Radii:

Pu cells: PuO2 pins .254 cm; SS clad .292 cm; Na coolant .388cm

Tc cells : Tc pins .296 cm; SS clad .341 cm;

I cells : Bel2 pins .231 cm; SS clad .266 cm;

SPECTRUM- average neutron energy 200 keV

effective 1G fuel fission cross section 1.53 barns

effective 1G Tc99 capture cross section 0.56 barns

effective 1G I129 capture cross section 0.32 barns OPERATION -

LWR park serviced : 19 1000 MWe LWRs

```
initial core load : 5720 Kg LWR waste Pu
                  8790 Kg LWR waste Tc99
                  2100 Kg LWR waste 1129
incineration period: 1.0 year
energy extraction : 125000 MWd/T (initial Pu)
                  .950 (BOC) - .800 (EOC)
core criticality :
power density : 1.06 (BOC) - .20 (EOC) MW/Liter
                  8.1 (BOC) - 1.8 (EOC) 10**15 n/cm<sup>2</sup>/sec
average flux
             :
net power to grid : +3600 (BOC) - -600 (EOC) MW th
< > power to grid :
                         +400 MW th
Na void reactivity :
                       +1.3\%
weight incinerated : 480 Kg Tc99 (Tc99 waste of 19 LWR)
                  67 Kg |129 (1129 waste of 11 LWR)
reload
```

at the end of the incineration period 480 Kg Tc99 had transmuted mostly to the stable isotope Ru100; and 67 Kg of 1129 had transmuted to the stable isotoe Xe130. The Ru, Xe, and Pu are removed from the core and the latter is supplied with 5720 Kg Pu, 480 Kg Tc99, and 67 Kg 1129, the newly accumulation of waste by, respectvely, 19, 19, 11, LWRs. The initial core load is reestablished and the next incineration period may commence. The 1129 waste of 8 LWRs remain 'unserviced'. It is allowed to accumulate for about 36 years until another incinerator, described below, can be started to burn out the '8 LWR' 1129 remainder.

#### **III. INCINERATION OF THE REMAINDER I129**

Incinerator II, above, was tuned to burning out the Tc99 waste from 19 LWRs. Since the I129 cross section was only 60% of the Tc99 cross section 8 LWR from the 19 LWR 'park' remain with their I129 waste. Below is a description of an incinerator for this 'remainder' iodine. This incinerator is loaded with 1.64 tonnes of I129, therefore its operation has to be delayed for about 36 years until 47.2 Kg of non-incinerated I129 for every 19 LWRxYEAR accumulate to provide a first loading. About 1/5 of the Pu used in the operation of incinerator II, and discarded at the end of each incineration period, is used in the incenerator III operation. No fission products are removed from this Pu prior to its loading into incinerator III. BEAM - protons of 1600 MeV, 25 mA.

CORE - cylinder of 73 cm radius by 146 cm height (2.4 cubic meters)

LATTICE- PuO2 and Bel2 pins uniformly interspaced in a graphite matrix. The PuO2 pins are cooled by annular Na channels.

```
Number of pins in core - 12390 (2:1 - 1:Pu pins)
pin pitch
                      - 1.162 cm
Radii
 Pu cells: PuO2 pins .254 cm; SS clad .292 cm; Na coolant .388cm
 I cells : Bel2 pins .381 cm; SS clad .425 cm;
SPECTRUM- average neutron energy 50 keV
    effective 1G fuel fission cross section 3.59 barns
    effective 1G I129 capture cross section 0.73 barns
OPERATION -
  LWR park serviced : 8 LWRs of 1000 MWe
  initial core load :
                      1050 Kg Pu, waste of incinerator II
                     1640 Kg LWR waste 1129
  incineration period: 1.0 year
  energy extraction : 120000 MWd/T (initial HM)
  core criticality : . 955 (BOC) to .820 (EOC)
  power density : . 40 (BOC) to .08 (EOC) MW/Liter
  average flux : 3.4 (BOC) to 0.7 (EOC) 10**15 n/cm2/sec
  net power to grid : +600 (BOC) to -140 (EOC) MW th
  < > power to grid :
                            +45 MW th
  Na void reactivity : -0.7%
  weight incinerated : 47 Kg I129 (I129 waste of 8 LWR)
```

#### Reload:

at the end of the incineration period 47 Kg I129 had transmuted to the stable isotoe Xe130. The Xe and Pu are removed from the core and the latter is supplied with 1050 Kg Pu and 47 Kg I129: the former from the incinerator II 'waste'; the latter from the 8 LWRs which were not 'I129 served' by incinerator II.

## 6. DISCUSSION

a. The proposed incinerations require relatively large amounts of the transmutables present in the core (Eq. 1), therefore the cores are rather large. We assume that, in order to avoid hot spots at proton beam entry points, as well as for flux flattening throughout the core, the proton beam can be split {4,5} to enter the core at a number of points around its surface.

b. The cores are with heights about equal to diameters, to minimize leakage and thus insure an in-core maximal usage of the flux generated. These cylindrical cores present only a first assessment of core shape and volume.

c. The lattice proposals for the cores are, in particular in the case of Tc and I incineration, only preliminary ideas. Lacking a true heat transfer analysis, S.S and Na volumes cannot be pinpointed. However the more important are the C/Pu and Pu/Tc,I ratios: this study shows that these ratios can be determined to result in viable transmutations.

d. Since a large amount of the transmutables is present at all times in the core, a meaningful incineration can be established only when the operation of the incinerators is guaranteed for many years, rendering relatively unimportant the in-core non-transmuted load.

e. The incinerators, as proposed, generate time dependent powers. For the Tc/l, in particular, the average power is only about 35% - 40% of the initial power. But if the constant proton currents needed for these incinerators can be assumed to be only fractions of the capability of envisaged beams, then by 'structuring' a time dependence for the beam current, one could establish a constant power operation of the fission cores.

f. For fission lattices, as assumed in this study, the limit of MWd/T to which the fuel is exposed is of prime importance. Eq. 1 is a rough, 1G-type, estimate of the weight that is incinerated in a 100000 MWd/T operation.

For the M.A. incinerator, and with the 1G effective cross sections of section 5.I, it predicts that 30% of M.A. load will be transmuted. The actual calculation establihes a 2.5 year period for an energy extraction of 100000 MWd/T, and a transmutation of 25% during this period.

For the Tc incinerator (5.II) each incineration period requires that the Pu waste from the LWR park destroys the Tc99 waste from that park; since the waste ratio of Tc99 to Pu is 1/12, Eq. 1 entails

MWd/T needed to incinerate the Tc99 = 0.74  $\frac{\sigma_{f}}{\sigma_{Tc}}$   $\frac{Load, Pu}{Load, T_{c}}$ 

With the 1G effective cross sections and the load figures of 5.III, the formula predicts 130000 MWd/T, the detailed analysis coming up with a close 125000. We found it impossible to get down to a 100000 MWd/T operation: if , e.g, the Tc cross section is augmented by the addition of carbon (softening the spectrum) the k-infinity of the Pu decreases, requiring a larger Pu load; these two effects outbalance each other, as can be appreciated from the above formula.

For the iodine incinerator (5.III) the weight to be incinerated is simply the amount unincinerated by the operation of incinerator II. An application of Eq 1. then results in

MWd/T needed to incinerate th I129 = 8.85  $\frac{\sigma_f}{\sigma_l} \frac{\text{Destroyed,I}}{\text{Loaded, I}}$ 

With the effective 1G cross sections and the iodine load and discharge figures of 5.III, the formula predicts 124000 MWd/T, the detailed analysis coming up as close as 120000.

g. No attempt is made presently to propose an incineration of the Pu of LWR waste. The operation of incinerator II destroys 14% of the Pu waste, the operation of incinerator III - another 3%. The Pu vector emanating from the Tc/l incineration is 2%-51%-26%-12%-5% for, respectively, Pu238-239-240-241-242. It is somewhat degraded, compared with the LWR waste percentages (2%-55%-26%-12%-5%). These 83% remaining Pu waste perhaps may be further used in incinerators to transmute also some 'limited hazard' {4} isotopes like Zr93 and Pd107. Otherwise, by blending this plutonium with a possible 'clean' Pu from fast breeders, a MOX operation can be reestablished.

h. A number of aspects clearly require future studies. Required are accurate measurments of neutron cross sections, especially for capture, and especially of actinides, in the range from 0.1 MeV to 10 MeV. The existing disagreements

between data from different evaluations prohibit a determination of core power to better than 40%. Required are analyses of flux distribution in the core, to design flux flattening where necessary, and of heat transfer, to establish in detail the structure of the incinerator lattices. The envinronment of the envisaged intense, hard spectra fluxes will result in considerable material damage: this too is a concern for study.

## REFERENCES

#### 1. T.Takahashi, H. Rief

Concepts of Accelerator-Based Transmutation Systems PSI-Proceedings 92-02, PSI, Switzerland, March 24, 1992

## 2. T. Takizuma et al.

Conceptual Design of Transmutation Plant Workshop on Nuclear Transmutation of Long-Lived Nuclear-Power Radiowastes (IAEA) Obninsk, Russia, July 1, 1991.

#### 3. T. Mukaiyama et al.

Conceptul Study of Actinide Burner Reactors Proc. Reactor Physics Conf., Jacson-Hole, U.S.A., 1988

#### 4. C. D. Bowman et al.

Nuclear Energy Generation and Waste Transmutation using an Accelerator -Driven Intense Thermal Neutron Source LA -UR- 91-2601

## 5. G.J. Van Tuyle, M. Todosow, M.J. Geiger, A.L. Aronson, H. Takahashi Accelerator-Driven Sub-Critical Target Concept for Transmutation of Nuclear Wastes. BNL-46365 DEC 1991.

#### 6. M.Segev

A Neutronic Methodology for the Analysis of Proton-Driven Fission Cores Sub. to Nucl. Sci. Eng.

## 7. P. Cloth et al.

HERMES - A Momte Carlo Program System for Beam-Materials Interaction Studies.

KFA Report Jül - 2203 May 1988

## 8. J. Briesmeister et al.

MCNP(3B) Monte Carlo Neutron and Photon Transport Code NEA Data Bank Aug. 1989

## 9. U. Fischer, H.W. Wiese,

Verbesserte Konsistenete Berechnung des Nuklearen Inventars Abgebrannter DWR-Brennstoffe auf der Basis von Zell-Abbrand-Verfahren mit KORIGEN KfK 3014, Jan 1983

## 10. H.Takahashi

Survey of Codes Relevnt to Design, Engineering, and Simulation of Transmutation of Actinides by Spallation BNL report

## 11. F. Atchison , P.S.I, Private Communication

## 12. Swedish nuclear data committee

Compilation of Actinides Nuclear Data KDK-35 , SKI B32/78 Stockholm, 1979

## III. PHOENIX TYPE CONCEPTS FOR THE TRANSMUTATION OF LWR WASTE MINOR ACTINIDES

M. Segev

#### ABSTRACT

A number of variations on the original PHOENIX theme were studied. The basic rationale of the PHOENIX incinerator is making oxide fuel of the LWR waste minor actinides, loading it in an FFTF-like subcritical core, then bombarding the core with a high current beam of accelerated protons to generate considerable energy through spallation and fission reactions. As originally assessed, if the machine is fed with 1600 MeV protons in a 102 mA current, then 8 core modules are driven to transmute the yearly MA waste of 75 1000 MW LWRs into Pu238 and fission products; in a 2 years cycle the energy extracted is 100000 MWd/T.

This performance cannot be substantiated in a rigourous analysis. A calculational consistent methodology, based on a combined execution of the HERMES, MCNP, and KORIGEN codes, shows, nonetheless, that changes in the original PHOENIX parameters can upgrade its performance.

1. The original PHOENIX contains 26 tons MA in 8 core modules; the 1.15 m\*\*3 module is shaped for 40% neutron leakage; with a beam of 102 mA the 8 modules are driven to 100000 MWd/T in 10.5 years, burning out the yearly MA waste of 15 LWRs; the operation must be assisted by grid electricity.

2. If the 1.15 m\*\*3 module is shaped to allow only 28% leakage, then a beam of 102 mA will drive the 8 modules to 100000 MWd/T in 3.5 years, burning out the yearly MA waste of 45 LWRs. Some net grid electricity will be generated

3. If 25 tons MA are loaded into 5 modules, each 1.72 m\*\*3 in volume and of 24% neutron leakage, then a 97 mA beam will drive the modules to 100000 MWd/T in 2.5 years, burning out the yearly MA waste of 70 LWRs. A considerable amount of net grid electricity will be generated.

4. If the lattice is made of metal fuel, and 26 tons MA are loaded into 32 small modules, 0.17 m\*\*3 each, then a 102 mA beam will drive the modules to 100000 MWd/T in 2 years, burning out the yearly MA waste of 72 LWRs. A considerable amount of net grid electricity will be produced. A further utilization of the 44% neutron leakage can be contemplated.

## Kurzfassung

Einige Variationen der Original-PHOENIX Anlage wurden untersucht. Die Grundannahme von PHOENIX ist, aus LWR-Abfall minore Aktiniden in Oxidform zu erstellen und diese in ein dem FFTF ähnlichen unterkritschen Core mit hochenergetischen Protonen zu beschießen, wobei eine beträchtliche Energie durch Spallation und durch Spaltreaktionen erzeugt wird. Auf diese Weise können mit 1,6 GeV Protonen mit einer Stromstärke von 102 mA etwa acht Core Module bestrahlt werden und so den jährlichen Waste-Anfall von 75 eintausend MW LWR in Pu 238 und Spaltprodukte umgewandelt werden. Die extrahierte Energie in einem 2 Jahreszyklus beträgt 100000 MWd/T, Änderungen im Original-PHOENIX-Konzept können dieses Verhalten noch verbessern:

- 1. Ursprünglich enthält PHOENIX 26 Tonnen MA in 8 Core-Moduln. Dies 1,15 m<sup>3</sup> große Modul besitzt etwa 40 % Leckage. Mit einem Protonenstrahl von 102 mA kann in 10,5 Jahren eine Energie von 100 000 MWd/t erzeugt werden. Diese Operation verlangt die Anbindung an ein Elektrizitätsnetz.
- Falls der 1,15 m<sup>3</sup> Modul 28% Leckrate zuläßt, dann kann man mit einem Protonenstrahl von 102 mA 8 Module mit einer Energie von 100 000 MWd/T in 3,5 Jahren erzeugen, wobei der Abfall von 45 LWR jährlich verbrannt wird.
- 3. Wenn 25 Tonnen minore Aktiniden in 5 Moduln untergebracht werden, wobei jeder Modul 1,72 m<sup>3</sup> Volumen besitzt und 24 % der Neutronen ausfließen, kann ein Protonenstrahl von 97 mA kann in den 5 Moduln eine Energie von 100 000 MWd/T in 2,5 Jahren erzeugen und gleichzeitig den Abfall an minoren Aktiniden aus 70 LWR vernichten.
- 4. Wenn das Gitter aus Metall statt Oxid-Brennstoff besteht, und wenn 26 Tonnen minorer Aktiniden in 32 kleine Module angebracht sind, dann wird eine Energie von 100 000 MWd/T in 2 Jahren erzeugt, wobei der jährliche Anfall von minoren Aktiniden von 72 LWR vernichtet wird. Hierbei wird ein beträchtlicher Anteil an Elektrizität erzeugt, der ans Netz gegeben wird.

#### INTRODUCTION

In recent years the idea of utilizing spallation reactions in sub- critical cores has bred a number of accelerator-cores concepts { 1,2 }. Common to all these is the assumption that future accelerators will deliver protons of about 1600 MeV, in currents ranging from a few tens to a few hundreds of mA. Such high power proton beams will be required if the accelerator-core machines are to be used as practical transmuters of LWR waste isotopes.

The entry of a high energy proton into a target gives rise to a host of spallation reactions. The total neutron output per proton depends on the target volume, mass number of target nuclei, and proton energy. It is an increasing function of all three, and may reach 50. These neutrons emerge with a hard spectrum, hence their envisaged utilizations as transmutation agents proceed via two avenues, namely directly within the hard spectrum or after thermalization. The fractional transmutation rate of a nuclide, e.g. the percentage transmuted in a year, is proportional to the effective one-group absorption cross section of the nuclide and to the average total flux. Comparing hard with thermalized spectra, the former yield higher fluxes, the latter sustain higher cross sections.

An incineration machine combines a proton accelerator with a core. In most studies the core is divided into a 'target' and a 'blanket'. The former is usually the core center, made of high mass materials, target for the protons. The latter is the site where transmutation takes place. Such 'blankets' may contain also fuel, if fuel is utilized to enhance the flux of spallation neutrons streaming out of the target. In thermal concepts a moderator region intervenes between target and blanket.

In the PHOENIX concept, addressing the hazard of the long-lived minor actinides waste, the target and blanket are one and the same. The proton beam enteres an FFTF-type core, initiating the transmutation of americium and neptunium, the fuel in the core. By fission, capture, and decay the Np and Am transform into Pu238 and fission products { 2 }.

A consistent calculational methodology for proton-driven subcritical cores { 3 } was used to recalculate the original PHOENIX. The resulting performance falls short of the performance claim of the originators. Built of mostly intermediate and low mass number materials, and shaped for high leakage, the original

PHOENIX will deliver only 1/5 (to 1/3, if basic neutron data uncertainties are considered) of the claimed power and thus will serve a smaller LWR community than envisaged. Also in discord with the originators, the PHOENIX enjoys a very small reactivity swing during a 100000 MWd/T operation.

Varying design parameters in the original PHOENIX help regain most of its envisaged performance. Reshaping the PHOENIX 1.15 m\*\*3 module for minimum (28%) leakage increases the spallation neutron yield (below 20 MeV) from 20 to 30, and the B.O.C criticality from .825 to .880, resulting in a much shortened cycle. Utilizing a larger core module of 1.72 m\*\*3 further increases the neutron yield and criticality to, respectively, 32 and .905, bringing the cycle period down to 2.5 years.

If one allows the options of metal fuel  $\{4\}$  and a proton beam splitup to many (a few tens of) small current beams, then with a core module as small as 0.17 m\*\*3 the PHOENIX cycle can be trimmed down to 2 years, serving at the same time as a neutron source (each module with 44% leakage) for further incineration applications.

TABLE 1.	PHOENI	.OXIDE	.40% LEAK/	AG (OR	IGINAL	DESIGN)	
target	:	cylindri	al, height	t/diamete	r = 48.	5cm/174cm	
lattice	:	FFTF					- •
fuel mixt	ure :	oxide M./	A. from the	e CURE pr	ocess (	'PHOENIX' fue	1)
coolant	:	sodium					
Kg. m.A.	loaded :	3330					
leakage (	(BOC) :	40.0 %					
protons e	energy :	1600 MeV					
beam curr	rent :	13 mA					
beam requ	ires :	20.8 MW(	protons)*8	= 166 MW	th targ	et power	
	B.O.C.	730 d	3800 d	5530 d			
n-spall	20.8	20.6	20.6				
k-eff	.825	.829	.833				
leakage	39.9 %	40.0 %	41.2 %				
flux	5.34+14	5.53+14	5.96+14	0.0			
MW/Liter	.072	.075	.078				
trgt MW	83.5	86.3	90.3				
grid mW	-82	-80	76				
MWd/T	0	18600	100000				
		Kg. in	target		к	g. difference 5530d - BOC	
He 4		6.353e-01	4.084e+00	4.468e+0	0		
0 16	4.442e+02	4.441e+02	4.439e+02	4.439e+0	2		
Ne 22		2.258e-04	3.606e-03	4.375e-0	3		
Na 22		7.788e-04	1.863e-03	1.093e-0	3		
Na 23	3.390e+02	3.390e+02	3.387e+02	3.387e+0	2		
Mg 24		1.027e-02	5.594e-02	5.596e-0	2		
Mn 55		1.027e-02	1.664e-01	2.025e-0	1		
Fe 55		3.687e-02	9.002e-02	5.389e-0	2		
Fe 56	1.930e+03	1.929e+03	1.925e+03	1.925e+0	3		
Fe 57		3.497e-01	1.902e+00	1.902e+0	0		
U 232		1.119e-05	1.171e-03	1.616e-0	3		
U 233		7.639e-04	3.283e-03	4.016e-0	3		
U 234		4.211e-01	1.211e+01	1.667e+0	1		
U 235		4.940e-03	3.618e-01	3.656e-0	1		
U 236		4.556e-04	1.612e-02	2.111e-0	2		
Np235		3.809e-03	4.742e-03	1.321e-0	3		
Np236		7.213e-02	2.813e-01	2.813e-0	1		
Np237	1.372e+03	1.316e+03	1.093e+03	1.097e+0	3	-275.	
Pu238		5.928e+01	2.884e+02	2.940e+0	2	+294.	
Pu239		2.834e-01	6.860e+00	6.916e+0	0	+ 6.9	
Pu240		4.279e+00	2.175e+01	2.615e+0	1	+ 26.2	
Pu241		1.841e-02	4.035e-01	3.673e-0	1		
Pu242		8.558e+00	3.905e+01	3.910e+0	1	+ 39.1	
Am241	1.601e+03	1.512e+03	1.173e+03	1.169e+0	3	-432.	
Am242m		5.892e+00	2.276e+01	2.253e+0	1	+ 22.5	
Am243	2.884e+02	2.763e+02	2.293e+02	2.292e+0	2	- 59.2	
Cm242		1.245e+01	1.057e+01	5.320e-0	1		
Cm243		5.891e-02	2.948e-01	2.808e-0	1		
Cm244	5.605e+01	5.769e+01	6.079e+01	5.632e+0	1	+ 0.3	
Cm245	3.310e+00	3.637e+00	4.946e+00	4.945e+0	0	+ 1.6	
Cm246		1.765e-02	1.104e-01	1.103e-0	1		
Cm247		8.815e-05	2.564e-03	2.564e-0	3		
(total)	6.034e+03	5.970e+03	5.678e+03	5.678e+0	3		
fis.prd	0.000e+00	6.190e+01	3.462e+02	3.462e+0	2	+346.	

TABLE	2 - PH	OENIX .OX	IDE .28%	LEAKAGE			
target	*****	cylindri	cal, heigh	t/diameter	= 113cm/1	14cm	
lattice		FFTF					
fuel m	ixture :	oxide M.	a. from th	e CURE pro	cess ('PHOI	ENIX' fuel)	
coolani	: :	sodium					
kg. m.a	a.loaded :	3330					
leakagi	e (BOC) :	28.0 %				,	
protons	s energy :	1600 MeV					
beam ci	irrent :	13 mA					
beam re	equires :	20.8 MW(	protons)*8	= 166 MWt	h target po	ower	
	B.O.C.	365 d	730 d	1095 d	1300 d	2030 d	
n-spall	30.2	30.6	30.3	30.4	30.0		
k-eff	.881	.888	.900	.907	.908		
leakage	28.0 %	28.8 %	29.2 %	29.8 %	30.0 %		
flux	1.47+15	1.59+15	1.81+15	2.00+15	2.03+15	0.0	
MW/Lite	er .180	.202	.230	.255	.257		
trgt MV	208	233	265	294	296		
grid mv	42	67	99	128	130		×
MWd/T	0	24200	51500	82000	100000		
			Kg. in	target			Kg. difference
11 m /		E 216 01	1 495-100	2 410-100	2 0080+00	2 8030+00	20300 -000
ne 4	4 440-100	5.3100-01	1.4200+00	2.4190+00	2.9900+00	3.003e+00	
No 22	4.4420+02	4.4410+02	4.440e+02	1 2420 02	1 8550-03	3 37/10-03	
Ne ZZ		1.0580-04	0.23/e-04	1.3438-03	1.6556-03	2 150p-03	
Na ZZ	2 2000102	1.1928-03	2.2010-03	3.1556-03	3.0782-03	2.139E-03	
Na 23	3.3900+02	3.3900+02	3.3090+02	J. 5060 02	5.5570702	5 6610-02	
My 24		1.3050-02	2.00/2-02	4.5908-02	9.4580-02	$1.5/8a_01$	
		7.514e-03	2.0358-02	1 / 97 - 01	1 7/80-01	$1.047e_{-01}$	
Fe 55	1 0200+02	1 0200+02	1.0422-01	1 0260+03	1 9250+03	1.047CH01	
Fe 50	1.9306+03	1.9290+03	1.9200+03	1.5202103	1 9250+00	1 925e+00	
10 37		9 162e-06	7 8/00-05	2 6990-04	4 501e-04	1 106e-03	•
11 233		3.501e-04	6.712e-04	9.333e-04	1.065e-03	1.788e-03	
11 234		2 /100-01	1 0680+00	2 474e+00	3.486e+00	8.133e+00	
11 235		3 902e-03	2.226e-02	7.020e-02	1.171e-01	1.274e-01	
11 236		1.296e-04	6.240e-04	1.830e-03	3.075e-03	5.399e-03	
Np235		6.687e-03	1.034e-02	1.272e-02	1.373e-02	3.825e-03	
Np236		9.475e-02	1.763e-01	2.472e-01	2.814e-01	2.814e-01	
Np237	1.372e+03	1.293e+03	1.213e+03	1.127e+03	1.077e+03	1.080e+03	-292.
Pu236		1.207e-04	5.266e-04	1.242e-03	1.775e-03	1.092e-03	
Pu238		6.822e+01	1.514e+02	2.333e+02	2.765e+02	3.062e+02	+306.
Pu239		3.832e-01	1.696e+00	4.162e+00	6.087e+00	6.168e+00	+ 6.2
Pu240		2.254e+00	4.614e+00	7.047e+00	8.431e+00	1.382e+01	+ 13.8
Pu241		1.351e-02	5.472e-02	1.282e-01	1.850e-01	1.689e-01	
Pu242		1.139e+01	2.242e+01	3.342e+01	3.950e+01	3.958e+01	+ 39.6
Am241	1.601e+03	1.486e+03	1.371e+03	1.251e+03	1.182e+03	1.178e+03	-423.
Am242m	1	7.789e+00	1.456e+01	2.052e+01	2.343e+01	2.320e+01	+ 23.2
Am243	2.884e+02	2.722e+02	2.559e+02	2.388e+02	2.288e+02	2.288e+02	- 59.6
Cm242		2.734e+01	3.283e+01	3.499e+01	3.593e+01	1.677e+00	+ 1.7
Cm243		1.460e-01	3.967e-01	6.737e-01	8.311e-01	7.916e-01	
Cm244	5.605e+01	6.171e+01	6.686e+01	7.178e+01	7.437e+01	6.891e+01	+ 12.9
Cm245	3.310e+00	3.774e+00	4.304e+00	4.922e+00	5.307e+00	5.306e+00	+ 2.0
Cm246		2.421e-02	5.334e-02	9.026e-02	1.152e-01	1.151e-01	
Cm247		1.610e-04	6.965e-04	1.763e-03	2.669e-03	2.669e-03	
(total	) 6.034e+03	5.948e+03	5.853e+03	5.744e+03	5.676e+03	5.676e+03	
fis.pr	d 0.000e+00	8.395e+01	1.764e+02	2.830e+02	3.494e+02	3.494e+02	+349.
		~~~~~~~~					

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TABLE 3	- PH(	DENIX .OX	IDE .24% L	EAKAGE				
target	:	cylindri	cal, heigh	t/diameter	= 130 cm/1	30cm		
lattice	:	FFTF						
fuel mixi	ture :	oxide M.	A. from the	e CURE pro	cess ('PHO	ENIX' fuel]	)	
coolant	:	sodium						
kg. M.A.	loaded :	5000						
leakage	(BOC) :	23.6 %						
protons (	energy :	1600 MeV						
beam curi	rent :	19.5 mA		050 1414				
beam requ	uires :	31.2 MW()	protons)*8	= 250 MWtH	n target p	ower		
	b.o.c.	226 d	432 d	615 d	759 d	902 d	1632 d	
n-spall	32.2	31.8	31.4	31.0	31.1	31.6		
k-eff	.904	.911	.921	.936	.936	.954		
leakage	23.6 %	23.9 %	24.3 %	25.1 %	25.1 %	25.7%		
flux	2.16+15	2.39+15	2.71+15	3.38+15	3.47+15	5.12+15	0.0	
MW/Liter	.255	.280	.316	.401	.403	.603		
trgt MW	440	483	546	692	696	1040		
grid MW	190	233	296	442	446	790		
MWd/t	0	20000	40000	60000	80000	100000		
			Kg. in :	2/3 target	·			Kg. difference 1632d - BOC
He 4		3.509e-01	9.708e-01	1.636e+00	2.223e+00	2.849e+00	4.019e+00	
0 16	4.442e+02	4.441e+02	4.441e+02	4.440e+02	4.440e+02	4.440e+02	4.440e+02	
Ne 22		5.236e-05	1.870e-04	3.754e-04	5.740e-04	8.185e-04	1.875e-03	
Na 22		6.178e-04	1.159e-03	1.651e-03	2.120e-03	2.557e-03	1.501e-03	
Na 23	3.390e+02	3.390e+02	3.389e+02	3.389e+02	3.388e+02	3.388e+02	3.388e+02	
Mg 24		1.273e-02	2.562e-02	3.860e-02	5.133e-02	6.431e-02	6.440e-02	
Mn 55		2.639e-03	9.438e-03	1.897e-02	2.904e-02	4.144e-02	9.553e-02	
Fe 55		3.237e-02	6.086e-02	8.685e-02	1.116e-01	1.348e-01	8.067e-02	
Fe 56	1.930e+03	1.929e+03	1.928e+03	1.927e+03	1.926e+03	1.926e+03	1.926e+03	
Fe 57		4.348e-01	8.732e-01	1.315e+00	1.748e+00	2.189e+00	2.1890+00	
U 233		2.082e-04	4.221e-04	6.253e-04	8.189e-04	1.0510-03	1./510-03	
U 234		1.247e-01	5.062e-01	1.0/8e+00	1.6/60+00	2.39/e+00	/.440e+00	
0 235		1.300e-03	6.623e-03	1./868-02	3.490e-02	5.8940-02	0./510-02	
U 236		5.42/e-05	2.5//e-04	7.040e-04	1.4/50-03	2.7020-03	4.038e-03	
Np235		3.902e-03	0.5198-03	8.433e-03	2 2100 01	2 5200 01	2 5290-01	
Np230	1 270-102	1.300-102	1.3398-01	1.0320-01	1 1020+03	2.5296-01	1 0460+03	-326
NP237	1.3/20+03	5 6300+01	1.2300+03	1.1042+03	2 377 a+02	2 870e+02	3.376e+02	+338.
Fu230		2 001e-01	1 2100+00	2 7370+00	4 701e+00	7.077e+00	7.182e+00	+ 7.2
Pu239		1 4140+00	2 7760+00	4 034e+00	5.047e+00	6.084e+00	1.186e+01	+ 11.9
Pu240		7.8876-03	2.976e-02	6.305e-02	1.037e-01	1.509e-01	1.380e-01	
Pu241		1 050e+01	2.013e+01	2.888e+01	3.661e+01	4.369e+01	4.379e+01	+ 43.8
Δm2/1	1 6010+03	1 4970+03	1 3980+03	1.306e+03	1.222e+03	1.141e+03	1.137e+03	-464.
$\Delta m 242 m$	1.0010.00	7.224e+00	1.325e+01	1.822e+01	2.218e+01	2.539e+01	2.514e+01	+ 25.1
Am243	2.884e+02	2.736e+02	2.596e+02	2.464e+02	2.343e+02	2.226e+02	2.226e+02	- 65.8
(m242	2.0040.02	3.195e+01	4.401e+01	5.002e+01	5.625e+01	5.822e+01	2.686e+00	
Cm243		1.458e-01	4.253e-01	7.385e-01	1.057e+00	1.375e+00	1.310e+00	+ 1.3
Cm244	5.605e+01	6.199e+01	6.727e+01	7.193e+01	7.606e+01	7.965e+01	7.381e+01	+ 17.8
Cm245	3.310e+00	3.742e+00	4.208e+00	4.698e+00	5.189e+00	5.694e+00	5.693e+00	+ 2.4
Cm246		2.210e-02	4.668e-02	7.385e-02	1.029e-01	1.350e-01	1.349e-01	
Cm247		1.343e-04	5.416e-04	1.229e-03	2.176e-03	3.421e-03	3.421e-03	
(total)	6.305e+03	6.228e+03	6.150e+03	6.069e+03	5.990e+03	5.908e+03	5.908e+03	
fis.prd	0.000e+00	7.488e+01	1.516e+02	2.300e+02	3.076e+02	3.871e+02	3.871e+02	+387.1

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- PHOENIX .METAL .44% LEAKAGE TABLE 4 : cylindrical, height/diameter = 60cm/60cm target : FFTF lattice : metal, M.A. from the CURE process ('PHOENIX' fuel) fuel mixture : sodium coolant kg. M.A. loaded : 3330 (into 4 unites) leakage (BOC) : 44.2 % protons energy : 1600 MeV : 13 mA (divided between 4 unites) beam current : 20.8 MW(protons)\*8 = 166 MWth target power (from 4 unites) beam requires 737 d 1467 d B.O.C. 218 d 462 d n-spall 31.1 30.7 30.3 30.0 .942 .936 .930 .930 k-eff 45.5 % 45.5 % 44.2 % 44.7 % leakage 2.40+15 0.0 2.85+15 2.62+15 2.37+15 flux .585 .746 .592 MW/Liter .669 397 trg MW \*4 506 454 402 288 236 231 grd MW \*4 340 66667 100000 MWd/T 0 33333 Kg. difference Kg. in 4 targets 1467d - BOC 3.181e-01 9.625e-01 1.726e+00 2.473e+00 He 4 5.043e-05 2.082e-04 4.819e-04 1.129e-03 Ne 22 1.994e+02 1.994e+02 1.994e+02 1.993e+02 1.993e+02 Na 23 6.731e-03 1.366e-02 2.075e-02 2.077e-02 Mg 24 2.135e-03 8.832e-03 2.048e-02 4.834e-02 Mn 55 2.712e-02 5.041e-02 6.940e-02 4.154e-02 Fe 55 1.135e+03 1.135e+03 1.134e+03 1.134e+03 1.134e+03 Fe 56 2.690e-01 5.448e-01 8.265e-01 8.265e-01 Fe 57 1.271e-03 2.605e-03 3.966e-03 3.989e-03 Y 89 6.599e-02 1.336e-01 2.027e-01 2.027e-01 Zr 90 5.566e+02 5.563e+02 5.559e+02 5.555e+02 5.555e+02 Zr 91 Zr 92 1.830e-01 3.705e-01 5.619e-01 5.619e-01 1.967e-04 4.510e-04 7.759e-04 1.527e-03 U 233 1.035e-01 4.998e-01 1.272e+00 4.243e+00 U 234 1.604e-03 8.017e-03 2.239e-02 3.075e-02 U 235 5.889e-05 3.394e-04 1.075e-03 2.561e-03 U 236 6.120e-03 9.767e-03 1.142e-02 3.182e-03 Np235 1.119e-01 2.038e-01 2.774e-01 2.774e-01 Np236 -250. 1.372e+03 1.284e+03 1.199e+03 1.118e+03 1.122e+03 Np237 +199. Pu238 4.861e+01 1.095e+02 1.688e+02 1.988e+02 2.238e-01 9.467e-01 2.178e+00 2.243e+00 + 2.2 Pu239 1.402e+00 2.960e+00 4.697e+00 9.496e+00 + 9.5 Pu240 7.439e-03 2.884e-02 6.346e-02 5.831e-02 Pu241 9.634e+00 1.837e+01 2.617e+01 2.623e+01 + 26.2 Pu242 1.596e+03 1.473e+03 1.357e+03 1.248e+03 1.244e+03 -352. Am241 + 16.36.624e+00 1.206e+01 1.642e+01 1.626e+01 Am242m - 49.7 2.901e+02 2.728e+02 2.562e+02 2.404e+02 2.404e+02 Am243 2.964e+01 3.670e+01 3.453e+01 1.596e+00 + 1.6 Cm242 1.034e-01 2.850e-01 4.527e-01 4.312e-01 Cm243 5.773e+01 6.141e+01 6.425e+01 6.621e+01 6.135e+01 + 3.6 Cm244 + 1.0 3.410e+00 3.723e+00 4.047e+00 4.370e+00 4.370e+00 Cm245 1.990e-02 4.147e-02 6.469e-02 6.467e-02 Cm246 1.537e-04 6.155e-04 1.383e-03 1.383e-03 Cm247 (total) 5.211e+03 5.083e+03 4.954e+03 4.824e+03 4.824e+03 1.241e+02 2.497e+02 3.763e+02 3.763e+02 +376. fis.prd

## References

- C.D. Bowman et al. Nuclear Energy WITHOUT LONG-TERM HIGH-WASTE Trns. A.N.S. 63, 1991 (p. 80).
- G.J. Van Tuyle et al.
   Accelerator-driven Subcritical Target Concept for Transmuation of Nuclear Wastes
   BNL-46365 Dec. 1991.

- 3. Chapter 1 of this report
- T. Mukaiyama et al.
   Conceptual Study of Actinide Burner Reactors
   Proc. Reactor Physics Conf. Jackson-hole, U.S:A., 1988