SESSION 3: SYSTEMS STUDIES

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POTENTIALITIES AND COSTS OF PARTITION AND TRANSMUTATION OF LONG-LIVED RADIONUCLIDES

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Abstract

The purpose of this conceptual study is to analyse the different strategies of waste management aiming at reducing contents of long lived radionuclides by partitionning and transmutation. Different scenarios are studied; they are classified in 3 phases : the first one concerns scenarios without separation of minor actinides, the second one concerns scenarios of separation and transmutation (Np and Am) with the present technologies and the last one concerns scenarios of separation and transmutation with innovative technologies. We compare the different scenarios in term of waste material and radiotoxicity balances.The separation process is presented and we estimate a comparison between the present fuel cycle costs and the fuel cycle costs in case where we separate and transmutate Np and Am with the present technologie.

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1. OBJECTIVE AND SCOPE

The purpose of this conceptual study is to analyse the different strategies of waste management aiming at reducing contents of long lived radionuclides by partitionning and transmutation; technical ways and costs will be evaluated.

In a first step reference scenario will be defined to draw up an inventory of long lived radionuclides produce by the west European installed or foreseen reactors and estimate radiotoxicity of such nuclides.

In a second step will be estimated the expected decrease of radionuclides stockpile according to two mains scenarios : first one using presently known or available technologies, second one taking into account all foreseen innovative technologies.

After a recall of principal hypothesis, the paper presents the results concerning :

The material balance,

The potential radiotoxity (ingestion) balance,

The separation process,

An economic study.

This study is a part of the contract with the Commission of the European Communities.

2. PRINCIPAL HYPOTHESIS

2.1 COMMON HYPOTHESIS FOR THE GENERAL SCENARIOS

The scenarios last for 100 years (2000 - 2100) for installed reactor park of 120 GWe, this power is at a constant level during the scenario,

The radiotoxicity is evaluated, for every cycle station, up to 10^7 years,

All the results are normalised the reference scenario (R1),

Two radiotoxicity inventories were performed :

- "waste" inventory including the radiotoxicity due to nuclear materials suitable for ultimate disposal,

- "cycle" inventory including the radiotoxicity due to nuclear materials throughout the fuel cycle (industrial facilities, interim storage, reactors and wastes).

2.2. SELECTED SCENARIOS

The study includes three main phases :

The reference scenarios without partition and transmutation of wastes :

- R1, the reference scenario with a reactor park having pressurised water reactors (PWRs) solely and burning uranium oxide (UOX), the fuel cycle is open without reprocessing.

-        R2, with the same reactor park as R1, but burning UOX and MOX fuel, the cycle is closed with PUREX reprocessing (the losses are : 0.3 % for U and 0.5 % for PU) of UOX and MOX. All the plutonium is recycled in the MOX fuel. The reprocessing is made in dilution (MOX/UOX ≤ 25 %).

-        R3, similar to R2 until 2020, fast reactors (FR) are progressively installed after 2020. The fabrication of the MOX fuel for the PWRs, is stopped after 2020.

To use a partition and transmutation with available technologies :

-        RP1-1 is compared to the R2 scenario, it's similar until 2010. In the RP1-1 scenario, neptunium and americium are incinerated in PWRs either in homogeneous or in heterogeneous mode.

In homogeneous mode, a quantity of neptunium oxide or americium oxide representing 1 % of the total mass of heavy isotopes is mixed to the UOX fuel. The UOX + actinide fuel is reprocessed as the standard UOX fuel. The losses during reprocessing are 0.3 % for U, 0.5 % for Pu, 5 % for Np and Am and 100 % for Cm.

In heterogeneous mode, specific targets are placed in guide tubes of a UOX assembly ; the pins are made of either 100 % neptunium oxide or 30 % americium oxide with 70 % alumina. These targets are irradiated during 5 years and are not reprocessed.

-        The RP1-2 scenario is compared to the R3 scenario. As the minor actinide partitioning starts in 2010, Np and Am are stored before being recycled in FRs after 2020 either in homogeneous or in heterogeneous mode.

In the homogeneous mode, an amount of Np or Am representing 2.5 % of the total mass of heavy isotopes is mixed with the FR fuel. The fuel is reprocessed as the standard FR fuel. The actinide losses during reprocessing are the same as in the RP1-1 scenario.

In the heterogeneous mode, neptunium and americium oxide targets coated with alumina are placed at the first radial breeding row of the core. These targets contain 40 % of Np or 20 % of Am of the metal mass. They are irradiated during 15 years (3 cycles) and are not reprocessed.

To use partition and transmutation with the innovative technologis after 2030.

RP2 is similar to RP1-2 until 2030 ; CAPRA type fast reactors are progressively installed after 2030 to transmutate :

•   Neptunium in the homogeneous mode,

•   Americium in the heterogeneous mode ; the targets are placed in the radial periphery of the core,

•   Technecium and Iodine in the heterogeneous mode ; the targets are placed in the axial periphery of the core.

All the fuel and the targets are reprocessed ; the curium is separate and placed in interim storage. The losses during reprocessing are 0.1 % for U, 0.1 % for Pu, 0.5 % for Np, 0.5 % for Am, 0.5 % for Cm, 10 % for Tc and 10 % for I.

The code "COSI" (1) has been used to simulate these scenarios and to compare the material balances and the radiotoxicity.

3. RESULTS CONCERNING THE MATERIAL BALANCE

3.1. URANIUM BALANCE

• Uranium losses from reprocessing

Figure 2 shows the evolution of uranium losses from reprocessing. The values of RP1-1 respectively RP1-2, and R2, respectively R3, are comparable. The scenario RP2 permit to reduce the losses by a factor 2.5.

3.2. PLUTONIUM BALANCE

Figure 3 shows the evolution of the initial Pu content in the MOX-PWRs for scenario R2 and RP1-1. The damage of plutonium, due to plutonium recycle, results in increase the Pu content at the fabrication.

Table hereafter shows the evolution (%) of plutonium from reprocessing for scenarios R2 and RP1-1.

	SCENARIO R2 AND RP1-1				
	2020	2040	2060	2080	2100
Pu 238	4,1	6,2	7,5	8,5	8,9
Pu 239	43,8	41,9	40,4	39,2	38,7
Pu 240	26,5	26,5	26,6	26,7	26,8
Pu 241	14,7	14,1	13,4	13,0	12,9
Pu 242	10,4	10,9	11,8	12,2	12,4
Am 241	0,4	0,3	0,3	0,3	0,3

• Plutonium losses from reprocessing

Figure 4 shows the evolution of plutonium losses from reprocessing. The scenario RP2 permits to reduce the plutonium losses by a factor 4.

3.3. MINOR ACTINIDES BALANCE

Figures 5, 6 and 7 show the evolution of Np, Am and Cm mass for disposal :

- For the scenario R1, it's the mass contained in the irradiated fuels.

- The transmutation of Np permits to gain a factor 10.

- The transmutation of Am permits to gain a factor 10 in the FRS and a factor 15 in the PWRs.

- The transmutation of Am increases the production of Cm.

- For the scenario RP2, the minor actinides for disposal are principaly the mass produced before 2020 without separation.

3.4. THE MATERIAL BALANCE OF WASTES

The table shows the cumulative (tons) actinide wastes (Pu, Np, Am, Cm) in 2100.

Reprocessing wastes	R1	R2	R3	RP1-1	RP1-2	RP2
Pu	1 870	22	23	22	23	7
Np	140	100	94	12	11	7
Am	310	420	250	27	24	11
Cm	6	65	40	108	54	10

4. RESULTS CONCERNING THE RADIOTOXICITY

Figures 8 and 9 show the evolution of the potential ingestion radiotoxicity beginning in the year 2100 in the "waste" and "cycle" inventories.

The "waste" inventory takes into account :

R1 :- Losses (U) (Conversion, Fabrication) and Irradiated fuels for disposal,

R2, R3 :- Losses (U, Pu) (Conversion, Fabrication, Reprocessing) and Am, Np, Cm, F.P. (Tc 99, I 129, Cs 135) from reprocessing,

RP1-1, RP1-2 :- Losses (U, Pu, Np, Am) and Cm, FP (Tc 99, I 129, Cs 135) from reprocessing,

RP2 :- Losses (U, Pu, Np, Am, Cm, Tc 99, I 129, Cs 135).

The "cycle" inventory takes into account :

R1 :- "Wastes" inventory, Depleted Uranium and Fuel in reactors,

R2 - R3 :- "Wastes" inventory, Depleted Uranium, Uranium from reprocessing, Fuel in reactors and Interim storage (Pu, Spent fuel),

RP1-1, RP1-2 :- "Waste inventory", Depleted uranium, Uranium from reprocessing, Fuel in reactors, Interim storage (Pu, Np, Am, Spent fuel) and Targets for heterogeneous mode,

RP2 :- "Waste inventory", Depleted uranium, Uranium from reprocessing, Fuel in reactors and Interim storage (Pu, Np, Am, Cm, Tc 99, I 129, Cs 135 Spent fuel, targets).

4.1. COMMENTS ON THE "WASTE" INVENTORY

4.1.1. The reference scenarios (R2, R3)

For the short cooling times (between 10 to 1000 years) the contribution of the radiotoxicity is due to the Cm 244 (at the beginning), then to the Am 241 :

- The more important contribution of Cm 244 in the MOX fuel penalizes R2 in comparison with R1 (until 30 years).

• The Pu 241 presence in the waste for R1 ; induced a favorable balance for R2 and R3 ; the R3 balance is better because the production of Am 241 in the FR is less important than in the MOX-PWRs.

For the long cooling times (betwen 1000 to 100 000 years), the contribution is due to the Pu 239 and Pu 240. With a Pu recycle (R2 and R3) we have an important gain (5 to 10) in comparison with R1.

For the very long cooling times (between 100 000 to 5 000 000 years), the contribution is due to Np 237 and Th 229. The Pu recycle (R2, R3) reduces the Np 237 production. This reduction is more important in the fast reactor (R3).

4.1.2. The scenario using partition and transmutation with available technologies (RP1-1, RP1-2)

Significant gains (by a factor of 6) over scenario R2 and R3 are obtained for two time periods :

• between 10^2 and 10^3 years, a period when most of the radiotoxicity is attributable to Am,

• between 5.10^5 and 5.10^6 years, a period when most of the radiotoxicity is due to Np.

For the short periods, the increased Cm production appreciably limits the gains.

Transmutation of Np and Am changes little a period when most of the radiotoxicity is attributable to Pu.

Compared with scenario R1, the average gain is by a factor of 10 to 50 beyond the first 100 years.

For the heterogeneous mode who isn't showed in the figure, the comments are similar.

4.1.3. The scenario using partition and transmutation with innovative technologies after 2030 (RP2)

The separation of Cm permits to gain an important factor for the short cooling times.

The losse reduction at the reprocessing permits to reach a factor of 100 compared with scenario R1.

4.2. COMMENTS ON THE "CYCLE" INVENTORY

The results concerning the reference scenarios (R1, R2 and R3) show that the Pu recycling doesn't degrade the cycle inventory.

The contribution of "waste" inventory evoluates from case to case ; after the first 100 years, the "waste" represent :

- less than 50 % to reach 10 % after 10^7 years for the reference scenario (R2, R3),

- less than 20 % to reach 1 % after 10^7 years for the scenario RP1-1, RP1-2,

- less than 5 % to reach 0.5 % after 10^7 years for the scenario RP2.

The other important items are irradiated fuels (both in interim storages and in the reactors) ; up to 10^6 years, depleted uranium account for most of the radiotoxicity thereafter.

For the scenario R2 and RP1-1, the radiotoxicity increases during the first 100 years due to Cm production.

Transmutation of Np and Am doesn't degrade the "cycle" inventory.

5   RP1 PROCESS

5.1   PROCESS SPECIFICATIONS

The RP1 process is intended to separate U, Pu, Np, Am and the fission products. In the finished product, uranium is present as uranyl nitrate, while plutonium, neptunium and americium are found in oxide form. The process adopted must be based on existing knowledge and technology for industrial implementation by 2010.

In view of these constraints, we have designed RP1 as an extension of the PUREX process currently implemented in the UP2 and UP3 plants at La Hague. The necessity of using existing methods required us to propose some solutions that probably are penalizing in the field of process complexity and the capital and operating cost.

5.2   OPERATING PRINCIPLE

The RP1 operating principle is summarized in the flowsheet in Appendix 5.1. It is designed as an add-on to the PUREX process, with U/Pu partitioning during the first cycle.

Neptunium is retained with the uranium by redox adjustment, and U/Np partitioning occurs during the second U cycle. The Np solution is then concentrated and the Np is converted to oxide form by the same oxalate process now used for Pu. No other changes are implemented in the U and Pu lines.

Am and Cm are obtained from the refined stream of the first U/Pu cycle by coextraction with the lanthanides, followed by (Am,Cm)/lanthanide separation. Am and Cm are then separated using a selective oxidized Am extraction process; americium oxide is obtained by the oxalate process. The Cm is returned with the fission products for vitrification.

5.3   REPROCESSING PLANT DESIGN

The RP1 process is common to both PWR (UOx-MOx) and FBR reprocessing plants, with some minor differences: the head-end facilities (fuel entry, cutup and dissolution) must be adapted to the type of fuel reprocessed; the capacities of the U and Pu lines must be adapted to process requirements, and the geometry of the subcritical process equipment is different, since it depends on the fuel medium.

The plant design remains consistent with the basic RP1 options, i.e. the minor actinide separation and conversion units are situated around the periphery of a conventional PUREX plant. The actual design is based on the known concept of the UP3 plant at La Hague. Appendix 5.2 provides an overview of the integration of the new facilities in the reprocessing plant.

We have assumed that the conventional portion of the plant will not be upgraded before 2010. The technical and economic effects of implementing neptunium and americium separation will therefore not be hidden by other factors for which the economic implications are difficult to assess.

6   ECONOMIC ASSESSMENT

6.1   FINAL OBJECTIVE OF THE ECONOMIC ASSESSMENT

In addition to the technical considerations discussed in the preceding chapters, the question was also approached from an economic standpoint. The management cost of the fuel cycle R2b and R3 was compared with a management route including separation and transmutation of the minor actinides (neptunium and americium). The options assessed were those of the two Level 1 enhanced reprocessing scenarios:

- RP1-1, in which plutonium and the minor actinides (Np and Am) are incinerated in PWRs;

- RP1-2, in which the same nuclides are incinerated in FBRs.

The results are indicated relative to a reference fuel cycle management cost. The reference scenarios are the following[1]: reprocessing without separation of the minor actinides and recycling of plutonium (except for process losses) in PWRs (scenario R2) or in FBRs (scenario R3). The unit cost of each fuel cycle operation was determined for purposes of comparison.

Actinide transmutation is assumed to occur in the same types of power reactors found in the reference population, without requiring significant modifications. For the reactor portion of the fuel cycle, the RP1 scenarios therefore do not include any capital or operating cost supplement. It may be necessary to modify the fuel, however, notably the ^{235}U or plutonium enrichment, but these supplements are included in the overall fuel cycle cost.

6.2   METHOD

The fuel cycle costs were considered on an annual basis for this study, by multiplying the material flows for one year by the unit operating cost. Most of the figures are based on national or international assessments (except for reprocessing and fuel fabrication costs, which were evaluated as part of this study) and are roughly equivalent to current market prices. This approach, which does not allow for discounting, identifies the relative weight of each fuel cycle step in the scenario, but the result cannot be integrated into the cost per kWh, which is assessed using a different method.

Two or three successive steps are required:

- Step 1: determination of the capital cost of the production facilities for each fuel cycle item.

- Step 2: determination of the corresponding unit cost[2].

- Step 3: determination of the annual fuel cycle expenditure.

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[1]The direct disposal scenario without reprocessing (R1) is used as a reference only for radiotoxicity comparisons; refer to Chapter 2 for a detailed description of these scenarios.

[2]This is an internationally accepted approach.

For the reference scenarios, the unit costs for each fuel cycle item are available directly, as they have already been determined at a national (CEA[3], DIGEC[4]) or international (OECD[5]) level, and correspond approximately to market prices, although some adjustments were required to allow for special constraints (notably for Scenario R2[6]).

For the other scenarios, two items in the cycle were modified: reprocessing and actinide fuel fabrication. The method described above was applied to reprocessing to allow for process modifications. The cost of actinide fuel fabrication was estimated directly in terms of operating costs based on MOx fuel fabrication costs.

The scenarios may be compared from an economic standpoint at each step:

- Step 1: comparison of reprocessing plant capital costs (RP1-1/R2 and RP1-2/R3).

- Step 2: comparison of unit reprocessing costs.

- Step 3: comparison of fuel cycle expenditures (RP1-1/R2 and RP1-2/R3).

From a waste disposal standpoint, the scenarios were compared only in terms of the potential radiotoxicity hazard. Disposal costs for reprocessing waste were assumed constant for all the reprocessing scenarios, as the separation yields obtained by the RP1 process are not sufficient to allow decategorization of a fraction of the fuel cycle wastes. Inasmuch as the costs are compared with a reference scenario, the waste disposal cost should not have a significant effect on the results.

6.3 REPROCESSING PLANT CAPITAL COST ESTIMATE (Step 1)

6.3.1 R2 and RP1-1 Plants

This step begins with an examination of the engineering design of the R2 and RP1 processes to conceive the overall structure of the reprocessing facilities, to specify the functions they must implement and to determine their capital cost. The analysis assumes an annual plant capacity of 800 metric tons of heavy metal based on cost data for the UP3 plant at La Hague. This represents approximately half the capacity required to meet the total equilibrium reprocessing demand under scenarios R2 or RP1-1. A unit reprocessing cost estimate based on a plant of this size may be considered representative of the implications of these scenarios.

The functional structures of the R2 and RP1 plants are summarized in Appendix 5.2. As in the La Hague reprocessing plant, each function is performed in a workshop for which the capacity and functional structure were assessed. The R2 and RP1 plants implement existing techniques, and the structures (although not the capacity) of virtually all the workshops are therefore

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[3] *French Atomic Energy Commission.*

[4] *French Gas, Electricity and Coal Utility Management Board.*

[5] *Organization for Economic Cooperation and Development.*

[6] *Scenario R2, in which all the plutonium (except for process losses) is recycled as MOx fuel in PWRs, results in a Pu/(U+Pu) ratio 3.5 higher for the reprocessing plant input stream than standard UOx fuel; this implies that the corresponding adjustment of the Pu reprocessing capacity.*

comparable to those of existing facilities. The costs of the new workshops are thus extrapolated from the known figures for reprocessing plants.

The plant construction cost is then the sum of the unit costs for each workshop, together with the site and utility costs.

6.3.2 R3 and RP1-2 Plants

A different approach was used to assess the capital cost of FBR MOx fuel reprocessing plants; no industrial facilities of this type exist for which reference costs validated by experience could be itemized, as for a PWR UOx reprocessing plant.

However, in 1986 the CEA, COGEMA and SGN conducted a joint preliminary design study to estimate the capital cost of a reprocessing plant of this type with a annual capacity of 400 metric tons. The figures from that study were discounted to the economic conditions prevailing in 1992 and used as the reference for the R3 plant. The 400 t.yr^{-1} capacity corresponds relatively well to the demand postulated under this scenario, in which the quantities of FBR MOx fuel to be reprocessed range from 80 to 600 metric tons per year as fast breeder reactors go online between 2030 and 2100.

The estimated cost of the RP1-2 plant is based on the assumption that the RP1 chemical process is valid for both PWR and FBR reprocessing plants, and that the differences between them are related to mechanical, thermal or criticality constraints. The differences concern mainly the head-end workshops and the first extraction workshop; the remaining facilities differ only by the magnitude of the process flows. In fact, the RP1-2 plant is an R3 plant with the addition of neptunium and americium separation and conditioning units. The flowsheet is the same as for the RP1-1 plant, and uses the same types of facilities. The cost supplement of the RP1-2 plant over the R3 plant is therefore the sum of four components:

- The neptunium line (dimensioned according to the neptunium flow).

- The fission product and lanthanide line (dimensioned according to the fission product flow, which in turn is proportional to the MWd equivalent throughput of the plant).

- The americium line (dimensioned according to the americium flow).

- The final product interim storage facilities (dimensioned according to the sum of the neptunium and americium flows).

The cost of each component may be estimated from the cost of the corresponding component in the RP1-1 plant with allowance for differences in capacity.

6.4 UNIT COST ASSESSMENT (Step 2)

6.4.1 Reprocessing

The basic principle was to assess the cost of the reprocessing operation from the capital cost of the plant, allowing for all the outlay required throughout the life of the facility and for the total cumulative production. The method was applied to the reprocessing plants included in the reference scenarios (R2 and R3) and to the Np-Am separation scenarios (RP1-1 and RP1-2).

For the lifetime of the reprocessing plant (construction, operation, decommissioning) the unit cost of the reprocessing step was determined from the following relation:

$$\text{Reprocessing cost} = \frac{\text{Discounted (capital + renovation + decommisioning + operating costs)}}{\text{Discounted (total production)}}$$

The capital, renovation, decommissioning and operating costs were determined from the construction cost with the following hypotheses:

• Capital cost = construction cost + interim interest

The plant construction was assumed to last 10 years; the annual capital cost breakdown was based on observed figures for typical nuclear facilities; the expenditures were discounted to the date of commercial startup.

• Renovation

¥ First renovation beginning the 11th year of operation, representing 30% of construction cost, amortized over 15 years and discounted to the date of commercial startup.

¥ Second renovation beginning the 26th year of operation, representing 5% of construction cost, amortized over 5 years and discounted to the date of commercial startup.

• Final shutdown after 35 years of operation

• Decommissioning over a 5-year period after final shutdown, representing 35% of construction cost, discounted to the date of commercial startup.

• Annual operating cost representing 5% of construction cost, discounted to the date of commercial startup; a 1% annual increase in the operating cost was assumed.

The discounting rate (5% for this study) naturally affects the absolute value of the result, but has no effect on the relative value.

Estimated Cost

The reprocessing cost is directly proportional to the construction cost:

Reprocessing cost = (Construction cost) x K

The unit cost supplements for RP1-1 relative to R2, and for RP1-2 relative to R3, are therefore the same as those determined for the construction costs:

Relative Cost Supplement of Enhanced Reprocessing	
$\dfrac{(RP1.1 - R2)}{R2} = 0.35$	$\dfrac{RP1.2 - R3}{R3} = 0.48$

6.4.2 Actinide Fuel Fabrication

A different approach was used to assess the actinide fuel fabrication step. It was assumed that the fabrication process itself will not raise insurmountable difficulties, and attention therefore focused on the radiation protection requirements imposed by the addition of neptunium and americium to existing fuel fabrication processes.

Homogeneous recycling of neptunium with UOx fuel in a mixed oxide fuel fabrication facility such as MELOX appears to be a viable solution, subject to enhancement of biological shielding, notably during the initial process steps.

The feasibility of fabricating a homogeneous fuel containing 1-2% americium in the same type of facility remains to be demonstrated. In any event, the entry of AmO_2 powder into the facility would require a very high degree of automation. Even after blending, americium recycling would increase the specific activity of the process stream by a factor of 10 compared with actual MOx fuel fabrication. Under these conditions, the maximum specific activity level specified for the initial container discharging station in MELOX would prevail throughout the fuel fabrication cycle.

A detailed assessment of the cost of these modifications is beyond the scope of this investigation.

The use of MELOX or similar technology for the fabrication of fuel containing neptunium or americium would require a number of modifications (additional shielding, automation) to avoid higher occupational doses; these modifications could result in a fabrication cost supplement of 20% over MOx fuel fabrication. **This value will be used as the reference for the remainder of this investigation**, although two additional values (+ 10% and + 100%) were considered in a sensitivity study. The first reflects the fact that the fabrication of UO_2 + NpO_2 + AmO_2 fuel will cost at least as much as MOx fuel, and that even under ideal circumstances, some modifications would be necessary, resulting in a minimum cost supplement of 10%. The second figure assumes that the glove box technology implemented in the MELOX plant must be abandoned, and that a different shielded line technology must be developed, with a major impact on fabrication costs. A figure of + 100% must not be taken too literally, as it was not obtained by a serious evaluation but is only postulated to assess the sensitivity of the overall results to a significant increase in the actinide fuel fabrication cost.

The relative cost supplement over the reference scenario, which could be the cost of either UOx or MOx fuel fabrication, is indicated below:

Relative Unit Cost Supplement of Actinide Fuel Fabrication Compared with:	
UOx fuel	2.6 < **2.9** < 5.5
MOx fuel[7]	0.1 < **0.2** < 1.0

6.5   COMPARATIVE FUEL CYCLE COSTS (Step 3)

The expenditures are calculated year after year by multiplying each item in the fuel cycle material or service balance by the unit cost. All the calculations are performed in constant French Francs under the economic conditions prevailing in 1992, without discounting.

Tow material and services balance was done :

- A cumulative balance throughout the duration of each scenario, reflects the relative importance of each scenario because it integrates all the transient phases.

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[7]The cost of MOx fuel fabrication is roughly 3.25 times that of UOx fuel.

- A typical steady state annual balance, minimizes the inertia factor in the fuel cycle especially for the FBR scenarios that change for the duration of the study.

The comparison of fuel cycle expenditure, were made from three types of balances without any discounted cost :

* Yearly balances, showing the evolution of cycle expenditures year by year.

* Cumulated balances over the duration of the scenarios, which take into account the effects of inertia of the scenarios.

* Current year balances, taken at the end of the period to be as close as possible to stable state. This type of comparison is significant of the differences of nature between scenarios. It is the only one we will discuss about here after.

At the end we carry out a sensitivity study, in order to assess the impact of a variation of the unit operating costs on the global results.

6.5.1 Typical Steady-State Annual Balance

6.5.1.1 R2 and RP1-1 scenarios

This assessment concerns a typical year of steady-state operation after 2040. The graph in Appendix 6.1 indicates the absolute outlay assigned to each cycle item for scenarios R2 and RP1-1.

The expenditures for scenario RP1-1 systematically exceeded those of scenario R2. At the front end of the cycle, this is due to the larger material balance and to the fabrication of UOx fuel containing actinides; for the back end, it is attributable to higher reprocessing costs.

Reprocessing (15%) and actinide fuel fabrication (17%) are the most significant additional expenditures incurred by transmutation of the minor actinides. Uranium mining and enrichment each represent 2.5% cost supplements, while the UOx fuel fabrication item diminishes by about 4% (again, it must be noted that the value of uranium and plutonium was deliberately considered nil, and not as material assets).

Under steady-state conditions, the total fuel cycle outlay under scenario RP1-1 exceeds that of R2 by 33%. Appendix 6.2 details the relative weight of each item in the overall steady-state annual balance for both scenarios.

Reprocessing is the dominant item in both scenarios; scenario RP1-1 also includes actinide UOx fuel fabrication. The following remarks may be made concerning these points:

• The importance of reprocessing (45%) is not surprising, and is consistent with the annual expenditure approach adopted for this study. This result cannot be directly compared with previously published economic assessments in which reprocessing represents only about 30% of the conventional fuel cycle cost: such figures clearly show the effect of discounting, which minimizes the value of outlay in the distant future compared with immediate outlay. No such effect is observed in this investigation, as the costs were not discounted. The

reprocessing cost is also increased over that of conventional reprocessing[8] because of massive plutonium recycling (R2) and the implementation of enhanced reprocessing methods (RP1-1).

- Recycling of minor actinides in UOx fuel is certainly not the best solution from a purely economic standpoint, as it subjects a large fraction of the UOx flow to fabrication costs comparable to those of MOx fuel, representing a three-fold cost supplement. If it were technically feasible, it would be preferable to recycle the largest possible actinide fraction in MOx fuel, the fabrication of which is already penalized by radiation protection constraints, and for which a few additional percent of neptunium or americium should have a lower impact on the fabrication costs.

6.5.1.2 R3 and RP1-2 Scenarios

These scenarios involve both PWR and FBR plants, and the assessment is therefore compiled using cycle costs estimated from different types of sources:

- Data based on current industrial experience for PWR cycle costs.

- Research data that have not been qualified industrially for FBR cycle costs.

Because of the different reliability of these estimates, errors may ensue that cannot be estimated at the present time; the following results must therefore be interpreted cautiously.

This assessment concerns the year 2090. The graph in Appendix 6.3 indicates the absolute outlay assigned to each cycle item for scenarios R3 and RP1-2.

The expenditures for scenario RP1-2 systematically exceed those of scenario R3. The higher front-end cycle costs of scenario RP1-2 are due mainly to increased outlay for actinide MOx fuel fabrication (2%). This supplement is minimal compared with the back-end cost supplements for PWR reprocessing (12%) and FBR reprocessing (14%). FBR fuel reprocessing is considerably more penalizing that in the cumulative balance because at the end of the scenario the FBR cycle expenditure is not weighted by integration of the balances over the entire duration of the scenario. In 2090, the proportion attributable to FBR reprocessing continues to rise as PWR reprocessing expenditure declines.

In a typical operating year, the total outlay for scenario RP1-2 exceeds that of scenario R3 by 28%. The graph in Appendix 6.4 indicates the relative weights of each item in the overall cycle outlay during a typical year. Reprocessing is the principal item in both scenarios, accounting for about 60% of the total.

6.5.2 Sensitivity of Estimates to Unit Cost Hypotheses

The results of the economic assessment depend directly on the unit operating costs. The figures used for this study were obtained from a variety of sources and are not all equally reliable. Three data categories were considered:

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[8] *Reprocessing of standard UOx fuel (33 GWd.t^{-1}) with a Pu/(U+Pu) ratio of about 1%, with separation of U, Pu and fission products.*
[9] *In this case, the increased productivity of industrial operation was factored into the final figure.*

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- The costs of operations now performed in the fuel cycle have been validated by industrial or semi-industrial[9] experience. For the purposes of this sensitivity study, these costs are considered to be known without any uncertainty margin. They include uranium mining, conversion and enrichment, PWR (UOx and MOx) fuel fabrication, FBR MOx (fertile and fissionable) fuel fabrication, spent fuel transportation and R'2 reprocessing. This category also includes waste transportation and disposal, which affect both the conventional cycle scenarios and the partitioning/transmutation scenarios in the same hypothetical manner.

- The cost of operations estimated from design studies but unconfirmed by industrial implementation are assigned an uncertainty margin.

- The cost of operations that have not been analyzed in detailed design studies are considered to have a very large uncertainty margin. These mainly include PWR UOx and FBR MOx actinide fuel fabrication costs.

To evaluate the effect of possible unit cost variations on the final result of the economic assessment, the results were recalculated to take into account the upper limit of the uncertainty margin (high estimate) and the lower limit of the uncertainty margin (low estimate) on the unit costs specific to partitioning and transmutation operations. All the other costs remained constant at their reference values. The results are indicated in the table below. Under the assumptions of this study, compared with the conventional fuel cycle scenarios, the partitioning/transmutation scenarios imply a cycle cost supplement ranging from 20% to 55% for the PWR scenario, and from 10% to 50% for the FBR scenario.

| Basis	$\dfrac{\text{RP1.1 - R2}}{\text{R2}}$		$\dfrac{\text{RP1.2 - R3}}{\text{R3}}$	
	Typical Year	Cumulative	Typical Year	Cumulative
High Estimate	54%	44%	52%	29%
Reference	33%	27%	28%	17%
Low Estimate	21%	17%	11%	7%

7 CONCLUSION

The study of different transmutation (Np and Am) scenarios have permit to show that it's possible to gain an important factor on the waste radiotoxicity generated by a reactor park and cycle plants. Compare with scenario R1 (open cycle), the gain is by a factor 40 to 100 beyond the first 100 years and according the cooling time. The gain is better when we use the fast reactors to transmute Np and Am. If we want to increase the gain, the separation of Np, Am and Cm must begin as soon as possible because in the future, the past will have an important weight for the evolution of the waste radiotoxicity. Transmutation of Am and Cm doesn't degrade the "cycle" inventory.

The average fuel cycle cost supplement resulting from the implementation of partitioning and transmutation of Pu, Np and Am (scenarios RP1-1 and RP1-2) above the cost of the reference scenarios (R2 and R3) in which only plutonium is recycled, should be approximately from 20% to 55% for the PWR scenario and 10% to 50% for the FR scenario.

The average cost supplements are slightly higher for a fuel cycle based on transmutation of the minor actinides in PWRs than for a cycle using fast reactors; however, the uncertainty margins on unit operating costs are such that this difference is probably not significant.

These results are based on relatively unfavorable hypotheses (implementation of complex processes, recycling of minor actinides in UOx fuel, reprocessing plant design). It is not unreasonable to expect that ongoing research and development work - notably in France under the SPIN program - will allow simpler processes to be designed in the future to achieve the same objectives.

URANIUM LOSSES FROM REPROCESSING
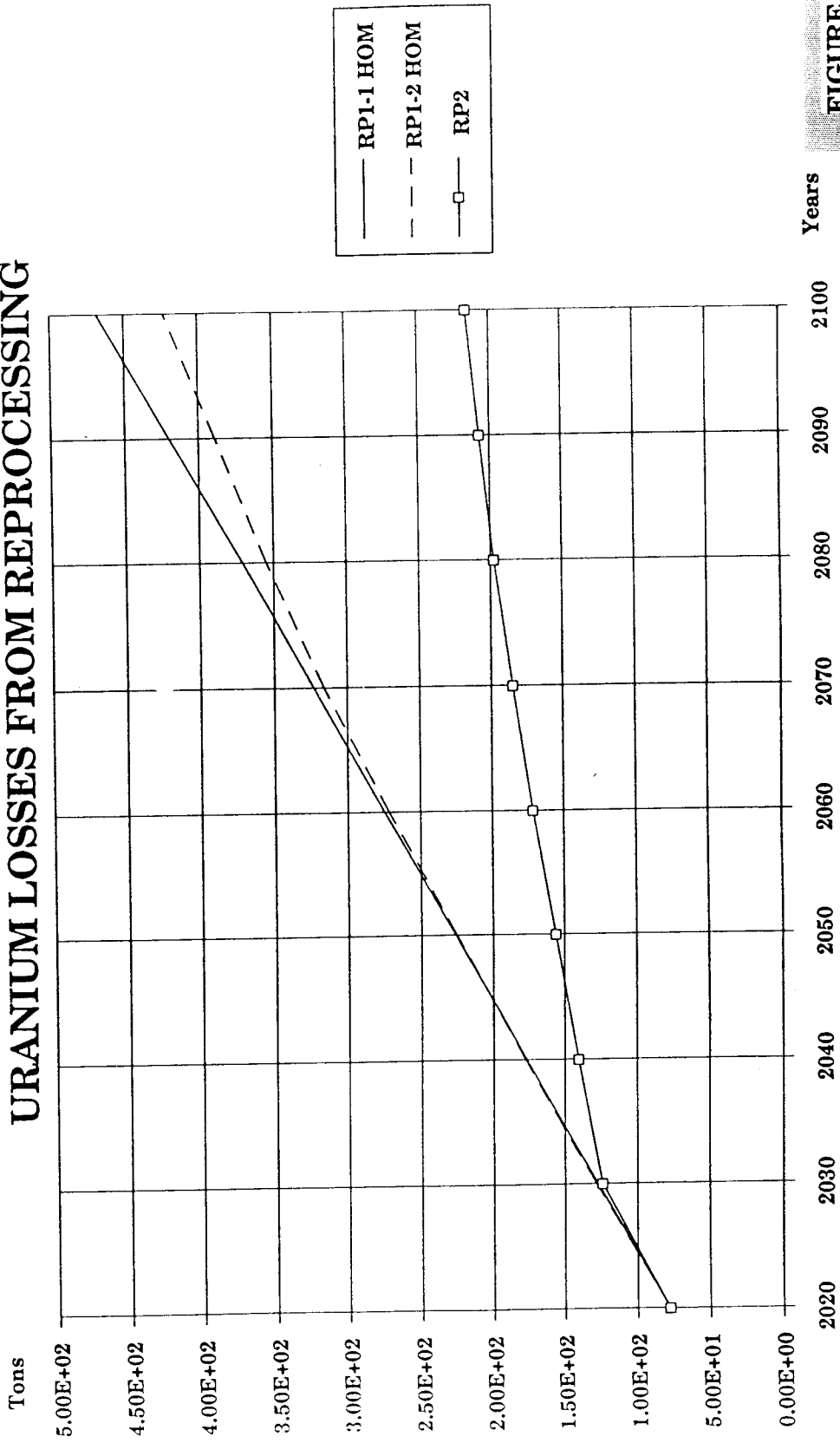
FIGURE 2

SCENARIO R2 : INITIAL PU CONTENT IN THE MOX-PWRS
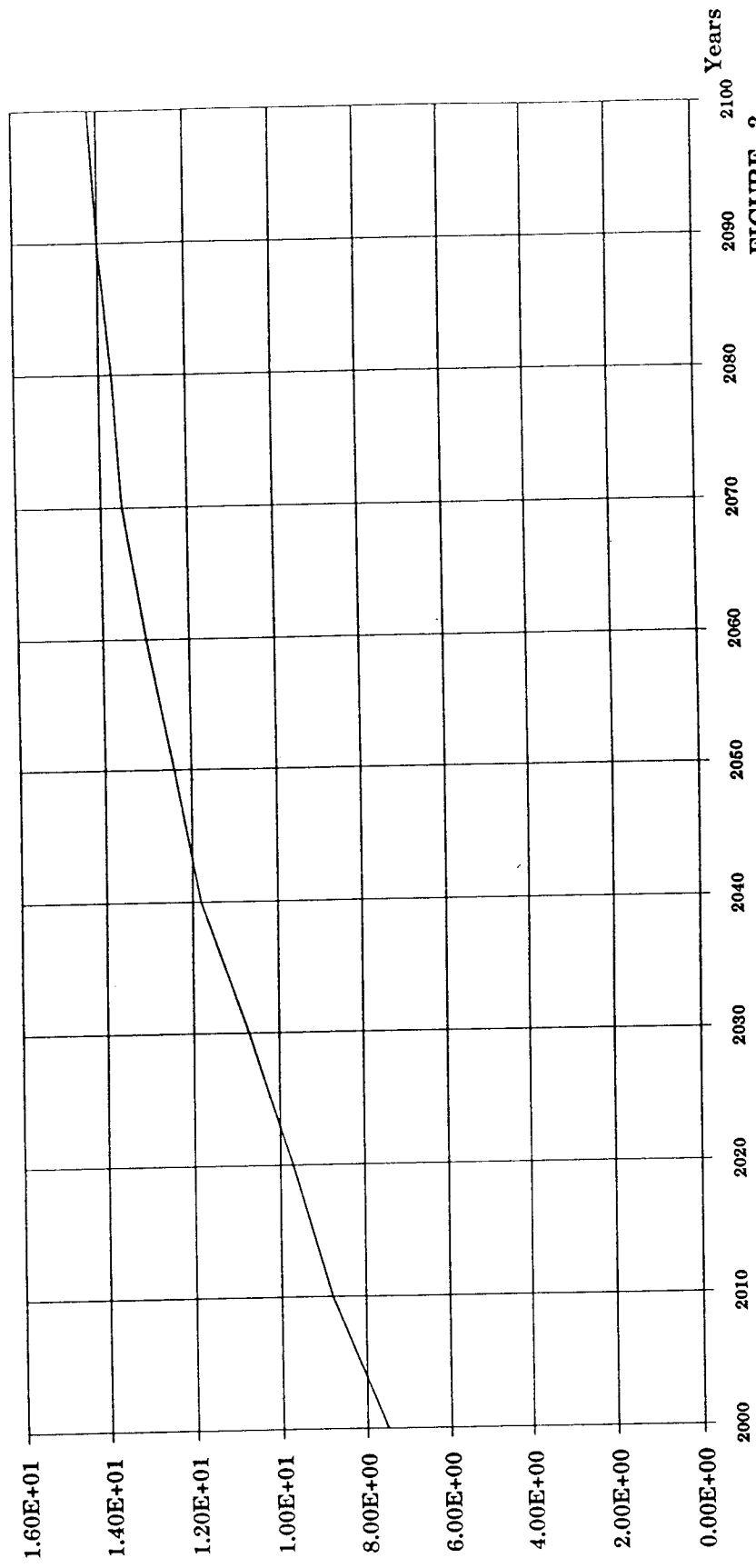
FIGURE 3

PLUTONIUM LOSSES FROM REPROCESSING

Tons

2.50E+01

2.00E+01

1.50E+01

1.00E+01

5.00E+00

0.00E+00

2020　2030　2040　2050　2060　2070　2080　2090　2100

Years

RP1-1 HOM
RP1-2 HOM
RP2
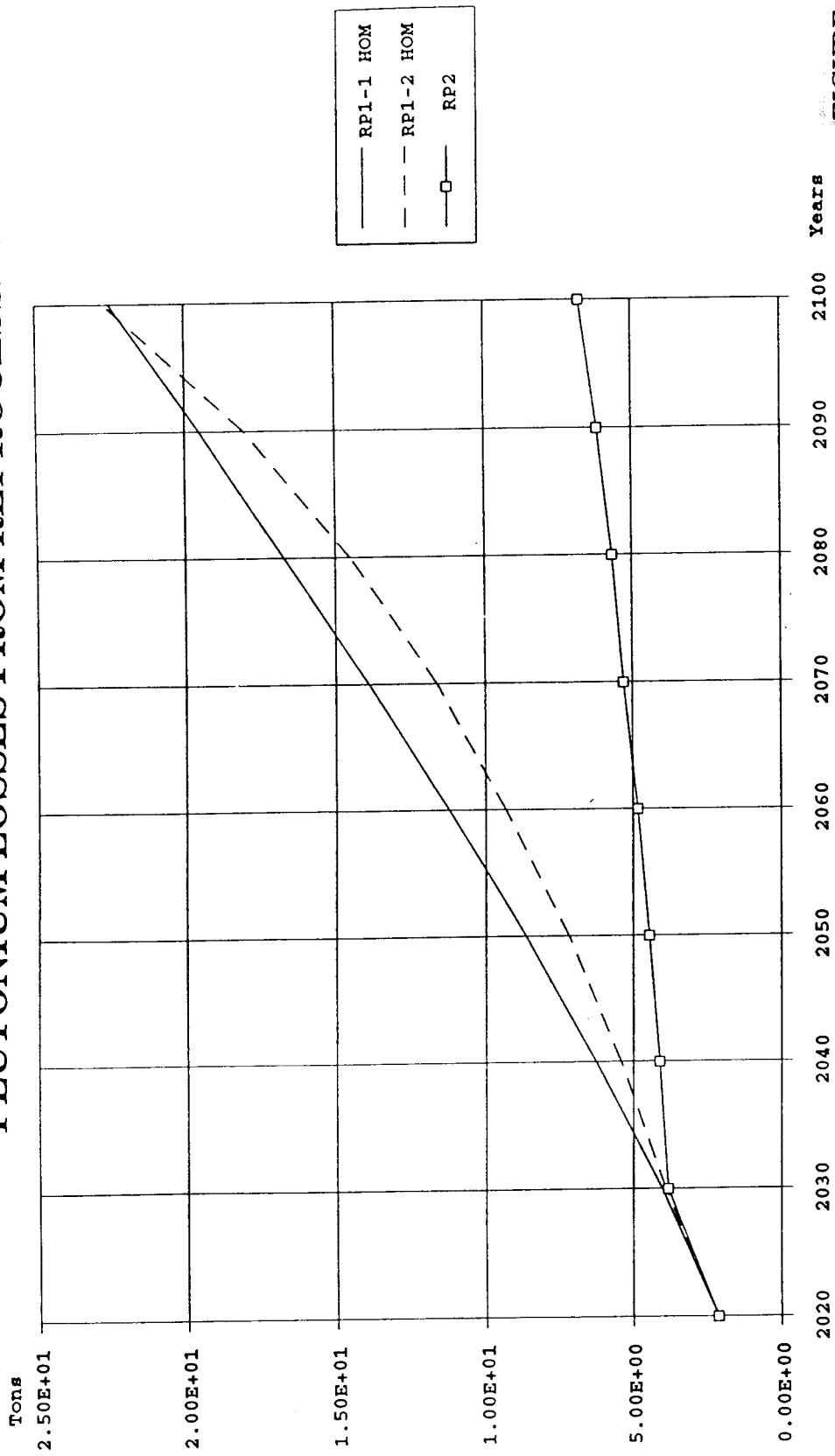
FIGURE 4

NEPTUNIUM FOR DISPOSAL
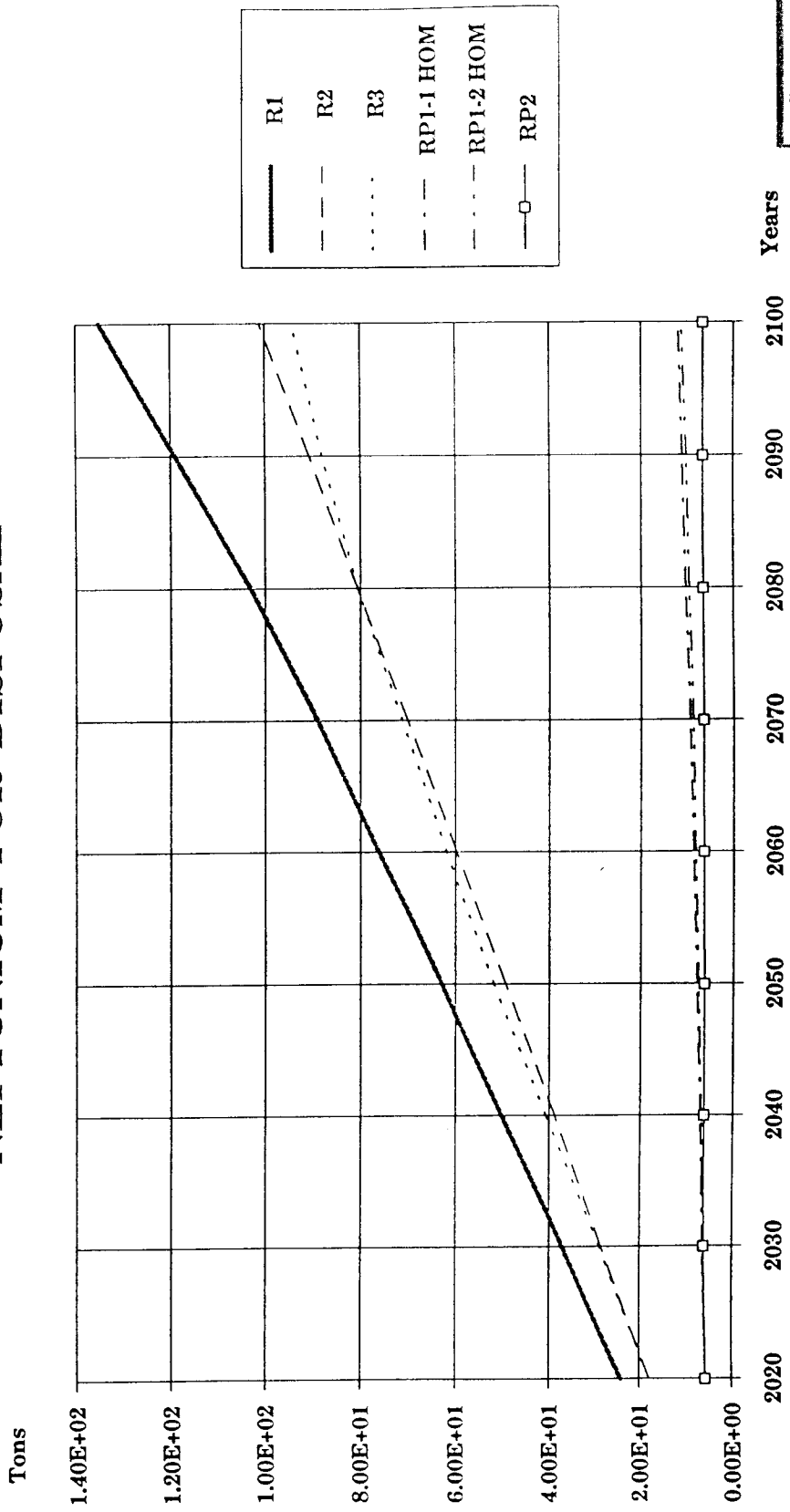
FIGURE 5

AMERICIUM FOR DISPOSAL

FIGURE 6

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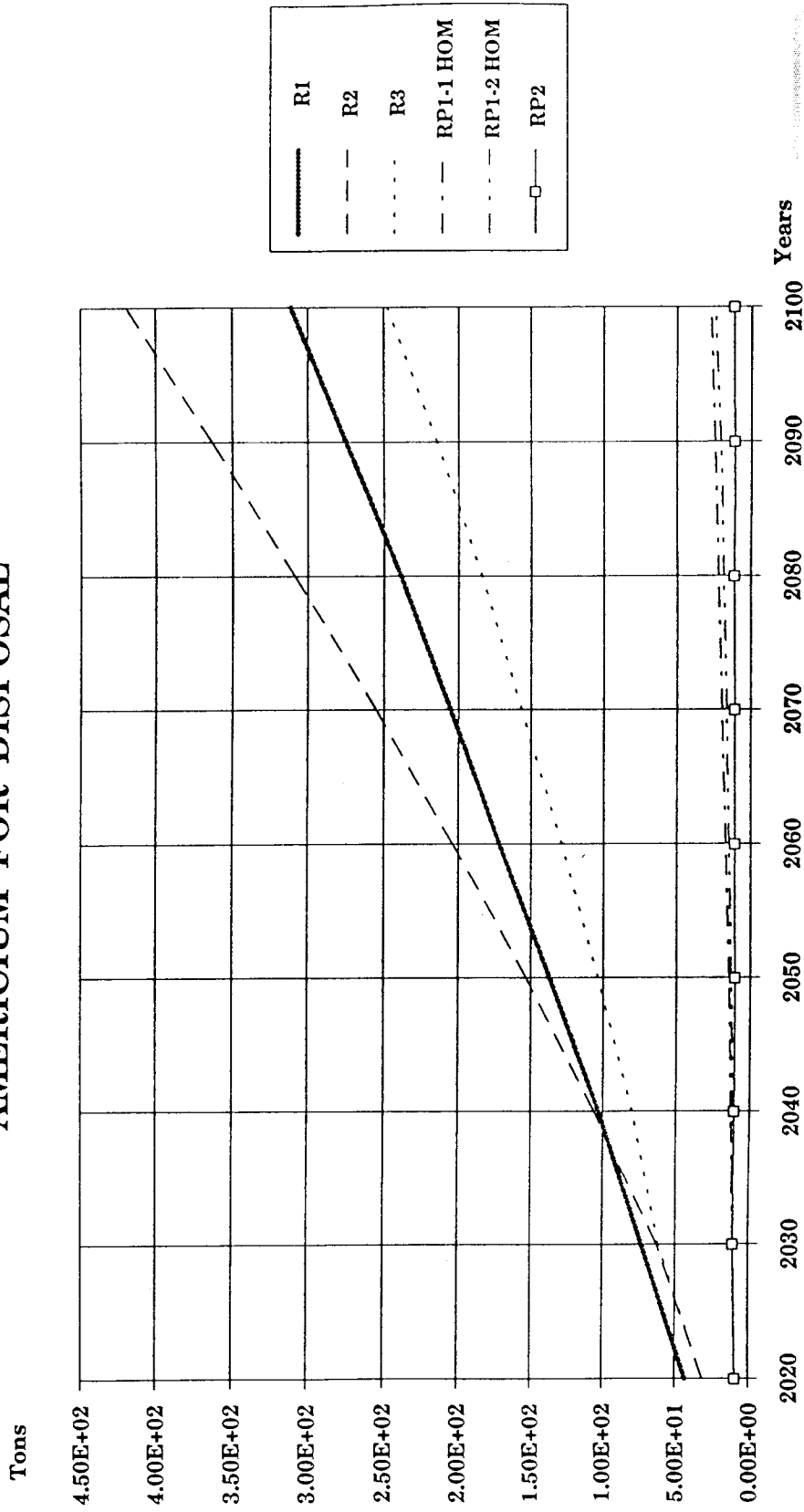
CURIUM FOR DISPOSAL

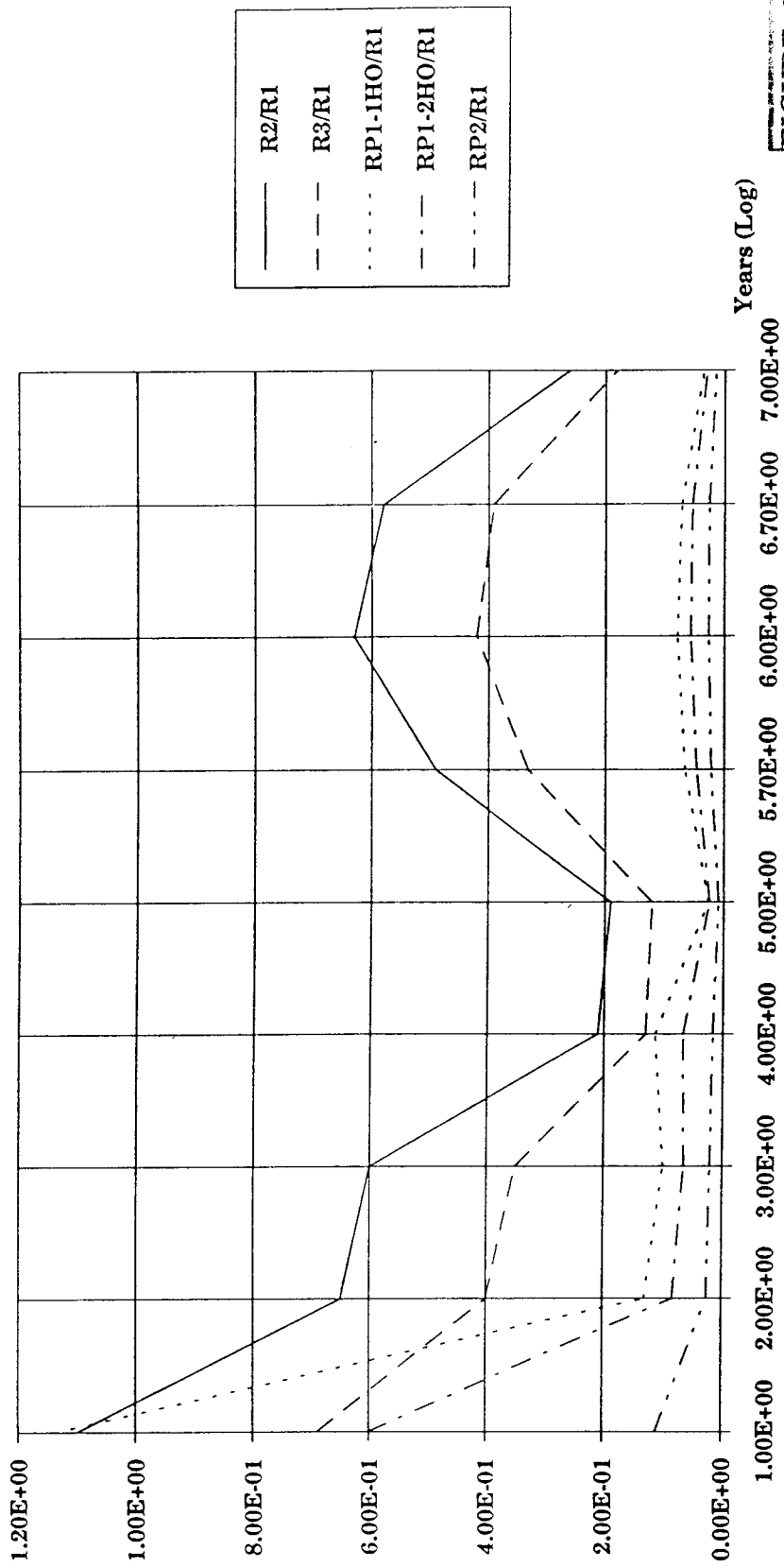FIGURE 7

140
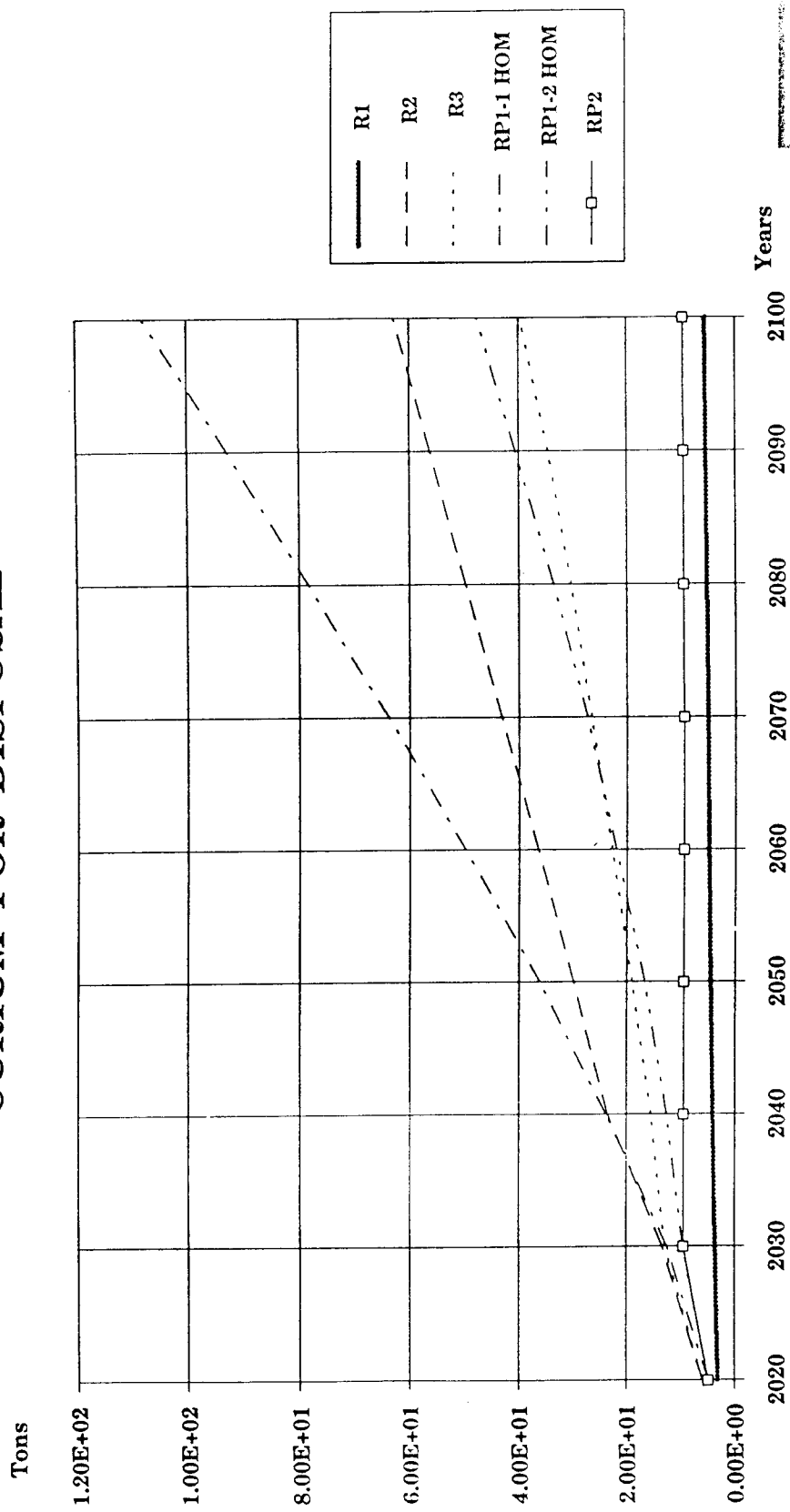
POTENTIAL INGESTION RADIOTOXICITY : "WASTE" INVENTORY

FIGURE 8

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POTENTIAL INGESTION RADIOTOXICITY : "CYCLE" INVENTORY

FIGURE 9

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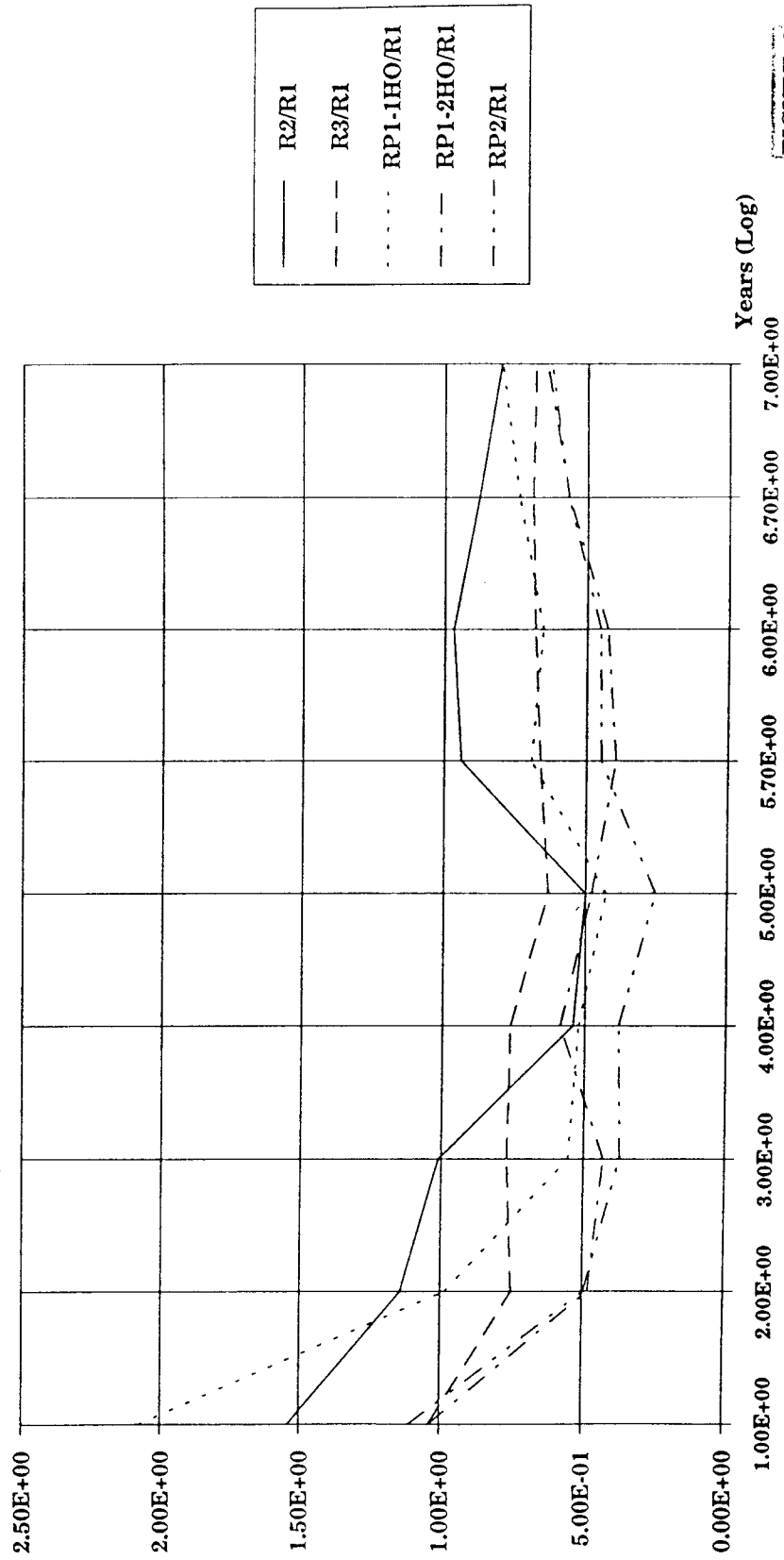
Référence :

(1) COSI, a simulation software for a pool of reactors and fuel cycle plants - Application to the study of the deployment of F.B.R.
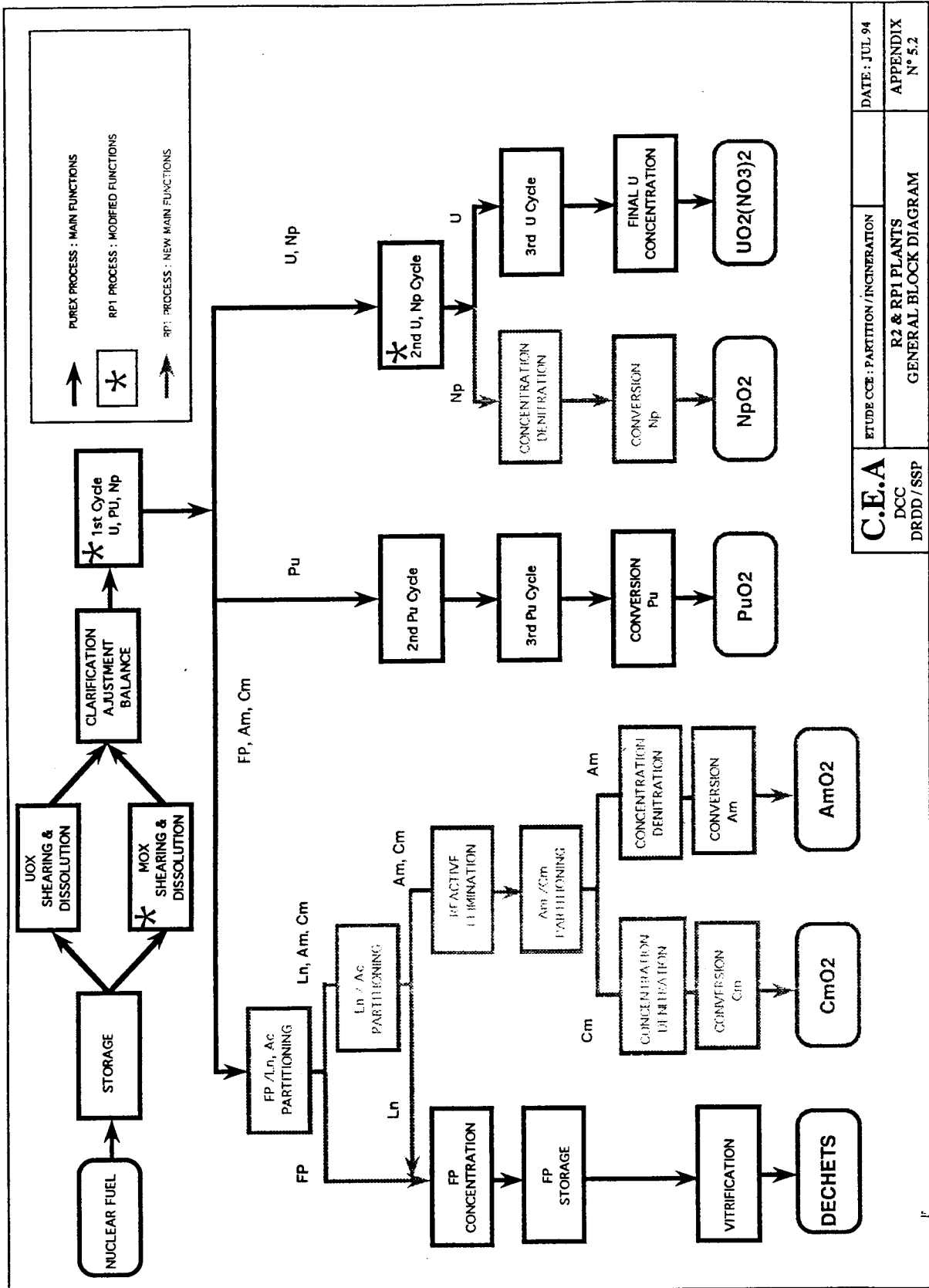
Log n° PA 185 KYOTO, Japan 1991

List of Appendices

Dissolution solution → oxidation NpV->NpVI

Modified PUREX Solv TBP → PUREX (cont'd) → Pu 99,5% yield

FP, Ln, Am, Cm ←

Extraction Ln+Am+Cm Solv Diamide → FP

Am+Cm+Ln

Séparation Ln/Am-Cm Solv DIDPA → Ln

Change media Solv Diamide

Separation Am/Cm Solvent DIDPA or HDDIBMP → Cm

Am

Concentration denitration

Oxalic Precipitation calcination

AmO2 95% yield

Cm → Concentration denitration → Oxalic Precipitation calcination → CmO2 95% yield

Vitrification

U+Np

oxidation NpV->NpVI

Modified PUREX (cont'd) → PUREX (cont'd) → U 99,5% yield

Np → Concentration denitration → Oxalic Precipitation calcination → NpO2 95% yield

Cm+Am → Concentration denitration → Oxalic Precipitation calcination → AmO2+CmO2 95% yield

C E A DCC/DRDD/SSP	ETUDE CCE : PARTITION / INCINERATION		DATE : JUL 94
	RP1 PROCESS FLOWSHEET		APPENDIX N°5.1

PUREX PROCESS : MAIN FUNCTIONS

RP1 PROCESS : MODIFIED FUNCTIONS

RP1 PROCESS : NEW MAIN FUNCTIONS

*

NUCLEAR FUEL

STORAGE

UOX SHEARING & DISSOLUTION

MOX SHEARING & DISSOLUTION *

CLARIFICATION AJUSTMENT BALANCE

1st Cycle U, PU, Np *

U, Np

Pu

FP, Am, Cm

2nd U, Np Cycle *

U

Np

3rd U Cycle

FINAL U CONCENTRATION

UO2(NO3)2

CONCENTRATION DENITRATION

CONVERSION Np

NpO2

2nd Pu Cycle

3rd Pu Cycle

CONVERSION Pu

PuO2

FP / Ln, Ac PARTITIONING

Ln, Am, Cm

Ln / Ac PARTITIONING

FP

Ln

Am, Cm

REACTIVE ELIMINATION

Am /Cm PARTITIONING

Am

Cm

CONCENTRATION DENITRATION

CONVERSION Am

AmO2

CONCENTRATION DENITRATION

CONVERSION Cm

CmO2

FP CONCENTRATION

FP STORAGE

VITRIFICATION

DECHETS

TYPICAL STEADY STATE BALANCE : RELATIVE COST SUPPLEMENT (RP1-1-R2)/R2TOTAL
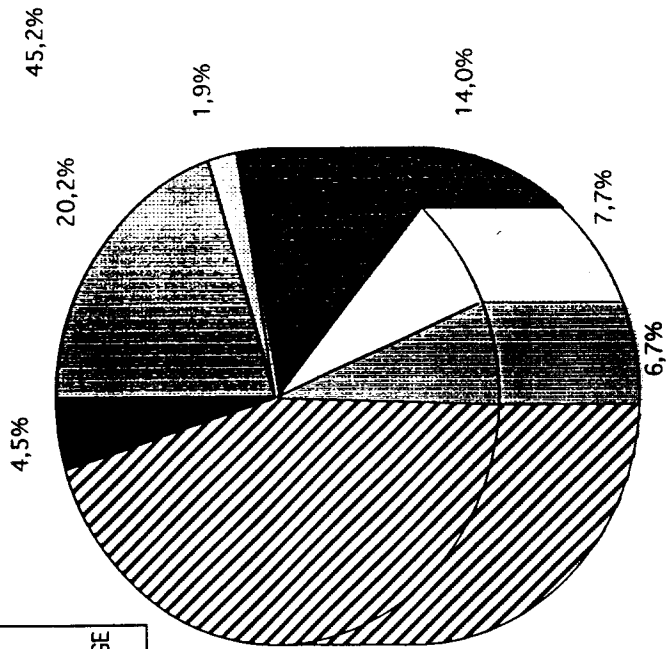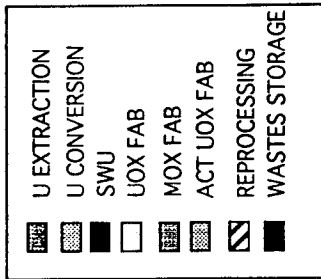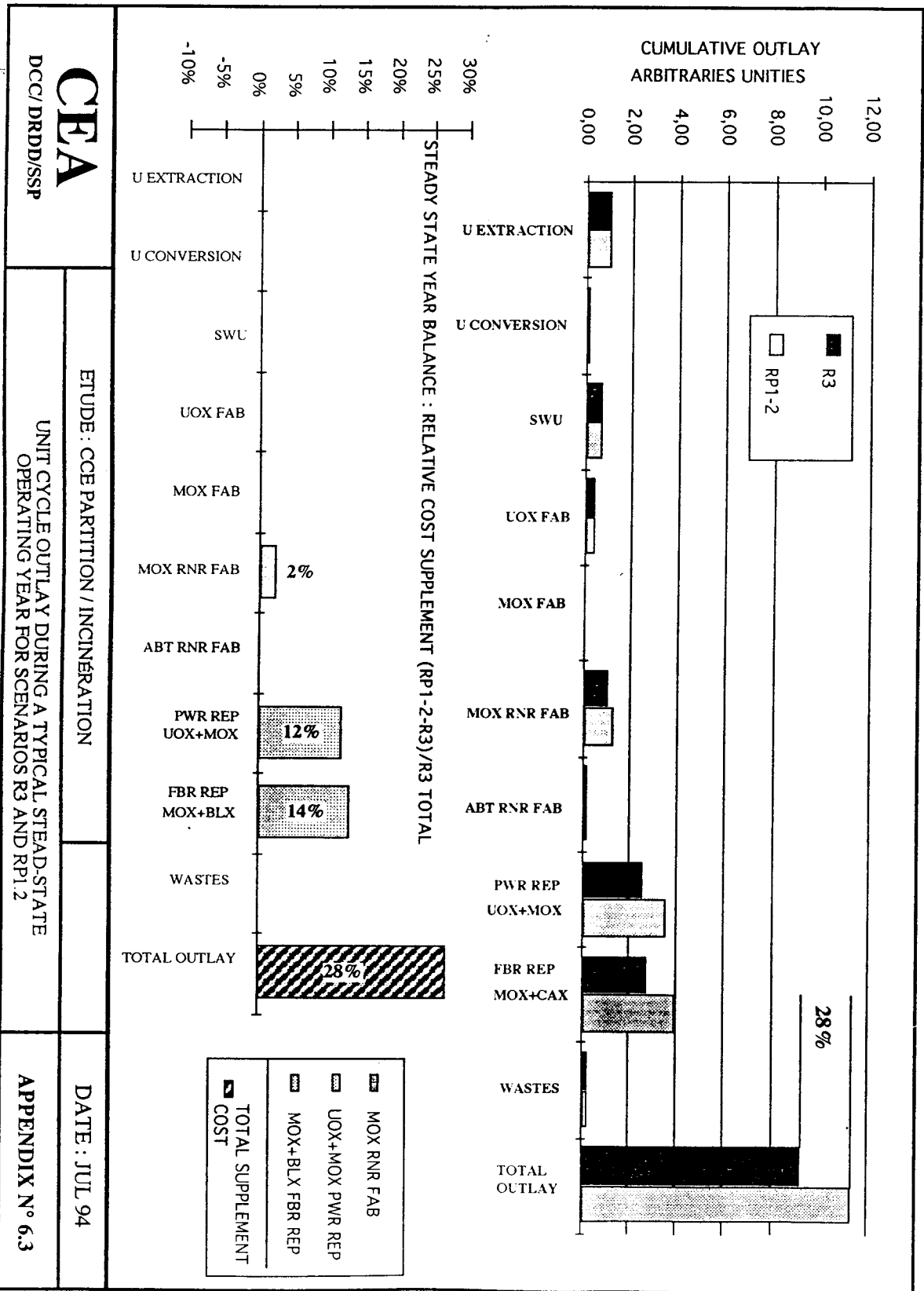
ETUDE CCE : PARTITION / INCINERATION	DATE : JUL 94
UNIT CYCLE OUTLAY DURING A TYPICAL STEADY-STATE OPERATING YEAR FOR SCENARIOS R2 AND RP1-1	
CEA DCC/ DRDD/SSP	APPENDIX N° 6.1

Legend:
- U EXTRACTION
- U CONVERSION
- SWU
- UOX FAB
- MOX FAB
- ACT UOX FAB
- REPROCESSING
- WASTES STORAGE

RP1-1

17,1%
1,6%
12,6%
3,4%
2,5%
5,0%
12,7%
45,2%

R2

20,2%
1,9%
14,0%
4,5%
7,7%
6,7%
44,8%

CUMULATIVE OUTLAY
ARBITRARIES UNITIES
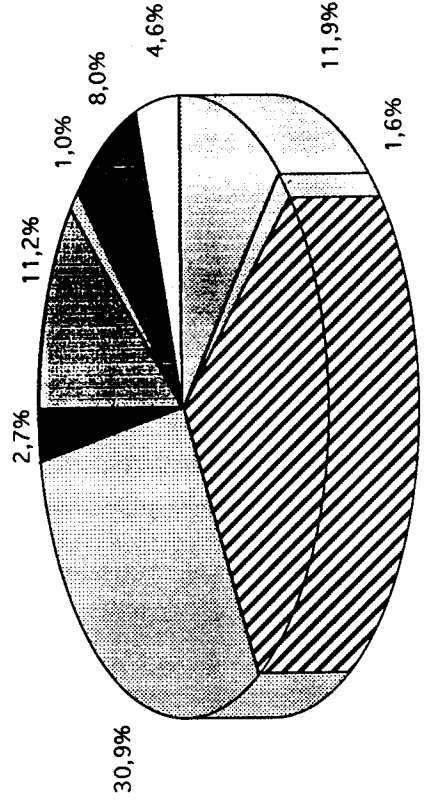
STEADY STATE YEAR BALANCE : RELATIVE COST SUPPLEMENT (RP1-2-R3)/R3 TOTAL
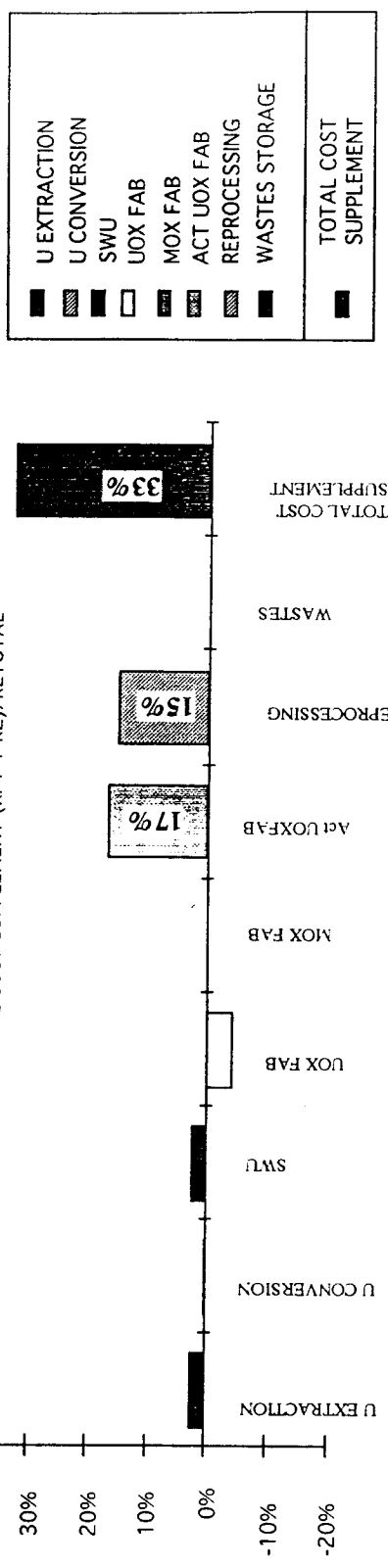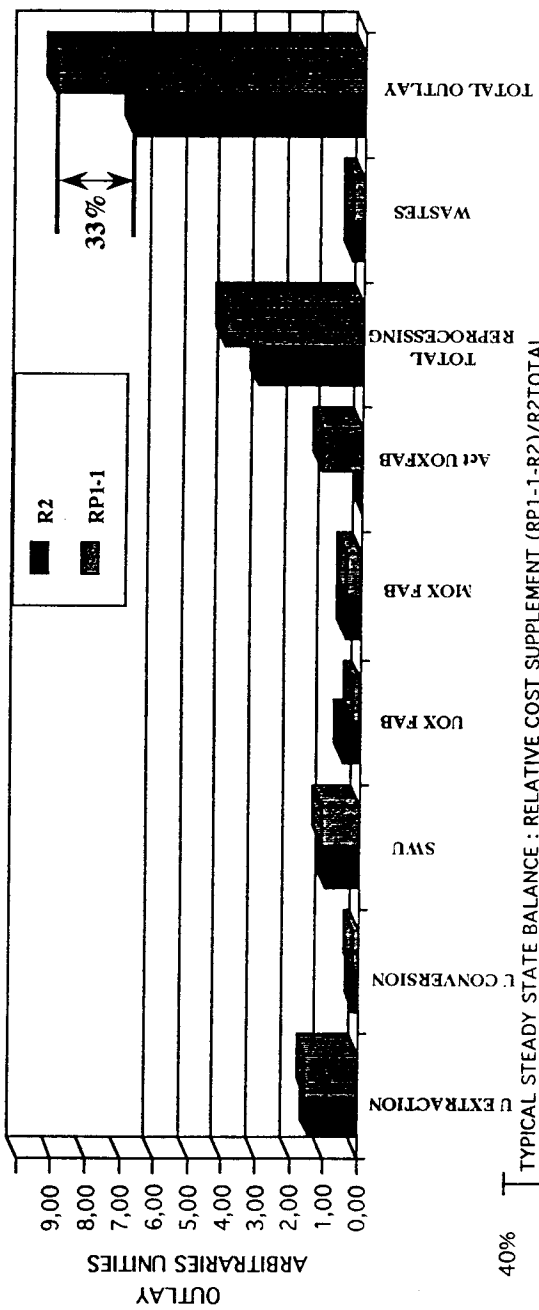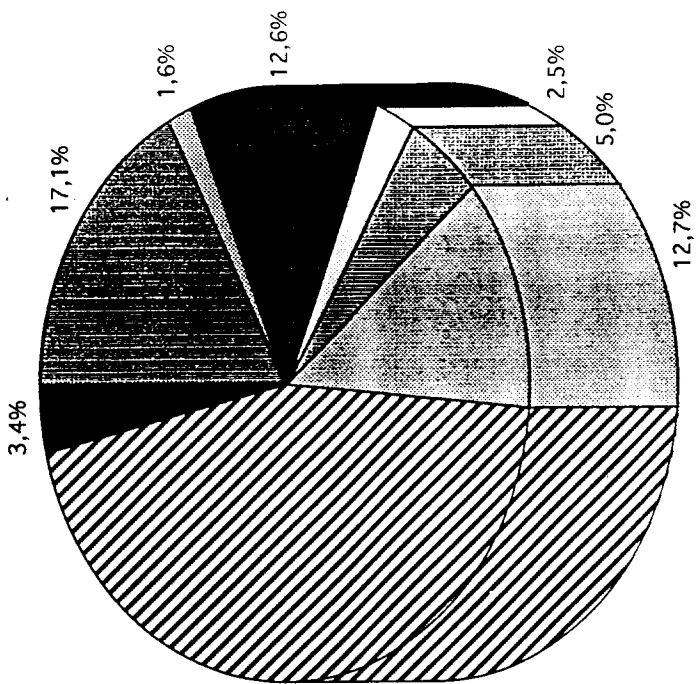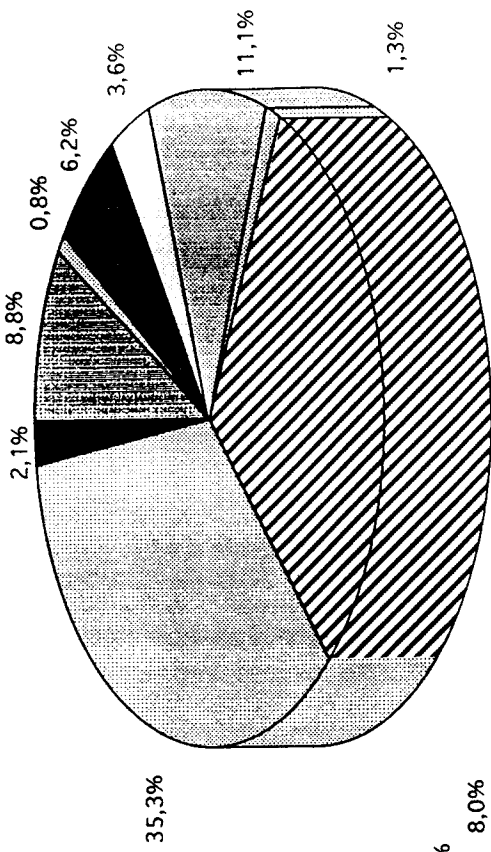
Legend:
- U EXTRACTION
- U CONVERSION
- SWU
- UOX FAB
- MOX PWR FAB
- MOX FBR FAB
- BLT FBR FAB
- PWR REP
- FBR REP
- WASTES STORAGE

RP1-2

3,6%
6,2%
0,8%
8,8%
2,1%
11,1%
1,3%
30,9%
35,3%

R3

4,6%
8,0%
1,0%
11,2%
2,7%
11,9%
1,6%
28,1%
30,9%

CEA DCC/DRDD/SSP	ETUDE CCE : PARTITION / INCINERATION	DATE : JUL 94
	UNIT CYCLE OUTLAY DURING A TYPICAL STEADY-STATE OPERATING YEAR FOR SCENARIOS R3 AND RP1.2	APPENDIX N° 6.4

Systems Study on Partitioning and Transmutation at JAERI

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T. Ogawa, and H. Yoshida

Japan Atomic Energy Research Institute
Tokai-mura, Naka-gun, Ibaraki-ken 319-11 Japan

Under the framework of Japanese OMEGA program, the Japan Atomic Energy Research Institute (JAERI) has studied P-T technologies aiming at a goal to provide a different HLW management from geological disposal. Technology of the proposed P-T system is based on the double stratum fuel cycle concept. The partitioning process has a potential capability to separate long-lived nuclides from HLW. The dedicated transmutation systems have a potential capability to burn minor actinides efficiently. Technology results so far obtained are sufficiently encouraging. P-T system requiments are discussed in view of risks due to P-T. The reduction of the risks demands a high transmutation rate, a high burnup and a low leakage factor.

1. INTRODUCTION

One of the most important problems with nuclear energy is the management of high-level radioactive waste (HLW) arising from the reprocessing of spent nuclear fuel. The hazard potential of actinides and fission products in the HLW is high due to their radioactivity. Of particular concern are the nuclides with very long half-life whose hazard remains high for millions of years. The major candidate scheme for the long-term waste management in most nuclear countries is the permanent disposal of unpartitioned HLW, or unreprocessed spent fuel, into a stable geological formation to isolate them from the human environment. There is, however, concern about the uncertainties in the long-term performance of an underground repository.

To mitigate the issues in the present waste management scheme, partitioning and transmutation (P-T) technology can play various roles according to the stages of advancement of the technology, such as a supporting technology by reduction of waste volume and heat generation, a complementary technology by mitigation of natural barrier uncertainty, and a new technology different from the geological disposal.

Under the framework of the long-term research and development program on P-T (OMEGA program) in Japan, the Japan Atomic Energy Research Institute (JAERI) has studied P-T technologies aiming at an ambitious goal to provide a different HLW management from geological disposal. The technologies include a partitioning process to separate HLW into four element groups together with minor-actinides group, and two different kinds of dedicated transmutation system; actinide burner fast reactor and proton accelerator-based transmutation system, both of which use fast neutrons and have comparable transmutation capabilities.

The proposed P-T system is based on the concept of double stratum fuel cycle, where the fuel cycle for partitioning and transmutation (P-T cycle) can be separated completely from the conventional commercial fuel cycle for power reactors. Technologies of the proposed P-T system are presented. The leakage of target nuclides in separation process and fabrication process has a very negative impact on the effectiveness of the P-T. The reduction of the overall leakage demands both a high burnup and a low leakage factor. Data needs and R&D issues to achieve the goal are discussed.

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2. JAERI R&D ON PARTITIONING AND TRANSMUTATION UNDER THE OMEGA PROGRAM

JAERI is carrying out R&Ds for advanced partitioning technology, transmutation with burner fast reactor, and proton accelerator-based transmutation under the OMEGA program[1]. Aims of the program are to widen options of future waste management and to explore the possibility to utilize HLW as useful resources. The program is conceived as a research effort to pursue benefits for future generations through the long-term basic R&D, and is not to seek a short-term alternative for established or planned fuel cycle back-end policies.

An advanced partitioning process has been developed to separate elements in HLW into four groups; transuranium elements (TRU), Sr-Cs, Tc-platinum group metals and the other elements.

Conceptual design studies have been carried out for actinide burner fast reactors and accelerator-based transmutation systems. To support the design studies of these transmutation systems, several basic researches are in progress or planned. These include the nuclear data measurements of minor actinides, thermodynamic and thermophysical studies on minor-actinide compounds, spallation integral experiment, and thermal-hydraulic experiment.

Construction of a high-intensity proton linear accelerator with an energy of 1.5 GeV and an average beam current of 10 mA, named the Engineering Test Accelerator (ETA), has been proposed to perform various engineering experiments and demonstration tests for accelerator-based transmutation system. R&D work for the full-scale mockup of ETA front-end, called the Basic Technology Accelerator (BTA, 10 MeV-10 mA), is in progress.

3. DESCRIPTION OF TECHNOLOGY

3.1 Concept of Double Stratum Fuel Cycle

JAERI is aiming to develop an advanced partitioning process of TRUs and long-lived fission products from HLW and dedicated systems for their subsequent transmutation. The dedicated transmutation system is specially designed to burn minor actinides efficiently in a very hard neutron energy spectrum and high neutron flux. In this context, JAERI has been pursuing the concepts of accelerator-driven system and actinide burner reactor as dedicated transmuter, rather than the transmutation scenarios using commercial power reactor.

With the dedicated transmuter, the troublesome MA from the waste management view point could be confined in a P-T fuel cycle for transmutation, which forms the second stratum separated from the first one of conventional commercial fuel cycle for power generation. The concept of double-stratum fuel cycle is illustrated in Fig. 1. It could offer several advantages over the recycling of MA to the commercial power reactor. These are higher transmutation rate, effective confinement of MAs, and no impact on the power reactor operation. There will be no additional shielding and cooling requirements to the existing commercial fuel cycle facilities.

3.2 Four-Group Partitioning Process

An advanced partitioning process has been developed to separate elements in HLW from reprocessing of spent fuel into four groups (TRU, Tc-platinum group metals, Sr-Cs, and the others)[2]. The flow sheet of the four-group partitioning process is shown in Fig. 2.

In this process, Am and Cm are extracted with the solvent diisodecylphosphric acid (DIDPA), and Np is extracted with addition of hydrogen peroxide. The DIDPA solvent is also be applied to the separation of Am and Cm from rare earths. The separation of Tc is made by adsorption with an active carbon column. Tc is adsorbed from HLW adjusted to 0.5 M nitric acid concentration and adsorbed Tc is eluted from the column by using alkaline potassium thiocyanate solution (2M KSCN-4M NaOH) as eluant. Sr and Cs are separated by adsorption with the inorganic ion exchangers of zeolite and titanic acid. Iodine will be recovered from the reprocessing offgas by adequate sorption methods.

3.3 Transmutation Systems

(1) Accelerator-based Transmutation System

Two types of accelerator-based transmutation system concept have been proposed; solid system and molten-salt system. In either system, a minor-actinide loaded subcritical core is driven by a high-intensity proton linear accelerator and uses fast neutrons to burn actinides efficiently.

The conceptual design of the solid target/core system [3] is shown in Fig. 3. The design of the solid system is based on the status of Na-cooled fast reactor technology. The fuel is actinide alloy or nitride clad in ODS steel. A high-energy proton beam is injected through beam window into the spallation target at the center of the core. The target consists of multi-layers of solid tungsten, which is designed to maximize the number of emitted neutrons and to flatten the axial neutron flux distribution. The heat generated in the target and core is removed by Na flow, and is recovered by a steam turbine plant into electricity. The solid system, having an actinide inventory of 3160 kg and an effective neutron multiplication factor of 0.89, produces 820-MWt thermal power with a 1.5 GeV-39 mA proton beam.

Another innovative option is the molten-salt target/core system [4]. Chloride salt (64NaCl-5PuCl$_3$-31MACl$_3$) is used both for fuel and for target material, and at the same time it also serves as primary coolant. This significantly simplifies the target/core configuration, as schematically shown in Fig. 4. A proton beam is injected into the central target/core region through the beam window. Intermediate heat exchangers and salt pumps are installed in the annular region around the internal reflector that surrounds the target/core region. This in-vessel heat exchanger design minimizes the total actinide inventory in the system. The molten state of the fuel salt offers the possibility of continuous on-line processing of actinides and reaction products.

The molten-salt system has an actinide inventory of 5430 kg and an effective neutron multiplication factor of 0.92, and produces 800-MWt thermal power with a 1.5 GeV-25 mA proton beam.

(2) Actinide Burner Reactor

The guidelines for designing an actinide burner reactor (ABR) as a dedicated transmuter are; minor actinide as the major fuel constituent, very hard neutron spectrum, very high neutron flux, and dry reprocessing of actinide fuel.

Two types of ABR concept were previously proposed [5], namely Na-cooled metallic fuel ABR and He-cooled particle-bed ABR. Design modification has been attempted to improve the safety characteristics [6]. To increase the effective delayed neutron fraction, Pu in the fuel is replaced with enriched U (90% enrichment). To avoid the large positive void coefficient for Na cooling, liquid Pb is used as coolant instead of Na. Also, metallic fuel is replaced with nitride fuel because of relatively low melting point and low thermal conductivity of metallic minor-actinide fuel.

The Pb-cooled ABR (L-ABR) has ductless, pin-bundle type fuel assemblies (Fig. 5). The plant will be consist of six reactor modules, each having a thermal power of 180 MWt and a minor-actinide inventory of 573 kg.

The He-cooled particle-bed ABR (P-ABR) has fuel elements of particle bed formed in the annular space of concentric porous frits (Fig. 6). Fuel particles are directly cooled with He gas. Very high power density is obtained since a large heat transfer surface per volume is very effective for heat removal. A fuel particle consists of actinide nitride microsphere and thin TiN coating layers. P-ABR has a thermal power of 1200 MWt and a minor-actinide inventory of 1865 kg.

3.4 Actinide Fuel Fabrication and Reprocessing

For a dedicated actinide transmuter, a fuel form with a high heavy-metal atom density is desirable in view of the neutron spectrum and the fuel cycle efficiency. One of the promising candidate fuels is the mixed mononitride: (U, Np, Pu, Am, Cm)N. The mutual solubility of the actinide mononitrides is expected to be sufficiently high since they have the same NaCl-type structure with lattice parameters which are close to each other. Fabrication process of the nitrides may follow that of UN and PuN. Figure 7 shows a proposed fuel cycle scheme of the TRU nitride fuel.

The nitrates of the minor actinides (MA) recovered in the partitioning of HLW could be readily converted to solid micorspheres consisting of the actinide oxides and carbon with a sol-gel process. In a sol-gel process being studied at JAERI, an ammonia donor, hexamethylenetetramine (HMTA), is added to the nitrate solution. By heating the liquid droplets, HMTA decomposes to form ammonia which causes gelling of the droplets. The solid microsphere is then converted to the nitride by the carbothermic reduction. The technique to fabricate the nitrides of transplutonium elements by the carbothermic reduction would be similar to those for UN and PuN, since the Gibbs free energy of formation is estimated to be roughly the same [7].

For reprocessing the irradiated MA nitride fuel, a pyrochemical process with the molten-salt electrorefining could be applied [8]. The procedure would be very similar to that of the molten-salt electrorefining

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of alloy fuels, which has been developed at Argonne National Laboratory[9]: the fuel is anodically dissolved into a molten LiCl-KCl salt, and the actinide metals are recovered on either solid cathode or liquid-Cd cathode. The liquid-Cd cathode will be used for recovering the TRU elements.

4. TECHNOLOGY RESULTS

4.1 Partitioning

The four-group partitioning process have been developed through laboratory-scaled experiments since 1985. In the mixer-settler experiments with the actual HLW, it was demonstrated that more than 99.99% of Am and Cm were recovered with the extractant DIDPA. It was also verified that more than 99.95% of Np was extracted in a series of experiments with the synthesized HLW. The separation of Tc was demonstrated with a high recovery over 99% in the tests with synthesized HLW.

These experiments with actual or synthesized HLW indicate that the proposed partitioning process has great promise of realizing the target separation efficiencies for the important elements in HLW without undue difficulty. The partitioning process will be tested with actual HLW at the Nuclear Fuel Cycle Safety Engineering Research Facility (NUCEF) that was constructed at JAERI Tokai, and will come into hot operation in 1995. The chemical engineering test mainly with a simulated HLW is also planed to start in 1996. Fundamental data for designing a partitioning pilot plant will be accumulated consistently until about 2000.

4.2 Transmutation

The proposed accelerator-based transmutation systems have hard neutron spectra with core averaged neutron energies around 700 keV. The core averaged neutron flux is 4×10^{15} n/cm^2/s for the solid system and 5.9×10^{15} n/cm^2/s for the molten-salt system. The actinide burnups per 1 GWt per year in the solid system and molten-salt system are 360 kg (10 %/y) and 370 kg (6 %/y), respectively. The solid and molten-salt systems produce 246-MWe and 360-MWe electricity, respectively. The system can provide enough electricity to drive its own accelerator.

As the first step toward the intense proton accelerator development, the four main accelerator components, ion source, radio-frequency quadrupole (RFQ), drift-tube linac (DTL) and radio-frequency (RF) source, have been developed[10]. The beam acceleration test with the ion source and RFQ was made successfully at a beam current of 52 mA (peak) and 5% duty. High power tests of the DTL hot-test model began in 1994. As the next development step, the design work for the BTA is in progress.

Significantly hard neutron spectra are achieved for the proposed ABRs, core averaged mean neutron energies being around 700 keV. The core averaged neutron flux is 3.1×10^{15} n cm^2/s for the lead-cooled ABR (L-ABR) and 5.9×10^{15} n/cm^2/s for the particle-bed ABR (P-ABR). The minor-actinide burnups per 1 GWt per year in the L-ABR and P-ABR are 190 kg and 200 kg, respectively. These values are decreased by 30 to 40% from those of the original ABR designs because of the contribution of U-235 fission. The minor-actinide burnup ratio is 11 %/cycle for the L-ABR with a cycle length of 550 days and 13 %/cycle for the P-ABR with a cycle length of 300 days.

4.3 Actinide Fuel Fabrication and Reprocessing

Studies on actinide fuel fabrication and reprocessing technology have been made. PuN and NpN have been successfully fabricated with a carbothermic reduction process, where the mixture of the oxide and carbon is heat-treated in flowing $N_2 + H_2$[11,12]. The study of the chemical vapor deposition of low-density TiN (buffer) coating for the nitride particle fuel is underway; initial attempts to deposit the low-density TiN on stationary substrates have been successfully made. The study on the sol-gel processing of the nitride fuel is also being made.

5. GOAL OF PARTITIONING AND TRANSMUTATION

Partitioning and transmutation technology can play various roles in HLW management depending on the stages of technology development. So, the goal of P-T will vary with P-T introduction timeframes of near term, mid-term, and long term. At present, technical feasibility of P-T is already established to reduce the volume and heat generation of HLW to some extent. In the near term, P-T technology could support the HLW

management in improving the safety of geological disposal through the reduction of the HLW volume and heat generation. In the mid-term, P-T can be a complementary technology by mitigation of concerns about natural barrier uncertaity, leading to further improvement in the long-term radiological safety assurance. The long-term goal of P-T is the most ambitios and challenging. The P-T technology can be a new waste management technology alternative to the geological disposal.

In the preliminary strategic study [13], priority of nuclides to be separated for transmutation was determined and target separation efficiencies were preliminarily defined for minor actinides and fission products in HLW.

Priority of nuclides, determined based on the half-lives and radioactive toxicity of nuclides in HLW, is as follows:

1; TRU nuclides including residual Pu
2; Tc-99 and I-129
3; Sr-90 and Cs-137
4; Zr-93, Cs-135 and Sm-151
5; long-lived activation products (C-14, Ni-59 etc.)

If neutron reaction is applied to transmute long-lived nuclides into shorter-lived or stable nuclides, TRU can be effectively transmuted to fission products by fission reaction, and Tc-99 and I-129 can be transmuted to stable nuclides by neutron capture reaction. However, it is rather difficult to transmute the other nuclides with lower priority by applying neutron reaction.

If potential radioactive toxicity should be balanced before and after reactor operation, the potential radioactive toxicity allowed for residual waste including long-lived nuclides should be equivalent to that of uranium ore. Target separation efficiencies were preliminarily defined for TRUs and fission products in HLW, comparing their radioactive toxicities with that of uranium ore. The target separation efficiencies defined for important elements from HLW are as follows;

Pu:	99.9%,	Np:	99.5%,	Am:	99.99%,
Cm:	99.9%,	Tc & I:	99.0%,	Sr &Cs:	99.9%.

The laboratory-scaled experiments at JAERI shows that the proposed partitioning process has high possibility to realize these target separation efficiencies.

6. P-T SYSTEM REQUIREMENTS

Performance of P-T systems should meet several requirements to achieve a goal of P-T. These requirements will be quantified through the systems and strategy studies including cost and benefit evaluation of P-T systems. Here, the P-T system requirements are discussed in a qualitative manner.

First, the system should has the sufficient capability of reducing the overall risks including both long-term and short-term risks caused by radiological toxicity of waste, and at the same time it should be economically feasible. This requires large transmutation capacity and rate together with reasonably good energy balance of the transmutation plant.

The transmutation rate should be large so as to make the time period required for transmutation (effective half-life) much shorter than the half-life of natural decay of the target nuclides, and also than the plant lifetime. Thus, the largest possible transmutation rate is desirable for the effective reduction of hazard. The transmutation capacity should be as large as or larger than the production of target nuclides. The number of power reactors supported by a transmutation plant is proportional to its transmutation capacity.

In case of accelerator-based transmutation, the accelerator consumes considerably large electric power to deliver the beam to the plant. The accelerator electricity input to be required for transmutation should be sufficiently small than the total electricity output of power reactors that are supported by the transmutation plant. The positive energy balance may be preferred for the transmutation plant, that is, it is to be desired that power output from the transmutation plant is larger than the power required to operate the plant including the accelerator.

It is impossible to achieve a complete transmutation in a single irradiation, target nuclides should be recycled many times to burn them up sufficiently. Each time the target nuclides pass through one cycle, it is also impossible to recover untransmuted nuclides completely in separation process. There will be some unavoidable leakage of untransmuted nuclides to various secondary waste streams. This leakage have a very negative impact on the effectiveness of P-T. The reduction of the overall leakage demands both a high burnup and a low leakage. The higher burnup in a single cycle needs the fewer cycles to burn up a given amount, and thus the less untransmuted nuclides will be lost.

.These requirements may be explained to some extent in terms of the radiological risks associated with the P-T system. The long-term risks can be assumed to be proportional to the leakage of target nuclides and the short-term risks, to the target nuclide inventory and resident time in transmuters and fuel cycle facilities. Here, the discussion is limited to TRU nuclides and they are dealt with collectively.

Risk due to waste leakage R_W is proportional to the total leakage factor. For batch processes repeated infinitely in a P-T fuel cycle shown in Fig. 8, R_W is approximately given as

$$R_W \propto \alpha_P + (1 - \alpha_P)[(\alpha_F + \alpha_S(1 - \varepsilon)]/[\varepsilon + (\alpha_F + \alpha_S)(1 - \varepsilon)]$$ (1)

where, ε is the burnup per cycle, α_P, α_F, and α_S are the leakage factor in partitioning of HLW, the leakage factor in fuel fabrication, and the leakage factor in separation, respectively. Assuming $\alpha_P = 0.1\%$, $\alpha_F = 0.15\%$, $\alpha_S = 0.1\%$, and $\varepsilon = 18\%$, the total leakage factor becomes about 1.4%. These assumed values represent the target loss factors of current process technology development. To achieve the most challenging goal of P-T, it is necessary to develop the technology to further reduce the loss and to recover the loss so that the eventual leakage to secondary waste streams can be minimized.

For continuous process for a liquid fuel system shown in Fig. 9, R_W becomes

$$R_W \propto \alpha_P + (1 - \alpha_P)\alpha_S/(\varepsilon + \alpha_S)$$ (2)

where, constant fuel flow rate through processing is assumed and the cycle length is defined as the ratio of the fuel inventory in the system to the fuel flow rate through processing. The liquid fuel system has an advantage of complete avoidance of fuel fabrication process that is rather laborious and responsible for a larger fraction of leakage. Since the liquid fuel system is free from the radiation damage of cladding material, a very high burnup can be expected. In such a case, however, the system in equilibrium has to be operated at a very high fission product concentration of the magnitude of ε.

The transmuter inventory risk R_T can be assumed to be proportional to the fuel inventory and resident time in the transmuter, and is expressed as

$$R_T \propto t_T(1 - \varepsilon/2)/\varepsilon$$ (3)

where, t_T is the cycle time. The transmuter inventory risk is nearly in inversely proportion to the burnup per unit time, or the transmutation rate (the ratio of the transmuted mass per unit time to the fuel inventory at the begining of cycle). This means that a higher power density (higher neutron flux) is desirable to reduce the risk. The proportional coefficient is dependent on the types of transmuter and its fuel.

In the early ATW concept[14] proposed by Los Alamos, efficient transmutation was expected at a very high thermal neutron flux of the order of 10^{16} $n/cm^2/s$ in a liquid fuel with a very small inventory. This approach is attractive to reduce the transmuter inventory risk significantly, but may involve many severe technical problems.

In the liquid fuel system, the power density is generally not a limiting factor for the core design, because its core can be designed to contain no solid components. Although a very high power density is achievable in the core, the liquid fuel system can not entirely circumvent the heat removal problem that determines the transmutation rate. The heat removal problem usually appears instead in intermediate heat exchangers, and it can become still more difficult. The fuel contained in the primary fuel loops other than the core region occupies a considerably large fraction of the total inventory. If the design had followed a conventional design approach with external shell-and-tube type heat exchangers, it would be practically impossible to achieve a sufficiently high transmutation rate.

The fuel cycle risk R_F can be assumed to be proportional to the fuel inventory and processing time in the fuel cycle facilities, and is expressed as

$$R_F \propto t_p + t_F/\varepsilon + t_S(1 - \varepsilon)/\varepsilon \tag{4}$$

where, t_p, t_F and t_S are time lengths per cycle required for partitioning, fuel fabrication and separation, respectively. A high burnup and short processing times are desirable to reduce the fuel cycle risk. In case of liquid fuel system, there is no risks due to fuel fabrication, that is, $t_F = 0$ in Eq. (4).

Another major contributor to the short-term risks is transportation of radioactive materials. Collocation of dedicated transmuters and P-T fuel cycle facilities in the proposed double stratum fuel cycle concept could substantially reduce the risk due to transportation.

7. UNCERTAINTIES AND DATA NEEDS

One of the most important problems in the P-T systems study is that the data and methods are insufficient. Their uncertainty affects the predicted system performance.

The accuracy of present nuclear data is considered to be satisfactory for actinide transmutation in LMFBRs where relatively small amount of actinide is added to the ordinary fuel. However, it is not satisfactory yet in actinide burner reactors loaded with minor-actinide enriched fuel[15]. The JENDL Actinide File[16] is being compiled for the use in the transmutation study and contains neutron induced reaction data for about 90 nuclides from Tl-208 to Fm-255.

In the accelerator-based transmutation system, the data and methods for high-energy nuclear reaction are essential. In our design study, the nuclear reaction in the energy range above 15 MeV is calculated with a Monte Carlo simulation code NMTC/JAERI[17,18]. The proton-induced spallation process in a target is the primary source of neutrons. The most important quantity is the neutron yield of spallation reaction from the designer's standpoint. The yield of spallation products is also important for the estimation of their toxicity. The neutron yield from the minor-actinide target can be estimated in the error range of ±20%[19]. With this level of confidence, it is possible to make approximate evaluation of the concept of an accelerator-based system. To make more accurate evaluation, further experiments and developments of code with more accurate nuclear data are required.

As well as the nuclear data, physical property data for minor actinide fuel are absolutely lacking. The uncertainty in fuel data also affects significantly to the transmutation rate of the system. Data of thermal conductivity and melting point of fuel are most influential. High-temperature thermal conductivity data are totally missing for Np, Am, Cm, AmN and CmN. In the case of fluid fuel, specific heat also has a major influence.

There are also little data of damage in structural materials irradiated by high energy particles. Although they do not have direct effects on the transmutation rate, they are essential in the design of accelerator-based systems.

Uncertainties and data needs are large in fuel fabrication and reprocessing in the P-T fuel cycle. Thermodynamic and thermophysical databases of the TRU alloys, nitrides and chlorides are still limited. Compounds of Am have the problem of evaporative loss or vapor-phase transport of Am during fabrication and irradiation. Thermodynamic data of the molten salts containing actinide chlorides, particularly $AmCl_3$ and $CmCl_3$, have to be better defined.

As discussed in Sec. 6, technology development aiming toward higher-burnup transmutation and lower-leakage partitioning is the most important to reduce the risks associated with the P-T system. Scientific and engineering data concerning to the high burnup and low leakage should be of top priority.

To fulfill a wide spectrum of data needs for P-T, it is necessary to follow a systematic approach. The approach may consist of a series of processes: sensitivity analysis of system performance to the uncertainties, identification of technical priority issues due to their impact on system design, examination of existing database, assessment of critical data needs, review experimental and testing capability, estimation of schedule and cost, and formulation of experimental and testing plan. These processes will be iterative through design study and systems study.

8. ECONOMICS

It is premature to estimate the cost of the P-T system. Here, some qualitative discussions are made on economics of the P-T system.

The mass flow within the P-T fuel cycle in the double stratum fuel cycle concept is much smaller compared to that in the conventional fuel cycle. The amount of mass flow determines the scale of the fuel cycle facilities, and hence their construction and operation costs. HLW is about 3% of spent fuel by mass; TRUs correspond about 0.1%. Compared to a reprocessing plant with a throughput of 4000 kg/day, the construction and operation costs of a corresponding partitioning facility are estimated to be only about 1% and 2.5%, respectively [20]. Fuel flow through the P-T cycle is 0.2 - 1% depending on the fuel burnup. Cost of actinide particle fuel fabrication and partitioning facilities in the P-T fuel cycle is estimated approximately $970M for an annual throughput of 4130 kg.

Cost of the actinide burner reactor will be roughly the same as that of a commercial fast reactor with equivalent thermal power. The thermal power of the actinide burner reactor corresponds to about 2.5 - 5% of the total thermal power of the supported commercial reactors. Cost of transmuter could be covered by electric power generation. For the accelerator-based transmutation system, cost of the target/core system will be comparable to that of the equivalent actinide burner reactor. Cost of the accelerator is not yet estimated, but it may increase the total cost by 50 - 100%. Detailing the design of the target/core would assure the operation at a higher neutron multiplication factor; this could considerably reduce the cost of the accelerator. Design, construction and operation of the proposed ETA will provide more detailed information for the accelerator cost estimation.

Implementation of P-T will significantly decrease the volume, heat generation and toxicity of radioactive waste, resulting in a reduction of the cost of geological disposal. This could compensate for a substantial portion of the extra cost due to introduction of P-T.
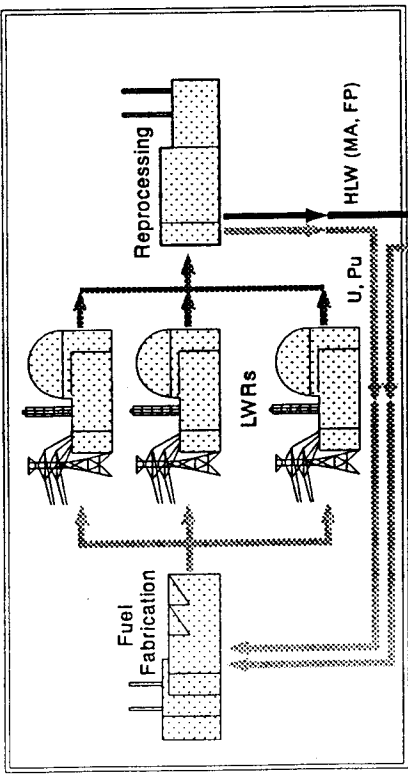
9. CONCLUDING REMARKS

Technology of the proposed P-T system based on the double stratum fuel cycle concept was presented. The partitioning process has a potential capability to separate long-lived nuclides from HLW, leaving final solution virtually free of both ultra-long-lived nuclides and high-decay-heat nuclides. The dedicated transmutation systems have a potential capability to burn minor actinides efficiently. Technology results so far obtained are sufficiently encouraging. Although technical feasibility of P-T is regarded as established, challenge still remain. Further R&D should be needed for other long-lived fission products than minor actinides. To achieve the ultimate goal of providing a different HLW management from geological disposal, technology development aiming toward higher-burnup transmutation and lower-leakage partitioning is of prime importance. R&D on P-T should be steadily carried out on a long time scale.

Systems studies should be needed to show its effective contribution to HLW management, not only in one specific country but also in more broad area. In this context, the P-T systems study under the framework of the OECD/NEA collaboration program will play a central role in finding out our common understandings on incentives for introducing P-T and technical issues to be developed further.

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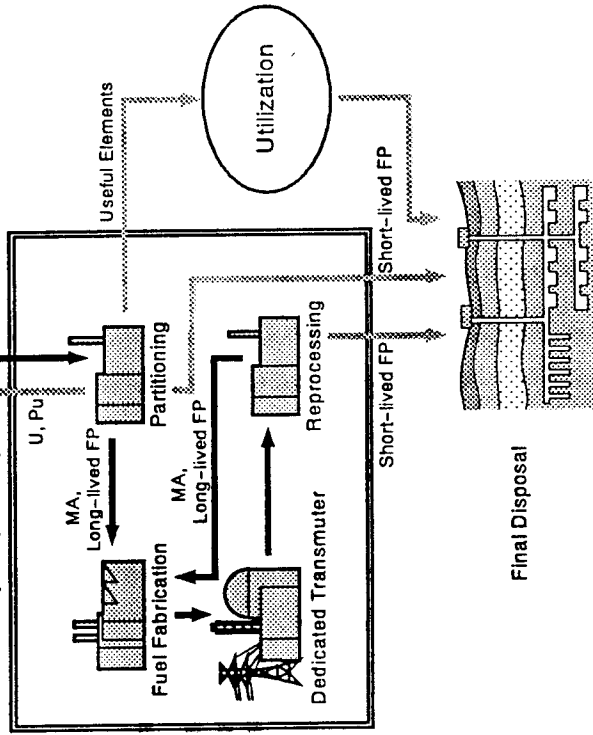First Stratum of Fuel Cycle (Commercial Power Reactor Fuel Cycle)

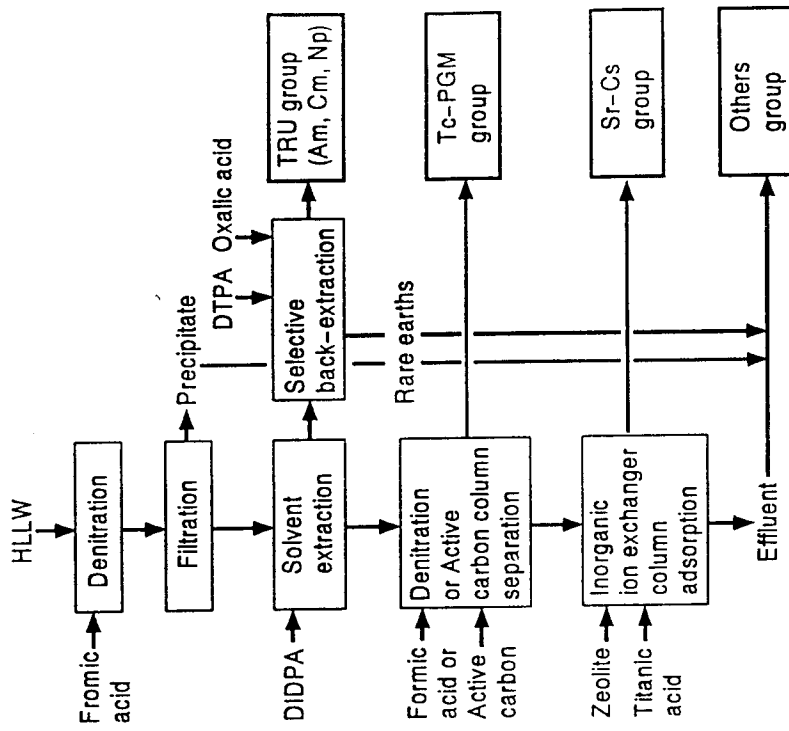Fig. 1 Concept of Double Stratum Fuel Cycle

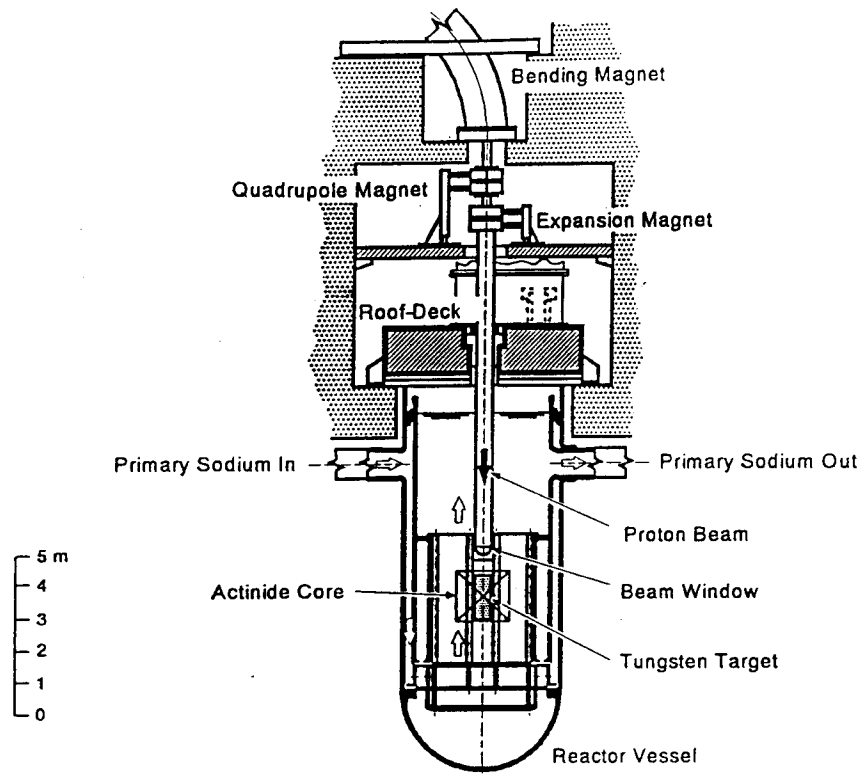Fig. 2 Four Group Partitioning Process
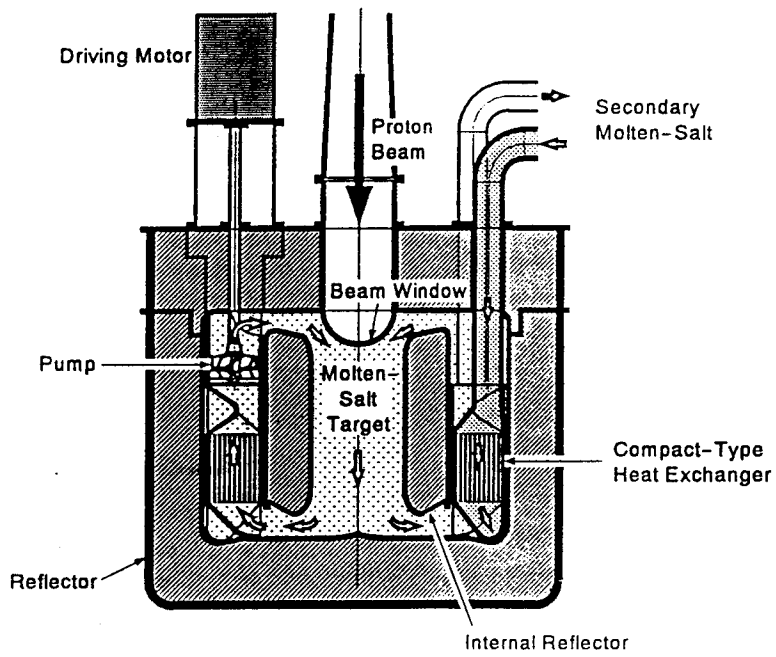
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Fig. 3 Solid Target/Core System

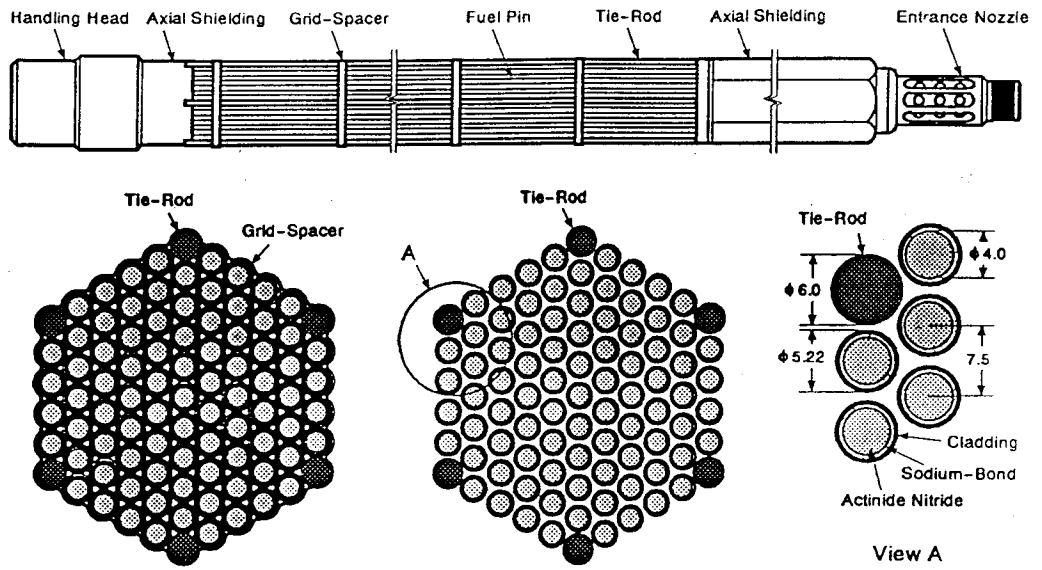Fig. 4 Concept of Molten–Salt Target/Core System
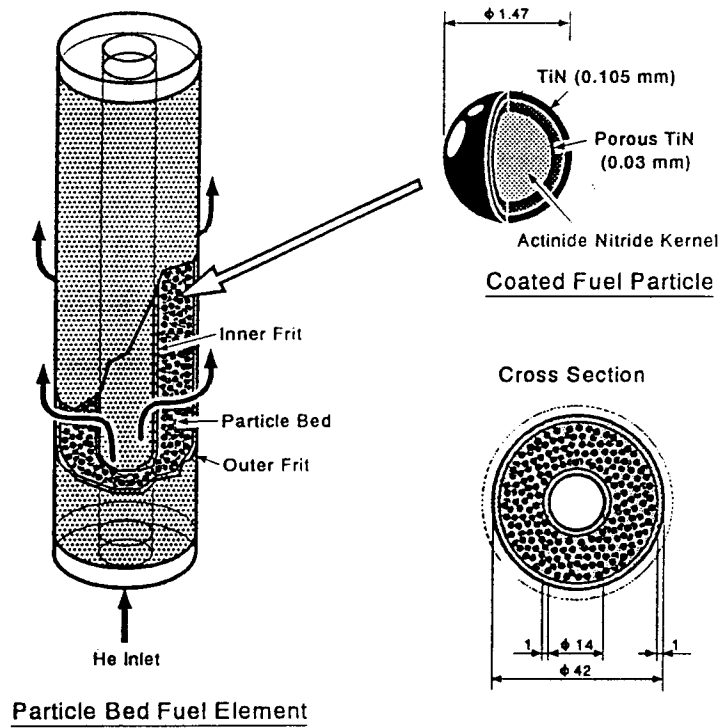
Fig. 5   Actinide Fuel Subassembly of L–ABR

Fig. 6   Coated Fuel Particle and Particle–Bed Fuel
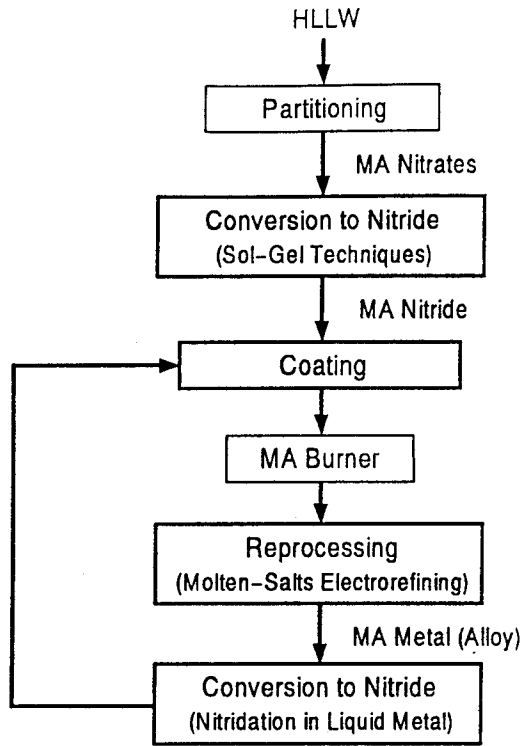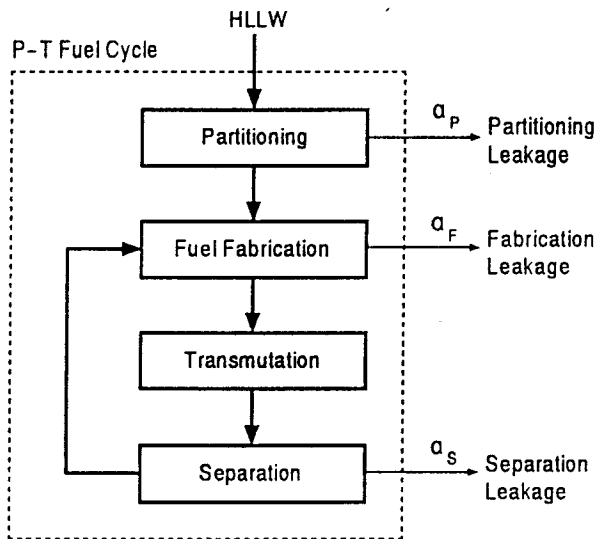Element for P–ABR

Fig. 7 MA Nitride Fuel Cycle

Fig. 8 P–T Fuel Cycle with
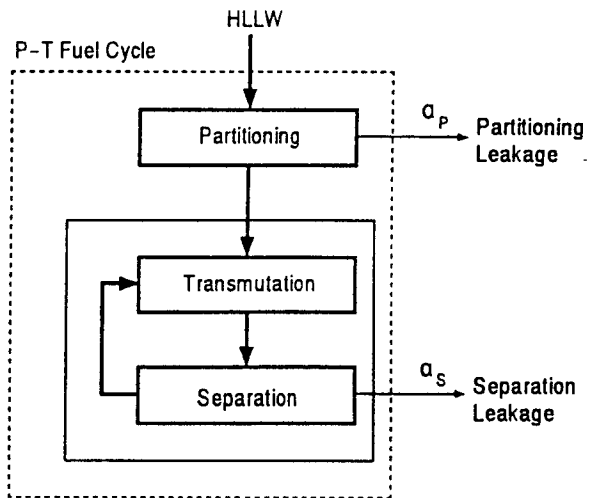Batch Processing

Fig. 9 P–T Fuel Cycle with
Continuous Processing

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"System Studies on the Advanced Fuel recycle at PNC"

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1.Introduction

In June 1994, the Atomic Energy Commission (AEC) of Japan formulated a "Long-Term Program for Research, Development and Utilization of Nuclear Energy". It entails that a development of advanced nuclear fuel recycling technology should be developed in the long range, on the basis of fast breeder reactor technology. In accordance with this new policy, PNC is starting development of advanced fuel recycle system. The advanced fuel recycle system is featured by the recycling of Minor Actinides (MA:Np,Am,Cm) in the fuel cycle.

This new approach to the future fuel-cycle stands on the following principal objectives ; (1) Reduction of the burden to the environment ; as the result of improvements both in cumulative production and distribution to the wastes of MA and Plutonium, by their high recovery and burning in the fast reactor core, (2) Improvement of nuclear fuel cycle economy ; by drastic modification and simplification of fuel cycle system in the FBR commercialization stage, (3) Enhanced proliferation resistance ; by adapting the co-processing of the plutonium with other actinides. (see Fig.1)
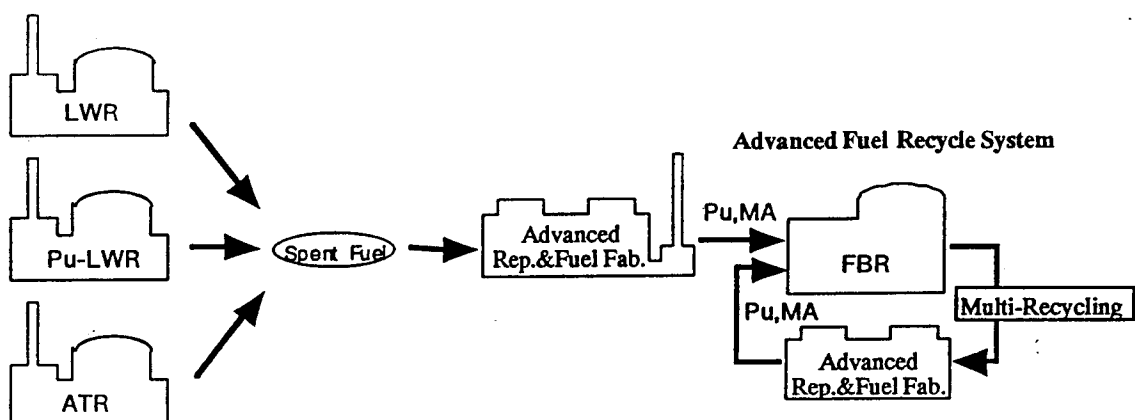

Fig.1 Concept of the actinide recycle system

2. Purpose of the Advanced Fuel Recycling

① Reducing burden to the global environment

Because MA and the residual plutonium dominate the toxicity of High Level Waste (HLW) for a long period, actinide recycle ensuring its high recovery of MA and Pu , can reduce long lasting toxicity in the HLW. The recovered MA is recycled and burnt in the FBR, thereby the total MA inventory in the fuel cycle does not simply increase but is maintained constant at a low level.

Through these features of actinide recycle, it is expected to make the waste disposal and the fuel cycle be more understandable and acceptable to the public.

② Improvement of nuclear fuel cycle economy

The nuclear power generation cost of fast reactor cycle mainly depends on the capital cost of the reactor, moreover, the influence of MA loading to the FBR core is negligibly small to the core performance and efficiency . Thereby, MA recycling to the core does not affect the generation cost so much.

Though the backend cost for fuel reprocessing and fabrication is possibly affected by the treatment of MA, if new design criteria of the cycles are applied which encourage the simplification of the process(e.g. imperfect purification of plutonium, rationalized waste handling etc.), it is possible to reach a reasonable and more efficient back-end costs in the FBR commercialization stage.

③ Enhancing proliferation resistance

It has been proposed that the closed fuel cycle with low decontamination has an proliferation resistance. The advanced fuel recycling system has the same feature by the co-recovery of Pu with other actinides or low decontamination of Pu from FPs. In this fuel cycle Pu never exist alone, which provides high barrier against its diversion and theft, and hence provides high reliability to the non-proliferation.

3. Production of Minor Actinides

Minor actinide nuclides are produced in the fission reactor (light water reactor and fast reactor) core by the neutron reaction and its chain. Especially, almost all Am-241 is generated during spent fuel cooling by reaction of the beta decay of Pu-241.

As a result of the operation of a 1000 MWe LWR for a year, production of 24 tons of spent fuel containing 22.6 tons of uranium, 1120 kg FPs, 250 kg Pu, and 20 kg MA is expected.

The production rate of ^{237}Np is large compared with that of Am and Cm in the LWR core using enriched uranium fuel. Almost all ^{237}Np is generated in thermal neutron spectrum by the following reaction.

$$^{235}U(n, \gamma)^{236}U \quad , \quad ^{236}U(n, \gamma)^{237}U \quad , \quad ^{237}U \xrightarrow[6.75d]{\beta^-} {}^{237}Np$$
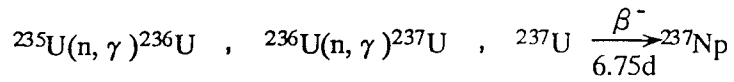
Am and Cm are produced by the reaction of neutron capture of Pu. Therefore, the production rate of MA in the thermal reactor increases according to the progress of burning.(Fig.2)

On the other hand, a fast neutron spectrum, with generally big fission-to-capture cross section ratio $\sigma f / \sigma c$ for minor actinide nuclides, can make more efficient use of neutrons for burning of MAs than a thermal spectrum. Therefore, the production rate of MA in the fast reactor decreases according to higher burn up.(Fig.3)
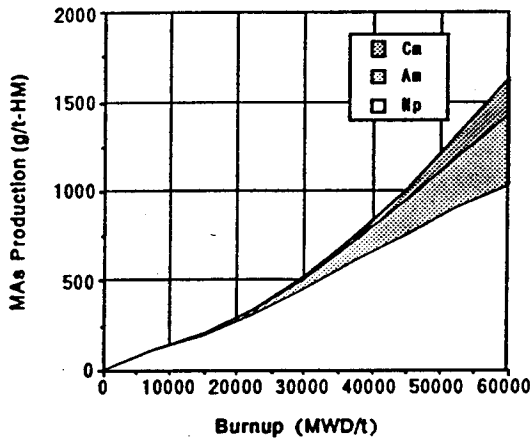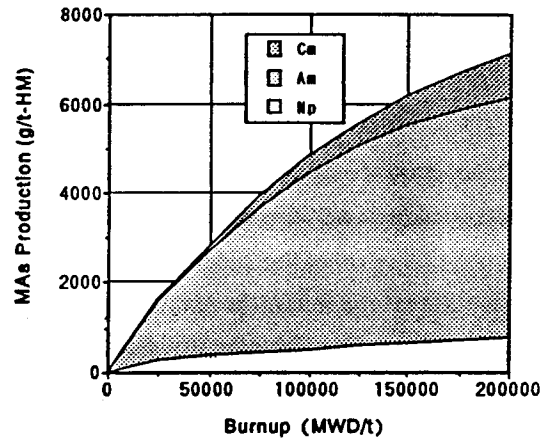

Fig.2 MAs Production in LWR core

Fig.3 MAs Production in FR core

4. Burning of MA through Fast Reactor

The objective of this section is to analyze Minor Actinide(MA) mass balance, such as the effect of MA accumulation reduction by the advanced fuel recycle and the MA concentration in FBR core fuel, according to the future foreseen nuclear energy production in Japan.

MA mass balance is studied on the assumptions that Plutonium and MA is recovered from the LWR and Pu-thermal reactor and recovered Pu and MA is recycled into fast reactor in multi number of times. Nuclear power generation is assumed to increase to 1000 MWe/y (Fig.4) or 1500MWe/y and introduction of commercial the fast reactor will start in the year 2030. New reactors are assumed to be totally FBR, and LWR reprocessing is assumed to be applied to all spent fuel discharged from LWR and Pu-thermal reactor.
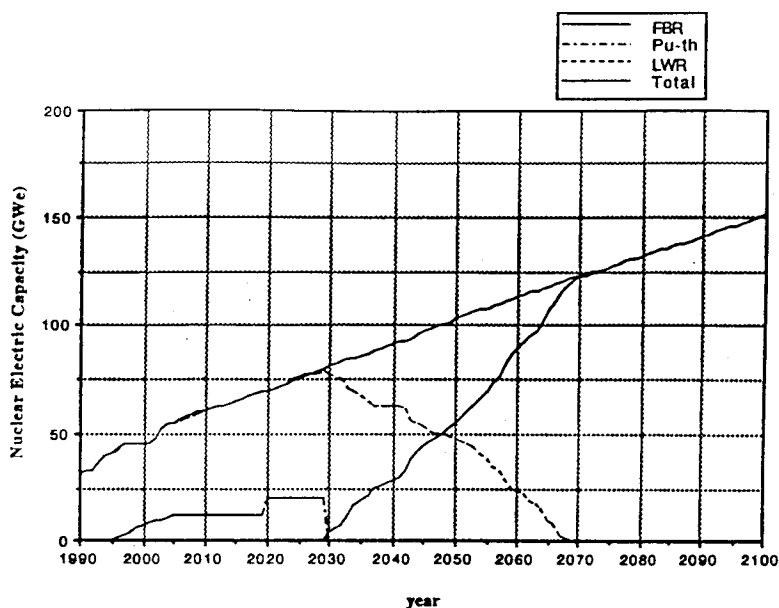
Fig.4 Assumed of Nuclear Electric Capacity

(1) Effect of MA Accumulation reduction

In Case-1(1000 MWe/y), the total MA transferred into the high level waste is calculated to be 310 tons from LWR, Pu-thermal LWR and FBR without recycling of MA. In the case of recycling MA into commercial FBR after the year 2030, the MA existing in the fuel cycle in the year 2100 is reduced to about 60 tons, reducing 80 % of no-recycle value.

The result of this analysis is indicated in Table 1.

And the residual accumulated MA are kept contained in the fuel cycle .(see Fig.5)

In this result, the maximum accumulation of total MA will be indicated about in the year 2065. Reduction ratio of curium is relatively lower than those of americium and neptunium because of the generation of curium by the neutron capture reaction of americium in the FBR core.

Table 1. Effect of MA Transmutation

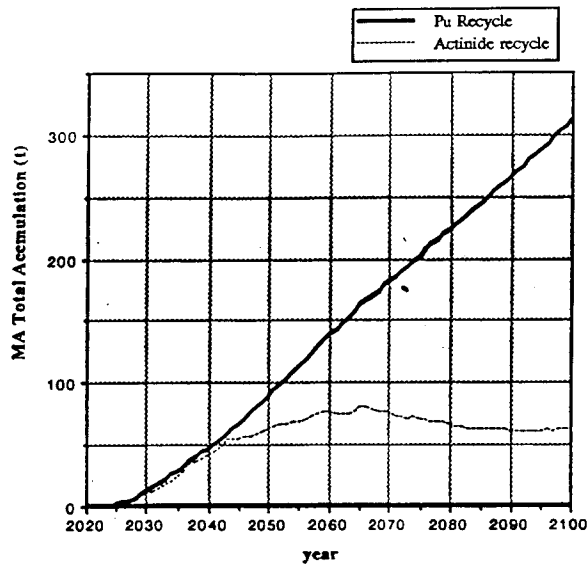|	Growth of Nuclear Power Generation	
	1000 MWe/y	1500 MWe/y
year 2025-2100 MA Generation (ton)	312 (Np 67, Am 230, Cm 15)	389 (Np 87, Am 283, Cm 19)
MA Accumulation in the year 2100 (ton)	63 (core 40, fuel cycle 22, waste 0.5)	87 (core 56, fuel cycle 30, waste 0.7)
MA Reduction Ratio (%) at the year 2100	80 (Np 88, Am 82, Cm 20)	78 (Np 87, Am 79, Cm 12)

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Fig.5 Effect of Minor Actinide Burning in the Advanced Fuel Recycling

(2) MA concentration in FBR fuel

Loading the MA in all-FBRs core homogeneously, MA concentration in FBR fuel is directly influenced by ratio of reprocessed spent fuel among LWR, Pu-thermal and FBR. , Especially, the higher ratio of Pu-thermal spent fuel reprocessing arises this concentration, therefore Pu-thermal spent fuel indicate high content of MA and small ratio of Pu-f.

Due to the MA multi-recycling, the concentration of the loaded FBR fuel and its spent fuel varies in every recycle. MA concentration will decrease gradually by the increasing FBR ratio and the effect of MA burning in FBR. In the year 2100, MA concentration will reach about only 0.7%.(see Fig.6)
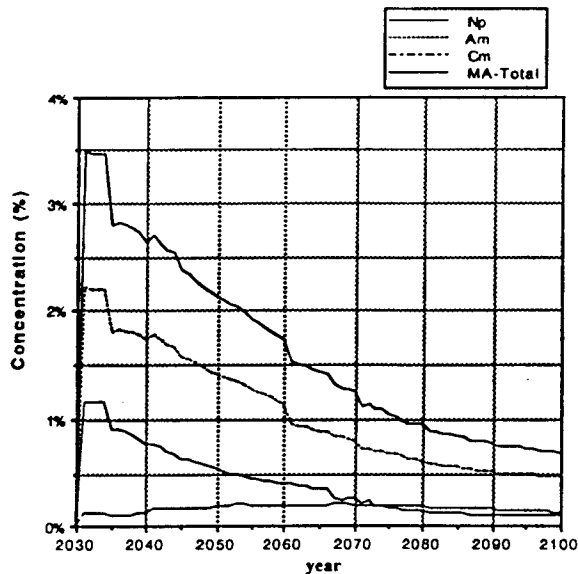

Fig.6 MA Concentration in the FBR Loading Fuel

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(3)Effect of potential hazard reduction

MA and the residual plutonium are the dominant toxicity from the high level waste (HLW) for a long time. In the present fuel reprocessing, high level waste is estimated to contain 100% minor actinide and about 0.5% plutonium from spent fuel. In addition, spent cladding hull and MOX secondary wastes contaminated from small quantity of plutonium.

The advanced fuel recycle intends to reduce the long-lived radiotoxicity by recovering MA and reducing Pu losses from not only HLW but also other wastes. (Fig.7)
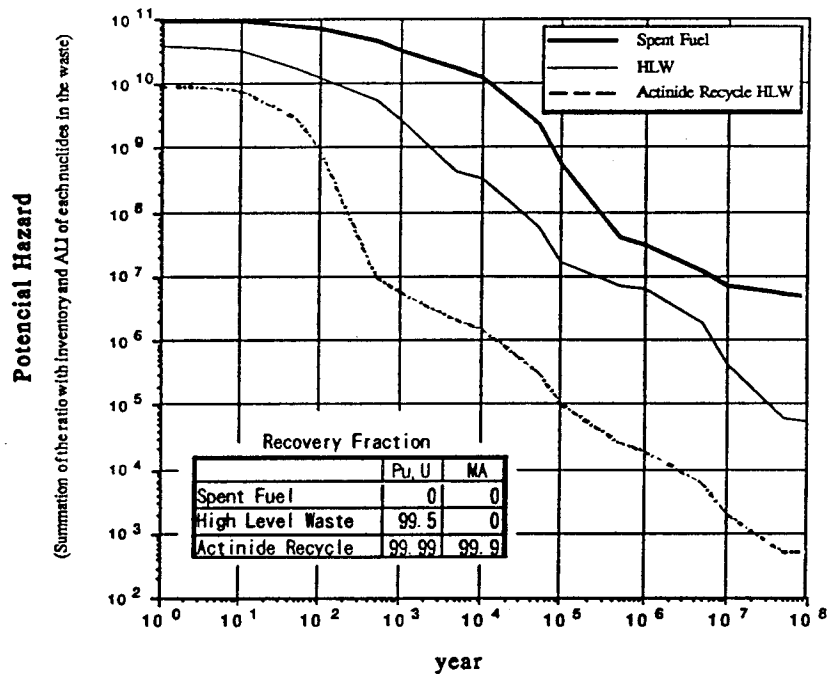
Fig.7 Effect of the Actinide Recovery on the Potencial Hazard of FBR-HLW (150,000MWD/t)

5. Conclusions

The development of advanced fuel recycling is expected in the next generation technology to incorporate various purposes such as to reduce the burden to global environment, improvement of economy and additional proliferation resistance. By recycling MA into commercial FBR, the MA existed in fuel cycle is reduced by 80% of no-recycle value. The long-lived radiotoxicity is reduced by recovering MA and reducing Pu losses in the fuel cycle.

PARTITIONING AND TRANSMUTATION SYSTEM OF TRANSURANIUM ELEMENTS WITH PYROMETALLURGICAL SEPARATION AND METAL FUEL

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1. Definition of criteria

1-1. Aims in terms of toxicity of waste

-Toxicity reduction of spent fuel, reprocessing waste after removing U and Pu with 99.5% and waste by recovering 99% of actinides; see figure 1

   Toxicity of 4.5 % enriched uranium fuel of 1t and toxicity of uranium ore to make 1t of uranium fuel are also indicated.

-Reprocessing waste contaminated actinides (Project Gewahr 1985, NAGRA)

   α -activity

      HLW: ca. 1.6 x 10^3 Ci/canister

      cladding after leaching; ca. 10^{-3} - 10^{-4} less than the activity of HLW

      undissolved residue;

      solvent scrubbing solution;

-Separation rate of TRU by 99 - 99.9 % from HLW; Not so large effect on safety analysis of vitrified waste disposed under geologic formation.

-Decontamination factor (DF) to be needed to make the same concentration with uranium ore; ca. 10^6 - 10^7 for HLW, and ca. 10^3 - 10^4 for cladding after leaching

-DF to be needed to make the non-TRU waste: ca. more than 10^9 - $10^{10}$for HLW

-The important issues of actinides recovery: To reduce the accumulation outside of the fuel cycle in order to maintain the nuclear-benefit world. Thus, actinides as much amount as possible should be confined and managed in the fuel cycle.

1-2. Aims for the reduction in CRIEPI

-The first target of the CRIEPI process; To recover more than 99% of each actinide in the high level radioactive liquid waste by dryprocess

-Kinds of wastes to be applied; HLW, waste from solvent scrubbing, undissolved

residue, cladding after leaching

1-3. Concept of fuel cycle with partitioning and transmutation

Object: To reduce the release of actinides as wastes as low as possible from both of LWR and FRB cycles.

Concept: Actinides from both of LWR and FBR fuel cycles are confined in the FBR fuel cycle. See figure 2, which shows the future fuel cycle including the partitioning of TRU, which is under developed in CRIEPI. We aim through this fuel cycle the release of TRU less than 1% from the fuel cycles.

-Actinides cycle instead of U and Pu cycle.

-Dry process for TRU recovery from wastes produced by purex type reprocessing

-Metal fuel, U-Pu-Zr, with minor actinides

-Dry reprocessing of FBR spent fuels to recover all of actinides

See figure 3, which shows the material flow at an equilibrium, in which metal fuels with 5% of minor actinides are used for FBR fuel.

1-4. Major process for increasing doses by introduction of PT

-Partitioning process; additional facility for separation of TRU

-Fuel fabrication process; high decay heat, High radioactivity; see figure 4 & table 1

2. Timings

2-1. Research for process development

[Partitioning]

- Establish the process flow and material flow by laboratory scale experiment; till 2000

-Establish the technology to recover more than 99% of all actinide elements; till 2000

[Transmutation]

- Target of fuel study; completed

    Measurement of major characterization for irradiation analysis

- irradiation test with MA; till 2000

    Fuel fabrication for irradiation test; completed

    Irradiation starts; 1995

    Complete the PIE; till 2000

- Computer code study

2-2. Further development for the engineering demonstration

[Partitioning]

   – Demonstration by engineering scale test after 2000

[Transmutation]

   – Development of fuel fabrication method for commercialization

   – Irradiation test for fuel optimization; after 2000

3. Description of technology

3-1. Partitioning, see figure 5

  –Pre-treatment process

    Denitration process; oxides conversion from nitrate aqueous solution

    Chlorination process; converting oxides to chlorides

  –Pyrometallurgical process

    Reductive extraction process; reduction of actinide chlorides to metals

      Noble metal recovery

      Separating actinides from alkali-, alkaline earths and lanthanides

    Electrorefining process

      Purify the actinides by separating from lanthanides

  –Waste salt treatment process;

    The substantial process for minimizing secondary wastes. Most of all solvent can be recycled to main stream. See figure 6.

3-2. Transmutation

  –Fuel fabrication

  –Evaluation fuel behavior

  –Fuel reprocessing used in FBR

  –Computer simulation for transmutation rate

  –Core analysis for the fuels with minor actinides

4. Predictions of technology results

4-1. Partitioning

  –Major thermodynamic date for pyrometallurgical separation: mostly completed

  –More than 99% recovery of actinides from HLW; possible. The point is the mixing rate of lanthanides in products, which increases with increasing the recovery rate of actinides. Experiments to attain the same amount of lanthanides with TRU

recovered more than 99% are underway.

-The amount of secondary waste ; can be expected to be lower amount of production, however, has to be evaluated precisely.

4-2. Transmutation

-Major properties of metal fuel with minor actinides to be need for analysis for reactor irradiation; mostly prepared by measurements.

-Content of minor actinides in the fuel: Maximum is less than 10% from points of safety parameters and fuel characterization. 5% is proposed. 2% is enough to recycle self-generated minor actinides.

5. Uncertainties

5-1. Partitioning

-Numerical estimation of secondary wastes to be produced.

-Selection of materials for equipment; corrosion-resistant materials at elevated temperatures.

5-2. Transmutation

-How many recycles of Pu and minor actinides can be possible from points of nuclear data analysis

-How behave minor actinides, especially Am, in fuels during irradiation.

6. Data needs (scientific, engineering, economic)

- Precise process and material flows for separation prior to the engineering scale test

- Estimation of process loss, and production of secondary waste

- Economic analysis for dry partitioning facility

- Nuclear data analysis after multiple recycling

- Am behavior at fuel fabrication
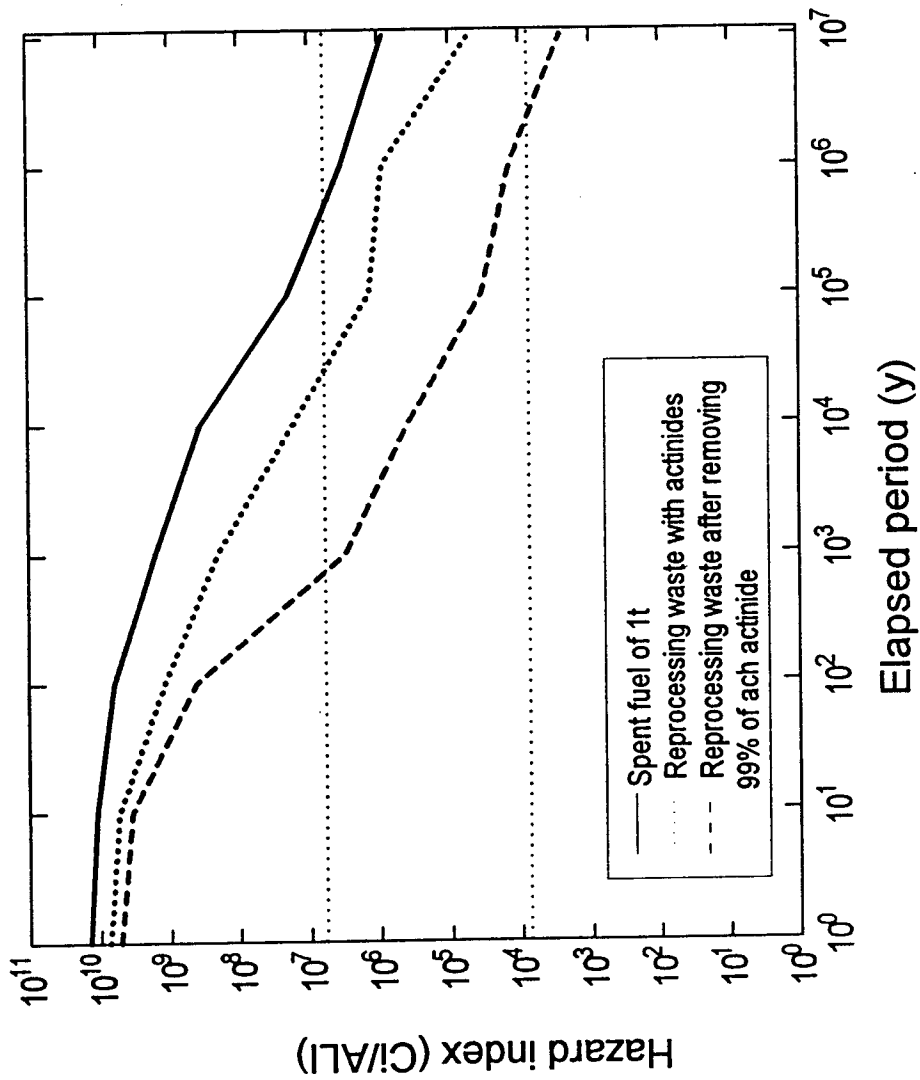
- Irradiation data of metal fuels with minor actinides

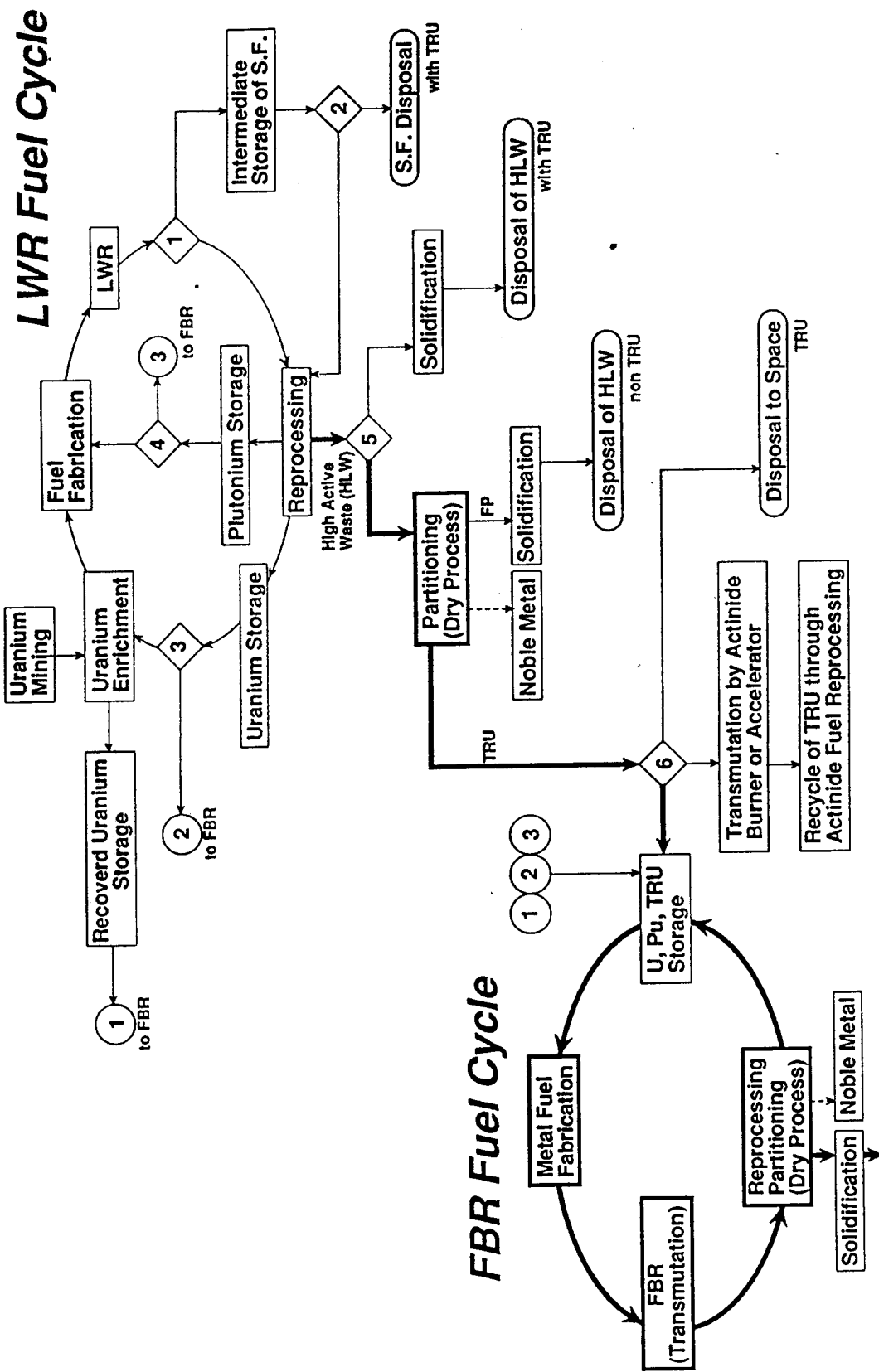Fig. 1   Radio-toxicity of spent fuel and reprocessing wastes

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LWR Fuel Cycle

Uranium Mining → Uranium Enrichment

Recoverd Uranium Storage → ① to FBR

Uranium Enrichment → ◇3 → ② to FBR

Uranium Enrichment → LWR → Fuel Fabrication

Fuel Fabrication → ◇4 → ③ to FBR

Plutonium Storage → ◇4

LWR → ◇1 → Intermediate Storage of S.F. → ◇2 → S.F. Disposal (with TRU)

◇1 → Reprocessing

Uranium Storage → ◇3

Reprocessing → Plutonium Storage

Reprocessing → High Active Waste (HLW) → ◇5 → Solidification → Disposal of HLW (with TRU)

◇5 → Partitioning (Dry Process)

Partitioning (Dry Process) → FP → Solidification → Disposal of HLW (non TRU)

Partitioning (Dry Process) → Noble Metal

Partitioning (Dry Process) → TRU → ◇6

◇6 → Disposal to Space (TRU)

◇6 → Transmutation by Actinide Burner or Accelerator

◇6 → Recycle of TRU through Actinide Fuel Reprocessing

① ② ③ → U, Pu, TRU Storage

◇6 → U, Pu, TRU Storage

FBR Fuel Cycle

U, Pu, TRU Storage → Metal Fuel Fabrication → FBR (Transmutation) → Reprocessing Partitioning (Dry Process) → U, Pu, TRU Storage

Reprocessing Partitioning (Dry Process) → Noble Metal

Reprocessing Partitioning (Dry Process) → Solidification

Fig. 2 Partitioning and transmutation concept of transuranium elements(CRIEPI).
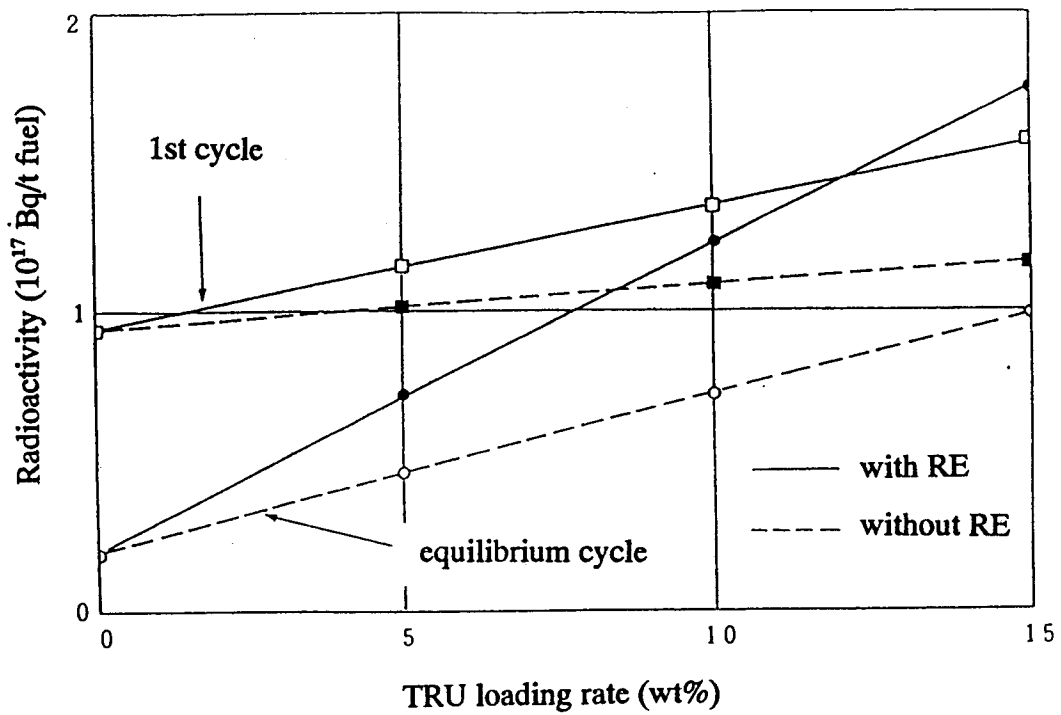TRU produced in LWR and FBR cycles are confined in the FBR cycle.

1000MWe Metallic fuel FBR

Spent fuel

U : 4935(kg)
Pu : 1278(kg)
MA : 251(kg)
RE : 605(kg)
FP : 710(kg)

Cooling: 3years.

Reprocess and partitioning

U : 4935 (kg)
Pu : 1280 (kg)
MA : 248 (kg)
RE : 248 (kg)

RE separation efficiency :59%

Burnup:3years.

Fuel prefabrication

U : 5618(kg)
Pu : 1385(kg)
MA : 389(kg)
RE : 389(kg)

Depleted U

U : 683 (kg)

Pu : 105 (kg)

MA : 141 (kg)
RE : 141 (kg)

Burnup : 3years.
Cooling : 4 years.
Partitioning.

x 6 plants

1000MWe LWR

Fig.3   Equilibrium recycle of minor actinides from LWRs in a metallic fuel FBR.
The metal fuel contain 5% of minor actinide and 5% of lanthanide.

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Fig. 4. Radioactivity of fresh metal fuel with minor actinides as a
function of loading ratio

Table 1. Contribution of nuclides to radioactivity of fresh metal fuel with 1t
(TRU loading; 15%, RE loading 15%)

(a) Actinides   (b) lanthanides

Nuclide	Contribution to radioactivity(%)
Cm 244	61
Pu 238	17
Pu 241	17
Am 241	3
Others	2

Nuclide	Contribution to radioactivity(%)
Pm 147	52
Ce 144	36
Eu 155	6
Eu 154	4
Others	2

Total; 9.84 x 10^{16} Bq/t-fuel   Total; 7.55 x 10^{16} Bq/t-fuel
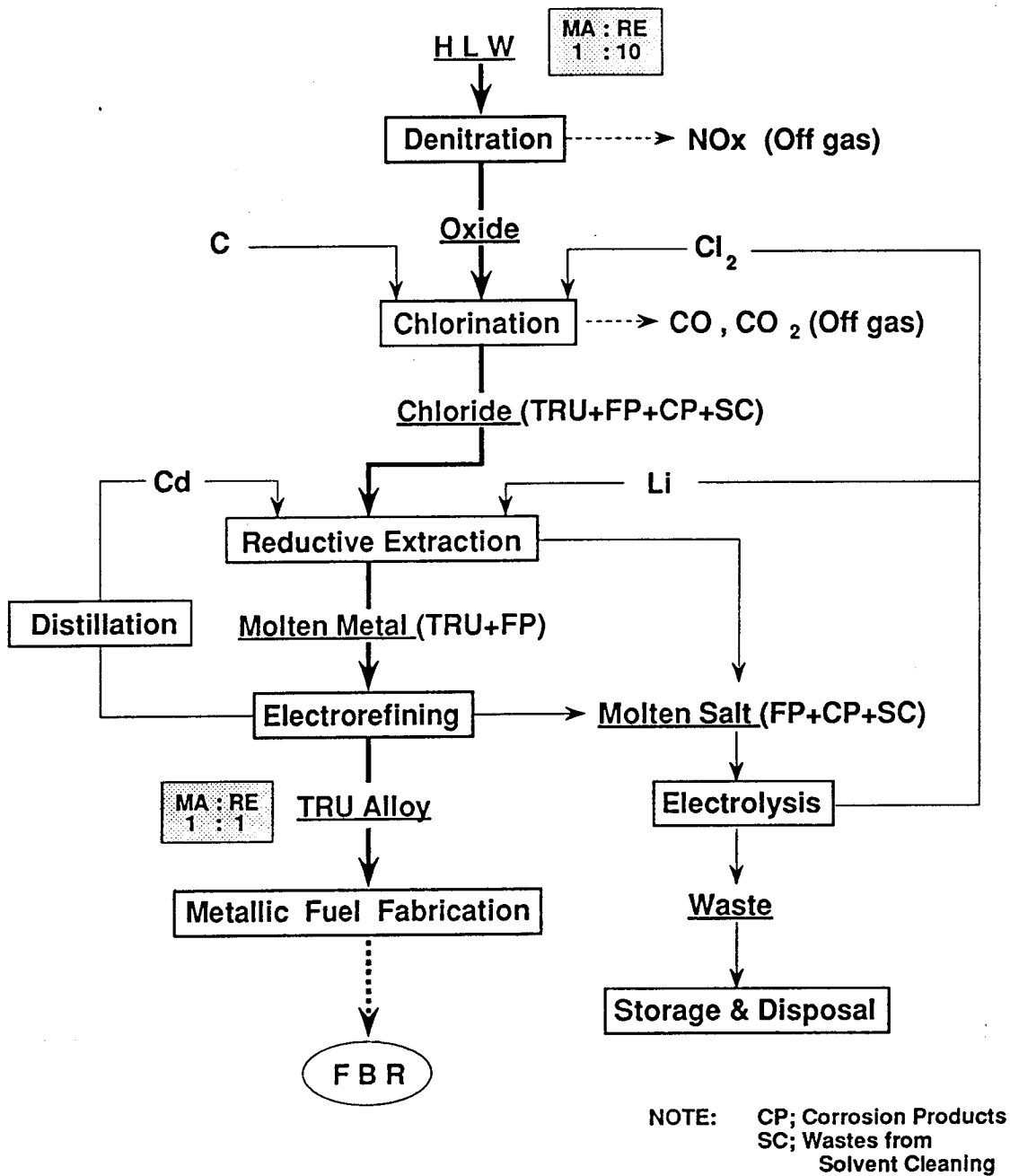
Fig. 5 Flow diagram of dry process of actinide separation from high level waste

Main separation process

Waste treatment process
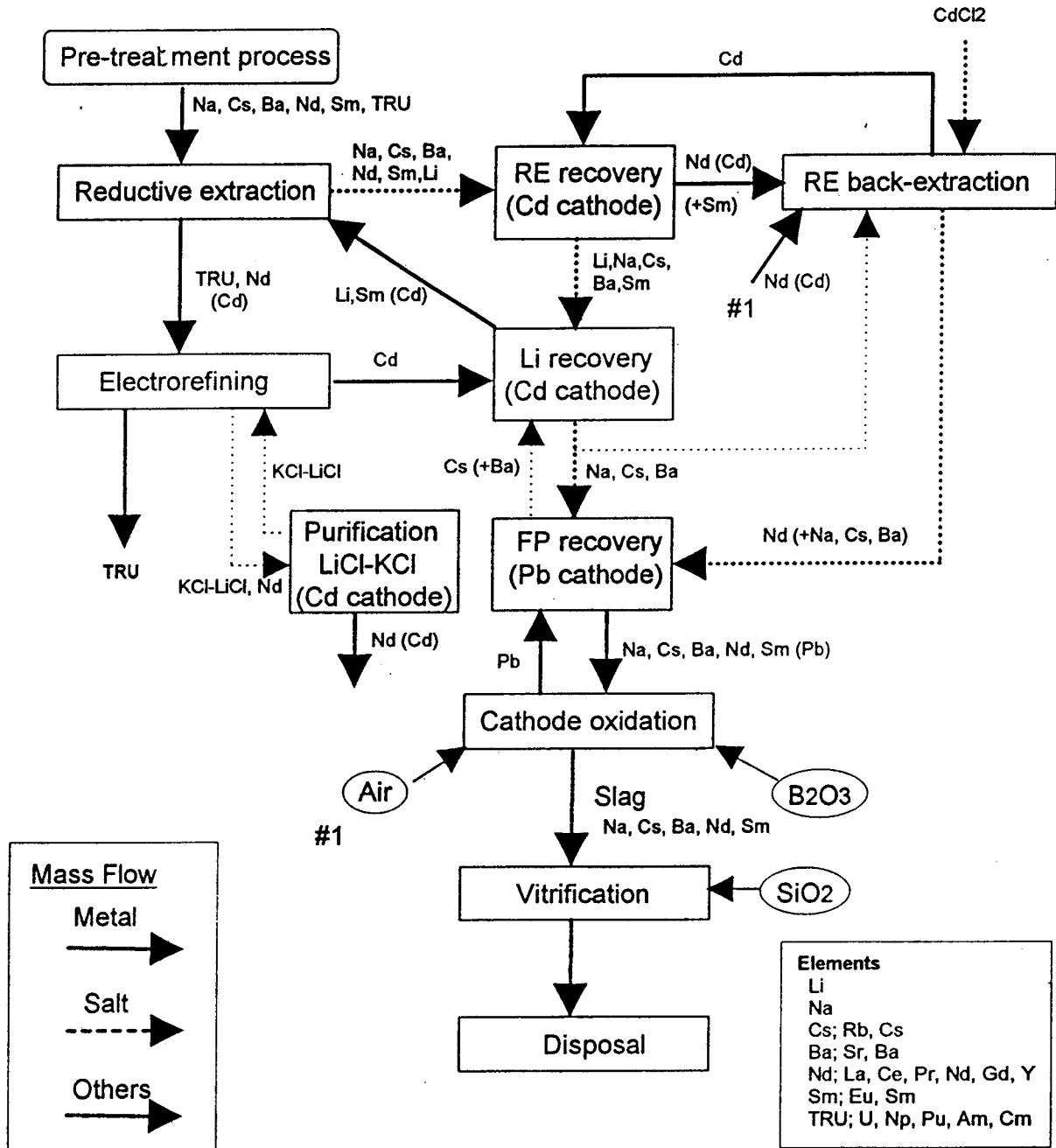

Fig. 6   Flow diagram of salt waste treatment

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WASTE MANAGEMENT CONCEPT
FOR FUTURE LARGE-SCALE NUCLEAR POWER

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ABSTRACT

The concept of radwaste management is discussed as part of the general concept of a naturally safe nuclear technology for the large-scale power industry. Deterministic exclusion of reactor accidents with fuel failure allows burning MA within the main fuel, as well as I and Tc. On attaining a certain efficiency of radiochemical separation of actinoids and FPs, it becomes possible to bury radwastes after their long-term cooling, without upsetting the natural radiation level. The paper presents the results of studies performed by RDIPE, covering the reactor proper, the radioactivity of fuel and wastes, the storage and disposal of radwastes, and the utilization of Sr and Cs.

I. INTRODUCTION

Along with evolutionary improvement of traditional nuclear technologies in the next few decades, nuclear power also needs a definite concept of its long-term development, the absence of which deprives it of a clear perspective and of major arguments in its favour. The possibility to arrest the growth and then to reduce the consumption of conventional fuels has from the outset been a major incentive to research and developments on peaceful nuclear energy. The global fuel problems came to be resolved in this century without great involvement of nuclear fuel, and forecasts up to the year 2020 show its share in the total fuel resources consumed to remain at about 5 %.

However, the alarming estimates of the available resources, such as oil and gas, and the looming environmental and international problems which are bound to be caused by the inevitable increase of the global population and fuel demands in the next century, make us go back to the concept of large-scale nuclear power, since other energy technologies still fall short of offering a realistic alternative to the traditional power engineering. But for nuclear power to be accepted as a conventional technology for mass application, it is essential to develop a nuclear technology which would fully take into account the lessons of the first stage as well as the emerging new conditions and new requirements to be met.

Of primary importance among the latter is the exclusion of reactor accidents with catastrophic radioactive releases. Their estimated probabilities are quite acceptable for predicting safe short-term development which will not go too far away from the accumulated experience of about 6×10^3 reactor-years. However, such extrapolation of experience to $\sim 10^6$ reactor-years, which may be reached in the century to come, is not convincing due to the use of the estimated event probabilities much smaller than 10^{-6}, which usually have no experimental or trustworthy theoretical basis to support them. One cannot agree with A. Weinberg in that the "Second Nuclear Era" calls for a new nuclear technology capable of deterministically excluding catastrophic accidents by virtue of natural laws which, unlike engineered systems and barriers, are entirely reliable.

But a too expensive technology, even if quite safe, has no prospects for large-scale application in the energy sector where economic criteria are of equal fundamental importance. The main reasons for the multiple increase in the cost of NPPs lie in the growing complexity of design, requirements to equipment and personnel, and licensing procedures, resulting from the mounting safety requirements. The utmost safety attained due to technical features protected from catastrophic accidents by their inherent properties and natural laws rather than through building up engineered systems and requirements, can be combined with simplification of design and requirements, and reduction of NPP costs.

Another factor of fundamental importance for economy consists in economical consumption of natural uranium which, with the present-day LWRs having operated for 10^6 reactor-years, would amount to about 200 men. t - a figure 20-40 times as large as the currently expected or explored U resources of acceptable cost. In order to keep

consumption within their limits and to avoid considerable expansion of the uranium mining industry, the new technology should ensure a reduction in the specific consumption of uranium by at least an order of magnitude.

Fast reactors with liquid metal cooling, in our opinion, have the greatest potential for coping with this problem, as well as with other challenges of a large-scale power industry (safety, wastes, economy), although some types of thermal converters in the Th-U cycle are also acceptable in terms of the fuel balance. But modern fast reactors proved to be much more expensive than LWRs despite the greater fuel and energy efficiency and simplicity of design and control principles. This may be accounted for by the choice of the highly reactive Na made at an early stage, which results in a more complicated design of the reactor, cooling and refuelling systems, SG, structures, etc.

The chemical activity and insufficient boiling margin of Na do not fit in with the requirements of inherent safety as well as the low density and heat conductivity of the oxide fuel. We see the new reactor technology as a fast reactor with a chemically inert high-boiling coolant (Pb) and a high-density, heat-conducting, heat- and radiation-resistant fuel (UN-PuN) with good radionuclide retention, efficient self-regulation by feedbacks, high natural circulation of coolant, small reactivity margin, etc.

The principles of deterministic (inherent) safety should be extended to include other links of the closed fuel cycle, primarily the final disposal of radwastes, due to the questionable reliability of engineering, geological and "historical" forecasts for tens of thousands of years ahead. Fission of all actinoids in fast reactors, transmutation of the most long-lived FPs, and gradual radioactive decay of the remaining products are fundamental physical processes which reduce the hazard of the buried wastes to that of U extracted from the earth together with its α-active decay products. The technological arrangement based on the above can be convincingly proved to be safe. The main prerequisites for implementing such an arrangement include elimination of reactor accidents with fuel failure and fractionation of the burnt-out fuel composition into U, Pu and MA for fresh fuel fabrication, I and Tc for in-pile transmutation, Sr and Cs for utilization, and radwastes to be cooled and buried.

The risk of proliferation of nuclear weapons has turned into an acute problem which is barring adoption of the concept of large-scale global nuclear power with fast reactors, closed cycle and Pu. In conjunction with political safeguards, nuclear

technology has great, as yet untapped potentialities for ruling out thefts of Pu and its military uses. Development of a nuclear technology - economical, safe and protected against Pu thefts - would be an economically and strategically more effective application for funds and efforts, including efforts of nuclear weapon experts, than would be the development of special Pu burners and technologies for its vitrification and disposal.

After Chernobyl, we undertook a search for and studies on a new nuclear technology which would meet the varied requirements of a large-scale power industry, without going too far away from what has already been mastered by nuclear engineering.

While sharing A. Weinberg's opinion that creation of a deterministically safe nuclear technology is worth the efforts commensurate to those spent already, we, nevertheless, see possibilities for developing and demonstrating such a technology within a limited stretch of some 15-20 years, based on the experience amassed in peaceful and military nuclear engineering[*].

In team with other institutes, RDIPE has carried out conceptual development work for a lead-cooled fast reactor with UN-PuN-MA fuel, designed to operate in a closed transmutation fuel cycle. Consideration has been also given to an option proposed by C. Rubbia - a slightly subcritical reactor with a cyclic proton accelerator as a neutron source - which offers convincing proof of safety with respect to reactivity accidents. The results are discussed above, with emphasis laid on the problem of radwastes. Investigations performed in many countries in the recent years have clarified the concept of the transmutation technology in terms of its physics, for which 2- or 3-fold uncertainties in the radiation characteristics have no critical implications. We shall try and describe it here in fairly simple terms, without going into design details. Its engineering development is our main objective today.

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[*]   This is a normal time scale for nuclear developments. Such time, for instance, was taken to develop and bring in the BN-350, BN-600 and other reactors.
Inordinately protracted creation of a commercial fast reactor must have been caused, in our opinion, by the original choice of technical features (Na, oxide fuel) which cannot give effect to the potential advantages of fast reactors in terms of either safety or economic efficiency.

It stands to reason that the start of a new stage in nuclear power development around new reactors should be tied in with solving an urgent problem of the next few decades, which consists in utilizing the Pu accumulated in storages as a result of reduction of nuclear armaments and fuel reprocessing. Several reactors of the type in question are capable of converting within reasonable time the Pu accumulated in Russia into the theft-proof form of spent fuel, thus following the CISAC recommendations, while simultaneously gaining experience for subsequent large-scale deployment.

II. LEAD-COOLED FAST REACTOR

Two-circuit 300, 600 and 1000 MWe options of lead-cooled fast reactor were studied. Main characteristics of these reactors are given in Table 1 and discussed in detail in the reports presented at the ARS-94 Conference (Pittsburgh, 1994).

300 MWe was found to be the lowest power level at which it proved possible to use nitride fuel, do without uranium blanket and ensure CBR ~ 1 and total reactivity margin ΔK to t < 1 \$.

The feature of low neutron moderation and absorption in lead allowed the following :

- increase lead inventory in the core from ~ 40 % (Na) to ~ 60 % ;
- reduce lead heating to 120°C and maximum flow rate to 1.8 m/s ;
- reduce pamping power to less then 1 % of reactor power ;
- provide high level of natural circulation more than 15 % of nominal circulation.

Large pitch in the fuel lattice makes it possible to use unducted fuel assemblies (FA) and hence avoid local loss of FA cooling.

Lead temperature at the core inlet/outlet (420°C and 540°C respectively) ensures the required margin to lead freezing (327°C) and acceptable operating conditions for fuel claddings, reactor vessel, steam generators and pumps. To avoid lead freezing under abnormal conditions, it was decided to use supercritical steam parameters with the feedwater temperature of 340°C and a jet mixer and steam pump drive in the secondary circuit. These features result in the thermal efficiency of ~ 44 %.

Besides, the use of high-density heat-conductive nitride fuel in combination with lead coolant allows :

*   - to limit average operating fuel temperature to 700°C,
    -. to confine most of the fission products (FP),
    - to relieve stressis on fuel claddings ;

*   - to minimize power reactivity coefficient while preserving the temperature coefficient ;

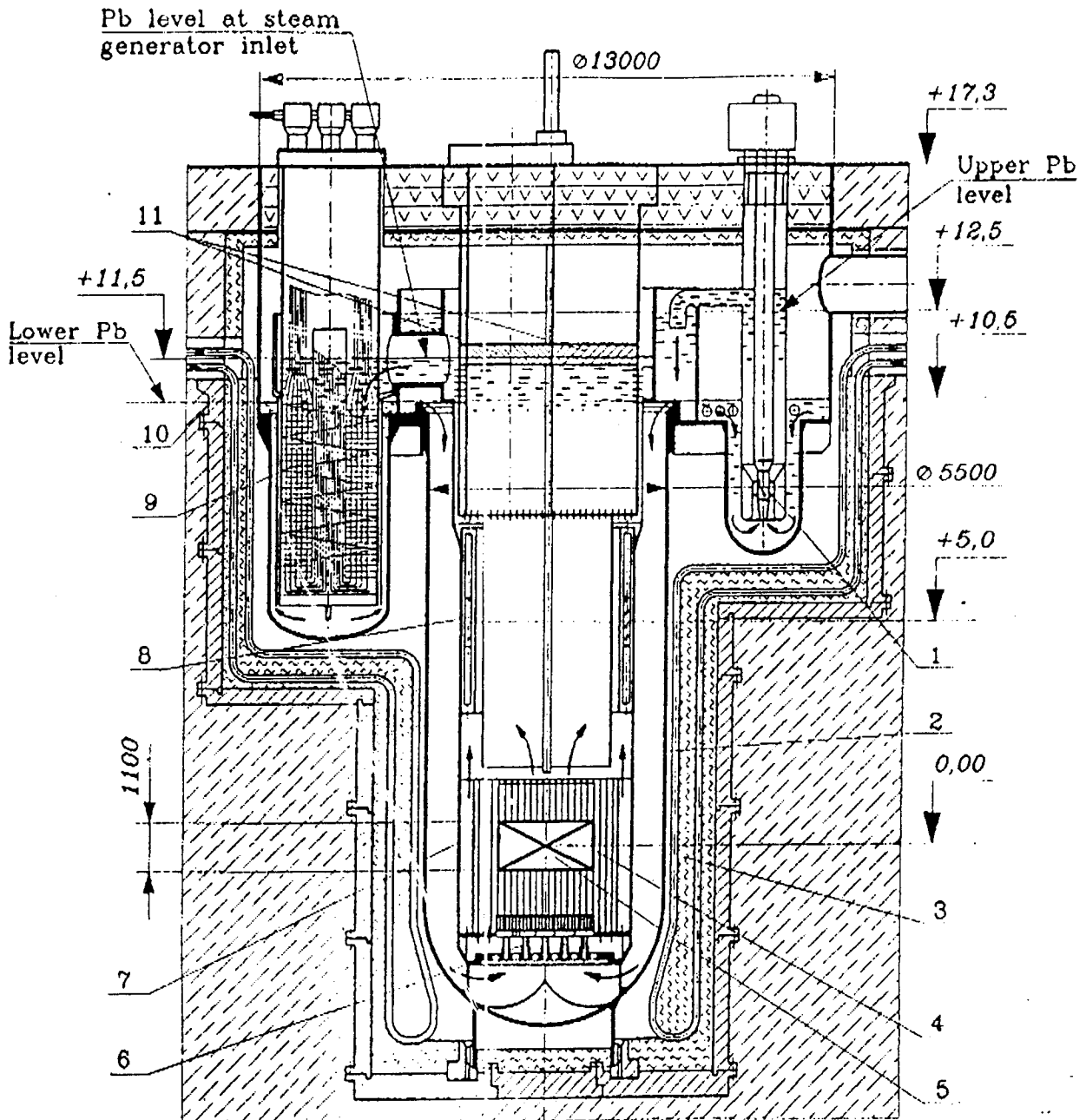   - to simplify control system and avoid fast runaway under all accidents in the system.

The absence of traditional uranium blanket prevents production of weapon-grade plutonium. Void reactivity effect becomes deeply negative in case of lead draining and is < 1 $ in case of steam or gas bubbles injection in the core. Taking into account high boiling temperature of lead, this prevents dangerous local positive void reactivity effect.

Control means are located outside the core. They operate based on simple reactivity control principles consisting in varying neutron leakage, which simplifies refuelling as well.

The reactor has an integral layout of the primary circuit (Fig. 1). Refuelling is performed via an in-core storage pool with the help of two rotating plugs. There is no need to wash removed FA during refuelling.

A single reactor vessel is located in a reinforced concrete vault 3 m thick. The gap between the walls of the vessel and the vault prevents loss of lead flow in case of vessel failure and partial loss of lead. Long-term emergency core cooling is based on natural air circulation in the vault.

As estimated, design simplification and natural safety features make the cost of such a plant comparable to that of LWR and even lower.

Pb level at steam generator inlet

∅13000

+17,3

Upper Pb level

+12,5

11

+11,5

Lower Pb level

+10,5

10

9

∅5500

8

+5,0

1

2

1100

0,00

3

7

4

6

5

1—pump   7—separating shell
2—vessel   8—fuel storage
3—thermal shielding 9—steam generator
4—CPS   10—safeguard vessel
5—core   11—rotary plug
6—support posts

Figure 1 - BREST-300 layout

Extreme accidents including vessel/containment failure, maximum reactivity insertion, pump trips, break in the secondary circuit etc. without scram will not result in fuel failure with catastrophic radioactive releases (over 10^4 Ci in 1 equivalent).

Fuel performance was calculated for equilibrium isotope composition obtained as a result of multiple recycling, with fission products extracted in each cycle and produced actinoids returned in the reactor for burning.

The calculations show that some 0.3 neutron in a fission is absorbed outside the core. These neutrons can be used for example for transmutation of long-lived FPs such as I-129 and Tc-99, which are released in total ~ 0.07 atoms per fission. The channels with long-lived FPs recovered during reprocessing are placed in lead reflector. Special channels with Sr and Cs are planned to be installed in the reactor as stable heat sources with the power of ~ 0.02 % N nom.

Studies are under way on the option of a subcritical reactor with a neutron source created in lead by a beam of protons injected by a cyclic accelerator.

The feature of ~ 0.02 ΔK/K subcriticality with CBR~1 allows to do without reactivity control means while avoiding reactor runaway both under credible and hypothetical accidents.

If accelerator adds about 10 % to the cost of the plant then such design could be economic acceptable.

The proton beam is injected vertically in an axial channel in which the lead level is regulated by gas pressure. With :

- proton energy 1 GeV,
- average current 7 mA,
- proton-neutron multiplication factor ~ 20,
- neutron importance in the core center ~ 1.5,
- subcriticality 0.02 ΔK/K,

the thermal power of the reactor will be close to 700 MW, with thermal and neutronic characteristics close to these of BREST-300.

Figure 2 - Cross-section of BREST core

MAIN CHARACTERISTICS OF REACTORS

Characteristic	BREST-600	BREST-300
Electric power, MWe	600	300
Number of FA in the core, pcs.	357	185
Core diameter, mm	3190	2300
Core height, mm	1200	1100
Length of a fuel rod, mm	9.1 ; 9.6 ; 10.4	9.1 ; 9.6 ; 10.4
Lattice pitch, mm	13.6	13.6
Fuel	UN + PuN + MA	UN + PuN + MA
(U+Pu)N load, t	28	16
Pu/Pu-239 + Pu-241 load, t	3.73/2.72	2.2/1.6
Fuel life, years	5-6	5
Time between refuellings, years	1-2	1
CBR	~ 1	~ 1
Inlet/outlet lead temperature, °C	420/540	420/540
Maximum cladding temperature, °C	650	650
Maximum lead flow, m/s	1.8	1.8
Power effect, % ΔK/K	0.17	0.16
Total reactivity margin, % ΔK/K	0.33	0.32
β eff, %	0.35	0.35
Steam temperature at SG outlet, °C	520	520
Steam pressure at SG outlet, MPa	24.5	24.5
Thermal efficiency, %	~ 44	~ 44

III. SCENARIO OF NUCLEAR POWER DEVELOPMENT IN RUSSIA

It was assumed that after 2010 the growing demand for electricity production will be met mainly due to construction of large NPPs with the discussed reactors (up to 150 GWe by 2050 and 300 GWe by 2100) with an on-site closed transmutation fuel cycle. Besides, small nuclear plants with thermal reactors will be constructed for local electricity supply and district heating, such as LWR, HTGR, etc. (equivalent to 50 GWe). Spent fuel from these reactors will be reprocessed at central reprocessing plants, with recovered U returned to thermal reactors and Pu and MA recycled in fast reactors (together with weapon-grade Pu stockpiled as a result of arms reduction). As the growth rate slows down, thermal reactors will gradually be converted to mixed U-Pu fuel due to certain overproduction in fast reactors. Another open option is

turning to Th-U cycle. After 2100 the growth of nuclear capacity and natural Uranium consumption is halted and over T years nuclear energy remains at the level of 350 GWe, consuming only stockpiled depleted uranium (which will be sufficient for T~3 K years). The stable period might last longer, then it may prove necessary to resume uranium production at a minor level (some 400 t per year) in case T > 3 K years.

About 900 K tU (Mu) will be produced up to 2100 (including already mined uranium).

The stable period T may prove much shorter if some novel energy technology, preferable in this or that respect, is developed. In this case, T might be followed by a fairly long final period of a step-by-step decommissioning of NPPs and transmutation of radioactive materials. Dedicated reactors (burners) may prove necessary to allow completing this stage in 100 years.

Assuming that 1 GWe entails 0.8 t FP per year, nuclear power of N(t) GWe will produce annually 0.8 x N(t) tFP/y, with total production up to T : 18(1+0.016 T)K t. Consumption of natural uranium per 1 tFP (currently amounting to about 200 tU/tFP) will decrease by 2100 to 50 tU/tFP and to 1 tU/tFP in ~ 3000 years.

Calculations were performed based on the VVER-1000 fuel performance as representing thermal reactors. For convenience, estimates below refer only to fast reactor fuel.

IV. FUEL COMPOSITION AND IRRADIATION CHARACTERISTICS

If MA is used in the main LCFR fuel, in 10-15 cycles the fuel composition will be close to equilibrium as summarized in Table 1 (15 cycles, stored during 1 year, % at.).

Calculations show that Cm transmutation drastically increases radiation level and heat release during fuel fabrication. Hence the Cm fraction ($\varepsilon \sim 1$ %) should better be extracted and placed into storage so that Cm-242 and Cm-244 would decay.

Table 1

Th-230	U-232	U-238	Np-237	Pu-236	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	Am-243	Cm-244	Cf-249	FP
2	.3.2	74	0.36	6	0.55	9.5	5	0.72	0.56	0.32	0.2	0.11	8.7	8.7
(-5)	(-5)			(-6)									(-5)	

Table 2 describes irradiation characteristics of the fuel at the fabrication stage under two options : transmutation cycle with MA (without Cm) recycling and traditional cycle without MA recycling (γ and n radioactivity at 1 m distance from FA).

Table 2

	α, Ci/kg	q, W/kg	γ, μrem/s	n, μrem/s
U-Pu-MA	72	2.3	45	16
U-Pu	28	0.9	14	7

Adoption of the transmutation fuel cycle entails higher heat generation and radiation exposure which can be compensated by protection and engineered measures.

V. FUEL REPROCESSING AND FRACTIONATION

Development effort for the IFR concept (ANL, INEL) revealed that small on-site reprocessing should preferably be based on the electrochemical technology of fuel fractionation and species extraction from molten salts. Such studies are currently under way also in Russia (NIIAR) and Japan (JAERI). VNIINM studied applicability of electrochemical technology for the LCFR nitride fuel, including laboratory experiments.

Calculations were performed for the following loss levels. Promising values are given in brackets ; on the right-fraction management.

P, Pu - 0.1 % (0.01 %), MA - 1 % (0.1 %). Fabrication of new fuel
Sr - 5 %, Cs - 15 %. Utilization, sources of heat and γ radiation
I, Tc - 10 % (1 %). Transmutation in the reactor

Radwastes are compacted and stored on site for 200 years.

Transmutation of I-129 and Tc-99 was studied for a 300 MWe reactor. Channels with I (0.4 g/cm^3) and Tc (5 g/cm^3) are located in lead reflector in the second row. Calculations taking into account neutron flux depression and resonance self-shielding showed that 90 % of Tc-99 atoms burn up in 15 years, I-129 atoms - in 23 years.

Equilibrium Sr-90 and Cs-137 amount to 205 kg (5.6×10^7 Ci, 187 kW) and 913 kg (1.5×10^8 Ci, 384 kW) per 1 GWe. They are placed in steel channels, probably together with I and Tc, to create a steady heat source in lead (about 600 kW, 0.025 % of the nominal power) to prevent its freezing during long outages.

Part of Cs can be used for setting up an on-site irradiation processing shop.

As radwastes are stockpiled, a RW storage will be constructed on site phase-by-phase over 200 years. Phase I should accommodate over the initial 50 years approximately 40 t FP (based on 1 GWe) and should be designed to provide heat removal of about 100 kW. Cm is located in special channels.

FPs in metal matrix with the concentration of $\sim 3 \times 10^3$ Ci/l and heat generation up to 20 W are placed in steel tubes D * H = 0.2 * 5 m. 120 such tubes are located in a concrete vault (30 cu. m) cooled by natural closed circulation of dray air heated to $\sim 150°C$, with heat removed in air-air and air-water heat exchangers. The storage volume is $\sim 600 \text{ m}^3$, estimated cost - less than 1 % of the plant cost.

The absence of U blanket and production of weapon-grade Pu, low reactivity margins, U-Pu-MA fuel, on-site fuel cycle prevent Pu thefts and its military application. Other security measures are planned, which together with political measures will ensure non-proliferation.

VI. RADIATION EQUIVALENT RADWASTE DISPOSAL

Uranium production is planned to be carried out with co-extraction of long-lived α active decay products (Th, Pa, Ra etc.) transmuted in reactors. This will contribute to recultivation of uranium mining areas while "giving space" for subsequent RW disposal in naturally radioactive formations.

Radioactivity of 1 t of natural uranium together with its decay products is worth 5.2 Ci. Mass of U equivalent in terms of radiation risk to the amount of radionuclide X (Λx Ci/tFP) accounting for 1 t of FP is :

$$m_x^0 = \frac{A x . \dfrac{Ci}{t_{FP}}}{5.2 . \dfrac{Ci}{tu_{Ci}} K_z^x} \left[\frac{t_u}{t_{FP}} \right],$$

where K_z^x is ratio of permissible concentrations (in Ci) in water with radionuclide X and uranium. Recently Annual Limits of Intake (ALI) were used in calculations. After reprocessing (εx is the portion of radionuclide X remaining in RW) and storage over time τ the mass of uranium equivalent to disposed RW is :

$$m_x^1 = \frac{m_x^0 . \varepsilon x . fx(\tau)}{K_m^x} \left[\frac{t_u}{t_{FP}} \right],$$

where K_m^x is the factor accounting for the difference in radionuclide X and uranium migration from repository to the surface.

Short-lived radionuclides with T 1/2 ≤ 100 years :

Sr (29 years), Cs-137 (30 years), Sm-151 (90 years), U-252 (72 years) etc. are stored in the repository in equilibrium amounts.

$$Mx = \frac{0.8 . m_x^1 . N(t) \dfrac{t_{FP}}{year}}{\lambda_x} [t_u],$$

where λ_x is decay constant.

The time of RW storage was taken as $\tau = 200$ years since over this period the amounts of Sr-90 and Cs-137 are reduced by a factor of two orders, while the further storage is ineffective due to slower decay of Sm-151 and actinoids.

Radionuclide migration is the most uncertain factor. The data show that most radionuclides (except for I) feature much greater retention as compared with U, however, it is difficult to account for various disturbances, both natural and man-made. With short-lived radionuclides, however, stable physical and chemical forms given to RW before disposal could reasonably be expected to hold for the whole of their life and hence $K_m \sim 10$ can be taken as a minimum estimate. Maximum estimated equivalent masses of short-lived nuclides are given below for N = 350 GWe.

Table 3

Nuclide	Sr-90	Cs-137	Sm-151	U-232
$m_x^0 \frac{t_u}{t_{FP}}$	$2.4\ 10^5$	$6.7\ 10^4$	$1.1\ 10^3$	$1.6\ 10^3$
ε, %	5	15	100	0.1 100
M_x, tU	$1.6\ 10^5$	$1.2\ 10^5$	$7\ 10^5$	600 $6\ 10^5$

Hence, the terms of radiation equivalent disposal of short-lived portion of radwastes adopted in the assumptions are fairly closely satisfied.

Long-lived I-129, Tc-99 and Cs-135 contribute rather insignificantly to the radioactivity of the disposed RW even under long-term development of nuclear power with uranium consumption at the level of 1 tU/tFP.

Table 4

Nuclide	I-129	Tc-99	Cs-135
ε, %	10	10	100
m_x, tU/tFP	1	0.1	0.08

It was adopted in the calculations that $K_I = 0.3$, $K_{Tc} = K_{Cs} = 1$. Kr was taken based on permissible concentration in water. Calculations based on ALI show significant reduction of m_I and increase of m_{Tc}.

If stored for 200 years, actinoid fraction of RW will be reduced in terms of equivalent mass of U only five-fold ; the level of RW treatment (ε) is a major importance.

Assuming for long-lived nuclides $Km = 1$, equivalent mass of U for actinoids amounts to : 91-180 tU/tFP with $\varepsilon = 0.1\%$ and 9.1-18 tU/tFP with $\varepsilon = 0.01$, as calculated based on permissible concentration in water and ALI. If $\varepsilon = 0.1\%$ the results fall by a factor of 2-3 beyond the radiation equivalency, even with $T = 0$ (specific U consumption ~ 50 tU/tFP), which is within the uncertainty of the present estimates. We see that long term nuclear development will strongly depend on improvements of reprocessing technology.

VII. THE FINAL PHASE

On this stage the use of U-Pu fuel (reactors with Pu reproduction) delays the termination of nuclear power.

Accelerated termination of nuclear power is possible due to disposal of U, introduction of burners operating mainly on MA (for instance, fast reactors with liquid fuel or, probably, accelerator burners). This allows to reduce the final phase to 100 years or even shorter.

VIII.COMPARISON WITH THE Th-U CYCLE

Drastically smaller production of MA as compared with the U-Pu cycle is considered as a major argument for adopting Th-U cycle in the future. Since this cycle shows advantages over U-Pu in its neutron and fuel balance only in case of thermal reactors (with fast reactors it is much inferior to U-Pu cycle), fast reactors with a transmutation fuel cycle were compared with heavy water reactors with a closed Th-U fuel cycle with and without MA transmutation under recycling.

As follows from Fig. 3, in terms of waste radioactivity the Th-U cycle is inferior to transmutation U-Pu cycle of anyway, is not superior to it. Since in the Th-U cycle thermal reactors are inferior to fast reactors in many aspects such as neutron and fuel balance, many safety features (reactivity margin, βeff etc ...), burnup and reprocessing volume, fast reactors are preferable for large NPPs provided they become cost effective.
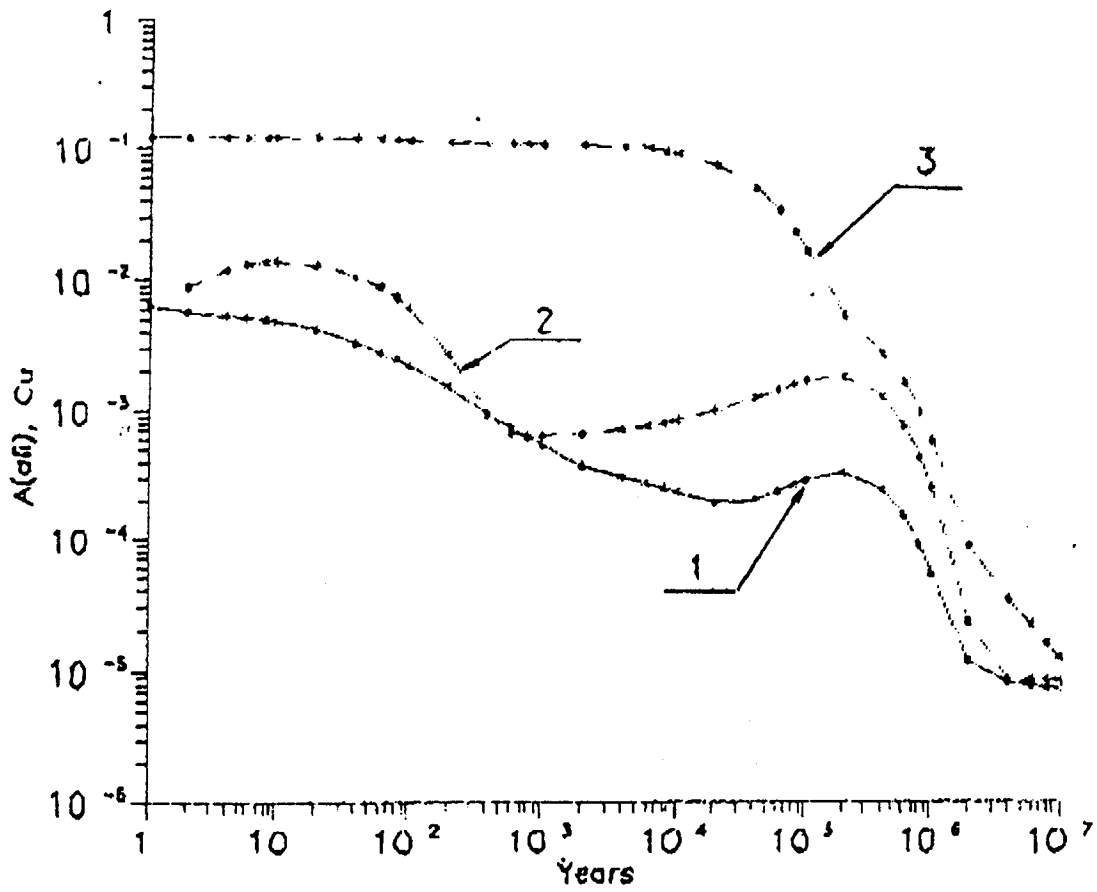
Figure 3
Actinoide waste activity per 1 MW.day (ALI equivalent)
from LCFR (U-Pu) [1] and CANDU (Th-U) [2] P/T fuel cycle
and CANDU (TH-U) cycle without transmutation [3]

At the same time fast reactor looses some of its advantages in case of small power, hence thermal reactors of different types may prove preferable for local heating and electricity production. For them Th-U cycle shows certain advantages.

IX. CONCLUSION

Analysis of the concept of the U-Pu fuel cycle with transmutation in fast reactors in terms of physics points to the feasibility of radiation-equivalent radwaste disposal without upsetting the natural radiation levels, provided a number of technical problems are solved.

These primarily include creation of a reactor which would rule out accidents with fuel failure and catastrophic radioactive releases.

A major prerequisite is the development of a radiochemical technology capable of removing Pu and MA from radwastes with a residue of no more than 0.1 % and of isolating the fractions of I and Tc (< 10 %) and Sr and Cs (5-15 %). Cm should be advisably separated from the fuel for long-term cooling in a storage. If nuclear power stays in service for several centuries, still finer fractionation will be needed. Utilization of Sr and Cs, and maybe other radionuclides, will make it possible to reduce the time of radwaste cooling to an acceptable stretch of about 200 years.

Both rehabilitation of U mining sites and radiation-equivalent disposal of radwastes call for mastering the technology of U mining with co-extraction of its long-lived decay products (Th, Pa, Ra, etc ...).

There is still much time left for developing a new technology of radwaste disposal, but the possibilities of creating compositions to prevent migration of radionuclides from burial grounds need investigation and demonstration.

THE CONSUMPTION OF ACTINIDES IN ADVANCED LIQUID METAL REACTORS (ALMR)

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This Work was performed by the organizations represented by the authors.

ABSTRACT

This paper discusses an actinide recycle system, the ALMR, with its fuel cycle, capable of consuming actinides produced in existing light water reactors. The ALMR has a wide range of capabilities, including the use of existing actinides to produce additional actinides for future electrical energy production, if needed; to the consumption of actinides. Due to the worlds' nations attention on what to do with actinides, the focus of this paper is the consumption of actinides. This results in the elimination of safeguards of nuclear waste produced in nuclear reactors that is scheduled for storage in repositories. Rates of actinide consumption are identified.

In US nuclear energy scenario analyses for the 1991 National Energy Strategy (NES), a "lower reference" case was developed which assumed a modest growth of nuclear power capacity from the presently installed capacity of about 110 GW(e) to 195 GW(e) by 2030. This capacity growth is primarily with advanced light water reactors (ALWR) but assumes a modest growth of advanced liquid metal reactors (ALMR) starting after 2010. The specific ALMR growth depends on the assumptions associated with the "breeding" or "converting" of actinides in ALMR designs. Breeding creates more plutonium than is "burned" and converting burns more plutonium than is created.

To gain an understanding of the potential for modest growth of ALMRs, a case was analyzed for the NES in which a growth of ALMRs was chosen sufficient to consume essentially all of the LWR origin transuranics produced from ALWRs introduced coincident with the ALMR growth. An ALMR can consume *actinides* (elements of atomic number 89 and greater). Spent fuel from LWRs consists of roughly 96 percent uranium, one percent transuranics (TRU - plutonium, neptunium, americium, curium, etc.) and the remainder (3 percent) is fission products. Thus, if processing of LWR spent fuel is pursued, a system exists in which energy can be produced from LWR-origin spent fuel material that would otherwise be disposed of as waste. The energy produced in ALMRs from LWR "waste" can easily exceed the original LWR energy production by factors as high as 60 to 100.

In producing power, an ALMR annually consumes a small fraction of its core fissile/fertile material content (i.e., out of about 100 tonnes of core material, approximately one metric ton is needed to produce a gigawatt-year of electrical energy in a large ALMR plant). Therefore, during ALMR growth, the majority of actinides available from spent LWR fuel will be utilized for initial core inventories. Nevertheless, approximately one metric ton of actinide material per gigawatt-year will always be converted to fission products, the source being either uranium if the ALMR operates as a *breeder*, or make-up transuranics and uranium if the ALMR operates as a *converter*. Although current NES analysis to 2030 uses an ALMR "converter", economic performance of a "breeder" appears somewhat better than a "converter".

Therefore, commercialization of the ALMR may be achieved more readily in the "breeder" or "near-breeder" design. Nevertheless, a sufficient number of ALMRs could eventually be deployed offering the potential for operation as "converters" to significantly reduce actinides from LWR spent fuel.

To illustrate possible breeder and converter deployment, one breakeven and two converter cases (see Figures 1 through 3) are presented in which ALMR growth of 27 GW(e) to 2030 was assumed as part of the NES growth scenario. The independent variable indicated in Figures 1 through 3 was taken as the conversion ratio (CR) for the reactors, ranging from a low of 0.6 up to 1.06 (i.e., threshold "breeding"). The general plant design used for ALMRs was that of the current GE advanced liquid metal reactor concept (1866 MWe). Table 1 shows the key TRU (transuranic) parameters for the three cases and Figures 1 to 3 present the results. The three cases generally show that with modest ALMR growth all the transuranic material from LWR spent fuel produced during ALMR growth can be utilized in the ALMRs, thereby essentially being removed as material requiring long-term high level storage. This is true even for a breeder growth case, because of core inventory growth.

After a certain growth period, such as up to 2075, sufficient ALMRs, could be in existence so that if they were operated as converters, their internal consumption would be adequate to consume TRU from all existing LWR spent fuel. A ratio of about one ALMR at CR = 0.7 for each two-to-three LWRs could provide a symbiotic state regarding TRU production and use to that time frame (see Figure 4). At these ALMR deployment and TRU consumption rates, fissile material from some other source would be required for additional ALMR deployment to occur.

It should be noted that uranium is a major by-product of processing LWR-origin spent fuel. Significantly more uranium is recovered than is required for the ALMR growth analyzed for the NES. At the present time, the proper future role for the excess recovered uranium, which is slightly enriched to about 0.8%, could be storage as potential future ALWR or ALMR fuel or could be disposal as waste. Utilization of this uranium in ALWRs would require reenrichment and is being evaluated as part of the LWR actinide recycle program

For LWR spent fuel actinide recycle in the ALMR to be beneficial (and accepted) it is important that the system not create more waste and/or it must improve the waste form compared to the conventional once-through LWR system. Through the use of pyroprocessing (an innovative process for recovery of actinides from irradiated fuel) the waste generated per unit of electricity produced is estimated to be less than direct disposal of spent fuel in the repository. However, more importantly, the repository heat load after about 300 years approaches zero and the waste form is improved over the direct disposal of spent fuel. It is estimated that isotopics of concern (e.g., Tc99 and I129) released to the environment from the repository are decreased about three orders of magnitude. Additional analysis of the pyroprocess waste form is needed in the development program but preliminary results are favorable, as they are for the pyroprocess planned for recycle of ALMR spent fuel.

There is an estimated cost penalty of one to two mills/kwh for recovery of LWR spent fuel actinides over exclusive recycle of ALMR spent fuel. This penalty is for the initial cores for ALMR startup which would need to be fabricated using some fissile source other than LWR TRU if it were not available which would also represent some penalty. If recycled ALMR fuel or weapons grade plutonium were used, the penalty should be less. In these cases, however, there is only enough excess weapons plutonium to start up about three ALMRs and there will be no recycled fuel from ALMRs until there is an ALMR infrastructure and a significant number of ALMRs producing electricity.

It is concluded that there is a major incentive to recycle LWR spent fuel TRU to the ALMR to: (1) recycle a valuable resource capable of producing electricity indefinitely in the ALMR; (2) improve the waste form to impede release to the environment, and (3) remove fissile material from the repository to preclude the requirement for safeguards in perpetuity.

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Table 1. ALMR Alternative Core TRU Data

	Heterogeneous Core	Homogeneous Cores	
Breeding or Conversion Ratio	1.06	0.7	0.6
Total Pu Consumed (makeup)/yr by 1866 MWe (kg)	-174	500	725
Total Pu Core Inventory for 1866 MWe (MT)	16.1	15.3	23.3
Total Pu Core Inventory Plus 2 Reloads for 1866 MWe Startup (MT)	25.9	23.4	35.6

DISPOSITION OF LWR-ORIGIN TRU WITH SMALL BURNER ALMR DEPLOYMENT
TO CAP AT FIRST REPOSITORY
(NES Final Scenario - 195 GWe by 2030)

TOTAL TRU FROM LWRS

TRU IN STORAGE

TRU IN-PLANT IN ALMRS

TRU CONSUMED

LWR-ORIGIN TRU, MTHM

1200
1000
800
600
400
200
0

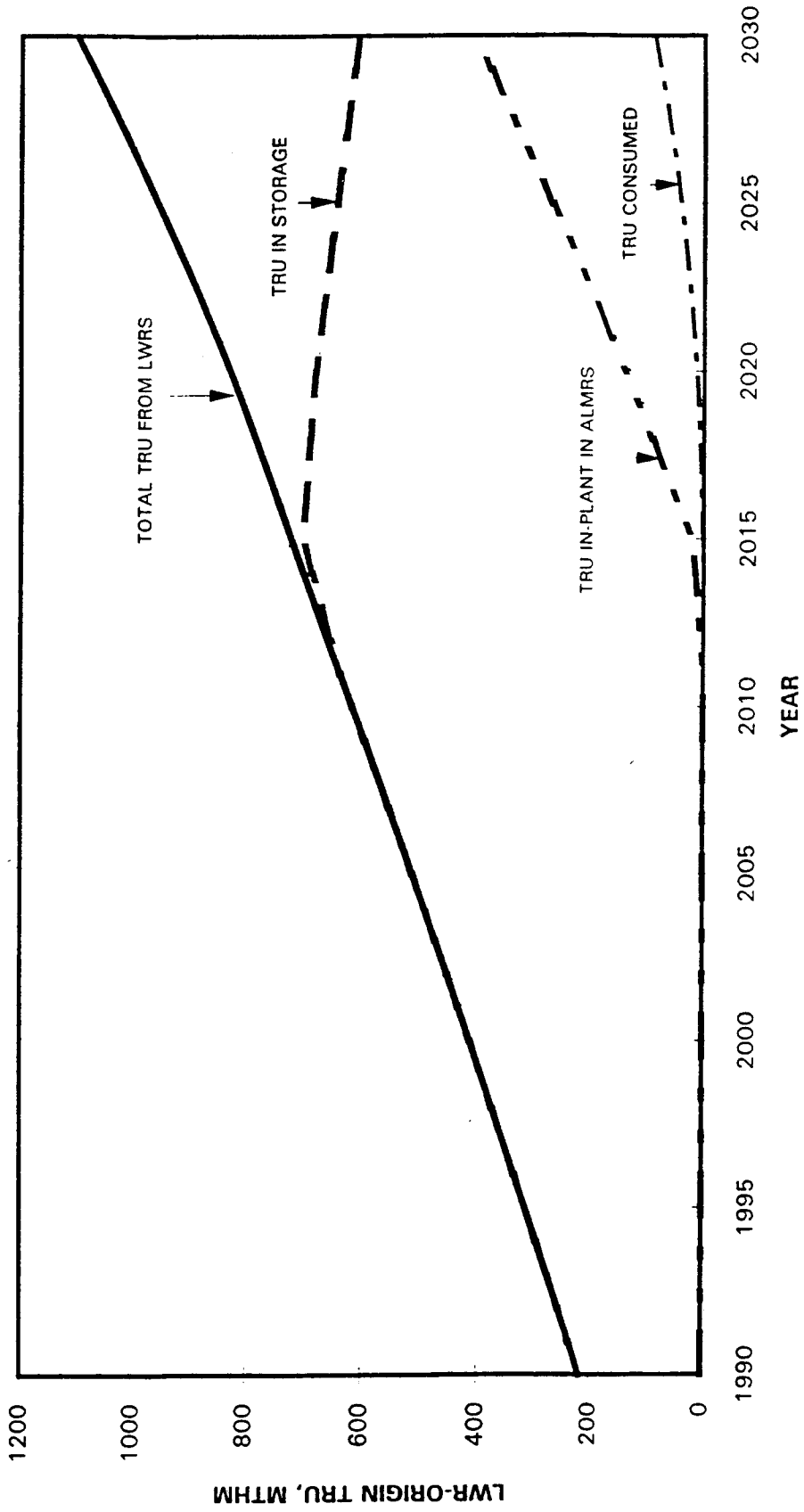1990   1995   2000   2005   2010   2015   2020   2025   2030

YEAR

Figure 1

SMALL BURNER ALMR, CR = 0.72, DEPLOYMENT RATE TO CAP AT FIRST REPOSITORY

DISPOSITION OF LWR-ORIGIN TRU WITH LARGE BURNER ALMR DEPLOYMENT TO
CAP AT FIRST REPOSITORY
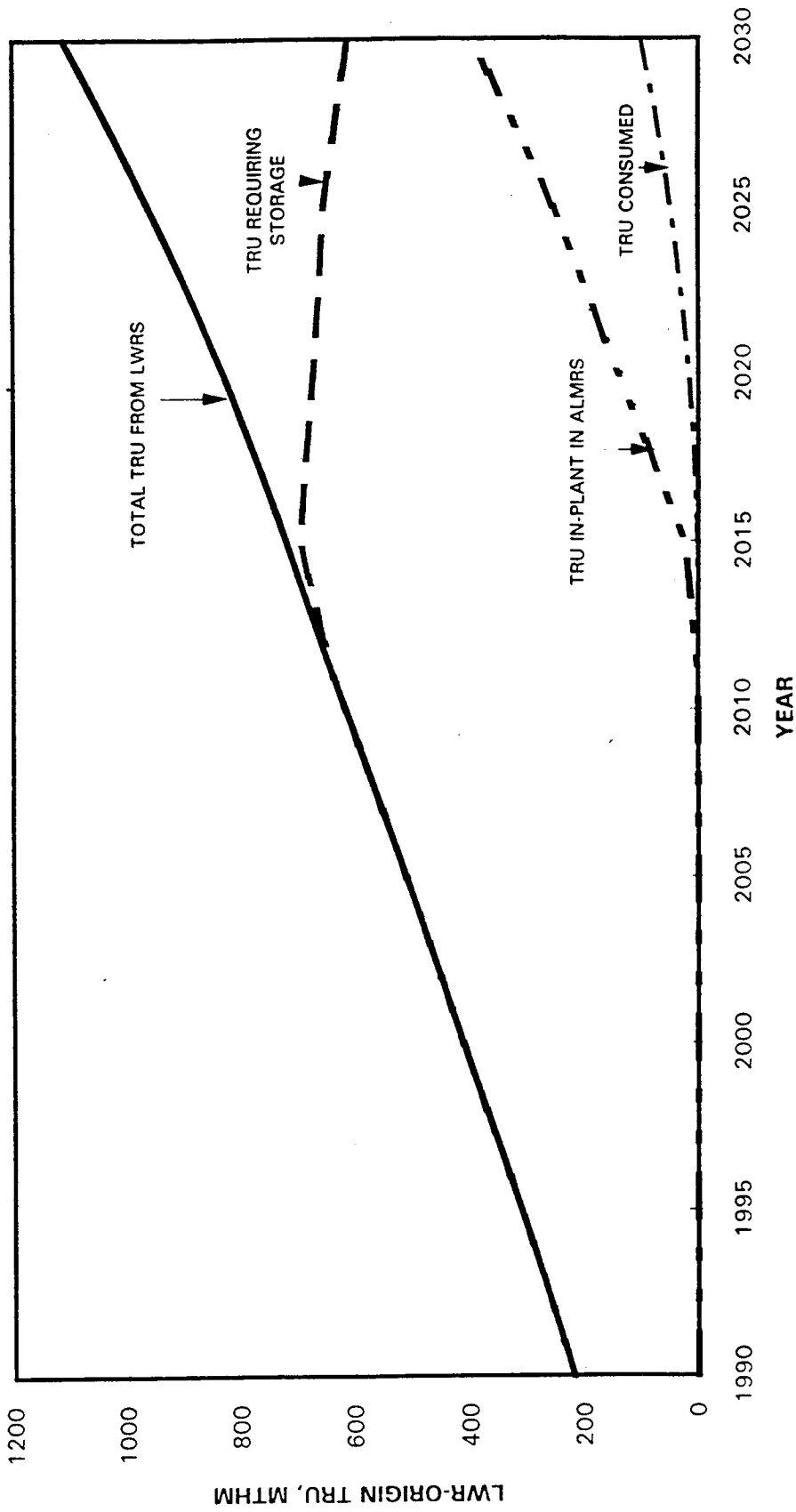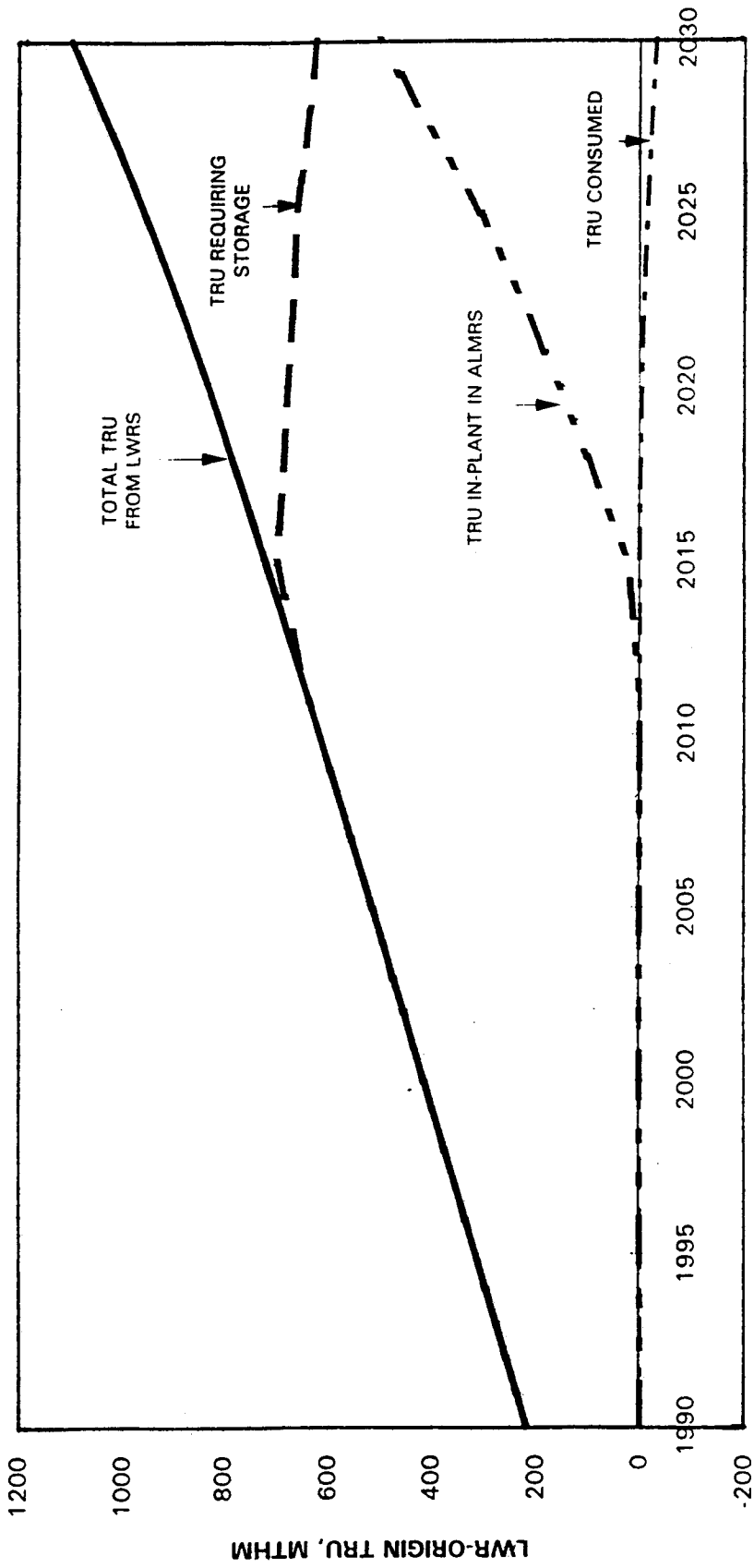(NES Final Scenario - 195 GWe by 2030)

Figure 2

LARGE BURNER ALMR, CR = 0.59; DEPLOYMENT RATE TO CAP AT FIRST REPOSITORY

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DISPOSITION OF LWR-ORIGIN TRU WITH BREAKEVEN ALMR DEPLOYMENT TO
CAP AT FIRST REPOSITORY STRATEGY
(NES Final Scenario - 195 GWe by 2030)

TOTAL TRU
FROM LWRS

TRU REQUIRING
STORAGE

TRU IN-PLANT IN ALMRS

TRU CONSUMED

LWR-ORIGIN TRU, MTHM

YEAR

Figure 3

BREAKEVEN BREEDER ALMR, CR = 1; DEPLOYMENT RATE TO CAP AT FIRST REPOSITORY

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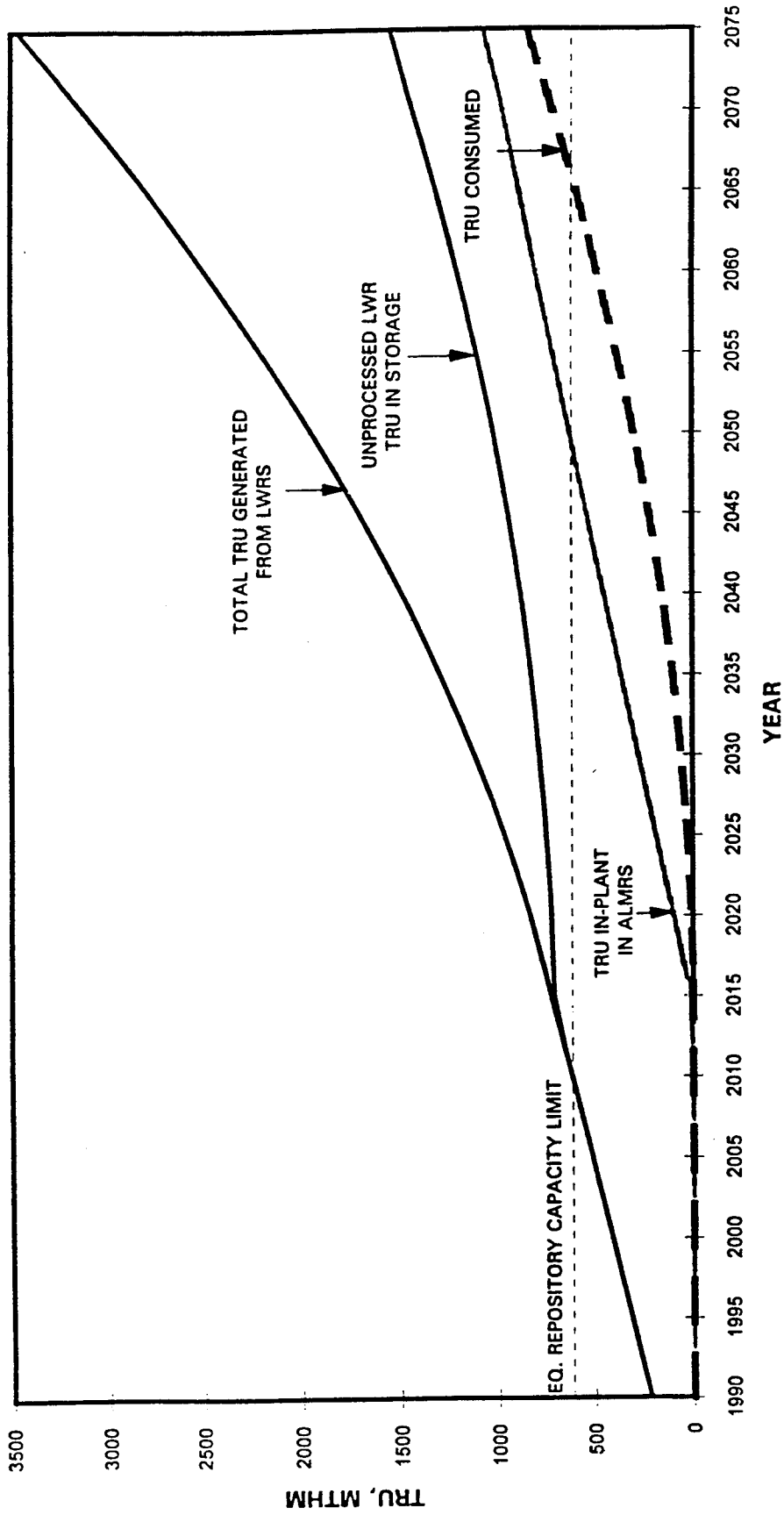EFFECT OF ACTINIDE RECYCLE ON LWR-ORIGIN TRU INVENTORY, SMALL BURNER ALMR DEPLOYED (27 GWE BY 2030)

Figure 4

SMALL BURNER ALMR; DEPLOYMENT RATE AFTER 2030 SAME AS AT 2030; 20-YR PLEX FOR BOTH ALMR LWRS

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