CEA AND PNC VIEWS ON CRITERIA AND RESEARCH GUIDELINES ON PARTITIONING AND TRANSMUTATION

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1. INTRODUCTION

Recognizing the importance of the long-term effort to maximize the utilization efficiency of uranium, both France and Japon have a similar nuclear energy strategy toward the 21-st century which is based on the closed fuel cycle option to reprocess spent LWR fuels and to recycle recovered plutonium in LWRs and fast reactors (FRs).

And now, both countries have been starting to develop an advanced fuel cycle to reduce long-lived radiotoxity in the deposed waste applying minor actinide (MA) recycling and burning. Long-lived radioactive waste management is of great concern for the population as well as for the environment. Therefore, this development is technically satisfactory and socially acceptable for the final wastes from today's and future nuclear industry, and is one of the major challenges of the coming decades.

In France, the December 30, 1991 law stipulated that three lines of research were to be pursued over a 15-year period :

- Partioning and transmutation of long-lived radionuclides,
- Conditioning of waste with a view to long-duration interim storage,
- Deep geological disposal.

In Japan, starting as a part of the effort of the OMEGA project, PNC has been conducting research on partitioning and transmutation. Efforts in this area have been recently upgraded as the "Advanced Fuel Recycle Program" which intends to optimize the future fuel cycle by putting further efforts to reduce burden to the environment from disposed wastes, and to burn minor actinides in fast reactors. PNC envisions that the effort to positively include minor actinides in the plutonium fuel cycle would result in the enhancement of resistivity against plutonium diversion.

PNC also intends to reduce fuel cycle cost by modification and simplification of the fuel cycle system drastically in the FBR commercialization stage, introducing low decontamination reprocessing, changing the fuel form, and so on.

The programs of both organizations share similar philosophies and have common technical targets in many areas. The collaboration between CEA and PNC in these areas have started to promote efficient progress of R&D in both organizations.

2. CRITERIA FOR PARTITIONING AND TRANSMUTATION

2.1 CEA Criteria

RADIONUCLIDES TO BE CONSIDERED :

The starting point of the study is the inventory in radionuclides of the fuel from a PWR 900 irradiated to 33 $GWDt^{-1}$ (UOX enriched at 3.25 %) and cooled for three years, the assessment of its radiotoxicity and the search for ways to reduce it.

The irradiated fuel discharged per year from a PWR 900 MWe contains 23 t of Uranium (with 0.9 % of ^{235}U), 240 Kg of Pu, 800 Kg of fission Products, 18 Kg of minor actinides.

Its radiotoxicity can be defined outside any notion of confinement barrier, by calculating a "source term" obtained by weighing the activity of each radionuclide by its specific toxicity coefficient (upon ingestion or inhalation) and then summing the resulting values.

Table I gives the value of the potential radiotoxicity of a fuel and the contribution of each long-lived radioisotope, brought back to its precursor present today. These values are expressed in Sieverts per TWhe produced.

After decay of the highly radioactive fission products (Cs 137 - Sr 90) estimated at 300 years, the main contribution comes from plutonium up to 10^5 years. Neptunium becomes important starting from about 10^5 years. Curium makes a large contribution before 10^4 years.

The contribution of the fission products is negligible.

In the event the confinement barrier is adopted, for deep disposal for instance, it is necessary to assess the return of the radionuclides to the biosphere according to the degradation of the waste packages with time and to the transport of radionuclides through the geological layers.

These models are complex and depend upon the sites. A study performed in 1990 (PAGIS) for glass packages (containing neither Uranium, Plutonium nor Iodine) in a granitic site (AURIAT) showed that no dose was released before 10^4 years, that it subsequently remained lower than the limit recommended by the ICRP and that the elements to be considered for storage safety were the most mobile ones (Technetium, Cesium) and the most radiotoxic (actinides) (See Figure 1).

STRATEGY DEVELOPED, THEORETICAL GAINS

Reducing radiotoxicity consists in not leaving with the waste the radioisotopes concerned, first of all plutonium and then the minor actinides.

Figure 2 quantifies the theoretical gains thus obtained. They may reach a factor of 500 between the once-through cycle where the irradiated fuel is sent to waste and that where the plutonium and the minor actinides are separated with respective yields of 99.9 % (Pu) and 99 % (minor actinides).

ADVANTAGE OF REPROCESSING

Reprocessing irradiated fuels, thereby recovering the energetic material, already makes it possible to separate 99.5 % of the plutonium (guaranteed value), soon to reach 99.9 % thanks to the effort made in the PURETEX program to improve reprocessing waste management in order to reduce their volume and activity.

Reprocessing also allows to consider strategies for separating the other radionuclides in line with today's techniques, either by modifying the processes (Np, Tc, I, Zr), or by adding separative operations on the high activity waste (Am, Cm, Cs).

PLUTONIUM

The separated products must be used or eliminated. Use of Plutonium in power reactors is a well established technique. However, multi-recycling and overall optimization of the system are necessary to achieve the best result in terms both of resources utilization and limitation of radiotoxic isotopes production during multi-recycling.

Figure 3 shows that fast neutron reactors allow to come closer to the theoretical case as far as radiotoxicity source term reduction is concerned. PWRs are effective over one cycle (radiotoxicity divided by three).

If one wants to reduce Pu stocks, FNRs accept highly degraded Pu isotopic vectors and may be adapted to higher consumption of Plutonium. For example, by suppressing the fertile blankets in a SUPER PHENIX type reactor, with some adaptation of the core, the consumption could reach 20 Kg/TWhe. In France, the CAPRA project aims at defining a reactor consuming from 70 to 110 Kg/TWhe (theoretical limit) of Plutonium.

MINOR ACTINIDES

Minor actinides (Americium, Neptunium) may be transmuted in a homogeneous form, uniformly distributed in the fuel or in targets, according to the so-called heterogeneous mode. A physics analysis shows that FNRs burn actinides better than PWRs in the sense that they produce fewer higher isotopes, due to higher σ_f/σ_c ratios for the different isotopes and a better neutron economy in the core.

2.2 PNC Criteria

PNC has similar technical objectives as CEA. In view of reduction of long-lived radiotoxicity, not only recovering minor actinides (MA) but also decreasing the residual plutonium in the recovery process.

In the PNC Actinide Recycles scenario, MA from the LWR and Pu-thermal reactor in the far future are recycled into fast reactor together with recovered Pu. In the advanced fuel recycle system, Pu and MA aré recycled in multi number of times (Fig. 4).

REDUCTION OF BURDEN TO THE ENVIRONMENT

Direct impact to the environment from radioactive waste disposal is identified by the two following concepts.

- Potential hazard of waste (radiotoxicity)
- Isolation of the radiotoxicity from ecological environment (waste treatment and disposal)

Waste treatment and disposal are aimed at technology that actualize no radiotoxicity at the surface of the ground, regardless of level of the hazard potential in the disposed waste.

The advanced fuel recycle intends to essentially reduce the long-lived radiotoxicity and to contain the total MA inventory inside the fuel cycle.

EFFECT ANALYSIS

MA and the residual Pu are the dominant toxicity from the high active waste (HAW) for a long time. The advanced fuel cycle system is aimed at further reduction of the toxicity by recovering MA and reducing Pu losses in the fuel cycle (Fig. 5).

Fig. 6 shows the result of the MA mass balance simulation in Japan, nuclear power generation is assumed to increase 1000 MWe/y and introduction of commercial fast reactor will start in the year 2030. Without recycling of MA, the total MA that is transferred into HAW is calculated to be 310 tonsq from LWR, Pu-LWR and FBR. In the case of recycling MA into commercial FBR from the year 2030, the MA existing and contained in fuel cycle in the year 2100 is reduced to 60 tons. Most of the 60 tons of the MA is contained in the fuel cycle and not into the waste.

3. RESEARCH GUIDELINES

3.1 CEA Guidelines

The CEA research program and guidelines are given in a companion paper presented at this meeting /1/. In fact, in the frame of the SPIN program, the ACTINEX program is devoted to the research and development in the field of partitioning and transmutation.

As far as research guidelines, the transmutation research program has the following objectives :

a) Basic transmutation physics understanding.
b) Basic nuclear data assessment and validation.
c) Reactor concept studies, using both homogeneous and heterogeneous recycling modes for minor actinides. Fission product transmutation in the subject of exploratory studies.
d) Fuel related studies.
e) Innovative system studies, such as hybrid accelerator based systems.

Major achievements are given for point a) in reference /2/ ; for point b), most work is in progress and in the frame of international collaborations.

For point c), a first indication of interesting options (e.g. homogeneous recycling of Np in MOX fuel ; heterogeneous recycling of Am in targets at the periphery of a FR ; use of moderating S/A for fission products in the blanket of a FR etc) is given in ref. /3/.

As far as point d), the SUPERFACT experiment analysis has been completed. Present studies are devoted to both homogeneous recycling (e.g. burn-up increase in specific irradiations in PHENIX ; experiments in PWR environment in the irradiation reactor OSIRIS) and to heterogeneous recycling. For this last program, CEA efforts are made in the frame of PNC-CEA collaboration, of a European collaboration EFFTRA /4/ and in collaboration with PSI-Switzerland.

For point e), results are given in ref. 2. In particular, the present analysis indicates a potential role of these innovative systems in the long-lived fission product transmutation, but not for minor actinide transmutation.

Finally, most of the results and guidelines obtained in this program, are transferred to the CAPRA project, devoted to Pu consumption in FR.

The partioning programme has the following objectives :

a) Behaviour of actinides and long-lived fisison products in Purex reprocessing.
b) Basic chemical data for partitioning long-lived radionuclides in nitric and acidic medium.
c) Studies of slight modification of Purex as far as possible (Tc, Np, I).
d) Developments of new processes for wet recovery of minor actinides and cesium on H.A. wastes.

Here also, more indications are given in companion papers presented at this meeting.

3.2 PNC Guidelines

BASIC POLICY

- The advanced fuel recyling in the FBR commercialization stage is a closed cycle system featuring actinide recycling in the most systematic and efficient way.

- PNC is continuously working on R&D to simplify technology related to the fuel cycle and to close the fuel cycle introducing FBRs which have the potential to burn actinides including these from LWRs.

CHARACTERISTICS OF ACTINIDE RECYCLE TECHNOLOGY

Major items for development in the theree main areas of the fuel cycle is targeted for research and development.

(1) Reprocessing

Achievements of the simplified reprocessing process which does not recover Pu separately but together with minor actinides.

2) Fuel

- Design of fuel containing minor actinides.
- Remote and simplified fuel fabrication technology to produce fuels containing minor actinides.

3) Reactor

Design of an advanced FBR core to burn minor actinides loaded efficiently with U and Pu.

Technologies mentioned above are presently under the development program of the current MOX fuel cycle technology. The progress of development are divided into two streams ; (1)Upgrading the Fuel Cycle Processes, (2)Introduction of Minor Actinides. Introduction of Minor Actinides is a challenging matter requiring progress of engineering. On the other hand, upgrading fuel cycle processes is indispensable for the commercialization use of FBR cycle.

PRESENT STATUS OF R&D, AND FUTURE PLAN

PNC is pursuing the following R&D on the advanced actinide recycling.

(1) Development of Aqueous Reprocessing Technology of Minor Actinides.
(2) Fabrication of Fuel Pellets Containing Minor Actinides.
(3) Irradiation Tests utilize "JOYO".
(4) Developement of Advanced Fuel Fabrication Technology such as by the Sol Gel Method
(5) Analysis and Evaluation of Actinide Burning in the advanced FR.

PNC is planning work on acquisition of basic data on actinides by use of existing hot facilities.

PNC also has plans to utilize "JOYO" and "MONJU" to carry out irradiation tests of fuels containing actinides, and to perform the feasibility studies on a reactor and a fuel recycle plant to demonstrate the advanced fuel recycling.

4. PRESENT COLLABORATION

Table 2 gives the collaborative issues between CEA and PNC.

4.1 Partitioning

Based on the common recognition for long-lived nuclides partioning in the high level liquid waste (HLLW), CEA and PNC have been developing new separation technologies using different bidentade ligands ; DIAMEX process employed with DMDBTDMA and CMPO-based TRUEX process respectively. Collaboration to develop partitioning process has been initiated since November 1991, to evalutate each partitioning process in the various technological aspects to give fundamental extractability, extraction mechanism, molecule design, etc .. for assessing their compatibility as a future nuclear fuel cycle technology. Collaboration has been going on through information exchange by specialists meetings annually and exchanging of assignees to each establishment. This collaboration originally focused only on actinide partitioning by new solvent extraction methods, but agreeably

developed to include the improvement of conventional PUREX process to enhance Pu and Np recovery of long-lived fission products by functional macrocyclic compounds.

The counter-current demonstrations by CMPO and DIAMIDE, using actual HAW arising from the PUREX process in each site, currently proved reasonable MA/FP separation corresponding to their original extraction functions obtained in tube tests.

Comparative test for two extractants has been carried out in satisfactorily by both parties.

The selective extractability were newly reported for functional crown ethers and calixarens analogs capable of maintaining excellent extraction for Cs, Sr and actinides in the nitric acid environments.

4.2 Transmutation

CEA and PNC have been carrying out R&D programs for MA and FP transmutation and the collaboration has been going on since 1993 under the CEA-PNC bilateral agreement. The main subjects of the programs are fuel studies, core physics and core safety.

In the field of fuel development, the experiments and investigations have been implementing to determine the most suitable materials in order to burn long-lived radioactive nuclides for heterogeneous recycling. Irradiation tests in JOYO and PHENIX will be performed to clarify the in-pile behavior of the fuels and targets aiming at transumting and fissioning MA in a fast reactor core.

Feasibility of minor actinide burning will also be demonstrated by operation on utilizing SUPER PHENIX.

For the core physics, the validation of basic nuclear data relevant to radioactive waste transmutation and of appropriate calculation schemes has been implemented using existing experimental data. Additional integral experiments will be performed using MASURCA, JOYO, PHENIX and SUPERPHENX in support of the transmutation studies. Concerning design studies, an intercomparison of the results of parametric survey calculations already performed by CEA and PNC has been made to clarify the basic characteristics for MA and FP transmutation. A common optimized core will be designed by the results of the comparison.

Fundamental studies for core safety on the selected cores will be carried out in design studies.

5. CONCLUSION

Criteria and research guidelines at CEA and PNC are based on similar philosophies and have common technical targets.

The collaborative studies performed of both organizations in the area of partitioning and transmutation have started in many areas to provide an efficient progress of nuclear fuel cycle development.

6. REFERENCES

1. M. VIALA, M. SALVATORES
"The SPIN Program" This meeting

2. M. SALVATORES et al.
Nucl. Sci. Eng. 116 1-18 (1994)

3. J. TOMMASI et al
To be published in Nucl. Technology

4. J.F. BABELOT et al
Paper presented at this meeting

TABLE I

SOURCE TERM
(TRANSURANICS AND LONG-LIVED FISSION PRODUCTS)
AND ITS COMPONENTS : EVOLUTION WITH TIME
(In Sv/TWhe)

TIME (years)	10^2	10^3	10^4	10^5	10^6	10^7
TOTAL (Sv/TWhe)	$1.1\ 10^9$	$3.1\ 10^8$	$7.7\ 10^7$	$4.2\ 10^6$	$5.2\ 10^5$	1.410^5
COMPONENTS (%)						
URT	/	/	0.1	6	29	79
Pu	85	90	97	88	50	17
Np	/	/	/	1.3	13	3
Am	10	9.2	2.5	2.7	6.8	1.4
Cm	0.4	0.3	0.4	/	/	/
FP	4.2	6.10^{-4}	2.410^{-3}	3.210^{-2}	9.610^{-2}	1.410^{-1}

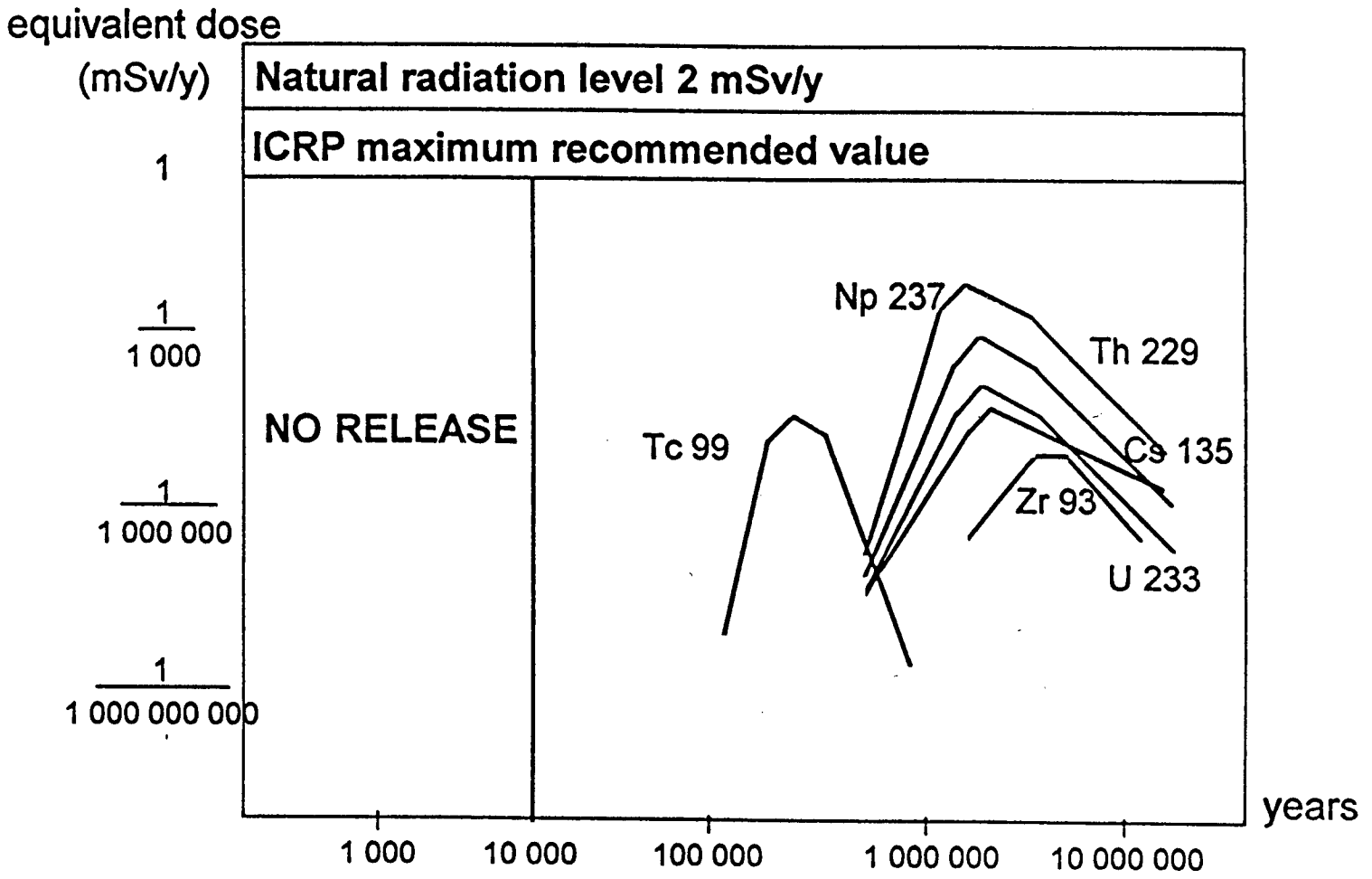
TABLE 2

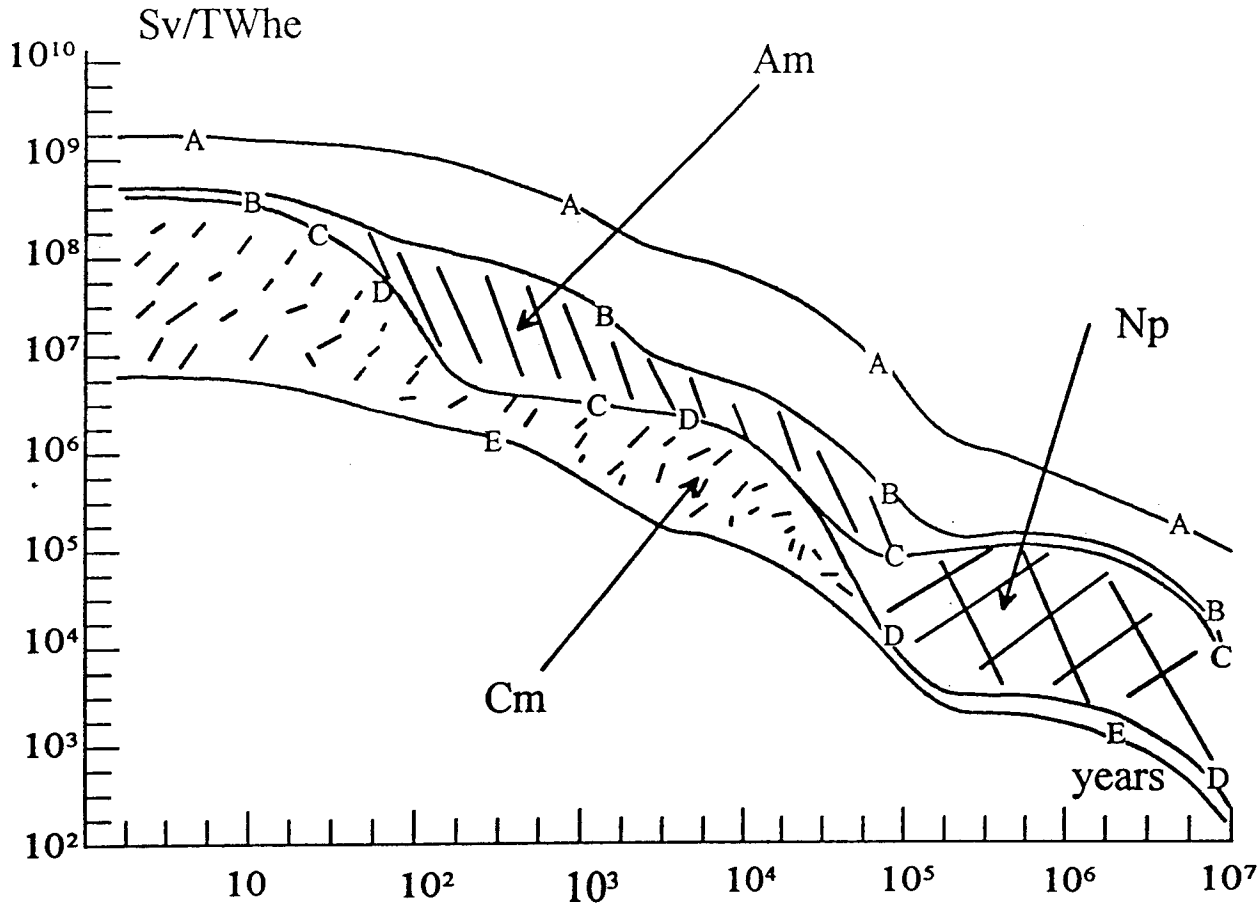
PRESENT COLLABORATION

FIELD	COLLABORATIVE ISSUES
	◊ Asses TRUEX and DIAMEX process as MA partitioning tools. ◊ Research bidentade extractants, CMPO and DIAMIDE ◊ Molecular structure study for innovate ligands ◊ Improve PUREX, to enhance Pu/Np recovery, salt-free process ◊ Separation Long-lived FPs by macrocyclic compounds
TRANSMUTATION	<u>Fuel</u> ◊ Basic Studies and Data on Fuels and Targets ◊ MA Transmutation in a Fast Reactor Core <u>Core Physics and Safety</u> ◊ Basic Studies : Method and Data ◊ Experimental studies ◊ Parametric Studies ◊ Core Safety Studies

RESIDUAL RADIOTOXICITY

Fig.1 AURIAT site (concept A) standard scenario

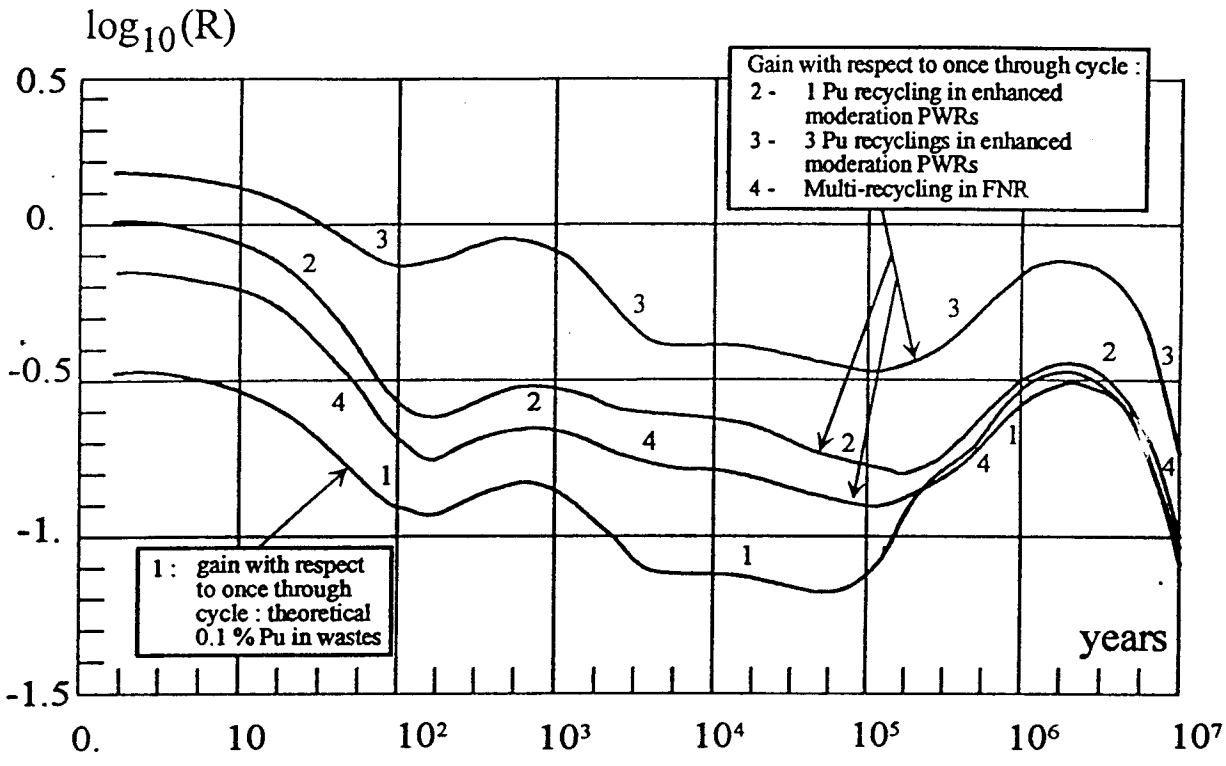




A : once through cycle
B : 0.1 % Pu ; 100 % Am in waste
C : 0.1 % Pu ; 1 % Am ; 100 % Np, Cm in waste
D : 0.1 % Pu ; 1 % Am, Np ; 100 % Cm in waste
E : 0.1 % Pu ; 1 % Am, Np, Cm in waste

RADIOTOXICITY : Theoretical cases

FIGURE 2



$$R = \frac{\text{Radiotoxicity in case (X)}}{\text{Radiotoxicity in once through cycle}}$$

FIGURE 3

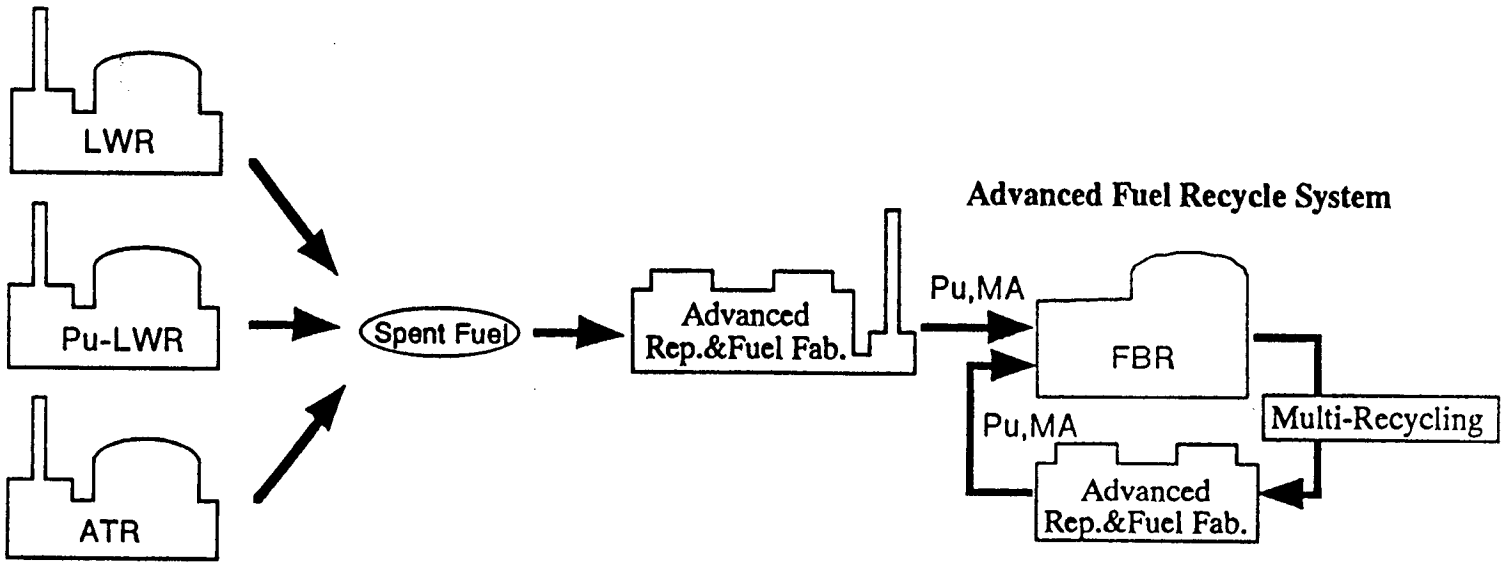


Fig.4 Concept of Advanced Fuel Recycle

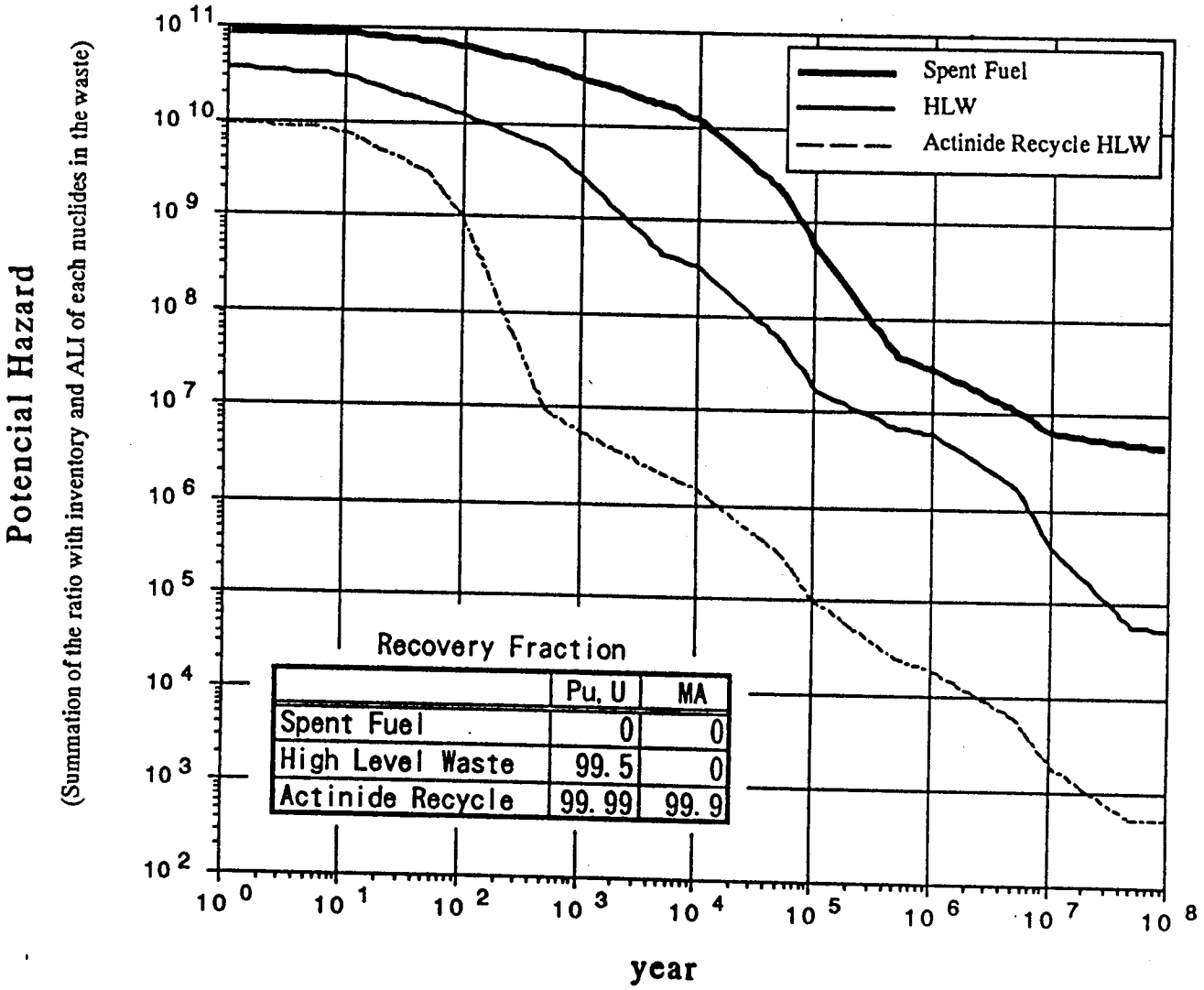


Fig.5 Effect of the Actinide Recovery on the Potencial Hazard of FBR-HLW (150,000MWD/t)

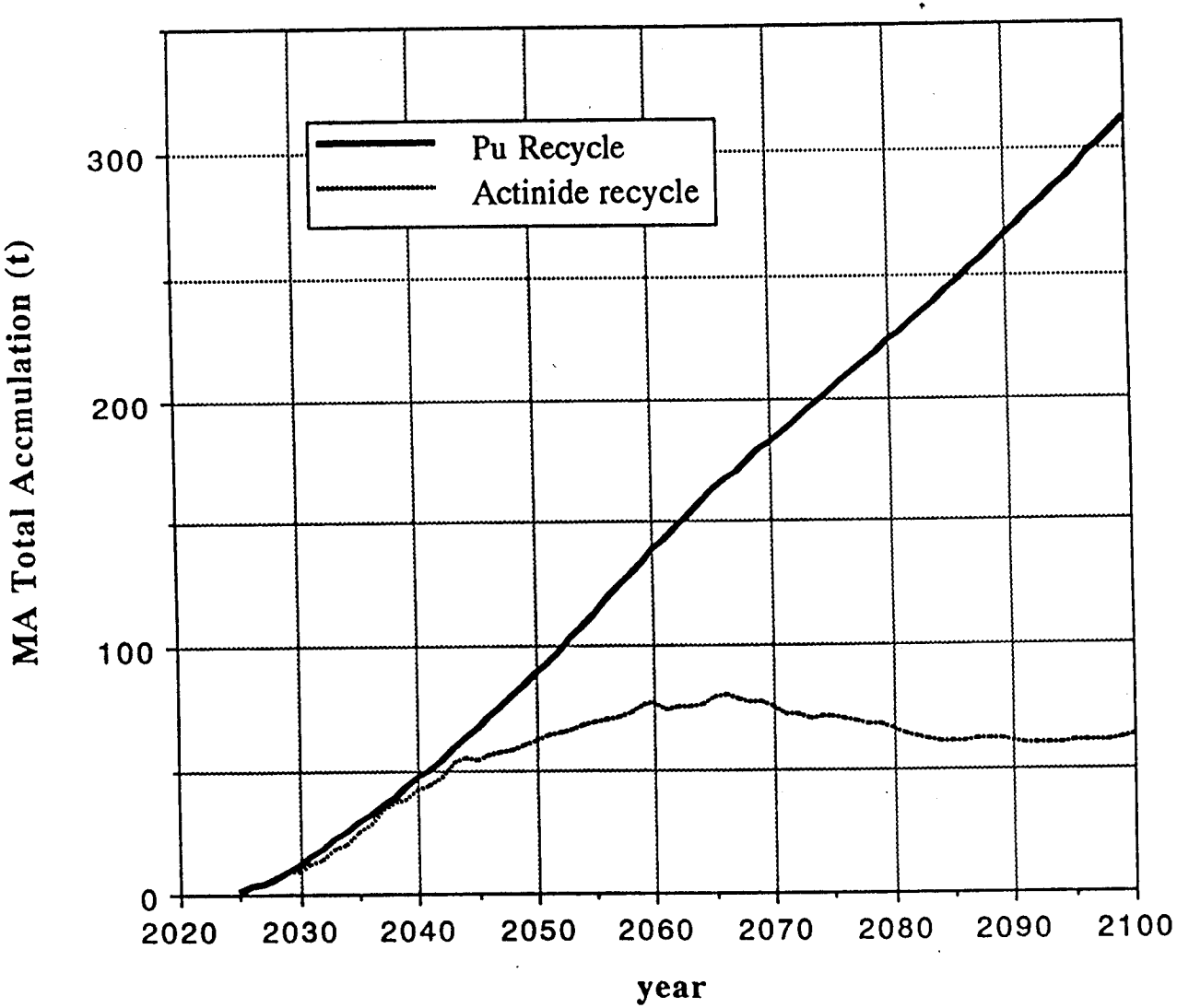


Fig.6 Effect of Minor Actinide Burning in the Advanced Fuel Recycle