

**SESSION 2: MAJOR PROGRAMMES AND  
INTERNATIONAL CO-OPERATION**

**CHAIRMAN: G.H. STEVENS (OECD/NEA)**



# Long-Term Program for Research, Development and Utilization of Nuclear Energy and Partitioning/Transmutation Technology in Japan

Takao Kuramochi, Koji Saeki  
Office of Nuclear Fuel Cycle Back-End Policy  
Atomic Energy Bureau, Science and Technology Agency, Japan

## I. Introduction

Atomic Energy Commission, which is responsible for the planning, deliberating and decision-making necessary for Japan's development and utilization of nuclear energy, revised the Long Term Program on 24 June 1994. The last Program was published in 1987, and AEC started its deliberation for the revision in 1992.

The circumstances among Atomic Energy has changed drastically since the publication of the 1987 Program, such as the collapse of cold war structure, global concern for environmental problems and the loss of public reliance on the safety of nuclear power plants after the Chernobyl accident. The new program was consolidated through careful and energetic study of AEC, taking public opinion into account.

The new Program attaches particular importance on treatment and disposal of radioactive wastes and decommissioning of nuclear energy facilities, which are defined as "back-end measures", from the stand point of accomplishing a consistent system of nuclear power generation. The basic research and development on Partitioning and Transmutation technology is to be promoted as a future technology in this area.

This report shows the outline of the new program and how the technology is described in the report briefly.

## 2. Long-Term Program for Research, Development and Utilization of Atomic Energy

### (1) Back Ground for the Revision

During 7 years from the publication of the 1987 Long-Term Program, the situation inside and outside Japan concerning the atomic energy development and utilization has changed dramatically, which necessitates careful deliberation for the revision of the Program. Followings are major international changes recognized by the AEC.

#### (a) Collapse of the cold war structure

Reduction and destruction of nuclear weapons is welcoming change, however, the treatment of the nuclear materials such as plutonium has become an important issue. Concern for proliferation of nuclear material is deepened and the safety of the nuclear plants in former East world also becomes major issue.

(b) Awareness of Global Environmental Problem

Global environmental problems such as global warming and acid rain are now of the international concern, and sustainable development consistent with environment becomes very important task.

(c) Increase in the Global Energy Consumption

Global energy demand is increasing especially among developing countries. It is important to secure the supply of energy for long-term.

(2) Major Role of Nuclear Energy

Energy is indispensable fundamental for economic and social activities as well as daily life. Presently more than 90% of the world's energy consumption depends on fossil fuels. Because of the limitation of fossil resources and environmental impact by the use of them, non-fossil fuels are expected to play a larger role in the years to come. New energy such as solar energy and wind power are seemed to be promising as dispersal energy. There are, however, many problems to be solved because of their characters, such as low energy density, being easily affected by changeable natural conditions.

Nuclear energy, on the other hand, has already overcome all basic problems in technical, economical and other terms and already occupies the position of a practical and stable source of energy. Nuclear energy can supply large amount of energy without emitting carbon dioxide, nitrogen oxide etc. Treatment and disposal of radioactive wastes is one of the important issue to be addressed for nuclear energy, but the amount of wastes is relatively small (an analysis shows about 85g/person-year in Japan) and the safe disposal of the wastes is technologically feasible.

International contribution is another important aspect of the development and utilization of nuclear energy. If the developed countries of economic power monopolized easy-to-use fossil energy, it would disturb the development of other countries. Nuclear energy enlarges options of energy supply and avoids friction related to energy resource allocation. Nuclear energy requires certain level of technology, therefore, it may not be feasible for every country. But if the advanced countries reduce the emission of carbon dioxide by introducing nuclear energy, it would be a remedy for the global environmental problem.

(3) Basic Principle for the Development and Utilization of Nuclear Energy

The new Program shows the basic principle of Japanese nuclear energy development and utilization:

(a) Development of Nuclear Energy Policy as a Nation Committed to Peaceful Use of Nuclear Energy

Japan today enjoys fruits of its past efforts in development and utilization of nuclear energy with consistent limitation to the peaceful purposes. It is vitally important for Japan to keep its commitment to peaceful use of nuclear energy in the future. That being the case, Japan intends to redouble its efforts with respect to gaining international confidence on nuclear non-proliferation regime, developing nuclear technology for peaceful use, acting internationally in a way that befits an advanced nation committing to peaceful use of nuclear energy and enhancing transparency and availability of information.

(b) Establishment of a Consistent System of Nuclear Power Generation by Light Water Reactors

Light water reactors, which today account for nearly 30% of Japan's total electric generation, have come to be considered a highly reliable type of reactor. Light water reactors are expected to continue to be the main-stream of nuclear power generation for a considerably long period in view of the basic trend of easier supply of natural uranium in recent years and the technical difficulties in developing commercial FBRs.

Therefore it is necessary to work for further improvement of their economic performance while ensuring their safety and reliability. Particularly important may be to enhance safety measures for aged nuclear power plants, and wastes disposal and decommissioning measures. The most important remaining task for the sake of ensuring a consistent system of nuclear power generation is the establishment of ways of accomplishing appropriate treatment and disposal of radioactive wastes and decommissioning of nuclear facilities. Our generation which enjoys the benefit of nuclear energy should be responsible for carrying out this task.

(c) Progress in Nuclear Fuel Recycling on the Basis of a Clear Future Outlook

As a country practically without its own energy resources, it is indispensable for Japan to secure its energy supply on the basis of the future outlook in order to maintain and develop its economic and social activities. Uranium resources are limited just as fossil fuel resources are, and it cannot be denied that uranium supply-demand relation will be tight by around the middle of the next century if light water reactors are continued to be used. Japan intends to secure its future energy supply by recycling of nuclear fuel involving reprocessing of spent fuel and reuse of recovered plutonium, uranium, etc. Therefore, Japan is steadily carrying forward research and development efforts aimed at future commercial nuclear fuel recycling. Another reason for doing so is that recycling of nuclear fuel is also meaningful in terms of sparing resources and the environment and of improving the management of radioactive waste.

It is necessary to recycle nuclear fuel based on the principle of not having plutonium in excess of the amount required to implement the program, as well as having very strict management of nuclear materials, implementing rational and consistent program and keeping its transparency so as not to give rise to any international doubts on Japan's program.

(d) Promoting Nuclear Sciences and Technologies in Various Ways and Strengthening Basic Research

Nuclear power generation by utilizing nuclear fission is a well-known form of nuclear technology, but not the only one. The range of application of nuclear technology is extremely broad. Nuclear fusion, heat supply by high temperature gas-cooled reactors, propulsion power for ships and utilization of radiation are also important area of nuclear technologies. Japan intends to continue to engage in development of various nuclear technologies.

From the understanding shown above, the new Program arranges the research and development themes into following four fields.

- i) Basic Research and Underlying Technology
- ii) Production of Energy
- iii) Radiation
- iv) Nuclear Fusion

### 3. Partitioning and Transmutation Technology in the New Program

#### (1) Back-End Policy and Partitioning and Transmutation Technology.

Back-End measures, which consist of treatment and disposal of radioactive waste and decommissioning of nuclear energy facilities are regarded as the most important task for the sake of ensuring a consistent system of nuclear power generation. As mentioned above, the new Program attaches particular importance to the back-end measures, especially to ensuring the smooth accomplishment of disposal of high-level radioactive waste.

In that context, the new Program stipulates procedure, schedule and responsibilities of entities concerned for the disposal enterprise. The entity to implement the disposal project will be established around the year 2000. The target for starting operation of a repository is in the 2030's or by mid-2040's at the latest.

The Partitioning and Transmutation technology, which would reduce the environmental impact of the disposal by utilizing useful nuclides in the high-level radioactive waste, is considered to be future technology in the new Program. The Japan Atomic Energy Research Institute (JAERI), the Power Reactor and Nuclear Fuel Development Corporation (PNC) and other organization such as Central Research Institute of Electric Power Industry (CRIEPI), are carrying out the basic research and development of these technologies. The check and review based on the progress of these activities will be carried out sometime in the second half of this decade.

#### (2) Partitioning and Transmutation as Nuclear Science and Technology

Partitioning and Transmutation technology will be promoted from two aspects. As basic research, the new Program urges the promotion of researches on TRU nuclides and on generation and utilization of various kind of beams. Beam technology is given a priority among underlying technologies.

Partitioning and Transmutation technology is also regarded as a technology for promoting effective production and utilization of energy.

### 4. OMEGA Project and Future Plan

As discussed above, the new Program supports the research and development of Nuclide Partitioning and Transmutation technology and R&D activities are being continued according to the Long-Term Program for Partitioning and Transmutation, which was published by the Advisory Committee for Radioactive Waste Management of AEC in 1988. The activities which follows this program is called OMEGA project (Options for Making Extra Gains of Actinides and Fission Products generated in Nuclear Fuel Cycle). The program selected the following areas for study and R&D are being carried out by three major organizations, namely JAERI, PNC and CRIEPI.

#### (a) Partitioning

- Partitioning of nuclide in HLW
- Recovery of metals from insoluble residue
- Utilization of recovered metal

(b) Transmutation

- Transmutation by using reactor
  - Reactor physics
  - FBR (PNC)
  - Actinide Burner Reactor (JAERI)
- Transmutation by using accelerator
  - Proton accelerator (JAERI)
  - Electron accelerator (PNC)

About six years has passed since the establishment of the program, and the check and review by STA/AEC is being planned according to the new Long-Term Program. One of the major consideration is the establishment of consistent program for OMEGA projects and actinide-recycle system which would recycle actinide elements (Np, Am, Cm etc) and is introduced to the new Long-term Program. Actinide-recycle system is aiming at improvement of resistance to nuclear proliferation as well as reduction of environmental impact.

## AN OVERVIEW OF THE SPIN PROGRAMME

M. VIALA, CEA, Direction du Cycle du Combustible  
M. SALVATORES, CEA, Direction des Réacteurs Nucléaires

### I. BACKGROUND

The SPIN programme fits into the important and increasing popular concern regarding high activity and long-lived nuclear waste management.

Subsequent to Mr. Christian Bataille's report, the National Assembly and the Senate discussed and adopted a law aimed at ensuring radioactive waste management in compliance with the protection of nature, the environment and health, taking the rights of future generations into consideration.

The French Law of December 30, 1991 explicitly requests that three subjects be studied over a period of 15 years:

- The search for alternatives enabling the separation and transmutation of long-lived radioactive elements present in these wastes.
- The study of reversible or irreversible storage possibilities in deep geological formations, and particularly thanks to the implementation of underground laboratories.
- The study of conditioning and long duration interim surface storage processes for these wastes.

Upon completion of these studies in 2006, Parliament will possibly have to come to a decision on the construction of an underground disposal site and the continuation of other lines of research.

#### *1.1. Radionuclides to be taken into consideration:*

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

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

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

Table 1 gives the value of the potential radiotoxicity of an irradiated fuel and the contribution of each long-lived radioisotope, on the basis of its parent product present today. These values are expressed in sieverts per  $\text{TWh}_e$  generated.



After the decay of highly radioactive fission products (Cs 137 - Sr 90) estimated at 300 years, the major contribution comes from plutonium, up to  $10^6$  years. Americium has a preponderant place among the minor actinides between  $10^2$  and  $10^5$  years. Neptunium becomes important after about  $10^5$  years. Curium makes a large contribution before  $10^4$  years. The contribution of fission products is negligible.

When the notion of confinement barrier is adopted, for deep disposal for example, the return of radionuclides to the biosphere must be assessed according to the degradation of the waste packages with time and to the transport of radionuclides through the geological media. This release of activity, weighted by the values of specific toxicity coefficients, leads to the notion of residual toxicity. The models are complex and site-dependent. A study performed in 1990 (PAGIS) for glass packages (containing no uranium, plutonium or iodine) in a granitic site (AURIAT) and for a standard scenario, showed that no dose was released before  $10^4$  years, that it then remained lower than the limit recommended by the ICRP and that the elements to be taken into consideration for storage safety were the most mobile ones (technetium, cesium) and the most radiotoxic (actinides). (See Figure 1). Incident scenarios are also to be considered.

### *1.2. Strategy Developed:*

A range of concern is developed below:

Concerning **potential radiotoxicity**, the high priority is to use or destroy plutonium, and then the minor actinides. The theoretical gains thus obtained may reach a factor of 500 for a standard fuel from a PWR 900 between the open cycle in which the irradiated fuel is sent to waste and that in which the plutonium and the minor actinides would be separated and destroyed with respective efficiencies of 99.9% for Pu and 99% for minor actinides.

But there is a wide gap between theoretical gains and real gains, separation and transmutation having limited yields and the latter being accompanied by the creation of radiotoxic elements (generally by neutron capture). Multi-recycling and global optimisation of the system is necessary to reach the best results concerning the use of resources and the limitation of the production of radiotoxic isotopes during multi-recycling.

Concerning **residual radiotoxicity**, the true to life model for deep disposal will be available only once the storage site is known and only tendencies can be seen which give, for example, iodine and cesium as the most soluble elements in reducing environments.

### *1.3. Advantage of Reprocessing*

The reprocessing of irradiated fuel, which recovers energy-bearing materials, already allows 99.5% of the plutonium to be separated (guaranteed value), soon to be 99.9% thanks to the efforts made in the PURETEX programme to improve the management of reprocessing wastes so as to reduce their volume and activity.

Reprocessing also allows to envisage strategies for separating other radionuclides in line with current techniques, either by modifying the process (Np, Tc, I, Zr) or by adding separation operations to the high activity stream (Am, Cm, Cs).

## II. GOALS AND ASSETS OF THE SPIN PROGRAMME

The goals of the SPIN programme are divided into two sub-programmes:

PURETEX, for the short and medium term, endeavours to reduce the volume and activity of reprocessing wastes from  $1.5 \text{ m}^3$  to  $0.5 \text{ m}^3$  per ton of reprocessed heavy metal ( $1.7 \text{ m}^3/\text{t}$  for direct disposal involved in the open cycle).

ACTINEX, for the long term, aims at assessing the various separation and transmutation strategies, their feasibility and their insertion in nuclear power generation.

II.1. PURETEX:

II.1.1. PURETEX - Solid Wastes

There are four types of waste not acceptable for surface-storage:

- Structural wastes from assemblies, hulls and end-pieces, embedded in grout.
- Coprecipitation sludge from effluent processing stations currently embedded in bitumen.
- Technological wastes embedded in concrete.
- Vitrified high activity effluents (F.P. and minor actinides).

The table below gives the volumes of these wastes specified in the La Hague plant process book, those produced in 1993 and the goals for 1995 and 2000 (see Figure 2).

**Table 2 - Volume (per ton of U in the fuel) of residues in each category:**

		Packaging Modes	UP3 rated values	1993 achieved values	1995 expected values	2000 expected values
Waste not compatible with present surface-storage requirements	Fission products	Glass blocks (C)	130 l/tU	115 l/tU	115 l/tU	115 l/tU
	Hulls and end-pieces	Concrete (B)	600 l/tU	600 l/tU	<600 l/tU	150 l/tU
	Sludges	Bitumen (B)	630 l/tU	450 l/tU	0	0
	Technological wastes	CAC concrete blocks (B)	1700 l/tU	200 l/tU	200 l/tU	<200 l/tU
TOTAL			3060 l/tU	1365 l/tU	<915 l/tU	<465 l/tU

The strategy adopted to obtain these performances consists in replacing the grouting of the hulls and end-pieces by a process still to be defined, in re-designing the whole management of liquid effluents to eliminate their processing by sludge and then bitumen-generating precipitation, in sorting the technological wastes, demoting them by decontamination whenever possible, or redefining methods for their processing.

Report on the Studies:

- The hull fusion process has been improved in the  $\alpha \beta \gamma$  laboratory, making it possible to lower the  $\alpha$  activity from  $3.5 \cdot 10^{11}$  Bq/t (9.4 Ci/t) to  $3.3 \cdot 10^{10}$  Bq/t (0.9 Ci/t) (about 10 times the surface standard).

A precise characterisation of the end-pieces may possible put demoting within reach.

Compacting of hulls and end-pieces is proposed by COGEMA, as a safe and reversible interim storage alternative.

- System studies have shown that the sodium ion is the dominant cation that prevents effluents from being processed by distillation and their activity from being sent back to vitrification, which would eliminate the precipitation of sludges and the production of bitume.

Replacing sodium carbonate by cetomalonic acid in the treatment of solvent has been demonstrated in the  $\alpha$  laboratory.

- The leaching process with silver II to decontaminate metallic waste highly contaminated by  $\text{PuO}_2$  has reached its industrial implementation phase.
- The evolution of vitrification towards a series of specific types of glass has begun: a sodium glass is under study as a complement to the R7/T7 glass.

#### II.1.2. PURETEX waste streams:

The PURETEX programme is also involved in limiting releases of activity into the sea.

Current release authorisations are 1700 TBq  $\beta\gamma$  (45 000 Ci  $\beta\gamma$ ) exclusive of tritium and 1.7 TBq  $\alpha$  (45 Ci  $\alpha$ ) for the La Hague reprocessing plants. The released values are much lower. Thus, releases were 0.15 TBq  $\alpha$  (4.1 Ci  $\alpha$ ) and 110 TBq  $\beta\gamma$  (3100 Ci  $\beta\gamma$ ) respectively in 1991, 0.1 TBq  $\alpha$  (2.9 Ci  $\alpha$ ) and 76 TBq  $\beta\gamma$  (2067 Ci  $\beta\gamma$ ) in 1992, 0.1 TBq  $\alpha$  (2.7 Ci  $\alpha$ ) and 73 TBq  $\beta\gamma$  (1980 Ci  $\beta\gamma$ ) in 1993, i.e. about 6% and 4% of the authorised values.

COGEMA and CEA have assigned R&D ambitious reduction goals aimed at releasing only about 1% of authorised values.

#### II.2. *ACTINEX:*

To evaluate the various separation and transmutation strategies, a hierarchy must be given to the concerns (and regularly updating them) and the technical feasibility of separation and transmutation must be assessed.

The hierarchisation is more advanced in the study dealing with potential toxicity. Actinides are involved (plutonium, neptunium, americium, curium) and, starting from  $10^5$  years, their radiotoxicity is of the same order of magnitude that the one of wastes containing uranium. Fission products are negligible.

Case studies are made based on available information: recycling of plutonium in PWR, in FBR, incineration of neptunium and of americium.

FBRs come closer to the theoretical scenario than PWRs. Furthermore, they accept the degraded isotopes of plutonium, may be adapted to a higher consumption of plutonium and are better suited for the incineration of minor actinides in the sense that they produce fewer higher isotopes, owing to a higher ratio of fission cross sections to capture cross sections for different isotopes and a better economy of neutrons in the core.

The CAPRA programme was launched to better evaluate these performances and to master the technology of an incinerator FBR.

Hierarchisation remains very difficult in the case of residual toxicity which brings out the fission products I, Cs, Tc, Zr, Pd.

The Interim Storage - Disposal and Storage Safety programmes must define the radionuclides that can return to the biosphere.

#### Technical feasibility of separation

The separation of plutonium is a reality and has been industrially mastered. The PURETEX programme will allow a maximum loss of 0.1% to be guaranteed.

The separation of neptunium may be mastered in the PUREX process itself by enhancing its extraction during the first cycle and its separation during the second uranium cycle. The accessible performance remains to be quantified (greater than 70%).

The separation of americium and of curium is much more difficult, especially if totally incinerable solvents are to be used. The DIAMEX process (under development) uses amides, extracts 99% of the americium and curium with the lanthanides. The SESAME process aims at selectively extracting the americium alone with an oxidized valency. Other fundamental studies are undertaken to separate these minor actinides.

Iodine is presently separated in the reprocessing process.

Technetium and zirconium are fission products with a behaviour already known in reprocessing. Some studies are performed on the solubility and separation of technetium which is greater than 70% for a standard PWR fuel. The main effort has concerned cesium for which a specific macrocycle extractant of the calixarene type has been defined at the fundamental research laboratory scale.

Palladium is part of the dissolution sludges. Its behaviour is unknown.

#### Technical feasibility of transmutation

The basic nuclear data are not all available and experiments are necessary, either for transmutation in a fission reactor (integral experiments to validate the European base, complementary data for technetium), or for hybrid system calculations (intermediate energy data and cascade codes).

Data are introduced into the calculation models used to predict the behaviour of radioelements in power reactors and the acceptable limits for these reactors without consequences on safety.

In a first analysis, the following indications may be given:

- the Increased Moderation Array PWRs have the best performance of all the PWRs in reducing the production of minor actinides and increasing the consumption of plutonium.
- FBRs give better performances than PWRs. They enable increased consumption of plutonium and incinerate the minor actinides with a lower production of higher isotopes.
- Various calculations have shown consumptions of minor actinides Am, Np from 7 to 8 kg/TWh<sub>e</sub>.
- The homogeneous mode (diluted in all the rods) is the most promising method for the recycling of neptunium, the heterogeneous mode (as targets) is preferable for the recycling of americium.

These calculations have to be backed up by irradiation experiments:

The result of neptunium transmutation calculations with limited burnup has been validated in a Superfact 1 (Phenix).

Superfact 2 (Phenix) is planned for the transmutation of neptunium and americium with a higher burnup.

A demonstration will be made in Superphenix.

ACTINEAU is an experiment programmed in the OPERA (PWR) loop to constitute a database on the transmutation of neptunium.

In the case of fission products, the calculations and experiments are not as advanced. Technetium may be transmuted into stable ruthenium in a fast reactor (technetium loses 50% of its mass in 3 years in radial blanket and thermalised flux).

The accelerator-sub-critical medium hybrid systems show that these systems offer no supplementary potential for the transmutation of minor actinides. Their use for the transmutation of fission products requires additional studies.

### III. FIVE-YEAR PLAN

The SPIN programme is planned over fifteen years with a more detailed definition over a five year rolling plan.

In financial terms (MF):

	1994	1995	1996	1997	1998
SPIN	250	300	300	300	290
including (Investments)	30	64	70	55	40

#### PURETEX

#### Years of R&D

- Hull fusion 92-98
- Hull compacting 93-97
- Elimination of the bitumen 92-96
- Sodium glass 94-96
- Decontamination of the wastes 92-97
- Reduction of releases 95-?

## ACTINEX

- Separation of the actinides 92-98
- Separation of the fission products 94-99
- Basic data for transmutation 93-97
- Transmutation studies in fission reactor 93-97
- Fuel studies and transmutation experiments 93-98
- Hybrid transmutation system studies 93-97



# Table 1 - POTENTIAL RADIOTOXICITY OF FUEL

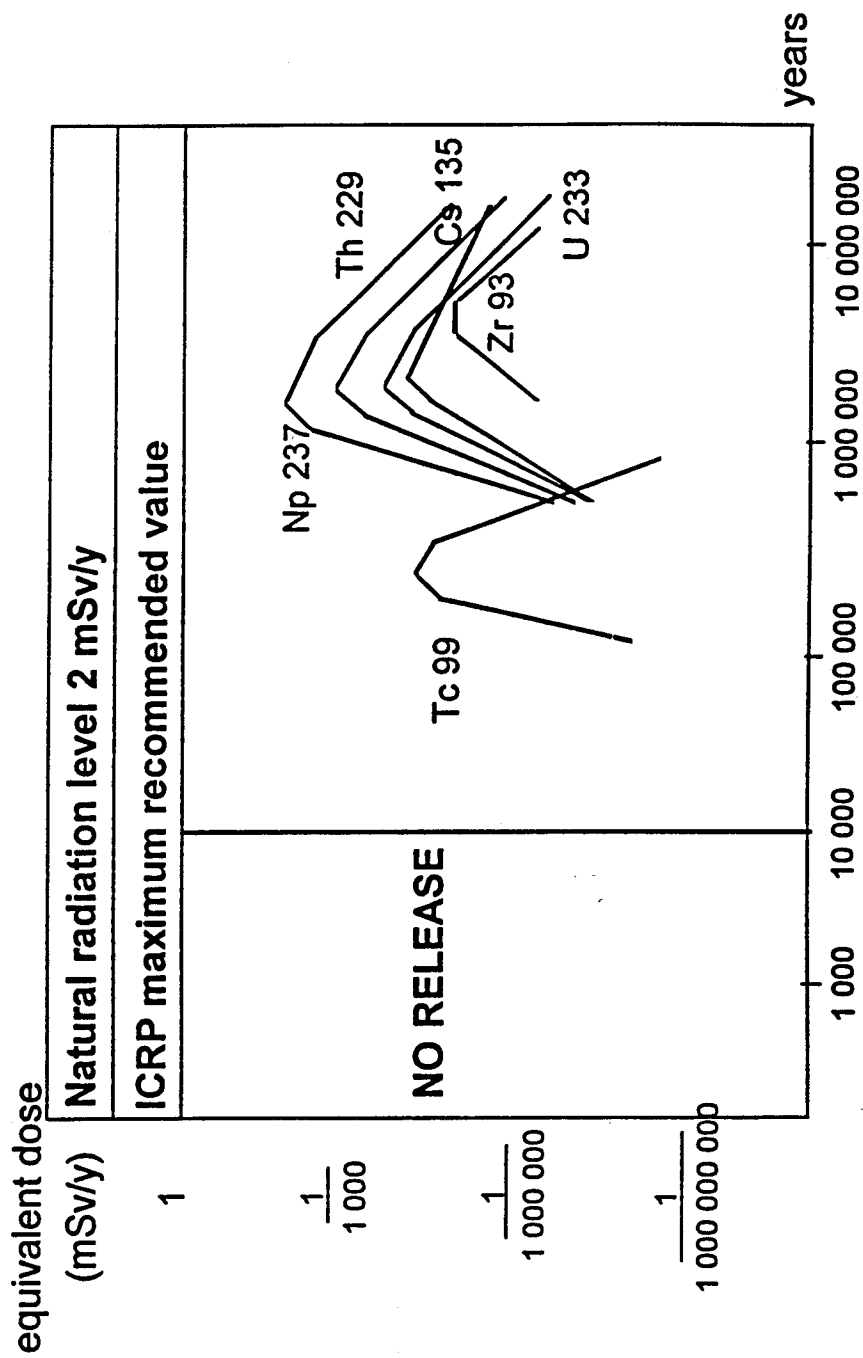
SOURCE TERM AND ITS COMPONENTS  
EVOLUTION VERSUS TIME

	10 <sup>3</sup> years	10 <sup>4</sup> years	10 <sup>5</sup> years
TOTAL (Sv/TWhe)	3.1 10 <sup>8</sup>	7,7 10 <sup>7</sup>	4.2 10 <sup>6</sup>
COMPONENTS %			
Pu	90	97	88
Np	/	/	1,3
Am	9.2	2.5	2.7
Cm	0.3	0.4	/
P.F.	6.0 10 <sup>-4</sup>	2.4 10 <sup>-3</sup>	3.2 10 <sup>-2</sup>



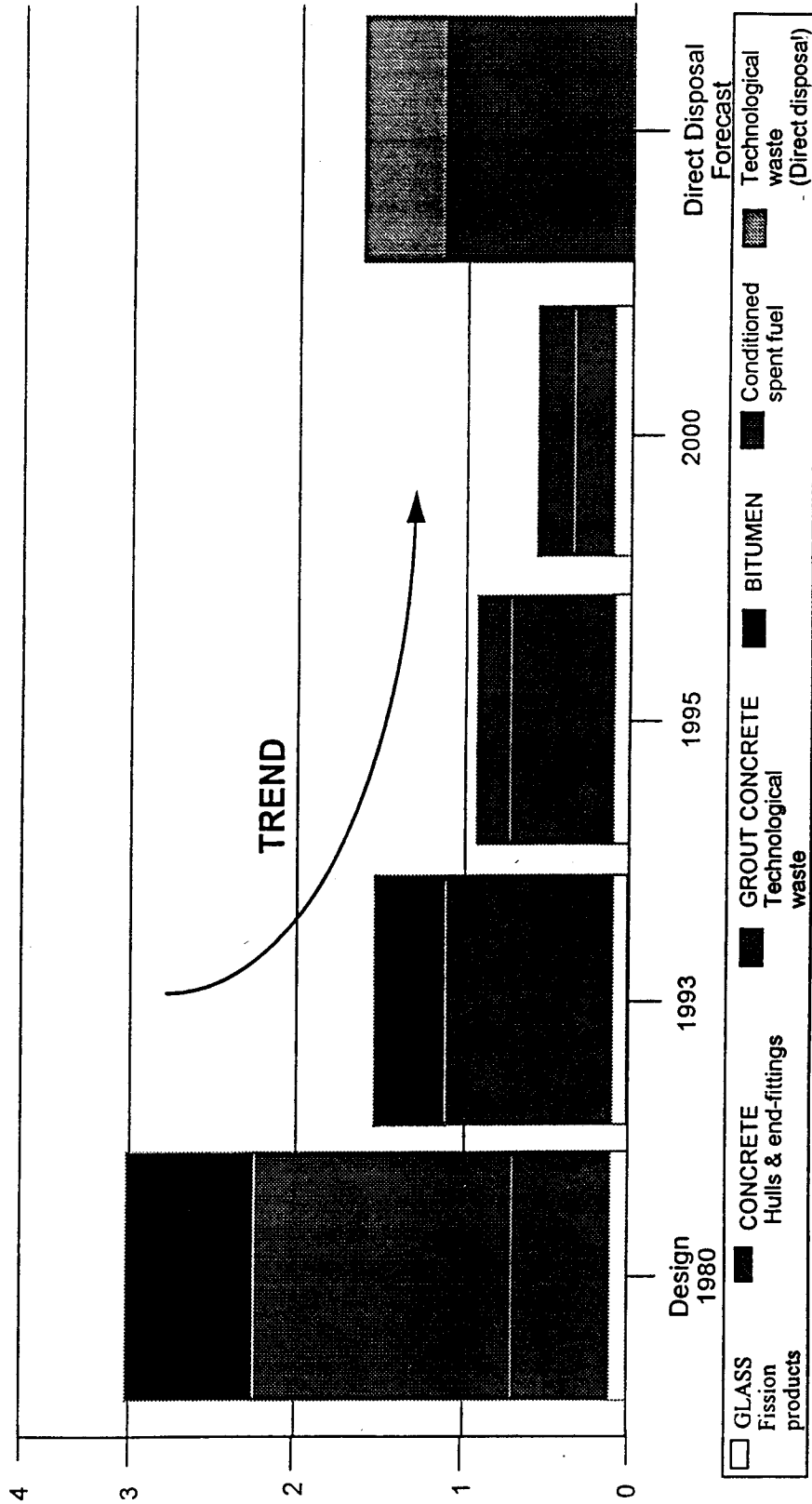
# RESIDUAL RADIO TOXICITY

Fig.1 AURIAT site (concept A) standard scenario





# CEA Volumes of Final Residues Generated in UP3 (m<sup>3</sup>/tU) (Long-lived waste after conditioning)



# **CURRENT STATUS OF THE SCIENTIFIC ACTIVITY IN RUSSIA ON HLW TRANSMUTATION AND USE OF WEAPON-GRADE AND POWER PLUTONIUM IN SUBCRITICAL SYSTEMS DRIVEN BY PROTON ACCELERATOR**

G.V. Kiselev, Head of Reactor Department  
Institute of Theoretical & Experimental Physics, Moscow, Russia

Features of neutron fuel cycles with ENF as well as FP's and actinides transmutation in ENF are analyzed in the paper: fuel type, fertile materials, neutron consumption, second radioactivity, change in FP's and radiotoxicity of actinides. Use of the weapon-grade and power plutonium in the ENF is considered too. Information on different design version of the ENF blanket including study of the sectional blanket with neutron valves, its performance, problems of the ENF design, R&D program including neutron source driven by 56 MeV "Istra" proton linac is given. Proposals on joint scientific cooperation are discussed.

## **1. MAIN IDEAS**

1. Decrease in the amount of the long-lived radioactive waste (LLRW) produced by the atomic power and defence industries with the help of a transmutation process.
2. The use of electro-nuclear facilities (ENFs) in combination with specialized fast reactors for LLRW nuclear transmutation.
3. The use of ENFs in the promising atomic power industry to close the nuclear fuel cycle (NFC) as to fissionable materials and fission products with concurrent power generation.
4. The application of new, more safe technologies in the ENFs implementation compared to NPPs reactors, particularly in the case when weapon-grade plutonium is used.
5. Possible support of the LLRW incineration (transmutation) by public and population.
6. A need for joining forces of and co-operation between different countries to implement the LLRW transmutation processes.

## **2. SHORT INTRODUCTION**

There are two important problems in modern atomic power industry: NPPs' safety and high level radioactive waste (HLW). In most countries including Russia the main strategy of the HLW management is HLW storage in solidified form in surface burial with further geological disposal. The data about HLW amount in Russia could be cited as an example (see Table 1) [1].

It can be seen from this Table that the main contribution constitute HLW produced in spent nuclear fuel (SNF) reprocessing plant. At the same time HLW produced by NPPs are several orders of magnitude less than those from a SNF reprocessing plant. On the basis of data from Table 1 an unexpected conclusion could be made that nations possessing NPPs and not performing SNF reprocessing or delaying it could not seemingly to hurry up with development of alternative HLW management technologies. At the same time as reserves of the natural uranium become more depleted

the SNF reprocessing will be necessary. Therefore all nuclear nations should be interested in developing alternative HLW management technologies even now since development of new technologies is time consuming. At the same time in those nations where SNF reprocessing is performed development of alternative technologies is necessary since the HLW amount will constantly rise with time. In this case the social aspect has to be taken into account implying that part of the population is against the construction of storage facilities on the territory where they live.

In the last few years attention of experts from various nations is attracted by a transformation (transmutation) of the long-lived HLW in stable nuclides in a neutron flux. A large number of papers and reports devoted to transmutation and presented at international conferences demonstrates that.

146 papers have been presented at ICENES-93 conference in Japan including 33 dealing with HLW transmutation (12 papers on transmutation in fast reactors, 20 papers on transmutation in electro-nuclear facilities). At GLOBAL-93 International Conference held in Seattle, USA on September 12-17, 1993 there were:

- 19 papers on transmutation in fast reactors;
- 13 papers on transmutation in thermal reactors;
- 26 papers on transmutation in electro-nuclear facilities;
- 2 papers on transmutation in fusion facilities;
- 12 papers on nuclear data for transmutation.

A large number of interesting papers on transmutation in proton accelerator driven subcritical systems have been presented at International Seminar organized by ITEP in Moscow in May 1994 and at International Conference in Las Vegas, USA in July 1994 organized by LANL.

For purposes of clarity of further presentation let us explain the term of "electro-nuclear facility" which is used in present paper. The ENF consists of a subcritical core (blanket) with external neutron source in the form of neutron generating target and proton accelerator. ITEP and its Director, Professor I.V. Chuvilo is a scientific supervisor in R&D on ENFs in Russia. ITEP combines forces of 7 Russian research institutes and design organizations: Moscow Radio Technical Institute, Institute for Inorganic Materials (Moscow), Radium Institute (S. Petersburg), Institute of Experimental Physics (Arzamas-16), Institute of Technical Physics (Chelyabinsk-70), Power Physical Institute (Obninsk), Design Bureau for Machine Building (N. Novgorod), Design Institute for Power Technology (S. Petersburg) with the total number of people taking part in this activity accounting for appr. 300.

Besides, work on different ENF versions is being conducted in Moscow Physical Engineering Institute, Research and Design Institute for Power Technology (Moscow), Atomic Power Institute (Obninsk), High Energy Physics Institute (Protvino, Moscow Region), and Joint Institute for Nuclear Research (Dubna).

ENF R&D program is supported by the Russian Ministry of Atomic Power.

International Science and Technology Center (ISTC) in Moscow has made a decision to fund over a period of 2 years conceptual investigations on HLW transmutation and weapon-grade plutonium conversion in ENF conducted by Russian institutes. Unfortunately, the means allocated for this account for 30% of necessary amount.

Because information concerning the status of work on ENFs in Russia is presented at International Information Exchange Meetings for the first time, the presentation of general concept approaches to ENFs development is given in this paper along with particular results and proposals.

### **3. ITEP APPROACHES TO ENF DEVELOPMENT**

ENF represents a new type of nuclear power facilities which in ITEP opinion should be introduced in atomic power industry of the 21st century. While conducting conceptual investigations and developments on ENF the ITEP experts are guided by the following criteria:

1. It is expedient to use the positive experience available and technical approaches verified in the atomic power industry, properties of inherent self-protection in particular, passive safety systems, "in depth protection" principle, etc. The main criterium in this case is the rise in the ENF nuclear and technical safety compared with NPP.
2. Experimental investigations should be conducted to substantiate new technical approaches proposed, first of all ENF blanket as the most complex and important unit for the purpose of safety.
3. Experimental work should be done on nuclear data base primarily for radionuclides to be transmuted.
4. At the stage of conceptual investigations it is expedient to study various versions of targets and blankets with estimates of their possible implementation, determination of technical and economical characteristics and choice of one version for further development.
5. The ENF operation should be performed with positive power balance and decrease in radiotoxicity compared with initial level.

### **4. ENF POSSIBLE OPERATION MODES AND FUEL CYCLES**

The availability of the external neutron source makes its possible to consider different modes of the ENF operation and fuel cycles correspondingly. The following ENF operation modes could be noted:

1. A transmutation mode without power utilization.
2. A transmutation mode with concurrent power generation and utilization.
3. A mode with power generation and utilization as well as new fissionable materials production and HLW transmutation.
4. A mode of power generation and new fissionable materials production.
5. An after-burn mode for power generation with use of the NPP spent fuel assemblies as nuclear fuel.

The ENF can also serve as high intensity neutron source to produce radionuclides with high specific activity and for research purposes.

The ENF can have various fuel cycles depending upon operation mode. The following materials could be used as fuel materials for transmutation modes: enriched uranium, power and weapon-grade plutonium, actinides (in mixture with plutonium and without it), and  $^{233}\text{U}$ . The depleted uranium and thorium could be used as fertile materials for production of the new fuel.

The possibility to implement in the ENF the following fuel cycles (FCs) could be indicated:

1. The uranium FC. In this FC version various uranium fuel could be used in blanket: 90% enriched uranium released as the result of nuclear disarmament, regenerated uranium produced after multiple irradiation and reprocessing cycles of the SNF with high content of  $^{232}\text{U}$  and  $^{236}\text{U}$ . Actinides and fission products (FPs) could be loaded into the blanket in various combinations and proportions for transmutation purposes. The depleted uranium without any restriction in  $^{235}\text{U}$  content should be loaded into the blanket in plutonium production mode.  $^{233}\text{U}$  could be used as a nuclear fuel in the ENF blanket too.
2. The plutonium FC. The plutonium fuel with different isotope composition: weapon-grade plutonium only, or power plutonium only, or their mixture could be loaded into the blanket in this version. Actinides and FPs could be loaded into the ENF blanket for transmutation purposes. The depleted uranium could be used for the purpose of nuclear fuel reproduction in the ENF blanket. Neptunium or  $^{238}\text{U}$  included into plutonium fuel could be used for special purposes of the weapon-grade plutonium de naturization.
3. The uranium-thorium and plutonium-thorium FCs. The enriched uranium or plutonium is used as a nuclear fuel in these FC versions (thorium is used as fertile material for  $^{233}\text{U}$  product ion purposes).
4. The actinide FC. In this version alternative actinides in various combinations with FPs are used as a nuclear fuel of the ENF blanket for transmutation.

In what form the fuel, fertile, and target materials are expected to be used has been of interest. Two main alternatives of fissionable fuel materials application is possible: in a solid and a liquid form. Each alternative has its own advantages and disadvantages. If the available experience in production of the nuclear fuel for NPPs is kept in mind, it is advisable to chose dioxide fuel in zirconium cladding. It will be a MOX-fuel in the case of power or weapon-grade plutonium. In recent years Russian experts are conducting investigations on cermet and nitride fuel with increased burnup level. In the last few years some research groups in LANL, JAERI, ITEP are being conducted investigations on liquid fuel based on fluoride molten salts of Li-BeF<sub>2</sub>-ThF<sub>4</sub>-PuF<sub>4</sub> type.

However, in the case of 3-5% actinides addition into the MOX-fuel its radiation resistance should be experimentally verified which requires substantial expenses and is time consuming.

The advantage of liquid fuel is the absence of radiation resistance problem, the possibility to break with metallurgical production of fuel assemblies, reduction in amount of FPs and fissionable materials in the core, etc. The main objection in Russia against liquid fuel lies in the absence of technological basis for its implementation in atomic power industry, rather complex technical problems emerging in reactor facility, etc. It could be argued that the problem of liquid fuel application advances the needs of the atomic power industry at the present time. However, R&D should be conducted on prospects and possibilities of the liquid fuel application in ENFs to have a basis for decision making.

The variety of the above mentioned operation modes and fuel cycles in the ENF requires certain priorities to be determined to concentrate small resources available in one or two R&D lines. For this purpose, systematic investigations have to be pursued which are not performed yet because of the lack of funding. But priorities in the fuel cycles development could be determined even now. In author's

opinion the highest priority has the ENF plutonium cycle where plutonium is used as nuclear fuel to produce power and excessive neutrons which in its turn are directed toward transmutation of actinides and FPs.

It is explained by the availability of large stocks of power and weapon-grade plutonium with high power potential. In accordance with the decision of the Scientific and Technical Council of the RF Ministry of the Atomic Power Industry fast reactors are considered as the main nuclear and power facilities for utilization of the Russian weapon-grade plutonium. At the same time RF Ministry of the Atomic Power Industry supports the ENF development as an alternative line keeping in mind the possibility to operate the ENF blanket in a subcritical mode which increase the nuclear safety level in the event of its loading with weapon-grade plutonium. Hence the transmutation mode with power generation and use of the weapon-grade and power plutonium wherein plutonium FC with actinides and FPs as target materials is implemented has a priority in conducting promising R&D.

The second priority has a plutonium FC wherein the ENF operates in the mode of power generation and plutonium production of depleted uranium. The priority of this mode and FC is explained by the availability of large amounts of depleted uranium at storages of the nuclear nations. The involvement of depleted uranium makes it possible to increase the nuclear fuel resources substantially as resources of cheap natural uranium will be exhausted. All necessary technologies are available for this variant which are verified in industrial scale.

The third priority in author's opinion has a plutonium FC wherein the ENF operates in the mode of power generation and  $^{233}\text{U}$  production of thorium. However, the involvement of  $^{233}\text{U}$  into the FC implies the development of new technologies which will be necessary after all resources of natural uranium will be exhausted.

## 5. TRANSMUTATION PROCESS PECULIARITIES

The process of the FPs and actinides transmutation in neutron fluxes has its own peculiarities which has been taken into account in the ENF development. The FPs transformation is performed mainly by  $(n, \gamma)$ -reactions through sequential capture of neutrons being transmuted by a nuclide and formed by an intermediate nuclide. The destruction of actinides occurs mainly through fission reaction. Despite this difference the following HLW transmutation processes are characterized by the following parameters:

1.  $A_i$  - the actinides destruction rate which is determined by neutron flux density  $\phi$  and  $\sigma_i$ , interaction effective cross-section.
2. The process power efficiency which is determined by the value of spent power for the destruction of one nucleus of the nuclide being transmuted or by a number of spent neutrons for the destruction of one nucleus.
3. The secondary radioactivity which is formed as a result of the HLW transmutation.
4. The value of the nuclides radiotoxicity before and after the transmutation.

Investigation of the mentioned above features allows to determine the list of nuclides which are advisable for transmutation. The short analysis of the mentioned above features is given below.

## 1. $A_i$ - destruction rate of nuclides being transmuted

Since the value of  $A_i$  is proportional to  $\phi \cdot \sigma_i$ ,  $A_i$  for FPs will be the higher, the higher will be  $\phi$  and  $\sigma_i$  values. The upper limit of  $\phi$  which can be reached with present-day knowledge and experience is probably in the range of  $5 \cdot 10^{15} - 10^{16} \text{ cm}^{-2} \cdot \text{S}^{-1}$ . The  $\sigma_i$  value depends on neutron spectrum. It is advisable to use thermal neutron spectrum for FPs transmutation through  $(n, \gamma)$ -reaction where cross-section follows the law of  $1/V$  ( $V$  is the velocity of neutrons). Comparison of  $\sigma_i$ , effective cross-sections of actinides for fast reactors and ENF blanket thermal spectrum performed by ITEP scientists is tabulated in Tables 2 and 3 borrowed from [2]. It is seen from results given in Tables 2 and 3 that  $\sigma_i$ , effective cross-section of actinides in thermal spectrum is in most cases much higher than the same values for fast reactor.

ITEP specialists have estimated the value of the neutron flux density necessary for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  [3] incineration. For  $^{90}\text{Sr}$  the radiation capture cross-section accounts for  $\sigma = 0.014$  barns according to the latest data (Einwei, 1983) or 0.0097 (Lone, 1993). According to older data it was believed that  $\sigma = 0.9 + 0.5$  (Zets, 1966). If we are adhered to contemporary data and believe that  $\sigma = 0.01$  barn, then neutron flux of  $7.6 \cdot 10^{16} \text{ cm}^{-2} \cdot \text{S}^{-1}$  is required for  $^{90}\text{Sr}$  transmutation rate to be equal to its radioactive decay rate ( $T_{1/2} = 28.8$  years). In this case the "half-destruction period" will be twice less than the half life, i. e. 15 years. For more rapid destruction higher flux densities (more than  $10^{17} \text{ cm}^{-2} \cdot \text{S}^{-1}$ ) would be required. Even though such flux densities could be achieved in ENF, it is unlikely that this will be economically justified.

For  $^{137}\text{Cs}$   $\sigma = 0.11$  barn (Stupetta, 1960) or 0.25 barn (Harada, 1990). For transmutation rate to be equal to the radioactive decay rate ( $T_{1/2} = 30$  years) the flux density of  $6.7 \cdot 10^{15}$  or  $2.9 \cdot 10^{15} \text{ cm}^{-2} \cdot \text{S}^{-1}$  is accordingly necessary which is achievable in modern reactors. In this case the "half-destruction period" will be 15 years. Flux densities higher than  $10^{16} \text{ cm}^{-2} \cdot \text{S}^{-1}$  are required for more rapid destruction. It should be kept in mind that radiotoxicity of  $^{90}\text{Sr}$  is significantly (37 times) higher than that of  $^{137}\text{Cs}$  (the maximum permissible concentration of  $^{90}\text{Sr}$  in water is  $4.0 \cdot 10^{-16} \text{ Ci/l}$ , and that of  $^{137}\text{Cs}$  is  $1.5 \cdot 10^{-8} \text{ Ci/l}$ ). Therefore destruction of  $^{137}\text{Cs}$  only without  $^{90}\text{Sr}$  reduces initial radiotoxicity of  $^{90}\text{Sr} + ^{137}\text{Cs}$  no more than 4% according to existing data on their interaction with neutrons.

## 2. *Transmutation process power efficiency*

It is quite obvious that power consumption for HLW destruction should be substantially less than amount of power generated by NPPs. According to investigations carried out by specialists from Moscow Physics Engineering Institute the FPs transmutation process is effective if power consumed for transmutation is appr. 30% of that generated in the NPP [4]. It is obvious that power balance in actinides destruction is positive. The power consumption for HLW transmutation could be determined after development of particular ENF design only. Therefore such characteristics as  $R_i$ , consumption of neutron necessary to destruct one FP's nuclei could be used for estimation of the power consumption. The  $R_i$  calculation results are tabulated in Table 4 [5]. It is evident from Table 4 that considerable amount of neutrons is required for transmutation of  $^{93}\text{Zr}$  and  $^{151}\text{Sm}$ . This demonstrates that their destruction in a neutron flux is ineffective from the power viewpoint. At the same time it is advisable to state a problem of their recycling in the FC. The transmutation of other radionuclides listed in the Table 4 is energetically advisable.

### 3. *Secondary radioactivity under transmutation*

The process of neutron generation for transmutation is accompanied by electric power consumption from external source for accelerator supply. If we believe that this electric power is generated in the NPP, then neutrons are produced as the result of fission reactions of the nuclear fuel in the reactor of the NPP which energize the accelerator. Moreover, nuclear reactions generating neutrons occur also in the blanket of the ENF itself. Hence the transmutation process, i.e. the process of one radioactivity unit destruction is accompanied by generation of FPs, that is secondary radioactivity. Because in the first 100 years after unloading of the NPP's spent nuclear fuel (SNF) its activity will be determined to appr. 90% by activity of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  the secondary activity is mainly the activity of the medium-lived nuclides -  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . The calculations of the secondary activity performed by ITEP specialists show that with elimination of all primary activity of the FPs (except for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ ) the secondary radioactivity is generated which accounts for 10-12% of the  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  primary radioactivity [5]. In the event of elimination of 100% of the primary activity including  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  the secondary activity is generated equal to 40% of the  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  primary activity. This result can be interpreted in the following way. First, the long-lived activity is transformed to a medium-lived with the help of an ENF. Second, we have to pay with generation of additional 10-12% of the secondary activity of the medium-lived  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  radionuclides ( $T_{1/2}$  up to 30 years) for elimination of the total primary activity of the long-lived FPs. This is a very serious result. It follows from this result that with the use of an ENF all long-lived FPs could be destructed. The cost of this is not too high. As for destruction of the medium-lived FPs,  $^{137}\text{Cs}$  in particular, the ENF permits to decrease its amount 2-2.5 times compared with its initial content.

### 4. *Radiotoxicity*

The main radiobiological hazard represent actinides with high half lives. The relative indexes of toxicity,  $TI_i$  for long-lived actinides depending upon their storage time are tabulated in Table 5.  $TI_i$  is the amount of water required for dilution of the  $i$  nuclide up to a maximum permissible concentration. It is evident from Table 5 that the most radiation hazard in first 100 years represent  $^{232}\text{U}$ ,  $^{244}\text{Cm}$ ,  $^{238}\text{Pu}$ ,  $^{241}\text{Am}$  and  $^{241}\text{Pu}$  ( $TI$  for  $^{239}\text{Pu}$  is assumed to be equal to 1). After 1000 years some redistribution occurs:  $^{241}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{240}\text{Pu}$ , and  $^{244}\text{Cm}$  come in the first place from radiation hazard point of view.

Taking into consideration high radiation hazard of actinides a comparison of the radiotoxicity has been performed for fuel loads of the thermal blanket, ENF, and fast blanket [2]. For comparison to be a representative one a stationary operation mode has been chosen such that the change in concentration of nuclides in the next cycle is the same as for the present one. The effective neutron flux density for fast reactor was assumed to be equal to  $1.2 \cdot 10^{16} \text{ cm}^{-2} \cdot \text{s}^{-1}$ , the effective cross-sections are listed in Table 2.

The cross-sections are listed in Table 3 for a thermal blanket, and effective, average over Maxwellian neutron spectrum neutron flux density was assumed to be equal to  $5 \cdot 10^{15} \text{ cm}^{-2} \cdot \text{s}^{-1}$ . The results of  $TI$  values calculations are tabulated in Tables 6, 7, 8. The following conclusions could be made from these Tables:

1. The equilibrium mass of all actinides in the fast reactor stationary mode is 43 times higher than in thermal blanket.



2 The main contributions into actinides mass are made by:

$^{237}\text{Np}$  (18%),  $^{238}\text{Pu}$  (26%),  $^{241}\text{Am}$  (15%),  $^{242}\text{Cm}$  (4%),  $^{243}\text{Cm}$  (9%) in fast reactor;  
 $^{237}\text{Np}$  (15%),  $^{238}\text{Pu}$  (2.9%),  $^{241}\text{Am}$  (4.9%),  $^{242}\text{Cm}$  (37%),  $^{244}\text{Cm}$  (16%),  $^{246}\text{Cm}$  (13%) in thermal blanket.

3. The long-lived activity of actinides in fast reactor is 43 times higher than in thermal blanket. The long-lived activity in fast reactor is governed for 34% by  $^{238}\text{Pu}$  and for 54% by  $^{244}\text{Cm}$ , and for 90% by  $^{244}\text{Cm}$  in thermal blanket.

4.  $\text{TI}_i$ , toxicity indexes for long-lived actinides in fast reactor is 140 times higher than in the ENF thermal blanket. The  $\text{TI}_i$ , the toxicity index corresponding to a stationary amount of long-lived actinides in fast reactor is 33 times higher than the toxicity index of actinides in its own annual replenishment. For thermal blanket the  $\text{TI}_{\text{AC}}$  is 4.3 times less than  $\text{TI}_{\text{AC}}$  in the annual replenishment. The results obtained demonstrate the ecological hazard of actinides transmutation in fast reactors and advantages of the ENF thermal blanket. Hence the reduction of actinides toxicity for several orders of magnitude occurs as the result of actinides transmutation in the ENF.

The mentioned analytical studies made it possible to chose FPs and actinides for transmutation. With consideration for the above mentioned characteristics it is advisable to transmute the following FPs and actinides:  $^{79}\text{Se}$ ,  $^{99}\text{Tc}$ ,  $^{107}\text{Pd}$ ,  $^{126}\text{Sn}$ ,  $^{129}\text{I}$ ; Np, Am, Cm,  $^{232}\text{U}$  using plutonium for generation of excessive neutrons.

## 6. THE USE OF THE WEAPON-GRADE AND POWER PLUTONIUM IN ENF

The investigations carried out in ITEP and other institutions demonstrates that existent power reactors and reactors under development despite technical measures taken are not guaranteed from accidents of such class as accidents dealing with reactivity (accidents with a change in the core reactivity). The probability of the accident associated with reactivity increases when plutonium fuel is used in thermal reactors because the decline in safety parameters takes place. According to calculations performed by specialists from Physical Power Institute (Obninsk) for example the temperature and power coefficients of reactivity are increased in the WWER-500 reactor when MOX-fuel is used (4.8% and 2.7% instead of 3.6% and 2.1% respectively), the  $\beta_{\text{eff}}$  value is decreased (from  $6.0 \cdot 10^{-3}$  to  $4.4 \cdot 10^{-3}$  with a condition hold that efficiency of the operating groups of the CPS rods does not exceeds  $\beta_{\text{eff}}$ ), the efficiency of the CPS rods is decreased by a factor of 1.3 [6].

The decline in nuclear safety level of fast reactors with Na coolant and plutonium fuel should be particularly emphasized. First, there is rather high error in determination of the breeding ratio and critical mass of fast reactors reaching 7-10% and 5% respectively. This situation leads to inaccuracy in the choice of the core volume (average power intensity) and reactivity margin between overloads. Second, the reactivity void coefficient for sodium (RVCS) could be positive. For instance, RVCS of BN-800 reactor was previously positive. To change the RVCS sign from positive to negative the BN-800 reactor designers have excluded the upper and side shields. This method of solving the problem resulted in a decrease in the breeding ratio to 1 instead of 1.23.

The same disadvantage takes also place for Na fast reactors in the case of weapon-grade plutonium application.

The features of using weapon-grade and power plutonium in nuclear reactors are the following:

- part of delayed neutrons ( $\beta_{\text{eff}}$ ) for Pu which is equal to  $2.7 \cdot 10^{-3}$  is smaller than for U ( $6.82 \cdot 10^{-3}$ ) and for WWER-1000 spent fuel with a burn-up of 40 kg/t ( $4.37 \cdot 10^{-3}$ );
- very high radiotoxicity of plutonium requires remote control technology of fuel management;
- small critical mass depending from plutonium isotope composition
- problem of the non-proliferation.

Addition of minor actinides to plutonium fuel for fast reactor with Na coolant leads to:

- decrease in  $\beta_{\text{eff}}$ , effective part of delayed neutrons;
- decrease in  $l_n$  average lifetime of prompt neutrons;
- positive reactivity void coefficient for Na.

The result is the decrease in safety level for a fast breeder.

These features of plutonium, especially of the weapon-grade one require special approach to the use of plutonium as nuclear fuel for atomic power industry.

In ITEP specialists' opinion the ENF operation with subcritical blanket permits to exclude accidents associated with reactivity and first of all such accident as reactor runaway on prompt neutrons. Therefore the ENF safety level is substantially higher than that of power reactors with a critical core. This fundamental advantage of the ENF gives grounds for use in ENF weapon-grade and power plutonium as nuclear fuel for power generation and excessive neutrons production.

ITEP experts have made analytical study of various alternatives of the ENF plutonium mode with power generation:

- weapon-grade plutonium burnout (Table 9);
- $^{237}\text{Np}$  transmutation (Table 10)
- $^{233}\text{U}$  production and transmutation of  $^{99}\text{Tc}$  and  $^{129}\text{I}$  (Table 11).

The results of these investigations have been presented at various international conferences, in particular at GLOBAL-93 conference in Seattle (USA) and ICENES-93 conference in Japan. It seems likely that they are well known to international scientific community. We would like to call attention to the possibility of further increase in nuclear safety level through the ENF stationary operation mode (concerning burnout). This mode could be implemented through regular continuous replenishment by newly made fuel. The salient feature of this mode is the possibility to maintain the reactivity margin virtually at the unchanged level. And the fundamental matter is that burnout of  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  fissionable in thermal spectrum and increase in  $^{240}\text{Pu}$  and  $^{242}\text{Pu}$  portions occurs which results in impossibility to use plutonium for nuclear weapons production after its unloading from the ENF.

## 7. THE ENF STRUCTURAL DESIGN VERSIONS

There are two ENF versions:

- with horizontal blanket(s) and target(s)
- with vertical blanket(s) and target(s).

The proton beam could be a single one (the first version) or can be divided into several beams where each beam interacts with a single module (a module consists of a target and a blanket - the second version).

The target material could be solid (W for instance) or liquid (for example, Pb, Pb-Bi, molten salt). The target may have a window to divide plenums of the target and accelerator (the first version) or may have it not (the second version).

The blanket design may be of heterogeneous or homogeneous type with a thermal or fast neutron spectrum.

To provide the thermal neutron spectrum ITEP specialists are now in the process of investigation of heavy water as a moderator for two different versions:

- with a solid fuel and targets made of fission products and actinides and a heavy-water coolant (the same as "CANDU" heavy-water reactor) - a heterogeneous version. In this version application of target materials in liquid form in special circuits that go through blanket so as to exclude the process of solid target manufacturing;
- with liquid fuel and targets made of materials such as nitrate salts dissolved in a heavy-water moderator - a homogeneous version.

ITEP specialists plan to investigate the possibility to use fluorine molten salts in future.

From the above versions the most preferential is the version with combined target and blanket wherein the problem of radiation resistance of walls separating target and blanket is excluded. ITEP specialists have developed in 1985 a combined target and blanket with Pb-Bi coolant and spherical fuel elements.

To provide fast neutron spectrum the specialists from PPI (Obninsk) investigate the possibility to use Pb-Bi coolant in the blanket.

At the present time the Design Bureau of Machine Building (N. Novgorod) is conducting study of various structural designs of the ENF. After finishing this study one ENF version could be chosen.

It should be pointed out that a solid fuel version is based on the verified technical approaches which increase the possibility of its implementation. A repeated radiochemical reprocessing of the solid fuel is necessary at the same time because of limitations on the fuel burnout value which rise the irretrievable losses during reprocessing. The liquid fuel version is deprived of this disadvantage, however rather complex technical problems of their own arise in this version.

Within recent years ITEP specialists suggested a number of interesting ideas, namely:

1. A double-purpose ENF consisting of two modules (each module involves an homogeneous blanket and a target) for power generation and transmutation of the HLW with liquid fuel proposed by Dr. B.R. Bergelson from ITEP.  $^{238}\text{U}$  is loaded in one module to produce  $^{239}\text{Pu}$ .  $^{239}\text{Pu}$  producing actinides and FPs from power reactors are directed into the second module for power generation and incineration. A continuous chemical reprocessing of the circulated liquid fuel is performed in this case. The results of this study have been presented at the International Conference in Las Vegas in July 1994 [7].

2. Unique liquid fuel elements placed in the form of channel array in the ENF heavy-water moderator [8] are suggested by ITEP specialists Drs. V.D. Kazaritsy, V.R. Mladov et. al. for use in the ENF with a liquid fuel. The liquid fuel assembly design is shown in Fig. 1. As is seen in Fig. 1 there are two circuits inside a channel: a fuel circuit (a solution or suspension of powder in the heavy-water) and a circuit of the heavy-water coolant under pressure. The coolant serves to cool the fuel which circulates in the channel and leaves blanket for chemical reprocessing only. Part of the fuel composition is continuously extracted for reprocessing, fractionated and actinides are returned into fuel circuit and all the rest undergoes further processing: fractionation, storage, transmutation and disposal.
3. Application of a sectioned blanket with neutron valves which permits to rise the neutron multiplication in blanket and decrease requirements imposed on proton current [9].

## 8. TECHNICAL PROBLEMS OF THE ENF DEVELOPMENT. NEW TECHNOLOGIES

The blanket is the most complex unit of the ENF. Its complexity lies in the fact that high thermal neutron flux density equal to  $5 \cdot 10^{15} \text{ cm}^{-2} \cdot \text{S}^{-1}$  and higher needs to be provided. Firstly, the possibility has to be found to rise the power generation density compared to those achieved in existing power reactors without decrease in reliability level. Secondly, there is a need in experimental substantiation of possibility to use section blanket with neutron valve without decrease in reliability level. Thirdly, in the case of use of the fuel and target material in liquid form the express technology of irradiated materials purification from non-metallic impurities needs to be developed. It is necessary to substantiate the radiation resistance of plutonium fuel with addition of minor actinides too. There is a need to check the possibility of the feasibility of the ENF accident protection from accelerator switch off signal.

In the case of initial target application the problem of membrane (window) development arises which divides the volumes of target and accelerator, has minimal proton absorption, must be manufactured of a heat-conducting material or has a cooling, must be remotely replaced when a given neutron fluence is reached. It is expedient to study the possibility to abandon a separating membrane. This problem is however more complex than a problem of the membrane design development. The technology to purify the target material has to be developed too. To provide the acceptable power generation in target during interaction with proton beam two complex technical problems have to be solved:

1. to split the proton beam into several beams with lower intensity;
2. to widen the beam to the size needed.

Rather complex technical problems face the proton accelerator designers. One of these problems is a problem associated with development of a reliable RF accelerator power supply system. Existing 1 MW clystron generators with 60-70% efficiency have low service life and reliability. The second problem is to guarantee the required level of proton losses in various stages of their acceleration. According to calculations made in ITEP by Prof. Kapchinsky the beam losses have to account for appr.  $10^{-6}$  of a nominal value. Specialists of ITEP and other institutes dealing with these problems believe that existing technical problems in the ENF development could be solved provided that means and efforts of specialists are concentrated on this.

## 9. R&D TO BE PERFORMED TO SUBSTANTIATE THE ENF FEASIBILITY

To substantiate the ENF feasibility a number of experimental investigations should be performed which verify the design parameters, safety level and reliability. The main design approaches for the following systems and technologies should be verified:

- blanket;
- target;
- accelerator;
- control and protection system (CPS);
- technology of express purification of fuel and target materials in liquid state;
- technology of selective partitioning of FPs and actinides.

A tremendous work in nuclear data bases compiling should be done.

According to the decision of the Scientific & Technical Council of the RF Ministry of the Atomic Power Industry a R&D program for 1995-2000 is elaborated which makes provisions for works both with ENFs and fast reactors for incineration of actinides.

As for ENFs the following experimental works are incorporated in this program:

1. Modelling of different versions of a blanket with a 56 MeV proton accelerator driven subcritical assembly. For this purpose the program calls for construction of an experimental facility named "Transmutation neutron source" using the building and biological shielding of the HWR research heavy-water reactor. Berillium is supposed to be used as target material. Istra-36 experimental proton accelerator already existing in ITEP and tested at 200 mA pulse current will be used too.

The neutron source main goals are [9]:

- application as a full-scale physical model of the power transmutation facility blanket;
- check and improvement of the control and protection system for electro-nuclear complex;
- fundamental research (actinide constants for fast neutron parameters of proton interaction with blanket and accelerator materials, production of ultra-cold neutrons and use of them, etc.)

The neutron source will have the following parameters:

- proton energy, $E_p$	36 MeV
- proton pulse time, $T_p$	150 $\mu$ s
- proton pulse current, $I_p$	150 mA
- mean proton current, $I_p$	0.5 $\mu$ A
- proton pulse power, $W_p$	5.4 MW
- intensity of the target fast neutrons	$1.5 \cdot 10^{14}$ neutrons/s
- K of the start-up blanket	0.95
- start-up blanket power	25 kW
- thermal neutron flux reflection	$1.5 \cdot 10^{12}$ neutrons/cm <sup>2</sup> S
- start-up blanket load <sup>235</sup> U (90%)	2 kg

Commissioning of this facility will be determined by extent of funding which are scarce now.

2. Investigation of power generation in various targets as a result of interaction with protons, neutron and  $\gamma$ -quanta yields in spallation reactions, and reaction rates.
3. Investigation of cross-sections of the proton and neutron interaction with FPs and actinides (a number of Russian institutes are involved in this activity: Physical Power Institute, Radium Institute, All-Russian Research Institute of Experimental Physics, Institute of Theoretical and Experimental Physics).
4. Study of liquid target hydrodynamics on models.
5. Experimental verification of accelerator and some other units.

Pursuance of these research works allows to initiate the development of the ENF design not earlier than in 1998. At the same time the ENF feasibility study and construction site selection will be performed.

## 10. NON-PROLIFERATION PROBLEM

The problem of non-proliferation arises in the context of use of weapon-grade plutonium as nuclear fuel. This problem is determined by political, economical and technical considerations. Let us dwell on one technical aspect only relating to the requirement of impossibility to manufacture nuclear weapons from a stolen weapon-grade plutonium. There are two ideas which implementation allows in our opinion to prevent or at a last resort complicate weapon-grade plutonium theft considerably. The first idea is in mixing of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  with weapon-grade plutonium and storage of this mixture in a storage. The fuel elements for loading into ENF blanket are then produced without separation of Cs and Sr from Pu. Because neutron capture cross-section of Cs and Sr are small their effect on neutron balance in blanket will be insignificant. It is obvious that first a mixing technology and second a technology of remote production of such fuel should be developed. If we manage to substantiate this idea for ENF, then a possibility to abandon purification of plutonium and some FPs with low absorption during the process of SNF reprocessing.

The second idea originally proposed by Prof. A.N. Shmelyov (Moscow Physical Engineering Institute) is that  $^{237}\text{Np}$  is admixed to weapon-grade plutonium with further irradiation in the ENF's blanket. The Np content will rise with irradiation time of this type of fuel as a result of which plutonium contained in SNF will be unsuitable for nuclear weapon production. We propose to study these two ideas and hope to make some contribution in solving this complex political problem using technical means.

We would like to dwell on one issue. In the case of MOX-fuel use in the ENF's blanket (i.e. the use of fuel with natural or depleted uranium) the possibility to produce weapon-grade plutonium could not be excluded. The similar note refers to existing and promising power reactors with MOX-fuel and to ENF operating mode used for plutonium production of depleted uranium.

## 11. PROPOSALS FOR INTERNATIONAL COOPERATION

In recent years for a variety of reasons a large number of experts in nuclear field were relieved of their duties in different nations. Part of them became involved in HLW transmutation and utilization of the weapon-grade and power plutonium.

In connection with this it is expedient to put for consideration as the first proposal to arrange an exchange of research reports on the themes discussed at this meeting.

The second proposal is to conduct scientific seminars and international conferences on themes discussed here systematically. In this respect the activity of the NEA/OECD which conducts the 3rd International Meeting should be supported. ITEP is ready to propose itself as a candidate for the 4th or 5th Information Meeting on the problem discussed to be held in Moscow in 1995-1996 provided that NEA will give its financial support. It would be useful to provide in the NEA program to conduct twice a year meetings of experts of different countries on some specific problems.

The third proposal is to prepare the international program on partitioning and transmutation. This would permit to coordinate the activity of scientists and specialists from various countries. In the case of positive attitude to this proposal ITEP is ready to submit the relevant proposal. ITEP believes that the problem discussed is of no less significance than an international project on thermonuclear synthesis. Should the NEA/OECD managed to unite the efforts of specialists from different nations in an international project this would be an exceptionally important political and technical decision. If this decision will find support, it is also advisable to consider the issue of organizing an international council to coordinate activity in this field.

The fourth proposal is to perform analytical studies of various versions of the ENF blanket and target including the problem of non-proliferation as important international problem including pursuance of calculations of various fuel compositions above.

The fifth proposal is to conduct experimental work to substantiate the ENF including measurements of nuclear constants study of physical and heat engineering features of the target blanket, accelerator, etc. In connection with this ITEP makes proposal to consider a possibility of the NEA/OECD and any other nation participation in funding of the "Transmutation Neutron Source" construction in ITEP and conducting the relevant physical experiments.

## **12. EXPECTED RESULTS**

ITEP is of the opinion that analytical and experimental studies to be conducted in the next 4-5 years will make it possible to develop the design of the ENF which main purpose will be the safe power generation when using weapon-grade and power plutonium with concurrent transmutation of the part of the long-lived FPs and actinides. ITEP believes that international cooperation will permit to save material resources and intellectual efforts of experts. It is seen even in this stage that as a result of this cooperation fundamentally new ideas and technologies could be implemented which are aimed at increasing the safety level of the nuclear power industry.

## **GENERAL CONCLUSION**

1. Physical features of the transmutation process allow implementation of the HLW incineration in neutron flux using specialized nuclear facilities.
2. Electro-nuclear facilities are preferable than fast reactors due to high level of nuclear safety, lower radiotoxicity of minor actinides and a possibility to close the nuclear fuel cycle as to fissionable materials and fission products.

3. International cooperation of specialists from various nations to develop and construct the electro-nuclear facility is necessary.

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Table 1. Radioactive waste in Russia

Radioactive waste from different industries	Liquid m <sup>3</sup> /Ci	Solid m <sup>3</sup> /Ci	Vitrified m <sup>3</sup> /Ci	Other
1. Ministry of the Atomic Power Industry of the Russian Federation	56·10 <sup>6</sup> / 6·10 <sup>5</sup>	-	-	
1.1 Uranium mines				
1.2 Production of UO <sub>2</sub> , uranium enrichment, fuel manufacturing	1.6·10 <sup>6</sup> / 9.3·10 <sup>4</sup>	-	-	
1.3 Reprocessing of the NPPs spent fuel	416·10 <sup>6</sup> / 2.65·10 <sup>9</sup>	-	250t glass/ 25·10 <sup>6</sup>	*)
- storage in tanks	/570·10 <sup>6</sup>	-	-	
- storage in ponds	/700·10 <sup>6</sup>	-	-	
- solid radioactive waste storage	-	/12·10 <sup>6</sup>	-	
1.4 NPPs	1.5·10 <sup>5</sup> / 3.5·10 <sup>4</sup>	1.0·10 <sup>5</sup> /	12·10 <sup>4</sup> /	**)
2. Shipbuilding	1.2·10 <sup>3</sup> / 2.5·10 <sup>2</sup>	1.9·10 <sup>5</sup> /	-	***)
3. Civilian ships	10 <sup>3</sup> / 10 <sup>1</sup>	5.2·10 <sup>2</sup> /	-	
4. Research institutes, hospitals, etc.	-	7.5·10 <sup>3</sup> / 8·10 <sup>5</sup>	-	

\*) 1000 t of the WWER spent fuel

\*\*\*) 5325 t of the NPPs spent fuel at site

\*\*\*) 15 spent cores with 15·10<sup>6</sup> Ci

Table 2.  $\sigma_r, \sigma_f, \sigma$  effective cross-sections and  $\nu$ , the number of secondary neutrons for fast reactor

Nuclide i	$\sigma_r, b$	$\sigma_f, b$	$\sigma, b$	$\nu$ , neutrons/fission
<sup>237</sup> Np	1.4	0.3	1.7	2.9
<sup>238</sup> Pu	0.5	1.1	1.6	3.0
<sup>239</sup> Pu	0.5	1.8	2.3	2.9
<sup>240</sup> Pu	0.5	0.4	0.9	3.1
<sup>241</sup> Pu	0.4	2.4	2.8	3.0
<sup>242</sup> Pu	0.4	0.3	0.7	3.0
<sup>241</sup> Am	1.7	0.3	2.0	3.4
<sup>242<sup>m</sup></sup> Am	0.4	3.2	3.6	3.2
<sup>243</sup> Am	1.0	0.2	1.2	3.6
<sup>242</sup> Cm	0.288	0.185	0.473	3.5
<sup>243</sup> Cm	0.3	2.6	2.9	3.8
<sup>244</sup> Cm	0.6	0.4	1.0	3.5
<sup>245</sup> Cm	0.4	2.8	3.2	3.8
<sup>246</sup> Cm	0.77	0.47	1.24	3.5

Table 3.  $\sigma_r$ ,  $\sigma_f$ ,  $\sigma$  effective cross-sections and  $\nu$ , the number of secondary neutrons for thermal reactor

Nuclide i	$T_{1/2}$ , yrs	$\sigma_r$ , b	$\sigma_f$ , b	$\sigma$ , b	$\nu^*)$ neutr/fiss
<sup>237</sup> Np	2.14+6	203.4	0.706	204.1	2.25
<sup>238</sup> Np	2.117 days	62	1773	1835	2.8
<sup>239</sup> Np	2.355 days	67.5	-	67.5	-
<sup>238</sup> Pu	87.71	433	17.1	450	2.9
<sup>239</sup> Pu	2.41+4	268	666	934	2.877
<sup>240</sup> Pu	6569	1050	0.93	1051	-
<sup>241</sup> Pu	14.35	316	910	1226	2.937
<sup>242</sup> Pu	3.76+5	126	0.7	12.7	-
<sup>243</sup> Pu	4.956 hrs	97	213	310	-
<sup>241</sup> Am	432.6	540	4.01	544	3.21
<sup>242g</sup> Am	16.1 hrs	-	1725	1725	3.26
<sup>242m</sup> Am	141	1637	6372	8009	3.26
<sup>243</sup> Am	7348	243	1.16	244	-
<sup>244</sup> Am	10.1 hrs	-	1856	1856	-
<sup>244m</sup> Am	26 min	-	1291	1291	-
<sup>242</sup> Cm	161.4 days	23.0	< 4	27	-
<sup>243</sup> Cm	28.5	657	127	784	3.43
<sup>244</sup> Cm	18.1	77.2	2.63	79.9	-
<sup>245</sup> Cm	8.5+3	292	1735	2027	3.717
<sup>246</sup> Cm	4.7+3	13.1	1.13	14.2	-
<sup>247</sup> Cm	1.56+3	99	142	241	-
<sup>248</sup> Cm	3.4+5	29.1	1.8	31	-
<sup>249</sup> Cm	64.15 min	1.3	-	1.3	-
<sup>249</sup> Bk	329 days	995	-	995	-

\* ) For nuclides which have no  $\nu$  data the following values were assumed: <sup>242</sup>Pu, <sup>243</sup>Pu - 3.0; <sup>243</sup>Am, <sup>244</sup>Am, <sup>244m</sup>Am, <sup>242</sup>Pu - 3.3; <sup>244</sup>Cm - 3.5; <sup>245</sup>Cm, <sup>247</sup>Cm - 3.8; <sup>248</sup>Cm - 3.9

Table 4. Neutron consumption for the FPs transmutation

Nuclide i	$T_{1/2}$ , years	$\gamma_1$	$R_1$
<sup>90</sup> Sr	28.8	5.94-2	2
<sup>137</sup> Cs	30.17	6.23-2	3
<sup>151</sup> Sm	93	4.16-3	30-250
<sup>99</sup> Tc	2.14+5	6.15-2	1.15-1.5
<sup>93</sup> Zr	1.5+6	6.35-2	150
<sup>126</sup> Sn	1.0+5	5.72-4	
<sup>79</sup> Se	6.5+4	4.49-4	1.5
<sup>135</sup> Cs	3.3+6	6.55-2	
<sup>107</sup> Pd	6.5+6	1.42-3	
<sup>129</sup> I	1.6+7	7.68-3	2

Note: The entry of 5.94-2 type means  $5.94 \cdot 10^{-2}$  and 1.5+6 means  $1.5 \cdot 10^{+6}$ .

Table 5. Secondary radioactivity in incineration of nuclides contained in 1 tonne of the WWER-1000 spent nuclear fuel

Nuclide i	$m_i$ , g/t	$m_i Q_i^{(0)}$ , Ci/t	$m_i Q_i^{(2)}$ , Ci/t	$m_i Q_i^{(2)}$ , Ci/t
$^{90}\text{Sr}$	678	9.4+4	2.2+4	4.3+4
$^{137}\text{Cs}$	1460	1.3+5	4.8+4	9.1+4
$^{151}\text{Sm}$	14.9	405	(4.5+3)- (2.2+4)	(0.75-7.0)+4
$^{99}\text{Mo}$	950	16.2	2.0+6	4.0+6
$^{93}\text{Zr}$	907	2.3		
$^{126}\text{Sn}$	22.4	0.64		
$^{79}\text{Se}$	5.9	0.41	165	320
$^{135}\text{Cs}$	422	0.486		
$^{107}\text{Pd}$	254	0.13		
$^{129}\text{I}$	220	0.039	5.0+3	9.7+3
Total without Zr, Sm		2.4+5	9.2+4	1.7+5
Without Zr, Sm, Cs, Sr			(2.1-2.7)+4	(4.1-5.1)+4

Note: The secondary radioactivity  $m_i Q_i^{(2)}$  is caused basically by  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ .

Table 6. Characteristics of fast reactor stationary mode

Nuclide i	$x_i$ , nuclei	$M_i$ , kg	$q_i$ , Ci	$TI_i$ , l of water
$^{237}\text{Np}$	6.91+25	27.2	19.2	1.28+9
$^{238}\text{Pu}$	1.0+26	39.7	6.84+5	2.74+15
$^{239}\text{Pu}$	2.17+25	8.63	535	2.43+12
$^{240}\text{Pu}$	1.57+25	6.26	1421	6.46+12
$^{241}\text{Pu}$	2.68+24	1.07	1.11+5	1.01+13
$^{242}\text{Pu}$	2.29+25	9.19	36.1	1.51+11
$^{241}\text{Am}$	5.75+25	23.0	7.89+4	8.32+13
$^{242m}\text{Am}$	2.72+24	1.09	1.15+4	1.04+13
$^{243}\text{Am}$	2.70+25	10.9	2182	1.98+12
$^{242}\text{Cm}$	1.58+25	6.35	2.12+7	3.54+15
$^{243}\text{Cm}$	1.60+24	0.646	3.33+4	2.66+13
$^{244}\text{Cm}$	3.31+25	13.4	1.08+6	6.02+14
$^{245}\text{Cm}$	6.21+24	2.53	433	5.10+11
$^{246}\text{Cm}$	2.0+24	0.817	251	2.80+11

Table 7. Characteristics of thermal reactor stationary mode

Nuclide i	$x_i$ , nuclei	$M_i$ , kg	$q_i$ , Ci	$TI_i$ , l of water
$^{237}\text{Np}$	$1.36+24$	0.536	0.379	$2.53+7$
$^{238}\text{Np}$	$1.09+23$	0.043	$1.12+7$	$1.12+15$
$^{239}\text{Pu}$	$8.94+21$	0.0036	$8.24+5$	$1.12+12$
$^{238}\text{Pu}$	$2.58+23$	0.102	1764	$7.06+12$
$^{239}\text{Pu}$	$1.26+23$	0.050	3.11	$1.41+10$
$^{240}\text{Pu}$	$3.21+22$	0.0128	3.08	$1.73+10$
$^{241}\text{Pu}$	$2.75+22$	0.011	1137	$1.03+11$
$^{242}\text{Pu}$	$2.61+23$	0.105	0.412	$1.72+9$
$^{243}\text{Pu}$	$4.09+21$	0.016	$4.30+6$	$1.26+14$
$^{241}\text{Am}$	$4.30+23$	0.172	590	$6.22+11$
$^{242g}\text{Am}$	$5.97+22$	0.024	$1.94+7$	$2.98+14$
$^{242m}\text{Am}$	$3.53+21$	$1.42+3$	14.9	$1.35+10$
$^{243}\text{Am}$	$3.72+23$	0.150	30.1	$2.74+10$
$^{244}\text{Am}$	$8.2+20$	0.0003	$4.23+5$	$1.76+11$
$^{244m}\text{Am}$	$9.5+20$	0.004	$1.14+7$	$4.74+12$
$^{242}\text{Cm}$	$3.21+24$	1.29	$4.31+6$	$7.18+14$
$^{243}\text{Cm}$	$9.44+22$	0.0381	1967	$1.57+12$
$^{244}\text{Cm}$	$1.38+24$	0.559	$4.51+4$	$2.50+13$
$^{245}\text{Cm}$	$5.50+22$	0.0224	3.84	$4.52+9$
$^{246}\text{Cm}$	$1.14+24$	0.465	143	$1.59+11$
$^{247}\text{Cm}$	$6.17+22$	0.0253	$2.35-3$	-
$^{248}\text{Cm}$	$1.97+23$	0.0812	0.344	$3.12+8$
$^{249}\text{Cm}$	$1.6+20$	$6.6-5$	$7.78+5$	$6.76+11$
$^{249}\text{Bk}$	$5.8+21$	$1.83-4$	3825	$2.54+10$

Table 8. Characteristics of fast and thermal high-flux reactor stationary mode

Characteristics	Fast reactor	Thermal reactor	Fast
			Thermal
M, kg	151	3.52	43
Q <sub>sl</sub> , kg	6.34+7	4.57+7	1.4
Q <sub>m1</sub> , kg	2.12+7	4.31+6	4.9
Q <sub>ll</sub> , kg	2.00+6	5.00+4	40
TI <sub>sl</sub> , l HO	3.67+15	1.31+15	2.8
TI <sub>m1</sub> , l HO	3.53+15	7.18+14	4.9
TI <sub>ll</sub> , l HO	3.48+15	2.46+13	140
M <sub>replen.</sub> , kg/yr	39.2	39.2	
Q <sub>replen.</sub> , Ci/yr	1.32+5	1.32+5	
TI <sub>replen.</sub> , l/yr	1.04+14	1.04+14	
Q <sub>unload. ll</sub> , Ci/yr	1.7+5	1.7+5	
TI <sub>unload. ll</sub> , l/yr	1.1+14	1.14+14	

M - total nuclide mass;

Q<sub>sl</sub> - total short-lived activity determined by short-lived nuclides with half life less than appr. 2 days (<sup>238</sup>Np, <sup>239</sup>Np, <sup>243</sup>Pu, <sup>244g</sup>Am, <sup>244</sup>Am);

Q<sub>m1</sub> - medium-lived activity determined by <sup>242</sup>Cm with half life T<sub>1/2</sub> = 161.4 days;

Q<sub>ll</sub> - long-lived activity determined by other long-lived nuclides;

TI<sub>sl</sub>, TI<sub>m1</sub>, TI<sub>ll</sub> - toxicity indexes for these 3 nuclide groups;

M<sub>replen.</sub> - total mass of actinides being replenished per year;

Q<sub>replen.</sub> - total activity of the annual replenishment;

TI<sub>replen.</sub> - total toxicity index of the annual replenishment;

Q<sub>unload. ll</sub> - total activity of <sup>90</sup>Sr and <sup>137</sup>Cs (in this case nuclide yields were assumed 2.2% for <sup>90</sup>Sr and 6.8% for <sup>137</sup>Cs) removed in one year from fission products produced in stationary mode;

TI<sub>unload. ll</sub> - their toxicity index.

Table 9. Blanket performance data for weapon-grade Pu.

ENR thermal power	2200 MW
ENR electrical power	680 MW
Linac electrical power	100 MW
Coolant pressure	10 MPa
Coolant temperature	
input	226° C
output	310° C
Number of linacs	1
Number of target and blanket modules	2
Proton beam power	30-60 MW
Proton energy	800 MeV
Proton current	28 mA
Target:	
material (solid target)	Cu or W
cylinder size	3000 x 6000 mm
Neutron source intensity	$3 \cdot 10^{18}$ neutrons/s
Power density in target	30 MW
Multiplication factor, k	0.97
Blanket factor	1100 MW
Mode of the fuel replenishment	continuous
Number of channels in blanket	380
Lattice pitch	280 mm
Weapon-grade plutonium consumption	850 kg/year

Table 10. ENF main performance data for  $^{237}\text{Np}$  transmutation

ACCELERATOR	
proton energy	1.6 GeV
proton current	0.3 A
TARGET	
inside diameter	0.38 m
material	Pb
proton to neutron conversion ratio	50 neutrons/proton
neutron source intensity	$9.36 \cdot 10^{19}$ neutrons/s
BLANKET	
thermal power	1.8 GW
core height	2.5 m
diameter of the core with Np	2.12 m
multiplication factor, k	0.598-0.572
thermal neutron flux density	$3 \cdot 10^{15} \text{ cm}^{-2} \cdot \text{s}^{-1}$
Np incineration rate	484.6 kg/year

Table 11. Performance data of the ENF modules

TARGET		
material		Pb
BLANKET		
height		2.5 m
diameter (with reflector)		5 m
type of assemblies		CANDU with pressure tubes
number of assemblies		250
pressure tube size:		
inside tube radius		5.42/5 cm
outside tube radius		6.23/5.81 cm
pitch (hexagonal lattice)		23.8 cm
tube material		Zr - Nb
moderator	D <sub>2</sub> O with dissolved nitrite salts,	Tc, Th
Module thermal power		1800 MW
Module electric power		600 MW
ENF life cycle		30 years
Pu initial load in each module		305 kg
Subcriticality level		0.97 - 0.98
Pu consumption in 30 years		100 tonnes
<sup>233</sup> U production in 30 years		78 tonnes
Transmutation of <sup>99</sup> Tc, <sup>129</sup> I, and partially of Sm		from ENF

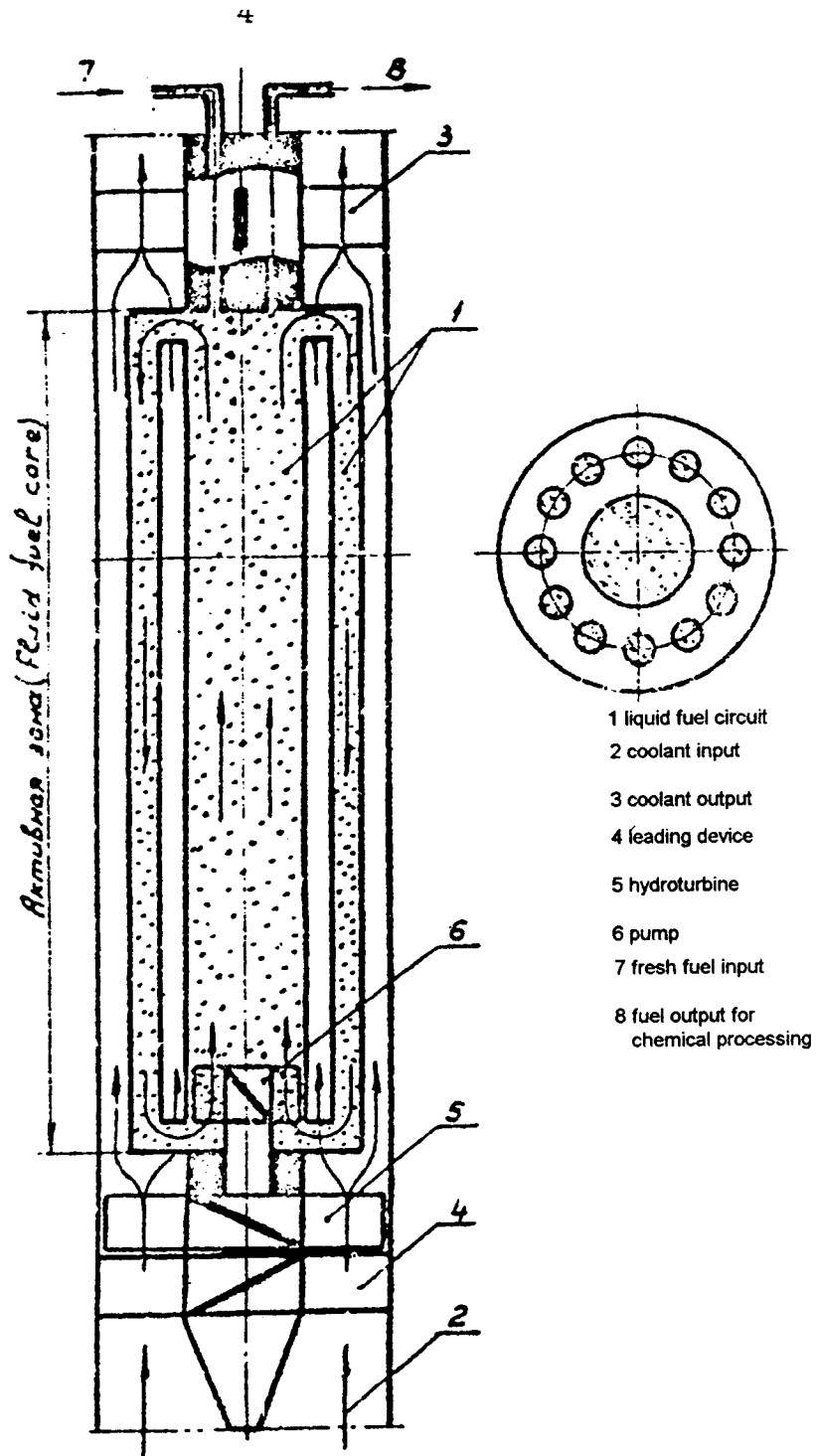


Fig. 1 Principle diagram of the liquid fuel element



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

M. Salvatores (a), M. Viala (a), C. Prunier (a), A. Zaetta (a)  
N. Sasao (b), H. Kaneko (b) K. Takahashi (b)

a) Commissariat à l'Energie Atomique - FRANCE  
b) PNC - Japan

### 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 radiotoxicity in the deposited 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 :

- Partitioning 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<sup>-1</sup> (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 <sup>235</sup>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 TWe 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<sup>5</sup> years. Neptunium becomes important starting from about 10<sup>5</sup> years. Curium makes a large contribution before 10<sup>4</sup> 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<sup>4</sup> 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  $\sigma_f/\sigma_c$  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 are 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 partitioning programme has the following objectives :

- a) Behaviour of actinides and long-lived fission 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 recycling 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 three 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) Development 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 partitioning in the high level liquid waste (HLLW), CEA and PNC have been developing new separation technologies using different bidentate ligands ; DIAMEX process employed with DMDBDMA and CMPO-based TRUEX process respectively. Collaboration to develop partitioning process has been initiated since November 1991, to evaluate 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 transmuting 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 SUPERPHENIX 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**

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"The SPIN Program" This meeting
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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 <sup>2</sup>	10 <sup>3</sup>	10 <sup>4</sup>	10 <sup>5</sup>	10 <sup>6</sup>	10 <sup>7</sup>
TOTAL (Sv/TWhe)	1.1 10 <sup>9</sup>	3.1 10 <sup>8</sup>	7.7 10 <sup>7</sup>	4.2 10 <sup>6</sup>	5.2 10 <sup>5</sup>	1.410 <sup>5</sup>
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 <sup>-4</sup>	2.410 <sup>-3</sup>	3.210 <sup>-2</sup>	9.610 <sup>-2</sup>	1.410 <sup>-1</sup>

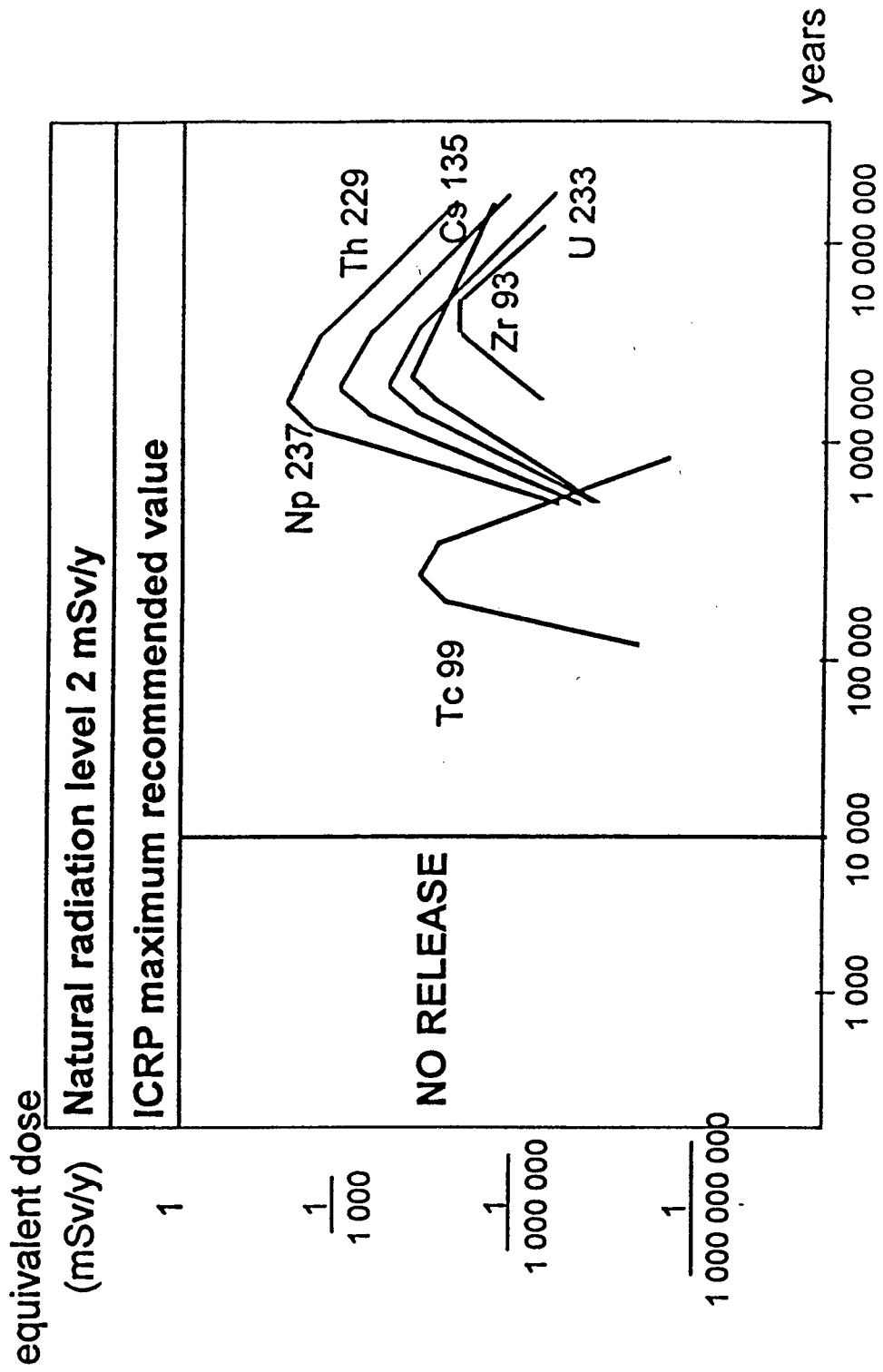


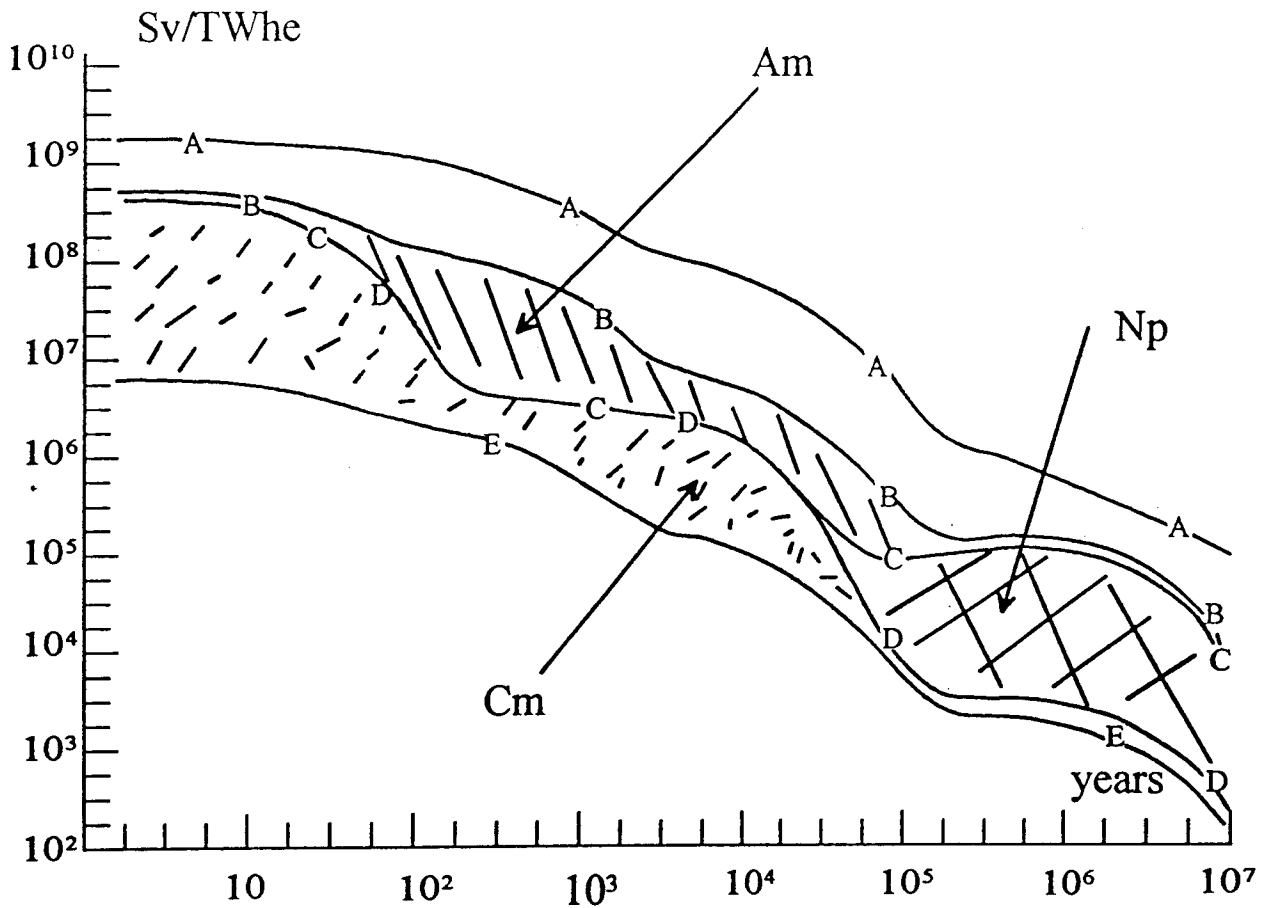
**TABLE 2**  
**PRESENT COLLABORATION**

<b>FIELD</b>	<b>COLLABORATIVE ISSUES</b>
	<ul style="list-style-type: none"> <li>◇ Asses TRUEX and DIAMEX process as MA partitioning tools.</li> <li>◇ Research bidentate extractants, CMPO and DIAMIDE</li> <li>◇ Molecular structure study for innovate ligands</li> <li>◇ Improve PUREX, to enhance Pu/Np recovery, salt-free process</li> <li>◇ Separation Long-lived FPs by macrocyclic compounds</li> </ul>
TRANSMUTATION	<p><u>Fuel</u></p> <ul style="list-style-type: none"> <li>◇ Basic Studies and Data on Fuels and Targets</li> <li>◇ MA Transmutation in a Fast Reactor Core</li> </ul> <p><u>Core Physics and Safety</u></p> <ul style="list-style-type: none"> <li>◇ Basic Studies : Method and Data</li> <li>◇ Experimental studies</li> <li>◇ Parametric Studies</li> <li>◇ Core Safety Studies</li> </ul>

# RESIDUAL RADIOTOXICITY

Fig.1 AURIAT site (concept A) standard scenario





A : once through cycle

B : 0.1 % Pu ; 100 % Am in waste

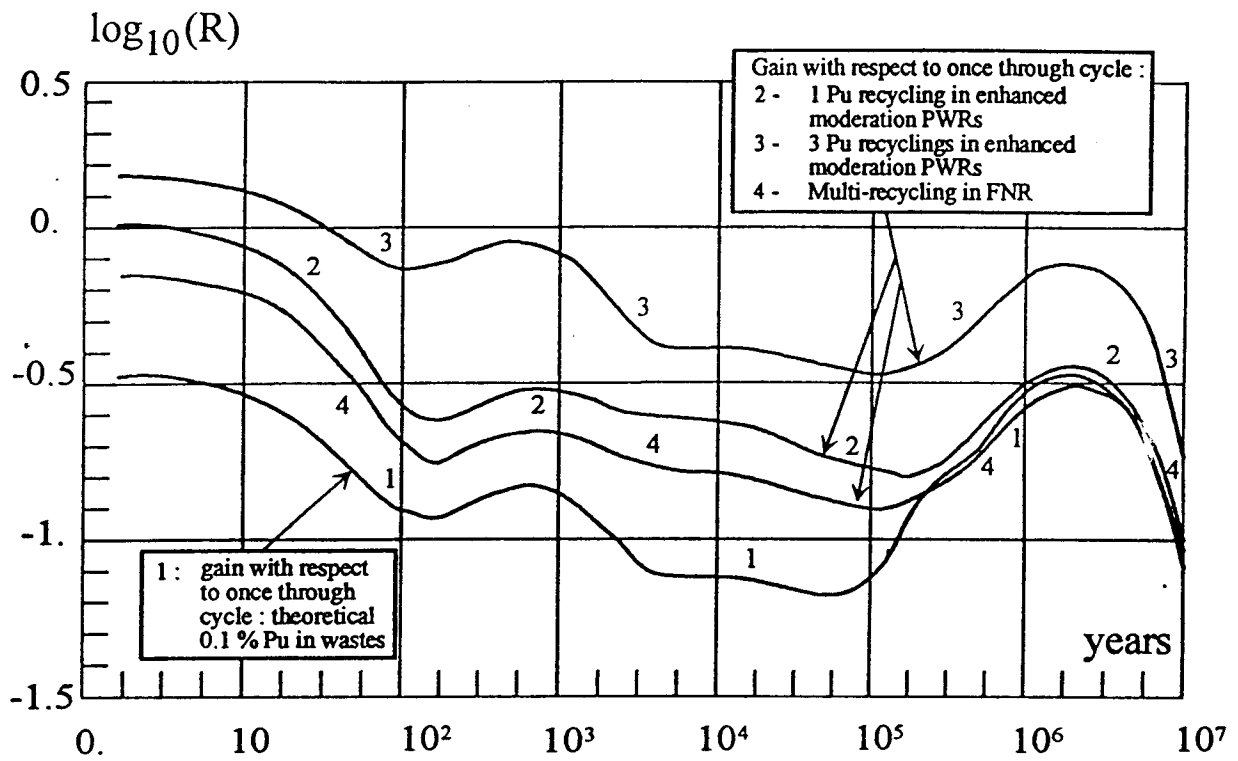
C : 0.1 % Pu ; 1 % Am ; 100 % Np, Cm in waste

D : 0.1 % Pu ; 1 % Am, Np ; 100 % Cm in waste

E : 0.1 % Pu ; 1 % Am, Np, Cm in waste

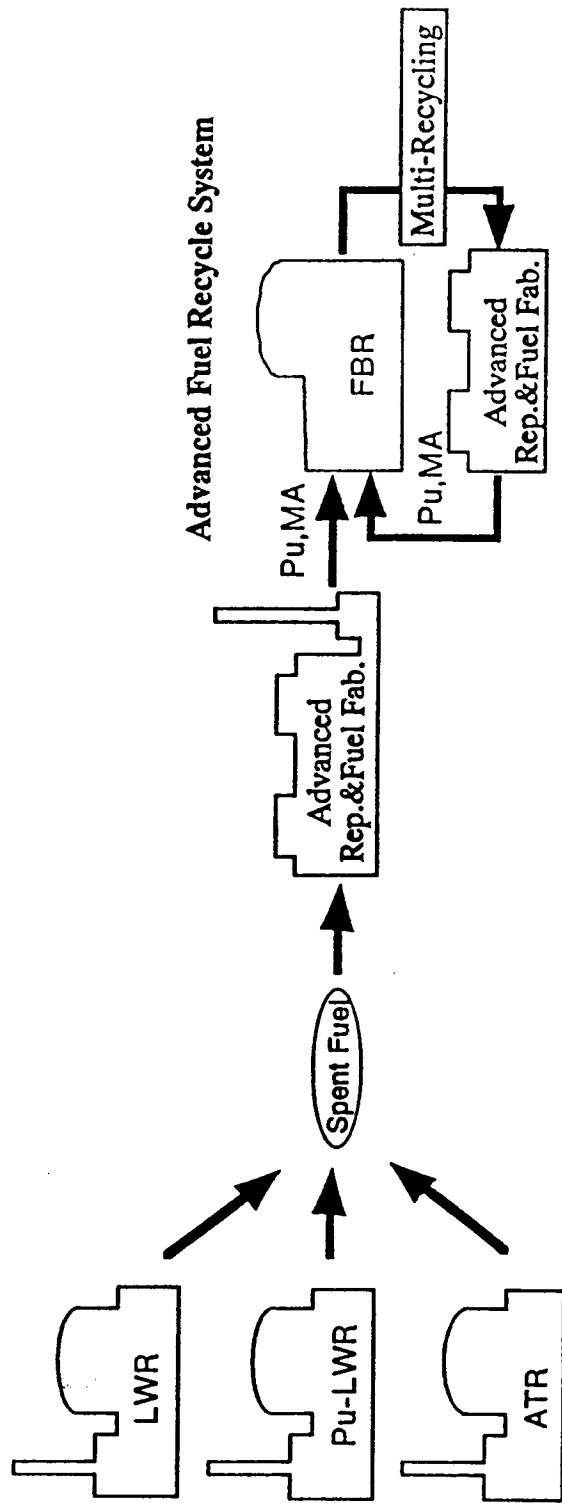
## **RADIOTOXICITY : Theoretical cases**

***FIGURE 2***



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

**FIGURE 3**



**Fig.4 Concept of Advanced Fuel Recycle**

## Potencial Hazard

(Summation of the ratio with inventory and ALI of each nuclides in the waste)

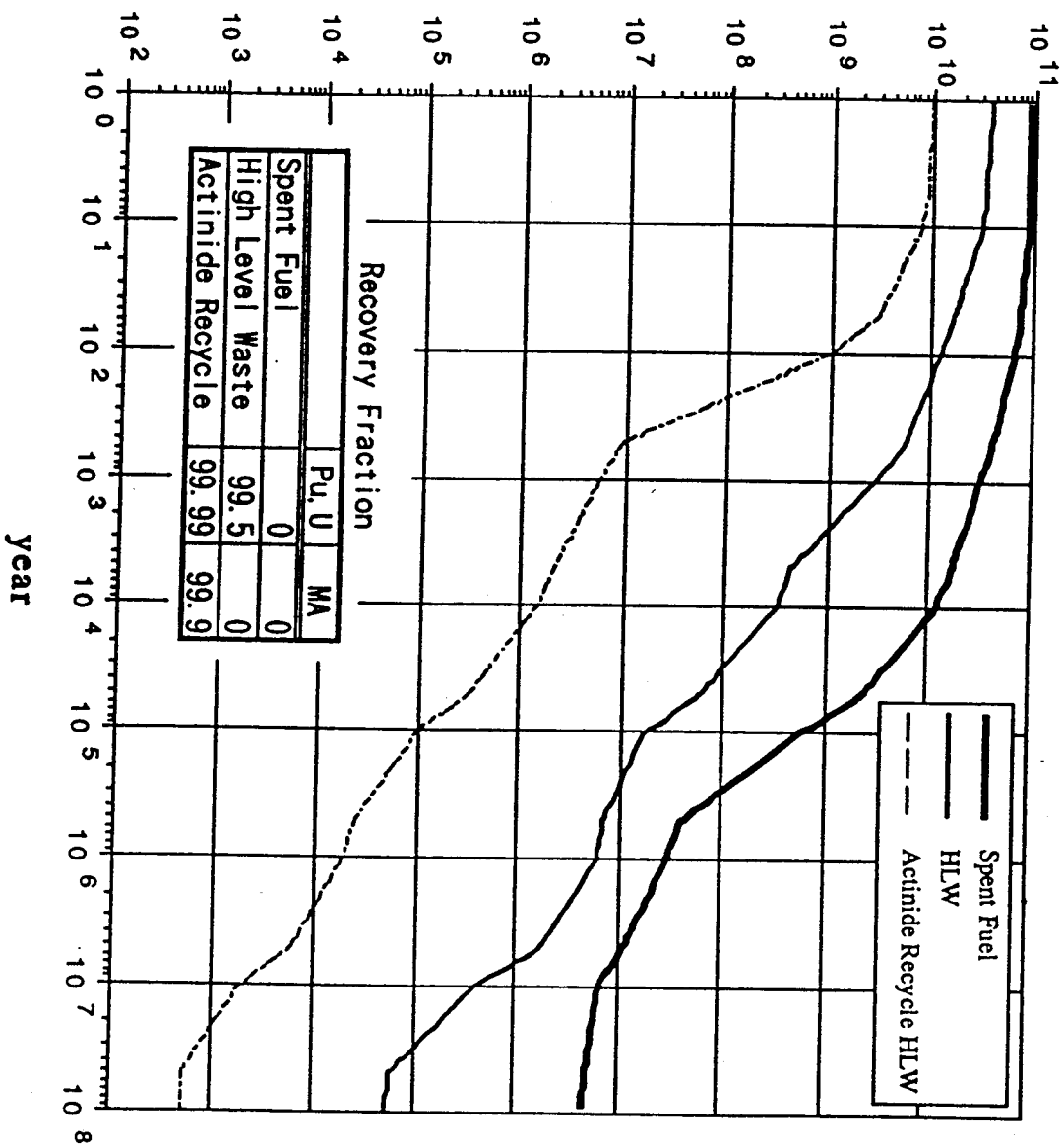
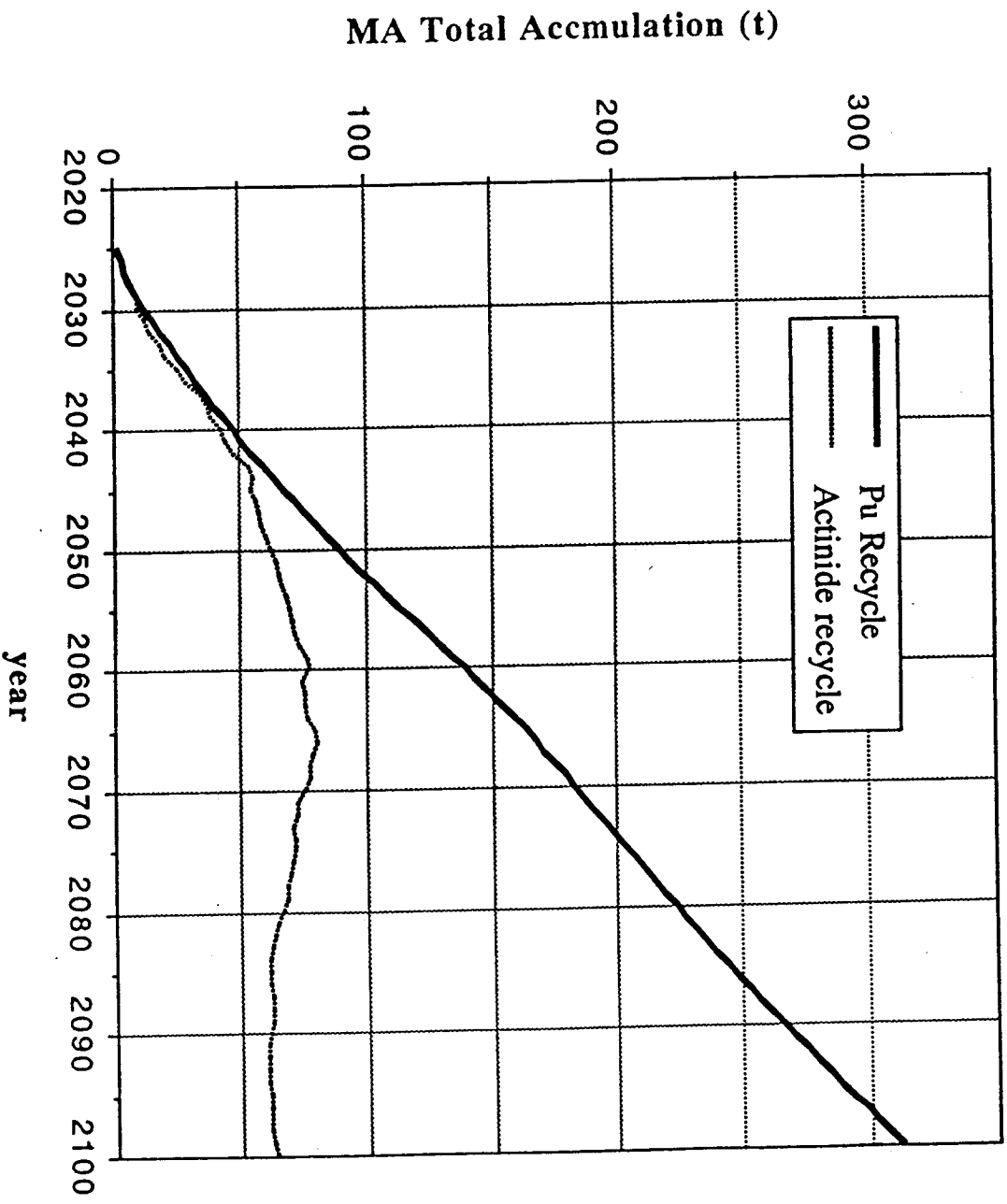


Fig.5 Effect of the Actinide Recovery on the Potential Hazard of FBR-HLW (150,000MWD/t)



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

## EFTTRA, A EUROPEAN COLLABORATION FOR THE DEVELOPMENT OF FUELS AND TARGETS FOR THE TRANSMUTATION

J.-F. BABELOT <sup>1)</sup>, H. GRUPPELAAR <sup>2)</sup>, G. MÜHLING <sup>3)</sup>, C. PRUNIER <sup>4)</sup>, M. ROME <sup>5)</sup>,  
M. SALVATORES <sup>4)</sup>

<sup>1)</sup>European Commission. Joint Research Centre

Institute for Transuranium Elements, Postfach 2340, 76125 Karlsruhe, Germany

<sup>2)</sup>ECN, Nuclear Energy, P.O. Box 1, 1755 ZG Petten (N.H.), The Netherlands

<sup>3)</sup>Kernforschungszentrum, PSF (Bau 419), Postfach 3640, 76021 Karlsruhe, Germany

<sup>4)</sup>CEA, DRN, CE/Cadarache, 13108 Saint-Paul-Lez-Durance Cedex, France

<sup>5)</sup>EDF / SEPTEN, 12-14, avenue Dutrievoz, 69628 Villeurbanne Cedex, France

### ABSTRACT

In the frame of the research programmes on the transmutation of long lived nuclides, many experimental or theoretical investigations have to be carried out within European collaborations, owing mainly to the costs of such studies. Therefore, a group named "Experimental Feasibility of Targets for Transmutation" (EFTTRA), has been formed, with participants from CEA (France), ECN (The Netherlands), EDF (France), KFK (Germany) and ITU (European Commission), to organise joint experiments for the study of materials for the transmutation. So far, it was decided to focus the work on the transmutation of <sup>99</sup>Tc (metal), of <sup>129</sup>I (compound), and of Am (in an inert matrix). Irradiations will take place in parallel in the Phenix fast reactor in France, and in the high flux thermal reactor HFR in the Netherlands. These experiments, together with the related post-irradiation examinations, constitute the first phase of the EFTTRA collaboration. In subsequent phases, EFTTRA will contribute to the development of fuels and targets.

### INTRODUCTION

In the field of the research on nuclear wastes management, the problem of the long-lived radioactive nuclides requires careful thought. The possibility of separating and transmuting these long-lived radioactive nuclides, with the aim of reducing the radiotoxicity of the final waste, has to be investigated. To carry out such experimental or theoretical investigations in the frame of an international collaboration presents many advantages, one of them being to split the costs of the studies. Therefore, the decision to form a group devoted to technical problems, together with European partners, was taken.

### BUILDING-UP OF EFTTRA

Following the Second Meeting of the Working Group on Targets and Fuels organised by ITU on June 1992 in Karlsruhe [1], participants from CEA (France), ECN (The Netherlands), EDF (France), KFK (Germany) and ITU (European Commission), decided to start a collaboration with the aim of setting up joint experiments for the study of materials for the transmutation. The group was named "Experimental Feasibility of Targets for Transmutation" (EFTTRA), and meets regularly since September 1992. A contract has recently been signed, and the support from the Human Capital and Mobility programme of the European Commission is under consideration.



The experimental programme itself, which is divided into phases, is defined in common ; the tasks are distributed according to the potentialities of the partners, and described in technical annexes corresponding to each phase. The balance of the contributions of the different partners is an essential condition for a smooth development of the collaboration ; each partner should bear the costs for his own actions, unless specific arrangements, agreed upon by all partners, are made to restore the equilibrium of the different contributions. More generally, it was decided to share the costs for the purchase of basic materials, for transport, and for the waste treatment. The extension of the collaboration to further partners willing to contribute to the experimental programme, is open, provided they agree with the terms of the contract.

### EFTTRA PROGRAMME

The goal of the EFTTRA collaboration is the study of materials for transmutation, including the fabrication and characterisation of fuels and samples, their irradiation, and the test of their in-pile behaviour. The work should be limited to the basic study of fundamental aspects of the problem. Being the subject of other programmes, the reprocessing and the partitioning as such are not concerned by EFTTRA, but their interrelation with transmutation should of course be taken into consideration. The same remark applies to strategies, where the type of reactor to be used for transmutation is, among others, an important parameter.

The first meeting reflected the concern of the participants to deal with concrete, precise problems : after an evaluation of the needs, and of the subjects already studied otherwise, three topics were finally selected, namely the behaviour of americium, technetium and iodine under irradiation, in the frame of the heterogeneous recycling route ; the homogeneous recycling of Am is the field of other international collaborative efforts, as illustrated for example by the irradiation experiments SUPERFACT 1 [2] and SUPERFACT 2 (in preparation). Irradiations of  $^{99}\text{Tc}$  (metal), of  $^{129}\text{I}$  (compound), and of Am (in an inert matrix) were discussed. The first phase of the EFTTRA collaboration was defined as the irradiation of Tc samples,  $^{127}\text{I}$  compounds, and "empty" (without Am, but partially using U to simulate Am) inert matrices, and the related post-irradiation examinations. The irradiations take place in parallel in the Phenix fast reactor in France, and in the high flux thermal reactor HFR in the Netherlands. Possibly, the thermal reactor Osiris will be used at a later stage for some additional experiments, for simulating PWR conditions. In subsequent phases, and more generally, with a feed-back from the results obtained besides on the reprocessing and the partitioning, EFTTRA will contribute to the development of fuels and targets. The phases of the collaboration being defined by objectives, and not by dates, an overlapping of different phases is possible. Fig.1 shows a tentative planning of the EFTTRA activities.

The objective of the irradiation of Tc is the knowledge of its behaviour under irradiation, the determination of the importance of the self-shielding effect, and a first estimation of the transmutation rate. Concerning I, samples of  $\text{PbI}_2$ ,  $\text{CeI}_3$ , and  $\text{NaI}$  (replacing the initially foreseen  $\text{YI}_3$ ) will be irradiated in HFR, for a verification of the transmutation rate, and for an investigation of the chemical interaction with the cladding.

The comparison of the production of plutonium isotopes through transmutation of actinides for  $\text{UO}_2$  based fuels (homogeneous recycling) and for MgO targets (heterogeneous recycling) shows that for the same transmutation rate, the produced plutonium is lower by a factor of 2 in the case of an inert matrix. Therefore, the possibility of utilising inert matrices for the transmutation of Am is worth investigating. The irradiation of several candidate matrices, scheduled for 94/95 in Phenix (MATINA irradiation) [3], will serve as preliminary tests (in some cases with uranium to simulate actinides), mainly of their behaviour under irradiation ; it will allow the selection of the matrices which will be used for a later irradiation experiment with Am.

## STATE OF THE ART

### Irradiation of Tc and Iodine compounds in HFR

In April 1994, the irradiation of 3 technetium samples and 6 iodine compounds has started in the HFR in Petten (Fig.2). The technetium samples, fabricated at ITU, consist of casted metallic rods which are encapsulated in 15-15 Ti stainless steel. For the iodine compounds, three different metal iodines have been selected on the basis of an analysis of their neutronic and physico-chemical properties : cerium triiodide ( $\text{CeI}_3$ ), sodium iodide NaI, and lead diiodide ( $\text{PbI}_2$ ). The samples, fabricated at ECN, are also encapsulated in 15-15 Ti stainless steel. The thermal neutron fluence rate in the core position is about  $2 \cdot 10^{18} \text{m}^{-2} \text{s}^{-1}$ , the irradiation temperature is about  $430^\circ\text{C}$ . At this temperature,  $\text{PbI}_2$  is in a molten state. The irradiation will be stopped after 8 HFR cycles, in January 1995. The expected transmutation rates are about 5% for the technetium samples, and about 3% for the iodine samples.

After irradiation, the samples will be distributed among ITU, CEA and ECN for post-irradiation examination (PIE). The examinations of the three technetium samples will focus on the material behaviour (swelling), the examination of the iodine samples on the interaction of the samples and the cladding. In addition, the transmutation efficiency will be determined by the analysis of the produced ruthenium (radial distribution and absolute quantities) in case of technetium and of the produced xenon in the plenum of the iodine capsules.

### Irradiation of matrices in Phenix

The rig containing the candidate inert matrices (MATINA experiment) for Am transmutation (heterogeneous recycling mode), has been mounted in the Phenix reactor for irradiation ; Phenix has diverged in September 1994, and operation at 2/3 of the power should start before the end of 1994. Fig.3 shows the arrangement of the pins in the Phenix rig.

Five ceramics (oxides, nitrides), and four refractory metals have been selected according to their thermodynamic, physico-chemical, and mechanical properties and to the (scarce) knowledge on their behaviour under irradiation.  $\text{Al}_2\text{O}_3$  and MgO have been chosen for their good basic properties, in spite of a large swelling under irradiation ; MgO being soluble in the conditions of the PUREX process, and inert with the sodium, appears to be the best candidate for the fast reactors. For the PWRs, the best oxide seems to be the spinel ( $\text{MgAl}_2\text{O}_4$ ), because of its good behaviour under irradiation.

### Additional studies on the matrices

New matrices, not included in the MATINA experiment, have been proposed as candidate for the fabrication of Am targets. For these matrices, a lack of knowledge of their properties hindered a strait forward irradiation in Phenix ; preliminary out-of-pile studies are required, and have been started at ITU, with a financing by EDF. A bibliographic study led to a first selection of these new matrices ; ion implantation experiments have been started for a better understanding of possible irradiation damages both in matrices already selected for MATINA (like spinel), and in new matrices. At a later stage, compounds of these matrices with Am will be examined for defects after 1 year storage.

### Irradiation of Tc in Phenix

The irradiation of 3 Tc samples, identical to the samples of the HFR irradiation will be irradiated in Phenix from 1995 to 1998 (ANTICORP 1 irradiation). The fabrication by ITU of these 3 samples should be completed by the beginning of 1995, depending on the progress in the operation of the Phenix reactor.

## CONCLUSION

The EFTTRA collaboration has now reached its cruising speed, with the first irradiation experiments on their way ; the results of each experiment will be reported separately as soon as available. EFTTRA demonstrates the possibility for European partners, to share their resources on a fair basis in order to reach well defined common goals in relation with practical problems. Regular meetings allow an up-dating of the experimental programmes, on the basis of an exchange of information on the latest technical developments in the given field or in correlated fields. The adaptation to the evolution of the individual programmes of the partners, concerning for exemple new priorities, or the budget, is also an important point of discussion ; actually, the time scale of this research project is larger than that of most of the individual programmes of the partners. The way EFTTRA operates seems to be an answer to this paradox.

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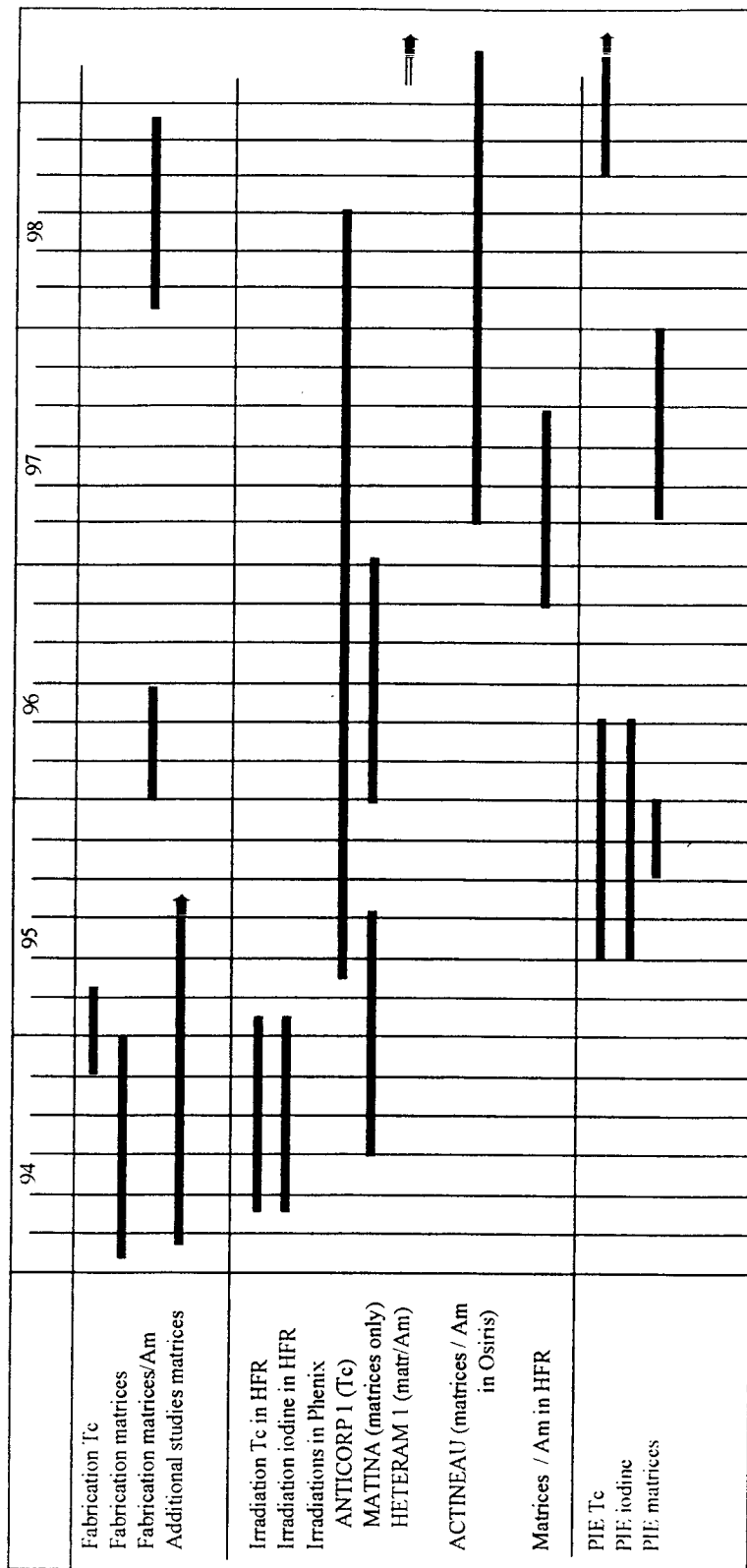


Figure 1 : Planning EFTTRA (tentative)

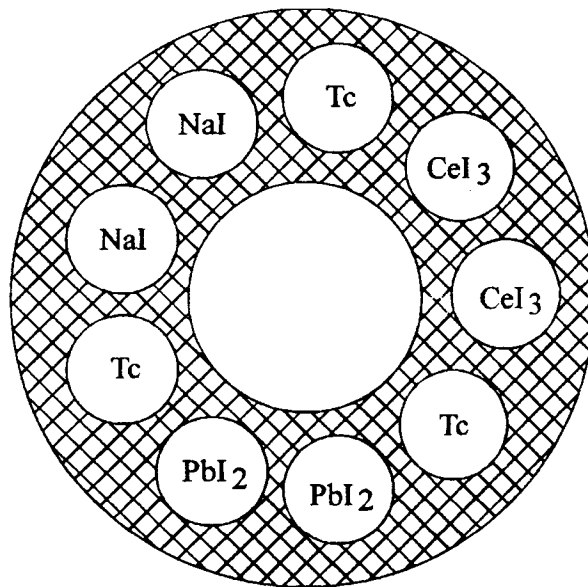


Figure 2 : Position of the samples in the rig for the irradiation of Tc and iodine compounds in HFR

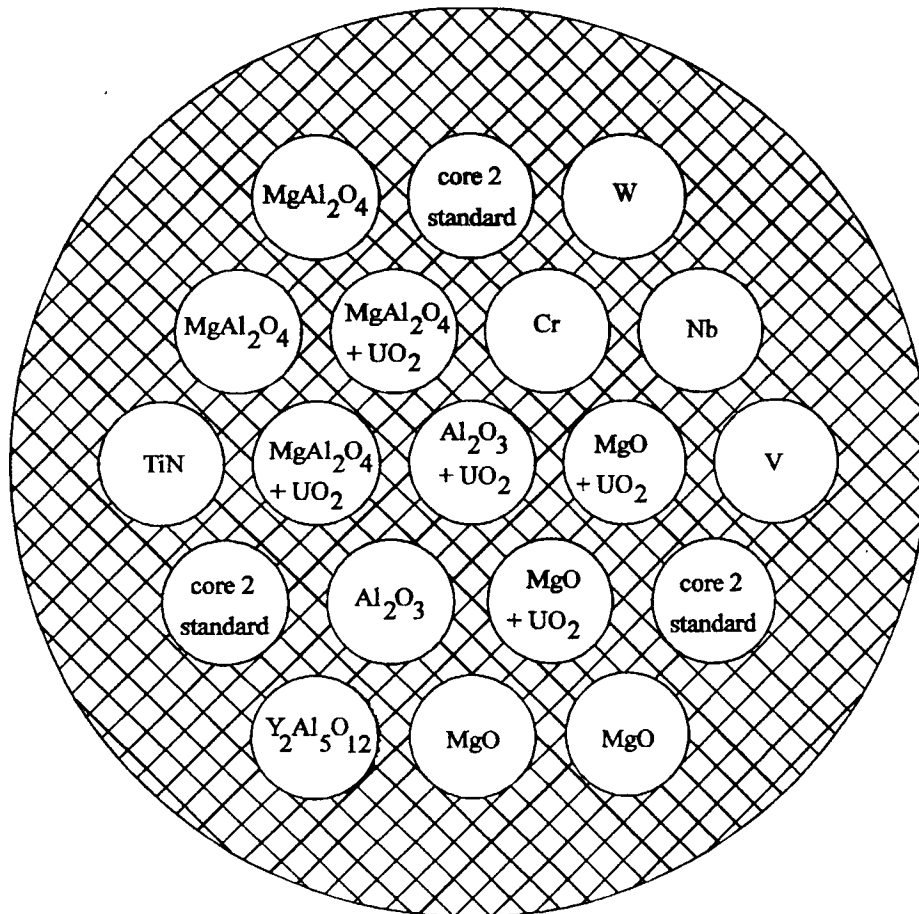


Figure 3 : Arrangement of the samples in the Phenix rig for the MATINA experiment