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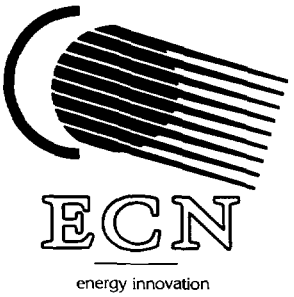

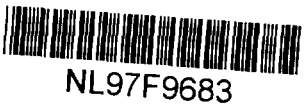

TRANSMUTATIONS OF NUCLEAR WASTE

Progress Report RAS Programme 1995: Recycling and Transmutation of Actinides and Fission Products

H. GRUPPELAAR
E.H.P. CORDFUNKE
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- A review study of the use of the thorium cycle as a means for nuclear waste reduction, has been completed. A follow-up of this work is embedded in an international project for the 4th Framework Programme of the EU.

- A systematic investigation has been initiated on non-proliferation issues and radiological impact associated with partitioning and transmutation options.

CONTENTS

1. INTRODUCTION

In 1991 the Netherlands Energy Research Foundation ECN has defined a programme on recycling and transmutation of long-lived nuclides, in particular actinides and fission products. This programme is known under the Dutch acronym RAS [1-3]. Since 1991 a large number of studies has been carried out in the framework of this programme, and international cooperation and networks have been established. In 1994, a status report on the RAS programme for the period 1990-1993 [1] was issued in the framework of a government report on nuclear energy in the Netherlands ("Dossier Kernenergie 1993"). The RAS-programme is regularly reviewed by a committee set up by the Dutch Government [2]. An extensive status report over the year 1994 was issued in 1995 [3]. The present report describes the work performed in 1995 and is written as a follow-up report of Refs. [1,76], to enable the review committee to monitor the progress of the RAS programme. ECN's current views on international developments with respect to recycling and transmutation will be described in a separate report, to be issued later [95].

Previous work

During 1991 to 1994 a great deal of experience has been gained. Reviews have been made of Partitioning and Transmutation (P & T) in general, the separation and partitioning issues, the possibilities of the "evolutionary route" using thermal and fast reactors, the potential of the Integral Fast Reactor, accelerator-based systems and the thorium cycle. Experimental research has been accelerated by building irradiation facilities in cooperation with IAM, Petten. In addition a special Actinide Laboratory has been designed for supporting the experimental activities in the coming years. A number of samples of Tc and I were inserted in the HFR research reactor and plans had been made to continue the irradiations with inert matrices and actinides.

In the status report over 1994 [76] it was recommended that further research on P& T in the framework of the RAS programme should concentrate on the following items:

1. Evaluate benefits and drawbacks of P & T, including risk assessment and non-proliferation issues.
2. Follow and review international partitioning developments. Contribute to the development of the Supported Liquid Membrane (SLM) technique for the separation of trivalent actinides and lanthanides.
3. Concentrate on fuel research for targets for transmutation, with emphasis on the role of inert matrices in combination with Pu and Am, in cooperation with EFTTRA and other partners.
4. Concentrate on the study of a higher percentage of MOX in LWRs, preferably in the framework of industrial cooperation.
5. Review possible advantages of CANDU and HTR reactors from the perspectives of waste and Pu-burning, in cooperation with AECL and KfA.
6. Contribute to the CAPRA programme on fast burners with reactor physics and fuel studies.
7. Follow developments of accelerator-based systems and give a special R & D contribution, in cooperation with NEA, EU and KVI, Groningen.

8. Review options of thorium cycle for various reactor systems,including acclerator-based systems, in cooperation with EU partners and AECL.
9. Finalize fission-product studies in 1995, apart from HFR irradiation and post-irradiation investigations.
10. Continue contributions to strategy studies, in particular those related to stable nuclear energy production and phasing-out options.

In the programme over 1995 these recommendations were taken into account.

Review committee

The review committee is chaired by Dr. J.N.C. van Geel (ITU, Karlsruhe) and is made up of nuclear experts from government departments, research institutes and environmental organisations. Although the committee largely agreed with the results of the status reports 1993 and 1994, they did make some critical remarks and recommendations. One of the recommendations was to separate the status report over 1995 into two parts as outlined above. A general remark was to concentrate the programme to less topics. Efforts have been made to narrow down the options and to shift part of the analytical work towards experimental irradiations of transuranics with inert matrices, in cooperation with European partners. The analytical work is been directed more towards MOX options in LWRs and the evaluation of thermal burners. The committee appreciates work on thorium and accelerator options; a possible combination of these topics may be practical in future work. Finally, the committee recommended to rescrict the study of risks of P & T and of proliferation risks by concentrating on a qualitative description of possible radiological effects of P & T for the present generation and of technical non-proliferation measures for future fuel cycles, respectively. These recommendations have been taken into account in the programme for 1995-1997.

Progress 1995

The progress of work performed in 1995 is described in chapter 3, following the subdivision of projects as given in chapter 2. In chapter 5 all publications over the period 1990-1995 are listed.

2. RAS PROGRAMME 1994-1997

This chapter describes the current 4-year programme as defined in 1993. The progress over 1994 is given in Ref. [76]; the progress over 1995 is described in the next chapter.

Objectives
The general objective of the RAS programme is to contribute to international research on recycling and transmutation of nuclear residues and to indicate possibilities of including this option in an acceptable waste management strategy. Two goals are considered: the reduction of the overall radiotoxicity of nuclear waste over a very long period and the reduction of the radiological effects to the population due to leakage of long-lived radionuclides.

First of all it will be investigated whether it is feasible to reduce the actinide production itself, e.g. by application of the thorium cycle or by replacing U-238 by inert matrix material.

Secondly, investigations will be made on reducing Pu losses during reprocessing and on separating (partitioning) the minor actinides (Np, Am and perhaps Cm), and some long-lived fission products (Tc-99, I-129).

Thirdly, investigations will be made on methods to deal with the reprocessed and partitioned products, e.g. by studying
. re-use of U and in particular Pu as fuel in thermal and fast reactors ("evolutionary" route, including the use of MOX);
. direct fission of minor actinides in fast spectra;
. transmutation, followed by fission of minor actinides in thermal spectra;
. transmutation of long-lived fission products in intense thermal spectra;
. alternative treatment by conditioning or isotopic dilution (in case of I-129).

At the end of this 4-year programme the technical feasibility and the risks of P & T will be evaluated and a proposal for the implementation of P & T in an acceptable waste management strategy will be presented.

Questions to be answered
In particular the following questions should be answered:

1. Does the "evolutionary route" lead to an important reduction in the long-term radiological hazard? In other words: Is it possible to further improve the current fuel cycle and reprocessing strategy by application of MOX and possibly MINOX fuel in thermal and eventually fast reactors?

2. Can advanced and innovative reactor systems or even accelerator-driven reactor systems supplement the "evolutionary route"?

3. Can the introduction of other fuel cycles, such as the thorium cycle lead to a better solution? Are accelerator-based thorium fuelled reactors (CERN-proposal) feasible?

4. What are the competing demands of safety, economy and non-proliferation? What are the risks, costs and benefits of P & T and how should such quantities be defined and evaluated?

5. What are the consequences for waste management should the period of nuclear energy production be terminated?

2.1 Detailed programme 1994-1997

A detailed description of the RAS-programme proposal for 1994-1997, subdivided into seven projects, is given below. A short characterization of these projects, a global time schedule and of the deliverables are given in Table 1. The progress over 1995 is summarized in chapter 3.

Project 1: General activities

It is important to follow actively the international developments in the field of P & T. To this end ECN is involved in a number of networks for international co-operation, both within the EC and within the NEA (Nuclear Energy Agency of OECD). This project aims to further enhance national and international cooperation and successful participation in EFTTRA and CAPRA project. An additional objective of this project is to inform government and the public on recent developments of the programme. Finally, this project will coordinate all other activities and a yearly status report will be issued such as this report. These status reports will be issued in the English language. At the end of the 4-year period a report will be issued, that should address the questions mentioned above.

Project 2: Reactor physics and scenario studies

Evaluation of nuclear data and the accelerator option
For reactor physics research and scenario studies a good data base is essential. This is even more important to assess the safety of transmutation facilities. An important start to this project was given in the period 1991 to 1993 by work performed under contract of the Ministry of Economic Affairs and also by work for the European Strategy Study. From these studies it followed that more work is necessary on fission yields and decay constants of actinides in working libraries and also on nuclear cross-sections for various matrix- and cladding materials. For the assessment of safety parameters more data are needed for minor actinides and even for Pu isotopes. This also applies to nuclear data which are needed for the evaluation of the thorium cycle. Finally, more work is needed on high-energy cross sections in the 20 MeV to 1.5 GeV range to evaluate the accelerator-based transmutation option. On a number of these items contributions will be made in close cooperation with the NEA Data Bank and the Nuclear Science Committee of NEA. In 1995 the possibility to perform a Ph-D study on the accelerator option will be investigated. Support of the EC programme will be requested, in particular to evaluate the recent CERN proposal.

Reactor physics Ph-D studies
The current Ph-D study on actinide transmutation concerns scenario and inventory calculations of actinides in various reactor types. A major part of the

thesis is devoted to ALMR studies, related to the IFR/PRISM development and has been realized by means of a detachment at GE, supported by GKN. In 1994, the work was concentrated on the potential of molten-salt reactor systems. After finishing this study by the end of 1994, a follow-up Ph-D study on the potential of High Temperature Gas-cooled Reactors for Pu and minor actinide burning will be initiated.

Reactor physics research and scenario studies
It is intended to give a rather complete survey of the possibilities of the various reactor types and accelerator-based options. In-depth studies are needed with emphasis on reactor physics consequences of large-scale recycling and transmutation, in particular with respect to safety features. One important aspect will be the study of possibilities for use of MOX and MINOX in existing reactor types ("evolutionary route"). Further use of MOX in LWRs contributes to the consumption of a growing amount of plutonium. Other solutions to reduce the plutonium stock will be investigated as well. Developments at CEA (CAPRA) and GE (PRISM) with respect to fast reactors for incineration of actinides will be followed and the capabilities of HTGRs for Pu-burning will be investigated. Finally, the evaluation of accelerator options, and of accelerator-driven reactor systems, will be continued with special emphasis on the recent proposal made by CERN.

Systematics of transmutation in various neutron spectra
In 1994 a systematic study on the possibilities of actinide transmutation in various neutron spectra and reactor types was started. The goal of the study is to offer means for optimization of transmutation facilities for actinides. A PC code will be issued with data for easy calculation of transmutation effects of various components of neutron flux spectra, such as Maxwellian, 1/E and fission spectra. In this study effects of self-shielding will be accounted for as far as possible.

European Strategy Study and follow-up
The European Strategy Study started in 1992 and will be finished mid 1995. This study is supported by the European Commission. Partners are: CEA, Siemens, ECN and since 1994 also AEA and Belgonucleaire. In this collaboration ECN contributes with an extensive status report on long-lived fission-product transmutation and by delivering improved nuclear data for transmutation. At present the research is focused upon large-scale possibilities for transmutation of long-lived fission products in various reactor and accelerator systems. ECN also studies the features of heavy water reactors for efficient transmutation of Tc-99, including safety aspects (moderator void coefficient). In cooperation with ECN's subcontractor Belgonucleaire the transmutation of actinides in MOX (MINOX) in PWRs is being studied. A number of possible items for a follow-up programme will be prepared in cooperation with Belgonucleaire and other partners, in particular to study the full MOX option. In the 4th Framework programme this work will be continued.

Physics research on inert matrices
To support the irradiation experiments in the HFR (see project 5) a number of reactor physics calculations will be performed. In these experiments the actinides are embedded in an inert matrix material. The calculations are needed to determine optimum irradiation conditions as well as parameters for the interpretation of the measurements. In particular radial burnup effects will be

evaluated in Tc and for uranium oxide, plutonium oxide and americium oxide in various inert matrices.

A second phase of this project is a joint investigation with AECL-Canada to evaluate the option of actinide- and/or Pu-burning in a CANDU reactor. ECN will calculate nuclear constants for 3D calculations as a function of burn-up and perform a number of benchmark calculations with Monte Carlo methods to determine burn-up parameters and reactivity coefficients. The potential of other Pu burners with inert matrices and burnable poison will be studied as well. Also an investigation will be made on inert matrices with a significant contribution to the Doppler effect.

Project 3: Fuel cycle and non-proliferation aspects

In the search for possible transmutation systems the study of proliferation aspects plays an increasingly important role. Therefore, ECN has started a systematic re-evaluation of the INFCE-study. In the coming years the INFCE re-evaluation will be continued and a discussion on possible safeguards of the IFR/PRISM reactor will be initiated. Furthermore, the proliferation aspects of other reactor cycles will be studied focusing on their P & T possibilities and Pu burning. The instrumental possibilities to detect the isotopic composition of actinide samples is also subject of study. ECN participates in the OECD Working Party of NEA's Nuclear Science Committee. Connection with the CAPRA fast reactor programme is investigated.

Project 4: Thorium cycle

Application of the thorium cycle could lead to a reduction of higher actinides. Within the RAS programme attention will be paid to this item: in 1994 a new project has been started to study the perspectives of the thorium cycle for reducing the long-term radiotoxicity of the actinides in the waste. Physical and chemical aspects will be considered for open and closed fuel cycles. After the generic study the specific features of thorium-fuelled CANDU and HTGR reactors will be studied in detail. Collaboration with AECL has been established and in 1995 the transmutation possibilities of the CANDU reactor will be investigated in detail. Support of the EC will be requested for study of PWR and fast reactor options.

In many accelerator-based reactor systems thorium is proposed as a fertile fuel. This is an additional motivation for studying the thorium cycle.

Project 5: Experimental verification of transmutation

Construction of irradiation facilities
In 1993 an irradiation facility has been constructed in cooperation with JRC-Petten to investigate material properties and transmutation rates of long-lived fission products in the HFR. A second facility has been constructed in 1994 to irradiate actinides diluted in various types of inert matrices. The available ECN Hot Cell Laboratories (LSO) will be extended with specific facilities to handle alpha radioactive samples. This actinide laboratory will become operational in 1996.

Irradiation programme in the HFR as part of the EFTTRA collaboration
A European collaboration, EFTTRA, with partners CEA, EdF, ITU, KfK and ECN and JRC has been established to execute a common irradiation programme in HFR and Phénix on transmutation of long-lived fission products as well as of actinides in inert matrices. ECN, together with JRC-Petten, will perform the scheduled irradiations in the HFR. Supporting physical studies are performed within project 2. This part of the programme includes the preparation of the irradiations, the actual irradiations in the HFR and post-irradiation examinations. ECN will be involved in similar irradiations on identical samples in the fast reactor Phénix. The experiments will require long-term efforts, scheduled beyond 1997. In 1994 the fission products Tc and I were irradiated. Post-irradiation examination has been performed in 1995. In 1996 inert matrices with enriched uranium will be irradiated. An irradiation of Am in inert matrices is also scheduled for 1996. Possibly also materials with good Doppler characteristics will be investigated. Irradiations and postirradiation work may extend beyond 1997. Support from the EC has been obtained to perform experiments on Am transmutation in the HFR.

Project 6: Chemical research on actinides and fission products

Chemistry of actinides and fission products
The actinides will most likely be irradiated as oxides; alternatives are nitrides and carbides. The physico-chemical properties of these actinide compounds should be determined and their interaction with the proposed inert matrix materials should be investigated. This information is necessary to predict the chemical stability of the targets for the transmutation experiments and to predict the behaviour of the fission products and actinides in the targets.
In addition, knowledge of the reprocessing of the actinides and long-lived fission products should be built up. In the first half year of 1994 an employee of ECN participated in the research programme of CEA at Fontenay-aux-Roses where he investigated the partitioning of the actinides and lanthanides using the DIAMEX process. At ECN laboratories advanced reprocessing processes such as Selective Liquid Membranes (SLM) will be investigated in collaboration with the university of Twente (see below).

With respect to the fission products, alternatives for transmutation such as dilution and immobilization will be studied, especially for I-129. Also chemistry of technetium is studied.

Chemistry of inert matrix materials
Transmutation of actinides can only be realised by fission with neutrons. During this process heat will evolve. To obtain a realistic heat production, the actinide-targets cannot consist entirely of actinide compounds but dilution with an inert matrix is required. The material is used as a diluent and must exhibit an inert behaviour during the irradiation. It is called an inert matrix. A number of candidate materials have been selected within the EFTTRA collaboration. A final selection has not been made yet since their behaviour under irradiation, their stability towards the lanthanide elements (common impurities in americium from commercial reprocessing) and their stability are not well defined. These items will be investigated in the coming years.

Ph-D research programme
A Ph-D investigation has been initiated on the possibility to separate lanthanides and actinides by means of organic supermolecules. These molecules can be used as carrier in liquid membranes. The synthesis of the molecules will be done at the University of Twente; the tests with radiuoactive materials will be performed in the actinide laboratory of ECN.

Project 7: Risk analysis of P & T

A study will be initiated on the risks involved in P&T of nuclear residues ("waste") for the present human generation as well as on the reduction of risks of stored waste for future generations. A first step in this direction will be the definition of relevant risk parameters. Secondly, methods have to be developed to quantify these parameters. In a number of cases standard PSA techniques can be used, certainly for the evaluation of additional risks in operating the reactors used for recycling or transmutation. The risks of storage of nuclear waste with and without P&T are evaluated in a separate Dutch programme (CORA programme).

2.2 National and international cooperation

A large number of national and international contacts have been established. On the national level there are excellent contacts with the Joint Research Centre (IAM) at Petten, the Technical University of Delft (IRI), the University of Twente (UT) and the Dutch utility GKN. Possible cooperation with the Dutch utilities' research institute KEMA (Arnhem) and with the nuclear accelerator institute KVI (Groningen) are investigated.

ECN participates in the NEA OMEGA Exchange Programme within NEA-OECD and in activities organised by the NEA Nuclear Science Committee like the "Evaluation Coordination Working Party", the "Task Force on Physics Methods for Transmutation" the "Task Force on Pu Recycling" and a number of activities initiated by the NEA Data Bank. Within the EC, ECN participates in the European Strategy Study on P & T and in the EFTTRA cooperation between CEA, EdF, ITU, KfK, IAM and ECN for target preparation and experimental irradiation in the HFR and Phénix. From 1996 on a number of 5 contracts has been established with the EU in the framework of the 4th Framework Programme.

ECN participates in IAEA activities and has cooperation agreements with GE (San Jose, USA) and AECL (Canada). Detachments of ECN personnel have been arranged at the following institutes: GE (San Jose), CEA (Fontenay aux Roses), CEA (Bruyères-le-Châtel) and Framatome. Participation in the CAPRA programme to investigate burning of actinides in fast reactors has been started in 1995 and will be continued in the following years.

Table 1 Global programme survey 1994 - 1997 **(Adjusted January 1996)**

Cat.	Subject	1994	1995	1996	1997 (tentative)
1. General		co-ordination status report public info scoping studies	co-ordination status report public info scoping studies	co-ordination status report public info CANDU status	co-ordination final report public info Acc. status
2. Reactor physics and Scenario Studies					
2.1	Nuclear data + accelerator option	evaluation data for transmutation	CERN option, co-op KVI, Ph-D study definition	Medium-E data, ECNAF, NEA contr.	continuation validation
2.2	Ph-D studies	1. ALMR thesis: final report	2. Start HTGR study 3. Start ACC. KVI study	2. HTGR Pu burning 3. Acc. KVI theory	2. HTGR Th 3. Acc. KVI exp.
2.3	Scenario studies, CAPRA (phys+chem.)	supporting research	CAPRA fission products + chemistry	CAPRA temp coeff. Inert matrix chem.	Cont. CAPRA final report
2.4	Systematics of Transmutation systems	PC code	final report March	----	----
2.5	EU studies	1. Fission product transm.	1. Final report	2. full MOX data 3. Acc. option 4. MOX contr.	Cont. until Dec. 1988
2.6	Inert matrices	Rim effect	Pu-burning, CANDU	Pu burning thermal reactors Doppler, full core	Cont.
3. Non-proliferation		INFCE re-evaluation	Supporting studies detection methods	New cycles	conclusion, final report
4. Thorium cycle					
4.1	Study Th-cycle (Phys.+Chem.)	generic studies	CANDU	Acc. option (Rubbia)	conclusion, final report
4.2.	EU-study	----------	---------	Co-ord. EU-study PWR contr.	cont. until Dec. 1988

Table 1 (continued) Global programme survey 1994 - 1997 **(Adjusted January 1996)**

Cat.	Subject	1994	1995	1996	1997 (tentative)
5. Experimental verification					
5.1	New facilities	HFR irradiation facilities, actinide laboratory	start building actinide laboratory	completion actinide laboratory	___
5.2	Irradiation exp. + EFTTRA (inc. Phys.)	Tc, I irradiation	Analysis Tc,I Inert matrix + U	U in inert matrices In HFR Continuation Tc irr.+PIE Coord. EFTTRA int.	Continuation + PIE, exp. progr. until 1999
5.3	EFTTRA-EU (inc. Phys)	---------	Definition EU-proposal	Coord. EU-study Am prep.+irr in HFR	Cont. until Jan.99
6. Chemical research					
6.1	Actinides	survey separation	review separation methods	continuation techn. watch.	conclusion, final report
6.2	Inert matrices	research inert matrices	Continuation U Start irr. in HFR	Continuation Irratiation inert matrices	PIE + inert matrices
6.3	PH-D investigations	Definition	Separation lanth. and actinides	continuation Def. second study	continuation
7. Radiological Effects + Risk		Definition risk-quantities	risk-evaluation	global risk-evaluation	conclusion

3. PROGRESS IN 1995

This chapter contains a summary of progress made in 1995 according to the programme 1994-1997, described in chapter 2.
Publications within the RAS programme are presented in chapter 5, the present chapter only reviews the 1995 activities.

3.1. Project 1: General activities

The objective of this project is to coordinate the RAS programme, to enhance national and international cooperation and to inform public and government of status and progress of the programme.

The coordination of the programme was facilitated by means of an internal steering committee, meeting at least monthly, to monitor progress, to review actions and to control the programme.

National cooperation was enhanced by intensifying contacts with the universities of Delft, Groningen and Twente. Early 1995 J. Bultman defended his thesis [42, 63] on Actinide Transmutation in Nuclear Reactor Systems, emphasizing two innovative burner systems, one fast (the ALMR) and one thermal (the molten salt transmuter). Two new Ph.D students were hired, one to study Pu burning in High Temperature Gas Cooled Reactors (cooperation with Delft Technical University) and one to study Nuclear Data for Accelerator-based systems (cooperation with Groningen University). With the University of Twente a collaboration was started to perform a PhD study on the utilization of Supported Liquid Membrane (SLM) techniques for the partitioning of actinides in waste.

On the international scene, ECN continued to collaborate with other partners in many ways. Two papers were presented at the NEA exchange meeting in Cadarache [43, 44]. The contract on P & T with the European Union was completed with two papers [62, 77] and an extensive final report [87]. Follow-up contracts were entered in the framework of the Fourth Framework Programme of the European Union in the fields of Strategy Studies (MOX and Nuclear Data for MOX), Am irradiation (EFTTRA), Thorium (PWR studies) and Accelerator-Based Transmutation (Nuclear Data). The EFTTRA cooperation was further enhanced and ECN chairs this important activity at present. Various conference papers were prepared and presented [51-60, 68, 72], in particular for the GLOBAL'95 conference. In 1995 ECN entered the CAPRA cooperation by signing a contract with CEA and performing physical and chemical investigations. In order to study the possibilities of CANDU type of reactors for burning Pu and minor actinides, the existing contract between ECN and AECL was utilized to realize a detachment of an ECN staff member in the summer of 1995. Continuation of this cooperation is envisaged in 1996. Another staff member was detached to Framatome Industries, Paris, for work on 50% MOX cores in PWRs.
The progress of the work performed in the framework of these cooperations is described in the following sections.

With respect to presentation of the RAS-programme to government and public, first of all the extensive status report covering the work in 1994 [76] should be

mentioned. This report is a follow-up of the 1993 status report [1], of which an English translation was issued in 1994. This status report was considered by the review committee, installed by the Dutch government and recommendations were included in the 1995 and 1996 programmes. There were a number of press contacts, lectures and a publications in a Dutch journal, e.g. Ref. [79].

3.2. Project 2: Reactor Physics and Scenario Studies

General supporting work

To gain insight in transmutation studies undertaken, an easy to use P.C. code has been developed named STAR (Show Transmutation of Actinides in Reactors) [69,70]. It solves analytically the differential equations describing the buildup and removal of actinides and it has powerful capabilities to plot masses, reaction rates, decay rates, radiotoxicity and other parameters as a function of irradiation time or as a function of the storage time. Path-way analysis can be used to plot the most important routes along which nuclides are produced.

Burnup calculations are generally done by a code with two- or three-dimensional geometric capabilities and only a limited number of decay and transmutation chains or by a code with limited geometric capabilities (one dimensional) and an extensive number of decay and transmutation chains. However, transmutation studies and experiments are often characterized by small samples in a complicated geometry and a need for a complete set of decay and transmutation chains (e.g. to calculate the concentrations of neutron emitting Cf isotopes). To meet these requirements, a new burnup code system has been developed, named OCTOPUS, which consists of neutron spectrum codes to calculate the neutron spectrum and nuclide cross sections at the beginning of each time step and of point-depletion codes to calculate the nuclide concentrations at the end of each time step. For the neutron spectrum calculations we can use our SCALE-4, WIMS-6 or MCNP-4 code systems, for the burnup calculations we can use ORIGEN-S or FISPACT-4. This latter code has capabilities for path-way analysis and sensitivity studies and makes use of our extensive activation and transmutation ECNAF95 library. For the purpose of transmutation studies, this library has been extended with the higher actinides [73] and has been used to update the data libraries of the ORIGEN-S fuel depletion code [55,61].

CAPRA fast burner reactors

The French CAPRA programme (CAPRA is an acronym for "Consummation Accrué de Plutonium dans les réacteurs RApides") aims at developing a fast reactor optimized for the burning of plutonium and minor actinides. In support of this project, a program has been written to calculate pseudo fission products for five actinide isotopes (^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu). Preliminary pseudo fission products based on fission product concentrations calculated for the SNR-300 reactor have been delivered to CEA. The same program has been used to calculate lumped fission-product cross sections for the SWG17 benchmark which has been organized by a Working Party of the NEA Nuclear Science Committee. This benchmark aims at quantifying the status of fission-product cross sections (both absorption cross sections and the inelastic scattering cross section) for use in fast reactors. This work is necessary to be able to calculate the uncertainty in the burnup reactivity swing of fast reactors. The results of this benchmark are expected in 1996.

In 1996, the above-mentioned OCTOPUS system will be used to calculate new fission-product concentrations for pseudo nuclides in CAPRA fuel types. To this end, ECN will investigate the possibilities to include the ECCO cell code for fast reactors in the OCTOPUS system.

Transmutation of plutonium in thermal and fast reactors
It has been shown that a CAPRA fast reactor without uranium in the fuel can have sufficiently large negative reactivity coefficients if plutonium with a large fraction of ^{240}Pu is used. This means that the plutonium has to be recycled one or more times in a PWR before it can be transmuted in a fast reactor. For this reason and others, (multi-) recycling plutonium in PWRs remains an interesting option. Special attention has been put to the development of uranium-free matrices with a sufficiently large negative fuel temperature coefficient. In support of this work, a program named VAREX has been written to calculate the contribution of individual isotopes to the fuel temperature and the moderator density reactivity coefficients [80]. It was found that the fuel temperature coefficient of matrices containing antimony or tantalum can have a sufficiently large negative value. In 1996 the use of burnable poisons with a large negative contribution to the fuel temperature coefficient will be studied.

In the coming years, studies will be undertaken to investigate the possibilities of multi-recycling plutonium in PWR reactors. In cooperation with European partners within the EU Fourth Framework programme, it will be investigated how many times plutonium can be recycled without penalizing the safety of the reactor too much. Also the influence of enhanced moderation on the reactivity coefficients and the plutonium transmutation rate will be investigated. To investigate the influence of cross section uncertainties on the reactivity coefficients, a sensitivity study will be performed.

To familiarize with PWR core calculations and to gain experience with MOX fuel, a six month detachment of an ECN employee at Nuclear Power International (NPI) in Paris (Framatome) is underway.

Transmutation of americium
Upon irradiation of americium, curium isotopes are produced. Whether or not the curium can be partitioned and recycled determines to a large extend the scenario to be followed for the transmutation of americium. If curium cannot be recycled, it is probably best to irradiate the americium target for a very long time in a thermal high flux environment and to dispose of the irradiated target. If the curium can be partitioned and recycled again, irradiation of americium in fast reactors has preference [54,77,84,87].

It can easily be shown that the transmutation of plutonium or the minor actinides can best be done in the absence of uranium [63]. In such case inert matrices can be used for the dilution of the actinides to be irradiated. However, the development of inert matrices requires a long time and it has been proposed to use uranium-dioxide instead. The penalties accompanied by the use of uranium-dioxide as a matrix are quantified in a parametric study for the transmutation of americium [85]. When uranium-dioxide is used as a matrix, plutonium will be produced by neutron capture reactions in the ^{238}U, which contributes to the plutonium density and to the fission power density in the sample and to the radiotoxicity of the irradiated sample. The contribution of the ^{238}U to the plutonium density and the fission power density in the sample are limited to 25% for

the irradiation of americium in a PWR or in a fast reactor. When americium is irradiated in a thermal high flux reactor like the Petten HFR, the contribution of the ^{238}U to the fission power density reaches a value as high as 50% (see figure 3.1). From these results, it has been concluded that the use of inert matrices for the transmutation of americium has preference above the use of uranium-dioxide.

An introductory study has been completed which aimed at the calculation of the radial burnup dependence of an americium sample irradiated in the Petten HFR [44,48]. Further studies on transmutation of americium in the Petten HFR will be undertaken in the coming year in the framework of the Fourth Framework Programme of the EU in support of the experimental work described in Section 3.5.

Transmutation of long-lived fission products

The work related to transmutation of long-lived fission products performed within the Fourth Community Research Programme on the Management and Disposal of Radioactive Waste has been finalized with several ECN reports [41,61,62,73] and a common report of ECN and Belgo-Nucléaire [84,87]. The fruitful cooperation between these two companies and the other partners of the research programme will be continued within the Fourth Framework Programme of the EU. The conclusions of the present study are that the transmutation rates of Tc-99 in present-day reactor systems are only marginal. The most suitable reactor types are the HWR reactor (CANDU) and the fast reactor [62,84].

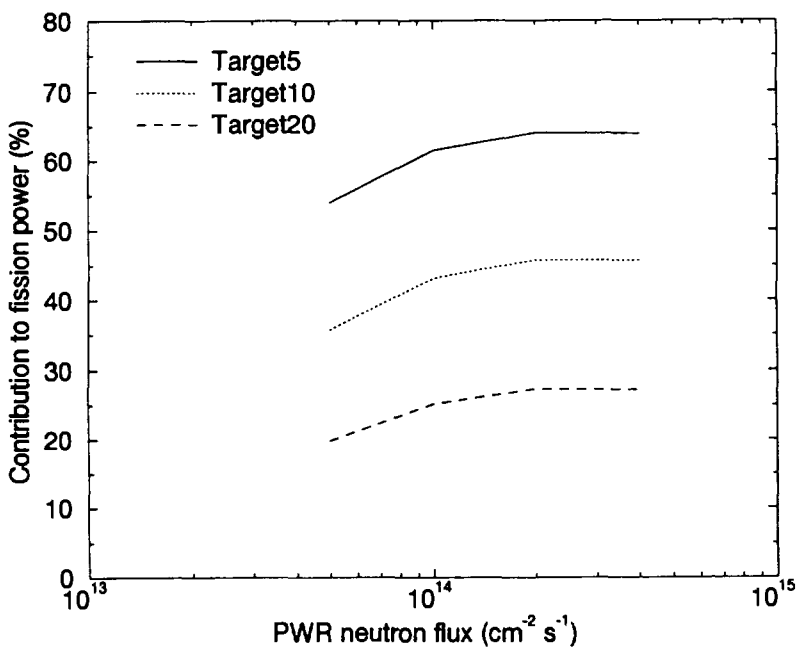

Figure 3.1 *Contribution of the uranium isotopes to the fission power density in a target made AmO_2 mixed in depleted uranium after an irradiation time of 1000 days in a PWR as a function of the neutron flux.*
Target 5 contains 5 wt.% Am, target 10 contains 10 wt.% Am, target 20 contains 20 wt.% Am.

Three-dimensional Monte Carlo calculations have been performed to calculate accurately the transmutation rate of the Tc-99 samples which have been irradiated in the Petten HFR for eight cycles (RAS1 experiment, see section 3.5). The difference between the measured and calculated transmutation rates is less than 10%. However, the measured radial dependence of the transmutation rate cannot be reproduced, which may indicate uncertainties in the Tc-99 resonance parameters.

Transmutation in CANDU reactors

An introductory study on the transmutation of actinides in four reactor types [77] has shown that the transmutation of actinides in CANDU reactors is an interesting option which deserves more attention. Important features are: the high flux, the very thermal spectrum, the good neutron economy and the on-line refuelling capability. From the three considered thermal reactors the CANDU reactor has the best transmutation features and it also competes with fast reactors in Pu-burning. A drawback of thermal reactors as compared to fast reactors is that transmutation of most higher actinides requires more neutrons and that relatively more curium isotopes are produced. The first point is not too serious in a CANDU burner reactor and the last point is mainly a concern if reprocessing is necessary for a next cycle. Therefore, it was decided to direct the studies towards once-through transuranic burning in CANDU-type reactors. In this mode a very high burn-up of the actinides is the objective, while a minimum of new waste should be produced. Thus, an investigation was made to see whether a CANDU type of reactor without uranium was feasible. Instead, Pu and other actinides in an inert matrix (SiC) were used in this conceptual CANDU burner. It turned out that indeed very large Pu and minor actinide consumption is possible.

The statics and dynamics of CANDU reactors (partly) filled with plutonium or a mix of transuranic actinides has been assessed in cooperation with AECL [60]. A slightly positive local (bundle) fuel temperature coefficient is obtained during the burnup of the fuels considered. The coefficient is negative at the loading of the bundle. The void coefficient is also positive as in the case of natural U fueled CANDUs, but in general smaller. The sign of the fuel temperature coefficient is determined by the relative change in the fission rate (determined by ^{239}Pu) and in the absorption rate (determined by ^{239}Pu and parasitic absorptions in fission products and inert matrices) upon a fuel temperature change. When the fuel temperature increases, both rates become smaller. However, the parasitic absorption rate decreases relatively more due to the fact that ^{239}Pu has a large resonance just above thermal energies. So, when the parasitic absorption rate becomes more important (i.e. at higher burn-up or at lower initial ^{239}Pu loading) the fuel temperature coefficient is larger, eventually becoming positive [83]. The power increase during a loss-of-coolant accident is acceptably low [60], despite the slightly positive (local) fuel temperature coefficient, as can be seen in figure 3.2. In this figure, the power increase during a LOCA is depicted depending on void coefficient and fuel temperature coefficient. It shows clearly that the void coefficient determines the power increase, not the fuel temperature coefficient.

In conclusion, it seems that within the current CANDU safety philosophy it is possible to design a transuranic burner with a very high Pu or minor actinide transmutation rate. Further study, e.g. on the radiotoxicity reduction of such systems, is in progress [96]. In 1996/1997 an attempt will be made to see

whether negative void and fuel temperature coefficients can be obtained. In an earlier Dutch study the feasibility of obtaining a negative void coefficient was shown for uranium fueled CANDU reactors.

Transmutation in HTGRs

A Phd student started in 1995 with a study to the possibilities of High Temperature Gas-cooled Reactors for the transmutation of plutonium. This reactor has been chosen because of its flexible on-line refuelling capabilities and its high degree of safety. First results of this study are expected in the course of 1996. Cooperation with IAEA is envisaged.

Accelerator-driven systems and nuclear data

There is an ongoing effort to enlarge the insight in the use of hybrid systems for (a) incineration of long-lived waste and (b) energy production based on the thorium fuel cycle, with a minimal production of actinides (Rubbia-concept). The technical, economical and safety features of these relatively new systems are under study. ECN has made an extensive list of pros and cons [97] concentrating on safety issues [29,33] and waste reduction capabilities. It was concluded that more study is required, preferably by means of international cooperation [97]. To this end ECN will participate in 1996 in the EU 4th framework programme on the

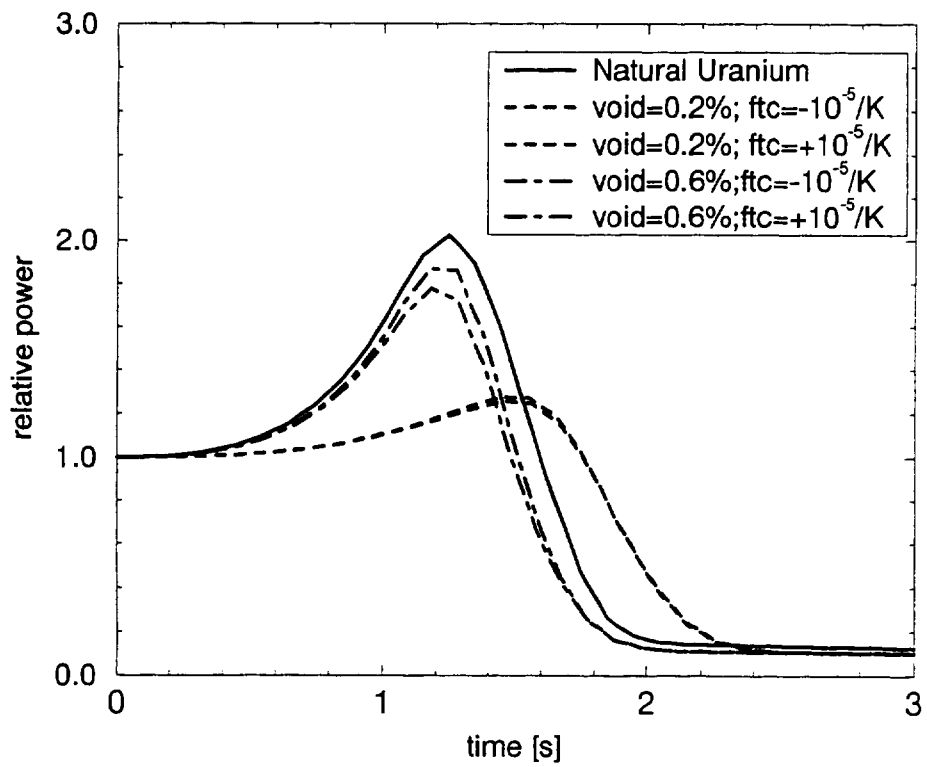

Figure 3.2 *Relative power during a loss-of-coolant accident for the natural uranium fueled CANDU, for a CANDU burner reactor with a void effect of 0.2%, and for a CANDU burner reactor with a void effect of 0.6%, both burners with two fuel temperature coefficients.*

Evaluation of the Accelerator-Based Transmutation Option, in a Coordinated Research Programme of the IAEA and possibly in a NEA benchmark. This work partially fits into the Thorium Cycle evaluation (section 3.4). The ECN theoretical studies and experimental work on transmutation of Am and long-lived fission-products (section 3.5) can also be viewed as valuable contributions to the international evaluation of the potential and accelerator-based transmutation [97]. A specific contribution from ECN to the research of accelerator-driven systems is the provision of nuclear data for hybrid-system studies. In ref. [58], the present global status of this field is outlined. Two proton nuclear data files for ^{208}Pb and ^{90}Zr up to 200 MeV have been produced. These files are meant to replace intranuclear cascade codes which are inadequate for transport calculations at these energies. They can be applied in future releases of MCNP that can handle protons. ECN has participated in the NEA/OECD Activation Yields Benchmark. With the nuclear reaction code MINGUS95, isotope production for iron, cobalt, zirconium and gold has been predicted. The results of this code-intercomparison are expected in 1996. The European Activation File EAF has been transformed to the ECN Activation File ECNAF. This data library will be used to calculate activation and the production of residual isotopes in both hybrid systems and conventional reactor-based transmutation concepts. The file is being used at CERN and Los Alamos. In collaboration with Los Alamos National Laboratory, high-energy neutron data files will be produced in 1996.

Within the Netherlands a cooperation with Groningen University (KVI) was initiated to investigate important reactions for nuclear waste transmutation experimentally, by means of the new AGOR facility. A PhD student will perform theoretical studies at ECN and experimental work at KVI, starting January 1996.

3.3. Project 3: Non-proliferation aspects

In order to provide the necessary background for the justification of investigations in this field a position paper [98] has been written. This paper reviews four important international non-proliferation studies and discusses consequences for the RAS programme.

The main conclusions about counter measures supported by the four discussed studies are:

- Technical barriers will be effective for reducing the risk of theft but will only in a limited way be effective for reducing the risk of proliferation.
- Safeguards measures are more effective than technical measures.
- Institutional measures are potentially more effective than technical measures.

It is suggested that this will have the following operational consequences for the proliferation R&D in the RAS programme:

- Focus analysis on the safeguardability of innovative fuel cycles. Safeguardability depends on how easy the flows of nuclear material can be tracked. Hence applied technologies and design of fuel cycle facilities, and type, quantities and chemical form of nuclear materials present therein, are all relevant. Note that integrated analysis of fuel cycle facilities and nuclear materials is required, since both are strongly interrelated.
- Focus technology development on the development and design of safeguards methods and techniques.

In 1995 the efforts were concentrated on safeguards methods of Pu fuel with admixtures of minor actinides (Np and Am) [72] and on the possible misuse of minor actinides for weapon material.

The application of existing Pu NDA methods to fresh MINOX fuels will encounter a series of problems [72]. These arise from the presence of curium and lanthanides in the extraction streams because of limited decontamination. The gamma-ray and neutron emission from lanthanides and curium in the fuel will deterioate the performance of some Pu Non-Destructive Analysis (NDA) methods, and will cause other methods to be useless, in particular the methods for the Pu total measurement. Internal X-Ray Fluorescence methods (XRF) may present a suitable replacement.
The MINOX fuels will not be directly accessible due to their high radiation levels. Pu NDA methods therefore will have to be remotely operated and the handling and distribution of reference materials will be hampered.

In future Partitioning & Transmutation (P & T) fuel cycles, minor actinides (MAC) will be available in various forms. A direct proliferation aspect of P & T fuel cycles may be the strategic value of the minor actinides Np, Am and Cm. During the past period the possible misuse of Cm was studied. It was found that reactor grade Cm is not possible to use as weapon material, mainly due to the heat production of ^{244}Cm.

3.4. Project 4: Thorium Cycle

The usual spent uranium fuel contains substantial amounts of transuranic elements, i.e. various plutonium, americium, neptunium and curium isotopes, which treatment is subject to waste strategy studies, such as RAS, for reduction of the long-term radiotoxicity by advanced P&T techniques. Thorium cycles may offer a complementary approach to handle this actinide waste problem. If thorium can act as fertile fuel, the use of U-238 can (partly) be avoided, thus reducing the transuranics production considerably. On the other hand some non-natural uranium and protactinium isotopes, typical for spent thorium fuel, will be produced and may contribute to the long-term radiotoxicity of the waste.
In order to assess the specific perspectives and drawbacks of thorium fuel cycles as waste management option an ECN programme has been started in 1994 and running till 1998 with the following activities:

A: A review study, assessing the general aspects of various types of thorium fuels and fuel cycles,
B: Reactor specific studies:
   1. CANDU-Thorium in collaboration with AECL
   2. HTR-Thorium and Accelerator-Based Thorium Fuelled Systems
   3. Participation in a European assessment on thorium fuels in LWR and FBR.

Activities A and B1 have been terminated in 1995, B2 is scheduled for 1996 and the European study B3 will run for the period 1996-1998.

3.4.1 Review study

The perspectives and drawbacks of thorium cycle options have been analysed with special attention to the nuclear waste reduction, both reduced actinide

production and enhanced burning capability. This study included reviews on the basic reactor physics characteristics, the fuel-cycle technology. Also a generic computational analysis on long-term radiotoxicity of the waste of thorium fuel in comparison to uranium fuel is given. The results are reported in detail in the ECN reports [64-66, 82]. The conclusions are summarized below.

Basic reactor physics characteristic:
The evaluation of reactor physics characteristics [64] included a review of the basic nuclear data of the fissile and the fertile components and an analysis of steady state behaviour and dynamic characteristics of reactor cores of a typical LWR and HWR with different fuel types, including equilibrium thorium/uranium fuel, once-through uranium fuel and equilibrium uranium/plutonium fuel.
It was concluded that the use of thorium instead of uranium fuel will not have a serious impact on the reactor physics safety characteristics of present day LWR and HWR, although minor adaptations may be required to handle typical aspects such as protactinium holdup and decreased fraction of delayed neutrons.

Fuel cycle technology
The chemical aspects of the thorium fuel cycle [82] have been assessed with respect to the various cycle processes: thorium resources and ore processing, fresh-fuel fabrication, irradiation performance of thorium based fuel elements, fuel reprocessing (THOREX process), fuel refabrication and waste management. It was concluded that most of the chemical aspects, from mining to reprocessing, have been studied in the past, technical solutions for all cycle steps have been indicated and sometimes even tested on pilot scale. The technologies have not been developed to large-scale, commercial applications. Due to the very hard gamma radiation, the handling of reprocessed uranium and thorium will require severe shielding and remote control operation for all process steps. The glass volume of wastes from the THOREX process is estimated to be considerably larger than of the wastes from a comparable PUREX process.

Actinide waste
The production of actinide waste [65] has been studied for various thorium cycles in comparison to the once-through uranium cycle. The evaluation is based on burnup calculations for the case of a HWR because of its good neutron economy, thus enabling better conversion ratios, better utilization of the thorium itself, and reduced requirement of additional fissile makeup fuel. The once-through natural uranium fuel cycle, standard for present CANDU operation, has been used as reference cycle.

Once-through fuel modes with fresh fuel consisting of thorium and additive makeup fuels of high enriched uranium (HEU) or plutonium have been considered in the burnup calculation. In this case all actinides in the discharged fuel are considered to be waste. The total actinide radiotoxicity of this waste is presented in figure 3.3. In comparison with natural uranium fuel, the thorium/HEU system shows a significant reduction of the radiotoxicity, at least for a period of 10^4 years (up to one order of magnitude reduction). After that period the radiotoxicity will even exceed the natural uranium case, mainly due to decay of ^{233}U and, to a minor extent, of ^{231}Pa. It is noted that the radiotoxicity of mining waste and of U tails from HEU enrichment have not been included in this figure. The horizontal line represents the radiotoxicity of the uranium ore.

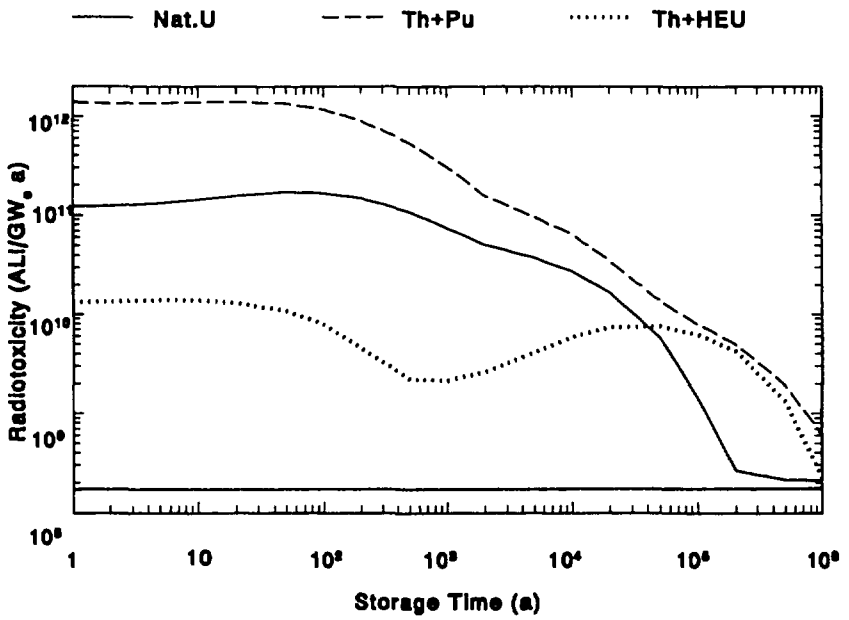

Figure 3.3 *Radiotoxicity of actinide waste of HWR once-through systems, exlucing mining waste and U-tails from enrichment. The horizontal line represents the radiotoxicity of the uranium ore.*

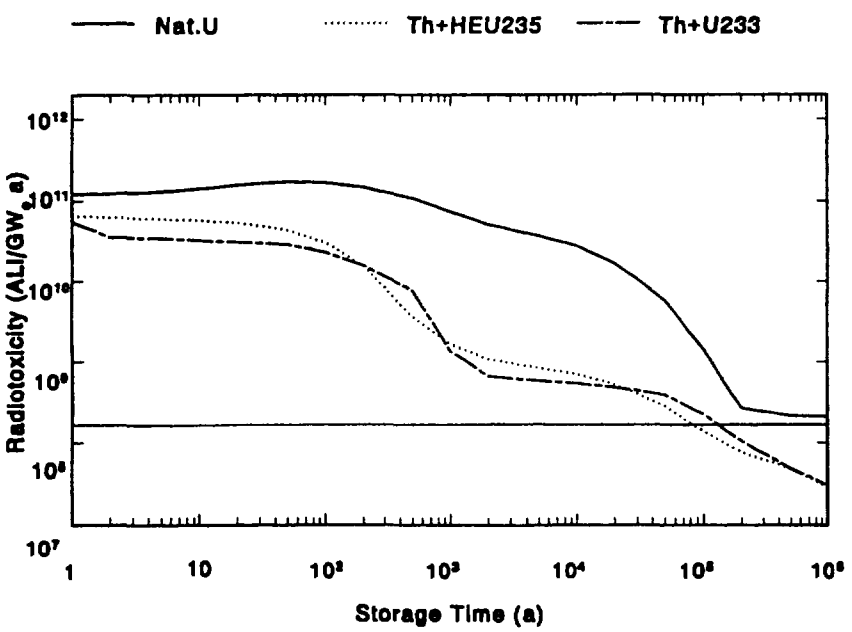

Figure 3.4 *Radiotoxicity of actinide waste of HWR cycles with U-recycling, excluding mining and U-tails from enrichment. The horizontal line represents the radiotoxicity of the uranium ore.*

For the thorium/plutonium cycles the radiotoxicity of the waste exceeds the value of the natural uranium case during the full storage period. The plutonium, fed into the fresh thorium fuel, represents, however, a certain amount of radiotoxicity that will be mostly destroyed during the reactor operation. This thorium/plutonium cycle is most efficient for net plutonium burning; a consumption of about 900 kg/GWe.a can be realized, much higher than achievable in present LWRs with MOX fuel loaded in 1/3 of the core.

Closed cycle operation was considered with recycling of the uranium. Only highly enriched uranium is considered as makeup fuel, in order to avoid the cycle to be transferred to a standard uranium cycle by multiple recycling.
As shown in figure 3.4 a reduction of the radiotoxicity can be achieved by a factor of 100 (compared to once-through natural uranium), at least for the storage period from 10^3 to 10^5 years. ^{231}Pa, originating from a (n,2n) reaction with thorium, dictates the threshold value for the long-term radiotoxicity. The horizontal line represents the radiotoxicity of the uranium ore.

In conclusion, the major advantages of thorium fuel cycles seem to be: a reduction of the volume of the actinide waste, a possible reduction of actinide radiotoxicity by a factor of about 50 to 100 and an improved utilization of uranium resources under the condition that efficient partitioning and multiple recycling of uranium and thorium would be feasible. This has still to be proven for large-scale reactor operation. It is noted that to complete this picture the radiotoxicity of mining residuals and of enrichment U-tails has to be taken into account. Moreover, a comparison with the LWR once-through cycle should be made.

3.4.2 Reactor specific studies

CANDU
ECN has performed, in collaboration with AECL, specific analyses on physics characteristics of the CANDU/Thorium combination, cf. figures 3.3 and 3.4 [65, 91]. Three items were treated in detail:
- intercomparison of reactivity calculations at AECL and at ECN,
- acquirement of the specific techniques for CANDU fuel management and its application for thorium based fuel and for actinide burning,
- suitability of thorium fuel in CANDU-type reactors and the strategy for switch-over from an uranium to a thorium-fuelled core.

HTR
A first assessment of thorium fuelled High-Temperature Gas-cooled Reactors was made by Bultman [75], using results obtained in the CANDU study mentioned above [65]. A PhD study has been started on the subject of High Temperature Gas-cooled Reactors with advanced fuel types. Thorium-based fuels will be one of the candidates to be investigated. This work is scheduled for 1996-1997.

Accelerator-Driven Reactors
Accelerators could provide additional neutrons without the necessity of using makeup fuel, avoiding waste from this additional fuel and avoiding possible non-proliferation problems from this fuel. This offers the possibility to have radiotoxicity curves as given in fig. 3.4. Even better performance is predicted for the "Fast Energy Amplifier" concept of CERN by the enhanced recycling of all actinides. A review of these possibilities is scheduled for 1996 in cooperation with

a Coordinated Research Programme of the IAEA. Also in the framework of EU studies (see below) information on this subject will be examined.

European Collaboration
Supported as part of the 4th Framework Programme of the European Community an European assessment on thorium cycles will be made. The project, named "Thorium cycles as nuclear waste management option", is supported by seven European partners: KFA (Germany), BN (Belgium), CEA and IN2P3 (France), ENEA (Italy), ITU (EC) and ECN (coordinator). The study will be focused on the role of LWR and FBR and, to a less extent also accelerator-driven systems, with thorium fuel as a contribution to reduce long-lived nuclear waste. For the study of accelerator-driven systems a second EU programme has been defined, also with ECN participation. The project will start in 1996 and will be terminated in 1998.

3.5. Project 5: Experimental verification of transmutation

In 1995 the fabrication of the irradiation facilities has been finished and the irradiation programme in the HFR has been defined in further detail [78]. According to the present plans, the programme now consist of four irradiation experiments of which one (RAS-1) has been completed in January 1995. These experiments are part of the international EFTTRA cooperation [53], which looks into methods for the transmutation of the fission products technetium and iodine as well as the minor actinide Americium.

The RAS-1 experiment was prepared in 1993 and the irradiation has been performed in 1994. Nine targets have been irradiated in a REFA rig during 8 reactor cycles, which corresponds to 192.95 full power days. The three technetium samples each contained two casted rods of Tc-99 metal, 25 mm long and 4.8 mm in diameter. These samples were prepared at the Institute for Transuranium Elements (ITU) in Karlsruhe. The six iodine (I-127) samples contained three different metal iodide powders: cerium triiodide (CeI_3), sodium iodide (NaI) and lead iodide (PbI_2), all in duplicate. All samples were contained in 15-15 Ti stainless steel cladding.

The results of the post-irradiation examination have become available in the course of 1995 and a preliminary report has been prepared for the GLOBAL'95 meeting [53]. The diameter of the technetium samples was measured to check whether swelling of the metal had occurred. The results are summarized in Table 3.1 and show that a small but insignificant increase of the dimensions has been found. The transmutation rate has also been determined by measuring the Ru-content of the irradiated rods. This has been done by two independent methods. Electron probe miroanalysis (EPMA) showed that the Ru-content varied from 6.1 to 6.5 % in the center of the rods, depending upon the radial position in the rod, Isotope Dilution Mass Spectrometry (IDMS) gave (6.4 $\pm$ 0.2) % for the mean transmutation rate. The EPMA analysis also showed a strong increase of the transmutation rate in the outer 50 μm of the rod, where the Ru-concentration was as high as 17.5 % (figure 3.5). This effect is due to the contribution of shelf-shielding of epithermal neutron capture.
With various techniques such as visual inspection, X-ray photographs, micrography and gas-puncturing it was shown that the cladding of the two

capsules containing lead iodide had failed during the irradiation. A reaction has taken place between the salt, which was molten at the temperature conditions of the experiment, and the stainless steel cladding. The samples containing erium iodide and sodium iodide were still intact and the transmutation rate has been measured by mass spetrometric analysis of the plenum gas. For the two NaI samples we found 5.14 and 5.12 % and for the two CeI_3 5.86 and 5.87 %.

Table 3.1. *Diameter measurements of the irradiated technetium rods*

sample	diameter (mm)		length (mm)	
	pre-test	post-test	pre-test	post-test
A	4.80 ±0.01	4.83 ± 0.01	25.05	25.09
B	4.81 ± 0.02	4.84 ± 0.01	25.05	25.12
D	4.81 ± 0.02	4.83 ± 0.03		

In addition to the examination of the targets, analysis of the gamma-scan wires [71] and flux-monitors [93] have been performed.

The RAS-2 experiment was prepared in 1995 and the irradiation will start in 1996 (HFR cycle 2). The experiment comprises two legs of a TRIO irradiation facility, each containing identical sample holder. The samples in RAS-2, which will be irradiated during 4 HFR cycles, contain four different inert matrices which have been selected as diluent for heterogeneous americium fuels: spinel ($MgAl_2O_4$), yttrium aluminum garnet (YAG = $Y_3Al_5O_{12}$) and alumina (Al_2O_3) in a Zircaloy cladding and cerium dioxide (CeO_2) in a stainless steel cladding. The goal of this experiment is the investigation of radiation damage in these materials due to neutrons. The RAS2bis experiment, which will be irradiated for about 22 cycles, contains identical samples of spinel, YAG and alumina and, in addition, a technetium sample. The latter sample is made of two rods that have been irradiated in the RAS-1 experiment which have been refabricated into stainless steel cladding. The goal of the re-irradiation of this Tc-sample is the investigation of the effect of a high burnup (20-25 %) on the material properties.

The RAS-3 experiment has been defined together with the EFTTRA partners [53]. The goal of this experiment is also the investigation of radiation damage in ceramic inert matrix materials but not only due to the impact of neutrons but also of fission product recoil. The effect of fission product recoil on the stability of the materials, which is thought to be much larger than that of neutrons, will be obtained by inclusion of a small quantity of 20% enriched uraniumoxide. In the present proposal 16 samples will be irradiated, consisting of pairs of 8 materials with and without UO_2. The test matrix is shown in Table 3.1.

The RAS-4 experiment is a joint proposal by the EFTTRA group for the 4th Framework Programme of the EU, to demonstrate the feasibility of the transmutation of americium in an inert matrix. This proposal has been granted by the EU and the work will begin in 1996.

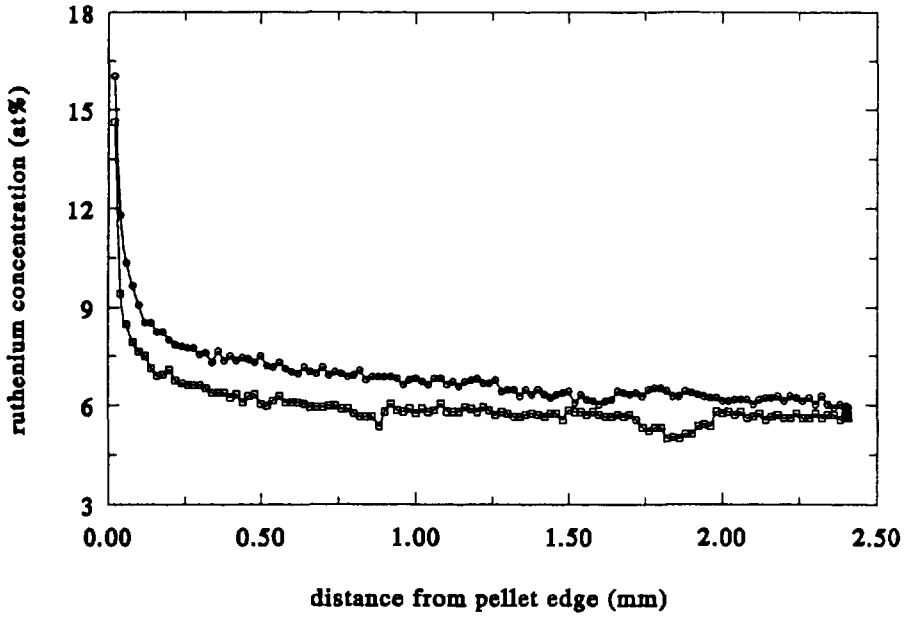

Figure 3.5. *The radial distribution of ruthenium in two sections of the irradiated technetium target.*

3.6. Chemical research on actinides and fission products

In 1995 the investigation of inert matrices for the incineration of actinides has been the central theme of this project. This work is performed within the frame of the international cooperations EFTTRA and CAPRA. Experiments with plutonium and/or minor actinide compounds could still not be performed since the construction of the actinide laboratorium was only started in the last quarter of 1995 due to delay in licensing.

In the EFTTRA cooperation, the possibilities of the transmutation of americium is being investigated. For the fuel design mixtures of americium oxide and ceramic or metallic materials, the inert matrix, are being considered. Most emphasis is placed on oxidic materials such as spinel ($MgAl_2O_4$), YAG ($Y_3Al_5O_{12}$), ceria (CeO_2) and magnesium oxide (MgO). It is important that these inert matrices have good thermal properties, are stable in strong radiation fields and are compatible with existing reprocessing processes. However, this information is not available for all materials that have been proposed by various institutes, and experiments are being performed to obtain sufficient information for a final selection.

For the RAS-2 irradiation experiment in the HFR (see project 5) the preparation and characterization of pellets of spinel, YAG, ceria and alumina have been undertaken. For all materials methods have been developed to obtain pellets with a density greater than 90 % of the theoretical one.

The preparation and characterization of an additional candidate material, cerium phosphate ($CePO_4$), has been undertaken. Cerium phosphate is an analogue of the mineral monazite, which is well known for its radiation resistance and its chemical stability. The material has been prepared succesfully from a cerium sulphate solution and high density pellets have been fabricated. In addition the following experiments have been performed:

- dissolution test in hot nitric acid,
- thermal expansion measurements,
- heat capacity measurements.

The results of these experiments show that this material has promising properties, but information on the thermal conductivity and verification of the radiation resistance is necessary.

For the study of the chemical behaviour of the inert matrices at high burn-up, a method have been developed to study the interaction between fission products and the matrix by means of simulated fuels (SIMFUELS). Such fuels are made of a representative mixture of the non-active oxides that represent the non-volatile fission products and the fuel. This mixture is ground, spray dried and sintered, followed by a thorough analysis by means of scanning electron microscopy (SEM) and chemical analysis (WDX). The method for preparing SIMFUELS has been tested on samples for high burn-up fuels based on spinel, YAG, alumina and cerium phosphate. These experiments are being supported by thermochemical calculations. The results of this approach show that interaction between matrix and the lanthanides, the most abundant group of fission products, plays an important role.

To improve the thermochemical calculations, critical evaluations of the thermodynamic properties of a number actinide and lanthanide compounds (halides, oxides) [56], and thermodynamic measurements have been made. To assist the interpretation of the RAS-1 irradiation experiment, a number of thermodynamic measurements on CeI_3 and PbI_2 have been made [88-90]. Special attention was given to the phase diagrams Pb-PbI_2 and Ce-CeI_3, since metal/iodine ratio decreases with inceasing burn-up. As an example, the Pb-PbI_2 diagram is shown in figure 3.6.

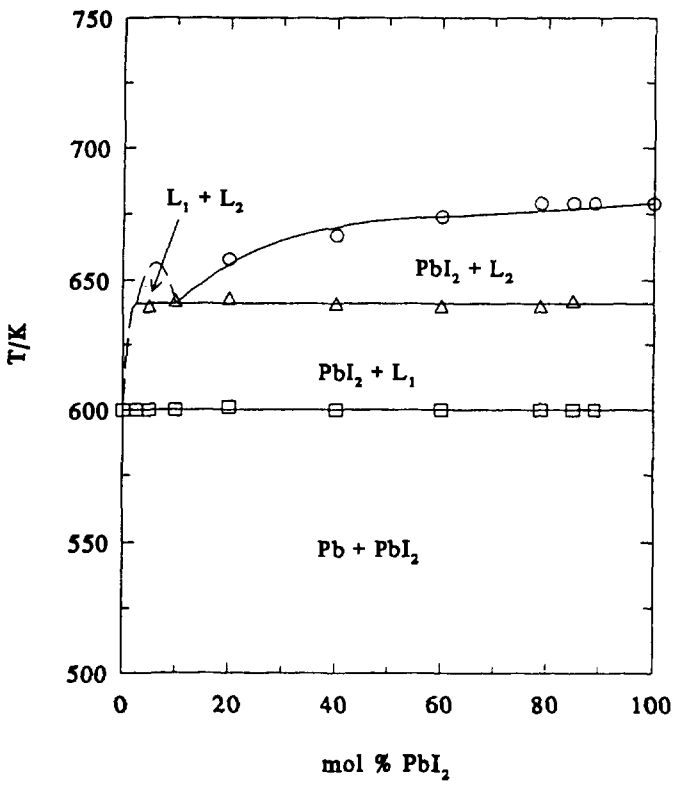

Figure 3.6. *The Pb-PbI_2 phase diagram as determined by DSC measurements.*

In the frame of the collaboration with CEA on CAPRA, the preparation of a ceramic nitride inert matrix for burning of plutonium (as PuN) in a Pu-without-U ore has been investigated. Nitride fuels are being considered since MOX-fuels with high Pu-content, which are considered for CAPRA, are not compatible with the PUREX reprocessing technology. It is therefore very important to find an inert nitride matrix that, like PuN, dissolves in hot HNO_3. Cerium nitride has been suggested as a possibility. Two different routes have been tested for the preparation of this compound. The preparation via reaction of cerium metal with nitrogen proved to be succesful. Dissolution tests showed that CeN indeed dissolves in nitric acid, the solution medium of the PUREX process. Further characterization of the material has been undertaken of which the results will become available in 1996.

In addition to the above, a Ph-D study on the separation of trivalent actinides and lanthanides from high level waste by means of selective liquid membranes (SLM) has been started. This work is being performed in close cooperation with the University of Twente. In the first stage of this study, the synthesis of functionalised marocylic compounds that can be used as carrier in a SLM, is being investigated. The first results have been presented at the GLOBAL'95 symposium.

3.7. Project 7: Radiological Impact associated with P&T

In 1995 work was carried out on two studies in the framework of the RAS Radiological Impact project: a study of the methodology to be used when assessing the radiological impact of nuclear fuel cycles and; a general, qualitative assessment of the radiological impact of a number of nuclear fuel cycles incorporating P&T technologies. The results of these two studies are described below.

Assessment Methodology
The objective of this study was to develop a methodology to assess the radiological risks associated with the P&T technologies being studied in the RAS research programme. The assessment methodology study comprised of three parts and the results of this study are presented in [86]. An overview of these results is given below.

Firstly, the nature of the radiological impact associated with (P&T) nuclear fuel cycles was analysed. This was done by considering the processes used, the inputs and outputs, the important waste streams and the locations where the activity takes place for each stage in the existing nuclear fuel cycle. Potential modifications to the existing fuel cycle following the adoption of one of the main research elements in the RAS programme were then briefly considered. It was concluded that the radiological impact associated with nuclear fuel cycles is complex in nature and that the radiotoxicity concept is not a satisfactory measure of this impact.

Secondly, a selection was made of measures which can be used to quantify the radiological impact associated with nuclear fuel cycles. The basic framework for judging the radiological impact was defined in terms of ethical considerations, radiation protection guidelines and social considerations. A number of existing

radiological impact measures were then defined: basic dose quantities; individual dose and risk; group risk; collective dose and risk; and measures based on comparisons with other risks. These existing measures were then compared against criteria derived from the basic framework for the selection of radiological impact measures: acceptability, practicality and completeness. It was concluded that the existing set of radiological impact measures do not fully meet these criteria. In particular they do not give sufficient information related to the qualitative attributes of the impacts. However, a limited picture of the radiological impact can be obtained using measures of collective dose integrated over a number of population groups and integration times. Research is being performed at ECN into the development of additional radiological impact measures. In the near future however, the evaluation of the radiological impact associated with P&T fuel cycles will be based on measures of collective dose.

Thirdly, the methodology for quantifying the radiological impact associated with nuclear fuel cycles was analysed. Each stage in a nuclear fuel cycle can be treated as a "module" and each module can be defined in terms of the characteristics which are relevant to the assessment of the radiological risks associated with it (i.e. release data and location). Modules can then be combined to form nuclear fuel cycles. The radiological consequences associated with the normal and accidental releases from each module can be analysed using standard radiological consequence assessment methodologies.

Global Assessment

The global assessment study [94] was initiated in October 1995 in response to recommendations received from the RAS review committee. The objective of this study is to assess the radiological impact associated with fuel cycles that incorporate technologies for the P&T of the long-lived radionuclides present in high-level radioactive waste. Seven nuclear fuel cycles are being considered: two once through fuel cycles (i.e. with and without reprocessing) and five 'advanced' fuel cycles (based upon the fuel cycles defined in the recent fuel cycle study performed by CEA[1] for the Commission of the European Union). The radiological impact associated with the two once through cycles has been assessed in terms of collective dose. Attention was given to the breakdown of this dose over the regional and global populations and over the present and future generations. The impact associated with the five 'advanced' fuel cycles is being assessed against these two cycles. Currently, only a limited amount of information on these 'advanced' fuel cycles is available. The assessment of the radiological impact associated with these cycles is therefore by necessity of a general nature. This study shall be completed in the first half of 1996 and the results published in an ECN report. A short overview of the preliminary results of this study are given below.

The radiological impact (in terms of collective dose) incurred over the period to several 100,000 years associated with the normal operation of installations in the 'once through' nuclear fuel cycles is dominated by the mining, electricity generation and reprocessing (for the 'reprocessing without recycling' option) stages. Evolutionary light water reactor (LWR) cycles (i.e. recycling of plutonium and minor actinides) do not appear to offer major radiological impact gains at

[1] H. Boussier: *Potentiality and Cost of Partitioning and Transmuting Long-Lived Radionuclides*, CEA Report Number NT/SSP/94/05, July 1994.

either the front end or the back end of the fuel cycle. At most a reduction of 25% in uranium ore requirements (with respect to the 'once through' cycles) can be achieved. It is unlikely that any major changes in the radiological impact from the wastes to be disposed of could be achieved with these fuel cycles. In addition, it is unlikely that the radiological impact associated with the electricity generating stage will be significantly different for these cycles.

The recycling of plutonium in fast reactors (FBRs) implies that a significant reduction in the amount of uranium ore required can be achieved once the cycle has reached an 'equilibrium' situation. This can be assumed to have a proportional effect upon the radiological impact associated with the front end of the fuel cycle. Additional reprocessing of the minor actinides in FBRs implies that significant reductions in the actinide inventory over the relative near future (i.e. isotopes of plutonium and americium) and the far future (i.e. neptunium) can be achieved. The most important impact of this reduction likely to be on the radiological impact associated with certain altered evolution scenarios for waste repositories.

The normalised radiological impact incurred over the first few 100,000 years associated with fission product recycling is likely to be similar to that associated with an identical fuel cycle without fission product recycling. Fission product and minor actinide recycling in FBRs will lead to a significant reduction in the inventory of some of the radionuclides which contribute to the long term radiological impact. However, several performance assessments for radioactive waste repositories show that the radionuclide ^{135}Cs makes the single most important contribution to the long term radiological impact associated with the repository. The inventory of this radionuclide is unaffected by the fission product recycling fuel cycle considered.

4. CONCLUDING REMARKS

The progress of the RAS programme at ECN has been described in the foregoing chapters. An important phase in the study of transmutation, the participation in a European Strategy Study, has been completed. A rather complete picture of the transmutation possibilities of LWRs and fast reactors was obtained. At ECN this resulted in the decision to continue to study the 100% MOX option and to participate in the CAPRA programme.

In 1995 special attention was given to the evaluation of transmutation options of HWRs (CANDU type of reactors). The potential of this reactor type for transmutation was found to be much larger than of LWRs.

The perspective of accelerator-based systems for radiological clean and safe energy production and transmutation of nuclear waste remains to be further examined. ECN gave important technical contributions that allow further participation in international evaluation studies to be entered in 1996.

Following the 1994 recommendations of the RAS review committee, the transmutation of actinides in inert matrices has become a central theme in the programme. A large effort in this field is made by the definition of a series of irradiations in the HFR that will continue up to 1999. This work is strongly supported by reactor physics calculations and fuel chemistry studies. The work on the transmutation of fission products has almost been completed with the analysis of the RAS-1 irradiation experiment for long-lived fission product transmutation.

Also, the advices of the RAS review committee with respect to radiological impacts and proliferation risks of P&T have been followed. A global assessment of the radiological impacts of P&T was started, and NDA safeguards methods for actinide fuels are being evaluated.

The work on thorium will be continued in the coming years, though the work will be narrowed down to the analysis of HTRs and accelerator-based systems.

In general, it can be concluded that the work has become an important input to the European programmes in this field, such as EFTTRA and CAPRA, and the studies of the 4th Framework Programme of the European Union.

Final conclusions with respect to strategies to be followed will be given in a forthcoming report [95], which will summarize the status of the international R&D on transmutation, Ref. [95] will also contain recommendations for further R&D in the framework of the RAS programme as well as preliminary observations with respect to a possible future waste management strategy, including P&T options.

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6. ACRONYMS

AEA	Atomic Energy Authority
AECL	Atomic Energy of Canada Limited
ALMR	Advanced Liquid Metal Reactor
BN	Belgo Nucléaire
CANDU	CANadian Deuterium Uranium reactor
CAPRA	Consommation Acrué de Plutonium dans réactuers RApide
CEA	Commissariat a l'Energie Atomique
CERN	Centre European Recherche Nucleaire
COVRA	Centrale Organisatie Voor Radioactief Afval (Central Organisation for Radioactive Waste in the Netherlands)
DIAMEX	DIAMide EXtraction
EAF	European Activation File
EC	European Community
ECN	Energieonderzoek Centrum Nederland (Netherlands Energy Research Foundation ECN)
EdF	Electricité de France
EFTTRA	Experimental Feasibility of Targets for TRAnsmutation, joint venture for experimental research with the following partners: CEA, ECN, EdF, KfK, IAM, ITU
EU	European Union
FBR	Fast Breeder Reactor
FR	Fast Reactor
FZK	ForschungsZentrum Karlsruhe
GE	General Electric
GKN	Gemeenschappelijke Kernenergiecentrale Nederland
GWe	
HEU	High Enriched Uranium
HFR	High Flux Reactor
HTGR	High Temperature Gas-cooled Reactor
HTR	High Temperature Reactor
IAEA	International Atomic Energy Agency
IAM	Institute for Advanced Materials (CEC, Petten)
IFR	Integral Fast Reactor
INFCE	International Nuclear Fuel Cycle Evaluation
IRI	Interfacultair Reactor Instituut (Interfaculty Nuclear Reactor Institute, Delft, the Netherlands)
ITU	Institute for Transuranic Elements (CEC, Karlsruhe)
JAERI	Japan Atomic Energy Research Institute
JEF	Joint Evaluated File
KEMA	NV tot Keuring van Elektrotechnische Materialen
KFA	KernForschungsAnlage (Jülich)
KfK	Kernforschungszentrum Karlsruhe (new name: FZK)
KVI	Kernfysisch Versneller Instituut (Nuclear Physics Accelerator Institute, Groningen, the Netherlands)
LSO	Laboratorium voor Sterk-radioactieve Objecten
LWR	Light-Water Reactor
MINOX	Mixed Oxide with 'minor' actinides
MOX	Mixed Oxide (U and Pu oxide mixture)
MWd	Megawatt-days
NEA	Nuclear Energy Agency of OECD
OECD	Organisation for Economic Cooperation and Development, Paris
OMEGA	Options for Making Extra Gains from Actinides, Japanese project, supported by NEA
P & T	Partitioning and Transmutation
Phénix	French fast research reactor with liquid sodium as a coolant
PRISM	American design for a fast liquid sodium-cooled reactor ALMR)

PSA	Probabilistic Safety Analysis
PUREX	Plutonium Uranium Refining by EXtraction
PWR	Pressurised Water Reactor
RAS	Integraal Programma voor Recycling van Actiniden en Splijtings-produkten (Integral programme for recycling and transmutation of actinides and fission products)
R & D	Research and Development
SLM	Supported Liquid Membrane technique
THOREX	THOrium EXtraction
XRF	X-Ray Fluorescence