Status report on actinide and fission product transmutation studies
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The management of radioactive waste is one of the key issues in today's political and public discussions on nuclear energy. In each country with nuclear power, the decision as to which strategy one should pursue in a national programme depends on a wide range of economic, political and historical considerations, but all have in common that the safety of the population and radiation exposure to humans is in accordance with radiation protection principles accepted worldwide. Public acceptance of nuclear power generation may be more easily obtained if the isolation time required for decay of most of the radionuclides in high level waste is reduced to 200-300 years.

One of the fields that looks into the future possibilities of nuclear technology is the neutronic transmutation of actinides and of some most important fission products. Rather than waiting for their radioactive decay it is in principle possible to reduce the period of toxicity of these isotopes by transforming them into short lived or stable nuclides in fission reactors or in accelerator-driven subcritical facilities.

This is a rather new field of activity for the IAEA. The Specialists Meeting on Use of Fast Reactors for Actinide Transmutation held in Obninsk, Russian Federation, 22-24 September 1992, may be considered a starting event. The Technical Committee Meeting in Vienna on Safety and Environmental Aspects of Partitioning and Transmutation of Actinides and Fission Products, 29 November - 2 December 1993, followed. A Special Scientific Programme on the Use of High Energy Accelerators for Transmutation of Actinides and Power Production in conjunction with the 38th IAEA General Conference was held in Vienna on 21 September 1994. More than one hundred participants attended the meeting.

Nowadays more and more studies are carried out on transmutation of actinides in various countries and at an international level. OECD/NEA has initiated recently a P&T Systems Study and it is expected that the results of the study will be published within the next two years. Since there are R&D activities in this field outside the OECD, the IAEA has taken the initiative to document the research activities on transmutation of actinides and fission products in those Member States. The Technical Committee Meeting on Feasibility of Transmutation of Actinides in Advanced Reactors, held in Vienna from 5 to 7 December 1994, has recommended the preparation of the status report on transmutation of actinides and fission products studies in non-OECD countries and provided an input for the preparation of the report.

The status report was prepared by a Technical Committee meeting held in September 1995. The aim of the report is to present an up-to-date general overview of current and planned research on transmutation in non-OECD countries, thus fostering bilateral and multilateral co-operation of interested Member States.

The IAEA would like to thank all those individuals who participated in the Technical Committee meetings and those who provided written contributions from the various countries. Special thanks are due to N. Rabotnov from the Russian Federation, who compiled the input from the experts and to I. Gibson from the United Kingdom, who reviewed and edited the final draft.
EDITORIAL NOTE

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1. INTRODUCTION

Management of radioactive waste (RW) in an environmentally safe manner is an important issue being addressed by all the countries developing a nuclear industry. In many countries it has become a serious political issue attracting intense critical attention of the general public. Therefore, working out of a safe acceptable solution is a technological challenge to international and national nuclear communities. Nuclear spent fuel (NSF) which is the main source of RW contains fissionable isotopes and therefore the issues connected with NSF handling are also proliferation sensitive.

Large scale spent fuel reprocessing has been carried out since the middle fifties by several countries. Countries such as the UK, France, Germany, Japan, India and the Russian Federation have been progressively stepping up their capabilities for spent fuel reprocessing. These countries, after carefully reviewing the various options have recognized that reprocessing of the fuel is the safer way to get rid of the problems associated with the long term storage and disposal of the spent fuel waste, at the same time ensuring augmented energy from a given initial fuel inventory. Several other countries however are storing the spent fuel for the time being and are yet to decide on the final option; the necessity for the decision becomes more urgent as time goes on and spent nuclear fuel continues accumulating on-site and in interim stores, some of them already operating above initially planned capacity.

Geological disposal is considered as the unavoidable final step in any scheme of RW management. But important questions which remain to be answered are:

- Whether the RW, which is usually considered dangerous, can be considered potentially or even presently a valuable commodity?
- What actions if any might be undertaken to turn the RW from nuisance into an asset, into secondary raw material, as it is routinely done in other industries?

Neutron transmutation of long lived radioactive minor actinides (MA - neptunium, americium, curium) by the fission process, producing energy and simultaneously turning them into shorter lived nuclides, is being intensely analysed and discussed as a possible answer to these questions. In the same way, neutron transmutation of selected long lived fission products (LLFP) is being proposed.

The concept of a closed nuclear fuel cycle (NFC) was traditionally considered as transmutation (burning) of only plutonium and recycled uranium, with minor actinides destined for final geological disposal. Now, a new understanding is emerging:

(a) In a long term case (multiple recycling and long total cooling times) plutonium and minor actinides are interconnected and mixed by decays ($^{241}$Pu into $^{241}$Am, $^{242}$Cm into $^{238}$Pu, $^{244}$Cm into $^{240}$Pu, etc.).
(b) Plutonium isotopes with small fission to capture ratios are close in their physical properties to minor actinides and degradation of plutonium isotopic composition in multiple recycling is in many ways similar to the admixture of minor actinides (neptunium americium and curium) both in neutronic properties and in the resulting difficulties in fabrication and handling of secondary fuel. Hence the experience gained in MOX fuel fabrication should be useful in the fabrication of fuel with minor actinides.
(c) Production of MA goes up in multiple recycling of plutonium while the mass of plutonium decreases.
(d) Reduction of actinide components would ease requirements for final repositories and make them relatively less expensive.

All this is attracting growing attention to the prospects of neutron transmutation of minor actinides and, also, of some selected fission products (FP). The proposed schemes include burning in advanced reactors, both thermal and fast and in accelerator driven subcritical facilities.
Although initially few nuclides (transuranium elements and $^{99}$Tc) have been identified as potential candidates for transmutation, additional ones are under consideration [1]. The extent to which they can be transmuted, is still an open question. Practically any isotopic mixture of minor actinides placed in an intense neutron flux for some time becomes highly radioactive. The period to cool it down to less than its initial radiotoxicity is one of the crucial parameters to be taken into account in planning transmutation schemes.

Several possibilities for the transmutation of long lived nuclides by nuclear reactions have been suggested [2]. In the beginning, the best choice appeared to be the fast breeder reactor [3], because accelerators, light water reactors or even futuristic fusion reactors did not appear economically viable. However, recently there has been a renewed interest in the accelerator driven transmutation (ADT) schemes which seems to show good promise.

The partitioning of nuclides had been seen mainly as an extension of the PUREX process. Technical feasibility studies (mainly in the USA), showed that the cost-benefit ratio for the net radiological risk reduction exceeded by $32,400 per man-rem the US guideline of $1000 per man-rem [4]. Here, the radiological risk reduction considered only minor actinides and not fission products, which, when included in the partitioning scheme, would have considerably reduced the cost per man-rem. Because of their low reaction cross-section, $^{99}$Tc, $^{129}$I, $^{135}$Cs could not be efficiently transmuted in the present nuclear power stations, so that the transmutation of those fission products appeared not to be feasible at this time. Later, it was proposed to use moderated assemblies in the blankets of fast breeder reactors or to employ accelerator driven systems having considerably higher neutron fluxes [5]. In 1982, the French Ministry of Research and Industry [6] encouraged continuation of research in this field, which, in France, led to the SPIN (separation and incineration) Programme where presently several options for partitioning & transmutation (P&T) are being studied. In the nineties the Japanese Government launched a similar initiative and started the Omega Programme that initiated broad research in P&T by several Japanese organizations.

It should be pointed out that technical feasibility, and especially the economic and radiological soundness of transmutation still needs to prove, so the arguments both for and against P&T should be carefully compared and evaluated.

The main arguments in favour of P&T are:

- burning of the actinides considerably reduces the necessary volume and relaxes the longevity requirements for final disposal stores which are very expensive and attract most intense public criticism and objections;
- multiple plutonium recycling which is the essence of a really efficient closed nuclear fuel cycle (NFC) is somewhat more difficult than first Pu recycle. However, additional complications due to the inclusion of MA recycling may not be critical;
- MA may become an additional source of fission energy.

The main arguments against P&T are:

- inclusion of highly radioactive transuranics into the NFC makes fuel refabrication more difficult and hazardous and necessitates the use of remotely controlled operations;
- many materials and technologies involved are proliferation sensitive;
- transmutation reducing the actinide component of high level waste (HLW) increases the total amount of medium level waste (MLW) and low level waste (LLW), but of shorter half-life.

Much stronger and more complicated interdependence of reactor physics on the one hand and the requirements for reprocessing, refabrication and final disposal on the other are characteristic of NFCs involving transmutation. Large variations in recycled fuel composition seem inevitable. Development of a detailed concept of a closed NFC including MA burning in advanced nuclear power systems is not just a multiparametric optimization problem. Some important input data, both in material science and nuclear physics still are not known with the necessary accuracy and should be
determined. Various proposed radiochemical processes need further investigations. For accelerator driven nuclear systems new technologies need to be developed.

Strategies of nuclear power development in every country are obviously dependent on historical roots and are strongly individual. The basic reactor types used in national nuclear power industries are quite diverse. Decisions once taken and implemented in each country have determined the path of development for many decades. National approaches to transmutation R&D also reflect this general situation as transmutation technologies are expected to be a logical continuation of the power reactor technologies adopted and proven in a particular country. However innovative approaches should be considered carefully so as not to block the realization of new and promising ideas.

A consistent concept for handling long term HLW must include both general principles, the description of methods chosen for realization of those principles and detailed explanation of the choice. The general points to be considered are:

1. Main strategic options of the nuclear fuel cycle which determine the rate of production, total volume and composition of HLW.
2. Sharing of risk and of responsibility connected with HLW between present, next and future generations.
3. The choice of optimum combination of passive (controlled storage and final disposal) and active (burning up and transmutation) methods in handling HLW.

Further details are related to:

(a) The composition of burnt fuel and the products of its radiochemical processing from presently operating and future plants.
(b) Individual parameters of HLW radionuclides determining the conditions of storage, the need and possibility of transmutation.
(c) The resulting list of the isotopes - candidates for transmutation and optimum storage conditions for the remaining radionuclides.
(d) Critical analyses of proposed transmutation schemes which may fall into the following main categories:
   - high flux reactors with thermal or slightly harder (epithermal, resonance) spectra;
   - fast reactors (FP) (standard power plants or specialized actinide burners);
   - subcritical assemblies and blankets, both thermal and fast, driven by high energy, high current accelerators or by other powerful neutron sources (for example, future fusion reactors).
(e) Using the results of above analyses, to choose the most promising designs and point out both their advantages and the problems to be solved.

There are several international programmes set up to study the P&T option. The OECD/NEA is co-ordinating the R&D activities of its interested Member States. The organization regularly arranges information exchange meetings and has started evaluation of the present activities [7].

In its cost shared action (CSA) programme, the European Union has supported and co-ordinated activities in several fields of partitioning & transmutation. A strategy study is being made to assess the benefits of partitioning & transmutation for the safety of the management and storage of waste. The participating organizations are CEA, Siemens, ECN, AEA Technology and Belgonucléaire. The CEA, FK, ECN, EdF, TUI co-operate together in EFFTRA to develop and jointly test heterogeneous mixed oxide fuels - which are to a large extent in inert matrices - to be irradiated in HFR and PHENIX. In a trilateral co-operation CEA, FK, TUI study mixed oxide fuels under irradiation in HFR, Osiris and PHENIX. In the frame of a CRIEPI commercial contract, the TUI is developing a minor actinide fuel using Zr-based alloys which is being irradiated by CEA in PHENIX (Metaphix).

The ISTC has accepted a research proposal from RIAR to demonstrate a minor actinide fuel cycle based on mixed oxides obtained by electro-deposition from NaCI-2CsCl melt. The oxides are
vibro-compacted and will be irradiated in BOR-60. The partners are PNC, BNFL and TUI. Two ISTC grants on transmutation related research were given to IPPE and the largest one ($3.2 million for two years) went to a collaboration, led by IPPE, on ADTT development.

There are considerations which indicate that now it is probably the right time to give further momentum to the research on the subject. First, the next generation of advanced reactors which will start operating in the first decade of the next century will have a lifetime of at least 50 to 60 years. This is long enough to require some flexibility in their fuel cycle options which should be planned now. Secondly, P&T is certainly a long term problem in the early stage of development and definite solutions may be reached only after a considerable amount of R&D work in this area. New facts are being accumulated fast and a lot of new, sometimes radically new, ideas and proposals are being formulated. At the same time, difficulties and contradictions are also being accumulated equally fast. The methods which are proposed to overcome them are often of purely theoretical and even hypothetical nature. Hence there is the necessity for constant analytical work with new information to avoid major mistakes and to correct and optimize plans.

Since there are also R&D activities in this field outside the OECD, the IAEA has taken an initiative to document the research activities in those Member States in order to assist in this programme. For the evaluation of the safety (environmental) and non-proliferation aspects of partitioning & transmutation of actinides and fission products, a co-ordinated research programme has already been set up [8].
2. CONCEPTS FOR THE FUTURE OF THE NUCLEAR FUEL CYCLE

2.1. INTRODUCTION

The options for the back end of the fuel cycle can be categorized broadly under three different approaches:

(A) Once through cycle or permanent disposal

In this option, the spent fuel is to be suitably disposed of as waste in a permanent manner. The plutonium and minor actinides present in spent fuel go unutilized. The amount of plutonium disposed of as waste is thus about 200 times that present in the equivalent waste after reprocessing. Disposing of spent fuel under a once through cycle strategy is yet to be standardized as a proven method. Presently, the strategy is to store for an interim period and work on permanent disposal methods such as site development, etc.

(B) Delayed reprocessing

In this option, the reprocessing is to be performed after the spent fuel has been kept in storage for several decades. While in this case, the reprocessing becomes relatively easier due to substantial cooling of the spent fuel during storage, this option entails the problem of safe temporary storage of spent fuel for a long period. This also requires construction of special spent fuel storage facilities, and involves passing on the cost and responsibilities for safety to future generations.

(C) Closed fuel cycle

This is by far the most extensively investigated option to date. It involves reprocessing and recycling. During reprocessing, unburned uranium and more than 99% of the plutonium formed are separated. In the case of low enriched uranium, this represents nearly 97% of the initial quantity of the fuel which can be recycled back for nuclear power generation. The reprocessing and recycling of Pu in a thermal reactor increases the fuel utilization factor by nearly 40%. A very substantial improvement of fuel efficiency can be achieved by feeding the recovered plutonium into fast reactors. The reprocessing and recycling option, i.e. the closing of the fuel cycle is certainly the best choice from a fuel utilization point of view.

Short summaries of activities in OECD countries have also been included in this chapter to outline the overall international perspective in this field.

2.2. SUMMARY OF NATIONAL ACTIVITIES

2.2.1. Belgium

With 60% of its total electricity production being of nuclear origin - (all nuclear power reactors are of the PWR type) - Belgium is strongly concerned with the nuclear waste issue. For this reason, studies are being performed and scenarios envisaged that are to lead to optimum spent fuel + waste handling and disposal policies.

Not only the minor actinides (MAs) and long lived fission products (LLFPs) are of concern, problems also arise as to the plutonium inventory in Belgium. Indeed, although the option of future utilization of MOX (mixed oxide) fuel has been taken, plutonium production will continue to exceed its consumption.

In this MOX utilization option, Pu is introduced into the MOX (thermal reactor) fuel, which hence consists of a mixture of about 95% (depleted) uranium and about 5% plutonium. At the present time, the total loading of a large PWR may consist of up to 30% MOX fuel assemblies (the
rest being the conventional U-oxide fuel assemblies), without major modification of the reactor components or operation mode (higher MOX fuel assembly contents could be used later, but this would need some additional adaptation of the reactor).

Recycling of the MOX fuel assemblies, after reprocessing, may be envisaged. During such a second passage the fraction of Pu in the fuel would increase from about 5% to about 8%. After this second passage the operation loses all interest because of the degraded quality of the Pu (high content of even isotopes) and of the increased production of long lived transplutonium actinides. The recycling of plutonium in MOX is therefore only an intermediate step, which allows its use as fuel and leads to a slowing-down of the increase of the plutonium inventory.

P&T of the MAs (mainly $^{237}$Np, $^{241}$Am, $^{243}$Am and the curium isotopes) is an option for the future, which can only be efficiently realized if all fuel is reprocessed.

In the context of improved reprocessing, Np extraction is the first additional separation, which can be carried out relatively easily by a slight modification of the current PUREX reprocessing process. The extraction of Am-Cm from high level liquid waste (HLLW) is still in its laboratory development stage. Several methods have been considered: TRUEX, DIDPA, DIAMIDES and TRPO. The last-mentioned seems the most promising. The main issue to be solved is nevertheless the Am-Cm/rare earth separation for which no fully satisfactory method is available.

Transmutation covers two distinct concepts: transmutation to a short lived actinide and/or incineration to fission products. Transmutation in LWRs and HFRs (high flux reactors) involves the transformation of $^{237}$Np into $^{238}$Pu and a buildup of a heavier isotope fraction of Am-Cm.

Among the LLFPs, $^{129}$I and $^{99}$Tc are the most important nuclides to eliminate. During reprocessing $^{129}$I is separated from the fuel and waste stream and could theoretically be transformed into a target type solid for partial transmutation before long term storage. Presently the iodine resulting from reprocessing operations is discharged into the ocean.

$^{99}$Tc is present as soluble and insoluble species and is accompanied by a number of noble metals from which it has to be separated before transforming it into a target for transmutation.

If the plutonium and the MA inventories are to be reduced by significant quantities, e.g. by a factor 10, it will be necessary to insert a new type of fast burner reactor into the global reactor park (probably in collaboration or exchange with other countries). Such reactors would be either fuelled with mixed oxide (or nitride) elements having a high (about 45%) plutonium content or fuelled with 100% plutonium in inert matrices. The elimination period of Pu and MAs would nevertheless remain very long (of the order of 100 to 200 years) and the related costs would be very high.

Accelerator driven transmutation is a potentially interesting but very complex technology which bears promise to reach much higher burnups than achievable in fast reactors and to accelerate the actinide transmutation.

In the framework of a possible transmutation of long lived nuclides, and possibly also of enhanced plutonium consumption, the Belgian materials testing reactor BR2 could play a useful role:

- for the investigation, on a technological scale, of the transmutation processes in various neutron spectra,
- for the investigation of the metallurgical behavior of the targets.

Belgonucléaire is also concentrating its efforts on the operational conditions in the MOX fuel fabrication plant.

Whatever the scenarios followed, the ultimate geological storage of a large part of the radionuclides produced by nuclear energy remains the main direction in which the Belgian
authorities will determine the long term policy. Since 1972, intensive investigations have been carried out in this field. Including work in the underground laboratory of SCK-CEN.

2.2.2. China

At the present time, the Chinese nuclear electricity generation rate is 15 TW·h/a (2 TW·h/a for Qinshan and 13 TW·h/a for Daya Bay). It may be expected to increase slightly and to reach a new rate later on. This nuclear electricity is produced by PWR type reactors using uranium oxide fuel assemblies. With such a type of fuel, the MA production rate is about 53 kg/a and the long lived fission product Tc is 55 kg/a. In order to reduce the long term radiological hazard associated with the disposal of high level radioactive waste, the problem of MA transmutation has been placed on the agenda in China.

In recent years, some studies on the subject of P&T have been carried out in China; the terms of the research activities are as follows:

2.2.2.1. The development of nuclear data preparation

CENDL-2 (The second version of the Chinese Evaluated Nuclear Data Library), one of the large evaluated nuclear data libraries, was developed by CNDC (Chinese Nuclear Data Center), completed and released worldwide in 1992. The most advanced nuclear data processing systems NJOY, MINX and AMPX-II were used to create various multigroup cross-section sets in China. The CCCPS processing system was used to create a multigroup cross-section set in ABBN format based on the LIB-IV library. The code system PASC-1 was used to create a neutron-gamma coupled multigroup cross-section set from the fine-group library VTTAMIN-C. The code NJOY and modules LINX and CINX have been used to create the multigroup cross-section set CLIB-2 in ABBN format. This library is suitable for transmutation calculations. The nuclear data of $^{241}\text{Am}$ has been evaluated for gamma production data, covariances and isomeric ratio of cross-sections by CNDC.

2.2.2.2. Studies on the feasibility of MA transmutation

2.2.2.2.1. Studies on the feasibility of MA transmutation in CEFR

Various methods of MA transmutation have been proposed during the last 20 years. Among the possible schemes, one of the practical methods is to recycle MA in fast reactors so as to incinerate them. Over the years a study on feasibility of MA transmutation in CEFR has been carried out.

2.2.2.2.2. Studies on the feasibility of MA transmutation in fusion-fission hybrid reactors

Some studies on the feasibility of MA and long lived fission products in fusion-fission hybrid reactors were carried out during recent years and some calculation results have been published. The results show that this may be a possible future method for actinide and fission product transmutation.

2.2.2.2.3. Studies on partitioning of MA

The research programme on partitioning and separation of actinides (An) and lanthanides (Ln) from HLW started more than ten years ago in China. Three different chemical flowsheets were demonstrated and compared, i.e., the reverse Talspeak Process, the DHDECMP process and the TRPO process. In the first process the An-Ln was extracted in 0.3 mol/L HDEHP-0.2 mol/L TBP-kerosene from a pH2-3 aqueous phase, then separation of An from Ln was performed with 0.05 mol/L Na5 DTPA-1 mol/L lactic acid with pH equal to 3. In the second process, the extraction of An-Ln from simulated HLLW with high acidity by DHDECMP-DEB or DHDECMP-TBP-kerosene was investigated. The third one is the TRPO process. TRPO is a mixture of trialkyl phosphine oxide with carbon number in the range of 6 to 8. 30 vol% TRPO-kerosene was used as extractant, the acidity of the aqueous phase was contained to around 0.20 mol /L-1.0 mol /L HNO3. Recently, separation of An from Ln in a re-extraction solution of DHDECMP, which had extracted An-Ln, was performed with HDEHP-kerosene.
At present, following the development of PWR nuclear electricity in China, the problem of alleviating the burden on the high level radioactive waste depository is on the agenda. An effective long term R&D programme supported by authority, for developing MA transmutation schemes is very necessary. All the past work mentioned above is the result of the effort of individual groups. Therefore, a unified programme to develop an MA transmutation scheme in China is important. Some details on the research described above may be found in Refs [9-17].

2.2.3. Czech Republic

2.2.3.1. Current status

As an initial stage of an organized national research programme in the field of transmutation of radioactive waste and spent fuel from thermal reactors, a national documentation center has been established in the frame of the Institute of Nuclear Physics of the Academy of Sciences of the Czech Republic (ASCR). One of the main directions in which the effort will be oriented and concentrated in this initial stage is an overall critical study of the field mentioned above under the specific conditions of the national nuclear power programme and the role of an accelerator driven subcritical reactor system in it. In the frame of this programme, the following subprogrammes are to be tackled and work on some of them has already started this year (1995):

1. Theoretical and experimental investigations of an optimal combination of a target and a projectile particle. (Research studies have been based on corresponding activities of international institutes in CERN and Dubna.)

2. Choice of a suitable target type and assembling of the necessary set of corresponding data (including those on target depletion, its service life and yield decrease).

3. Study of problems of long lived fission products and a blanket formed from them (global balance of those isotopes, possible utilization of (gamma,n) and (gamma,p) reactions for radioactive waste management).

4. Collection of cross-sections, fission product yields from transuranic isotopes, spectra and similar characteristics prediction for actinides and fission products from burnup calculations for the system.

5. Choice of a suitable accelerator (linac versus cyclotron, summarization of requirements, problems of development and manufacturing, financial questions, advantages and disadvantages of different systems).

6. Principle study of various types of reactor systems.

7. Solving of the chosen system kinetics.

8. Problems of an entry of the beam into the reactor vessel (service life of the window, embrittlement problems and possible accidents).

9. Basic balance study of the generation of actinides and fission products in the frame of the national nuclear power programme based upon the WWERs (PWRs) and following determination of the size and power of the designed devices.

10. Identification of chemical methods and other processes involved in a continuous reprocessing system.

Supported by the Czech Power Generating Board and the Ministry of Industry and Trade, the programme started with a conference organized in June 1994 which summarized all the potential capacities in the country and their abilities to take part in the programme. The following institutions
presented their contributions and proposals to the programme and announcements of their intention to participate:

- Institute of Nuclear Physics ASCR, Rez,
- Nuclear Research Institute Rez, plc, Rez near Prague,
- Skoda Nuclear Machinery, Ltd., Pilsen,
- Czech Technical University, Faculty of Nuclear and Physical Engineering, Praha,
- Charles University, Faculty of Mathematics and Physics, Praha,
- Institute of Thermomechanics and Hydraulics, Brno,
- Institute of Special Inorganic Chemistry ASCR, Rez.

2.2.3.2. Proposed activities in the near future

On the basis of the results and experience obtained in the frame of the already mentioned first stage of the national programme the second stage will start in the near future (1995-1996):

1. Engineering and design development of a chosen version of the accelerator driven reactor system (first variant of an initial design):
   a. Solving of the problems of driving the accelerator and its control.
   b. Determination of thermal power generation during operation as well as after shut down, calculations of power distribution in the system and heat transfer from the core and target.
   c. Temperature distribution and designs of circuits.
   d. Solutions of safety problems.
   e. Main design work (vessels, exchangers, circuits, pumps, turbine, condenser, etc.)
   f. Material properties problems.
   g. Technological designs.

2. Radiation shielding (both accelerator and reactor) designs.
3. Overall plant design of the accelerator driven reactor system.
5. Designs of auxiliary devices. Fuel handling systems, etc.
6. Fuel system design.
7. Evaluation of a plant for continuous chemical separation and development of suitable methods.

2.2.3.3. Plans and recommendations

While proposing a national research programme in the field of transmuters, the real situation in the national nuclear power programme has to be taken into account. There are four power reactors in operation and two additional ones under construction in the Czech Republic at the moment. The question of further development of nuclear power in the country is still open, however, there has not been any decision made either to erect some more NPPs or to stop the nuclear power development programme. Current strategy for the back end of fuel cycle is based on storing of spent fuel in pools in the main operational block for the first stage and then in dry storage for the second stage before a final solution will be accepted. The question of a final solution is again open and will be decided on the basis of overall complex conditions as are technical, economical, political, etc. in the corresponding term.

In such a situation, the spent fuel stored is not utilized as a secondary raw material and a source of energy and moreover forms a drag on the economy of electric power generation. The level of costs connected with this spent fuel management is largely determined by ecological considerations. These considerations depend closely on the reliability and credibility of predictions of spent fuel behavior and its influence on the environment. These are based in most cases upon mathematical modelling of radionuclide transport in the biosphere which is very often a subject of doubt by the public.
From these given facts, we should start with some attempt to formulate a research programme in the given region. First of all, we need to form a minimum research programme so as to create the necessary background for discussion and decision making. This is a typical situation for a small country and the behavior of a series European countries of a comparable size and overall conditions may serve as a pattern. On the other hand, if our country wishes to use some of the fruits of the expected long term international effort in this field of research it is necessary to be creatively employed in it. From this point of view we appreciate the role of the IAEA in the general field of nuclear power which is very helpful. Therefore we propose to take into account that beside the effort devoted to the elaboration of technical documents, a national research effort supported by international co-ordination is most effective. We may offer to insert into such a co-operative programme the essential part of our national programme outlined in this Chapter.

2.2.4. France

In order to respond to the public concern about wastes, and in particular the long lived high level ones, a French law issued on December 30, 1991 identified the major objectives of research for the next 15 years to provide a sound basis for a new debate and possibly a decision on final waste disposal options in Parliament in the early 21st century.

The objectives of research put forward are:

- improvement of waste conditioning; extraction and transmutation of the long lived wastes in order to minimize their long term toxicity;
- research performed in underground laboratories in order to assess the capability of geological structures to confine radioactive wastes (2 sites have to be selected for these underground laboratories, in concentration with the local population);
- the study of conditioning and extended surface storage of wastes.

To comply with the requirements of this law related to the management of long lived, high level wastes, the CEA has launched an important long term programme called SPIN (an acronym for separation and incineration).

Studies are being performed in order to reduce the potential source of radiotoxicity in a deep geological repository and/or to reduce the residual radiotoxicity risk presented by mobile long lived fission products. These studies are performed consistently with parallel studies devoted to plutonium management (as Pu is not considered as a waste, but as a useful - even if quite "cumbersome" - element for further energy production). These studies on Pu management are performed both on thermal reactors (MOX recycling in PWR’s and its optimization) and in fast reactors (the CAPRA project to burn Pu in fast reactors).

The transmutation programme covers the three following areas:

1. Basic data studies (i.e. how reliable are the calculations?)
   - sensitivity and uncertainty analysis;
   - minor actinide and fission data evaluation and measurement intermediate energy data and codes for accelerator-based systems;
   - Integral experiments for minor actinide and fission product data validation;

2. Fuel studies
   - fuel fabrication and irradiation for homogeneous recycling of Np and Am both in fast and thermal reactors;
   - inert matrix basic research, fabrication and irradiation for targets in the reactors;
   - heterogeneous recycling mode, in particular for Am;
   - experiments for $^{99}$Tc irradiations in power reactors;
3. Reactor and fuel cycle studies

- core characteristics modifications (e.g. reactivity coefficients) when minor actinides and/or fission products are added to the fuel/core constituents, fuel cycle impact: $^{238}$Pu content, decay heat, gamma and neutron sources, complete mass inventories;
- strategic studies for different scenarios (cost/benefit analysis);
- hybrid systems physics and conceptual studies (accelerator based systems).

A significant irradiation programme has been defined, both in fast reactors (PHENIX) and in thermal reactors (experimental reactor OSIRIS). The irradiation programme is devoted to the validation of homogeneous/heterogeneous recycling of minor actinides and fission products. Some examples of planned irradiations:

- an irradiation device for inert matrix tests (MATINA experiment in PHENIX) the capsule for a new SUPERFACT-2 experiment in PHENIX, aiming to reach higher burnups (> 10% h.a.) than in the previous SUPERFACT-1 experiment, the ACTINEAU experiment related to transmutation studies in PWR's (in the OSIRIS reactor);
- $^{99}$Tc irradiation in a subassembly of PHENIX located at the core periphery, using a moderator to increase the ephithermal capture.

Demonstration experiments are foreseen in SUPERPHENIX which will involve 1 to 4 fuel subassemblies loaded with about 2% Np homogeneously mixed into the fuel and a few Am targets in an inert matrix.

EdF is engaged on the same R&D programme as CEA on partitioning and transmutation in the frame of the French law issued on 30 December 1991. The main objective of this law was to manage, in the long term, high radioactive wastes such as minor actinides and fission products.

But it soon appeared that Pu management was a necessary step before dealing with MA studies, as the greatest amount of radiotoxicity was brought in by Pu. Especially in France, an excess of Pu may appear if the arrival of FBRs in the park is postponed. So, Pu burning in PWRs or in a few FR burners could be considered. Until now, EdF has concentrated on core studies, because of their possibility to undertake a great number of neutronic calculations.

As French policy is to recycle Pu at least once in a MOX PWR, a great number of scenarios have been considered, such as Pu or Pu+MA multicycling either in PWR or in FR, in order to retain the best solution with regards to a few criteria: radioactivity reduction, safety and economics. As yet there is no optimized solution, comparing FR to PWR. In these two types of reactors, however, a heterogeneous mode seems better than a homogenous one from many points of view (enrichment, safety, but not radioprotection). Pu multiple cycling does not seem impossible in a PWR as several solutions exist to limit Pu content in MOX fuel (i.e. $^{233}$U enrichment in the MOX, or use of a high moderation ratio, connected with the mixing of multirecycled Pu with Pu of the first generation).

Although FR have a better fission/capture ratio than PWR, the global difference is feeble because the Pu is degraded by its first passage in a MOX PWR. For the next year, radioprotection and separation studies will be emphasized, because transmutation seems feasible but could imply a great amount of reprocessing.

2.2.5. India

The decision taken in the early sixties in India to base its nuclear programme on the use of pressurised heavy water reactors (PHWRs) included spent fuel reprocessing for plutonium recycling in the fast breeder reactors in the second stage and $^{233}$U breeding from the large reserves of Thorium available in the country to utilise the $^{232}$Th $^{233}$U cycle in the third stage within the scope of the three stage programme. Reprocessing was also considered necessary for final waste disposal in terms of radiation safety.
The Indian nuclear power programme which is modest at present is expected to expand rapidly. A number of power reactors, initially based on pressurized heavy water reactor design followed by breeder reactors using plutonium and subsequently uranium fuel in the future, are planned to be set up. India has adopted a closed nuclear fuel cycle and has indigenously developed the necessary technologies right from Uranium mining to reprocessing and waste management.

Safe management of radioactive waste which is generated at every stage of the fuel cycle has been accorded the highest priority. The philosophy adopted for the radioactive waste management programme is to concentrate and contain as much radioactivity as possible and releases to the environment are kept to a bare minimum consistent with the principle of ALARA.

The processes adopted for the treatment of gaseous and liquid streams are to isolate the radioisotopes and condition them in suitable inert solid forms. Solid wastes too are subjected to volume reduction by techniques such as compaction and incineration. Based on the activity content and the radionuclides, the wastes are disposed of in engineered near-surface disposal facilities or in deep underground geological repositories.

In the complete fuel cycle, excepting the reprocessing step, only low and intermediate level waste are generated. These are treated by employing suitable processes such as precipitation, ion-exchange, reverse osmosis, evaporation, etc. At sites having favourable climatic conditions such as high ambient temperature, low humidity and high wind velocity, solar evaporation is employed. The concentrates from the above processes are subsequently immobilized in inert solid matrices such as ordinary Portland cement, modified cement, polymer, etc.

The high level liquid waste (HLW) generated during reprocessing contains about 99% of the radioactivity associated with the entire fuel cycle. For management of these wastes, India has adopted a three stage programme.

In the first stage, the HLW is immobilized in a borosilicate glass matrix. In the second phase the vitrified high level waste canisters are stored under continuous surveillance in an air cooled vault for a period of about 25 to 30 years. This interim storage helps in assessing the integrity of the waste package besides bringing down the activity and decay heat. In the third stage, the vitrified HLW canisters are planned to be disposed off in deep underground geological repositories.

Our experience thus far in managing low and intermediate level waste has been good and our efforts presently are being directed towards establishing a very reliable and safe disposal systems for immobilized HLW.

The Indian programme of nuclear power generation is conceived to consist of three stages of nuclear fuel cycle development:

- The first stage consists of setting up a number of power reactors based on PHWR design. The plutonium generated in PHWRs and coming out of the reprocessing plants is to be utilised in the second stage consisting of the fast breeder reactors to produce further Pu fuel from uranium and/or $^{233}\text{U}$ fuel from the thorium blankets.

- In the second stage advanced heavy water reactor systems utilising plutonium fuel are also being considered as an option and R&D on the design work in this direction is in progress.

- The third stage running in parallel with the second stage will consist of thorium $^{232}\text{U}$ fuelled reactors utilising the vast reserves of thorium in the country for nuclear power generation over the years.

India has developed necessary technologies right from uranium mining to spent fuel reprocessing and waste management relevant to all the three stages in this closed nuclear fuel cycle.
Thus, by the very initial formulation, the Indian nuclear energy development and utilisation programme calls for extensive recycling of nuclear material. India's first nuclear power station consisting of two boiling water reactors of 210 MW(e) each went into operation in 1969 at Tarapur. However, a decision was taken at this stage to switch over to PHWR as the main reactor system for deployment in the first stage of our nuclear power generation programme. Therefore, the second nuclear power station located near Kota in Rajasthan consisted of two PHWRs of 220 MW(e) each which went into operation in the seventies. The third plant at Kalpakkam near Madras and the fourth at Narora are each of 2 x 220 MW(e) PHWRs.

Recently two PHWR units of 220 MW(e) has been commissioned at Kakrapar in Gujarat and a similar second unit is expected to go critical shortly. Additional power stations consisting of 2 x 220 MW(e) and 500 MW(e) PHWR units are in the various stages of planning/construction at various locations.

Development of Fast Breeder Reactor (FBR) technology is an important step in India's nuclear energy generation programme. While Commercial development of FBRs in developed countries has been slowed down because of easy availability to them of low cost uranium, the condition in India is different due to the limited availability of uranium.

The Indian FBR programme was initiated in the early seventies with the setting up a 40 MW(th) fast breeder test reactor (FBTR) at Kalpakkam. The FBTR used a core of plutonium-uranium carbide fuel. Based on the experience in operating the test reactor, the design of a 500 MW(e) prototype fast breeder power reactor (PFBR) has been initiated.

Thus, India's nuclear energy development programme includes spent fuel reprocessing at the back end of the nuclear fuel cycle to achieve both waste conditioning for safe final disposal and plutonium recycling in fast breeder reactors to derive maximum possible energy output. The reprocessing programme in India was launched with the commissioning of the plutonium plant built at Trombay in 1964 to reprocess the spent fuel from the research reactor CIRUS. This facility is now catering for the processing of spent fuels from the DHRUVA reactor as well.

A Power Reactor Fuel Reprocessing Plant (PREFRE) was set up in the seventies at Tarapur for reprocessing the Zircaloy-clad oxide fuel from the power reactors at Tarapur (BWRs) and Rajasthan (PHWRs). To cater for the reprocessing of spent fuel from the Madras Atomic Power Station (MAPS), a new plant has been built at Kalpakkam near the power station.

The feasibility of recycling plutonium in the Tarapur boiling water reactors was also considered in the late seventies and a programme was initiated to work out schemes which would expedite such a recycle. The objectives were to sustain the fuelling of a BWR utilising plutonium with the constraints of not altering the operational modes and not making major changes to the mechanical design of the fuel bundle. Fabrication of experimental mixed (Pu-U) oxide (MOX) fuel rods was carried out in BARC and irradiation tests were carried out in the research reactor CIRUS.

The utilisation of plutonium in a PHWR recycle mode has also been considered and found to have many advantages such as improved fuel utilisation, extended burn up and very significant front end and back end economics. The quantitative requirements of plutonium for this scheme are quite small and easily met. A viable fuel design has been worked out and the relevant safety studies related to bulk implementation of this scheme have been conducted. Based on the results obtained detailed plans have been worked out for utilisation of MOX fuel at Tarapur. A beginning has been made by charging two MOX-BWR assemblies last year.

Plants for the treatment of low and intermediate level wastes from the reprocessing plants are operational at the various sites in India. A waste immobilisation plant for the treatment of high level waste (HLW) has been set up and is operational at Tarapur. It is a semi-continuous pot glass process involving calcination followed by melting in the processing vessels headed by an independently controlled multizone induction furnace. A solid storage and surveillance facility (SSSF) has also been
set up for the interim storage or vitrified HLW prior to disposal at a permanent site. Two more waste immobilisation plants are being set up at Trombay and at Kalpakkam, which will employ a Joule heated ceramic melter.

Research and development activities for the improvement of the existing processes and systems in waste management include actinide partitioning and separation of long lived fission products. Partitioning of long lived alpha emitting actinides from the HLW originating from fuel reprocessing operations is considered very useful from the point of view of reduction of alpha activities of the waste. A long term policy on the final utilisation/transmutation of the partitioned actinides will be evolved at an appropriate time taking into consideration the evolving technologies on the different aspects of partitioning and transmutation (P&T) schemes.

To summarise, in the Indian context, reprocessing and closing the back end of the fuel cycle have become an industrial reality for full exploitation of the Indian uranium resources and waste disposal with long term safety. Considerable experience is being gained in plutonium recycling in thermal as well as fast breeder reactors. India has assessed the various options in the light of her energy demands and waste disposal requirements and decided that reprocessing and closing of the fuel cycle is the only responsible approach with respect to future generations.

Some details on Indian P&T activities may be found in Refs [18-28].

2.2.6. Japan

2.2.6.1. Introduction

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

The situation in Atomic Energy has changed drastically since the publication of the 1987 Programme, including the collapse of the 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 Programme was consolidated through careful and energetic study by the AEC, taking public opinion into account.

The new Programme attaches particular importance to 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. Basic research and development on partitioning and transmutation technology is to be promoted as a future technology in this area.

2.2.6.2. Partitioning and transmutation technology in the new programme

2.2.6.2.1. 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. The new Programme 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 Programme stipulates procedures, schedules and responsibilities of entities concerned in 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 2030s or by the mid-2040s at the latest.
The partitioning and transmutation technology, which would reduce the environmental impact of disposal by utilizing useful nuclides in the high level radioactive waste, is considered to be a future technology in the new Programme to be executed by the Japan Atomic Energy Research Institute (JAERI), the Power Reactor and Nuclear Fuel Development Corporation (PNC).

PNC and other organizations, such as the Central Research Institute of Electric Power Industry (CRIEPI), are carrying out the basic research and development of these technologies. A check and review based on the progress of these activities will be carried out sometime in the second half of this decade.

2.2.6.2.2. Partitioning and transmutation as nuclear science and technology

The partitioning and transmutation technology will be promoted from two aspects.

As basic research, the new Programme urges the promotion of research on TRU nuclides and on generation and utilization of various kinds 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.

2.2.6.2.3. OMEGA project and future plans

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

(a) Partitioning
- Partitioning of nuclides in HLW
- Recovery of metals from insoluble residue
- Utilization of recovered metal

(b) Transmutation
- Reactor transmutation
- Reactor physics
- FBR (PNC)
- Actinide Burner Reactor (JAERI)

(c) Accelerator driven transmutation
- Proton accelerator (JAERI)
- Electron accelerator (PNC)

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

Nuclear power plants have been part of the national energy system of the Republic of Korea ever since the start of commercial operation of the country's first nuclear power reactor, Kori-1, in 1978. The nation's nuclear power programme has continuously expanded since then.

Nine nuclear power plants (8 PWRs and 1 CANDU) are in operation with a generating capacity of 7,616 MW(e). The nuclear share of total installed electrical capacity is about 36% but about 50% of electricity is produced by these nine nuclear power reactors. In addition, 14 more nuclear power reactors are planned to be constructed by the year 2006.

The safe management of radioactive waste generated from nuclear power plants has become an important part of the nuclear energy policy and attracts strong public interest. Radioactive waste management should, therefore, be carried out systematically to achieve public confidence.

The Atomic Energy Commission (AEC), which is the country's top policy-making body on nuclear problems in the Republic of Korea, set the main goals for spent nuclear fuel as follows: spent fuel will be temporarily stored at an away-from-reactor (AFR) interim storage facility until the decision for future reprocessing or direct disposal is made.

In order to respond to the public concern about the long lived radionuclides in spent fuel or high level radioactive wastes, the Republic of Korea embarked on a long term research programme in 1992. The objectives of the research programme are to set up a methodology to assess and reduce the radiotoxicity risk of the long lived radionuclides in high level radioactive waste.

The transmutation study programme covers following areas:
- data evaluation for minor actinide fission cross-sections and spectra;
- intermediate-energy nuclear data evaluation;
- sensitivity and uncertainty analysis for nuclear data;
- feasibility study on the transmutation of minor actinides in a PWR;
- development of computer codes for the transmutation rate;
- set up the conceptual and basic design of a fission transmutation system;
- generation of nuclear and thermal hydraulic design parameters;
- analysis of sensitivity and uncertainty for safety and design parameters;
- hybrid system physics and conceptual studies.

The partitioning study programme covers the following areas:
- 4 group separation process, to be developed;
- coprecipitation of MA and Ln by use of oxalic acid;
- mutual separation of MA and Lanthanides by extraction with HDEHP;
- purification of MA.

2.2.8. Russian Federation

In the former Soviet Union countries including the Russian Federation political aspects of the RW problem have specific complications due to:
- Chernobyl accident aftermath;
- existence of large areas badly contaminated by radioactive substances as a result of military nuclear activities going on for decades;
- appearance of new independent states which made many operations with nuclear materials, including RW, into a trans-border activity;
- nuclear disarmament resulting in a large release of nuclear materials posing serious security, safety and non-proliferation problems;
- general economic difficulties.
All this should be kept in mind in assessing the present and future status of RW oriented R&D in the Russian Federation. This R&D is intensive, covers a wide range of subjects with many institutions involved but at the same time meets considerable financial and organizational difficulties.

Three main types of actinide containing nuclear materials and RW stored in temporary repositories in the Russian Federation may be singled out and ordered according to the complexity of their composition:

a. Military production reactor waste left after efficient extraction of U and Pu from very low burnup spent fuel. $^{237}$Np is practically the only actinide component of the composition.

b. The complicated mixture of minor actinides (MA) left after reprocessing of well burnt spent nuclear fuel (SNF) from commercial power reactors. $^{237}$Np forms about half of its mass, the rest being isotopes of Am and Cm.

c. Complete spent fuel elements, the product of an open NFC.

Each of the groups a-c is potential input for P&T processes requiring individual approach depending on composition. As was already mentioned, plutonium forms the bulk of the material to be recycled in any case, that is why other actinides are called "minor".

Military HLW is a specific problem of the Russian Federation as one of nuclear weapon powers. It is produced in radiochemical processing of the irradiated uranium and differs from the spent fuel from NPPs in that:

1. To produce weapon grade plutonium low burns are needed with the resulting plutonium contents of not more than 1 kg per ton of natural uranium, so the volume of the uranium to be reprocessed and resulting physical volumes of HLW are very high.

2. The extraction of plutonium is very effective, not more than 1 mg/l of it goes to the waste.

3. The accumulation of other actinides is relatively low except $^{237}$Np.

4. The spent fuel of inner cores is close to spent fuel from NPPs, it takes special reprocessing lines and their HLW are more like commercial HLW.

5. The disarmament process has stopped most of the plutonium producing reactors with resulting reduction of radiochemical reprocessing and HLW volume.

6. Actinide waste handling is closely interconnected with the most important and acute problem of safe utilization of the weapon grade plutonium.

It is also worth pointing out that the situation with secondary nuclear fuel in the Russian Federation differs in one important respect from that in OECD countries. For them uranium is just one of many things they can easily buy. For the Russian Federation it is rather important export product.

In the case of enriched uranium or ready reactor fuel, what is sold is not mainly a raw material but a high technology product. Thus, the use of plutonium with its presently "negative value" on the international market, instead of uranium may be a potentially very profitable process in the Russian Federation.

A closed NFC remains Russian national strategy for the peaceful use of nuclear power with plutonium, both weapon and commercial grade, considered as a key element of the cycle and not as a RW component. "Closed nuclear fuel cycle" and "transmutation of plutonium" are in fact synonyms.
As Minister V.N. Mikhailov pointed out: "The efforts connected with plutonium utilization in nuclear power, the uranium-plutonium fuel cycle and related technologies, investigations of physical and other problems, analyses of fast and thermal reactor potential were extensive enough in our Ministry from the very beginning" [29].

General conceptual work on transmutation, co-ordinated and funded by Minatom, includes:

- collection, systematization and analyses of the information published worldwide on neutron transmutation and using the results for working out and realization of corresponding national plans;
- analyses of alternative methods of managing high level waste, direct disposal first of all, to evaluate economic and ecological soundness and competitiveness of transmutation;
- tracing of the trends in the world nuclear power industry at large, first of all in the countries accepting a closed nuclear fuel cycle strategy;
- analyses of the progress of national research efforts in the field supported by theoretical and numerical investigation and compilation of corresponding reviews, status reports and updated proposals.

The post-Chernobyl period changed the pace of nuclear power development but not its main direction: thermal uranium fuelled reactors supported by sodium cooled fast reactors as utilizers of secondary fuel materials. That is what is usually called the evolutionary approach. Hence prevailing plans on actinide transmutation try to incorporate it into this general scheme in as natural way as possible [30-32]. They are supported by research and design institutions active in the development of PWR (WWER type) and fast (BN type) reactors.

There are also innovative approaches to actinide transmutation based on emerging concepts still to be tested on a large scale. In the Russian Federation two such concepts may be singled out: accelerator driven subcritical blankets [33, 34] similar to LANL proposals [35] and fast reactors cooled by liquid heavy metal coolants [36]. They are promoted by organizations which up to now have dealt mainly with heavy water and water-graphite designs.

Actinides are clearly among the first candidates for transmutation for two reasons: they all are, in the long run, alpha emitters and as such very radiotoxic; they all are fissionable, if the neutron spectrum is hard enough, and thus may be in principle used as components of nuclear fuel. The importance of this possibility is fully recognized in the Russian Federation.

Many Minatom organizations have shown interest in transmutation oriented research and development recently. Only the largest and most active in the field are listed below with a very short outline of main directions of their P&T research. Abbreviations of their Russian names are transliterated in brackets.

1. Institute of Physics and Power Engineering, IPPE (FEI), Obninsk: neutron physics, fuel and material science research with associated radiochemistry, thermohydraulics of fast sodium cooled reactors; plutonium utilization; core design and composition optimization on various critical assemblies and research reactors; nuclear data; technology of various liquid metal coolants.

2. Research Institute of Atomic Reactors, RIAR (NIIAR), Dimitrovgrad: reactor physics studies based on a few high flux research reactors; plutonium utilization; fuel fabrication technology on pilot plant scale; high fluence and high burn up irradiation experiments with post-irradiation examination; associated radiochemistry.

3. Institute of Theoretical and Experimental Physics, ITEP (ITEF), Moscow: neutron physics of accelerator driven energy producing and transmutation blankets with special emphasis on heavy water systems; accelerator and target technology.
4. A.A. Bochvar Research Institute of Inorganic Materials, BRIIM (VNIINM), Moscow: wide range of comprehensive reactor fuel studies, including material science, technology of fabrication, post-radiation investigations and radiochemistry.

5. Research & Design Institute of Power Engineering, RDipe (NIKIE), Moscow: conceptual studies on a lead cooled fast reactor proposed by the Institute; calculations and design of accelerator driven blankets and targets.

6. V.G. Khlopin Radium Institute, RI (NPO RI), St. Petersburg: chemical reprocessing and partitioning of spent fuel, development of corresponding radiochemical technologies.

7. Research Institute of Experimental Physics, RIEP (VNIIEF), Sarov: comprehensive study of nuclear, material and chemical properties of actinides; experiments with powerful neutron sources; nuclear data.


The last two organizations are engaged in reactor design and construction and have all kinds of experimental and testing facilities.

Two major international meetings on transmutation were held in the Russian Federation in recent years, one devoted primarily to accelerator driven transmutation technologies [37] and another to transmutation in fast reactors [38]. Regular Russian-American seminars on transmutation in accelerator driven blankets are held in ITEP.

Transmutation and especially plutonium utilization were high on the agenda of all the recent Annual Conferences of the National Nuclear Society [39-41]. These Conferences are the largest and most representative meetings on nuclear power problems held in the Russian Federation with wide international participation.

2.2.9. International organizations

2.2.9.1. Summary of P&T activities in the European Union

At the request of the European Parliament, the Council of Ministers of the European Union adopted in 1989 its multi annual Research Programme on "Radioactive Waste 1990-1994". Parallel to this, research and development on the transmutation of minor actinides in fast reactors is carried out in the European Institute for Transuranium Elements in Karlsruhe. In the framework of shared cost activities, the European Commission placed contracts with various research organizations in its member states that covered Partitioning and Transmutation of actinides from high level waste [42].

In partitioning, the studies were basically concerned with the separation of minor actinides from high level liquid waste resulting from the reprocessing of spent LWR fuel by the PUREX process. It followed the classical route of separating neptunium together with residual plutonium and the transplutonium actinides together with rare earths, from which they are separated later by a special extraction. Among the studies underway are the separation of actinides together with lanthanides using the DIAMEX process as developed by CEA, France, and using malonamides synthesized by the University of Reading, United Kingdom.

The group separation of lanthanides/actinides has been under investigation using tritertiary-butyl-PTZ that seems to be more efficient than the formerly applied TPTZ. Also other separations have been studied such as the use of soft donors by KfK Karlsruhe or using Ph₂P₃ or CMPO by ENEA, Italy, as well as the Talspeak process. These two later known processes have also been looked at by the European Institute for Transuranium Elements where the CMPO process has been
compared to the TRPO (as developed in China) and the DIDPA (as developed in Japan) processes [43].

2.2.9.2. Fuel and target development

In the seventies the Institute for Transuranium Elements started to develop mixed oxides containing uranium, plutonium and minor actinides. This so-called SUPERFACT experiment comprised the irradiation of 8 fuel pins in a standard PHENIX assembly, which contained the minor actinides neptunium and $^{241}$Am in the range from 2 to 40% [44]. The experiment has been evaluated and the material will soon be recovered for a subsequent irradiation.

In collaboration with CEA (F), ECN (NL), EdF (F) and FK (D), ITU will develop and produce several fuel materials for test irradiation in thermal as well as in fast neutron fluxes. The irradiation will comprise minor actinides as well as $^{99}$Tc and natural iodine. The concept of an inert matrix is included [45].

2.2.9.3. Strategy studies

In the framework of its shared cost actions, the European Commission has placed contracts with CEA (France), Siemens (Germany), ECN Petten (Netherlands) and more recently with AEA Technology (UK) and Belgonucléaire (Belgium) on a strategy study of Partitioning and Transmutation to assess its benefits for the safety of the management and storage of radioactive waste [46]. The main results obtained so far in these contracts are summarized below.

The aim of a study carried out by CEA is to analyse a strategy for reducing the inventory of long lived radionuclides by P&T and to assess its technological requirements and costs. Reference scenarios without and with conventional reprocessing and scenarios using P&T are compared to assess the potentialities of P&T. The main results of this study are described in Ref. [47].

The three reference scenarios are:

- A reactor park having pressurized water reactors (PWRs) solely, producing a total power of 120 GW(e), which is close to the present European capacity, and burning uranium oxide (UOX); the fuel cycle is open without reprocessing;

- The same reactor park as before, but burning UOX and MOX (uranium and plutonium oxide) fuel; the fuel cycle is closed with PUREX reprocessing of UOX and MOX; the losses during reprocessing are 0.3% for U and 0.5% for Pu;

- As above until 2020 and then fast reactors (FR) are progressively installed; the losses during FR fuel reprocessing are 0.9% for U and 0.25% for Pu.

Three scenarios are considered for partitioning and transmutation, two with available technologies, and one with future technology:

- The first scenario foresees the transmutation of neptunium and americium from 2010 in PWRs in homogeneous or in heterogeneous mode; in the homogeneous mode, the UOX + actinide fuel is reprocessed in the same way as standard UOX fuel; the losses during reprocessing are 0.3% for U, 0.5% for Pu, 5% for Np and Am and 100% for Cm; in the heterogeneous mode, the specific targets containing either neptunium or americium oxide are irradiated during 5 years and are not reprocessed;

- In the second scenario the minor actinide partitioning starts in 2010, Np and Am are stored before being recycled in FRs after 2020 either in homogeneous or in heterogeneous mode. In the homogeneous mode, the fuel is reprocessed as the standard FR fuel; the actinide losses during reprocessing are the same as before; in the
heterogeneous mode, neptunium and americium oxide targets are irradiated during 15 years (3 cycles) and are not reprocessed;

- The last scenario is the same until 2030; FRs are progressively starting operation after 2030 to transmute neptunium in homogeneous mode and americium, technetium and iodine in heterogeneous mode; the fuel and targets are reprocessed with losses of 0.1% for U and Pu, 0.5% for Np, Am and Cm and 10% for Tc and I; curium is placed in interim storage.

The implementation of P&T of U, Pu, Np and Am with available technologies leads to a global cost increase of the overall fuel cycle between 10 and 50% with respect to the reference case where only U and Pu are recycled. This increase mainly reflects the higher reprocessing and minor actinide fuel fabrication costs for the P&T scenarios.

In another study Siemens Interatom has analysed the potential of a fast reactor, EFR, for transmutation with a thermal power output of 3600 MW(th), MOX fuel and a 1 m core height. The effects of enhancing transmutation by neutron spectrum hardening as well as metallic and oxide fuel (homogeneous or heterogeneous) are compared. The influence of the sodium void effect and the Doppler effect to the safety behavior of the different cores is evaluated.

The study shows: in keeping the sodium void effect at an acceptable level and maximizing the MA transmutation in a FR with a large diameter core and a reduced height (0.7 m is proposed), that it would be able to take 860 kg of MA and to transmute 65 kg of MA per year, which represents the production of about three PWRs.

The MA transmutation rate for oxide or metal fuel is nearly the same with a transmutation half-life of 11 years, taking into account the limitation of $^{239}$Pu buildup of 5% of the total Pu imposed at present by reprocessing. There is however a slight advantage of about 15% for the metal fuel concerning the transmuted mass per year because of the higher metal density in metal fuel.

The positive sodium void effect is much more favourable for oxide than for metal fuel, but the negative Doppler constant is strongly reduced in metal cores. This is beneficial for loss of heat sink accidents, but a disadvantage in case of transient overpower. On the other hand, metal fuel has a lower loss of reactivity and a longer residence time for the same maximum fuel burnup as oxide fuel. There are thus no clear advantages for oxide or metal fuel for transmuting MA in a FR.

The MA transmutation efficiency in fast reactors is similar for heterogeneous and homogeneous recycling, when considering the whole core, i.e. the MA containing subassemblies and the standard fuel subassemblies. About 30 to 35% of the initial MA content are transmuted during a subassembly lifetime of 6 years. The safety parameters, sodium void effect and Doppler constant, are much more favourable for heterogeneous than for homogeneous recycling.

The mass ratio of lanthanides to (Am+Cm) in spent UOX fuel is about 20-50 depending on the cooling time. The lanthanides have a similar influence on the core behavior as the MA: they increase the sodium void effect and reduce the Doppler effect. The admixture of MA and lanthanides has therefore to be limited to an upper value of 5% of the total mass of heavy metals. This ratio requires separation factors between MA and lanthanides around 30-50 in order to achieve transmutation efficiencies equivalent to those obtained without lanthanides.

Large quantities of $^{99}$Tc in the breeder zones of a FR core could be transmuted in special moderated target subassemblies. For a 1.2 m core height with 84 $^{99}$Tc target subassemblies and a thermal power output of 2600 MWth, about 116 kg of $^{99}$Tc can be transmuted per year, which corresponds to the $^{99}$Tc production of five to six 1 GWe PWRs.

The effective transmutation half-life is around 26 years. The safety parameters of such a transmutation device are in the scope of conventional FR layouts. The presence of target
subassemblies inside the core region even leads to a sodium void effect reduction. A macrocell study shows that, with an appropriate design of the moderated $^{99}$Tc subassemblies, a moderator volume of up to 20% does not disturb too much the fuel pin power distribution in the vicinity of the moderated subassemblies.

When replacing some of the $^{99}$Tc target subassemblies by $^{129}$I ones, the transmutation rate of $^{99}$Tc is not affected and about 22 kg of $^{129}$I could be transmuted in a FR with a transmutation half-life of around 44 years.

A study carried out by ECN Petten included:

- the preparation of a nuclear database for transmutation of actinides and fission products by inspecting the European JEF-2 library and by paying special attention to long lived fission product capture and (n, 2n) data;
- the assessment of the ORIGEN nuclear data library for transmutation studies;
- sample burnup calculations for a few scenarios using the ORIGEN code and the updated ORIGEN nuclear data library;
- investigation of transmutation of long lived fission products ($^{129}$I and $^{99}$Tc): in a very high thermal neutron flux reactor such as the European High Flux Reactor (HFR) at Petten.

The main results were:

- The cross-section database was updated for PWRs and FRs with data from the JEF2-2 and EAF-3 libraries.
- Three-group cross-sections for the ORIGEN-S fuel depletion code and one-group cross-sections have been calculated.

For PWRs, the cross-sections strongly depend on burnup, because the variations of the nuclide densities cause changes in the neutron spectrum and resonance integrals. The FR cross-sections only show a slight dependence on burnup. For both types of reactors, the cross-sections at average burnup are used for the updated databases.

Burnup computations for PWRs show that the nuclide densities obtained from the new cross-section database agree with those derived from the regular updated cross-section base within 20% and even much less than 10% for most nuclides.

A strategy study on transmutation of long lived fission products in different reactors is being performed [48, 49]. Computations indicate that the transmutation half-life of $^{99}$Tc (cf. $^{129}$I) is about 40 years (cf. 20 years) with a rate of 38 kg/year (cf. 46 kg/year) in heavy water reactors. These figures have to be compared with the annual production of $^{99}$Tc (20 kg) and $^{129}$I (4.6 kg) of a 1000 MW(e) LWR. With a special subassembly in the Petten high flux reactor, the calculated transmutation half-lives for $^{99}$Tc and $^{129}$I are about 8.6 and 5.7 years. In the case of transmutation in fast reactors, the results of computation for $^{99}$Tc are in agreement with Siemens.

In another study Belgonucléaire identifies the potentialities and constraints of actinide and long lived fission product recycling in PWRs. This study includes homogeneous recycling of Pu and Am; impact on fuel refabrication; physics of fission product recycling in dedicated assemblies in a PWR core having a better moderation. The first results show that the recycling of Am with Pu in MOX fuel limits the number of recycling steps to one instead of two or three [50]. The Pu + Am recycling strategy increases the dose rates by a factor of 4.5. An extra 25 mm steel shielding is required to reduce the dose rates to the values corresponding to Pu recycling only.

AEA Technology is assessing the secondary waste arising from partitioning of actinides and long lived fission products and from the fuel and target fabrication [51]. The plant requirements, indicative costs and doses to operators for these processes will be compared with a reference fuel cycle without P&T. The separation schemes under investigation are the CTH, TRPO, TRUEX and DIAMEX processes and a typical non-aqueous (pyrochemical) process.
3. TRANSMUTATION RESEARCH

3.1. NUCLEAR PHYSICS AND NUCLEAR DATA

With the introduction of MA into reactor fuel the nuclear data for these nuclides gain new importance because what previously was a small admixture now becomes a significant component of fuel composition. This fact is well understood in all the countries engaged in transmutation research and reflected in their corresponding programmes. Not all the important data may be measured directly for radioactive - sometimes highly radioactive - targets so theoretical calculations and integral experiments play an important role in producing a new and better set of evaluated data.

3.1.1. China

The Chinese Nuclear Data Center (CNDC) was established in 1975. CENDL-2 (The second version of the Chinese Evaluated Nuclear Data Library), one of the world's large evaluated nuclear data libraries, based on CENDL-1, was developed at CNDC completed, and released worldwide in 1992. CENDL-2 contains evaluated neutron reaction data of 54 elements or isotopes from H to $^{249}$Cf in the neutron energy range from $10^5$eV to 20 MeV in ENDF/B-6 format.

The most advanced nuclear data processing systems NJOY, MINX and AMPX-II have been introduced into CNDC from the USA. These codes were used to create various multigroup cross-section sets used in thermal and fast reactor calculations in China.

The LDB-IV, a library of group constants for nuclear reactor calculations has also been introduced into CNDC from NNDC of the USA. A processing system CCCCPS which contains three utility codes SIFP (coded by CNDC), BINX, and CINX was used in co-operation with LIB-IV to create a 46 group neutron cross-section set CLIB-1 with ABBN format for use in LMFBR calculations and especially for CEFR (China Experimental Fast Reactor) design calculations.

In addition, VITAMIN-C, a fine-group cross-section library (171 groups of neutrons and 36 groups of gamma rays) has been introduced from RSIC of the USA and a variant of the PASC-1 code system for fast reactor calculations was used to create a neutron-gamma coupled cross-section set from VITAMIN-C for use with the ANISN, DOT3.5 and CITATION codes. The PASC-1 system is a co-operative achievement of CNDC and ECN of the Netherlands.

At present, the study on actinide and fission product transmutation appears more and more important with the aim of reducing the long term radiological hazard associated with the disposal of high level radioactive waste. In order to meet the requirements of multigroup cross-section constants for transmutation calculations and in addition, to provide a new version of CLIB-1, the code NJOY, module LINX and CINX have been used at CNDC to create a multigroup cross-section set CLIB-2 which is based on the nuclear data libraries CENDL-2, ENDF/B-3, ENDF/B-6, JEF-1 and JENDL-3.1. Besides general reactor calculations, the new library CLIB-2 can especially be used for transmutation calculations because CLIB-2 contains more actinide nuclides data than the LIB-IV library.

The nuclear data of $^{241}$Am have been evaluated for gamma production data, covariances and isomeric ratio of capture cross-section by CNDC, these data are included in CENDL-2 and in ENDF/B-6 as well.

The current status of nuclear data activities in China is described in Refs [52-53].

3.1.2. India

The requirements for nuclear data are numerous and varied depending on the transmutation concept. Some of the important radionuclides involved in the transmutation schemes including fission products and actinides (L. Koch, IAEA-TECDOC-693, March 1993) are the following:
Nuclide/half-life (years):

\[ ^{14}\text{C}/5.7\text{E}3, \quad ^{36}\text{Cl}/3.0\text{E}5, \quad ^{129}\text{I}/1.6\text{E}7, \quad ^{135}\text{Cs}/2.3\text{E}6, \quad ^{78}\text{Se}/6.5\text{E}4, \quad ^{92}\text{Zr}/1.5\text{E}6, \quad ^{90}\text{Sr}/28.5, \quad ^{121}\text{Sn}/50, \]
\[ ^{126}\text{Sn}/10.5, \quad ^{137}\text{Cs}/30.17, \quad ^{99}\text{Te}/2.1\text{E}5, \quad ^{235}\text{Np}/2.1\text{E}6, \quad ^{238}\text{Pu}/87.7, \quad ^{239}\text{Pu}/2.4\text{E}4, \quad ^{240}\text{Pu}/6.5\text{E}3, \quad ^{241}\text{Pu}/14.4, \]
\[ ^{242}\text{Pu}/3.7\text{E}5, \quad ^{241}\text{Am}/432, \quad ^{242m}\text{Am}/141, \quad ^{243}\text{Am}/7.3\text{E}3, \quad ^{245}\text{Cm}/7.3\text{E}3, \quad ^{246}\text{Cm}/4.7\text{E}3, \quad ^{247}\text{Cm}/1.5\text{E}7, \quad ^{248}\text{Cm}/3.4\text{E}5. \]

The above list is not complete. Other nuclei of interest include Ta, W, Pb, Bi as target nuclei, molten salt components Li, F, Be, Zr + He coolant, structural materials and other fission products, etc.

For transmutation methods based on the use of intense neutron fluxes, detailed data providing knowledge of double differential neutron production cross-sections for the neutron producing target is essential. For transmutation schemes involving direct spallation of minor actinides and fission products by the primary proton beam, nuclear data of high energy fission cross-sections for the actinides is required.

The construction and shielding of the accelerator, producing an intense neutron source near the target area will need nuclear data for activation and material damage calculations.

Presently, the calculations of accelerator based transmutation generally involve use of the evaluated neutron-nuclear reaction and decay data from the nuclear data libraries such as ENDF/B-6 (USA), JEF-2 (OECD/NEA), JENDL-3 (Japan) and BROND-2 (Russia) which cover neutron energies below 20 MeV. However, the data in many cases are somewhat discrepant and need to be improved upon. The design studies on recycling of minor actinides produced in the \(^{238}\text{U}/\text{Pu}\) cycle in fast or thermal reactors will require the nuclear data of minor actinides to be known with further improved accuracy.

For all the important minor actinides, in addition to the neutron cross-sections, the data of fission product yields, delayed neutron yields, group parameters and spectra and prompt neutron yields and spectra are at present based on poor or sparse experimental data and calculations based on models. Prediction of safety related reactivity coefficients such as the Doppler reactivity coefficient and the sodium coolant void reactivity coefficient in fast reactors demands an accurate knowledge of resonance cross-sections for the minor actinides recycled in the core. Presently, the irradiation experiments in fast cores necessitate a more conservative design in order to accommodate uncertainties in decay heat, Doppler and coolant void reactivity coefficients arising from nuclear data uncertainties of minor actinides.

The high energy transport codes such as HETC which is based upon the classical intra-nuclear cascade model followed by evaporation from 1.5 GeV down to 20 MeV has been employed. It has been felt that the nuclear data files need to be extended beyond 20 MeV say up to 100 MeV as the validity of the intra-nuclear cascade model fails below 100 MeV.

A review of the most important nuclear data needs for accelerator based transmutation and the present status of the relevant available experimental data, systematics, theories and model codes has been recently given by Koning. (NAEA report NEA/NSC/DOC(92), 12 1992; report ECN-C-93-005 (1993)). One of the conclusions of this report is that there are sufficient nuclear data tools and bibliographic databases containing nuclear data in the energy region 20-1500 MeV, which can be used for construction of evaluated intermediate energy nuclear data files. It was also suggested that the extension of the existing low energy data files to energies of up to 100 MeV will be important for accelerator based transmutation research.

In the current nuclear data situation the overall calculations of transmutation processes from about 1.5 GeV proton beams down to thermalized particles in the shielding is based on the use of evaluated data files and their associated processing and transport codes for neutron induced reactions below 20 MeV, and high energy transport codes for all other reactions and energies.
The high energy transport codes produce unreliable results for incident energies below 100 MeV, and this may be due to the fact that it is not sufficient to describe the 20 to 100 MeV region with a single intranuclear cascade plus evaporation model. Instead, one needs to use suitable more refined nuclear models for different processes.

For example, an optical model and coupled channel calculations for elastic scattering and reactions to discrete states, a pre-equilibrium and equilibrium model for reactions to continuum and a proper fission model for the intermediate energy region, etc. are needed to describe the 20-100 MeV region. It is an important priority that as a complement to high energy transport codes, the region 20-100 MeV should be subjected to similar evaluation routines as the 0-20 MeV region. Preparation of evaluated nuclear data files for all nuclei of interest beyond 20 MeV, extending at least up to 100 MeV is an important priority. A possible further step is the construction of a data library to 1500 MeV.

Associated with development of such basic data files are the corresponding issues in the development of processing code system such as the NJOY and the neutronics code, such as the MCNP which is based upon the Monte Carlo Method.

In the case of using an accelerator driven intense thermal neutron source for transmutation, C.D. Bowman et al., point out that the nuclear data are needed for a number of short lived fission products and spallation products. Since there is a substantial epithermal neutron flux in the system, nuclear data are also needed in the epithermal range for a number of nuclei. Particularly for few-day half-life $^{232}$Pa, $^{239}$Np and $^{242}$Am nuclides, fission cross-section data are critically needed.

### 3.1.3. Russian Federation

Development and validation of a reliable transmutation concept depends strongly on the completeness and accuracy of nuclear data used in numerical modeling of the transmutation processes going on in various nuclear installations. Each of the main options considered now - burner-reactors and accelerator driven blankets - have distinct specific features as far as necessary nuclear data are concerned.

#### 3.1.3.1. Burner-reactors

Important aspects of the nuclear safety of transmutation installations are determined by the effective yields of delayed neutrons. These yields and the energy spectra of delayed neutrons for minor actinides are not known accurately enough but that is only a part of the problem. Another point is that the efficiency of delayed neutrons in fissioning the fuel nuclides with relatively high fission thresholds depends strongly on the inelastic scattering cross-sections of those nuclides, for which only calculated estimates are known. In this connection significant improvement of the accuracy of nuclear data for transuranics in the reactor range of neutron energies is needed, especially if the concentration of the nuclides in the fuel reaches dozens per cent. Primary needs: delayed neutron yields and spectra, fission fragments yields and inelastic scattering cross-sections for $^{237}$Np, $^{241,243}$Am, $^{238}$Pu, fission and capture cross-sections for Cm isotopes. One group estimates of required and achieved accuracies of fast neutron cross-sections for key nuclides are given in Table I (based on [54] with some additions).

More accurate thermal fission and capture cross-sections are also needed for short lived nuclides $^{237,239}$U, $^{238,239}$Np, $^{242}$Pu, $^{242}$Am.

Significant discrepancies exist in the evaluated spectra of prompt fission neutrons and their accuracy should be also improved.

Capture cross-sections present another problem. Their measurements are very difficult if not impossible. Heavier actinides are formed by successive neutron captures, so existing compounding of errors for capture cross-sections results in growing uncertainties in calculations of their accumulation and improvement of the theoretical models for calculations of capture cross-sections is one of the most important tasks.
<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Capture</th>
<th>Fission</th>
<th>Inelastic scattering</th>
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<tr>
<td>$^{237}$Np</td>
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<td>5</td>
<td>7</td>
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<tr>
<td>$^{238}$U</td>
<td>5</td>
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<td>$^{238}$Pu</td>
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<td>$^{241}$Am</td>
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<td>$^{242}$mAm</td>
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<td>$^{242}$Am</td>
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<td>$^{242}$Cm</td>
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<tr>
<td>$^{242}$Cm</td>
<td>50</td>
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<td>15</td>
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<tr>
<td>$^{244}$Cm</td>
<td>50</td>
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</table>

The importance of nuclear data for curium isotopes is sometimes underestimated because the total mass of curium accumulated in SNF is less than that of Np and Am. The very necessity of especially burning Cm is argued and not without reason - it is very difficult to handle in prefabrication. But Cm isotopes play a special role in burning $^{241}$Am and $^{243}$Am. Those two isotopes are mainly neutron absorbers and turn into curium faster than fission because $^{242}$Am and $^{244}$Am are short lived. The situation partly resembles the U-Pu case. About 30 to 40 per cent of energy produced in uranium fueled reactors is provided by fissioning plutonium. In an "Americium-fueled" reactor up to 80 per cent of the energy would come from fissioning curium because all its isotopes are rather long lived.

Microscopic capture cross-sections of Cm isotopes are practically impossible to measure directly and they may be only evaluated from fission cross-sections with the use of relevant theoretical models and estimated in integral experiments, hence the importance of Cm fission cross-section measurements.

### 3.1.3.2. Accelerator driven blankets

All the nuclear data mentioned above will be needed for this option also because neutron spectra in such blankets are close to reactor ones. But a very large volume of additional nuclear data is needed in the energy range 20-1500 MeV. The list includes double differential cross-sections of neutron production on target material, of gamma emission, activation cross-sections and other data for calculations of heat release, radiation damage and gas production.

Data for the following nuclides are needed:

1. Fuel actinides: $^{238}$U, $^{237}$Np, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu, $^{241}$Am, $^{242m}$Am, $^{242}$Cm, $^{243}$Cm, $^{244}$Cm, $^{245}$Cm, $^{246}$Cm.
2. The target: Ta, W, Pb, Bi.
3. Structural materials, shielding, coolants, etc.: H, C, N, O, Na, Mg, Al, Ar, K, Ca, Cr, Mn, Fe, Co, Ni, Cu, Zn, Zr.
Existing nuclear data in the 20-1500 MeV energy range are not systematic, are fragmentary and they need careful analyses and presentation in EXFOR computer format. Nuclear models and systematics available do not guarantee the required accuracy and need to be essentially improved. Russian experts plan to support international efforts in evaluation of the most important nuclear data for accelerator driven transmutation and to take part in the most important programmes:

1. Creation of a neutron and proton data library for key nuclides in the 20-100 MeV range.
2. Creation of reference (standard) data files for the energies up to 1500 MeV.
3. Development of an activation data library for neutrons and protons up to 100 MeV.

3.1.3.3 Immediate plans

IPPE (Obninsk)

1. Measurements of fission cross-sections of $^{238}$Pu, $^{242}$mAm, $^{243-248}$Cm for neutron energies 0.1-7.0 MeV;
2. Measurements of the yields of primary fragments in the fission of $^{237}$Np at 0.8-1.3 MeV, 3 MeV, 5 MeV including the region of large mass asymmetries;
3. Measurements of the neutron spectra of inelastic scattering and fission for $^{237}$Np;
4. Analyses of discrepancies of existing data for minor actinides and improvement of evaluations:
   a. Identification of the discrepancies in evaluated cross-sections and gamma radiation yields for Np, Am and Cm between BROND-2, JENDL-3,ENDF/b-VI and JEF-2.
   b. Description of total and elastic cross-sections for minor actinides in terms of a generalized optical model.
   c. Systematization of (n,2n),(n,3n) and fission cross-sections for minor actinides. Determination of level densities and fission barriers as input information for calculations of inelastic scattering and fission cross-sections, statistical evaluations of differences between $^{242}$Am cross-sections in ground and isomeric states.
   d. Improved evaluations of various cross-sections for Np, Am and Cm, first of all in the thermal and resonance region.
5. Measurements of total yields and calculations of group constants of delayed neutrons in fast neutron fission of $^{237}$Np.

RIEP (Sarov)

1. Nuclear data measurements for separated isotopes of transuranic elements:
   a. Preparation of mass separator and input materials, separation of Pu, Am and Cm isotopes, certification of samples, preparation of target layers.
   b. Preparation of measurement equipment, calibration of the targets, measurements of fission cross-sections on KS-1 and KS-2 critical assemblies.
   c. Measurements of the average number of fission neutrons and of the total energy of fission gamma rays and relative fission cross-section of $^{238}$U for 0.1 eV-12 MeV neutrons. Next step: the same for $^{235}$Pa and, probably, for $^{237}$U.
2. Evaluation of nuclear data for $^{236,237}$U and $^{233}$Pa.
3. Formation of specialized nuclear data library for transmutation (together with IPPE).

The most important of the recent experimental, theoretical and evaluation results on nuclear data which may be used in reactor and electronuclear transmutation facilities were presented at the International Conference on Nuclear Data for Science and Technology in Gatlinburg, Tennessee [55-65].
3.2. THERMOPHYSICS AND THERMOHYDRAULICS

The changes in core physics and technology needed to incorporate large admixtures of MA into the fuel make it necessary to reconsider some associated thermophysical problems also. Because fast neutron systems are most promising for transmutation the use of liquid metal coolants should be carefully studied which is also true for powerful accelerator targets needed to produce intense primary neutron fluxes to be multiplied in subcritical blankets. Some efforts in this direction are described below.

3.2.1. Russian Federation

Most of the basic problems in thermophysics and thermohydraulics of fast reactors cooled by liquid sodium (BN type reactors) were solved during some forty years of development of sodium coolant technology including operation of four reactors: BR-10 (Obninsk), BOR-60 (Dimitrovgrad), BN-350 (Kazakhstan), BN-600 (Ekaterinburg region), all in about 90 reactor-years. The figures in the abbreviated names of the reactors indicate their nominal power in MW, in the first two cases - thermal, in the last two - electrical.

Inclusion of Pu and minor actinides into the fuel, especially in the heterogeneous case, besides obvious changes in neutronics, results in some specific problems related to the geometry and other parameters of fuel assemblies which call for investigation of the temperature behavior of FAs in nominal, transient and accident regimes. This work is going on in IPPE liquid sodium rigs and includes:

- hydraulic optimization of FAs, determination of the margins to boiling;
- determination of temperature non-uniformities in FAs; calculations of the durability of FAs under thermal deformation;
- numerical modeling of coolant boiling due to power bursts or coolant flow drops.

Liquid heavy metal coolants (lead and lead-bismuth alloy) have attracted attention for a long time due to their excellent neutronics, satisfactory thermohydraulic properties and chemical inertness. There is a considerable experience of using Pb-Bi coolant in transport power units but the first publications began to appear only recently [66-70]. These systems are beginning to attract attention outside Russia [71]. So the thermohydraulics of fast reactors with heavy liquid metal coolants is an emerging issue. The practical feasibility of lead containing coolants has been proved but operation conditions and regimes and safety requirements are quite different for stationary NPPs so a lot of further development is needed.

High temperature differences the reactor wall and the coolant (50-100°C) and between the output and input (150°C and higher) are characteristic for such reactor concepts, so very reliable and accurate data are needed on heat exchange and on temperature non-uniformities in fuel sub-assemblies which have a larger pitch on a square lattice than traditional designs.

Intense conceptual investigations of the possibility to use lead as fast reactor coolant are going on in RIPE [72]. Lead coolant provides a number of attractive possibilities:

- high electric generation efficiency coefficient (up to 40 per cent);
- high fuel burn up (about 15 per cent) with low stresses in the fuel pin shells;
- large temperature margins to coolant boiling and fuel melting, high level of self-regulation and natural circulation, simple controls;
- possibility to exclude a large positive void reactivity effect and the formation of secondary critical masses;
- low induced radioactivity of lead enabling maintenance and repair works;
- long campaigns and relatively simple reloading;
- low radiation damage and low radioactivity of structural materials;
- simple and efficient systems for removing residual heat. These advantages may be used both in reactors and in the targets of electronuclear blankets.
A major problem is the rather high melting point of lead (327°C), so the technology of its use in nuclear power installations still has to be developed in detail and extensively tested.

Coolant flow is complicated by intense interchannel mixing and should be investigated in detail. Channel by channel flow calculations should be carried out and verified experimentally (distributions of velocities, pressures and pressure pulsations in the upper and lower reactor model chambers in nominal and other regimes, with different loop geometries, in heat exchangers and steam generators). Other problems:

- hydrodynamically suspended control rods;
- coolant technology;
- on-line coolant cleaning from radioactive polonium.

Considerable experience (over 30 years of work) has been accumulated in development of algorithms and computer codes and calculations of 2D- and 3D-flows and heat exchange in lead-bismuth coolant in reactor circuits. Those codes solve the Navier-Stokes equation in 2D (DUPT) and 3D (TUPT, SUPORT) geometries. Dynamic 1D (TRIANA, TRAKT-M) and 2D (DINCOR) codes were also developed for calculations of thermo-hydraulic processes in liquid metal circuits with the possibility of water and steam ingresses and of phase transitions taken into account.

Numerical and experimental investigations are under way to support the design of neutron producing targets and accelerator windows for transmutation applications. The designs were investigated earlier and continued for many years mainly to meet the requirements of the accelerator complex in the Institute of High Energy Physics in Protvino. It was demonstrated that solid targets may withstand power densities $10^7-10^{10}$ W/m$^3$ depending on the beam parameters, material chosen and planned lifetime. Effective methods of target cooling were developed with the use of many experimental facilities for investigations of the targets behavior under intense heating produced by electron gun, plasmotron or electric current. But the power levels needed for ADTT are much higher so the thermodynamics and technology of liquid lead and lead-bismuth targets for electronuclear transmutation systems are to be investigated.

A proton beam target producing neutrons, and coupled with a multiplying blanket, is one of the key elements of electronuclear transmutation systems. If beam powers around 100 MW are needed, liquid lead or lead-bismuth is the only realistic possibility for the target material. Target cooling is done in a most natural way and a dividing window isolating the accelerator vacuum volume may even not be needed. The radiation resistance of a window and the necessity for its frequent replacement present a major problem. Target design is much simpler if the same coolant is used both in the target and in the blanket.

Main research directions:

- completion of development of the technical concept of a transmutation fuel cycle for fast lead cooled reactors (including ADTT option with a cyclic accelerator);
- thermo-hydraulic and technological validation of fast reactors with heavy liquid metal coolants (Pb and Pb-Bi);
- experimental studies of heat exchange in pin lattices with Re > 1000;
- interchannel mixing of coolant in the lattices;
- temperature variations on the periphery of the assemblies;
- development and testing of a computer code to calculate channel-by-channel flow rates;
- investigations of the hydrodynamical parameters of coolant flow (velocity, pressure and temperature distributions, velocity and pressure pulsations) in nominal and other regimes;
- coolant flow in the vessel, heat exchangers and steam generators;
- hydrodynamics of passive control systems (suspended rods);
- methods and means of coolant quality control and stabilization;
- conceptual design of liquid metal proton target;
- conceptual design of Pb-Bi cooled accelerator driven blanket;
thermohydraulics of the target circuits including ones with a free surface;
- forecast of target properties as functions of beam parameters;
- protection of ion tract from vapors, gases and aerosols for operation without dividing diaphragm or in the case of its damage and replacement;
- mechanism and kinetics of coolant evaporation and aerosol formation and of its penetration into the accelerator volume;
- thermodynamics of the impurities getting into the target circuit and generated there, of their interaction with the coolant and structure materials;
- technology of the coolant, stabilization of its properties and protection of equipment surfaces;
- development of the methods of thermal, electrolytic and membrane cleaning of the coolant from polonium, theoretical and experimental investigations of the temperature dependence of polonium volatility; comprehensive comparison of lead-bismuth and lead as liquid target materials, including radiology, thermohydraulics, corrosion, coolant technology equipment.

So heavy liquid metal coolants are in fact of dual purpose - for reactors and for accelerator targets - and the results on their physics and technology may be used in both areas.

One of the most important problems of developing high power accelerators is reliable cooling of their heat releasing parts (ion sources, high frequency generators, etc.) and thermal stabilization of resonator sections (beam focusing requires high thermal uniformity in spite of intense non-uniform heating). Highly efficient non-standard cooling systems are needed to ensure acceptable operation parameters and lifetime of the whole accelerator.

This experience and software form a good base for forecasting the thermohydraulic parameters of liquid metal targets including transients and emergency processes. Straightforward use of existing software is however somewhat difficult for the following reasons:

- due to the specific geometry of the target circuitry, for example, due to the presence of a hemispherical window which is difficult to describe in terms of standard reactor \((r,z)\) of \((r,z,\varphi)\) geometries;
- due to the presence of a free coolant surface in windowless targets unlike the traditional reactor case;
- due to heat release into the volume coolant itself.

Hence both modernization or rewriting of existing codes and accumulating of experience in numerical modeling of thermohydraulics of target circuits is necessary. The 2D dynamic code SIMAKS was developed for calculations of thermohydraulic parameters (velocities, pressures and temperatures) of axially symmetric liquid metal targets with a separating diaphragm in the ion beam and, particularly, for the designed 10 MW target. Navier-Stokes and thermal energy equations are solved for viscous incompressible fluid in a curvilinear orthogonal co-ordinate system, allowing accurate enough description of the diaphragm geometry as well as the geometry of hydrodynamical flow separators included in the design. The TRESOR code was developed in IPPE in 1991-1994 for calculations of turbulent flow and heat exchange in linear channels and this experience and some blocks of the code were used in the development of SIMAKS.

The method and code were tested numerically for the case of laminar flow in a cylindrical tube. The results agree well with the analytical solution.

Calculations have now been started of velocity and pressure fields for the design of a 10 MW target with hemispherical diaphragm. Significant difficulties arise in numerical modeling of the thermohydraulical parameters of a windowless, free coolant-surface target and in designing the target. Investigations were carried out to adapt the DINCOR code for solving such problems. A number of calculations of flow hydrodynamics were done for the target designed for a 100 MW power level.

Schematically this target is a cylindrical tank 1m in diameter and 1.2 m high with an inner drift tube. The inner tube, 320 mm in diameter, is placed along the axis of the cylinder. The tube
broadens funnel-like toward its end to a diameter of 550 mm. The height of this transition part is 500 mm. The coolant is fed to the funnel entrance of the inner tube (which also accepts a proton beam 150 mm in diameter), flows down and is then pumped upward through the annulus between the cylindrical tank and the inner tube.

The DINCOR code provides a numerical solution of the non-stationary equations of hydrodynamics and heat exchange in multi-component media with possible melting and freezing of the components and, in particular, it can calculate one-phase flow of the coolant in circuits with free surfaces.

Two versions of the DINCOR code are available:

a. A finite difference version;

b. A version using a "particles in a cell" method and allowing tracing of the movement of the media as represented by marker particles. An initial stationary state was postulated for the calculations (the coolant does not move, free surface and pressure correspond to an immobile liquid) and it was assumed that a pump with fixed pressure pumping the liquid through the annulus is switched on at the initial moment of time.

The dynamics of the process was calculated up to the moment of establishing of stationary flow corresponding to a definite form of free surface and constant coolant flow rate. Pump pressure, tubing geometry and coolant level in the target were varied in the calculations.

The calculations demonstrated in particular:

- version B of the DINCOR code is preferable because in version A the free surface gets artificially diffused in the numerical calculations;
- the form of the free surface and its stability against various perturbations depend on many factors, the most important ones being drift tube geometry and hydraulic resistance of the heat exchanging loop;
- a lot of variants should be calculated to validate the geometry of the input section of the target.

3.3. REACTOR PHYSICS AND CORE DESIGN

Actinide containing fuels affect the neutronics of the burner reactor cores in many important respects, some of them safety related (coolant void coefficient, effective share of delayed neutrons, control rod worth, etc). All minor actinides absorb thermal neutrons intensely and their fission is usually a multi-stage process including formation of intermediate nuclides. The fission/capture ratios of MA are much better for fast reactors and the harder the spectrum the better, so special core designs and layouts comprise a considerable part of transmutation related R&D.

3.3.1. China

3.3.1.1. The study on feasibility of MA transmutation in CEFR

3.3.1.1.1. Introduction

CEFR (China Experimental Fast Reactor) is a 65 MW(th) (25 MW(e)) sodium cooled experimental fast reactor designed by the China Institute of Atomic Energy (CIAE) [73-74]. In order to develop the MA transmutation technique in China, it is necessary to consider the role of CEFR in MA transmutation. First, some irradiation tests can be performed on CEFR. For example, irradiation of MA containing samples or irradiation of special fuel pins with various contents of MA can be carried out. These tests can be used to study the properties of various MA containing fuel samples and fuel pins to validate theoretical work. The other role is that CEFR can be used for MA transmutation. Although the amount of MA transmuted is limited, it is still significant in scientific sense and calculational demonstration. The feasibility of MA transmutation has been studied.
Parameter survey calculations are performed for CEFR MA loaded core so as to investigate its basic nuclear characteristics. The comparison of nuclear characteristics between the reference core and a MA loaded core has been presented and the amount of MA transmutation in CEFR has been determined.

3.3.1.1.2. Calculational method

The basic constant set used in calculations is the 46 groups CLIB-IV. This 46 groups constant set is condensed to 6 groups constant set by the IDX code. Various nuclear parameters of the core are calculated by the two dimensional diffusion and burnup code 2DB.

3.3.1.1.3. CEFR reference core

The CEFR original core design is defined as the CEFR reference core. The main parameters of CEFR reference core are shown in Table II. The core layout is shown in Fig. 1.

3.3.1.1.4. CEFR MA loaded core

The MA fuel is assumed to come from PWR spent fuel which has been cooled for five years before reprocessing. The data on the composition of MA fuel are cited from a paper of T. Wakabayashi et al. (pp 99-108 of Ref. [168]) (see Table III).

The MA is homogeneously dispersed throughout the entire core. The ratio the amount of MA loaded in the fuel is 50% and the enrichment of $^{235}U$ is 90%. The nuclear characteristics of the CEFR MA loaded core are shown in Table IV and the reference core parameters are also listed for comparison.

As shown in Table IV, the reactivity worth of all control rods in a MA loaded core is smaller than that of the reference core. This is caused mainly by the decrease in the relative importance of the control rods due to the high cross-section of the MA fuel. Although the control rod worth decreases, the total worth is still sufficient for reactor control.

TABLE II. DESIGN PARAMETERS OF CEFR REFERENCE CORE

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Data</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal power (MW(th))</td>
<td>65</td>
</tr>
<tr>
<td>Gross electric output (MW(e))</td>
<td>25</td>
</tr>
<tr>
<td>Operation length (EFPD)</td>
<td>71</td>
</tr>
<tr>
<td>Number of fuel batch</td>
<td>3</td>
</tr>
<tr>
<td>Fuel material (first core)</td>
<td>UO$_2$</td>
</tr>
<tr>
<td>Enrichment of fuel</td>
<td>60%</td>
</tr>
<tr>
<td>Fuel pin diameter (cm)</td>
<td>0.6</td>
</tr>
<tr>
<td>Number of subassemblies</td>
<td></td>
</tr>
<tr>
<td>Fuel subassemblies</td>
<td>82</td>
</tr>
<tr>
<td>Control rods</td>
<td>8</td>
</tr>
<tr>
<td>Core height (cm)</td>
<td>45</td>
</tr>
<tr>
<td>Equivalent core diameter (cm)</td>
<td>58.5</td>
</tr>
<tr>
<td>Neutron absorber</td>
<td>B$_4$C</td>
</tr>
<tr>
<td>B-10 enrichment in B$_4$C</td>
<td>91% (at)</td>
</tr>
<tr>
<td>Assembly lattice pitch (cm)</td>
<td>5.85</td>
</tr>
<tr>
<td>Volume fraction</td>
<td></td>
</tr>
<tr>
<td>Fuel</td>
<td>0.381</td>
</tr>
<tr>
<td>Sodium</td>
<td>0.207</td>
</tr>
<tr>
<td>Structure</td>
<td>0.397</td>
</tr>
<tr>
<td>Gap</td>
<td>0.015</td>
</tr>
</tbody>
</table>
FIG. 1. Core layout of CEFR.
The decrease of $\beta_{\text{eff}}$ in the MA loaded core is due to the small value of the delayed neutron yield of MA. The decrease in sodium void effect in MA loaded core is caused by neutron spectrum hardening which results in an increase in the fission cross-sections of MA. The Doppler effect of the MA loaded core is smaller than that of the reference core. It is caused by the decrease of $^{238}\text{U}$ concentration. The burnup reactivity loss per cycle for the MA loaded core is smaller than that of reference core due to the production of $^{238}\text{Pu}$ from $^{237}\text{Np}$. It is advantageous in that it extends the reactor operation period and increases the fuel burnup.

Although the parameters related to reactor safety of the MA loaded core are not as good as those of the reference core, they are still acceptable from view point of reactor safety.
It can be seen from Table IV, that the MA transmutation rate in the MA loaded core is 9.4 kg/a. It is nearly equal to the MA production rate in the QINSHAN PWR nuclear plant (0.3 GW(e)). Therefore a CEFR MA loaded core has the capability to transmute the long lived radiotoxic nuclides produced in the QINSHAN nuclear power plant. CEFR is an experimental fast reactor, the thermal power is limited (65 MW(th)). Although the amount of MA transmutation in the CEFR MA loaded core is limited, it is still significant in a scientific sense and as a calculational demonstration.

CEFR also has its significant role in MA fuel irradiation test and MA transmutation technique studies, for instance, irradiation tests of samples containing MA and special fuel pins with various contents of MA. These tests can be used to study the properties of various MA samples and fuel pins and for the production of data to compare with theory.

3.3.2. Republic of Korea

3.3.2.1. Analysis of transmutation rates of long lived radionuclides in the Korea multi-purpose research reactor

3.3.2.1.1. Introduction

Several types of reactors and/or accelerators are being suggested as practical transmutation facilities in several countries. For example, as an actinide burner, both a hybrid system of a subcritical reactor combined with an accelerator, and conventional power reactors are being considered for nuclear transmutation. However, the best way of transmutation is yet to be determined. Both economic and technical advantages or disadvantages should be taken into account to select the most suitable way of carrying out transmutation.

This study is focused on the estimation of long lived radionuclide transmutation rates in the research reactor, KMRR which will be decommissioned early next year. Since the neutron flux in the core of KMRR is higher than those of conventional PWR's, it may provide more favorable conditions for the transmutation of Minor Actinides which have lower fission cross sections than those of fissile uranium and plutonium.

The computer code, TRIFON, developed in the Russian Institute of Theoretical and Experimental Physics (ITEP) was used to estimate the transmutation rates of MA and long lived fission products such as $^{237}$Np, a mixture of Am and Cm, $^{99}$Tc and $^{129}$I.

The fission products of $^{99}$Tc and $^{129}$I were assumed to be placed in the central flux trap. The minor actinides of $^{237}$Np and a mixture of Am and Cm were assumed to be either placed in the central flux trap or partially mixed up into six central fuel elements among in a 36-element assembly. Both cases were assumed to be irradiated during the period of six fuel cycles (203 days).

3.3.2.1.2. Reactor core and irradiation thimbles of the KMRR [75]

The reactor physics design was carried out to achieve the maximum neutron flux with the maximum available fuel enrichment, 20% LEU and proper heat removal. The core features a symbiosis of light water reactor lattices as well as heavy water reactor lattices. This combination provides extensive variety and flexibility of neutron quality in terms of its energy and spatial distribution. Table V shows the design concept of the KMRR core.

As shown in Fig. 2, the inner core, an assembly of light water reactor lattices, is enclosed by the corrugated parallelepiped inner shell of the reflector tank. Eight out of 31 sites are assigned for positioning 4 control absorbers and 4 shut off shrouds. Each of them has an 18-element fuel assembly and circular flow tube. The nominal core configuration allows 20 hexagonal flow tubes to accept 36-element fuel assemblies, and 3 vacant sites are reserved for capsule, rig, or loop installations.
### TABLE V. MAJOR DESIGN CONCEPT OF KMRR

<table>
<thead>
<tr>
<th>Reactor type</th>
<th>Open-tank-in-pool, LWR/HWR hybrid</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal power</td>
<td>30 MW(th)</td>
</tr>
<tr>
<td>Fuel</td>
<td>U₈Si-Al, 19.75 W/O 235U enriched</td>
</tr>
<tr>
<td>Coolant</td>
<td>H₂O</td>
</tr>
<tr>
<td>Moderator</td>
<td>H₂O/D₂O</td>
</tr>
<tr>
<td>Reflector</td>
<td>D₂O</td>
</tr>
<tr>
<td>Cooling method</td>
<td>Convective up-flow</td>
</tr>
<tr>
<td>Secondary cooling</td>
<td>Cooling tower</td>
</tr>
<tr>
<td>Reactor building</td>
<td>Confinement</td>
</tr>
<tr>
<td>Power regulation and</td>
<td>Four hafnium tubes sliding over the 18-element fuel assembly</td>
</tr>
<tr>
<td>Xenon poison compensation</td>
<td>Four hafnium tubes sliding over the 18-element fuel assembly. Dropped by gravity</td>
</tr>
<tr>
<td>Shutdown system</td>
<td></td>
</tr>
</tbody>
</table>

| Heat generated in reactor fuel   | 27.5 MW                                           |
| Heat generated in moderator and structure | Approx. 2.5 MW                                     |
| Total fission power              | 30.0 MW                                           |
| heat removed by primary coolant system | 27.5 MW                                           |

**FIG. 2. Schematic diagram of the KMRR core.**
The outer core has 8 circular flow tubes vertically passing through the reflector tank on the narrow sides of the inner core periphery. Loaded with 18-element fuel assemblies, the H2O cooled and D2O moderated outer core enhances total core excess reactivity. The outer core sites, used for irradiation purpose, provide high epithermal neutron fluence.

The central trap, one of the three flux traps in the inner core, has the highest flux, $5.3 \times 10^{14}$ n/cm$^2$s. It will be used to install the fuel and material test loop. The D2O filled reflector tank is equipped with a number of vertical irradiation thimbles. Their sizes and locations are optimized to maintain the required neutron quality and level without significant disturbance by changes in reactor operating condition and by other nearby experiments.

One fuel cycle consists of 28 days of burning and 7 days of shutdown which is necessary for fuel exchange. Most of the 36-element assemblies and all of the 18-element assemblies are burned up for 6 fuel cycles, but some of the 36-element assemblies are used for 7 fuel cycles. Material and specifications of KMRR standard fuel are shown in Table VI.

### TABLE VI. SPECIFICATIONS OF KMRR STANDARD FUEL

<table>
<thead>
<tr>
<th>Composition ratios in fuel meat (W/o)</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}$U</td>
<td>11.7</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>46.9</td>
</tr>
<tr>
<td>Si</td>
<td>2.4</td>
</tr>
<tr>
<td>Al</td>
<td>39.0</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Fuel meat</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>enrichment (w/o)</td>
<td>19.75</td>
</tr>
<tr>
<td>density (g/cc)</td>
<td>5.4</td>
</tr>
<tr>
<td>diameter (mm)</td>
<td>6.35/5.5</td>
</tr>
<tr>
<td>length (mm)</td>
<td>700.0</td>
</tr>
<tr>
<td>mass of uranium (g)</td>
<td>69.8/52.4</td>
</tr>
<tr>
<td>LEU density (g/cc)</td>
<td>3.15</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cladding</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>material</td>
<td>Al (co-extruded)</td>
</tr>
<tr>
<td>thickness mm</td>
<td>0.76/1.19</td>
</tr>
<tr>
<td>Fin</td>
<td>8</td>
</tr>
<tr>
<td>number</td>
<td>8</td>
</tr>
<tr>
<td>height (mm)</td>
<td>1.02</td>
</tr>
<tr>
<td>width (mm)</td>
<td>0.76</td>
</tr>
</tbody>
</table>

3.3.2.1.3. Trifon code [76]

The one-dimensional code TRIFON, which is based on the collision probability method, was developed in ITEP, Russia. The neutron energy spectrum and the spatial distribution of the neutron flux can be computed with this model and transmutation rates of various nuclides can also be estimated. The cross-section data used in this code, are mainly from the Russian ABBN and also partly from other additional information. They are composed of 26 group cross-section data and resonance parameters.

ITEP validated this code by comparing the calculation results from other codes, as shown in Table VII [77]. The results of this code are closer to the results of Monte Carlo simulations than that of the WIMS code. Also, the quantities of uranium and transuranium contained in LWR spent fuel
TABLE VII. COMPARISON OF THE PARAMETER VALUES CALCULATED FROM TRIFON AND OTHER CODES

<table>
<thead>
<tr>
<th>Lattice</th>
<th>Parameter</th>
<th>TRIFON</th>
<th>WIMS</th>
<th>EPRI-CELL</th>
<th>Monte-Carlo (%SD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NB-1, low enrichment</td>
<td>$K_-$</td>
<td>1.1344</td>
<td>1.254</td>
<td>1.1449</td>
<td>1.1471(0.14)</td>
</tr>
<tr>
<td></td>
<td>$\rho_{28}$</td>
<td>1.375</td>
<td>1.458</td>
<td>1.360</td>
<td>1.363(0.6)</td>
</tr>
<tr>
<td></td>
<td>CR</td>
<td>0.804</td>
<td>0.829</td>
<td>0.796</td>
<td>0.798(0.4)</td>
</tr>
<tr>
<td>NB-2, (PU/U=0.02)</td>
<td>$K_-$</td>
<td>1.1734</td>
<td>1.1640</td>
<td>1.1698</td>
<td>1.1748</td>
</tr>
<tr>
<td></td>
<td>$\rho_{28}$</td>
<td>2.526</td>
<td>2.372</td>
<td>2.613</td>
<td>2.612</td>
</tr>
<tr>
<td></td>
<td>CR</td>
<td>2.110</td>
<td>2.319</td>
<td>2.148</td>
<td>2.148</td>
</tr>
<tr>
<td>NB-4, PWR type</td>
<td>$K_-$</td>
<td>1.3363</td>
<td>-</td>
<td>1.3415</td>
<td>1.3424(0.26)</td>
</tr>
<tr>
<td></td>
<td>$\rho_{28}$</td>
<td>2.666</td>
<td>-</td>
<td>2.632</td>
<td>2.654(0.6)</td>
</tr>
<tr>
<td></td>
<td>CR</td>
<td>0.556</td>
<td>-</td>
<td>0.548</td>
<td>0.549(0.4)</td>
</tr>
<tr>
<td></td>
<td>$K_-$</td>
<td>1.1302</td>
<td>-</td>
<td>1.1421</td>
<td>1.1456(0.15)</td>
</tr>
<tr>
<td>NB-5, hard spectrum</td>
<td>$\rho_{28}$</td>
<td>8.452</td>
<td>-</td>
<td>8.534</td>
<td>8.503(0.4)</td>
</tr>
<tr>
<td></td>
<td>CR</td>
<td>1.015</td>
<td>-</td>
<td>1.006</td>
<td>1.006(0.3)</td>
</tr>
</tbody>
</table>

FIG. 3. Comparison of the experimental and calculated results for the amount of $^{239}$Pu with burnup in a PWR.

were also calculated with this code. The comparison of the calculated results with experimental results is shown in Fig. 3. The agreement between the calculated and experimental results is quite good. On the basis of this demonstration, ITEP carried out transmutation studies with this code for the heavy water reactor, where high enriched uranium is used as fuel, as well as the thorium fuel cycle reactor [78, 79].
3.3.2.1.4. Transmutation rate analysis in the KMRR

**Calculational model**

For the convenience of calculation, in the transmutation study with the central irradiation thimble, the shapes of the hexagonal thimble, fuel assembly tubes, and surrounding moderator were assumed to be annular rings with corresponding equivalent volumes. The schematic of the cell model reflected by these assumptions is illustrated in Fig. 4. The net current was assumed to be zero at the outer boundary of the six assemblies around the central trap in the core.

In the transmutation study with minor actinides partially mixed up into six central fuel elements out of the 36-element fuel assembly as shown in Fig. 5, the central support rod, moderator, cladding, structure, and MA and fuel were also assumed to be annular rings with corresponding equivalent volumes in the order of their locations.

**Calculations and results**

The transmutation rate of $^{237}$Np with burning time calculated by the TRIFON code was compared, in Fig. 6, with that obtained from the WIMS-KAERI code [80] which had been used in the design of KMRR. The two results were found to be in accordance within 7% error.

Small quantities of long lived radionuclides were considered to be loaded as the target to minimize the impact on core behavior. The atomic number density of a target nuclide is fixed as 0.0001/cm barn. Since the code was developed on the basis of one dimension, the thermal power was converted to the power per unit length. As noted in the previous section, one fuel cycle is composed of 28 days burning and 7 days shutdown in the KMRR. The thermal power during the irradiation period of six fuel cycles is assumed as $30 \times (168/203) f70 = 0.355$ MW/cm.

Figure 7 shows the transmutation rate of small quantity of $^{237}$Np in the central flux trap. The transmutation rate was estimated as 64.6% and the net burning rate, offset by the production of new transuranium from the Np target was 34.5%.

Figure 8 illustrates the transmutation rates of a mixture of Am and Cm in the central flux trap with burning time when loaded in the same ratio as in typical PWR spent fuel at 10 year cooling time. The amount of $^{241}$Am as well as $^{243}$Am was found to decrease with time whereas that of total Cm nuclides increases due to the transmutation of Am into Cm by neutron capture and then beta-decay during the irradiation. Consequently, the total net burning rate of Am and Cm was 27.3% during the irradiation period of six fuel cycles.

On the other hand, the transmutation rates of $^{99}$Tc and $^{129}$I loading in the central flux trap were 13% and 9.5%, respectively, as shown in Fig. 9. They were lower than those of minor actinides due to the lower neutron cross-sections of $^{99}$Tc and $^{129}$I.

In the cases of partially mixing up $^{237}$Np or a mixture of Am-Cm into six central fuel elements out of the 36-element assembly of KMRR fuel, the transmutation rate of $^{237}$Np is 26% for 17.5 g replacement and 20% for 52.4 g replacement as shown in Fig. 10. The rate of $^{241}$Am is 46% for 17.5 g replacement and 26% for 52.4 g replacement as shown in Fig. 11. As the amounts of test materials inserted in the fuel assembly are increased, there is strong neutron capture in the test materials and the transmutation rates in such cases are decreased.

The estimated transmutation rates are much lower than those from the transmutation in the irradiation thimble because of the 4-5 times lower neutron flux.

So it was found that transmutation of minor actinides in the KMRR can be achieved to some extent, showing the possibility for using KMRR as a facility for a transmutation test. The calculation results obtained from this study can also be used as basic information for target design or transmutation test.

FIG. 5. Cell calculation model of 36-element assembly with partly replacement of fuel by MA.
FIG. 6. Comparison of the calculated results of TRIFON and WIMS codes for transmutation rate of Np-237.

FIG. 7. The changes in the amounts of Np-237 and total TRU with burning time.

FIG. 8. The changes in the amounts of Am-Cm and total TRU with burning time.

FIG. 9. Transmutation rates of Tc-99 and I-129 with burning time.

FIG. 11. Transmutation of Am-Cm with loading quantity in the case of replacement of fuel.
3.3.2.2. Feasibility study on transmutation of minor actinides in conventional fission power reactors

Several types of reactors and/or accelerators are being suggested as practical transmutation systems in several countries. For example the actinide burner, hybrid system of a subcritical reactor combined with an accelerator or conventional power reactors are being considered for nuclear transmutation. However, the best way of transmutation is yet to be determined. Both economic and technical aspects should be taken into account to select the most suitable way of transmutation. This study was focused on the estimation of MA transmutation rates in the conventional PWR, Yonggwang 2 which was commissioned in 1989.

The CASMO-3 code used for assembly burnup calculations in this study is a multigroup 2-dimensional transport theory code. This code handles a geometry consisting of cylindrical fuel rods of varying composition in a square pitch array. CASMO-3 has the capability to handle four BWR bundle and PWR colorset cases. The Code NEM-3D used for the burnup calculation of the whole reactor core is a multidimensional burnup code based on the nodal expansion method. Main outputs from this code are the temperature distributions of moderator and fuel and the distributions of neutron flux and burnup. The code can also calculate the normal power distribution along the radial and axial axes.

3.3.2.2.1. Method

In the transmutation analysis with MOX fuel we performed the color set calculation. The color set consists of 1 MOX fuel assembly and 3 UO₂ assemblies. In order to avoid power peaking in the surrounding UO₂ assemblies, we adopted the loading pattern of MOX fuel as shown in Fig. 12. The plutonium concentration in the MOX fuel was determined from Siemens' MOX fuel design data [81].

In the transmutation analysis with UO₂ fuel minor actinides were assumed to be located uniformly in the assembly. The enrichment of ²³⁵U was 3.5 w% for this study. The procedure for the transmutation calculation is shown in Fig. 13.

3.3.2.2.2. Results and discussion

Minor actinides were assumed to be loaded in cycles 4, 5, and 6 in the Yonggwang unit 2 [82]. The transmutation rates of these isotopes in the MOX assembly and UO₂ assembly are shown in Table VIII. To reduce the uncertainty of the results, microdepletion calculations for MA isotopes should be further carried out. The optimized loading pattern in the MOX assembly should be investigated for future transmutation analysis.

It was found for a more correct transmutation calculation in a PWR that we need to develop the global core calculation code, more reliable cross-section data and more accurate decay chains for MA isotopes.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>UO₂ + MA (1.0 w/o)</th>
<th>MOX + MA (1.0 w/o)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isotope</td>
<td>EOC(g) cycle 4</td>
<td>EOC(g) cycle 6</td>
</tr>
<tr>
<td>Am-241</td>
<td>733</td>
<td>59</td>
</tr>
<tr>
<td>Am-243</td>
<td>356</td>
<td>136</td>
</tr>
<tr>
<td>Cm-244</td>
<td>101</td>
<td>248</td>
</tr>
<tr>
<td>Total</td>
<td>1190</td>
<td>443</td>
</tr>
</tbody>
</table>
FIG. 12. Loading pattern of the MOX fuel assembly.
FIG. 13. Computer program system of 3D PWR simulation for transmutation analysis.
3.3.3. Russian Federation

3.3.3.1. Actinide behavior in the reactors of different types

Thermal reactors will be the basis of nuclear power for 3-4 decades, with corresponding research and development taking a proper share of effort and funds. The next priority level is fast reactors. The task of more efficient utilization of natural uranium resources is postponed, not cancelled. The accumulation of reactor and weapon grade plutonium calls for the development of methods for its utilization. Burning of transuranics (minor actinides) formed in spent fuel is one of possible ways to manage high level radwaste. Fast reactors promise solutions of all these problems. Transmutation of plutonium and MA provides additional incentives for the development of fast reactors. They are real, industrial scale facilities with about a hundred reactor-years of experience behind them. They belong to the present just like thermal reactors. The material in Section 3.3.3 is based mainly on publications [83-97].

The distinction between U-Pu and minor actinides is only a chemical classification. Nuclear properties of even Pu isotopes make them look more like MA. The evolution of Pu isotopic composition (often called the "plutonium vector") during recycling and storage is in fact degradation. Even isotopes (neutron absorbers) accumulate in recycling, so weapon grade Pu, almost pure fissile \(^{239}\text{Pu}\), potentially may be used to enrich Pu mixtures the way \(^{235}\text{U}\) enriches natural uranium. Short lived \(^{241}\text{Pu}\) (14 year half-life) decays into \(^{241}\text{Am}\) during storage turning an excellent fuel isotope into an absorber, a hard gamma and alpha emitter, and complicating all recycling operations considerably. So transmutation is a multiparametric problem and its optimization is a very complex and challenging task.

First proposals on neutron transmutation were reactor based. Most MA are threshold fission nuclides and their fission to capture ratios are much more than unity at neutron energies around 1 MeV, so fast reactors were the obvious choice. But it soon became clear that averaging over the real neutron spectrum of a fast reactor results in fission to capture ratios 10-20 times larger than for standard LWR but still considerably less than unity. For the most important isotope, \(^{237}\text{Np}\) corresponding figures are: 0.02 (LWR), 0.2 (standard fast reactor, average neutron energy 400 keV), 0.7 (specialized fast neutron burner, average neutron energy 700 keV). So dedicated burners must have as hard a neutron spectrum as possible and that is an important task for designers.

The case of \(^{237}\text{Np}\) deserves special attention and first a general remark is needed. The terms "transmutation" and "burning" were used as synonyms until recently. But when they are applied to actinides there is an important distinction worth formalizing. "Transmutation" generally means any change in nucleonic composition of the affected nucleus, preferably leading to stable or short lived isotopes. Burning means fission of actinide nuclides, turning them into fission products. Results of an actinide transmutation scheme should be estimated by its burning efficiency.

A \(^{237}\text{Np}\) nucleus, absorbing a neutron, turns into \(^{238}\text{Np}\) which may either fission or decay rapidly into \(^{238}\text{Pu}\). The last event may hardly be considered a satisfactory result. First, it produces a very radioactive, heat releasing, relatively short lived alpha emitter (period 87.7 years) which is very inconvenient for both recycling and intermediate storage. Moreover, \(^{238}\text{Pu}\) decays into \(^{234}\text{U}\). Its period (245,000 years) results in a combination of specific activity and longevity which is a serious problem for final disposal. In addition neptunium has the largest share of the mass of the minor actinides. All that makes neptunium a touchstone in assessing the efficiency of transmutation facilities which is recognized in Russia. In addition, pure neptunium has a reasonably low critical mass and low specific radioactivity (0.7 Ci/kg). Those factors make neptunium a non-proliferation issue.

The considerations mentioned above are illustrated quantitatively by Figs 14 and 15 presenting different aspects of the time evolution of a unit mass (1 kg) of neptunium in different neutron spectra corresponding to operating Russian reactors:
FIG. 14. Total actinide mass $M$ as a function of residual mass of $^{237}$Np in the process of burning of 1 kg of Np for 10 000 days in various neutron spectra corresponding to different types of Russian reactors.

FIG. 15. Number of neutrons spent per one actinide fission in the same process as in Fig. 14.
- PWR of WWER-1000 type (thermal neutron flux $F = 4 \times 10^{14}$ neutrons/s cm$^2$);
- fast reactor BN-600 (the same as future BN-800, $F = 6 \times 10^6$ neutrons/s cm$^2$);
- research fast reactor BOR-60 (spectrum slightly harder than that of BN type reactors, $F = 3.7 \times 10^{15}$ neutrons/cm$^2$).

Let us designate the $^{237}$Np untransmuted mass as $m$ and the total mass of actinide products from the evolution of 1 initial kg of neptunium as $M$. Both quantities are functions of the time $t$ spent in the corresponding neutron flux: $m = m(t)$; $M = M(t)$. Excluding time $t$ from these two parametric equations we get a functional relation between two observables: $M = M(m)$. Such relations for the three reactor spectra mentioned and for a continuous 10 000 days in the core are given in Fig. 14. A straight line $M = m$ corresponds to an "ideal burner" - no radiation captures by actinides, fissions only, so transmutation means pure burning. The closer to this line is a "mass trajectory" $M(m)$ the more efficient is the actinide transmutation. Fig. 14 clearly illustrates the role of spectrum hardness in increasing this efficiency. It may be also expressed in terms of neutron economy which is done in Fig. 15 where the number of neutrons spent to induce a single fission is plotted again as a function of $M$ for the same three reactors.

The example of $^{238}$Pu illustrates the fact that from the point of view of final disposal there is no such thing as "short lived transuranics". Every nucleus heavier than uranium is a starting point of a decay chain and in the process of decay emits 6 to 9 alpha particles and 5 to 7 beta particles with varying rates. $^{238}$Pu decays practically completely in a thousand years but its daughter nucleus $^{234}$U in equilibrium with its shorter lived decay products stays dangerous for hundreds of thousands years with resulting a non-monotonous time dependence of radiotoxicity (see Fig. 16).

The two largest Russian research institutions investigating actinide transmutation in BN reactors are IPPE (Obninsk) and RIAR (Dimitrovgrad). The latest achievements and plans of IPPE in the field are shortly summarized below. The main activity in RIAR is described in Section 3.6.3.2.
3.3.3.2. IPPE activities

The Institute has a few laboratories specializing in the development and use of computer codes and nuclear constants sets for reactor calculations, a computer division and a Nuclear Data Center (a part of the world wide system of such Centers).

The experimental base includes:
- a few critical assemblies (BFS-1, BFS-2, KOBRA) for modeling the cores of full scale power fast reactors and for improvement of nuclear constants;
- research fast reactor BR-10 (10 MW(th) for technological studies under irradiation;
- six electrostatic accelerators of protons and other ions in the energy range 0.3-15 MeV for measuring cross-sections and spectra of various nuclear reactions;
- more than a hundred technological experimental facilities for thermohydraulics studies of the cores and heat exchanging equipment using most of the known liquid metal coolants and water;
- a complex of hot cells for material studies and radiation sold state physics;

Extensive calculations have been started on the neutronics of the prospective burners with three main directions of investigations:
- elimination of fertile blankets;
- increasing of enrichment;
- use of a new special fuel with inert matrix and without $^{238}U$.

The results demonstrate that even use of well studied MOX fuel may lead to efficient burning of Pu and MA. For example a 1 GWe reactor with a load factor of 0.8 can burn 500 kg of Pu annually if the enrichment is as high as 45 per cent. If $^{238}U$ is replaced by inert matrix the figure will be 750 kg.

Basic instruments are prepared for experiments on critical assemblies, first of all fission chambers with coatings of $^{237}Np$, $^{238,242}Pu$, $^{241}Am$, $^{243}Am$, $^{244}Cm$, then track detectors (glasses) for the same isotopes. The chambers were calibrated together with French ones in the experiments on the critical assembly "Harmony" (Cadarache) with an extremely hard spectrum.

These chambers and track detectors were used in critical assembly experiments aimed at improving the accuracy of MA neutron cross-sections for various reactor spectra.

Chamber experiments are accompanied by reactivity perturbation measurements with small specimens of MA (dozens of grams). The aim of the experiments is to improve the accuracy of fissioning and capture cross-sections by measuring central reactivity coefficients. Up to now the measurements have been made for $^{237}Np$, $^{239}Pu$, $^{241}Am$ for a series of assemblies (about 15) with different spectra. Experiments of the same type were done in Japan. Considerable discrepancy for $^{240}Pu$ was observed and the reasons are now being investigated.

Most detailed information including not only fission and capture cross-section but also inelastic scattering will be obtained in macroexperiments on critical assemblies with large amounts of $^{237}Np$ which are now under way and first results have been obtained. Besides improvement in neutron cross-sections such experiments will provide direct information on real reactor parameters (sodium void coefficients, power density, beta-eff, control rod efficiency, reactivity swing per cycle etc.) both for homogeneous and heterogeneous burner-reactors. From 50 to 100 kg of neptunium dioxide are needed for full scale experiments of this type. The Institute now has about 10 kg allowing the first stage of the experiments on BFS assembly to begin.

Along with critical assembly measurements, the experiments with capillary specimens of MA are under way in the BN-350 reactor. Their aim is the improvement of cross-section accuracy by tracing changes of isotope compositions. Specimens of $^{237}Np$, $^{238,240}Pu$ and $^{241}Am$ have already been irradiated and are now to be transported to IPPE for analyses. Some are still being irradiated. BN-350 irradiations will enable not only physical but radiochemical investigations as well.
There are concrete plans for using BN-800 reactor operation to reduce by transmutation the radiotoxicity of Pu and MA accumulated in reprocessing of WWER-440 reactor spent fuel and corresponding calculations were done. The main results are reproduced below. Evaluated volumes of actinides accumulated to the year 2000 are presented in Table IX.

TABLE IX. ACTINIDES ACCUMULATION IN WWER-440 REPROCESSED SPENT FUEL BY THE YEAR 2000

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Mass, kg</th>
<th>Nuclide</th>
<th>Mass, kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}\text{Pu}$</td>
<td>165</td>
<td>$^{241}\text{Am}$</td>
<td>420</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>11564</td>
<td>$^{243}\text{Am}$</td>
<td>146</td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>4173</td>
<td>$^{237}\text{Np}$</td>
<td>550</td>
</tr>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>2350</td>
<td>$^{244}\text{Cm}$</td>
<td>41</td>
</tr>
<tr>
<td>$^{242}\text{Pu}$</td>
<td>779</td>
<td>$^{245}\text{Cm}$</td>
<td>2</td>
</tr>
</tbody>
</table>

TABLE X. THE MASS OF RADIONUCLIDES ON THE SITE

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Mass at 2000, kg</th>
<th>Radionuclide mass at 2060, kg, including the mass gone to waste (brackets)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Scenario number:</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>$^{237}\text{Np}$</td>
<td>550</td>
<td>720</td>
</tr>
<tr>
<td>$^{241}\text{Am}$</td>
<td>420</td>
<td>2450</td>
</tr>
<tr>
<td>$^{243}\text{Am}$</td>
<td>150</td>
<td>150</td>
</tr>
<tr>
<td>$^{244}\text{Cm}$</td>
<td>40</td>
<td>5</td>
</tr>
<tr>
<td>$^{238}\text{Pu}$</td>
<td>165</td>
<td>110</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>17600</td>
<td>17600</td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>4370</td>
<td>4400</td>
</tr>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>2350</td>
<td>165</td>
</tr>
</tbody>
</table>

* At losses = 2% for Np and Cm, losses = 0.2% for Pu and Am.

TABLE XI. TOTAL RADIOTOXICITY INDICES OF THE ACTINIDES AFTER 1000 YEARS STORAGE IN DIFFERENT SCENARIOS (THE UNIT IS $10^3$ m$^3$ OF WATER NEEDED TO DILUTE THE MASS OF A NUCLIDE TO DRINKING STANDARDS)

<table>
<thead>
<tr>
<th>Element</th>
<th>Scenario</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Np</td>
<td>.03</td>
</tr>
<tr>
<td>Pu</td>
<td>100</td>
</tr>
<tr>
<td>Am</td>
<td>170</td>
</tr>
<tr>
<td>Cm</td>
<td>0.07</td>
</tr>
<tr>
<td>Total</td>
<td>270</td>
</tr>
</tbody>
</table>

* At losses = 2% for Np and Cm, losses = 0.2% for Pu and Am.
Three situations are considered:

1. BN-800 is not put into operation, the actinides are stored to 2060.
2. BN-800 reactor is put into operation at 2000 and burns traditional MOX fuel without MA.
3. BN-800 is put into operation at 2000 and uses MOX fuel containing MA.

Irretrievable actinide losses are taken fixed and equal to 2 per cent, but the results are also presented for a case when this key parameter is improved to 0.2 per cent for Pu and Am.

The resulting data on the radionuclide inventory and their total radiotoxicity at the year 2060 for the three scenarios considered are presented in Tables X and XI. It is obvious that fast reactor operation reduces significantly the volume and toxicity of the actinides going to the waste.

The possibility of using fast reactors cooled by molten heavy metal for actinide transmutation and general reduction of the radiotoxicity of a closed NFC has attracted attention recently. Calculations and analyses were made on the conceptual design of the BRUS-150 reactor cooled by Pb-Bi eutectics which would burn 100 kg of actinides in oxide form from three WWER-1000 reactors and produce about 90 kg of $^{233}$U annually, one third of it with less than 10 ppm of $^{232}$U, the rest with 3000 ppm. The quality of $^{233}$U produced may be drastically improved by placing a steel layer between the core and the thorium blanket.

Planned thermal power of the reactor is 500 MW, FAs campaign - 3 years. Reactivity variation during the campaign is small: -0.28 per cent. One of the limiting factors is the concentration of $^{236}$Pu in the Pu composition to be reprocessed - 7.2 per cent which is higher than the 5 per cent limit determined by decay heating. Observing this limit reduces the actinide burning capacity two-fold which still allows BRUS-150 to support a PWR capacity almost ten times higher than its own thermal power.

Main directions of research in the near future:

- development of the computer codes for reactor calculations taking into account the complete set of nuclides: actinides, activation and fission products.
- designing of oxide fuel cores for BN-600 and BN-800 reactors for efficient burning of transuranics including the cores without breeding blankets and with increased enrichment for utilization of reactor grade and weapon grade plutonium.
- development of new types of fuel - without $^{238}$U but with inert dilutants based on zirconium carbide, magnesia oxide, aluminum nitride.
- feasibility studies of using other fuel types (metal, nitride, carbide) for improving the technology and safety of burning transuranics.
- conceptual designs of cores without $^{238}$U with the emphases on safety (improved Doppler effect due to additions of resonance absorbers).
- experimental and numerical investigations of physical parameters of the core without $^{238}$U in a special core in the Large Physical Assembly (BFS).
- participation in the development of a specialized power actinide burner-reactor in the framework of the Russian-French co-operation on the EFR project.
- BFS experiments with the central part of the core containing 5-10 per cent of neptunium for improvement and verification of nuclear data and calculation methods and codes.
- conceptual and feasibility studies on transmutation of selected fission products in fast reactors.
- experimental and numerical investigations of fast reactor cores with steel blankets and boron carbide shielding layers.
- analyses of the possibility of applying existing and planned radiochemical technologies in fabrication and reprocessing of oxide fuel with very high plutonium enrichment (up to 50 per cent) and MA admixtures (up to 4 per cent) to be used in fast burner-reactors.
- preparations for testing fuel pins with zirconium carbide based fuel;
- verification and improvement of Pu group constants in integral experiments, evaluation of the accuracy of parameters of burner-reactors calculated in various nuclear data systems.
calculation of the radiation parameters of the fuel in various fuel cycle schemes;
verification of computer codes used for calculations of the evolution of isotopic compositions of "dirty" plutonium and minor actinides in the neutron spectra of a fast power reactor;
investigation of distribution of transuranics in the different stages of spent fuel reprocessing;
development and testing of the methods of radiochemical analyses of transuranic compositions;
study of dry reprocessing waste prepared for long term storage;
design of a plutonium burning core for BN-600;
conceptual design of a specialized sodium cooled transuranic burner-reactor;
thermohydraulic studies of sodium cooled burner-reactors:
determination of margins to boiling;
determination of temperature non-uniformities in fuel assemblies;
calculations of durability of thermally deformed elements;
numerical modeling of coolant boiling in accidents with power bursts or coolant flow failures.

3.3.3.3. RDIPE activities

A concept of a fuel cycle for future large scale nuclear power (with a one third share of the total primary energy production) was proposed by the RDIPE (NIKIET) team. The main points of the concept are:

- lead cooled fast reactor with UN-PuN fuel, breeding ratio about 1.05, without uranium blanket, total cost similar to a PWR;
- fuel enrichment using PWR discharge plutonium, weapon grade plutonium and slightly enriched uranium if necessary;
- concentration of fuel cycle facilities around NPPs;
- possible underground location of the reactors to protect them from the danger of external damage;
- electrochemical reprocessing with separation of U, Pu and the MA fraction not worse than 99.9 per cent followed by recycling, Cs and Sr separation at a 99 per cent level and their subsequent use on the site as heat and radiation sources, separation of 90 per cent of I and Tc with subsequent transmutation into stable isotopes, the rest of the FP bonded into porous matrices with concentrations up to $10^6$ Ci/1;
- 200 years intermediate storage for the FP with natural air cooling;
- subsequent vitrification and "radiationally equivalent" disposal to replace extracted radioactive ores;
- reduction of LLW and ILW volume, their concentration and returning to the NFC.

These proposals are part of a more general concept of radiation equivalent waste disposal also being developed in the Institute.

An open NFC based on Pb-Bi cooled fast reactors initially loaded by rather highly enriched (10-15 per cent) MOX or uranium fuel with subsequent feeding by slightly enriched, natural or even depleted uranium and probably thorium is proposed as a temporary concept for a transition period of a few decades before switching to a full scale closed NFC with multiple recycling. A high core breeding ratio is planned as well as high burn up and energy production per kg of refueled $^{235}$U (three times higher than in PWRs). It is in fact equivalent to "closing the nuclear fuel cycle inside the reactor" providing ample time for the development of safe and efficient reprocessing and refabrication schemes.

Fast reactor transmutation meets with rather serious problems. Radioactivity and heating of fresh MA containing fuels is many orders of magnitude higher than traditional uranium PWR fuel. That complicates all the operations with fresh fuel - fabrication, storage, transportation. A growing variety of fuel compositions is a common difficulty of all transmutation schemes. But the factor determining the success or failure of MA burning is the possibility to ensure low enough irretrievable losses of Pu and MA going to the waste. Partitioning of MA one from another as well as from U and Pu is not too important for fast reactors with their physics being benign to moderate variations in fuel composition.
The main problem of MA partitioning and burning in fast reactors seems to be the minimization of irretrievable losses and of the total volume of radioactive waste to ensure an acceptable economy of fuel cycles which include transmutation.

Among the innovative reactor concepts for transmutation, RDIPE proposals of molten fuel designs for actinide burning [87, 88] and FP transmutation [89] should be noted.

3.4. RADIOCHEMISTRY

Reprocessing of spent nuclear fuel undergoes significant and complicated changes if partitioning of neptunium, americium and curium from one another is needed as well as their separation from rare earths. Those processes and their input, intermediate and output products are the sources of major radiological risks, so radiochemical research is crucial to make transmutation schemes technically feasible and publicly acceptable.

3.4.1. India

Plants for the treatment of low and intermediate level wastes from the reprocessing plants are operational at the various sites in India. A waste immobilization plant for the treatment of high level waste (HLW) has been set up and is operational at Tarapur. It is a semi-continuous pot glass process involving calcination followed by melting in the processing vessels headed by an independently controlled multizone induction furnace. A solid storage and surveillance facility (SSSF) has also been set up for the interim storage of vitrified HLW prior to disposal at a permanent site. Two more waste immobilization plants are being set up at Trombay and at Kalpakkam, which will employ a Joule heated ceramic melter.

Research and development activities for the improvement of the existing processes and systems in waste management include actinide partitioning and separation of long lived fission products. Partitioning of long lived alpha emitting actinides from the HLW originating from fuel reprocessing operations is considered very useful from the point of view of reduction of alpha activities of the waste. A long term policy on the final utilization/transmutation of the partitioned actinides will be evolved at an appropriate time taking into consideration the evolving technologies on the different aspects of partitioning and transmutation (P&T) schemes.

To summarise, in the Indian context, reprocessing and closing the back end of the fuel cycle have become an industrial reality for full exploitation of the Indian uranium resources and waste disposal with long term safety. Considerable experience is being gained in Plutonium recycling in thermal as well as fast breeder reactors. India has assessed the various options in the light of her energy demands and waste disposal requirements and decided that reprocessing and closing of the fuel cycle is the only responsible approach with respect to future generations.

As per the present programme actinide separation is not included in any operational scheme. Our nuclear energy programme at present does not warrant the partitioning step. Presently only laboratory scale R&D is going on in the area of partitioning. Significant reduction in the alpha burden of these wastes would render the HLW much safer with respect to long term disposal.

For actinide extraction and partitioning from PUREX high level wastes, the TRUEX solvent, (CMPO) is one of the most promising reagents.

In India at the Bhabha Atomic Research Centre, solvent extraction and extraction chromatographic studies based on departmentally prepared CMPO are in progress to propose suitable extraction flowsheets for the removal of actinides from Purex high level waste concentrate (HLW) solutions.

The present day Purex process technology generally yields HLW solution mainly in nitrate form. However, the earlier Purex process utilised ferrous sulphamate as a reducing agent for
plutonium partitioning and this resulted in the generation of sulphate bearing HLW with low acidity. India is presently concerned with these two types of wastes.

Our initial approach is characterised by a prior extraction step with 30% TBP for the removal of residual uranium present in the waste followed by extraction with 0.2M CMPO/1.2 M TBP extractant mixture in dodecane.

From laboratory studies with actual HLW and simulated waste solutions, CMPO/TBP based flowsheets have been developed and the initial results indicate that near 100% recovery of alpha emitting actinides is possible from sulphate bearing HLW waste and also the HLW waste arising from PHWR fuel reprocessing.

Neptunium which generally exists as Np(V) in HLW has poor extractability in TBP and CMPO. But a change in its oxidation state to either (IV) or (VI) brought about by a suitable redox reagent would render it extractable in CMPO or TBP.

The CMPO based extraction chromatographic studies conducted with actual Purex HLW have also shown efficient uptake of actinides with near 100% recovery. This technique has a good potential for application in the removal of actinides from selected Purex waste streams.

Separation of minor actinides (Am and Cm) from the trivalent rare earths which accompany them in the above CMPO process is under investigation. PC-88A based extraction chromatographic techniques appear promising for this.

The secondary wastes generated in the above processes and the radiation stability of CMPO are also being assessed.

From a reprocessing angle, improvements in the Purex process to reduce Pu losses to the waste, recovery of neptunium and possibly $^{99}$Tc in the process are some of the aspects receiving attention. Alkaline trapping for $^{129}$I is also envisaged.

The advanced reprocessing cycle with additional P&T operations compared to conventional reprocessing facilities requires back fitting of additional facilities to existing plants. The facilities operating or under construction would require modifications to incorporate minor actinide partitioning and recovery flowsheets. This option will decrease considerably the alpha burden and the long term radiological hazards of active wastes leaving the plants. The extent of hazard reduction in HLW expected from the minor actinide partitioning flowsheet would depend on the fuel being reprocessed. At an appropriate time, a long term policy on the final utilisation/transmutation of the partitioned actinides will be evolved, based on the available state of the art technology at that point of time.

3.4.2. Russian Federation

Fuel recycling has a controversial influence on the various stages of the NFC. On the one hand, recovery and consumption of natural uranium is reduced drastically. If reprocessing plants are close to NPPs ("nuclear island" concept) then transportation of nuclear materials is minimized. The possibility arises of creating a full scale power industry almost without mining industry and fuel transportation. That is an obvious and important advantage. But on the other hand reprocessing and refabrication of fuel are much more complicated, expensive and hazardous than fresh fuel fabrication. An open cycle practically does not involve radiochemistry and associated hazards. This extremely important argument alone has induced some countries with a high level of nuclear power development (USA, Sweden, Germany) to drop recycling plans and concentrate on an open NFC. So development of reliable, safe and efficient methods of fuel refabrication is of crucial importance for recycling which is reflected in transmutation activities in Russia. Although aqueous technology is the basic method used in existing reprocessing facilities, more and more R&D attention is being paid to "dry" methods - electrochemistry and pyrometallurgy.
Material in this section is mainly based on publications [98-113].

The major part of the uranium mining industry of the former Soviet Union, a source of primary fuel material, was left outside Russia, but practically all radiochemical plants and research institutions are on Russian territory forming a good base for development and production of secondary fuel which is very important for transmutation projects. Many problems solved for defense purposes may now be used for fabrication of fuel containing minor actinides. Extensive and detailed have been obtained on the physical chemistry of uranium, plutonium, minor actinides and fission products in the following sectors:

- extraction radiochemistry;
- properties of dispersed systems;
- basic chemical reactions of actinides;
- hydrolytic properties of their compounds;
- complex forming reactions in aqueous and non-aqueous media;
- radiolysis and thermolysis of technological solutions;
- formation of actinide solid compounds, alloys, matrix materials;
- high temperature processes in molten salts, etc.

The main dilemma in planning a transmutation fuel cycle is the choice of the elements to be incorporated in the cycle (or, more correctly, of the isotopes, but isotope separation of highly radioactive mixtures is not considered as a serious option in most of the concepts although a few exceptions do exist).

The general strategies of managing medium lived (heat producing) and long lived nuclides are different and it is true also for transmutation. Because of the small thermal capture cross-section of \(^{90}\text{Sr}\) [104] its transmutation looks impossible which makes the transmutation of the rest of the medium lived fission products irrational. On the other hand large heat production and radiation damage to the matrices due to intense heating make immediate final disposal dangerous so it must be preceded by long term controlled storage, most probably for a few hundred years.

Among long lived fission products \(^{129}\text{I}\) presents a special problem due to its high mobility in the rocks and volatility in reprocessing. Its efficient transmutation in high flux reactors is in principle possible but solid compound targets may present problems. So \(^{129}\text{I}\) is a very important limiting factor in actinide recycling and burning. Its efficient capture and binding in reprocessing is a top priority. Most probable candidates for the transmutation list are minor actinides and technetium which has a large enough thermal capture cross-section, capturing up to three neutrons which turns it into stable isotopes of ruthenium and is a suitable material for reactor targets. This approach is adopted by both the main Russian radiochemistry laboratories involved in P&T research - Chlopin Radium Institute and Bochvar Institute of Inorganic Materials.

A first real step in closing the fuel cycle was done in 1976 when the RT-1 plant on the Production Association "Mayak" (PA "Mayak") site was put into operation. The plant regenerates spent fuel from WWER-440, BN-350 and BN-600 reactors of marine nuclear power units and research reactors. Its full capacity is 400 t a year which allows it to be used for regeneration of WWER-440 fuel from abroad before it was legally banned. More than 3000 t was reprocessed with regenerated uranium used as the secondary fuel.

A new RT-2 plant for WWER-1000 fuel reprocessing is in the belated process of construction near Krasnoyarsk. Its first stage capacity would be 1500 t/year. A pool type storage facility already built there contains more than a thousand tons of spent fuel.

It is planned to produce MOX fuel for fast reactors in "Shop 300" facilities also on the PA "Mayak" site but construction of the half completed plant is practically frozen. About 2000 MOX fuel pins were fabricated on a pilot plant and successfully used in BN-350 and BN-600 reactors with burnup to 10 per cent. Not a single pin was damaged. The technology of refabrication produces less
dust and aerosols than traditional technologies used in production of MOX fuel for PWRs in France, UK and Belgium.

The latest changes in the structure of the nuclear fuel cycle in Russia as described in Ref. [101] and calling for increased attention to the back end stages are:

- increased share of more expensive underground uranium mining;
- switching to centrifugal from diffusion separation which allows use of some previously depleted uranium;
- reprocessing of large volumes of depleted UF₆ ($^{235}$U concentration less than 0.01 per cent) into compact fluorneless products with regeneration of valuable fluorine agents;
- decommissioning of production reactors and old plutonium extracting plants which are hardly suitable for reprocessing of commercial fuel especially those with high transuranic contents;
- filling up of NPP spent fuel storage facilities;
- necessity to build new reprocessing plants, large stores for spent fuel and for final disposal of radwaste;
- increased share of regenerated uranium resulting in the hazards connected with $^{232}$U and $^{236}$Pu;
- plutonium coming from dismantled warheads;
- increase of volume and inventory of all kinds of radwaste including decommissioned equipment.

Intense investigations of reprocessing SNF of fast reactors started in BRIIM in the early seventies (see [109]). The progress of twenty years was recently reviewed in [112]. First industrial experiments were done in 1983 at the PA "Mayak" site where 28 FAs from the BN-600 reactor (burnup 5 per cent, cooling time 3 years) were reprocessed. The technological regeneration scheme included a nuclear-safe mixer-settler with 10 per cent solution of TBP in paraffin based dilutant. Losses of U and Pu were 0.01 and 0.1 per cent respectively, 3-5 per cent for Np. The decontamination factor of U from FP in two cycles was $10^6$.

As reported in [101] five full scale experiments on regeneration of MOX fuel irradiated in the fast reactors BR-10, BOR-60 and BN-350 were carried out in IPPE (in co-operation with BRIIM) in the eighties. Burnup varied from 1.5 to 10 per cent, cooling time - from 6 to 30 months. The main U-Pu extraction cascade contained 43 stages.

Three types of extractors for FR fuel reprocessing were developed and tested (time of contact between aqueous and organic phases indicated in the brackets): mixer-settlers (1 minute and more), pulsation columns (less than 1 min), centrifugal extractors (seconds). All three types demonstrated acceptable performance, major extraction coefficients being better than 99.99%, decontamination from fission products and uranium-plutonium separation coefficients are not less than $10^4$. Individual advantages and disadvantages of all three types are compared in detail in [112].

Composition of input solution: U-165 g/l; Pu-23 g/l (no more than 11 per cent of Pu-IV), beta-gamma activity - 6.5 Bk/l; alpha-activity $3.7 \times 10^{12}$ Bk/l; dose rate - 3960 mcrem/s l.

The following parameters were achieved:

- U extraction: 99.993 %
- Pu extraction: 99.996 %;
- Np extraction: up to 98 %.

The coefficients of decontamination from beta-gamma emitting fission products achieved were: U - $1.5 \times 10^5$; Pu - $4.1 \times 10^4$. The coefficients of actinide decontamination achieved were: U from Pu - 1100; Pu from U - from 40 to 660; U from Np - 11. The transplutonium and rare earth fraction is extracted by di-2-ethylhexil- phosphorous acid and by its zirconium salt.

The problem of extraction of the slightly radioactive valuable elements Pd and Rh from SNF is also being studied. Palladium concentrations in various spent fuels are: FR-MOX, 80 GW days/t -
4.7 kg/t; FR-MOX, 40 GW days/t - 3.5 kg/t; WWER-440, 30 GW days/t - 1 kg/t. A simple straightforward method allows only 90 per cent of Pd to pass into aqueous solution and it is considerably contaminated. Methods of obtaining needed decontamination coefficients of $10^{10}$ to $10^{11}$ are now being developed and are based on three stage extraction by a mixture of TBP and diheptisulphite.

The authors of Ref. [107] point out that the amounts of HLW and MLW in NSF reprocessing are determined by the amounts of nitrates and other salts introduced as reagents. Their replacement by salt-free reagents reduces waste volumes radically, does the use of alternative ways of stabilization of U, Pu and Np valences needed to extract and separate those elements, for example by electrochemical methods.

The most important problem in minimization of LLW volume is removal of tritium from the SNF.

Among the possibilities considered is incomplete separation of Pu from Np and U or even no separation at all but "collective" recycling. Incomplete separation from Am and some fission products should also be considered if remote control refabrication methods are used.

As was already mentioned, average fission to capture ratios for threshold nuclides are by an order of magnitude higher in fast reactors than in thermal ones and that is an important advantage. In high thermal neutron fluxes a considerable fraction of actinides turns into curium isotopes instead of fissioning. That may not be considered a satisfactory result of transmutation because medium lived $^{244}$Cm has very high activity and heat production. And it decays to $^{240}$Pu. So one gets a "squirrel wheel" - spending four neutrons to turn $^{240}$Pu into $^{244}$Cm and a return to the start with a very dangerous nuclide as an intermediate stage. And $^{244}$Cm is more radiotoxic than any other actinide in the spent fuel because 8 per cent of its decay is spontaneous fission and its half life is most inconvenient - 340,000 years. But curium recycling in fast reactors creates some serious problems due to its high activity and heat production. That stimulates looking for an efficient way of extracting it from the wastes and cooling it for a few decades.

The burn rate of 3.5 per cent of actinides added to BN-800 fuel (14 per cent to plutonium) is about 100 kg/year, so the support ratio to WWER-1000 is 3. An actinide admixture of a few per cent does not change fast reactor physics much. Higher burning rates may be reached in a specialized BN-800 core with $^{238}$U replaced by an inert matrix like zirconium carbide. Fuel composition may be optimized to provide zero sodium void coefficient and acceptable beta-eff. This optimum corresponds to a MA/basic fissionable isotope ratio in fresh fuel at the level of 1:3. Such a 2000 MW(th) reactor burns some 350 kg of MA a year (support ratio about 10).

Main topics investigated or included in immediate plans (mainly in Bochvar and Khlopin Radium Institutes; RIAR activities are described in Section 3.6.3.2):

- analyses and validation of strategies, concrete research targets and methods of radionuclides partitioning;
- partitioning of volatile elements (H, J, Kr, Xe, Cs, Ru) using thermochemical processes and oxidation treatment of spent fuel;
- development of the methods of deep partitioning of Pu, minor actinides and fission products from highly radioactive solutions of RT-1 (operating) and RT-2 (under construction) reprocessing plants with the use of extraction, extraction-membrane, ion-exchange, electrochemical, distillation and sedimentation methods;
- radiochemical problems in burning weapon grade and reactor plutonium;
- technology of solid fuel compositions and reactor targets containing actinides and fission products to be transmuted in reactors;
- methods of localization, compacting and inclusion into matrix materials of transmutation products oriented on intermediate and final storage conditions;
- development of the equipment for radiochemical and chemical metallurgy processes;
- analytical support of R&D (non-traditional methods of component determination in the P&T processes);
- partitioning technologies for treatment of wastes left from military nuclear programmes;
  radiochemical support of transmutation in water loops;
- technology of fabrication of Np, Am and Cm containing fuels by plasma-chemical methods;
- radwaste petrifaction and vitrification technologies;
- radiological and geochemical validation of the developed P&T methods;
- economical aspects of P&T schemes;
- conceptual development of the methods of fabrication and reprocessing of UN-PuN-MA fuel;
- quantitative methods of and equipment for solving metallic Pu (aqueous and molten salt systems)
  with the emphases on nuclear safety;
- development of an on-line radiochemical module for liquid fuel accelerator driven transmutation
  blankets;
- preparation of UO2-PuO2 fuel mixtures with plutonium concentrations from 5 to 100 per cent;
- radiological validation of various separation and decontamination factors;
- adaptation of "Mayak" technologies for P&T tasks;
- preparation of concentrated specimens of 129I, 99Tc and 237Np;
- fluoride free methods of dissolving transuranic oxides.

For some years investigations have been going on in the V.G. Khlopin Radium Institute on the
behaviour of "reactor" palladium in the flowsheet for NPP spent fuel reprocessing. As a result, the
potential points for withdrawal of palladium (raffinate or evaporated raffinates of the first extraction
cycle) are determined, the extraction and sorption methods for the recovery of palladium concentrate
are being developed. The extraction method has been tested on a laboratory rig in the hot cells of RI.
100 g of palladium were obtained by a sorption method in a pilot test facility of the Production
Association "Mayak".

A method for sorption purification of palladium has been developed and tested. Five g of
purified palladium were produced. The method allows production of palladium of practically
radiochemical purity.

The obtained specimen is being used to study some radiation-physical characteristics of
palladium. Isotope composition (15% of radioactive isotope 107Pd), thickness of semiabsorption
layer, specific activity of 107Pd per surface unit are being determined.

The possible fields of palladium applications in the radiochemical industry (fixation of iodine,
fixation of actinides in cermet compositions) are being evaluated.

The behaviour of 106Tc in extraction reprocessing of spent fuel from WWER-440 NPPs was
investigated.

A technology for the recovery of radiochemically pure Tc was developed. In the PA "Mayak" a
pilot facility for final elaboration of the process of Tc recovery from waste solutions generated in
WWER-440 spent fuel reprocessing was designed. During test operation more than 100 kg of Tc
were produced in the form of sodium pertechnetate. Methods for production of volatile compounds
of Tc and technology for applying thin metal coatings to the surfaces of different metals were
developed in the course of further investigations.

A technology for the complete capture of 129I, the major portion of which is present in gas
effluents of spent fuel reprocessing was developed and tested in the PA "Mayak". A combined
gas-cleaning scheme allows separation of 129I rather easily in forms suitable for subsequent
management.

A method was elaborated for chemical purification of separated iodine with the concentration of
129I not less than 87% and for obtaining its compounds with different chemicals.

The Radium Institute in collaboration with PA "Mayak" can recover and produce the necessary
amount of 129I in an appropriate form for extended experiments on the transmutation of this isotope.
3.5. ACCELERATOR DRIVEN TRANSMUTATION TECHNOLOGIES (ADTT)

3.5.1. International status

It has been recognized for a long time that high current (10-100 mA), high energy (1 GeV) proton accelerators can provide an intense spallation neutron source through bombardment of protons on suitable targets. Based on this, schemes have been proposed in the past to breed fissile material $^{233}$U, $^{239}$Pu or generate tritium or transmute troublesome long half life components of nuclear waste through the use of accelerators. In the recent years there has been renewed interest in hybrid accelerator driven subcritical reactor systems. As the accelerator produced neutrons can be considered to indirectly augment the average number of neutrons per fission, a subcritical reactor assembly of say $k < 0.9$ to $0.95$ can be operated as a self sustaining chain reaction system at any large thermal power with the input of accelerator produced neutrons. The system, however, will be a critical assembly only so long as the accelerator is on and is, therefore, very safe from the consideration of criticality accidents as with the accelerator off, the system again becomes subcritical. Such accelerator driven subcritical systems have been recently proposed for transmutation of troublesome long half life minor actinides and fission products present in the spent fuel waste. A similar proposal has recently been made by Carlo Rubbia & Collaborators to generate nuclear energy from the Th-$^{233}$U fuel cycle in an accelerator driven subcritical reactor assembly.

The physics of an accelerator driven reactor system is basically as follows:

As a high energy proton passes through a fissionable target, the number of fissions of the target nuclei is directly proportional to the number of neutrons released via the spallation and the evaporation processes. Each 1.6 GeV proton impinging on a minor actinide lattice releases about 50 neutrons and causes about 5 to 6 spallations (destruction by fission) of actinide atoms. If the target is a large subcritical assembly, the number of fissions of the actinide atoms will be much larger depending on the effective neutron multiplication factor $k_{eff} (=k)$ for the lattice. The additional number of fission neutrons due to multiplication in the lattice will then become $50k/(1-k)$. If $n$ is the average number of neutrons released per fission, the additional number of fissions will be $50k/(1-k)n$. Assuming $n = 2.7$, for $k_{eff}$ in the range of 0.9 to 0.95 the additional number of fissions of the lattice actinide nuclei will be 167 to 352, which is much more than the direct 5 or 6 fissions caused by the incident proton. Since most of the fissions are caused by the neutrons multiplied in the subcritical assembly and not by the direct beam, the basic characteristics of the neutronics is almost the same as in an equivalent reactor system. An important advantage of the scheme is that an accelerator linked chain reaction is possible by an indirect enhancement in the effective average number of neutrons per fission when one considers a complete cycle in which all or even a part of the fission generated electric power is returned to the accelerator.

The basic idea of transmutation of troublesome fission product and actinide waste to stable or short lived species has been under consideration for many years now [See Hebel et al., Rev. Mod. Phys. 50 (1978) Part II p.1]. It has also been well recognized that the minor actinides Np, Am, Cm can be effectively transmuted in a fast neutron flux, since the ratio of their fission to capture cross-section is quite large (about 100 for $^{237}$Np, about 20 for $^{241}$Am and 200 for $^{244}$Cm) for fast neutrons. Thermal neutrons have not been considered particularly useful for this purpose because the fission to capture ratio is quite small for the primary constituents $^{237}$Np and $^{241}$Am of the actinide waste, although thermal neutrons are suitable for the elimination by transmutation of the fission product waste by neutron radiative capture processes. However, a new approach has been proposed by Bowman et al., wherein it is shown that one can effectively transmute actinide waste in a large thermal flux of accelerator produced neutrons in the range of $10^{16}$n/m$^2$, making use of the rapid successive neutron capture which becomes possible in a large neutron flux. Hence, the proposed schemes of accelerator driven subcritical assemblies for actinide waste transmutation can be classified into two broad categories involving fast or thermal neutron spectra which are discussed in more detail below:

One of the accelerator driven subcritical target concepts for transmutation of nuclear waste called the PHOENIX concept proposed by the Brookhaven Group uses a large linear proton accelerator.
(linac) capable of producing a proton beam of about 100 mA of 1.6 GeV protons. The number of neutrons released is about 50 for each 1.6 GeV proton in a minor actinide lattice of the PHOENIX target modules. One of the 3600 MW (thermal) proposed machines of this type would be capable of transmuting the neptunium, americium, curium and much of the iodine produced in as much as 75 light water reactors (LWRs) and will also generate 850 MW of usable electricity of for the electric grid. Since the minor actinides fission efficiently only in a fast neutronic spectrum, the present scheme is based on generating an intense fast neutron flux, with the options to consider sodium, lead or helium coolants and metal, oxide or other suitable fuel forms. A multiple module concept was developed for the subcritical lattice, with each module resembling the core of the fast flux test facility (FFTF) with the minor actinides formed into oxide fuel rods, replacing uranium or plutonium in the FFTF fuel. Liquid sodium is used to cool the fuel rods which are bundled into 217 pin assemblies, with 124 such assemblies making up a 450 MW (thermal) large module.

The 1.6 GeV, proton beam with a current of about 100 mA from a linear accelerator is suitably expanded with a beam expander to strike from 1 to 8 of the target modules. It has been estimated that when the number of modules are 8, out of the 1260 MW electrical converted from 3600 MW thermal, about one third goes to run the accelerator and the remaining two thirds is available to the electric grid as net power generated. This ratio of net available power to the total power generated, however, decreases from about 65% to 25% as the number of modules decreases from 8 to 1.

Most liquid metal cooled reactors use oxide fuel and it is generally believed that minor actinides could be substituted for the uranium and/or plutonium in this fuel. However, the poor thermal conductivity and the softening on the neutron spectrum due to oxygen atoms are notable disadvantages. Thus development of metallic fuel will lead to the desired hardening of the neutron spectrum and improvement in thermal conductivity. However, substitution of Americium, Neptunium and Curium for Plutonium and Uranium in the metal fuel would require development effort. Since the subcritical lattice is driven by an accelerator, the inherent shut down characteristics of the metallic fuel in the reactor environment will not have much impact. It may be noted that the inclusion of minor actinides significantly increases the sodium void reactivity worth and decreases the Doppler reactivity feed back. In addition to metal and oxide fuel, it is also of interest to study carbide or nitride fuels for this purpose.

The concepts of JAERI, Japan, for the Japanese Omega process also includes use of fast neutrons generated through the use of fast reactors or accelerators. The concept proposed by Carlo Rubia and Collaborators for nuclear energy production in a $^{232}$Th-$^{233}$U fuel cycle with a particle accelerator is also quite similar and, in principle, can be utilized also for actinide waste incineration.

The concept of using an accelerator driven intense thermal neutron source of flux of about $10^{16}$ n/cm$^2$ for transmutation was proposed by C.D. Bowman et al., of Los Alamos National Laboratory, USA. In this proposal, the neutron production-transmutation system consists of an accelerator for the proton beam, a flowing heavy metal (liquid lead) target for neutron production by spallation of lead, and a surrounding blanket containing primarily heavy water for moderating the neutrons into the thermal range. The neutron flux is further enhanced by neutrons from actinide fission in the blanket. Molten salt carrying actinide material mixed with the carrier salt LiF-BeF$_2$ circulates in the heavy water blanket through double walled pipes. The heat generated by the fission of the actinides in the molten salt is suitably extracted and converted to electric power. While some of the power drives the accelerator, a major fraction is made available to the electric grid for distribution. Nuclear waste including that produced in the molten salt is also circulated through the blanket in a separate loop and transmuted by high thermal flux to stable and short lived nuclides which are extracted and stored. The high flux enables transmutation of isotopes such as $^{237}$Np and $^{241}$Am which in a low flux would result in other long lived radioactive species. In order to retain high efficiency and to remove stable or short lived transmutation end product, it is necessary to perform a chemical separation either as part of a slipstream or by processing the material in batches.

The basic mechanism of actinide destruction in a very intense thermal flux is through a two step capture process in which the target nucleus goes from $^{237}$NP to $^{238}$Np to $^{239}$Np on a time scale which
is much shorter than the 2.1 day half life of $^{238}\text{Np}$. Since the thermal fission cross-section (2100 b) of $^{238}\text{Np}$ is much larger than the thermal radiative capture cross-section (300 b), the $^{238}\text{Np}$ nucleus is ultimately destroyed by fission in most of the cases. Thus $^{237}\text{Np}$ which is a neutron poison in low flux, becomes a fuel in a high thermal flux. The situation for $^{241}\text{Am}$, although a little more complicated due to the presence of an isomer, is very similar and an efficient two step fission of $^{241}\text{Am}$ is also possible. Detailed feasibility calculations on this scheme have been reported by Bowman et al., using the code CINDER, which takes into account multistep capture and radioactive decay along all the possible routes.

Since the radioactivity of the spallation products also gives substantial energy generation, one needs to take care of the decay heat in the lead target even after the accelerator is switched off. The power level of the decay heat in the lead target with several years of operation at 30 mA is estimated to be about 1 MW, which is to be compared with the power level of about 50 MW during operation with a 30 mA, 1.6 GeV proton beam.

**Accelerator technology**

High power linac technology has been demonstrated long ago with the construction of the accelerator for the Los Alamos Meson Physics facility. Since then, new technology for the injector using a radio frequency quadrupole (RFQ) to accelerate and bunch the beam into packets for further acceleration in a drift tube linac to 20 MeV has been developed. These high intensity beams in the several tens of MeV range can be accelerated to the GeV range with linear accelerator technology. The room temperature 25% duty factor design of the LAMPF type is not expected to satisfactorily meet the current and other efficiency requirements, as the busbar efficiency is quite low. A room temperature 100% duty factor design was recently proposed by the US Department of Energy for the accelerator production of tritium (APT) at a current of 250 mA and an energy of 1.6 GeV. The expected busbar-to-beam efficiency of this is about 48%, which is quite satisfactory. The superconducting 100% duty factor design, which represents the latest in linac technology, is being incorporated into the CEBAF accelerator and was proposed for the SSC Project. The capital costs for this design are expected to be significantly lower. With an effective gradient of about 3 MV/m, an actual superconducting accelerator of 800 MeV would be about 270 m long. The efficiency of this would be higher as there is virtually no loss in the walls of the cavities. Of the three possibilities, the superconducting design has the advantage because of the higher efficiency and the lower capital cost.

In the scheme proposed by Carlo Rubia for an accelerator driven reactor system based on the $^{232}\text{Th} - ^{233}\text{U}$ Cycle, the requirement of the beam power is about 10 MW which is about an order of magnitude lower than for the schemes proposed for transmutation of waste. Carlo Rubia has proposed a three stage cyclotron accelerator, as a possible solution. The injector consists of two 5 mA compact isochronous cyclotrons working in parallel. The intermediate stage (ISSC) consists of a four separated sector cyclotron accelerating the injected beam up to 120 MeV. The final booster separated sector cyclotron (BSSC) has ten separated sectors and six cavities raising the final energy up to 1 GeV. The possibility of adopting cyclotron based compact, economic and efficient accelerators for waste transmutation applications needs further study.

For actinide waste transmutation through accelerator driven subcritical systems, some of the areas where further work needs to be actively pursued are as follows:

a. Development of compact, efficient, high energy (1 GeV) high current accelerators.
   - Study of basic technology of ion source, RFQ and accelerator tubes
   - Development of high power proton linacs
   - Possible development of a compact cyclic accelerator

b. Development of simulation and design codes
   - Development of nuclear spallation reaction and particle transport codes, nuclear heating and structure design codes
c. Physics and engineering studies for spallation target system

- Integral experiments using lead and uranium targets
- Thermo-hydrodynamics and material tests of spallation target design
- Design studies of target blanket fabrication and its cooling system

d. Other studies

- Evaluation of energy balance
- Risk analysis
- Fuel cycle cost evaluation.

3.5.2. Czech Republic

3.5.2.1. Introduction

The systems for nuclear waste management based on accelerator driven transmutation of long lived radionuclides (transuranic actinides and long lived fission products) are in recent period mostly, designed as systems which are naturally divided into three technological parts:

1. accelerator (neutron source).
2. reactor (blanket).
3. reprocessing (radiochemical).

Here a short and comprehensive review is given of the current status of research and development activities of different scientific, research and industrial institutions in the Czech Republic in the individual fields listed above. The principal approach is close to the one outlined in Ref. [114].

As a country having constructed and operated nuclear power plants and partly producing the corresponding technology (namely heavy and chemical machinery) the Czech Republic is highly interested in implementation of new techniques of radioactive waste management in its national nuclear power programme. As in other countries in a similar situation, the first reason for this high level of interest is the problem of finding a final solution for the disposal of radioactive waste generated by nuclear power which is coming to be a really key issue of the future destination of not only nuclear power but also the national economy itself.

The approach to solving these problems will be described in principal and the first results of attempts to formulate and start the organization of a focused activity will be outlined in principle.

For a better understanding of the national position, a brief survey of most the important types of reactor designs of such systems and their characteristics will be recalled in the paper. The problems of neutronic characteristics and their prediction both for steady state and long term time behavior will be described in principal. Their influence on the reactor part of the design as well as some still open problems will be mentioned, too.

There will be, at least very briefly, a quantitative analysis and an economical efficiency estimate given for this technology assuming it to have been introduced into a system of nuclear power based upon thermal nuclear reactors.

Taking account of the limited economical capacity of the country in the face of such a broad and complex branch of nuclear power technology, an effort to formulate a proposal for a suitable form of an international co-operation project in the given field has been made and this is discussed in the conclusion and recommendation part of the paper.
3.5.2.2. Philosophy of radioactive waste and spent fuel management

The issue of a definitive solution of long lived radioactive waste has become a crucial point in the future of nuclear power. The attempts to use more or less classical (i.e. mechanical, chemical and other non-nuclear) techniques for avoiding the potentially bad influence of long lived RW upon the environment have in practice failed. The underground disposal of such a waste has been doubted by not only the public but, recently, also by the scientific community.

The main cause for this skepticism is the long lifetime of some of the radionuclides which play a significant role in this issue. And this is the crux of the problem. The radioisotopes in question have been mostly produced by forced nuclear reactions in nuclear reactors. On the contrary, however sophisticated the engineering systems for conditioning the reactor waste may be, they rely on only one process for converting radioactive materials into stable ones not requiring engineered storage systems, and that is radioactive decay. This, of course, leads to an extraordinary long period of time (~10^5~6 years) for which the reliability of the engineering barriers being incorporated into the system has to be guaranteed. It is not only a great drawback from engineering and economical points of view but it is also going out of the frame of real possibilities to prove such a system in a creditable way.

The only acceptable principle of how to avoid troubles caused by long lived radionuclides created by nuclear reactions is to employ nuclear processes to accelerate their transmutation to stable materials. If such a technology of nuclear waste burning could be technically and economically feasible it would be a solution of the issue in question.

3.5.2.3. Principles of different transmutation systems

The first candidate for this purpose is the classical nuclear reactor itself. Its ability, to a certain degree, to burn fission products is natural and well known. The first limitation to this approach is the neutron economy of conventional reactor systems. The next candidate could be an actinide fueled (critical) reactor. Then other problems arise, for example, the problem of control of such a system. It is well known that the fissile isotopes of Neptunium, Americium and Curium have a much smaller fraction of delayed neutron emitters in comparison to the common fuels based on Uranium isotopes. It is not necessary to recall here the essential role which the fraction of delayed neutrons plays in the control of a nuclear reactor in its critical state. Beside this, there are other problems with a small Doppler effect and a positive sodium void coefficient which arise in this case.

To overcome these problems, proposals have recently been made of various concepts of subcritical systems driven by a suitable neutron source, aiming at the transmutation of actinides and long lived fission products. A suitable neutron source, one with a sufficient yield of neutrons avoiding the principle barrier of neutron economy, is possible in the concept of spallation reactions induced by highly accelerated charged particles, e.g. protons:

1. The idea of direct exploitation of the spallation process to transmute actinides and fission products had to be given up because the necessary particle currents are much larger than the most optimistic theoretical accelerator designs can achieve (around 300 mA). It has been reported [114,115] that the destruction rate of the largest possible proton accelerator would correspond only to a fraction of the amount of fission products generated by one thermal reactor of 1000 MW, in the same period of time.

2. The idea of a direct use of spallation neutrons was tested. The fission products would be placed around a proton target to use only the spallation neutrons as they are generated in the target. Depending on the characteristics of the material which is to be transmuted, either the fast neutrons would be used as they are emitted from the target or they would be slowed down by a moderator to energy levels with higher transmutation cross sections in the resonance or thermal energy ranges. The following simple estimate shows the substantial disadvantage of such a system. Assuming that it is possible to make all the spallation neutrons available for the transmutation process:
where \( E_p \) is the amount of energy which is necessary to transmute the fraction \( q \) of radionuclides per fission process in a nuclear energy system, \( P_b \) is proton beam energy, \( \eta_p \) is the number of neutrons generated by one proton, \( \eta_p \) is the efficiency of converting electricity into proton beam energy (\(-0.5\)) and \( \eta_t \) is the efficiency of converting thermal energy into electricity (\(-0.33\)). For example, in the case of a 1.5 GeV proton beam emitting 50 neutrons per spallation in a lead target, the transmutation of \(^{99}\text{Tc}, ^{129}\text{I}, ^{135}\text{Cs}, ^{85}\text{Kr and} ^{93}\text{Zr}\) (constituting 28% of all fission products) would require 51.3 MeV to transmute the fission product fraction of one fission process. This is \(-26\%\) of the total power production energy system under consideration and the real percentage of energy required will be even higher due to the very optimistic assumptions made in this estimate. Together with the cost for reprocessing it would make this type of accelerator transmutation prohibitively expensive, at least in a commercial nuclear energy system.

3. The idea of an accelerator neutron source driven subcritical reactor type assembly to improve neutron economy remained. Technically, this can be realized by surrounding a proton target region by fissionable material in a cooling system. For removing the high specific heat released in the target, a liquid metal alloy is mostly used. However, we should mention that the specific heat production per neutron is considerably lower than in a fission process (30 MeV against 80 MeV). We will concentrate on a deeper analysis of such a system in the following paragraphs.

3.5.2.4. Brief survey of reactor designs and their characteristics

3.5.2.4.1. Technical description of basic systems

There has been a series of deep analyses and evaluations of the feasibility of various transmutation systems performed in the last few years. One of them is a detailed and instructive estimation of different transmutation systems given in [114]. A very brief and compressed version of the conclusions of that analysis follows.

One of many possible conceptual designs for an accelerator based transmutation of waste (ATW) system of the target/blanket type is as follows. The target consists of a material that produces neutrons from either direct or spallation reactions. There is, in general, an annulus of high-Z material, such as lead, for further multiplication of the source through \((n,xn)\) and \((p,xn)\) reactions. The blanket contains a moderated heterogeneous lattice, using a flowing liquid which contains the actinides. There are separate regions in the blanket where the long lived fission products (LLFP) are transmuted.

The structure of the heterogeneous core is either a heavy water moderated aqueous slurry system or a graphite moderated molten salt system.

In the aqueous system, the actinides are in the form of an oxide (500 g/l \(\text{AcO}_2\)) slurry in heavy water (1/4% \(\text{H}_2\text{O}\) impurity). The complete slurry flows with a velocity of 12 m/s, temperature difference between 275 and 325°C, under an operating pressure of 13.1 MPa through 3m of tubing made of Zr/2.5% Nb alloy. The heavy water moderator outside the tubes is held at atmospheric pressure and 100°C. The neutron flux in the cell is determined to be \(1.7 \times 10^{15}\) n/cm²/s. The ratios of the times spent outside the neutron flux to the time spent in the neutron flux are 50% for hold up in the heat exchangers, and 50% (Am/Np) and 25% (Np/Pu) for chemical processing.
In the molten salt system, the fuel is in the form of fluoride salt (100 g/1 AcF₄), dissolved in a molten salt carrier whose composition is 60% ⁷LiF and 40% ⁶BeF₂. The carrier’s melting point and operating temperature are 500°C and 650°C, respectively. The molten salt flows over the outside of a hexagonal close-packed set of high-purity graphite balls, of 3 cm radius. The simplest unit cell is a tetrahedron. The neutron flux in the system is determined to be $1.1 \times 10^{15}$ n/cm²/s. The aqueous and molten salt systems are compared in Table XII.

**TABLE XII. COMPARISON OF AQUEOUS AND MOLTEN SALT SYSTEMS**

<table>
<thead>
<tr>
<th>configuration</th>
<th>aqueous</th>
<th>molten salt</th>
</tr>
</thead>
<tbody>
<tr>
<td>fuel capture/fission</td>
<td>1.62</td>
<td>1.74</td>
</tr>
<tr>
<td>neutrons/fission</td>
<td>3.05</td>
<td>3.06</td>
</tr>
<tr>
<td>neutrons/fuel absorption</td>
<td>1.164</td>
<td>1.117</td>
</tr>
<tr>
<td>parasitic capture/fission</td>
<td>0.11</td>
<td>0.015</td>
</tr>
<tr>
<td>fission product/fission</td>
<td>0.12</td>
<td>0.036</td>
</tr>
<tr>
<td>eigenvalue</td>
<td>1.070</td>
<td>1.096</td>
</tr>
</tbody>
</table>

The neutron balance of the ATW system is a function of the choices that are made for the burning of fission products. In both the aqueous and the non-aqueous case most of the neutrons used to transmute LLFPs are supplied by the accelerator. The size of the accelerator is determined by the choice of how many of the LLFPs are to be burned. If none are transmuted, then the accelerator is not necessary. The LLFPs of interest in commercial LWR waste transmutation schemes are listed in Table XIII.

**TABLE XIII. PROPERTIES OF MOST IMPORTANT LONG LIVED FISSION PRODUCTS**

<table>
<thead>
<tr>
<th>species</th>
<th>isotopic moles per LWR/a</th>
<th>elemental moles per LWR/a</th>
<th>half-life 1000s yrs</th>
<th>beta energy [keV]</th>
<th>gamma energy [keV]</th>
<th>elemental neutrons per atom</th>
<th>cross-section [barns]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Se⁷⁹</td>
<td>2.5</td>
<td>23.8</td>
<td>65</td>
<td>160.0</td>
<td>0.0</td>
<td>1.2</td>
<td>1.50</td>
</tr>
<tr>
<td>Zr⁹⁳</td>
<td>257.5</td>
<td>1297.4</td>
<td>1500</td>
<td>60.0</td>
<td>0.0</td>
<td>2.1</td>
<td>2.10</td>
</tr>
<tr>
<td>Te⁹⁹</td>
<td>259.5</td>
<td>259.5</td>
<td>210</td>
<td>293.0</td>
<td>0.0</td>
<td>1.0</td>
<td>17.00</td>
</tr>
<tr>
<td>Pd¹⁰⁷</td>
<td>67.9</td>
<td>427.3</td>
<td>6500</td>
<td>33.0</td>
<td>0.0</td>
<td>2.8</td>
<td>6.60</td>
</tr>
<tr>
<td>Sn¹²⁶</td>
<td>7.2</td>
<td>46.0</td>
<td>100</td>
<td>250.0</td>
<td>87.0</td>
<td>1.8</td>
<td>0.16</td>
</tr>
<tr>
<td>I¹²⁹</td>
<td>46.2</td>
<td>60.5</td>
<td>16000</td>
<td>150.0</td>
<td>39.0</td>
<td>1.0</td>
<td>15.00</td>
</tr>
<tr>
<td>Cs¹³⁵</td>
<td>74.2</td>
<td>588.1</td>
<td>3000</td>
<td>210.0</td>
<td>0.0</td>
<td>1.6</td>
<td>5.80</td>
</tr>
</tbody>
</table>

All the isotopes have low β⁻ decay energies. Some of the species have accompanying gamma rays, also of low energy. Most species occur generally with several isotopes. All the isotopes present in a mixture get transmuted no matter whether they are radioactive or not and neutrons are spent in the process. Irradiation of elemental mixtures to reduce them to mixtures of long lived radioactive components may be prohibitively expensive in neutrons or even impossible. For example, zirconium appears as 7 species, 90 through 96, not including the short lived 97. If isotope separation is not used, isotopes 90 through 94 must be transmuted to 95, which decays by two successive β⁻ emissions to ⁹⁵Mo. The isotopes 90 through 93 thus require more than 1 neutron per transmutation. Isotopes 94 and 96 require each 1 neutron. The net average per atom is 2.1 neutrons per transmutation. These are the quantities listed in the last but one column in Table XIII.
The amount of electrical power needed to operate the accelerator can be expressed as follows. The transmutation of the only species that appears as a single isotope (\(^{99}\text{Tc}\)) requires 16% of the power of an LWR. The transmutation of all 7 isotopes would require 45.3% of the electrical output of the LWR. The elemental Zr would require more power than is produced in the LWR. Elemental transmutations are still expensive for Se, Tc, Sn and I. The transmutation of all these combined would require 27% of the LWR's power.

3.5.2.4.2. Principal balance analysis

Let us quantify the power production \(P\) of a subcritical assembly fed by spallation neutrons:

\[
P = \eta_p \frac{a-k}{\mu(1-k)} \frac{iE_f}{E_f^P}
\]

(2)

where \(k\) is the multiplication factor, \(a\) is the importance of the target position (usually \(a > 1\) for a central target position), \(\mu\) is the mean number of neutrons in a fission process, \(E_f\) is the power release per fission \((= 3.2 \times 10^{11} \text{ W})\), \(\eta_p\) is a neutron yield from one proton, \(i\) is proton current, \(C\) is proton charge \((= 1.6 \times 10^{-19} \text{ A})\).

The additional neutrons from the subcritical system as well as its fission power, which can be also transformed into electricity, can be exploited to run the transmutation process. The energy required to transmute a fraction \(q_{TF}\) of fission products in such a system can be expressed as follows (for the case \(a = 1\)):

\[
E_{fp} = \eta_p \frac{k}{\mu(1-k)} \frac{P_k}{\eta_p \eta_T} \frac{E_f}{\eta_p ((1-k)\eta_p + \frac{k}{1-k}((1-k)\eta_p + \frac{d_{fp}}{\mu})}
\]

(3)

where

\[
\eta_p = \sum_a (FP + \text{fuel + structural materials})
\]

A positive sign of \(E_{fp}\) means that there is even a surplus of energy, while a negative sign indicates a need of energy which should be added to the system from outside.

In spite of the very promising features, the fact that the amounts of long lived fission products generated by currently operated thermal reactors are relatively very high, and correspondingly huge numbers of neutrons are required for their transmutation, results in a certain stress in the overall neutron economy requiring a relatively high power accelerator for the spallation neutron source. One of the possible improvements in this situation is a higher degree of multiplication of spallation neutrons from the source by the subcritical reactor. In a deeply subcritical transmutor with \(k_{sp} = 0.7\), a proton beam power of 300 MW is required to transmute 17% of LLFP, such as \(^{99}\text{Tc}, ^{129}\text{I}, ^{135}\text{Cs}, ^{85}\text{Kr}\) and \(^{92}\text{Zr}\), created by a 1 GWe LWR. When the spallation source neutrons are multiplied by a transmutor which is closer to criticality (with e.g. \(k_{sp} = 0.99\)) and when the ratio \(n\), of neutron capture by LLFP to the total is 20%, the proton power becomes 4 MW.

3.5.2.5. Problems of radiochemical reprocessing and connected technology

3.5.2.5.1. Mutual relations of physical and chemical aspects

The close relations between physical and chemical aspects are reflected in the series of fundamental reviews and basic work in the field in question. However, there exists a series of mutual relations which are not visible at first and this paragraph focuses attention on them, at least very briefly.
Nuclear transmutation is the main point of the technology discussed here but the nuclear properties of radionuclides have an important influence on the requirements for the chemical methods to be applied. For example, the fact that burning and transmutation go on in classical thermal and fast reactors (the minor actinides are better burned in a thermal reactor and on the contrary the major actinides in a fast one) results in a requirement to develop suitable methods for separation of the following different steps:

- minor actinides,
- major actinides,
- fission products with half-life greater than 30 years ($^{99}\text{Tc}$, $^{129}\text{I}$, and $^{135}\text{Cs}$),
- $^{96}\text{Sr}$ and $^{137}\text{Cs}$ which might be utilized as radiation sources,
- remaining fission products.

Beside this, the requirements on the efficiency of chemical separation depend on the physical properties of the process applied. E.g., the application of a fast reactor leads to the demand for a separation of 99.9% of Pu and Am, and 98.04 of Np and Cm. Separation with such a high efficiency is e.g. in the case of Am, a new and difficult task from the chemical point of view. Also different material and geometrical arrangements of reactor systems determine the requirements on chemistry (there is a necessity for development of a specific method of separation when transmutation is not carried out as "homogeneous" with equal content of actinides in all fuel elements of the system).

It would be possible to go on in this description of mutual relations and feed backs, however, even the few examples presented allow summarization that the concept of partitioning represents in fact a more developed form of reprocessing and its side effects, beside developing targets for transmutation, may be expected also in an improvement of radioactive waste conditioning techniques and may even create strategic reserves of e.g. platinum metals.

3.5.2.5.2. Time sequence of further steps of development

Many authors agree that the strategy of partitioning and transformation will require twenty or even forty years of development. However, some steps can be already made now and in the near future. According to ref. [116] it is already technologically possible to increase the yield of U and Pu in a reprocessing process and to separate minor actinides by use of certain chemical procedures and then once again to utilize U and Pu in the form of MOX fuel in classical reactors and to keep the minor actinides for future utilization. Also other authors [117] confirm that pressures and stimulation for improvement of the current PUREX process and its broadening into separation of Am are very strong.

There are various views on the problem of separation of long lived fission products and its possible solution. The global estimation is that at least 15–20 years will be necessary to have a sufficient amount of technical and economical information to be able to develop an optimal strategy and make decisions. The most demanding features of the chemical part are the following:

- high level of the elemental separation efficiency,
- necessity to separate isotopes so as to avoid transmutation of stable nuclides,
- relatively fast chemical separation.

3.5.2.6. Short analysis of economic efficiency

To evaluate the economical aspects of this system is an extraordinary difficult task as the majority of the innovations involved are complex. This is probably also the reason why there has appeared so few of publications in the literature so far. Nevertheless, some at least preliminary, conclusions can be formulated from the available studies in this field [118-120]. Besides from the conclusions of the well known Los Alamos paper a rough guess can be expressed as follows: even if a two order (the first estimation made in the paper is 55 times) higher price of the chemical processing involved in this system in comparison with the prices of reprocessing valid in France
nowadays, the competitiveness of electricity production by this system will still not be in danger. This is true in the case when $^{90}$Sr, $^{137}$Cs and $^{93}$Zr are not fully transmuted.

There is also a qualitative analysis dealing with the economical benefit of separation of precious metals in [121]. It is estimated that the isotopes of ruthenium, rhodium and palladium will become very lowly radioactive after fifty years and the amount of these elements in high level radioactive waste is estimated to represent about 30% of the known world wide reserves of the platinum group of metals. There is a condition leading to a possibility to utilize all of it. This needs the precursor – technetium – to be collected in the reprocessing process in one stream in spite of the current situation where about 50% remains in the non-soluble residues after dissolution of spent fuel before reprocessing. What is more, the separation of the platinum group elements would have one more advantage in improving the properties of vitrified waste where their originally high concentration causes the long term instability of the product.

Generally, it can be concluded that the economical effects of each step of realization of transmutation technology should be evaluated separately and the resulting values might be the subject of significant changes as the technology is developed. In any case, the cost benefits are much more difficult to estimate with desirable accuracy at the moment than the technical aspects of the system.

In the above outline the material of Refs [122-125] was also used.

3.5.2.7. Latest developments

On the basis of a broad study and a series of preliminary analyses, attention has been concentrated on R&D activities in the Czech Republic connected with a chosen heterogeneous blanket core structure of an accelerator driven transmutation system. The structures of heterogeneous cores which were competitive in the previous stages were either a heavy water moderated aqueous slurry system or a graphite moderated molten salt system. The former has been connected with demanding continuous reprocessing which became its main drawback. The latter can utilize mechanical (e.g. centrifuge) separation of individual components of the liquid fuel and has also a higher thermal efficiency. Therefore, these systems have become the center of attention of the national R&D program in the field of transmutation systems in the Czech Republic for the near future.

The neutron balance of the accelerator driven transmutation systems, namely for spent fuel from PWRs, is a function of the choices that are made for the burning of the fission products. The majority of the neutrons used to transmute long lived fission products (LLFP) is supplied by the accelerator driven neutron source. The strength of the neutron source and the corresponding size (parameters) of the accelerator are determined by the choice of how many of the LLFP are to be burned. If none are transmuted and only actinides from PWR spent fuel are burned, then an accelerator is not necessary at all. However, the advantages of a subcritical core operation regime would also be lost in that case. So there is an optimum hybrid system (subcritical blanket/core driven by a suitable accelerator source) for which a search is being made.

A preliminary concept call LA-0 has been elaborated in the Czech Republic in close collaboration with Los Alamos National Laboratory, USA, a brief description of which is given below.

In the molten salt system, fuel is in the form of fluoride salt $\text{AcF}_4$ dissolved in a molten salt carrier whose composition is a mixture of $^7\text{LiF}$ and $^9\text{BeF}_2$. The carrier's melting point and operating temperature are about 500°C and 650°C respectively. The molten salt flows over the outside of a close-packed set of cylindrical graphite blocks arranged in a regular triangular lattice. There have been elementary analyses made which lead to a preliminary accepted set of optimal values of the graphite rod radius $R_1 = 8.6$ cm and the elementary cell hexagonal shaped border size (inner circle radius $R_2 = 10.0$ cm.)
For the very first stage, the following scheme will be applied which will allow the first results to be achieved very cheaply and relatively soon. There will be an elementary but nevertheless sufficiently representative sample of the above described lattice (an elementary module consisting of 7 elementary lattice cells) inserted in the experimental reactor LR-0 which has been successfully operated for core analyses of thermal power reactors (e.g. light or heavy water moderated and uranium fueled systems were tested there) in the Nuclear Research Institute of Rez.

The chosen concept of the elementary module should not only fulfill its role as the first stage, it should allow progression to further stages and to compose more complex models of the principal parts of the investigated technological system and its functions (e.g. higher temperature, flows of given media and corresponding technological unit testing should be possible).

It is unlikely that verification of accelerator driven subcritical blanket core control properties will start by use of a spallation source. Therefore there has been the idea of a low level accelerator driven source incorporated into the LA-0 concept adopted. This can be either a proton accelerator with a $^7$Li-target or more likely a deuteron accelerator with a $^9$Be target. This would be either the existing cyclotron U-120 being currently operated in the Nuclear Physics Institute in Rez or a newly constructed 5 MeV cyclotron which would be incorporated into the LR-0 experimental reactor complex.

The concept described above will allow a complex investigation of realistic behavior of an ADT system together with development of individual components and structures as well as the complete system.

3.5.3. Russian Federation

First international meeting on transmutation in Russia devoted mainly to ADTT was held in Obninsk in 1991 and Proceedings containing 25 papers were published [37]. The main concepts were formulated there in an initial form and have been developed ever since.

ITEP is the leading center of ADTT related research in Russia, co-ordinating, in particular, a $3.2 million research project funded by the International Science & Technology Center and launched in November, 1994. A few institutions (ITEP, RIEP, RTTP, Bochvar Institute, IPPE, Khlopin Radium Institute) and about 400 researchers are engaged (mostly part time). The main task of the project is to prepare a preliminary design of a powerful proton accelerator target cooled by heavy liquid metal, most probably lead-bismuth eutectics.

The main features of the ITEP approach were outlined, for example in a series of papers [33, 34, 126], presented to the Willigen Specialists' Meeting [127]. The work is going on in close co-ordination with Los Alamos National Laboratory.

The latest developments and proposals were discussed at a Moscow Seminar [128] and are briefly outlined below.

In Ref. [129] the possibility is discussed of safe use of weapon grade Pu to fuel an accelerator driven blanket for energy production and transmutation. The parameters and operation regimes of different ADTT facilities are discussed. The conclusion is that they will be useful for closing the NFC with transmutation both of actinides and FP.

The authors of Ref. [130] propose to model a powerful ADT facility using an inexpensive prototype before designing it to full scale and for this purpose to combine the ITEP heavy water research reactor TVR (using it in subcritical regime) and the small ISTRA 36 MeV proton linac. Besides ADTT research this combination will be useful for some other applications. Its conceptual design is described in [130]. This work is supported in part by Grumman Aerospace Corporation and the Russian Fundamental Research Foundation.
Extensive calorimetric measurements of the energy released by fast protons in Be, C, Al, Cu, Fe, Pb and Bi were done by the ITEP group in the energy range 0.8-1.2 GeV and the results published previously are summed up and analyzed in [131].

Radiological hazards associated with the transmutations of actinides in fast reactors and accelerator driven blankets are compared in [132] with the conclusions in favor of the latter.

The advantages of the thorium based ADTT fuel cycle are discussed in [133] and again compared favorably with fast U-Pu breeders. A new concept is outlined in [134] of a blanket composed of a few multiplying sections separated by "neutron valves" (combinations of moderators and absorbers) making section reactivities almost independent with total multiplication factorized which may allow a decrease in the needed power of the accelerator beam by about an order of magnitude.

Lead-bismuth cooled ADTT facilities, their neutronics and coolant technology are considered in [135-136].

Chemical reprocessing of molten salt fuel in the ADTT units is discussed in RIAR paper [137]. Preliminary estimates are given of regeneration and waste management efficiency. Planned experiments are described. Specific blankets designs and calculations are discussed in RDIPCE paper [138].

There are no operating prototypes of accelerator driven blankets in Russia (or elsewhere) and existing plans are based on proposals to combine existing reactors or assemblies with existing accelerators to get inexpensive low power facilities and demonstrate feasibility of the concept.

The following main topics are included in current and near future research plans:

- investigations of technical safety of ADTT using weapon and reactor grade plutonium;
- comparison of various ADTT based fuel cycle options;
- conceptual design of a blanket with solid Pu fuel and solid W target (PER project);
- development and construction of an electronuclear neutron generator coupling the heavy water research reactor TVR (ITEP) and a modernized "Istra-36" proton accelerator;
- numerical and experimental studies of a sectioned blanket with almost factorized multiplication to reduce the necessary proton current;
- experimental studies of model proton targets and separating windows;
- investigations on thermohydraulic and technological aspects of solid and liquid metal target design (including liquids with a free surface);
- reliability and safety of powerful proton beams;
- conceptual design of a prototype facility based on the I-100 proton injector (IHEP, Serpukhov) and a subcritical assembly provided by IPPE (Obninsk);
- accelerator technology studies (continuous current proton sources, dynamic co-ordination of RFQ structures, high power resonators, minimization of beam losses etc.);
- interaction of intermediate energy protons with bulk beryllium and heavy metal targets;
- listing of initiating events, emergency situations and beyond design basis accidents for the target-blanket system;
- methods of protection of the accelerator's ion tract for the case without separating window, formation and transport of a gas phase in the target circuit;
- numerical and experimental study of thermodynamic properties and behavior of the admixtures getting into the liquid target or generated there, of interaction of the admixtures with the coolant and structural materials;
- technology for stabilizing the quality of the coolant and its surfaces;
- development of membrane and electrolyte methods of removing Po;
- comprehensive comparison of lead and lead-bismuth as liquid target materials;
- development and testing of cooling systems for thermal stabilization of accelerator heat releasing equipment for power levels 100-1000 kW per section.
- development of technologies for fast separation of Xe from I and Ru from Tc.
ADTT prospects depend strongly on whether the corresponding facilities in general will find their place in the nuclear power industry. Making, for example, only every tenth reactor subcritical improves neither general nuclear safety significantly nor economy because ADT systems may be more expensive than traditional reactors and consume a considerable part of the energy produced to feed the accelerator.

3.5.3.1. Special radioactivity and radiation resistance problems associated with heavy metal coolants in reactors and ADTT

The main subjects of investigations are reactions and mechanisms leading to formation of polonium from lead and bismuth in mixed radiation fields (protons, neutrons, alpha particles).

Polonium nuclides from $^{198}$Po to $^{210}$Po are formed in lead-bismuth coolant due to interaction of fast protons with bismuth nuclei. $^{210}$Po is also formed in neutron radiation capture $^{209}$Bi(n,$\gamma$)$^{210}$Bi decaying to $^{210}$Po, other polonium isotopes are products of (p,xn) reactions on $^{209}$Bi ($x = 1$-12). Analyses indicated that the most hazardous nuclei are $^{210}$Po ($T_{1/2} = 138.4$ days), $^{209}$Po ($T_{1/2} = 102$ years) and $^{208}$Po ($T_{1/2} = 2.9$ years). Other nuclides are relatively short lived.

There are three channels of polonium formation in lead coolant:

- as a result of radiation capture on bismuth impurities in lead;
- as a result of radiation capture on bismuth formed previously in the reaction $^{208}$Pb(n,$\gamma$)$^{209}$Pb with $^{209}$Pb decaying to $^{209}$Bi ($T_{1/2} = 3.25$ hours);
- as a result of (α,xn) reactions by cascade alpha particles on stable lead nuclides.

The yields of these reactions have been estimated for various energies of incident particles using existing information on microscopic cross-section data.

The estimates are given of polonium activity in liquid metal targets of electronuclear systems for various power levels. It was shown that in the most powerful systems the total specific activity of polonium isotopes may be as high as 1000 Ci/l. It is determined mainly by $^{210}$Po. The activity of polonium nuclides was evaluated for a 10 MW lead-bismuth target now being designed. The activities of various isotopes after three years of continuous operation are: 300 Ci/l for $^{210}$Po; 0.1 Ci/l for $^{209}$Po and 3 Ci/l for $^{208}$Po. Polonium activity is 3 orders of magnitude lower in a lead target than in a lead-bismuth target for identical conditions. But it is not decisive because other radioactive isotopes are formed in those conditions in larger quantities (see below).

Polonium does not present radiation danger in normal operation conditions and with a tightly sealed circuit. But if the sealing is damaged and coolant leaks, the radiation hazards may be essentially due to polonium evaporation from the coolant resulting in aerosol and surface radioactive contamination. Investigations of physico-chemical mechanisms of polonium release from the coolant into the atmosphere (or, for example, into the vacuum ion tract volume for windowless targets) and development of appropriate models are necessary for quantitative evaluation and forecasting of polonium activity and radiation danger. Such models have been practically absent up to now but are in the process of extensive development. For this purpose available published information on the release of $^{210}$Po from Pb-83-Li-17 eutectics [139] and lead-bismuth [71,140] was analyzed and used.

This and additional information indicates that polonium is released from molten coolant not as a free metal but in the form of intermetallic compounds like PoPb. Their volatility is essentially lower than that of metallic polonium. This result is very important for the development of prognostic models of polonium activity formation resulting from target circuit leaks. The release of polonium from the coolant depends strongly on pressure and temperature of the gas over the coolant surface. The rate of release is a thousand times lower at atmospheric pressure than in vacuum.

A programme has been developed for experimental investigation of $^{210}$Po evaporation from irradiated specimens of lead-bismuth alloy and of lead with 0.1 per cent of bismuth added.
Measurements are planned of the polonium evaporation rate from those specimens in vacuum, inert gas and air at temperatures 220-800°C. Sixteen specimens were irradiated in a reactor for 15 days.

The release of the \(^{203}\)Hg radionuclide will be investigated together with \(^{210}\)Po, so some quantity of mercury was added into the specimens. Mercury is formed in liquid lead and lead-bismuth targets in significant quantities in spallation reactions. Mercury is a volatile element and information on its release may be of interest for windowless targets.

Basic parameters of the targets being designed are being analyzed. The main part of the energy released by the beam in a thin window is connected with ionization losses. Nuclear interactions increase ionization losses by some 10-15 per cent.

The analyses demonstrated that replacement of stainless steel by titanium results in decrease of energy release by a factor of 1.7 for the same window thickness. Estimates of energy release done by various groups agree within the accuracy of calculations which is 15-20 per cent.

Significant discrepancies exist in published evaluations of radiation damage to target windows. In particular, the estimates carried out in JAERI and BNL for designs with comparable integral proton fluxes differ by more than an order of magnitude. The main reason for the differences in radiation damage evaluations is the discrepancy in the calculated spectra of secondary particles produced in nuclear collisions. Considerable work is needed to remove the discrepancy and to test the calculation results by comparison with experimental data.

There is a difference of a few times between the evaluations of hydrogen and helium accumulation in the window. To improve the situation more reliable evaluations are needed of the cross sections of the emission of all hydrogen and helium isotopes in the material of the window.

There is also a sharp discrepancy in the evaluations of long lived activities in the targets, sometimes by a few orders of magnitude. Calculations were carried out of the accumulation of the isotopes with periods longer than 1 day for lead and lead-bismuth targets irradiated by a 1800 MeV, 30 mA proton beam. The activation data library ADL-2 with MENDL-1 data added above 20 MeV was used to calculate the reaction rates. For one year of irradiation of the target without heat exchanger the \(^{210}\)Po concentration is \(11.1 \times 10^7\) g/kg for a lead target and \(2.9 \times 10^3\) g/kg for a lead-bismuth target which is about 1000 lower than the results obtained for this isotope in the same conditions in earlier calculations by the OINPE group. It was found that this discrepancy, as well as less important differences for other polonium isotopes, is due to the use in OINPE calculations of the cross section of \(^{209}\)Bi \((n,\gamma)\)\(^{210}\)Po reaction overestimated by a factor of 1000. Use of correct the cross section removes the discrepancies in the results for the accumulation of radioactive isotopes of Bi and Po.

It is necessary to point out that various Bi, Tl, Hg and Au isotopes make larger contribution to long lived activity \((T_{1/2} > 100\) days) than \(^{210}\)Po due to \((p,xn)\) and \((n,xn)\) reactions in the high energy regions of proton and neutron spectra. Testing and validation of such calculations will be an important part of further analyses of the accumulation of induced activity in the target.

A new version of the activation data library (ADL-3) was developed in IPPE Nuclear Data Center during the last year and decay data library was essentially updated for the nuclei with \(A < 210\). Some of the new ADL-3 data on the cross sections of \((p,xn)\) and \((n,xn)\) reactions on heavy nuclides differ strongly from previous evaluations. Cross sections for excitation of long lived isomers have also been re-evaluated and significantly corrected. The influence of these improvements on the results of calculations of the activities of lead-bismuth target irradiated by 1.6 GeV protons will be studied later.

Equilibrium intensities and spectra of alpha particles produced in the target were evaluated as well as the accumulation of polonium isotopes in the reactions induced by those particles. The results indicate that contribution of this channel of polonium isotopes formation by Pb(\(\alpha,xn\)) reactions may
be a few times higher than that of double neutron capture. It is necessary to point out that these processes are induced by high energy particles formed due to pre-equilibrium emission which is usually not taken into account in the models of intra-nuclear cascade. So further investigation of the role of this mechanism seems to be important for reliable evaluation of polonium activities accumulated in lead targets irradiated by high energy protons. Calculation results indicate also that taking these processes into account may lead to reassessment of the relative contribution of different polonium isotopes into total activity which may be important for estimates of residual heat, contamination of accelerator beam tract and for designing of on-line cleaning of liquid target (see also [141]).

3.6. FUEL CYCLES FOR P&T

The fuel cycle for P&T necessarily involves a closed fuel cycle with reprocessing including partitioning technology. R&D related to partitioning technology was described earlier.

The recycling of minor actinides, e.g. to reduce \(^{237}\text{Np}\) and its parent \(^{241}\text{Pu}\), \(^{241}\text{Am}\) and \(^{245}\text{Cm}\) will introduce further stages to the present fuel cycle, which are the partitioning of the minor actinides (and possibly fission products) from the spent fuel, their fabrication into appropriate targets and their transmutation through a reactor or accelerator. Although a precise P&T scheme has yet to be defined, these stages are technically essential and have to be considered in any case.

3.6.1. General issues of MA recycling

The inclusion in a fuel of minor actinides, particularly americium and curium, would increase the radiation dose levels of the fuel both before and after irradiation. In particular, the addition of \(^{241}\text{Am}\) increases the gamma flux from the fresh fuel and the neutron flux from the spent fuel since it gives rise to higher curium production during irradiation. Knowledge of the associates radiation dose rates would allow measures to be conceived for the handling of such fuels.

The issue of handling nuclear material that contains MA at charge time has been addressed in a number of studies [142-144]. Radiation dose levels have been calculated at fabrication and discharge for different recycling schemes.

In Belgium, scientists have examined the implications of recycling plutonium with minor actinides ( Americium, curium and neptunium) in the form of MOX fuel [142,143]. The problems of MOX fabrication were identified with reference to the operating conditions of the MOX fuel fabrication plant in Dessel, Belgium. The following fuel cycles were considered:

3.6.1.1. Recycling of plutonium with minor actinides

The subject of the work was to determine what would be the impact of actinide recycling on the MOX fuel fabrication plant [142]. The case of first recycling was considered. The quantities to be handled were taken equal to those resulting from the irradiation of one ton of enriched heavy metal in \(\text{UO}_2\) loaded initially in a PWR. The quantities are 10.5 kg \(\text{Pu}\), 0.5 kg \(\text{Np}\), 0.66 kg \(\text{Am}\) and 0.05 kg \(\text{Cm}\). Recovery yields of 99.5% for plutonium and 80% for the americium, neptunium and curium were assumed.

A reference plutonium powder was considered with an isotopic composition corresponding to that of first generation plutonium. The \(\text{PuO}_2\) powders were mixed separately with the neptunium, americium and curium powders in order to distinguish the radiation effects associated with the addition of each of the minor actinides to plutonium. A 3 year cooling period was considered prior to fabrication. The most critical stages, as far as radiation doses are concerned during fabrication, were the front end ones, i.e. storage of the material and blending the powders in a glove-box.

In comparison to the reference plutonium powder, the addition of neptunium slightly increased the dose rates. A small re-enforcement of the lead shielding (3 mm) would be required. The addition
of americium increases the dose rate by a factor of 4, thus making necessary the use of up to 20 mm lead. The addition of curium increases the dose rate by a factor of 30, the main contributor being neutrons, and demands the use of up to 300 mm polyethylene as shielding. The addition of americium and curium clearly demonstrates that additional shielding has to be allowed in order to reduce the occupational dose.

3.6.1.2. Multiple recycling of plutonium and americium in PWR

The recycling takes place in a 900 MW(e) PWR in the form of MOX fuel (Pu + Am) [143]. The plutonium and americium from the first core, loaded with UO2 and irradiated to 45 GWd/t, are recycled in parallel to UO2 (in ratio 1:7). The procedure is repeated with successive recycling. It is assumed that the one MOX assembly and the 7 UO2 assemblies are reprocessed together with recovery yields of 99.5% for Pu and 98% for Am. The reprocessing takes place 3 years after discharge, while the fuel is fabricated 2 years latter. The multiple recycling of plutonium only was also considered, on the same lines as the Pu + Am one, for reference.

The front end stages in the fabrication are the most critical ones in terms of radiation protection since blending of the powder material to be used in the fuel is handled in glove boxes. ORIGEN-2 [145] was used to calculate the gamma source at fabrication by simulating the steps of irradiation, cooling reprocessing and time to refabrication. Then, the gamma dose rates were obtained using the shielding computer code QAD-CG [146]. The neutron dose rate was calculated using the one dimensional transport program ANISN-ORNL with the EURLIB cross-section library [147].

The addition of plutonium for a second recycling increases the neutron dose by 20%, due to (α, n) reactions, over the material of a standard MOX fuel. However, the addition of americium and plutonium increases the gamma dose by a factor of 4.5 over the reference case. An addition of stainless steel (25 mm) is required to reduce the gamma dose to that of the reference case.

3.6.1.3. Experience at TUI

Several concepts of MA transmutation, relying on existing nuclear power stations, are being studied theoretically and experimentally. The recycling of MA with plutonium in the "self-generated mode" is being studied either in thermal (PWR) or fast reactors (FR) [148, 149]. Another concept, the transmutation of minor actinides in the fast reactor PHENIX has been studied in the irradiation experiment SUPERFACT [150].

3.6.1.3.1. Nuclear fuel cycles for the self-generated recycling of Pu and minor actinides

Several concepts, relying on existing nuclear power stations, have been proposed for the self-generated recycling of the nuclides of Np, Pu, Am, Cm and Tc [151]:

1. PWR and Fast Reactor reference fuel cycles (R1)

    A BIBLIS type PWR reactor is considered, fuelled with UOX or MOX (U, Pu) and enriched by either 4 wt % in $^{235}$U or by 3.7 wt % fissile Pu. The inventory of the spent fuel was calculated after a burnup of 50 and 33 GWd/t respectively and a cooling time of 7 years [148]. As fast reactor, a PHENIX type, fuelled with 25% first generation Pu and burnt to 60 GWd/t is considered.

2. PWR self-generated transuranium (TU) recycle (R2)

    All self-generated transuranium nuclides together with $^{99}$Tc are recycled in the same reactor and under the same conditions as above (limitations by reactor safety considerations would not allow such a scheme).
3. FR self-generated transuranium recycle (R3)

The inventory of spent fuel, having a burnup of 80 GWd/t, was calculated after the 16th cycle of transuranium nuclide recycling in the fast reactor SUPERPHENIX [149]. An out-of-pile-time of 2 years was considered in this case.

The fuel compositions corresponding to the above nuclear fuel cycles are given in Table XIV [151].

**TABLE XIV. FUEL COMPOSITIONS OF DIFFERENT REFERENCE NUCLEAR FUEL CYCLES**

<table>
<thead>
<tr>
<th>Cycle Description</th>
<th>Fuel Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>PWR-UX and -MOX, FR reference cycles (R1)</td>
<td>( \text{UO}<em>2, (\text{U}</em>{0.97-\text{Pu}<em>{0.03})O}<em>2 , (\text{U}</em>{1.27-\text{Pu}</em>{0.35})O}_2 )</td>
</tr>
<tr>
<td>PWR self-generated TU recycle (R2)</td>
<td>( \text{Pu}<em>{0.01-\text{Np}</em>{0.09}-\text{Am}<em>{0.02}-\text{Cm}</em>{0.01}-\text{Tc}_{0.01})O_2 )</td>
</tr>
<tr>
<td>FR self-generated TU recycle (R3)</td>
<td>( \text{Pu}<em>{0.01-\text{Np}</em>{0.09}-\text{Am}<em>{0.02}-\text{Cm}</em>{0.01}-\text{Tc}_{0.01})O_2 )</td>
</tr>
</tbody>
</table>

3.6.1.3.2. Minor actinide containing fuel for fast reactors: a case study (experiment SUPERFACT)

Minor actinide recycling in a Fast Reactor has been studied on a series of oxide fuels containing \( ^{237}\text{Np} \) and \( ^{241}\text{Am} \) at low and high concentrations [150]. Four types of mixed actinide oxide fuels (Table XV) were prepared at the Institute in accordance with the homogeneous (SF13, SF16) and heterogeneous (SF14, SF15) fuel concepts. The fuels have been irradiated in the PHENIX power plant and are currently undergoing post-irradiation examinations at the Institute.

**TABLE XV. FUEL COMPOSITIONS OF THE SUPERFACT IRRADIATION**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fuel Composition</th>
<th>Burnup</th>
</tr>
</thead>
<tbody>
<tr>
<td>SF13</td>
<td>( \text{U}<em>{0.01-\text{Pu}</em>{0.09}-\text{Np}_{0.07}} O_2 )</td>
<td>6.4 at%</td>
</tr>
<tr>
<td>SF14</td>
<td>( \text{U}<em>{0.05-\text{Am}</em>{0.02}-\text{Np}_{0.07}} O_2 )</td>
<td>4.1 at%</td>
</tr>
<tr>
<td>SF15</td>
<td>( \text{U}<em>{0.01-\text{Np}</em>{0.01}} O_2 )</td>
<td>4.5 at%</td>
</tr>
<tr>
<td>SF16</td>
<td>( \text{U}<em>{0.4-\text{Pu}</em>{0.04}-\text{Am}_{0.02}} O_2 )</td>
<td>6.4 at%</td>
</tr>
</tbody>
</table>

3.6.1.3.3. Methodology

The computer code KORIGEN [152] was used to calculate the evolution of the fuel during irradiation for each of the recycling concepts and minor-actinide-containing fuels. Appropriate burnup dependent fission and capture cross-sections were used for the studies in PWR. The cross-section libraries were adjusted to those for the PHENIX reactor in the case of the SUPERFACT fuels.

The neutron and gamma dose rates were calculated for the different fuels using the computer program PUDOL [153]. The calculations were performed on the basis of the fuel composition and the physical properties of the fuel and cladding. The spent fuel compositions obtained by KORIGEN were used as input to PUDOL for the dosimetry calculations. The program considers self-shielding effects in the fuel itself and attenuation in the cladding. Furthermore, it allows the use of a range of shielding materials for radiation protection purposes.

The gamma dose rates of the four SUPERFACT pins were measured during fabrication using a portable ionisation chamber. After discharge (cooling time 57 months), the gamma and neutron dose rates were measured using a passive neutron-gamma interrogation unit developed for the characterisation of spent fuel inside a hot cell at the Institute [150].
3.6.1.3.4. Comparison of the nuclear fuel cycles

In the present study, it is assumed that the fuels are fabricated one year after reprocessing. Thus, the build-up of large amounts of $^{208}\text{Tl}$ (2.6 MeV gamma) from the decay of $^{236}\text{Pu}$ (half-life 2.6 years) is avoided.

The dose rates per unit mass of the fresh fuel material corresponding to the recycling schemes for Pu and MA are compared to the PWR and FR MOX reference cycles (Table XVI). The distance of 50 cm corresponds to the working conditions when pellets are handled manually inside a glovebox. A dose rate limit for a radiation worker of 2 mSv/h is considered. The results confirm the expected increase in dose rates at fabrication due to the inclusion of Pu and minor actinides.

The difference between PWR MOX and self-generated Pu recycling schemes is due to the change in the Pu isotopic composition. Multiple recycling of self-generated Pu in PWR results in a Pu composition with a significant increase in the even isotopes. The $^{238}\text{Pu}$, increased by 3-4 times, contributes greatly to both gamma and neutron dose rates. The inclusion of minor actinides contributes to a further increase of the dose rates of the fuels R2 and R3 over the corresponding reference MOX fuels. The soft gamma spectrum from Pu and $^{241}\text{Am}$ which is important to the dose rate, can however be easily shielded by 1-2 mm of lead. In normal industrial practice, remote handling may be necessary when a large number of pellets are present inside the box.

**TABLE XVI. DOSE RATES AT 50 cm FOR ONE GRAM OF FRESH FUEL MATERIAL**

<table>
<thead>
<tr>
<th>Material</th>
<th>Gamma dose rate (mSv/h)</th>
<th>Neutron dose rate (mSv/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PWR-UOX</td>
<td>2.0 E-7</td>
<td>4.0 E-11</td>
</tr>
<tr>
<td>PWR-UOX</td>
<td>3.5 E-4</td>
<td>5.0 E-7</td>
</tr>
<tr>
<td>FR-MOX</td>
<td>2.0 E-3</td>
<td>3.0 E-6</td>
</tr>
<tr>
<td>R2</td>
<td>3.0 E-3</td>
<td>1.5 E-4</td>
</tr>
<tr>
<td>R3</td>
<td>9.3 E-3</td>
<td>7.5 E-4</td>
</tr>
</tbody>
</table>

The handling of a single PWR or FR fuel pin at fabrication involves dose rates which exceed the working limit by factors of up to twenty (Table XVII). Although the soft gamma spectrum is again easily shielded by thin Pb, the neutron dose rate demands up to 150 mm polyethylene. The above analyses have been performed for one year after reprocessing. At longer times, $^{208}\text{Tl}$ builds up and may eventually require the use of further Pb shielding. In the case of the R2 recycling scheme, the dose rate for one pin due to $^{208}\text{Tl}$ is just below (1 mSv/h) the working limit. At discharge, the gamma dose rate is determined by the burnup, i.e. the fission product content of the fuel. Dose rates comparable to the PWR-UOX and MOX fuels are observed at 1 m from the fuel, requiring 100-150 mm Pb of shielding. However, the inclusion of minor actinides in the charge composition of the fuels results in an increase of the neutron emission after irradiation. Polyethylene shielding of 200 mm is now required.

3.6.1.3.5. Verification of dosimetry predictions

The availability of computer programs enabling the prediction of dose rates is essential to estimate possible hazards in a given fuel cycle. Future industrial scale fabrication of minor actinide fuels will be planned according to calculations using computer programs. Verification of such predictions is therefore needed using the minor-actinide fuels available from small-scale irradiation experiments.

In order to assess the reliability of the dosimetry computer codes, measured radiation doses from fresh and spent fuels have been compared to the calculated ones. The gamma dose rates during fabrication are given in Table XVII for the minor-actinide fuels used in the SUPERFACT program.
The calculations were performed on the basis of the fresh fuel compositions obtained by chemical analyses. The level of agreement in Table XVIII is within 30%, which is an acceptable accuracy for radiation protection purposes. The high radiation doses from the mixtures containing americium can be seen in the results. The increased concentration of $^{241}$Am in the SUPERFACT fuels causes an increased gamma dose of 40 times for SF14 and 7 times for SF16. The former exceeds the dose limit by two and the latter by one order of magnitude. However, the soft gamma spectrum emitted by the fuels can be easily shielded by 1-2 mm Pb.

The presence of PWR and MA spent fuels in the hot cells at TUI, provided the opportunity to study their dosimetry. Furthermore, it allowed an assessment of the accuracy in predicting these rates by a computer code on the basis of the fuel compositions obtained by KORIGEN. The experimental and theoretical neutron and gamma dose rates for the spent fuels are shown in Table XIX. The corresponding final burnup and cooling times are included since they determine the radiation emissions at measurement.

### TABLE XVII. DOSE RATES AT 1 m AND NECESSARY SHIELDING FOR FRESH FUEL

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Gamma emission</th>
<th>Neutron emission</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Dose rate (mSv/h)</td>
<td>Pb shielding (mm)</td>
</tr>
<tr>
<td>PWR-UXO</td>
<td>2.0E-5</td>
<td>-</td>
</tr>
<tr>
<td>PWR-MOX</td>
<td>4.0E-3</td>
<td>1</td>
</tr>
<tr>
<td>FR-MOX</td>
<td>6.5E-3</td>
<td>1</td>
</tr>
<tr>
<td>R2</td>
<td>4.0E-2</td>
<td>2.5</td>
</tr>
<tr>
<td>R3</td>
<td>3.3E-2</td>
<td>2.5</td>
</tr>
</tbody>
</table>

### TABLE XVIII. PREDICTED AND MEASURED DOSE RATES AT 1 m FROM A FRESH FUEL

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Gamma dose rate (mSv/h)</th>
<th>Predicted</th>
</tr>
</thead>
<tbody>
<tr>
<td>SF13</td>
<td>7.0E-3</td>
<td>8.0E-3</td>
</tr>
<tr>
<td>SF14</td>
<td>2.6E-1</td>
<td>2.5E-1</td>
</tr>
<tr>
<td>SF15</td>
<td>&lt; 1.0E-2</td>
<td>2.0E-4</td>
</tr>
<tr>
<td>SF16</td>
<td>4.0E-2</td>
<td>5.6E-2</td>
</tr>
</tbody>
</table>

### TABLE XIX. PREDICTED AND MEASURED DOSE RATES FROM SPENT FUEL

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Burnup (at %)</th>
<th>Cooling time (a)</th>
<th>Gamma dose rate (mSv/h)</th>
<th>Neutron dose rate (mSv/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Predicted</td>
<td>Measured</td>
<td>Predicted</td>
<td>Measured</td>
</tr>
<tr>
<td>SF13</td>
<td>6.4</td>
<td>5</td>
<td>3.0E2</td>
<td>2.7E2</td>
</tr>
<tr>
<td>SF14</td>
<td>4.1</td>
<td>5</td>
<td>1.0E2</td>
<td>1.5E2</td>
</tr>
<tr>
<td>SF15</td>
<td>4.5</td>
<td>5</td>
<td>1.0E2</td>
<td>1.5E2</td>
</tr>
<tr>
<td>SF16</td>
<td>6.4</td>
<td>5</td>
<td>3.0E2</td>
<td>2.5E2</td>
</tr>
<tr>
<td>PWR-MOX</td>
<td>4.7</td>
<td>3</td>
<td>4.2E2</td>
<td>7.0E2</td>
</tr>
</tbody>
</table>
The increase in the dose rate following the inclusion of MA in the charge composition of fuels is demonstrated in Table XIX. The total radiation dose is dominated by the gamma dose rate. This is due to the large cooling times, particularly for the SUPERFACT fuels, and the consequent decay of $^{242}$Cm and $^{244}$Cm. Even for the SF14 fuel which contains 20% $^{241}$Am and for which most of the curium isotopes are expected to be produced, the gamma dose dominates. Nevertheless, at discharge the neutron dose rate is considerable, although still lower than the gamma dose rate. A shielding of 100-150 mm polyethylene would be required to reduce the occupational exposure at 1 m from SF14 and SF16 during unloading.

In order to assess the PUDOL predictions, the experimental and theoretical dose rates have been compared (Table XIX). An agreement within 50% is observed. The discrepancy can be attributed to different sources including the reproducibility of the experimental set-up and the fuel compositions obtained from KORIGEN predictions. The reliability of KORIGEN, for the source term nuclides, has been checked by the comparison of its predictions with chemical analyses [150]. The agreement is within 25% for the actinides and 50% for the fission products. This agreement, although sufficient for dose-rate calculations indicates limitations in its basic nuclear data libraries. The latter should be considered when neutron-physics calculations are performed.

The P&T fuel cycle involves extended reprocessing (spent fuel reprocessing for U and Pu and partitioning of minor actinides), advanced reactors, different fuel cycle operations at the front and back end of the cycle and transportation of additional nuclear material. All these aspects are expected to alter the occupational and public impact.

The radiological risk resulting from exposure to radiation can be either occupational or accidental. The occupational radiological dose would be due to gas releases from the fuel processing, fabrication and waste treatment plants. The accidental radiological risk would arise from accidents in the previously mentioned plants and during transportation resulting in the dispersion of radioactivity. An assessment of the radiological risk requires a precise knowledge of the fuel cycle under investigation.

Croff et al. [154] considered two closed LWR fuel cycles. The cycles are based on a single 1250 MW(e) PWR operating in the self-generated plutonium recycle mode. The reference fuel cycle comprises coprocessing for recovery and recycle of uranium and plutonium. The P&T fuel cycle includes, in addition to the reference cycle, partitioning processes for the recovery of all actinides in the wastes from refabrication and reprocessing plants.

The initial core of the reactor in the reference fuel cycle is made up of 33% MOX (plutonium-enriched uranium, with some neptunium) and 67% 3.2 wt% enriched UO$_2$. After an exposure of 33 GWD/t, the UO$_2$ and MOX fuel are discharged from the core, allowed to decay for 1.5 y and then transported to the fuel reprocessing plant. Uranium and plutonium are recovered by a coprocessing PUREX flowsheet. After an additional 0.5 y cooling, the recovered material is transported to the MOX fuel fabrication plant where 33% of the fuel for the next reactor loading is fabricated. The resulting waste from fuel reprocessing and fabrication are sent to a repository.

The P&T fuel cycle is basically the same as the reference fuel cycle with the exception that a partitioning plant is now included in the cycle. The waste is now sent to the partitioning plant where actinides are separated from it. The recovered actinides are incorporated into the MOX recycle streams for fuel fabrication for the next reactor loading.

The results of risk analysis for both the reference and P&T fuel cycles are shown in Tab.XX. The results in Tab. XX demonstrate that the risk from a P&T fuel cycle is higher than that from the reference cycle, generally by a factor 1.5 to 3.0. This is due to the increase in routine radiological releases, in processing and in the concentration in toxic actinides such as $^{241}$Am and $^{242,244}$Cm. The major contributor to the increase in the total risk of the P&T cycle is the risk due to routine operations.
TABLE XX. IMPACT OF P&T FUEL CYCLE ON OCCUPATIONAL AND PUBLIC RISK

<table>
<thead>
<tr>
<th>Risk source</th>
<th>Reference cycle</th>
<th>P&amp;T cycle</th>
<th>P&amp;T/Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>General Public</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Accidental</td>
<td>5.0E-7</td>
<td>7.0E-7</td>
<td>1.4</td>
</tr>
<tr>
<td>Routine</td>
<td>8.0E-4</td>
<td>4.0E-3</td>
<td>5.0</td>
</tr>
<tr>
<td>Total</td>
<td>8.0E-4</td>
<td>4.0E-3</td>
<td>5.0</td>
</tr>
<tr>
<td>Occupational</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Accidental</td>
<td>7.0E-8</td>
<td>1.0E-7</td>
<td>1.4</td>
</tr>
<tr>
<td>Routine</td>
<td>1.2E-3</td>
<td>2.0E-3</td>
<td>1.7</td>
</tr>
<tr>
<td>Total</td>
<td>1.2E-3</td>
<td>2.0E-3</td>
<td>1.7</td>
</tr>
<tr>
<td>Overall</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Accidental</td>
<td>6.0E-7</td>
<td>8.0E-7</td>
<td>1.3</td>
</tr>
<tr>
<td>Routine</td>
<td>2.0E-3</td>
<td>6.0E-3</td>
<td>3.0</td>
</tr>
<tr>
<td>Total</td>
<td>2.0E-3</td>
<td>6.0E-3</td>
<td>3.0</td>
</tr>
</tbody>
</table>

In another study [155], health risks for a P&T fuel cycle based on LMR technology are compared to the current US. nuclear fuel cycle. The reference fuel cycle is the LWR once-through one. The reactor is loaded with 4.2% enriched UO2 fuel which is operated to a final burnup of 50 GWd/t. After discharge, the spent unprocessed fuel is stored in a geologic repository.

The P&T fuel cycle is based on the advanced liquid metal reactor (ALMR)/integral fast reactor (IFR) system coupled with a uranium recycle. The relative contributions to the nuclear electricity production are assumed as 21% occurring in ALMRs and the remaining 79% in LWRs, within a centralised LWR-actinide recycle plant. The spent fuel from the LWR is reprocessed using a separation technology similar to the aqueous process. The recovered uranium is refabricated into fuel and recycled in the LWRs. The recovered TRU are used as fuel in the ALMRs. The processing plant, termed IFR, includes on-site fabrication and pyrochemical processing facilities of the ALMR metal fuel. The ALMR/IFR plant continuously recycles its own spent fuel, while the resulting HLW is stored in a repository. It is assumed further that: both front end and back end of fuel cycle are considered; the transport of LWR fuel to the repository involves more mileage than to a reprocessing plant; the ALMR fuel reprocessing and fabrication facilities will be collocated with the reactors. The calculated relative health impacts for the LWR once-through fuel cycle and for the actinide burning fuel cycle are shown in Table XXI. A decreased risk in the actinide burning cycle is observed. This is because the nuclear fuel cycle health risks are dominated by the front end uranium facilities. Transportation risk is dominated by spent fuel shipments due to the presence of volatile fission product compounds. The transport of HLW carries a lower risk because the HLW form immobilises the fission products, thus minimising releases in accident scenarios. The use of the ALMR/IFR fuel cycle reduces the dependence on uranium mines and mills as a source of fissile material. Furthermore, the ALMR/IFR health risk is expected to be lower due to the plans to recover radioactive gases (tritium, 85Kr) and the absence of significant quantities of 14C in the metal fuel. The health risk from transportation operations is significant and comparable for the reference and P&T fuel cycles.

TABLE XXI. IMPACT OF P&T FUEL CYCLE ON OCCUPATIONAL AND PUBLIC RISK

<table>
<thead>
<tr>
<th>Potential risk (health effects/GW(e)-year)</th>
<th>P&amp;T/Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>General public</td>
<td>0.70</td>
</tr>
<tr>
<td>Occupational</td>
<td>0.69</td>
</tr>
</tbody>
</table>

85
3.6.2. India

3.6.2.1. Th$^{233}$U fuel cycle

Computations have been carried out to determine the quantities of actinide waste generated for a given energy output in different reactor systems which show that by-product actinides of mass greater than $^{237}$Np generated in PHWR operating on the Th$^{233}$U cycle is about three orders of magnitude smaller than from reactors operating on a U/Pu cycle, for a given energy output. The third stage of the Indian programme based on the Th$^{233}$U cycle thus would have the advantage of considerable reduction of actinide waste; but the problem of $^{231}$Pa produced in these systems has to be suitably addressed.

However, the Th$^{233}$U fuel cycle would call for remote fuel fabrication and $^{233}$U handling technologies because of the presence of $^{232}$U contaminant. The decay of short lived daughter products of $^{232}$U is associated with high energy beta-gamma radiation.

3.6.3. Russian Federation

3.6.3.1. Plutonium problem in Russia as the most important aspect of transmutation and closing the nuclear fuel cycle in general

In all the industries the problems of raw materials and wastes are closely interconnected through secondary raw materials. Nuclear power is no exception with plutonium being the connecting link and the attitude towards plutonium is the most important factor determining various national strategies in nuclear power development. In fact, a "closed nuclear fuel cycle" is primarily plutonium transmutation. The plutonium problem is a basic concern of Russian experts dealing with transmutation, and the importance of this in closing the nuclear fuel cycle has more than once been expressed in the statements and papers on scientific and technological policy outlining Russian nuclear industry doctrine [29, 31] and research results supporting it (see for example [95, 156]).

Nuclear power infrastructure is very expensive and consequently changes slowly. Its particular national features influence attitudes toward plutonium decisively and must be taken into account in the discussions of plutonium problems at the international level. In view of the release of large volumes of weapon grade plutonium now under way, active supporters of the open NFC concept propose to consider weapon plutonium as a radwaste and a most dangerous one, the first to be neutralized. This approach is demonstrated by the Proceedings of a Moscow Seminar [157]. The subject is also extensively covered in [158]. The main argument there beside nonproliferation is economy: there still is plenty of cheap and safe natural fuel, uranium. Some of the counterarguments expressed in [95, 156] are summed up below.

In the open NFC only about one per cent of excavated uranium is used. Spent fuel is handled as waste after once-through use. In a closed NFC it is valued as a secondary fuel raw material and waste appears after reprocessing and extracting useful components, plutonium first of all. The analogy with the most valuable organic fuel, oil, may help to illustrate the main paradox of the situation. The figures below are rounded but are close to real ones. The energy content of a ton of oil is 40 GJ, price $160, specific price of energy $4/GJ. The energy content of a ton of uranium is $7.5\times10^7$ GJ, price $30,000, specific price of energy $4\times10^{-4}$/GJ, i.e. 10,000 times less. Of course, burning of uranium is much more complicated than burning of oil and relative cheapness of the fuel is a necessary component of nuclear power efficiency. But now the natural uranium contribution to the price of the final product, electricity, is next to negligible which is without precedent in organic fuels. "Uranium cream" is exhausted dozens of times faster than in a rational fuel cycle. This attitude the toward vital and urgent needs of our immediate descendants is in sharp contrast with the exaggerated and not always well founded care for the creatures who will inhabit the Earth in millions of years from now.
The quantitative side of the question looks like this (see [146]). The price $P_f$ of fresh uranium fuel cited in [147] may be presented as a linear function of two variables - the price of separative work unit $P_s$ and the price of a kilogram of natural uranium $P_{ram}$.

$$P_f = A + BP_{ram} + CP_s,$$

where $A = 240; B = 8.3; C = 5$ (all prices are given in US dollars per kilogram mass). Equating $P_f$ to the MOX-fuel price $P_{mox}$ we get an expression for the price of natural uranium at which prices of uranium fuel and MOX-fuel are equal, for different prices of a separative work unit

$$P_{ram} = (P_{mox} - A - CP_s)/B.$$

Corresponding figures are presented in Table XXII. It must be remembered that once, in the early seventies, the price of uranium (in fixed dollars) already was approximately an order higher than now without visible negative consequences for the nuclear power industry. On the contrary, it was a boom period. It is worth pointing out also that situation with secondary nuclear fuel in Russia differs in one important respect from the situation in OECD countries. For them uranium is just one of many things they can easily buy. For Russia it is one of the not so numerous things the country can sell on the international market. In the case of enriched uranium, to say nothing of ready made reactor fuel, what is sold is not mainly raw material but a product of high technology. So use of plutonium with its "negative" value on the international market instead of uranium may be a potentially very profitable process in Russia.

TABLE XXII. THE VALUES OF NATURAL URANIUM PRICE $P_{ram}$ AT WHICH THE PRICES OF URANIUM AND MOX-FUELS $P_f$ AND $P_{mox}$ ARE EQUAL, FOR DIFFERENT PRICES OF SEPARATION WORK UNIT $P_s$ (ALL PRICES IN USD)

<table>
<thead>
<tr>
<th>$P_s$</th>
<th>$P_{mox}$ 40</th>
<th>$P_{mox}$ 70</th>
<th>$P_{mox}$ 100</th>
<th>$P_{mox}$ 130</th>
</tr>
</thead>
<tbody>
<tr>
<td>1200</td>
<td>91.5</td>
<td>73.5</td>
<td>55.4</td>
<td>37.3</td>
</tr>
<tr>
<td>1400</td>
<td>115.7</td>
<td>97.6</td>
<td>79.5</td>
<td>61.4</td>
</tr>
<tr>
<td>1600</td>
<td>139.7</td>
<td>121.7</td>
<td>103.6</td>
<td>85.5</td>
</tr>
</tbody>
</table>

On the incentives of plutonium utilization

The doubts and fears of the opponents of peaceful utilization of plutonium may be briefly summarized as follows:

- channeling of weapon plutonium into the NFC does not relax but instead complicates the already far from simple technical safety problems connected with the use of reactor grade plutonium in MOX fuel;
- MOX fuel producing capacities are presently limited as well as the possibilities of using it in reactors in the near future;
- MOX fuel is uneconomic and will stay so for a long time;
- the best way to burn plutonium is in fast reactors but world interest in them is lost and will be revived not sooner than when the total nuclear power capacity reaches the level of 1000 GW(e). The initial loading for such a capacity is about 6000 tons so a few hundred tons of weapon plutonium do not solve the problem;
- weapon plutonium storage is expensive;
- the threat of plutonium use for nuclear terrorism and blackmail is increased sharply.
So the authors of Ref. [158] come to the conclusion that the troubles and dangers of plutonium utilization outweigh potential profits and the main task is not inventing ways of using plutonium (both weapon and reactor grade) but preventing it getting into the hands of potential nuclear terrorists. But there are also different points of view and corresponding national programmes. The French and Japanese first of all, attach considerable importance to the use of plutonium and minor actinides in their power industries. Some objective differences in present status and stimuli in the development of plutonium utilization methods in Russia and the USA are illustrated by Table XXIII.

### TABLE XXIII. PLUTONIUM RELATED FACILITIES IN RUSSIA AND USA

<table>
<thead>
<tr>
<th></th>
<th>USA</th>
<th>Russia</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Operating commercial NPPs with fast reactors</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>2 Operating experimental fast reactors</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>3 Operating radiochemical plants reprocessing LWR spent fuel</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>4 Same for fast reactor spent fuel</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>5 Fabrication of plutonium containing fuel pins:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>For experimental reactors</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>for commercial NPPs</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>6 Construction of a geological storage for spent fuel</td>
<td>Yes</td>
<td>No</td>
</tr>
</tbody>
</table>

It is necessary to discuss briefly the problems connected with the storage of large quantities of plutonium. Storage safety depends on the total mass of the stored material and on the reliability of the protective barriers preventing unauthorized access. Barrier penetrability (the probability of penetration) is usually determined by two factors, exponential and pre-exponential

\[ W = A(p)\exp(-B(p')) \]

where functions A and B depend only slightly, for example linearly, on the parameters p, p'. Let us postulate this admittedly oversimplified model for qualitative consideration. Factor A is approximately proportional to the mass of plutonium in this case. B(p') is an approximately linear function of the money invested in the protective barriers. The exponent must be small enough. Let us suppose it is equal to \(10^6\), for example. Then B(p)=20. It means that increase of A(p) (i.e. of the stored mass) by 20 per cent may be compensated by just a 1 per cent increase of B(p') (i.e. of barriers costs). So the continuation of small scale plutonium production in the remaining reactors is not very important if possible profits of reactor operation (heat, electricity, commercial isotopes) allow significant improvement in storage reliability. It must be also remembered that weapon grade uranium - its mass is five times that of plutonium - is not to be disposed of in any case, and before it is diluted for power industry use all the problems and dangers associated with storage will be determined by uranium. When its utilization begins some very safe storage capacity will start becoming available and may be used for the plutonium.

Being fully aware of the dangers of release of weapon plutonium we should at the same time stress that hasty attempts to get rid of it sometimes recommended, may be first of all unsafe themselves and secondly they are not clearly justified by the existing status of nuclear weapon materials storage and protection systems. Their annihilation is not dictated by any urgent necessity. Weapon plutonium incorporation in nuclear power fuel supplies should be a natural process. This material of relatively low radiation background is convenient for the initial stages of developing fuel recycling technologies and may be used for that purpose.
In any case it is clear that plutonium poses a long term, strategic problem. No prompt global solution of it exists. Hence reliable safe storage of plutonium, both weapon and reactor grade, must be ensured as before but with increasing volumes taken into account. And the ways and means of plutonium utilization must be selected within formulated national policy.

Policy may differ significantly in different countries depending on many conditions most important of which are:

- the concept of long term nuclear power development, including the strategy of transition from existing to emerging NPP designs;
- the status of the scientific and technological base of plutonium utilization in a particular country;
- historically formed details of the national nuclear industry structure.

3.6.3.2. Experience on development and production of vibropac MOX fuel for fast reactors in RIAR (Dimitrovgrad)

For the last 15 years in the State Scientific Center RIAR, a wide range of work has been performed on the validation of the fast reactor fuel cycle using pyroelectrochemical methods for nuclear fuel reprocessing and vibropac technology for fuel pin production. An experimental investigation complex has been created with remotely controlled production facilities located in shielded hot cells.

A fuel pin design was developed on the basis of uranium-plutonium oxide fuel. The fuel pin high performance features were confirmed by testing results in BOR-60, BN-350 and BN-600 reactors. The maximum burnup achieved was 26%.

3.6.3.2.1. (UPu)O₂ granulated fuel features

In RIAR, the pyroelectrochemical technology has been developed for production of UO₂ and UO₂+PuO₂ granulated fuel. The process includes dissolution of uranium and/or plutonium dioxides in molten NaCl-CsCl at 600-650 °C:

\[ \text{UO}_2 + \text{Cl}_2 = \text{UO}_2\text{Cl}_2 \]
\[ \text{PuO}_2 + 2\text{Cl}_2 = \text{PuCl}_4 + \text{O}_2 \]

Electrolysis of the melt allows for coprecipitation of uranium and plutonium dioxides:

\[ \text{PuO}_2^{+2} + e^- = \text{PuO}_2^{+} \]
\[ \text{PuO}_2^{+} + e^- = \text{PuO}_2^{(\text{solid})} \]
\[ \text{UO}_2^{+2} + 2e^- = \text{UO}_2^{(\text{solid})} \]

The process is performed in a chlorinator-electrolyzer. The maximum fuel loading into the apparatus is 30 kg. The produced cathodic deposit is crushed, salt residuals are removed and the product is certified. Table XXIV presents the granulate features.

This technology is designed for a fuel mixture with 30% PuO₂ content. Experimental investigation was made to study the possibility of production of fuel compositions containing up to 75% of PuO₂. It showed that, in this case, the granulate quality does not differ much from the standard one.

3.6.3.2.2. Development of fuel pins with vibropac (UPu)O₂ fuel

Since 1981 the BOR-60 reactor has been operated using vibropac uranium-plutonium oxide fuel. Table XXV shows the features of the tested BOR60 fuel assemblies.
TABLE XXIV. THE GRANULATE FEATURES (UO$_2$, UO$_2$+PuO$_2$)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particles density, g/cm$^2$</td>
<td>10,7-10,8</td>
</tr>
<tr>
<td>Number of fraction:</td>
<td>5</td>
</tr>
<tr>
<td>Size of particles, mm</td>
<td></td>
</tr>
<tr>
<td>-1,00 +0,63</td>
<td></td>
</tr>
<tr>
<td>-0,40 +0,40</td>
<td></td>
</tr>
<tr>
<td>-0,40 +0,25</td>
<td></td>
</tr>
<tr>
<td>-0,25 +0,10</td>
<td></td>
</tr>
<tr>
<td>-0,10</td>
<td></td>
</tr>
<tr>
<td>Mass content of isotopes, %</td>
<td></td>
</tr>
<tr>
<td>238Pu</td>
<td>0,2 - 1,7</td>
</tr>
<tr>
<td>239Pu</td>
<td>64,0 - 95,0</td>
</tr>
<tr>
<td>240Pu</td>
<td>3,3 - 21,5</td>
</tr>
<tr>
<td>241Pu</td>
<td>0,4 - 9,0</td>
</tr>
<tr>
<td>242Pu</td>
<td>0,2 - 4,0</td>
</tr>
<tr>
<td>Smear density, g/cm$^3$</td>
<td>8,8 - 9,8</td>
</tr>
</tbody>
</table>

TABLE XXV. THE MAIN FEATURES OF BOR-60 FUEL ASSEMBLIES WITH VIBROPAC (UPU)$_2$ FUEL

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel composition</td>
<td>UO$_2$ + PuO$_2$ + U (UPu)$_2$</td>
</tr>
<tr>
<td>PuO$_2$ content, %</td>
<td>20 - 28</td>
</tr>
<tr>
<td>Number of subassemblies tested</td>
<td>282</td>
</tr>
<tr>
<td>Getter mass fraction, %</td>
<td>3 - 10</td>
</tr>
<tr>
<td>Clad diameter and thickness, mm</td>
<td>6,0 x 0,3; 6,9 x 0,4</td>
</tr>
<tr>
<td>Maximum linear power, kW/m</td>
<td>52,0</td>
</tr>
<tr>
<td>Maximum clad temperature, °C</td>
<td>720</td>
</tr>
<tr>
<td>Maximum fuel burnup, % h.a.:</td>
<td></td>
</tr>
<tr>
<td>- standard subassembly</td>
<td>15,6</td>
</tr>
<tr>
<td>- experimental subassembly</td>
<td>26,0</td>
</tr>
<tr>
<td>number of subassemblies with burnup</td>
<td></td>
</tr>
<tr>
<td>10-15 %</td>
<td>163</td>
</tr>
<tr>
<td>15-20 %</td>
<td>5</td>
</tr>
<tr>
<td>more than 20 %</td>
<td>8</td>
</tr>
</tbody>
</table>

The in-pile investigations and material science examinations of the standard fuel pins containing getter of metal uranium powder as a part of fuel composition confirmed their high performance ability up to 26 % burnup. None of the analyzed cross-sections showed any thermal-mechanical or physical-chemical fuel-cladding interaction.

The analysis of radiation performance features of vibropac fuel pins showed that:

- usage of fuel composition with getter addition (UPuO$_2$ + U) allows for complete elimination of corrosion processes attributed to the presence of cesium, halogens and other possible impurities; this, in practice, excludes the possibility of any burnup limit due to physical-chemical fuel-cladding interaction;
- cladding stress in transients is several times lower and stress relaxation starts significantly earlier as compared to that of pellet fuel;
- increased smear density of a fuel column (>9.0 g/cm$^3$) provides for a sufficient temperature reserve up to the melting point;
- increased Cs migration to the low temperature fuel pin area and absence of the fuel cladding gap provide that in the case of a fuel cladding failure less volatile products will be released into the coolant in comparison with pellet fuel and the fuel will not be washed out (Table XXVI).

Two experimental subassemblies with 30 and 40% PuO₂ content were tested to study radiation aspects of plutonium utilisation (Table XXVII).

**TABLE XXVI. Cs-137 RELEASE FROM DAMAGED FUEL PINS**

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Time period</th>
<th>Average burn-up, % h.a.</th>
<th>Total Cs-137 activity increase, TBq</th>
</tr>
</thead>
<tbody>
<tr>
<td>UO₂-pellets</td>
<td>1971-1981</td>
<td>8,5</td>
<td>23,3</td>
</tr>
<tr>
<td>UO₂ (pellets) + UPuO₂ (vibropac)</td>
<td>1981-1982</td>
<td>2,8</td>
<td>1,7</td>
</tr>
<tr>
<td>UPuO₂-vibropac</td>
<td>1982-1986</td>
<td>7,2</td>
<td>38,9</td>
</tr>
</tbody>
</table>

\[a\] Total increase for a campaign. As a result of cesium capture in the cold trap during shutdowns the actual activity increase in the circuit is lower than the value indicated in the table.

**TABLE XXVII. THE MAIN FEATURES OF THE BOR-60 EXPERIMENTAL SUBASSEMBLIES WITH INCREASED PUO₂ CONTENT**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Subassembly number</td>
<td>1</td>
</tr>
<tr>
<td>Fuel composition</td>
<td>UPuO₂ + U</td>
</tr>
<tr>
<td>PuO₂ content, %</td>
<td>30</td>
</tr>
<tr>
<td>Getter mass fraction, %</td>
<td>10</td>
</tr>
<tr>
<td>Cladding diameter x thickness, mm</td>
<td>6.9 x 0.3</td>
</tr>
<tr>
<td>Maximum linear power, kW/m</td>
<td>60.0</td>
</tr>
<tr>
<td>Maximum cladding temperature, °C</td>
<td>740</td>
</tr>
<tr>
<td>Burnup, % h.a.</td>
<td>5.3</td>
</tr>
</tbody>
</table>

Pu isotope composition, % 238:239:240:241:242 = 0.5:71:19:7.5:2.0. Enrichment in \(^{235}\)U - 64.5%.

The specific feature of the radiation behaviour of fuel pins with more than 30% of Pu is its increased concentration in the periphery of the central fuel pin area that caused corrosion damage of the inner cladding surface to a depth of 70 \(\mu\)m. No chemical interaction was detected in the high temperature fuel cladding area. The severity of the effect depends on the fuel column temperature and oxygen coefficient of the fuel composition. Taking into account the importance of this aspect additional investigations are required to optimize the testing parameters and initial fuel column characteristics with different PuO₂ contents.

One of the advantages of the pyroelectrochemical method for nuclear fuel reprocessing and vibropac technology is a possibility to prepare a fuel composition both in the form of a mechanical mixture and as a homogeneous solution. This feature was utilized in preparing experimental fuel pins for investigation of minor actinide transmutation. Currently fuel compositions of (U + 5% Np)O₂ + U and UPuO₂ + Am₂O₃ types as well as actinide isotopes in oxide form are being investigated (Table XXVIII).
3.6.3.2.3. Preparation of the oxide fuel with neptunium additives

The methods of preparation of Np with UO2 or with UO2-PuO2 fuel are being developed on the basis of existing MOX fuel pyroelectrical technology.

On the basis of laboratory tests the conclusion was made that preparation of (U,Np)O2 by joint electrodeposition from melt is possible. A theoretical model of (U,Np)O2 and (U,Pu,Np)O2 electrodeposition from molten chlorides is being developed. With the purpose of preparation of experimental batches of of Np containing fuel for the irradiation tests in BOR-60 experiments are being carried out on the choice of conditions for co-deposition of about 5% of Np into the fuel.

As a result of NpO2 chlorination in the molten eutectic NaCl-2Cs-Cl at 923 K and in molten equimolar NaCl-KCl at 1000K, melts containing from 6 to 28 mass per cent of Np are obtained. The dissolution limit for NpO2 was not achieved.

The production of experimental batches of (U,Np)O2 fuel is carried out in a special laboratory facility. The working temperature of the (NaCl-2CsCl) salt was about 923 K.

The following cathode deposits were produced during electrolysis:

- 92.6 g of UO2 with 6.55 mass per cent of Np;
- 103.5 g of UO2 with 5.31 mass per cent of Np.

Fuel pins made from these mixtures are now being irradiated in the BOR-60 reactor.

For the first irradiation test of (U,Pu,Np)O2 fuel it was decided to prepare BOR-60 experimental fuel pins from mechanically mixed fuel (U,Np)O2 + PuO2. Two test batches of the fuel were prepared by electrochemical methods producing the following mechanical mixtures (in mass per cent):

- UO2-NpO2 (77-3%) and 20% of PuO2;
- UO2-NpO2 (74-6%) and 20% of PuO2.

The weight of each batch was about 200 g. The fuel pins from this fuel are being produced at present (1995).

3.6.3.2.4. Development of a method for preparation of UO2 with Am additive

Research on the electrolysis of molten salt containing U and Am has shown a possibility of Am extraction into a UO2 deposit.

The investigations on cerium (as an Am simulator) were carried out to study U and Am deposition processes from chloride melts. It was determined that in the presence of Ce oxychloride and UO2Cl2 in a molten alkali chloride mixture the formation of UO2-CeO2 solid solution occurs during electrolysis. Effects of the temperature and of the current mode on the Ce contents in the cathode deposit are being investigated.

Conditions for cathode deposit production with the Ce contents up to 2 mass per cent were found. On the basis of this research the pyroelectrochemical experiments on production of (U,Am)O2 specimens are being prepared. The investigations go on and the following tasks are planned:
- Pyroelectrical preparation of samples for irradiation tests:
  a. UO₂-NpO₂ with Np concentration about 20 per cent;
  b. Codeposited ternary fuel (U,Pu,Np)O₂;
  c. (U,Am)O₂ fuel;

- Manufacturing of a fuel with high PuO₂ concentration including fuel without UO₂.

The successful work on the development of a fuel pin of a conventional design allowed for starting a programme on the achievement of superhigh burnups (Table XXIX).

### TABLE XXVIII. ACTINIDE ISOTOPES IRRADIATED IN THE BOR 60 REACTOR

<p>| | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>232-Th</td>
<td>Zr + 241Am (20%)</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>233,235,238U</td>
<td>Al + 241Am (20%)</td>
<td></td>
</tr>
<tr>
<td>279Np</td>
<td>Mg + 241Am (20%)</td>
<td></td>
<td>235U + 237Np (60%)</td>
</tr>
<tr>
<td>238,239,240,242Pu</td>
<td>Al + 244Cm (5%)</td>
<td></td>
<td>235U + 237Np (60%)</td>
</tr>
<tr>
<td>241,243Am</td>
<td></td>
<td></td>
<td>239U + 241Am (20%)</td>
</tr>
<tr>
<td>244Cm</td>
<td></td>
<td>237Np (60%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>237Np (5%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>237Np (60%)</td>
<td></td>
</tr>
</tbody>
</table>

### TABLE XXIX. PROGRAMME OF WORK ON THE ACHIEVEMENT OF SUPERHIGH BURNUPS AND DAMAGE DOSES IN BOR-60

<table>
<thead>
<tr>
<th>Clad</th>
<th>Wrapper</th>
<th>Number of pins in SA</th>
<th>Burnup (prognosis), %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>IY</td>
</tr>
<tr>
<td>EP-450</td>
<td>EP-450</td>
<td>37</td>
<td>22,0</td>
</tr>
<tr>
<td>EP-172</td>
<td>EP-450</td>
<td>37</td>
<td>21,4</td>
</tr>
<tr>
<td>EP-450</td>
<td>EP-450</td>
<td>3*</td>
<td>24,5</td>
</tr>
<tr>
<td>EP-450</td>
<td>EP-450</td>
<td>3*</td>
<td>26,0</td>
</tr>
<tr>
<td>68 cw</td>
<td></td>
<td>4*</td>
<td>14,9</td>
</tr>
</tbody>
</table>

* - refabrication of irradiated fuel pins. Re-irradiation inside dismountable SA. The programme provides the use of promising structural materials as well as refabrication of irradiated fuel pins followed by their re-irradiation inside a dismountable fuel assembly.

3.6.3.2.5. UPuO₅ fuel pin irradiation in BN-350 and BN-600

To validate the vibropac fuel pin performance in power reactor conditions investigations were performed in BN-350 and BN-600 (Tables XXX and XXXI).
Based on the results of vibropac UPuO₂ fuel investigations in BN-350, one failed fuel pin was detected with a crack in its central part. The material science investigations of the defect fuel pin showed significant axial fuel mass transfer in some parts of the core. The fuel mass transfer appeared to be a result of a combination of increased linear rating and the minimum smear density of the fuel core (8.4 g/cm³). The fuel pins having a smear density higher than 8.6 g/cm³ did not show any anomalies in fuel behaviour.

TABLE XXX. INITIAL PARAMETERS OF THE BN-350 AND BN-600 SA

<table>
<thead>
<tr>
<th>Parameter</th>
<th>BN-350</th>
<th>BN-600</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Fuel composition</td>
<td>UpuO₂ + U</td>
<td>UPuO₂ + U</td>
</tr>
<tr>
<td>-Pu₂O₃ mass fraction, %</td>
<td>20</td>
<td>28</td>
</tr>
<tr>
<td>-Pu-239 content, %</td>
<td>95</td>
<td>95</td>
</tr>
<tr>
<td>-Enrichment in U-235, %</td>
<td>10</td>
<td>-</td>
</tr>
<tr>
<td>-getter mass fraction, %</td>
<td>5</td>
<td>10</td>
</tr>
<tr>
<td>2. Smear density, g/cm³</td>
<td>8.4-8.8</td>
<td>8.8-9.2</td>
</tr>
<tr>
<td>3. Core length, mm</td>
<td>1050</td>
<td>950</td>
</tr>
<tr>
<td>4. Cladding diameter x thick ness, mm</td>
<td>6.9 x 0.4</td>
<td>6.9 x 0.4</td>
</tr>
</tbody>
</table>

Gas leakage from two fuel pins from the second BN-350 fuel assembly appeared as a result of transport operations and was caused by degradation of the cladding mechanical characteristics due to increased swelling.

TABLE XXXI. CHARACTERISTICS OF THE INVESTIGATION OF UPuO₂ VIBROPAC SUBASSEMBLIES IN BN-350 AND BN-600

<table>
<thead>
<tr>
<th>Clad material</th>
<th>Burnup, % h.a.</th>
<th>Dose, dpa</th>
<th>Linear rating, kW/m</th>
<th>Clad temperature, C</th>
<th>Failed fuel pins</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr₁₆Ni₁₅Mo₃Nb</td>
<td>4.7</td>
<td>30.2</td>
<td>51.0</td>
<td>740</td>
<td>1</td>
</tr>
<tr>
<td>Cr₁₆Ni₁₅Mo₃Nb</td>
<td>6.8</td>
<td>42.0</td>
<td>48.0</td>
<td>690</td>
<td>2*</td>
</tr>
<tr>
<td>Cr₁₆Ni₁₅Mo₂Mn₂TiBx</td>
<td>6.9</td>
<td>53.0</td>
<td>45.0</td>
<td>670</td>
<td></td>
</tr>
<tr>
<td>Cr₁₆Ni₁₅Mo₂Mn₂TiBx</td>
<td>9.6</td>
<td>76.5</td>
<td>45.0</td>
<td>670</td>
<td></td>
</tr>
<tr>
<td>&quot;&quot;</td>
<td>9.6</td>
<td>76.5</td>
<td>43.8</td>
<td>680</td>
<td></td>
</tr>
<tr>
<td>&quot;&quot;</td>
<td>9.6</td>
<td>76.4</td>
<td>43.7</td>
<td>680</td>
<td></td>
</tr>
<tr>
<td>&quot;&quot;</td>
<td>9.6</td>
<td>76.7</td>
<td>42.3</td>
<td>680</td>
<td></td>
</tr>
<tr>
<td>&quot;&quot;</td>
<td>9.6</td>
<td>76.5</td>
<td>42.9</td>
<td>680</td>
<td></td>
</tr>
</tbody>
</table>

* As a result of transport operations.

The in-pile and material science investigations results led to the introduction of the following changes in the fuel pin design to improve its reliability and service life:

- minimum smear density value was increased up to 8.8 g/cm³;
- getter mass fraction was increased up to 10%;
- structural materials were used with higher radiation resistance.
All these changes were made in the course of manufacturing BN-600 subassemblies which were successfully tested up to 9.6% burnup.

3.6.3.2.6. Fuel pin and subassembly manufacturing

The fuel pin production procedure is rather simple and provides a minimum of technological and control operations in the hot cells. For its implementation an automated remotely controlled plant for fuel pin and assembly production on the basis of uranium and plutonium oxide fuel was developed in RIAR.

The plant equipment is located in two parallel hot cells and heavy boxes.

**Hot cell dimensions, m:**

<table>
<thead>
<tr>
<th>Hot cell 1</th>
<th>Hot cell 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>length</td>
<td>30,0</td>
</tr>
<tr>
<td>height</td>
<td>6,5</td>
</tr>
<tr>
<td>depth</td>
<td>2,5</td>
</tr>
</tbody>
</table>

The hot cells and boxes form a single chain by means of transport-technological channels. The biological shielding and engineered systems allow for working with highly radioactive materials. The equipment is designed on common principles and arranged in one line.

All technological modules can be controlled both automatically from the computer and manually from control consoles situated in the operator rooms.

The equipment located in the hot cells is used for preparation of a fuel batch, loading it into a fuel pin cladding, vibropacking, loading of upper blanket and upper end plug and sealing. The equipment of hot cell No.1 is designed for work with open fuel (except some stands). For this reason it is classified as a "contaminated" cell.

The equipment situated in hot cell No.2 is used for fuel pin inspection and subassembly assembling followed by inspection. Hot cell No.2 is considered relatively "clean". The main technological processes are implemented as a mass production line using the principle of free junction, piece-by-piece product processing and transport.

The production process starts with the delivery of granulated fuel to the stand for preparation of a fuel batch. Helium filling and fuel batch loading into a fuel pin cladding are performed within one cell. An electrodynamic vibrator is used for vibropacking of the loaded granulated fuel column. After vibropacking, the fuel pin is brought by the transportation mechanism to the stands where in succession an upper pellet blanket, a stopper and an upper end plug are loaded into the cladding. Argon-arc tungsten electrode welding is used for sealing the upper end plug to the thin-wall cladding.

Leak testing of each fuel pin is followed by inspection of the axial density distribution of the vibropac fuel column and that of plutonium along its height by measurement of $^{239}$Pu gamma emission. A combined cell is used for the inspection of surface quality and fuel pin geometry parameters: linearity, diameter, length and spacer wire pitch.

Subassembly production begins with assembling a fuel bundle. Then all necessary assembling and control operations are performed. The completed subassembly is loaded into a transport container.
In addition, at the Institute there are two technological lines situated in the glove-boxes for realization of any research programme on production of experimental fuel pins.

To date, at the facilities of the experimental-research complex, 24 BN-600 subassemblies with fuel of different compositions and more than 420 fuel assemblies of the BOR-60 reactor have been manufactured. About 3000 kg of granulated oxide fuel have been produced including 2000 kg with plutonium dioxide. The production equipment of the complex has high reliability and provides manufacturing up to 60 subassemblies per year for fast power reactors.

3.6.3.2.7. Conclusion

The large scale investigations of vibropac uranium-plutonium oxide fuel pins in the BOR-60 reactor along with successful investigations of fuel pins in the BN-600 reactor and reliable operation of the experimental-research complex allow it to be concluded that there is a good possibility for realisation of a safer, cost efficient fast reactor fuel cycle based on "dry" nuclear fuel reprocessing methods and vibropacking technology. These methods have possible application for utilization of both power and weapon grade plutonium and MA.

3.6.3.3. Some radiological aspects of the Th-based fuel cycle

The uranium-plutonium fuel cycle is adopted as the basic nuclear strategy in Russia but the Th-U cycle is considered as serious alternative for about a decade (see, for example, [159-162]). Both reactor opportunities and Th-based ADTT have been investigated [163]. Interest to the latter was intensified by publishing of the concept of an "Energy Amplifier" fueled by thorium [164].

The main advantage of Th-U cycle is the absence of Pu and heavier elements in SNF. But in fact every actinide nucleus including those formed and decaying in the Th-U cycle is a multiple alpha and beta emitter, only the "time-table" of this emission is different. This question is briefly reviewed below according to [165].

Even-even isotopes of U and Pu, if judged by their nuclear properties, (radioactivity, fission barrier height and resulting fission-to-capture ratios in different spectra, rôle in the fuel cycle) are closer to minor actinides. Isotopes of the main breeding elements, playing approximately equivalent roles in Th-U and U-Pu fuel cycles respectively are listed in Table XXXII. It is useful to compare the nuclides in the two columns of Table XXXII.

### TABLE XXXII. ISOTOPES OF MAIN BREEDING ELEMENTS IN Th-U AND U-Pu FUEL CYCLES

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Decay mode</th>
<th>Half-life (y)</th>
<th>Isotope</th>
<th>Decay mode</th>
<th>Half-life (y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}$U</td>
<td>alpha</td>
<td>68.9</td>
<td>$^{238}$Pu</td>
<td>alpha</td>
<td>87.7</td>
</tr>
<tr>
<td>$^{233}$U</td>
<td>alpha</td>
<td>1.59(5)</td>
<td>$^{239}$Pu</td>
<td>alpha</td>
<td>2.4(4)</td>
</tr>
<tr>
<td>$^{234}$U</td>
<td>alpha</td>
<td>2.45(5)</td>
<td>$^{236}$Pu</td>
<td>alpha</td>
<td>6.57(3)</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>alpha</td>
<td>7.04(8)</td>
<td>$^{241}$Pu</td>
<td>beta</td>
<td>13.9</td>
</tr>
<tr>
<td>$^{238}$U6</td>
<td>alpha</td>
<td>2.34(7)</td>
<td>$^{242}$Pu</td>
<td>alpha</td>
<td>3.76(5)</td>
</tr>
</tbody>
</table>

$^{232}$U vs $^{238}$Pu

Their primary decay properties are practically identical, almost pure alpha-emission with close half-lives, but $^{238}$Pu decays to long-lived $^{234}$U while $^{232}$U initiates a chain of 6 fast alpha decays interspersed with beta decays and ending with $^{208}$Tl, an emitter of an extremely hard gamma-line, so as a medium term hazard $^{232}$U is worse than $^{238}$Pu.
233U vs 239Pu

233U is some six time less radioactive than 239Pu, but the latter decays to practically stable 233U, while the 233U decay product 229Th starts a relatively fast chain. So total radioactivity of 233U and its products increases in the long run to almost that of 239Pu. This is important in considering a geological repository.

234U and 236U Vs. 240Pu and 242Pu

That is another clear case of a dilemma: longer periods against proportionally larger radioactivity, and it is not yet resolved. The difference is almost two orders of magnitude. The remaining line in the table is the definitive argument for the thorium cycle. 235U and 241Pu are both excellent fuel isotopes, but 241Pu decays rapidly to the highly radioactive absorber 241Am, one of the main hazards of the U-Pu cycle, together with other "minor actinides".

But is there anything in the Th-U cycle which may be considered as an analog to MA? In fact there is: radioactive thorium isotopes are listed together with MA in Table XXXIII. In the reactor case they are important only as decay products of uranium isotopes. But in the targets of accelerator-driven thorium blankets the situation may be drastically different. If the primary beam target is also made of thorium to ensure maximum efficiency of neutron production [164], reactions of the 232Th(n,xn) type become an intense source of radioactive actinides.

TABLE XXXIII. "MINOR ACTINIDES" OF Th-U AND U-Pu CYCLES

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life, years</th>
<th>Isotope</th>
<th>Half-life, years</th>
</tr>
</thead>
<tbody>
<tr>
<td>228Th</td>
<td>1.91</td>
<td>No short lived minor actinides</td>
<td></td>
</tr>
<tr>
<td>229Th</td>
<td>7.3(3)</td>
<td>235Np</td>
<td>2.14(6)</td>
</tr>
<tr>
<td>230Th</td>
<td>7.54(4)</td>
<td>241Am</td>
<td>432</td>
</tr>
<tr>
<td>231Pa</td>
<td>3.28(4)</td>
<td>243Am</td>
<td>7.38(3)</td>
</tr>
</tbody>
</table>

Cross-sections of 232Th-(n,xn) reactions (see, for example, [166]) all have maxima a few MeV above thresholds with maximum values decreasing approximately from 2 b to 1 b when going from 232Th(n,2n)231Th to 232Th(n,5n)228Th reactions. The accumulation of light Th isotopes in the region of a fast neutron cascade in a thorium proton target has been evaluated in terms of the following simple model:

1. A 10 mA, 1.5 GeV proton beam comes to a 232Th target, generating approximately 70 cascade neutrons for each incident proton.
2. Those neutrons are distributed uniformly in a 1m$^3$ block of Th producing a flux of 4.37 × 10$^{14}$/cm$^2$·s.
3. They induce 232Th(n,xn) reactions (n = 2, 3, 4, 5) producing isotopes from 231Th (decaying fast to 231Pa) to 229Th.
4. The accumulating isotopes absorb moderated neutrons in a thermal flux 10$^{14}$/cm$^2$·s and after absorption are considered lost for simplicity.

Data, needed for numerical evaluation, resulting equilibrium concentrations and accumulation half lives are given in Table XXXIV. Data in the table show that the equilibrium 230Th concentration is very close to that of the main breeding nuclide 233U, estimated in [164]. The lightest, short lived, nuclide 229Th is in the 232U chain with the same 2.62 MeV thallium gamma-line at the end, but some 30 times more radioactive (800 Ci/g) and presents an extremely serious intermediate term hazard. All in all, the Table XXXIV figures indicate that the use of a thorium primary target may create both long and intermediate term radiological problems which should be carefully evaluated. Most of this radioactivity is chemically unseparable from the bulk of the natural thorium. So low transuranics content does not automatically mean low alpha activity of thorium-uranium ADTT fuel cycle materials.
TABLE XXXIV. ACCUMULATION OF LIGHT THORIUM ISOTOPES IN Th-232(N,XN) REACTIONS DUE TO PRIMARY CASCADE NEUTRONS GENERATED BY 10 MA, 1.5 GeV PROTON BEAM

<table>
<thead>
<tr>
<th>Isotope</th>
<th>(n,xn) cross-section, spectrum averaged, mb(^a)</th>
<th>Absorption cross-sec., therm., b</th>
<th>Equilibrium concentration rel. to Th-232</th>
<th>Accumulation half-life, years</th>
</tr>
</thead>
<tbody>
<tr>
<td>Th-228</td>
<td>24</td>
<td>123</td>
<td>4.4(-4)</td>
<td>0.90</td>
</tr>
<tr>
<td>Th-229</td>
<td>38</td>
<td>84</td>
<td>1.9(-3)</td>
<td>2.63</td>
</tr>
<tr>
<td>Th-230</td>
<td>73</td>
<td>23</td>
<td>1.4(-2)</td>
<td>9.56</td>
</tr>
<tr>
<td>Pa-231</td>
<td>145</td>
<td>210</td>
<td>3.0(-3)</td>
<td>1.04</td>
</tr>
</tbody>
</table>

\(^a\) Averaging was done in [166] for half proton energy (750 GeV), but spectra vary slowly in this energy range, so available [166] figures were used for rough estimate.

It is interesting to note that a uranium target, which is an even better neutron producer than thorium, may prove to be also more acceptable radiologically, because (n,xn) reactions on depleted uranium produce lighter uranium isotopes, less immediately harmful than light thorium isotopes. One may say that uranium and the thorium situations for thermal and very fast neutrons are "mirror reflections" of one another as far as long term radiation consequences are concerned. For example \(^{238}\text{U}(n,4n)^{235}\text{U}\) reaction, as was pointed out in [167], means in fact ADT breeding of \(^{235}\text{U}\) from \(^{238}\text{U}\) in not negligible quantities. Its thorium analog leads to highly hazardous \(^{229}\text{Th}\).
4. FUTURE TRENDS

Safe management of nuclear radioactive waste is very important for the applications of nuclear energy, especially for exploitation of fission energy for electricity generation. While geological disposal in suitable repositories is considered as an unavoidable final step in any scheme of RW management, there have been particular concerns with respect to disposal of some very long lived fission product and alpha active minor actinide components of the spent fuel in nuclear power generation. Neutron transmutation of these problematic nuclides by a suitable method is being suggested as a possible way to attack this problem. It is also pointed out that these transmutation schemes may ultimately turn out to be quite attractive and economically viable since in these schemes minor actinides which are otherwise troublesome waste, also become additional fissile fuel. However, a number of important technological problems remains to be addressed and there is clearly the necessity for further active R&D in this field.

Partitioning and transmutation is a further extension of an efficient closed nuclear fuel cycle and, therefore, with future R&D in this field it is expected that this technology will be fully developed in the coming years to solve fuel problems of the nuclear industry and satisfy world energy needs. Rational use of uranium and thorium natural resources is impossible without serious, eventually multiple, recycling. It is especially true for large countries with large population and growing nuclear power programmes for electricity generation - China, India, Russia.

Active programmes in partitioning and transmutation research are now going on internationally, covering practically all the relevant aspects. Some of these important research areas are:

- radiochemistry of partitioning;
- fuel fabrication;
- physics of fast burner reactors;
- liquid metal coolants technology;
- nuclear data;
- advanced fuel cycles including those which are thorium based;
- accelerator driven transmutation technology.
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ABBREVIATIONS

(Names of computer codes, chemical processes, data libraries, research projects and programs as well as commercial companies are not included)

ADT     accelerator driven transmutation
ADTT    accelerator driven transmutation technology
ALMR    advanced liquid metal reactor
BARC    Bhabha Atomic Research Centre, India
BFS     large physical assembly at IPPE, Russian Federation
BWR     boiling water reactor
CEFR    Chinese experimental fast reactor
CNDC    Chinese Nuclear Data Center
CIAE    Chinese Institute of Atomic Energy
CRIEPI  Central Research Institute of Electric Power Industry, Japan
EDMB    Experimental Design Bureau of Machine Building, Russian Federation
FA      fuel assembly
FBR     fast breeder reactor
FBTR    fast breeder test reactor
FP      fission product
FR      fast reactor
HFR     high flux reactor
HLLW    high level liquid waste
HLW     high level waste
ICRP    International Commission on Radiological Protection
IHEP    Institute of High Energy Physics, Russian Federation
IPPE    Institute of Physics and Power Engineering, Russian Federation
ISTC    International Science and Technology Center
ITEP    Institute of Theoretical and Experimental Physics, Russian Federation
JAERI   Japan Atomic Energy Research Institute
KMRR    Korean multi-purpose research reactor
LANL    Los Alamos National Laboratory
LLFP    long lived fission products
LLW     low level waste
LWR     light water reactor
MA      minor actinides (neptunium, americium, curium)
MLW     medium level waste
MOX     mixed oxide
NEA     Nuclear Energy Agency, OECD
NFC     nuclear fuel cycle
NNDC    National Nuclear Data Center
NPP     nuclear power plant
NSF     nuclear spent fuel
OINPE   Obninsk Institute of Nuclear Power Engineering, Russian Federation
OECD    Organisation for Economic Co-operation and Development
P&T     partitioning and transmutation
PNC     Power Reactor and Nuclear Fuel Development Corporation, Japan
PWR     pressurized water reactor
KRI     V.G. Khlopin Radium Institute, Russian Federation
RIAR    Research Institute of Atomic Reactors, Russian Federation
RW      radioactive waste
SNF     spent nuclear fuel
BRIIM   A.A. Bochvar Research Institute of Inorganic Materials, Russian Federation
RIEP    Research Institute of Experimental Physics, Russian Federation
RDIPE   Research and Design Institute of Power Engineering, Russian Federation
TUI     Transuranium Institute (European Union)
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>TRU</td>
<td>transuranics (transuranium elements)</td>
</tr>
<tr>
<td>WWER</td>
<td>pressurised water reactor (Russian design)</td>
</tr>
<tr>
<td>Contributor</td>
<td>Institution</td>
</tr>
<tr>
<td>-------------------------</td>
<td>------------------------------------------------------------------------------</td>
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<tr>
<td>Arkhipov, V.M.</td>
<td>International Atomic Energy Agency</td>
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<tr>
<td>Borisov, V.V.</td>
<td>Ministry of Russian Federation for Atomic Energy</td>
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<tr>
<td>De Raedt, C.</td>
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<td>Gibson, I.H.</td>
<td>Mortec Associates, United Kingdom</td>
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<td>EDF, France</td>
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<td>Hron, M.</td>
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<tr>
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