

A COMPARATIVE SYSTEMS-ANALYSIS APPROACH ON FUEL CYCLES WITH PARTITIONING AND TRANSMUTATION

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Abstract

A system's analysis approach aims at the definition of the optimum strategy in future P&T research and development activities. Since all nuclear electricity production leads to the production of waste, a general discussion must lead to an agreement on the risk analysis assessment. Two concepts are in opposition: the intrinsic radiotoxicity versus the site specific risk analysis.

The objectives of the P&T systems analysis are:

- provide a synthetic and comparative overview of current projects
- investigate the feasibility of the proposed approaches
- bridge the gap between waste management issues and P&T options
- select promising developments for further international cooperation.

The fuel cycle with P&T should be compared with the currently practised fuel cycles: the once-through fuel cycle with direct disposal and the conventional fuel cycle with U+Pu recovery and MOX use in LWRs and disposal of vitrified waste.

In the once-through cycle the criticality issue and the long term stability of spent fuel are the most important radiological safety aspects to be assessed.

The conventional fuel cycle reduces Uranium mining needs and the resulting radiologic impact of the mill tailings. The short term increase of dose to man in LWR-MOX fabrication and the longterm impact of depleted and reprocessed uranium should also be discussed.

The separation and recycling of Minor Actinides from HLLW will reduce the intrinsic residual radiotoxicity of vitrified waste, but may not influence the risk analysis.

Separation of actinides and a recycling strategy to stabilize the total actinide inventory is probably the most ambitious goal of P&T in the present circumstances.

The longlived fission products can only partially be eliminated either by dilution e.g. I129, or by transmutation (Tc99), in dedicated very specialised facilities and at very high cost.

The advanced fuel cycle with a complete set of waste recycling steps should be analysed from comparative cost benefit point of view. Fully new technologies e.g. actinide burner reactors and accelerator-driven-transmutation facilities can only be assessed in a generic way since their development is still in their infancy. A very extensive set of parameters might influence the outcome and the credibility of the comparative analysis.

Adaptation of existing technologies will be more easily approached: a.o. reprocessing of high burnup fuel from LWR-MOX and FR-MOX, fabrication of concentrated actinide targets and fuels especially those containing Pu238, Am241, Cm244., processing losses etc but are fundamental issues in the judgment on P&T.

The realization of a comprehensive systems analysis study will require contribution from all involved countries and require the participation of a multidisciplinary task force.

1. DEFINITION OF PARTITIONING AND TRANSMUTATION

Partitioning is a complex series of Chemical and /or Metallurgical operations, intended to separate selected radiotoxic nuclides or groups of nuclides from the bulk of radionuclides occluded in the spent fuel elements which are periodically discharged from a nuclear reactor. The separated nuclides or group of nuclides can be stored as such or transformed into new fuel elements or irradiation targets.

Transmutation is the general term covering as well elementary nuclear transmutation as a result of single neutron capture or more complex phenomena as fission of heavy nuclei, spallation and other nuclear reactions involving neutrons produced in nuclear reactor cores or in subcritical nuclear facilities connected to high energy proton accelerators. The ultimate aim of the transmutation is to reduce the radiotoxicity by transforming the initial nuclide either into short lived radionuclides or into stable isotopes.

2. ROLE OF P&T IN THE FUEL CYCLE SCENARIOS.

2.1 The Once Through Cycle (OTC)

The OTC is the scenario by which all spent fuel discharged from a nuclear reactor is stored in engineered facilities till sufficiently cooled to be transferred to a permanent disposal site in deep geological formations.

The OTC scenario implies that with continuing nuclear power throughout the world the quantity of Uranium mining remains at present day levels, and that the residual fissile material content of the spent fuel (1% Pu 0.8%U235) is lost for ever.

The main longterm potential radiotoxicity is associated with the actinide elements particularly the TRU nuclides (Pu, Np, Am, Cm...) which constitute over periods of hundred thousands of years the radiological source term within the geosphere. However the intrinsic insolubility of actinides in deep geological formations reduces the effective contribution of the TRUs to the longterm dose to man.

The fission products (FP) are in the short term the most limiting factor in designing the repository facilities due to the decay heat emission which increases proportionally with the burnup. After some 300 to 500 years the major part of the FP have decayed except for some longlived nuclides (Cs135, Tc99, I129, Zr93...) with half lives of hundred thousands to million years. Some of them are relatively mobile in the geosphere and contribute to the dose to man in the vicinity of repository structures or may even enter a worldwide circulation.

The radiologic impact of the OTC can only be controlled by man made barriers which are as longlived as the radiological source term they confine. The very long time periods involved require a full public acceptance of such scenario which goes far beyond the reference lifetime of a geologic repository i.e. 10,000 years.

At the present time there is no worldwide agreement on the time intervals for confinement of high level radioactive wastes in a geologic repository. Periods of 100000 years and longer have in the past been considered as a necessary or a wishful target but no internationally accepted confinement periods have been established.

The working group might investigate the consequences of the duration of the confinement period in a geologic repository on the requirements for a P&T approach.

A number of specific regulatory and safety aspects are associated with the OTC scenario:

-the maximum inventory of spent fuel is (in the US) limited to 70000 THM per repository.
-there is an inherent criticality potential associated with the quantity of fissile material in the repository

-the longterm heat emission on the surrounding geosphere due to the TRUs.

Indirectly the OTC scenario implies the continuous production of mill tailings which accumulate at the mining sites.

By not recycling fissile or fertile materials the energetic significance of the nuclear energy in the global world energy economy is rather limited. **P&T is incompatible with OTC.**

2.2 Conventional Fuel Cycle with Reprocessing and U- Pu recycling. (CFC)

Since natural Uranium contains only 0.72% of fissile U235 isotope, the recycling of U -Pu from spent fuel through the CFC has been from the beginning of the nuclear era the standard scenario of nuclear energy production. Since 1972 this policy has been rejected by the US administration for economic and non-proliferation reasons.

The CFC scenario incorporates the "reprocessing" step which aims at the recovery of U and Pu from dissolved fuel elements and the transfer of FP and Minor Actinides (M.A.=Np,Am,Cm) to the High Level Liquid Waste(HLLW) which is stored for a number of years till vitrification of this highly active solution. The glass-blocks are stored in engineered facilities till their transfer as HLW to a geological repository.

By proceeding according to this CFC scenario the major fraction (99.7%) of the U and Pu streams are extracted and only a very minor fraction of the so called "major actinides" are transferred to the HLLW (and consequently to the HLW) and eventually to the geologic repository.

Partitioning of Minor Actinides (M.A.) from HLLW in order to further reduce the radiological potential of HLW has been studied since the 1970ies. Initially the R&D activities were focussed on the quantitative removal of MA in order to eliminate the need for any longterm storage or final disposal in geologic formations. This option was abandoned, because unrealizable. However if the public and/or political acceptance of very long term disposal of HLW could not be obtained, the removal of MA from HLLW is a technical solution which might reduce the residual radiotoxicity of the HLW. With increasing burnup the generation of MA becomes more and more important.

The addition of a MA Partitioning module to the standard reprocessing plant would in such a case be the most obvious step to be added to the current CFC.

Countries with a reprocessing infrastructure (France,UK,Japan,Russia,China) and their associated partners could in a medium term realize a partial partitioning scenario by which the actually produced HLW would be practically free from longlived TRUs.

However the question arises what to do with the recovered U,Pu, and TRU fractions ?

The countries which chose to reprocess their spent fuel did this with the main purpose to recover the major actinides (U and Pu), to save on fresh uranium purchase (20%) and to use

the residual fissile components of the spent fuel (1% U235,1%Pu) corresponding to about 25 % of the regular SWU expenses in the uranium enrichment step.

For a number of decades the Pu recycling was envisaged in a Fast Reactor (FR) option but for technical, economic and political reasons this longterm option of nuclear energy production has been slowed down and sometimes even put to an end.

The stock of Pu already accumulated at the reprocessing plants and which was intended to be used in LMFBRs became redundant in a cheap uranium market economy.

Some years ago the recycling in LWRs of Pu recovered in the reprocessing operations became an industrial practice. The use of LWR-MOX in a CFC option got industrial significance in Western- Europe where increasing quantities of PuO₂ were transformed into LWR-MOX fuel and irradiated in specially licensed reactors in France, Germany, Switzerland and Belgium.

The reuse of Pu is to a certain extent a first step in a global P&T scenario which has to be brought into broader perspective of reuse of resources and reduction of the radiotoxic legacy from the nuclear age. The recycling of Pu as LWR-MOX is a posteriori a step to recover the expenses spent during the conventional reprocessing step by producing a fraction of the electricity in a subsequent reactor core. But from radiotoxic point of view the overall gain is rather limited since only ~ 25% of the recycled Pu is fissioned and about 10% is transformed into a longterm radiotoxic MA source term.

Spent LWR-MOX is for the time being the most important radiotoxic source term which has been reduced in overall mass (a factor of about 5) but not significantly in total radiotoxicity. Multiple recycling of LWR-MOX is theoretically possible in LWRs but the resulting radiotoxicity increases throughout the subsequent recycling campaigns.

In order to reduce the global radiotoxicity it is necessary to transfer the TRU content of spent LWR-MOX fuel into a FR-MOX fuel cycle scenario which becomes a part of the next-generation scenario: the AFC.

In the meantime retrievable storage of spent LWR-MOX fuel is the most appropriate fuel cycle option till a fast Burner reactor technology will become industrially available.

Reprocessing of spent LWR-MOX fuel in view of recycling the TRUs in a FR energy scenario is conceptually possible in present reprocessing plants but these are equipped with extraction facilities for U+Pu recovery and transfer the MA fraction to the HLLW. Since the MA generation in spent LWR-MOX fuel is much higher than that in LWR-UO₂ fuel the radiotoxicity of HLLW resulting from LWR-MOX reprocessing is much more important.

The potential radiotoxicity of HLLW produced during reprocessing of LWR-MOX fuel would require an additional TRU separation module to significantly reduce the residual radiotoxicity of HLW in comparison with spent LWR-MOX fuel. The same technology as that proposed for HLLW from LWR-UO₂ would be adequate.

The recovery of U from spent fuel was from the beginning of the nuclear era an obvious option which was followed because the extraction in the PUREX process was carried out with TBP. For many decades U has been recovered during reprocessing but very little of this stockpile has been reused in subsequent reactor loadings.

Reprocessed U contains some disturbing radioisotopes (U234 and U236) which are the mother isotopes of natural decay chains with radiotoxic decay nuclides. Any delay in recycling reprocessed U leads to the buildup of the (4n+2). Radium chain from U234 with outspoken residual radiotoxicity due to Ra226. The buildup of U236 in reprocessed U, has economic consequences, since it is a neutron poison which decreases the reactivity of reprocessed U versus natural U. For all these reasons and particularly because fresh mined U became so cheap, reprocessed U has not been recycled on an industrial scale and might become a medium level waste type which cannot be disposed of in surface repositories.

If there is no medium term solution to this potential U waste stream, the residual radiotoxicity of reprocessed U will surpass that of Np237, one of the MA.

Recovery of Np237 from the U- Pu product stream which is technically possible in the PUREX process, might this way become unnecessary from radiotoxic point of view. This problem ought to be investigated from many viewpoints to end up in a recommendation for the elaboration of the AFC scenario. Indeed one of the TRU nuclides with the longest half life (2,140,000 years) is Np and very few natural barriers are capable to confine this radiotoxic element over such an extended time period.

The quantitative removal of Am241,243 from HLLW resulting from the reprocessing of LWR-MOX is without any doubt the most important task to be investigated, since these nuclides determine to a major extent the potential radiotoxicity of this type of HLW within the time interval of 500 to 4000 years. Part of the Np 237 problem is solved when Am241, its mother isotope, has been quantitatively separated from HLLW before vitrification.

As a conclusion we might say that the recovery of TRUs from spent LWR-MOX fuel would be a first significant step in implementing the P&T option for LWR fuel . The costs incurred in this option might become prohibitive unless the dose-benefit/cost-increment ratio remains acceptable in comparison with the Pu extraction and recycling.

2.3. Advanced Fuel Cycle with TRU recycling. (AFC)

P&T has to play an essential role in the future AFC which is intended to reduce as much as possible the residual radiotoxicity of the Waste streams. The ideal AFC scenario with P&T will be treated in this note as a long term objective to be reached, knowing that economic constraints will damp the enthusiasm for this futuristic option.

Any AFC scenario must rely on the use of reprocessing of spent fuel as an indispensable first step.

A comprehensive AFC scenario with **Partitioning and Transmutation** comprises the following steps:

- Conventional reprocessing of LWR-UO₂ fuel.
- Separation of MA from HLLW resulting from LWR-UO₂ reprocessing.
- Fabrication of MA targets for heterogeneous irradiation in LWRs.
- Separation of certain fission products with long half lives if required for the disposal step.
- Quantitative recycling of U and Pu into LWR-MOX fuel. (single or multiple recycling)
- Reprocessing of spent LWR-MOX fuel in adequate facilities (higher Pu inventory).
- Separation of MA from HLLW and conditioning of individual elements. (Np, Am, Cm)
- Fabrication of FR-MOX fuel with limited MA content.
- Irradiation of FR-MOX fuel in Fast Burner Reactors. (very high burnup)
- Reprocessing of spent FR-MOX fuel in specially designed and licensed facilities.
- Quantitative separation of all TRUs from the spent FR fuel solution.
- Revision of the fission product management: Tc99 separation (head end, HLLW)
- Investigation of the radiological impact of the minor long-lived Fission Products.
- Impact of Platinum metals separation on the overall economics of Partitioning.
- Multiple recycling of FR-MOX fuel with major TRU content until significant depletion.

The most important of these steps will be briefly commented:

2.3.1 Conventional reprocessing of LWR-UO₂ fuel.

The current separation factors obtained for major Actinides (99.7%) are in a first approach sufficient to reduce their content in HLLW. The only improvement which might have a significant influence on the longterm toxicity and waste management is the reduction of Medium Level Waste (MLW) which is not vitrified before disposal and occupies a rather large volume compared to HLW.

2.3.2 Separation of MA from HLLW.

The current reprocessing technology does not separate quantitatively all the MA. Am and Cm (+ shorter lived TRUs, Bk, Cf...) are quantitatively (>99.5%) transferred to HLLW but Np is partly transferred to the High Active Feed (HAF) solution and partly directed towards the HLLW.

The partitioning of Np needs further chemical extraction steps to insure a quantitative transfer to one single waste or product stream, from which further separations could be performed. Partitioning of all MA from HLLW is presently under investigation in many laboratories throughout the world. (Japan, France, China and some minor nuclear countries) and was studied formerly in the US national laboratories (ANL, ORNL, Hanford...).

Several processes have been studied at the conceptual level and tested in hot facilities among the most important are: the TRUEX, DIDPA, TRPO and DIAMEX processes.

The systems analysis should emphasize the merits and drawbacks of each of these approaches in existing reprocessing facilities or the adaptations to be made to the current flowsheets and their translation into technological modifications to be implemented in present facilities or to be designed in future reprocessing plants with incorporated Partitioning steps.

Separation of Am(+Cm) is from radiotoxic point of view the first priority, but the Am(Cm) fraction contains all the Rare Earth (RE) elements which are in terms of quantity, 10 times more important than actinides. (12.4Kg RE compared to 1.27 Kg MA). In order to obtain a TRU fraction with 90% purity a separation factor of 100 is required for the RE fraction. A 99% purity involves a separation factor of 1000, which is a technical limit for elements such as the Lanthanides and the Actinides, with very similar chemical properties.

2.3.3. Fabrication of MA targets for heterogeneous irradiation in LWRs.

In the medium term only thermal reactors and particularly LWRs are available for irradiation of MA targets. Fabrication of irradiation targets with industrially representative quantities of MA are difficult to prepare even in pilot-scale hot-cell facilities.

Experience has been gained in the production of isotopic heat sources, but the radiologic context and the ALARA limitations to be expected from regulatory bodies on industrial activities are probably very different from what has been done for military- and space applications.

The presence of large quantities of Am²⁴¹ accompanied by 1 to 10 % RE will require fully gamma shielded and remote operated fabrication facilities. The presence of 5% Cm²⁴⁴ in an Am²⁴¹⁻²⁴³ target will multiply the degree of technical complexity due to the additional neutron shielding resulting from the spontaneous fission rate and from the alpha-n reaction in oxide-type isotopic targets.

The report should emphasize the order of magnitude of advanced fuel fabrication capacity required to cope with the potential output from an industrial advanced reprocessing plant, equipped with an Am(Cm) separation module. Recycling of such targets to achieve a significant TRU depletion in the target could be discussed in connection with the issue of multiple recycling of fuels and targets (see section 2.3.9) in fast reactors.

2.3.4. Separation of long-lived fission products.

A number of radiologically important fission/activation products play a potentially important role in the assessment of a geologic repository. In order of importance the following nuclides have to be assessed: the fission products Tc99, I129, Cs135, Se79, Zr93, and the activation product C14.

Tc99 is a fission product with a half life of 210,000 years which occurs as Tc (TcO₂) metal in the insoluble residues and as Technetate ion in the HLLW solution. In order to effectively address the longterm radiotoxicity problem both fractions ought to be combined before any nuclear action is taken towards depletion by transmutation. The similarity between Tc and the Platinum metals and the nature of the separation methods (pyrometallurgical techniques) makes this partitioning operation very difficult. Tc displays only a radiotoxicological hazard when submitted to oxidative underground conditions (Tuff, Yucca Mountains). In reducing deep underground aquifers the migration of Tc99 is negligible.

I129 is separated from the HLLW during the conventional reprocessing operations. The separated fraction can either be stored on a specific adsorbent or discharged into the ocean. Since I129 has a half life of 16 million years it will in any scenario enter into a worldwide dispersion in the geo- or biosphere. I129 is one of the critical nuclides when considering landbased repositories of spent fuel. In a worldwide dispersion scenario its radiotoxic importance is rather limited.

Zr93 and **Cs135** are two longlived (1.5 resp. 2 million years half-life) nuclides occurring in spent fuel. Separation of these radionuclides from the other fission products for eventual transmutation is almost excluded since they are accompanied by other radioisotopes which are very radioactive (Cs137) or are present in much larger quantities (736 g Zr93 with 3327 g Zr per THM). In order to effectively reduce the radiotoxic potential by neutron irradiation a series of isotopic separation processes ought to precede any target fabrication and this route is presently considered as an almost impossible endeavour from economic point of view.

C14, with a half life of 5730 years, is a difficult case because it can potentially enter into the biosphere through its solubility in groundwater and play an important radiotoxicological role because of its incorporation into the biochemical life cycle. Its role in the longterm radiotoxicity is dependent on the physico-chemical conditions occurring in deep underground aquifers or in water unsaturated geospheres.

Some radionuclides discussed in this section ought to be examined in depth by the working group in order to establish **their risk and potential radiotoxic role** in comparison with the TRUs. Their radiotoxicity is between 1000 and 100,000 times less important than TRUs but their contribution to the long term risk is predominant because migration to the biosphere may be much more rapid and generate in the very long term a certain radiation dose to man.

2.3.5 Quantitative recycling of U and Pu into LWR-MOX fuel.

Up to now it has been assumed that in the CFC scenario all major actinides (U+Pu) were recycled and used in a next fuel loading scheme: LWR-MOX or FR-MOX. For reasons explained above this has not yet been realized and large quantities of reprocessed U and recovered Pu are still in the engineered storage facilities.

The management of Pu has recently been examined by a OECD-NEA working group and the conclusions should be used to establish the most probable scenario for the recovery and future use of this very controversial spent fuel constituent.

Three alternatives are open -CFC with partial use of U+ Pu as LWR-MOX

- CFC with use of U+ Pu as feedstock for FR-MOX

- AFC with quantitative recovery of U and Pu for LWR-MOX followed by its transfer to FR-MOX at a later stage.

The working group should assess whether the AFC scenario should preferably be situated as a follow up of the LWR-UO₂ reprocessing step or whether it is indicated to transfer all the Pu first to the LWR-MOX step before starting a FR-MOX recycle. If quantitative recovery of all major actinides is not a viable route, it will have a direct impact on the overall separation factor to be achieved for TRUs in general and MAs in particular.

Recycling, or not, of U has a direct influence on the significance of separating Np from spent fuel solutions. The degree of Pu recycling will directly affect the radiotoxic significance of TRU partitioning.

2.3.6 Reprocessing of LWR-MOX

Up to now the reprocessing of LWR-MOX has been done by diluting the LWR-MOX fuel with LWR-UO₂ fuel according to the ratio it occurs in the reactor-core. (UO₂/MOX=2). Reprocessing of spent LWR-MOX can be performed industrially if the reprocessing plant has been licensed for the treatment of increased Pu concentrations and a much higher total Pu inventory.

The radiotoxicity of spent LWR-MOX is ~ 8 times higher than that of LWR-UO₂. Conventional reprocessing will remove U+Pu which accounts for about 30% of the total alpha activity the residual 70% enters into the HLLW stream and is composed of Cm²⁴⁴ and Am²⁴¹⁺²⁴³.

In a perspective of P&T it would be indispensable to remove the TRUs from the HLLW before vitrification. The techniques to be used are in principle the same as for the LWR-UO₂ fuel (see 2.3.2) ,but the higher alpha activity level will interfere with the extraction because of increased radiation damage.

Another option is to store the spent LWR-MOX fuel for example during 50 or more years and to let Cm²⁴⁴ decay (18 years half life) to Pu²⁴⁰ before carrying out the reprocessing. The chemical extraction processes are after the "cooling" period much easier as the alpha decay heat is reduced by a factor of 7 or more.

2.3.7 FR-MOX Fuel fabrication with limited TRU content.

The largest industrial experience has been gained in the FR-MOX fuel fabrication since for several decades FR programmes were undertaken in many nuclear countries. The fabrication of

FR-MOX fuel with 15 to 25 % Pu has been realized routinely. But the Pu quality used for these purposes was derived from low burnup UO₂ fuel with low Pu²³⁸ and Pu²⁴¹ contents..

In a perspective of the use of advanced FRs (CAPRA) still higher Pu concentrations are envisaged. (up to 45%)

In the mean time the burnup of spent LWR-UO₂ fuel has increased to 45 MWd/THM and that of LWR-MOX may reach 50 MWd/THM. The isotopic composition of Pu resulting from the reprocessing of such fuels is seriously degraded. High Pu²³⁸ and Pu²⁴², low Pu²³⁹ and Pu²⁴¹. A thorough study of the issues involved was published in 1989 by OECD-NEA and is still valid as source book for the working group on P&T. (PLUTONIUM FUEL An Assessment)

The recycling of FR-MOX containing high Pu²³⁸ levels and limited amounts of MA is still more difficult and requires the design and construction of remotely operated fuel fabrication plants.

For homogeneous recycling of MA in FR-MOX, admixtures of 2.5% Np²³⁷ and/or Am²⁴¹ are currently studied. Since Np²³⁷ is a pure alpha emitter there is no handling problem involved; but the admixture of Am²⁴¹ at the 2.5% level will induce a gamma field around the glove boxes or hot cells. However the major interfering nuclide is Pu²³⁸ at the 3% level which is a heat and neutron source (5KWth/THM; 5.10 E8 neutrons/s per THM.)

The FR-MOX fuel fabrication with limited TRU admixture will also be influenced by the degree of separation of the Rare Earths (strong gamma emitters) and last but not least by Cm²⁴⁴ which will accompany Am²⁴¹ and Pu²⁴³ when separated from HLLW. The presence of even small Cm²⁴⁴ impurity levels will boost the neutron emission of the resulting fresh FR-MOX fuel.

The working group should establish which separation coefficients ought to be obtained from Rare Earths and from Cm²⁴⁴ to allow for industrial fuel fabrication operations.

Heterogeneous recycling of MA means to avoid the dilution of troublesome nuclides, e.g. Cm²⁴⁴, throughout the fuel fabrication step and carry this operation in small but dedicated facilities.

2.3.8 Metal Fuel fabrication for ALMRs and advanced fuels for Burner reactors

In the frame work of the Integral Fast Reactor project a specific fuel fabrication technology has been developed and tested on cold (and hot) pilot scale. At the FFTF facility metal fuel has been recycled by casting a UPu-Zr alloy on laboratory- and hot pilot scale.

It is obvious that these processes are still in the exploratory stage and cannot be considered as proven technology but their potential should be investigated since metal fuel permits to reach very high burnups and has good neutronic characteristics for transmutation of TRUs.

The working group could summarize the results obtained and draft a series of recommendations for further work in the countries (Japan, and possibly the US) involved in this type of pyrometallurgical development.

Very recently the attention was drawn on the potential of Nitride and Carbide fuels for Fast Burner Reactors. (FBuR)

Nitride TRU fuel containing macroscopic quantities of MA can be produced by a combination of an internal gelation method and a carbothermic synthesis. These nitride fuels can be reprocessed by electrorefining methods similar to the technology developed for metal fuel.

A short analysis of the potential and limitations of this type of new fuel cycle should be included.

2.3.9. Fast Burner Reactors (FBuR)

In the frame work of P&T the Fast Breeder Reactor (LMFBR) is not a goal in itself because it produces more TRUs than initially present in the reactor. But very much technological experience has been accumulated since thirty years of R&D&D all over the world which can be transferred to FBuR technology.

The decision of the french government to modify the objective of the Super-Phenix (SPX) reactor

will have far reaching consequences on the future FR programme. Gradually the large SPX prototype reactor working with FR-MOX fuel, will be used as a semi industrial tool to investigate the feasibility of accelerated TRU burning. Therefore it is necessary to modify the reactor configuration (removal of blankets) and even to fundamentally change the fuel and core design. The recently launched CAPRA project will primarily investigate accelerated Pu burning technologies (very high Pu concentrations in the fuel), but MA incineration is also considered. In order to reduce as much as possible the TRU formation, U free TRU fuel with inert matrices are receiving increasing attention.

Several years ago from 1988 on, the Japanese government launched new transmutation projects in the framework of the OMEGA project. The Minor Actinide Burner reactor (MABR), the Particulate Bed Burner Reactor (P-ABR). These projects are being further developed but apart from FR-MOX fuel other types e.g. Nitrides and Carbo-nitrides are investigated as possible alternatives.

The IFR project of ANL evolved into an industrial sodium cooled FR prototype design called the PRISM reactor which is supposed to use metallic fuel (UPuZr) with adjacent pyrometallurgical reprocessing facilities to recycle TRU fuel and discard the fission products. Core design studies and core performance analysis are underway to increase the TRU incineration.

The working panel should comparatively investigate:

- the transmutation capacity of TRUs as a function of neutron fluence and spectrum
- the influence of burnup on the fuel behaviour and the cycle time
- the overall mass balance of TRU input and output inventories
- the evolution of the residual radiotoxicity
- the fuel recycle aspects, total cycle inventory, TRU depletion, residual core content.

This analysis would lead to a better understanding of the relative merits and drawbacks of each conceptual design and help the international nuclear community to better grasp the significance and purpose of the many new reactor concepts which were launched in the frame of P&T.

2.3.10 FR-spent fuel reprocessing

The reduction of the radiotoxic inventory resulting from multiple recycling of spent FR-MOX or FR-Metal fuel is only possible if repeated reprocessing of FR-spent fuel is part of the AFC scenario. Irradiation of FR-MOX or FR-Metal fuel is carried out till very high burnup levels. Compared to LWR-MOX fuel the target burnups of FR-fuel are 2 to 3 times higher, i.e. burnups ranging from 120 to 180 MWd/THM are currently to be envisaged, and even higher burnup

levels should be attained in a more distant future. But even these very high burnup levels still correspond to a rather small transmutation or incineration %.

After a prolonged irradiation campaign of e.g. 5 years and a burnup of 150 GWd/THM the degree of depletion is only about 15 to 20 % of the initial TRU inventory. In order to reach an overall TRU depletion yield of 90% it is necessary to recycle the FR-spent fuel between 15 to 17 times according to the expected percentage of losses during each of the chemical extraction operations.

Any reprocessing campaign of spent FR-fuel based on the use of the PUREX process or on an equivalent aqueous extraction process cannot be carried out within a short time interval after the discharge from the fast reactor because the decay heat is much too high. Cooling times between discharge and reprocessing are, as a result, increased from 7 to 10 or 12 years.

The duration of the total AFC cycle becomes 15 to 17 years per cycle. The time interval to reach a global depletion of 90% is consequently of the order of 250 years.

Pyrochemical reprocessing of spent FR-fuel was developed in the frame of the IFR project. In this case FR-Metal fuel irradiated at very high burnup is transferred to a "on-site" pyrometallurgical hot-cell for reprocessing. The molten salt bath (CdCl₂, LiCl, KCl..) is not subject to alpha radiation damage and much shorter cooling times can be envisaged. However this type of reprocessing technology is still in its infancy and will require extensive R&D before reaching industrial maturity in the civil nuclear sector.

The process involves electrorefining of spent metal fuel with a Cadmium anode, solid and liquid cathodes, and a molten salt electrolyte (LiCl+KCl) at 500°C. The heart of the process is a pyrochemical dissolution process of mechanically degraded spent fuel.

Pure U (free of Pu) is electro-transported to the solid Cd cathode. The mixture of Pu+MA with some U is electro-transported to a second but liquid Cd cathode and separated from the bulk of fission products. The Pu+MA fraction is recycled as metal and casted into the new fuel pins.

Similar pyrometallurgical processes are being developed in Japan by the CRIEPI institute and may in a distant future become an alternative to aqueous reprocessing with the PUREX process for highly irradiated FR-Metal/Nitride spent fuel.

The fundamental advantage of the pyrometallurgical reprocessing processes is their insensitivity to burnup of the spent fuel and the possibility to reduce the overall AFC fuel cycle to 6 or 7 years. By doing so, a given depletion yield could be reached in half the time period scheduled for an AFC with aqueous reprocessing. The material problems due to corrosion, the degree of separation of the TRUs and the waste handling are the most important issues which await fully convincing answers before this type of pyrometallurgical process will become a serious contender for the reprocessing of FR-Metal fuel.

The P&T working group should assess the merits and drawbacks of each of the approaches taking into account that improvements are possible in the PUREX process and that the pyrometallurgical processes are still in their laboratory phase.

2.3.11 Transmutation issues of longlived fission products

Transmutation of longlived FP is a very difficult task, because the capture cross sections to transmute the radioactive nuclide into a short lived or stable isotopes requires dedicated and large reactor capacities. Thermal reactors and dedicated Accelerator driven Transmutation (ADT) facilities are the only possible choices for carrying out this very expensive endeavour.

From reactor- technological point of view **the transmutation of Tc99** is the easiest approach since the metallic Tc target is transmuted into metallic Ru100, a stable isotope. However the thermal n-gamma cross section for thermal neutrons is only 20 barns (b) which is extremely low for practical transmutation purposes. The transmutation "half life", i.e. the time necessary to deplete the target with 50 percent varies from one thermal reactor to another but amounts to about 30 years in the best conditions. The reactor must be overenriched to compensate the reactivity loss due to the antireactivity of the Tc targets in the fuel assemblies. The simultaneous production of Tc99 by fission in the LWR-UO2 driver fuel decreases the net transmutation rate and necessitates large Tc loadings. The reactor ought to be dedicated for that purpose.

Transmutation of I129 is from pure neutronic point of view very similar to that of Tc99, but is from chemical-metallurgical standpoint a much more difficult process since the target product is chemically unstable and the neutron capture reaction product is a noble gas : Xe 130 which has to be vented from the irradiation capsule during its stay in the reactor. Any temperature excursion would result in a release of I129 into the reactor off gases.

ADT technologies with extreme high thermal neutron fluxes ($10E16$ n/cm²/s) should be capable of reducing the transmutation half life. This technology is presently in the conceptual phase. However any type of thermal neutron transmutation will be energetically very expensive.

The radionuclides **Zr93 and Cs135 cannot be considered for reactor transmutation** since they have to be isotopically separated from the other Zr and Cs isotopes before being submitted to irradiation.

The transmutation of **C14** has up to now not yet been considered in the P&T context. Theoretically the C14 released from the spent fuel could be removed from the reprocessing off-gases and transformed into a solid target e.g. BaCO₃, but the cross section of C14 for thermal neutrons is nearly zero.

Transmutation by charged particles in high energy accelerators is a theoretical alternative in some cases, but the practical feasibility and the economy of such approaches is very questionable.

The working group should evaluate whether the transmutation of long-lived fission products is a realistic way to pursue and assess the dose benefit/ cost increment ratio for a significant (e.g. 90%), source term reduction of Tc99 and I129. Since these radionuclides have very long half lives, the effect on the dose to man is stretched over geologic periods and surpasses by far the reference lifetime of a repository and even the confinement times of natural barriers.

2.3.11 Conclusions on the role of P&T in the AFC option.

The P&T option within the AFC scenario, as described above, is the most comprehensive approach which can be reasonably proposed and constitutes a very important extension of the fuel cycle activities in comparison with the CFC and a priori with OTC scenario.

The Working Group on P&T should identify the most attractive of the subscenarios to be pursued in the future within a comprehensive and often wishful type of AFC scenario.