CHAPTER IV

GENERAL CONCLUSIONS AND OPEN ISSUES

CHAPTER I : THE FUEL CYCLE OPTION AND THE RADIOLOGICAL SOURCE TERM

Since the world is subdivided into two more or less equal blocks with an opposed fuel cycle option (once through cycle versus conventional reprocessing), the potential influence of a P&T option will have only limited radiological benefits which are yet significant on national or regional scale.

In order to have its full impact the P&T option has to be adhered worldwide and lead to a radiological impact reduction with a factor of 10 to 100.

If the NPP capacity associated with the once-through cycle has to engage into reprocessing in order to follow the P&T route, an additional reprocessing capacity of 3500 - 4500 THM/y has to be built. Moreover, the backlog of spent fuel expected at the end of the century (200.000 THM) corresponds to about 40 years of operation of the presently existing or planned reprocessing capacity (4800 THM/year).

The introduction of the P&T option on a worldwide scale is a long term issue with economical, technical and political aspects, which have to be weighed against the long term radiological benefit.

On a 1000 year horizon there is little difference between storage of spent fuel and storage of HLW since Am 241 determines the radiological hazard. Beyond 1000 and up to 100.000 years the Pu content of spent fuel is the most important radiological burden. In the case of vitrified HLW resulting from conventional reprocessing, Am 241 and Am 243 are the most important radionuclides.

On the very long term (10^5 to 10^6 years) Np 237, I 129 and Tc 99 constitute the most critical radionuclides whose confinement cannot be assured by any geological repository.

CHAPTER II : RADIOCHEMICAL AND NUCLEAR ENGINEERING DATA

The analysis of the 4n+1 series starting with Cm 245 and leading to Pu 241 - Am 241 - Np 237 is the radioactive family with the greatest radiological impact. The Cm 245 content could in a first approach be neglected as it represents only 0.47% of the total actinide inventory.
The radiological impact of Pu 241 - Am 241 - Np 237 is a very complex issue which depends on many factors as e.g. the time of reprocessing, the Pu decontamination factor, the Np distribution in the reprocessing streams... In order to reduce the Am 241 impact with a factor of 1000 it is necessary to reduce the Pu 241 content in HLLW and to adapt the Np flowsheet.

In the 4n+3 series Am 243 appears as the most important radionuclide not only because of its half life but also because it's the parent nuclide of Pu 239. Any action on Am 243 has to be planned in the perspective of a Pu 239 decrease in HLW.

The 4n+2 series is less important from radiological point of view.
The decay of Am 242 into Cm 242 - Pu 238 needs further investigation in order to examine whether Pu 238 with a half life of 87 y has still to be considered as a "long term" alpha emitter. this issue is of great importance for the selection of the Np 237 transmutation reaction.

In order to reduce MA concentration in HLLW to levels compatible with surface storage of HLW, decontamination factors of up to $8.7\ 10^4$ are to be aimed at. Such high values are unrealistic.
One option is to consider the target DF's without or with partial Cm removal. Under such hypotheses the overall DF's are reduced to the 200 - 900 range.
However, establishing technically and radiologically acceptable goals remains a difficult task which requires interdisciplinary consultation among nuclear chemists, reprocessing plant engineers and radiological experts.

The Actinide residues found in cladding hulls and clarification residues may be neglected as a significant MA stream when dealing with LWR - UO_2 fuel. However in the case of LWR-MOX fuel recycle, the insoluble residues may contain up to 0.4% of the total Pu content.

The real composition of HLLW which is the source term for any P&T option, is not yet very well documented in literature and the available data do not permit to draw general conclusions on the prevailing acidity and concentration levels of Actinides and Long lived fission products and on the occurrence of soluble and insoluble compounds in the stored liquids. An international sampling and analysis campaign would very much contribute to the definition of the HLLW source term which is very important for the P&T option but also very useful for vitrification purposes.

Important progress has been achieved in the separation of MA from HLLW solutions and in the adaptation of the PUREX flowsheet for improving the Np recovery.
The TRUEX process is the most advanced MA separation flowsheet which is compatible with a conventional TBP process. The bottleneck in the separation flowsheet is the great difficulty to separate MA and in particular Am-Cm from the Rare Earths. Major R&D efforts will be required to develop an integrated flowsheet, for MA separation from HLLW and Np isolation from the Pu product stream, compatible with the MA product specifications imposed by subsequent transmutation steps.

The acceptance or the rejection of geological disposal as a means to protect the biosphere from long term Actinide contamination is of crucial importance for the significance of the MA separation option in the overall waste management. If deep disposal is accepted for a period of up to 10.000 years, only partitioning of long lived fission products and Np 237 becomes an important task.

The separation of iodine (I 129) in the reprocessing process is standard industrial procedure. However, the separated I 129 is according to present practice diluted into the sea and dispersed in the biosphere where it slowly but steadily accumulates. If it would be transferred to a geological repository it would cause the highest impact among the fission products on the overall radiation dose.

Technetium 99 is the second most important long lived fission product which cannot be confined in a geologic repository. Isolation from HLLW and/or preseparation in the PUREX process are two alternatives for partition. The

presence of highly active nuclides (e.g. Ru 106) with similar properties excludes the use of a simple separation procedure. A delayed treatment would have advantages since noble metals are a strategic resource for the future. Transmutation of Tc 99 requires very intense thermal neutron sources.

The merit of partitioning other long lived radionuclides e.g. Cs 135, Zr 93, Se 79 and C 14 is a matter of radiological assessment in a specific geohydrologic configuration. Most of these radionuclides would require isotopic separation in order to convert them into a useful irradiation target.

CHAPTER III : MINOR ACTINIDES AND LONG LIVED FISSION PRODUCTS CONDITIONING AND TRANSMUTATION

The magnitude of the MA conditioning and transmutation is directly linked to the production capacity of existing and planned reprocessing plants in the OECD countries.

A maximum of about 1700 kg Np 237 per year is to be expected and this quantity requires a dedicated MOX production capacity of 85 THM LWR-MOX or 68 THM FBR-MOX.

The potential maximum production of Am-Cm amounts to about 1500 kg per year, but such an inventory of highly gamma irradiating nuclides cannot be manipulated in standard MOX production facilities. According to the present state of the art the separation of Am-Cm from bulk Rare Earths has not yet been realized at the industrial level. A 75 THM MOX fuel fabrication plant capacity based on sol gel technology with full-remote-handling and maintenance capability has to be built for these purposes around the existing reprocessing plant sites.

Heterogeneous recycle of MA is still in its infancy but encouraging results have been obtained in experimental fabrication campaigns of fuel pins with up to 45% Np 0_2 or Am_2O_3 contents in depleted $U0_2$.

An alternative fuel fabrication technology based on pyre-metallurgical processing of metallic fuel received considerable attention. The proposed

flowsheets have not yet been demonstrated at hot pilot scale and the handling of large quantities metallic MA-Pu-Zr type fuels require very elaborate pyre-metallurgical processing facilities which are sensitive to air and moisture ingress. The presently proposed flowsheets do not provide an adequate MA/Rare Earth separation step. The MAB reactors require MA-Rare Earth ratio's of 100 to 10. The small throughput of these processes (10-50 kg/day) will increase the cost but improve their flexibility.

A comparative conceptual design study ought to be made for the two approaches :

construction and operation of a MA-MOX-FBR fuel fabrication plant on the basis of a single capacity of 90 THM/year or 3 units of 30 THM/year;

construction and operation of several modular units of MA-metal fuel fabrication as conceived in the IFR concept with a total capacity of 90 THM/year.

Based on such a technical study the costs for reprocessing the fuel (7-14 times per initial charge depending on the transmutation system) could be evaluated on a more factual basis.

Minor Actinide Transmutation can be carried out in LWR, FBR, ABR and Accelerator driven reactor systems.

LWR reactors with slightly higher enrichments can transmute substantial quantities of MA but the high Pu 238 concentration in spent fuel will complicate the recycle fuel fabrication steps.
The degraded Pu isotopic composition will require subsequently higher Pu enrichments.

A 1 GWe-year FBR-MOX reactor can transmute the Np-Am output resulting from 6 GWe-year LWR's. However the mass balance of Cm is positive i.e. 22% more Cm is generated per year; other transmutation systems have to be found for this nuclide.
In order to transmute the total Np-Am output from the existing or planned

reprocessing units in the OECD countries, a total FBR-MOX capacity of 25 GWe-y would be required. In case all nuclear involved countries choose the P&T option, this capacity ought to be doubled.

- The ABR is the most attractive transmutation-incineration facility since it uses Pu-MA fuel mixtures without actinide breeding effect. The main transmutation process is fission due to the high mean neutron energy (= 700 keV).
  The MA inventory of a notional 400 MWe-ABR is 2,670 kg Np-Am-Cm which corresponds to the output of 100 - 120 units of 1 GWe-y LWR. The MA incineration throughput per year of a 400 MWe ABR amounts to about 195 kg Np Am Cm or one ABR 400 MWe-y for 8.7 GWe-y LWR units.
  In order to destroy the entire Np-Am-Cm production resulting from the annual LWR output in the OECD countries about 6.4 GWe ABR capacity should be constructed; this is a reduction with a factor of 4 of the requirements in comparison with FBR-MOX reactors.
  The main drawback of the ABR option if its fuel cycle which relies either on a metal type fuel and pyrometallurgical processing or on coated particle fuel with a very exotic type of reprocessing.

- The proton Accelerator driven reactor systems are based on direct heavy nuclide (Pu, U, MA, W, Pb) spallation sources and the use of induced high neutron fluxes which are capable of transmuting the actinides and the fission products. Depending on the beam current the reactor systems can produce neutron fluxes from 10^{15} to 10^{16} n/cm².sec in a narrow zone around the beam trajectory.

  .The JAERI project is a 1.5 GeV - 39 mA proton accelerator beam impinging on a solid tungsten target surrounded by a subcritical metallic Pu-MA reactor core. The design proposed is very similar to the IFR-ABR fast reactor design.

  .The PHOENIX concept is a 1.5 GeV - 104 mA proton accelerator beam impinging directly on a modular FBR-MOX subcritical reactor core. Due to the lower mean energy, neutron capture is the prevailing transmutation event. Advanced aqueous reprocessing based on PUREX and TRUEX can be used to recycle the spent fuel pins. Large quantities of Pu 238 are formed.

- The 'LOS ALAMOS high thermal neutron incinerator project is a 1.6 GeV - 250 mA proton beam impinging on a molten Pb-Bi target surrounded by a D_2O moderator. The extremely high" thermal flux (10^{16}) can induce double neutron capture and fission of fertile nuclides.

- All these accelerator projects are long term ventures which might become operational within one or two decades. The investment costs will be very high as a linear accelerator and a reactor core have- to be--jointly operated. The advantages of the Accelerator driven spallation sources lie in the operation flexibility and in the occurrence of very high neutron fluxes which can be used not only for Actinides but also and perhaps mainly for transmutation of long lived fission products (I 129, Tc 99,...) with very low thermal cross section.

A systems analysis study should be undertaken to investigate the overall transmutation costs, the time span necessary to carry out a MA incineration campaign, the residual MA quantities resulting from a multiple recycle programme.

These costs have to be weighed against the cost for constructing and operating an underground repository for the entire MA and long lived fission product inventory. The effect of inventory reduction on the residual cost of geological disposal is the final aim of this analysis.

The long term radiological impact of the MA and long lived fission products inventory should be examined and compared with the short time expenses in order to assess the net cost of the long term dose to man reduction.