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Partitioning and Transmutation:  
Radioactive Waste Management Option

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# **Partitioning and Transmutation: Radioactive Waste Management Option**

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**ICTP, Trieste, Italy, 17 - 28 October 2005**

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# Background

- **Growing world population with increasing energy needs, especially in the developing countries**
- **Threat of global warming due to CO<sub>2</sub> emissions demands non-fossil electricity production**
- **Nuclear will have to be part of a sustainable mix of energy production options**



# Nuclear Energy: Significant Figures

- **350 GWe worldwide capacity; ~ 100-120 GWe in each region USA/Europe/rest of the world**
- **10'500 t HM annual spent fuel discharge worldwide**
- **3'900 t HM industrial reprocessing capacity worldwide**
- **130,000 t HM cumulative stored spent fuel inventory**
- **70,000 t HM reprocessed and transformed into HLW and spent LWR-MOX**



# Repository Needs, Characteristics

- Present worldwide **spent fuel** (containing high Pu inventory) and **HLW** arising would need **2** and one **1** Yucca Mountain size repositories, respectively
- **Spent fuel** repository: high Pu content  $\Rightarrow$  non proliferation and criticality concerns
- Medium-lived fission products ( $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ) determine heat load  $\Rightarrow$  same in **spent fuel** and **HLW** repository
- Minor actinides (Np, Am, Cm) determine radiotoxicity potential  $\Rightarrow$  same in **spent fuel** and **HLW** repository



# P&T: Basics, Objectives

- **Renewed interest in P&T  $\Rightarrow$  numerous efforts worldwide to assess its potential as a radioactive waste management option**
- **P&T is a complex technology  $\Rightarrow$  advanced reprocessing and transuranics fuel fabrication plants, innovative and/or dedicated transmutation reactors**
- **In addition to U, Pu and  $^{129}\text{I}$ , “Partitioning” extracts from the liquid high level waste the minor actinides (MA) and the long-lived fission products (LLFP)  $^{99}\text{Tc}$ ,  $^{93}\text{Zr}$ ,  $^{135}\text{Cs}$ ,  $^{107}\text{Pd}$ , and  $^{79}\text{Se}$ )**



## P&T: Basics, Objectives (cont'd)

- **“Transmutation” requires fully new fuel fabrication plants and reactor technologies to be developed and implemented on industrial scale**
- **Present LWRs are not suited for MA and LLFP transmutation (safety consideration, plant operation, poor incineration capability)**
- **Only specially licensed LWRs can cope with MOX-fuel; for increased Pu loadings (up to 100%), special reactor designs (e.g., ABB80+) are required; a combination of these reactor types could allow Pu inventory stabilization, albeit at the price of increased MA production**



## **P&T: Basics, Objectives (cont'd)**

- **Long-term waste radio-toxicity can be effectively reduced only if transuranics are “incinerated” through fission  $\Rightarrow$  very hard neutron spectra needed**
- **New reactor concepts (dedicated fast reactors, Accelerator Driven Systems (ADS), fusion/fission hybrid reactors) have been proposed as transmuters/incinerators**



## **P&T: Basics, Objectives (cont'd)**

- **Significant Pu+MAs incineration rates can be achieved in symbiotic scenarios: LWR-MOX & dedicated fast reactors; fast neutron spectrum ADS could be reserved for MA incineration; very high thermal flux ADS concepts could also provide a significant transuranics destruction**
- **LLFP transmutation problematic:**
  - **occur in elemental mixtures (different isotopes of the same element) ⇒ isotopic separation required**
  - **transmutation yields small because of very low capture cross sections in thermal neutron fields ⇒ dedicated reactors required with very high loadings and/or high thermal flux levels**



## P&T: Basics, Objectives (cont'd)

- Reduction of **long-term hazard** of spent fuel or HLW by transforming long-lived radionuclides into short-lived or inactive elements is one of the main P&T objectives
- Reduction of **long-term radiological risk** (combination of potential hazard and confining properties) is “conventional” waste management objective



## P&T: Basics, Objectives (cont'd)

- Hazard reduction (P&T objective) requires very different and much more fundamental measures as compared to risk reduction:
  - long-term hazard of spent fuel and HLW is associated with the radioactive source, i.e., the transuranics
  - short and long-term risks are due to the mobility of fission products in the geosphere and the possibility to enter the biosphere



# Nuclear Fuel Cycle Options

- **Conventional**
  - **Once through fuel cycle with direct disposal of spent fuel (OTC)**
  - **Aqueous reprocessing fuel cycle with vitrification of high-level liquid waste (RFC)**
- **Advanced fuel cycle with partitioning of actinides (AFC)**



# Once-through Fuel Cycle

- Option considered in Canada, Spain, Sweden, USA, and some other countries
- Cheapest option at present low uranium prices
- Residual fissile material (1% Pu and 0.8%  $^{235}\text{U}$ ) and remaining fertile material ( $^{238}\text{U}$ ) in spent fuel is not recovered and considered waste material
- Spent fuel long-term radiotoxicity associated with transuranics (Pu, Np, Am, Cm, ...)  $\Rightarrow$  significant radiological source term in repository over hundred thousand years
- Effective isolation of transuranics in repository due to intrinsic insolubility of actinides in deep geological formations



## Once-through Fuel Cycle (cont'd)

- FP  $\gamma$ -radiation and decay heat are most limiting short-term repository design factors
- Most FPs have decayed after 300 - 500 years, except for a few long-lived nuclides ( $^{135}\text{Cs}$ ,  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{93}\text{Zr}$ , ...)
- Some long-lived FPs are relatively mobile in the geosphere



## Once-through Fuel Cycle (cont'd)

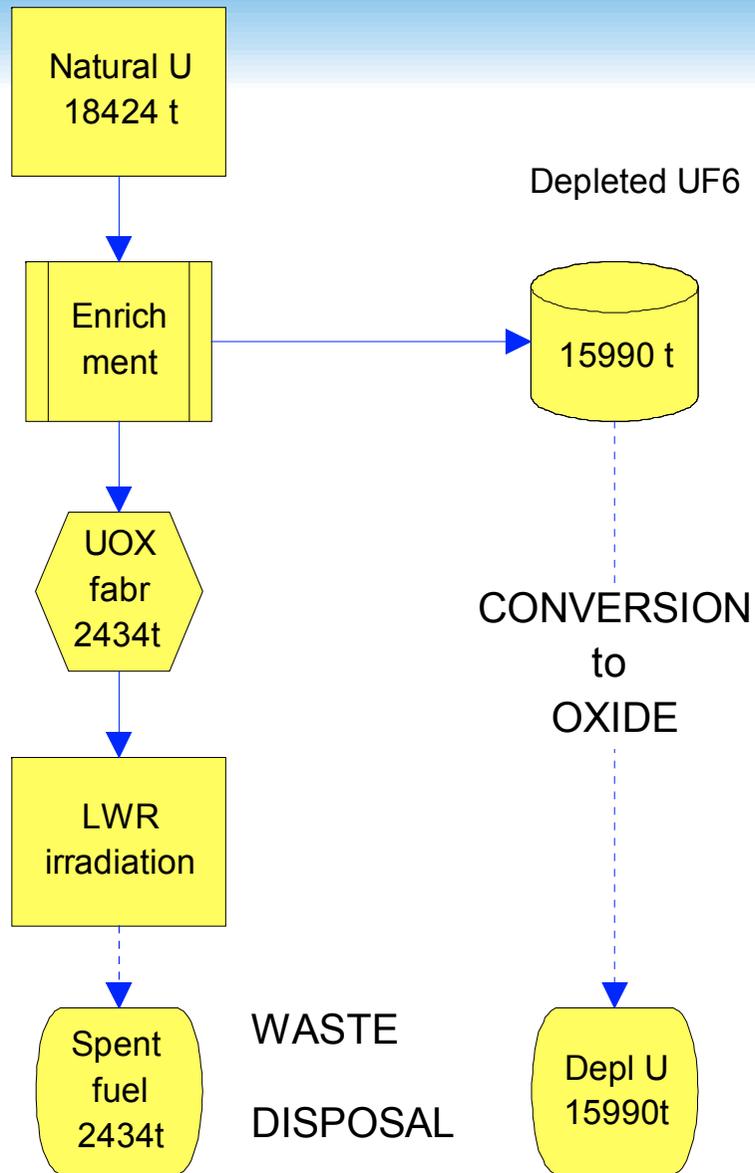
- Long-term OTC radiological impact can be controlled by engineering design and natural barriers which should provide protection for the time period defined by the life-time of the confined source term  $\Rightarrow$  long time periods involved require a careful analysis of the confinement technology and of the long-term consequences of accident scenarios
- There is no worldwide agreement on the time periods for confinement of HLW in a geologic repository; periods of  $10^3 - 10^5$  years, or even longer, have in been put forward, but no internationally accepted confinement period has been established



# Once-through Fuel Cycle (cont'd)

- **Safety and regulatory aspects of OTC**
  - Substantial quantities of fissile material are deposited, and also the decay of  $^{239}\text{Pu}$  to  $^{235}\text{U}$  has to be considered  $\Rightarrow$  potential for criticality has to be addressed in the repository licensing process
  - $^{129}\text{I}$  is the limiting nuclide in risk repository analyses: if no specific actions are taken to limit  $^{129}\text{I}$  solubility in the deep aquifers, the entire  $^{129}\text{I}$  inventory will be fully dispersed in the geosphere after a few  $10^4$  yrs
  - Beyond 500 years, spent fuel heat emission due to total actinide content is significantly higher than that of the FPs; absolute value of that long-term heat output is, however, significantly lower than that of the initially loaded FPs
  - 70'000 t HM per repository is limit in the USA  $\Rightarrow$  new repository every ~30 years





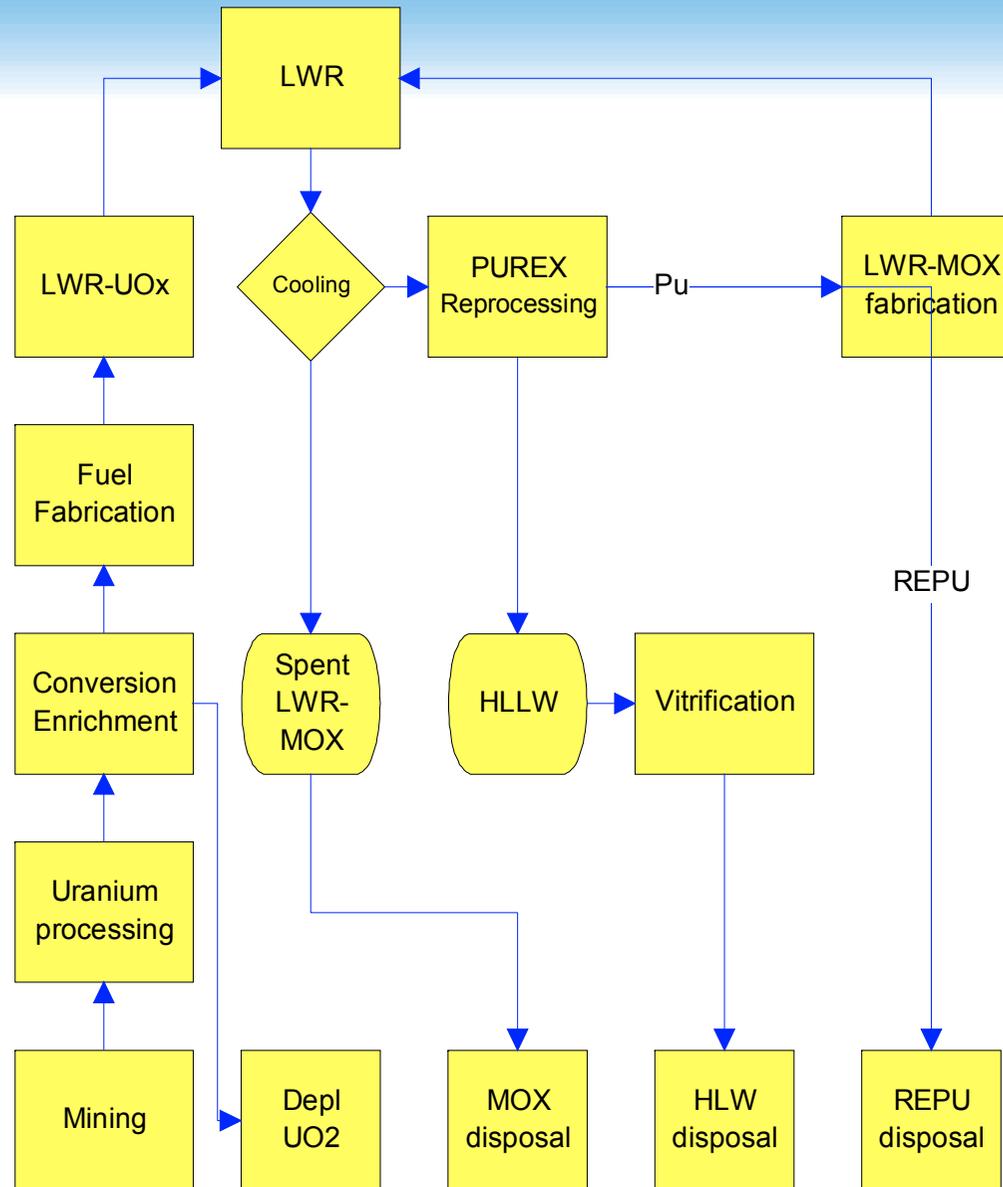
**Fig 1 ONCE-THROUGH FUEL CYCLE**



# Aqueous Reprocessing Fuel Cycle with Vitrification of HLLW

- Natural uranium contains only 0.72% of fissile  $^{235}\text{U}$   $\Rightarrow$  recycling of the “major actinides”, i.e., U and Pu, from spent fuel in the aqueous reprocessing fuel cycle (RFC) with vitrification of HLLW is standard scenario of nuclear energy production
- Reduced support for this option in many countries due to costs and proliferation concerns





**Fig 2 LWR REPROCESSING FUEL CYCLE**



# Aqueous Reprocessing Fuel Cycle with Vitrification of HLLW (cont'd)

- ~99.9% of the U and Pu streams are extracted, thus only a very minor fraction of the “major actinides” goes to HLLW, and thus to HLW, and to geologic repository
- Removal of MAs (that become more important with increasing burnup) from HLLW would reduce the residual HLW radiotoxicity
- Adding a MA partitioning module to the standard reprocessing plant would be the most obvious change to the current RFC ⇒ countries with reprocessing infrastructure (France, UK, Japan, India, Russia, and China) could implement, in a medium-term, a partial partitioning scenario by which the HLW would be practically free from long-lived transuranics



# Aqueous Reprocessing Fuel Cycle with Vitrification of HLLW (cont'd)

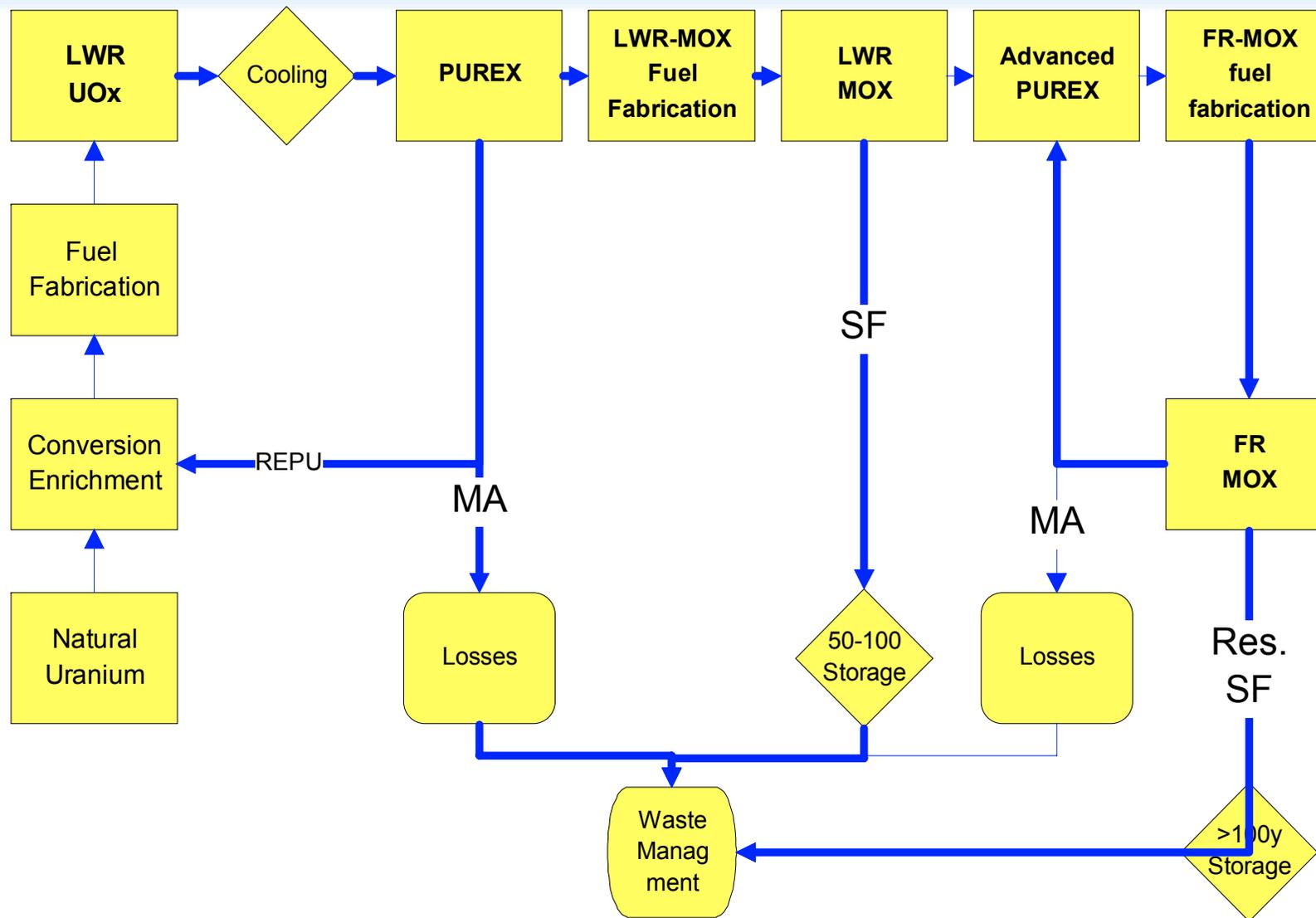
- Use of LWR-MOX in the RFC option is industrially significant in Western-Europe (specially licensed reactors in France, Germany, Switzerland and Belgium are fuelled with MOX)
- Within the broader context of resources utilization and waste management, the reuse of Pu is the first step in a global P&T scenario
- Recycling spent LWR fuel as MOX provides an overall mass reduction of a factor ~5, but the radiotoxicity is not significantly reduced, since only ~25% of the recycled Pu is consumed and about 10% is transformed into a long-term radiotoxic MA source term



# Aqueous Reprocessing Fuel Cycle with Vitrification of HLLW (cont'd)

- Further radiotoxicity reduction requires loading of the spent LWR-MOX transuranics into a fast reactor ⇒ part of the advanced fuel cycle (AFC)
- In theory, reprocessing the spent LWR-MOX fuel in view of recycling the transuranics in FRs is possible in present reprocessing plants
- Additional transuranics separation module is required to reduce significantly the radiotoxicity of the HLLW produced during reprocessing of LWR-MOX (up to levels comparable to those of non-reprocessed spent LWR-MOX), and this could be done with the same technology for HLLW from LWR-UO<sub>2</sub>





**Fig 3 CONVENTIONAL LWR-FR -MOX FUEL CYCLE**



# Aqueous Reprocessing Fuel Cycle with Vitrification of HLLW (cont'd)

- Recovery of U from spent fuel by TBP extraction in the PUREX process
- Very little of recovered U stockpile has been reused in subsequent reactor loadings
- Reprocessed U contains some troublesome radioisotopes ( $^{232}\text{U}$ , a parent of natural decay chains with radiotoxic daughter nuclides, and  $^{236}\text{U}$  forming  $^{237}\text{Np}$  in a neutron field [via  $(n,\gamma)$  reaction to  $^{237}\text{U}$  and its decay ( $\beta^-$ ,  $T_{1/2}=6.8$  d)])
- Recycling of reprocessed U in LWR fuel is done industrially  $\Rightarrow$  fresh fuel fissile enrichment increase



## **Aqueous Reprocessing Fuel Cycle with Vitrification of HLLW (cont'd)**

- The radiotoxic impact from stockpiling depleted and reprocessed U is greater than, e.g., Np  $\Rightarrow$  large amounts of depleted U (8 to 9 tons per ton LWR fuel produced), stored as  $UF_6$ , constitute an important long term radiotoxic and chemical hazard to be addressed**



# Advanced Fuel Cycle (AFC)

- **AFC scenario with actinides P&T of comprises the following steps**
  - **Improved reprocessing of LWR UO<sub>2</sub> fuel with additional Np removal**
  - **Separation of MAs from HLLW resulting from LWR UO<sub>2</sub> reprocessing**
  - **Fabrication of MA targets for heterogeneous irradiation in LWRs**
  - **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)**



# Advanced Fuel Cycle (AFC) (cont'd)

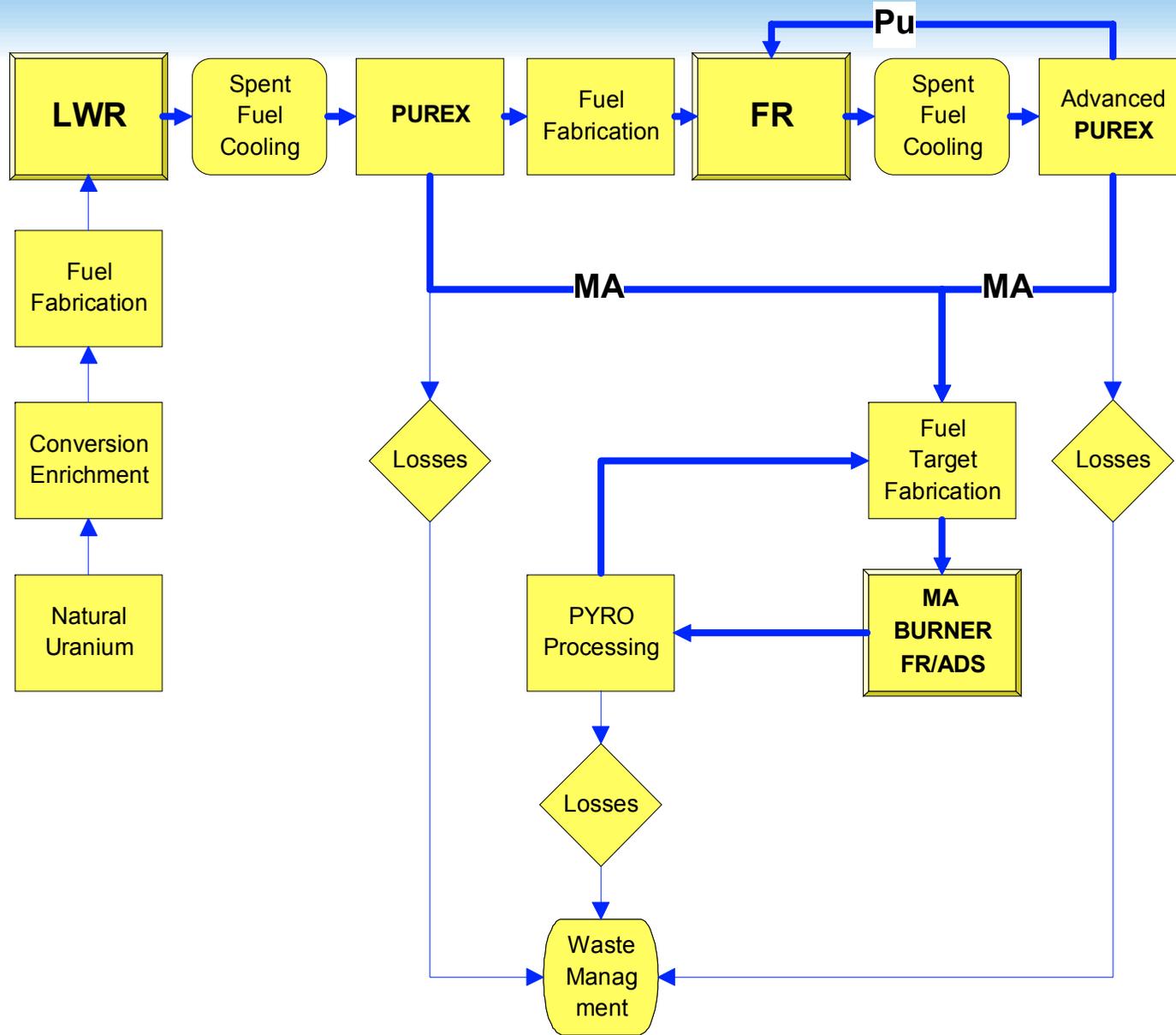
- Separation of MAs from HLLW and conditioning of individual elements (Np, Am, Cm)
- Long-term storage and eventual disposal of specially conditioned MA
- Fabrication of FR (MOX, metal, or nitride) fuel with a limited MA content
- Irradiation of FR-fuel in Fast Burner Reactors or dedicated hybrid facilities (very high burnup)
- Reprocessing of spent FR fuel in specially designed (aqueous and/or pyrochemical) and licensed facilities
- Separation of all transuranics from the spent FR fuel processing during multiple recycling



## Advanced Fuel Cycle (AFC) (cont'd)

- Multiple recycling of FR MOX fuel with major transuranics content until significant depletion
- Separation of certain long-lived fission products (if required for the disposal step)
- Revision of the fission product management  $\Rightarrow$   $^{99}\text{Tc}$  separation (head-end, HLLW)
- If wanted, platinum metals separation and recovery (economics)





**FIG 4 DOUBLE STRATA FUEL CYCLE**



# Advanced Reprocessing With Np Removal

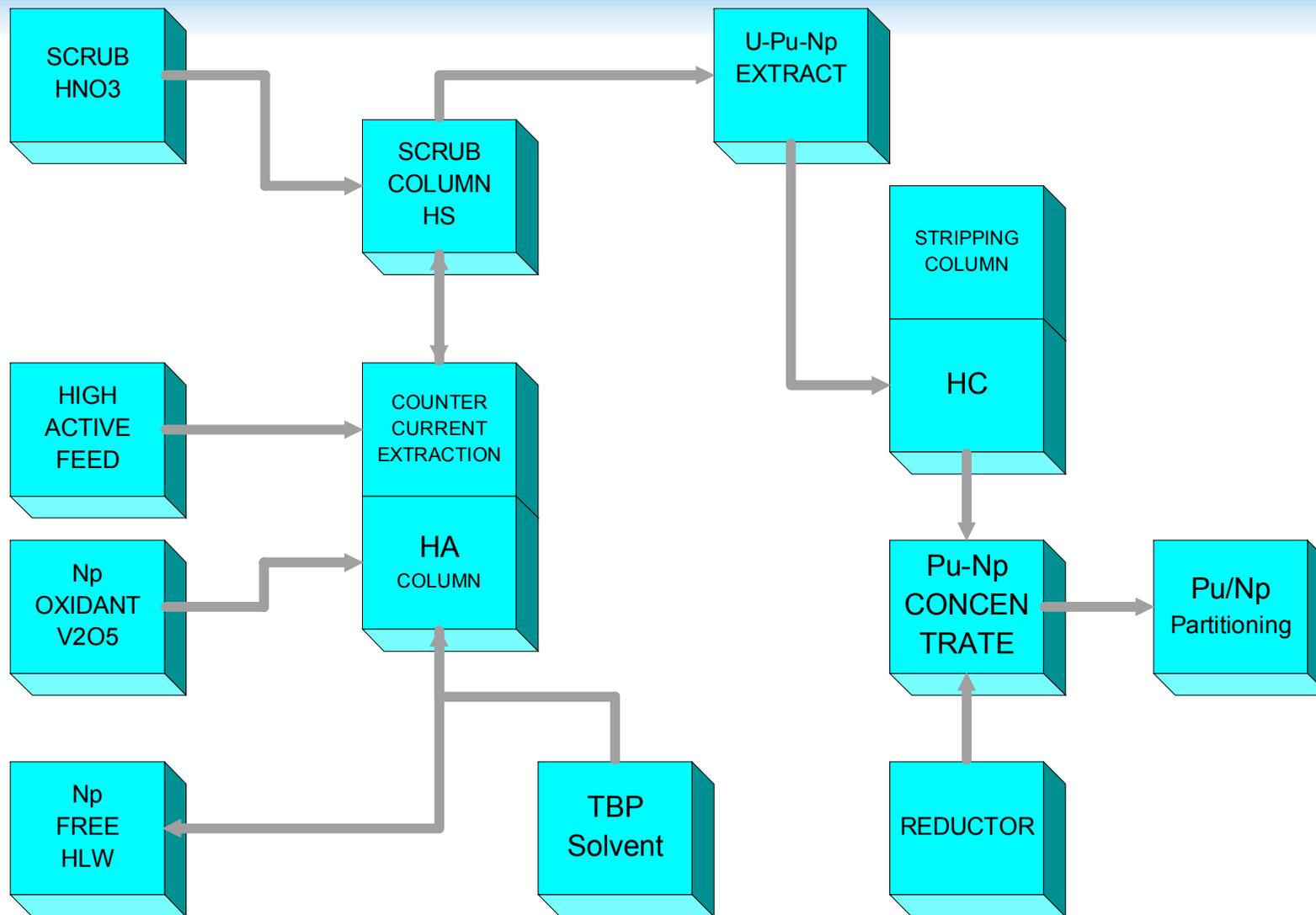
- $^{237}\text{Np}$  recovery from U-Pu technically possible in the PUREX (during the current reprocessing operations, Np is partly discharged with the fission products into the HLLW and partly associated with the U, Pu, Np stream in TBP; the purification of Pu and its quantitative separation from Np is achieved in the second PUREX extraction cycle)
- Presently, LWR-MOX reprocessing mainly done by diluting LWR-MOX with LWR-UO<sub>2</sub> fuel according to the ratio in which it occurs in the reactor-core (UO<sub>2</sub>/MOX=2)



# Advanced Reprocessing With Np Removal (cont'd)

- **Reprocessing of spent LWR MOX fuel without dilution in UO<sub>2</sub> fuel has been demonstrated at COGEMA La Hague UP2 in 1992 (~5 t); it is industrially feasible, provided the reprocessing plant has been designed for the treatment of increased Pu concentrations and licensed for a much higher total Pu inventory**
- **Conclusions**
  - **Np recovery from dissolved spent fuel streams can be realized in adequately designed and/or refurbished reprocessing plants**
  - **Separated Np has to be stored under strict safeguarded conditions since its critical mass amounts to 55 kg**





**FIG 5 MODIFIED PUREX EXTRACTION**



# Separation of Am-Cm From HLLW Resulting From Spent LWR Fuel

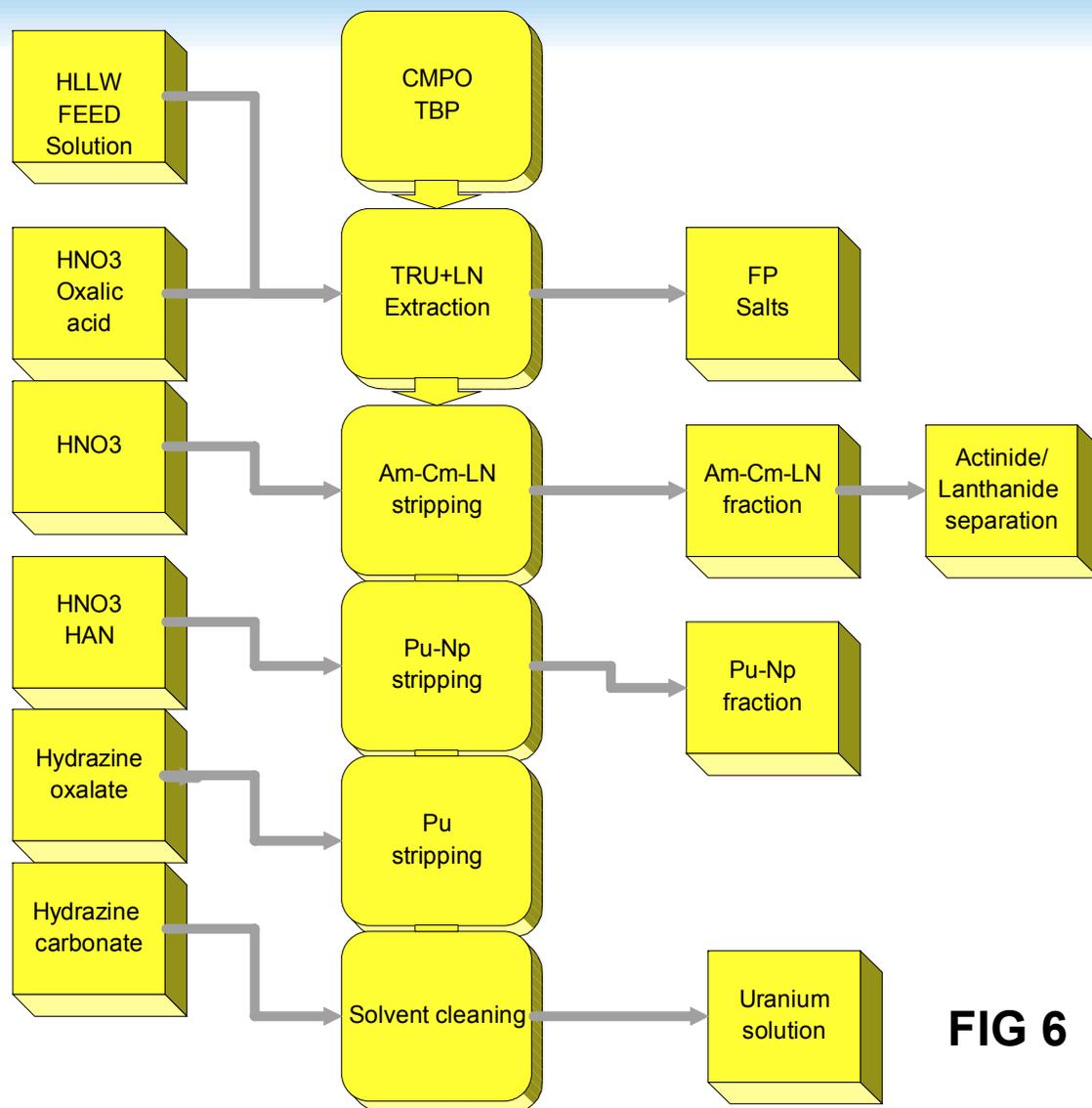
- **Conventional reprocessing: most MAs (Np, Am, Cm) transferred to the HLLW**
- **>99.5% of Am and Cm, and some shorter-lived transuranics (Bk, Cf, ...) transferred to HLLW**
- **Am-Cm separation from HLLW is the first priority from the radiotoxic point of view**
- **Also prerequisite for a significant reduction of the (very) long-term radiotoxicity due to  $^{237}\text{Np}$**
- **Separation of  $^{241}\text{Am}$  implies separation of the long-lived  $^{243}\text{Am}$  (parent of  $^{239}\text{Pu}$ )**



# Separation of Am-Cm From HLLW Resulting From Spent LWR Fuel (cont'd)

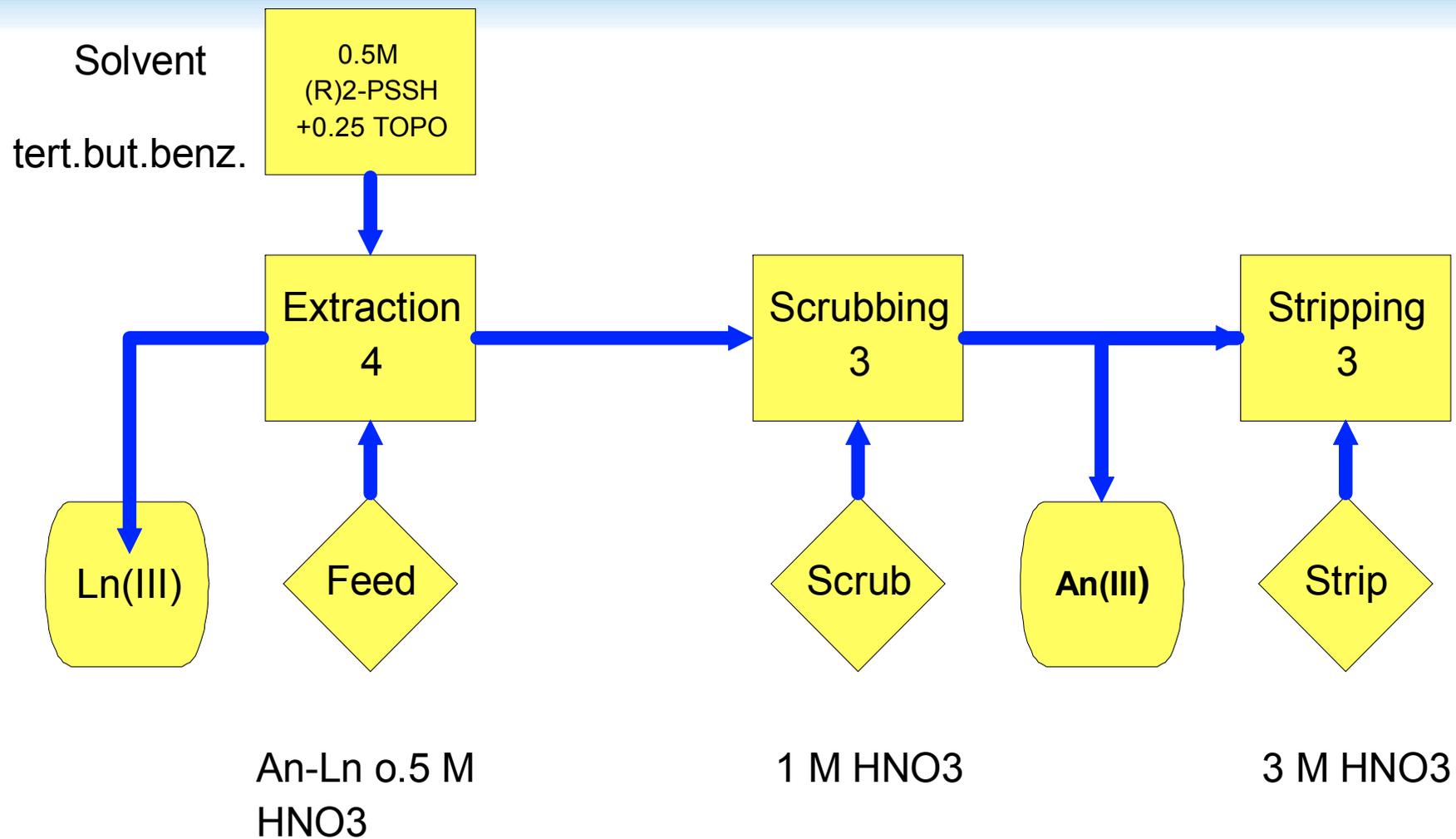
- **The Am-Cm spent fuel fraction contains all the rare earth (RE)**
  - RE constitute a 10 to 20 times higher inventory in the spent fuel than the actinides (depending on the burnup)  
⇒ in 1 t 45 GWd/t HM spent fuel there are 13.9 kg RE vs. 0.870 kg Am-Cm)
- **Several processes have been studied at the conceptual level and tested in hot facilities, e.g., TRUEX (Fig 6), DIDPA, TRPO, DIAMEX for An-Ln group separation, in conjunction with CYANEX 301™ SANEX, ALINA processes allowing the An/Ln separation (Fig 7)**





**FIG 6 TRUEX Process flowsheet**





**FIG 7 ALINA FLOWSHEET - FZJ**



# Separation of Am-Cm From HLLW Resulting From Spent LWR Fuel (cont'd)

- Basic criterion to judge the different methods is the overall decontamination factor (DF) obtained during HLLW extraction as compared to the required DFs in order to reach the 100 nCi level of  $\alpha$  active nuclides in the HLW
- The highest DFs must be reached for  $^{241}\text{Am}$  ( $3.2 \times 10^4$ , if immediate separation is considered)



## Required DF's to reach 100 nCi/g in vitrified HLW

Nuclide	Immediate	500 yrs	10 <sup>3</sup> yrs	10 <sup>4</sup> yrs
<b>238Pu</b>	<b>9.8×10<sup>4</sup></b>	<b>1.88×10<sup>3</sup></b>	<b>36</b>	<b>-</b>
<b>238Pu (repro)</b>	<b>326</b>	<b>6.25</b>	<b>-</b>	<b>-</b>
<b>239Pu</b>	<b>10<sup>4</sup></b>	<b>9.8×10<sup>3</sup></b>	<b>9.7×10<sup>3</sup></b>	<b>7.5×10<sup>3</sup></b>
<b>239Pu (repro)</b>	<b>33</b>	<b>32.6</b>	<b>32.3</b>	<b>25</b>
<b>240Pu</b>	<b>1.63×10<sup>4</sup></b>	<b>1.54×10<sup>4</sup></b>	<b>1.47×10<sup>4</sup></b>	<b>5.66×10<sup>3</sup></b>
<b>240Pu (repro)</b>	<b>54</b>	<b>51</b>	<b>49</b>	<b>18</b>



## Required DF's to reach 100 nCi/g in vitrified HLW (cont'd)

Nuclide	Immediate	500 yrs	10 <sup>3</sup> yrs	10 <sup>4</sup> yrs
<b>241Pu</b>	<b>31.4</b>  <b>(β<sup>-</sup>, 1% α)</b>	<b>(241Am)</b>  <b>1.04×10<sup>5</sup></b>	<b>(241Am- 237Np)</b>  <b>2.25×10<sup>4</sup></b>	<b>237Np</b>  <b>21</b>
<b>241Pu (repro)</b>	<b>92, as α</b>	<b>346</b>	<b>75</b>	<b>21</b>
<b>242Pu</b>	<b>69</b>	<b>69</b>	<b>69</b>	<b>69</b>
<b>242Pu (repro)</b>	<b>-</b>	<b>-</b>	<b>-</b>	<b>-</b>
<b>237Np</b>	<b>11.2</b>	<b>11</b>	<b>11</b>	<b>11</b>



## Required DF's to reach 100 nCi/g in vitrified HLW (cont'd)

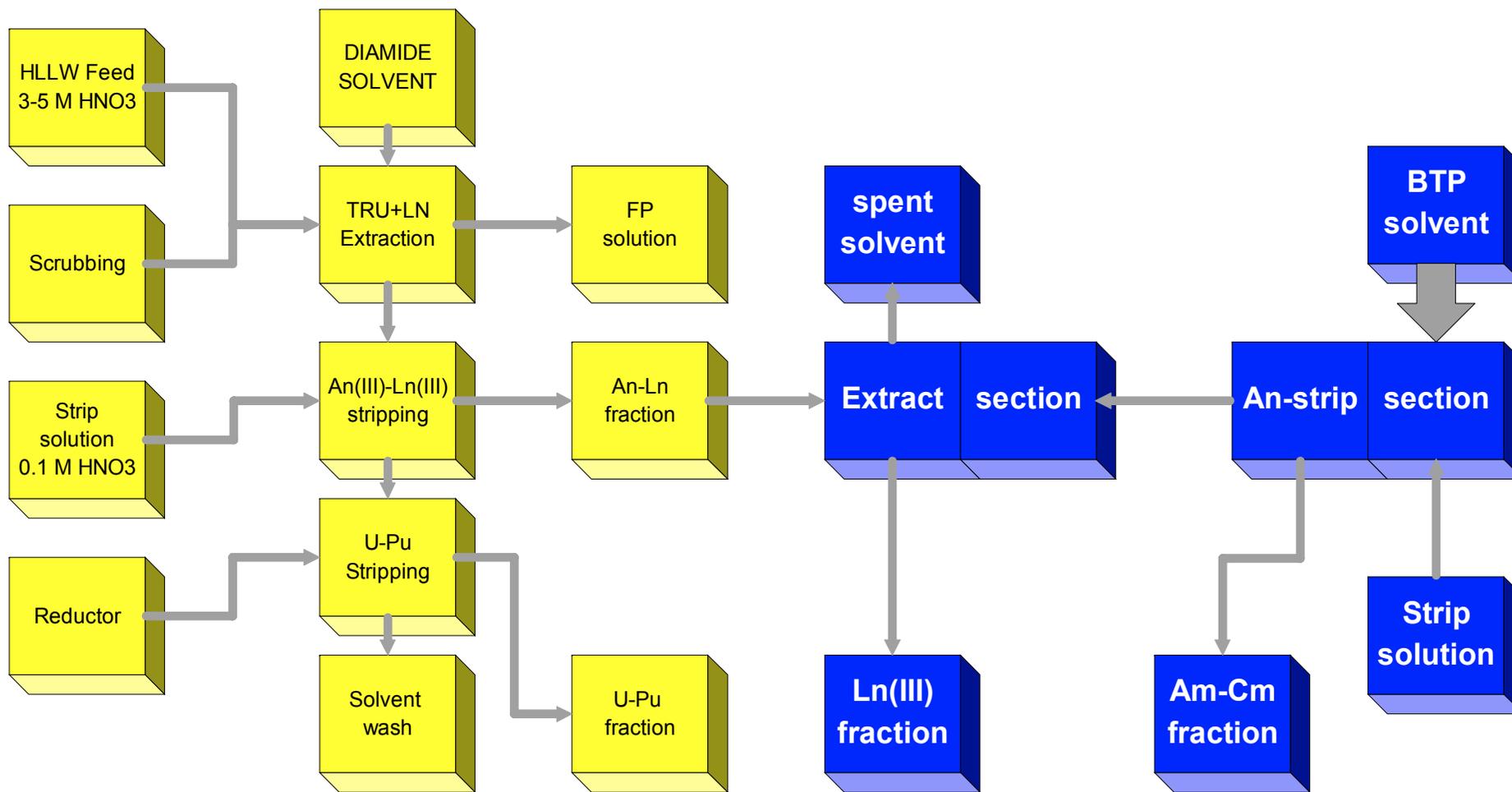
Nuclide	Immediate	500 yrs	10 <sup>3</sup> yrs	10 <sup>4</sup> yrs
<b>241Am</b>	<b>3.2×10<sup>4</sup></b>	<b>1.45×10<sup>4</sup></b>	<b>6.4×10<sup>3</sup></b>	<b>(Np) 17</b>
<b>243Am</b>	<b>710</b>	<b>678</b>	<b>647</b>	<b>276</b>
<b>244Cm</b>	<b>8.27×10<sup>4</sup></b>	<b>(240Pu) 229</b>	<b>(240Pu) 205</b>	<b>(240Pu) 80</b>
<b>245Cm</b>	<b>7.5</b>	<b>7.5</b>	<b>7</b>	<b>3.3</b>



# Separation of Am-Cm From HLLW Resulting From Spent LWR Fuel (cont'd)

- Hot cell tests carried out at the JRC-ITU at Karlsruhe indicate that DIAMEX represents the best compromise  $\Rightarrow$  DFs  $\sim 10^3$  were obtained for MAs in 3.5 M acid HLLW
- An/Ln separation was demonstrated in the laboratory with ALINA (based on the use of a new organo-sulfinic acid extractant)  $\Rightarrow$  DFs  $\sim 30$  were obtained in 0.5 M acid solution





**FIG 8 DIAMEX CONCEPTUAL FLOWSHEET**

**SANEX FLOWSHEET**



# Separation of Am-Cm From HLLW Resulting From Spent LWR Fuel (cont'd)

- **TRUEX is very effective for  $\alpha$  decontamination of medium-level and non-heating high active waste**
  - MA fraction with 90% purity requires An/Ln DF  $> 10^2$
  - MA fraction with 99% purity requires An/Ln DF  $> 10^3$
- **Research labs have strong limitations with regard to the MA quantities which can be handled in shielded facilities (e.g., the new JRC-ITU MA laboratory has an authorization for maximum 150 g  $^{241}\text{Am}$ , and 5 g  $^{244}\text{Cm}$ )**



# Separation of Am-Cm From HLLW Resulting From Spent LWR Fuel (cont'd)

- **100 GWe NPP park produces annually about 1'600 kg of each Np, and Am-Cm ⇒ large effort needed to upscale the laboratory methods to pilot scale (and, later, industrial prototype scale) in order to include MA separation rigs from the design phase on, in the future advanced reprocessing plants**



# Separation of Am-Cm From HLLW Resulting From Spent LWR Fuel (cont'd)

- **Considering Europe, industrial RFC can be taken for granted  $\Rightarrow$  sufficient reprocessing capacity (La Hague, Sellafield) to cover the European and some overseas needs for the next 20-30 years**



# Separation of Am-Cm From HLLW Resulting From Spent LWR Fuel (cont'd)

- **The first steps towards AFC implementation**
  - **Installation of the separation facilities for MA from liquid HLLW**
  - **Conditioning of these nuclides for intermediate storage or as potential target material for transmutation**
  - **"Actinide-free HLW" could be produced by the vitrification plants and stored for cooling in surface facilities followed by geological disposal. There are no objective arguments to oppose geological disposal of such a waste stream which decays by more than four orders of magnitude during 500 years**



# Separation Of Long-lived Fission And Activation Products

- P&T must consider some radiologically important fission and activation products given their important role in the geologic repository assessment
- Fission products:  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{135}\text{Cs}$ ,  $^{79}\text{Se}$ ,  $^{93}\text{Zr}$ , and  $^{126}\text{Sn}$
- Activation products:  $^{14}\text{C}$ , and  $^{36}\text{Cl}$
- Some of these radionuclides must be examined in depth in order to establish their risk and potential radiotoxic role in comparison with the transuranics: their radiotoxicity is between  $10^3$  and  $10^5$  times smaller than transuranics, but their contribution to the very long-term risk is predominant because migration to the biosphere may be much more rapid



# Separation Of Long-lived Fission And Activation Products (cont'd)

- **$^{99}\text{Tc}$** 
  - **$T_{1/2} = 213'000$**
  - **Occurs as Tc metal and  $\text{TcO}_2$  in the insoluble residues, and as soluble pertechnetate ion in the HLLW solution**
  - **Generation rate = 26.6 kg/GWe-yr**
  - **Overall specific concentration = 1.2 kg/tHM**
  - **Effective long-term radiotoxicity reduction by transmutation requires combination of the soluble (80%) and insoluble (20%) fractions**



# Separation Of Long-lived Fission And Activation Products (cont'd)

- The extraction of soluble  $\text{TcO}_4^-$  is relatively easy
- The similarity between Tc and the platinum metals in insoluble waste and the nature of the separation methods makes this partitioning operation very difficult  $\Rightarrow$  pyrometallurgical processes might be more adequate to carry out a group separation together with the platinum metals
- Separation from aqueous effluents is possible in an advanced PUREX scheme
- Recovery from insoluble residues is very difficult (the present recovery yield could approach 80% at best (DF = 5))
- A significant improvement of the  $^{99}\text{Tc}$  recovery from HLW is only possible if it is converted into a single chemical species  $\Rightarrow$  difficult task



# Separation Of Long-lived Fission And Activation Products (cont'd)

- **Iodine**
  - **Generation rate = 7.1kg/GWe-yr**
  - **Isotopic composition: 80%  $^{129}\text{I}$  ( $T_{1/2} = 16 \times 10^6$  yr), and 20% stable  $^{127}\text{I}$**
  - **$^{129}\text{I}$  occurrence in spent fuel: molecular iodine, soluble  $\text{CsI}$ , solid  $\text{ZrI}_{4-n}$ , and volatile  $\text{ZrI}_4$**
  - **$^{129}\text{I}$  is in most of the land-based spent fuel repository concepts the first nuclide to emerge in the biosphere due to its very high mobility in aquifers**
  - **$^{129}\text{I}$  is removed from the dissolver solution during reprocessing (PUREX, UREX) with a yield ~95-98% (DF ~20-50)**



# Separation Of Long-lived Fission And Activation Products (cont'd)

- $^{129}\text{I}$  radioactive concentration in spent fuel is, depending on the burnup,  $\sim 1.6 \times 10^9$  Bq/tHM and its Annual Limit of Intake (ALI) is  $2 \times 10^5$  Bq
- $^{129}\text{I}$  radiotoxicity is the highest among the fission products ( $1.1 \times 10^7$  Sv/Bq), and it is very soluble  $\Rightarrow$  increased separation yields should be aimed at (DF  $\sim 10^3$  would be a significant improvement)
- Higher separation yields require more complex chemical treatments (e.g., high temperature pyrochemical processes)



# Separation Of Long-lived Fission And Activation Products (cont'd)

- Separated  $^{129}\text{I}$  can either be stored on a specific (zeolite) adsorbent, or discharged into the ocean
- In a salt-dome (evaporated sea water) type of repository the dilution of eventually migrating  $^{129}\text{I}$  by the mass of natural iodine ( $^{127}\text{I}$ ) present in the body of the salt dome strongly decreases the radiological hazard
- Final storage in a salt dome is an alternative which undoubtedly has its merits but, conditioning and confinement are the preferred  $^{129}\text{I}$  management options to reduce its radiological impact



# Separation Of Long-lived Fission And Activation Products (cont'd)

- $^{79}\text{Se}$ 
  - $T_{1/2} = 65'000$
  - Generation rate = 0.16 kg/GWe-yr
  - Chemically it is a sulfate
  - $^{79}\text{Se}$  separation from HLLW is not obvious because of the very small chemical concentration in which it occurs, in comparison with natural sulfur compounds
  - Incorporated in vitrified waste
  - Radioactive concentration in spent fuel is  $\sim 2 \times 10^{10}$  Bq/tHM, and its Annual Limit of Intake (ALI) is  $10^7$  Bq



# Separation Of Long-lived Fission And Activation Products (cont'd)

- $^{93}\text{Zr}$  and  $^{135}\text{Cs}$ 
  - $T_{1/2} = 1.5 \times 10^6$  and  $2 \times 10^6$  yrs, respectively
  - Generation rate = 23 kg/GWe-yr and 12.5 kg/GWe-yr, respectively
  - $^{93}\text{Zr}$  and  $^{135}\text{Cs}$  are contained in the spent fuel by other radioisotopes which are either very radioactive ( $^{137}\text{Cs}$ ) or present in much larger quantities (23 kg/GWe-yr  $^{93}\text{Zr}$  among 118 kg/GWe-yr Zr)  $\Rightarrow$   $^{93}\text{Zr}$  and  $^{135}\text{Cs}$  separation presently excluded, both from a technical and an economical point of view



# Separation Of Long-lived Fission And Activation Products (cont'd)

- $^{126}\text{Sn}$ 
  - $T_{1/2} = 250'000$  yrs
  - Occurs partly in the insoluble residues, and is partly soluble in the HLLW
  - $^{126}\text{Sn}$  generation rate = 0.72 kg/GWe-yr (1.81 kg /GWe-yr for Sn)
  - $^{126}\text{Sn}$  radioactive concentration in HLLW is  $\sim 3.2 \times 10^{10}$  Bq/tHM, and its Annual Limit of Intake (ALI) is  $\sim 3 \times 10^6$  Bq
  - $^{126}\text{Sn}$  is accompanied by a series of stable isotopes ( $^{116}, ^{118}, ^{119}, ^{120}, ^{122}, ^{123}, ^{124}\text{Sn}$ )  $\Rightarrow$  transmutation difficult



# Separation Of Long-lived Fission And Activation Products (cont'd)

- $^{14}\text{C}$ 
  - $T_{1/2} = 5'730$  yrs
  - Soluble in groundwater, important radiotoxicological role because of its uptake into the biochemical life cycle
  - Its role in the long-term radiotoxicity is dependent on the physico-chemical conditions occurring in deep underground aquifers or in water unsaturated geospheres
  - The capture cross section in a thermal neutron spectrum is negligibly small



# Separation Of Long-lived Fission And Activation Products (cont'd)

- **$^{36}\text{Cl}$** 
  - 5 to 20 ppm chlorine impurity in Zircaloy cladding  $\Rightarrow$  producing  $^{36}\text{Cl}$
  - $T_{1/2} = 300'000$  yrs
  - Arises in the dissolver liquid and remains, in part, within the washed Zircaloy hulls
  - At 45 GWd/tHM  $\sim 2 \times 10^6$  Bq in the HLW and MLW
  - The Annual Limit of Intake (ALI) is  $\sim 2 \times 10^7$  Bq
  - $^{36}\text{Cl}$  is gradually dissolved in groundwater and could contaminate water bodies around a repository
  - $^{36}\text{Cl}$  cannot be considered in a recovery or transmutation scenario



# Conclusions On Advanced Aqueous Reprocessing Options With P&T

- $^{237}\text{Np}$  separation is technically feasible in refurbished reprocessing plants or in newly designed facilities
- Separation of MAs has been demonstrated at the hot-cell lab level; the corresponding flow sheets are very complex and need to be simplified in order to allow up scaling to technological levels
- The secondary wastes expected from MAs separation are of the same order of magnitude as from the second cycle of the PUREX process



## Conclusions On Advanced Aqueous Reprocessing Options With P&T (cont'd)

- Separation of MAs followed by conditioning as a ceramic waste form and retrievable storage is a valid option for the MAs management
- Among LLFP only  $^{129}\text{I}$  has a radiotoxicity comparable to that of the actinides (the radiotoxicity of the other FP is  $\sim 10^3$  smaller); isolation and specific conditioning of  $^{129}\text{I}$  is advisable
- Transmutation of  $^{99}\text{Tc}$  is technically feasible, but at very high economic burden



## Conclusions On Advanced Aqueous Reprocessing Options With P&T (cont'd)

- All the other LLFP cannot be effectively separated from HLLW, unless isotopic separation techniques are envisaged
- The activation products C-14 and Cl-36 are troublesome contaminants of medium level wastes



For more information, please visit

<http://www.iaea.org/inis/aws/fnss/>

*Thank You !*

