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Key Note Lecture: Why Partitioning & Transmutation (P/T)? ... and how to implement it

Concetta FAZIO

Program Nuclear Safety Research Forschungszentrum Karlsruhe D-76344 Eggenstein-Leopoldshafen Germany IAEA/ICTP School on Physics, Technology and Applications of Accelerator Driven Systems (ADS) Trieste, Italy, 19 – 30 November 2007



Key Note Lecture: Why Partitioning & Transmutation (P/T)?

.... and how to implement it

Concetta Fazio Program Nuclear Safety Research

Forschungszentrum Karlsruhe Hermann-von-Helmholtz Platz 1 76344 Eggenstein-Leopoldshafen Phone: +49 7247 825517, Fax: +49 7247 825508, e.mail: concetta.fazio@nuklear.fzk.de

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Outline



- Part 1
 - Energy demand and population
 - Waste arising from fuel irradiation in current reactors;
 - Role of Partitioning and Transmutation (P/T);
 - Reactor and fuel cycle options to implement P/T;
 - Potential impact of P/T
- Part 2
 - Challenges for P&T development and selected results
 - Cost considerations
 - A regional approach?
 - Conclusions

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Energy Demand and population growth





International Energy Agency IEA



Nuclear Power Plants world wide





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Waste arising from fuel irradiation in current reactors





Today options for the nuclear fuel cycle:

1- Direct disposal of Spent fuel

2- Spent fuel reprocessing and Pu recovery



Composition of Spent Nuclear Fuel e.g.: Standard PWR 33GW/t, 10 yr. cooling





Most of the hazard stems from Pu, MA and some LLFP when released into the environment, and their disposal requires isolation in stable deep geological formations.

A measure of the hazard is provided by the radiotoxicity arising from their radioactive nature.

1 tonne of SNF contains:

955.4 kg <mark>U</mark> 8,5 kg <mark>Pu</mark>

Minor Actinides (MAs) 0,5 kg ²³⁷Np

0,6 kg Am 0,02 kg Cm

Long-Lived fission Products (LLFPs) 0,2 kg ¹²⁹I

0,8 kg ⁹⁹Tc 0,7 kg ⁹³Zr 0,3 kg ¹³⁵Cs

Short-Lived fission products (SLFPs) 1 kg ¹³⁷Cs 0,7 kg ⁹⁰Sr

Stable Isotopes

10,1 kg Lanthanides 21,8 kg other stable

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Radiotoxicity of the spent nuclear fuel



Evolution of the radiotoxic inventory, expressed in sievert per tonne of initial heavy metal (uranium) (Sv/THM) of UOX spent fuel unloaded at 60 GW d/t, versus time (years).



What is Partitioning and Transmutation (P/T)



- **Objectives**: Minimization of long-lived nuclides in High Level Waste (HLW), thus alleviation of the burden of a final disposal.
- Partitioning: Chemical separation of long lived nuclides from HLW
- **Transmutation**: Use of nuclear reactions to transform long lived nuclides into stable or short-lived nuclides

P/T applies to **TRU** (**Pu** and **M**inor Actinides) and Long Lived Fission Products. **Plutonium** is a special case: it can be considered as a valuable resource or part of the wastes. However, P/T technologies apply to the most general case.

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A short historical perspective on P/T (1/4)



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Mid-80s

Early P/T studies, mostly in Europe and in the US.

The physics is first explored together with some pioneering partitioning studies.

Early studies on the impact of P/T on fuel cycle, P/T motivations, possible P/T "metrics" for cost/benefits evaluation.

> Both IAEA and EURATOM did issue extensive reports with lukewarm conclusions: the challenge did seem to be formidable, without a clear strategy.

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A short historical perspective on P/T (2/4)



Late 80s

Early-90s

The "OMEGA" initiative in Japan, motivated by a strong public opinion concern about waste management. Japan requests OECD-Nuclear Energy Agency to organize international cooperation and information exchange in the field of P/T.

First OECD-NEA Information Exchange meeting on P/T in 1990 at Mito, Japan. These conferences are still going on today, with a two years pace

At the same time in France, the waste management issue is discussed at the political level and a law is passed in 1991, in order to study possible strategies (including P/T) during a fifteen years period (1991-2006).

A National Commission of Evaluation, appointed by the Government, is put in place.

In the specific US context, the idea of a "Acceleratordriven Transmutation of Wastes (ATW)" is launched at LANL, based on previous work.

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A short historical perspective on P/T (3/4)



Early-90s

International discussions on **"metrics"** and motivations focus on the waste doses or "radiotoxicity". This notion is controversial:

• Geologists and repository experts point out that the potential return to the bio-sphere of wastes is dominated by a few LLFP (like I-129), more mobile than Transuranics (Pigford).

• Moreover, the contribution to the dose after long periods of time would be in any case very small.

However, safety experts point out that one has to consider:

"normal" scenarios of evolution in time of the geological environment of the repository, and

"abnormal" evolution scenarios, like human intrusion. These scenarios point out to the role of the "potential source" of radiotoxicity (e.g. at ingestion), which is dominated by the TRU contribution.

> <u>Japan & France</u>: significant resources in particular in the field of P, in order to achieve scientific demonstrations of feasibility of different separation processes <u>Europe</u>: R&D Framework Programs with projects focused on chemistry and on ADS-based transmutation. <u>US</u>: AFCI program is started.

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A short historical perspective on P/T (4/4)



2000

Key demonstration of the potential beneficial effects of P/T on a specific repository from the point of view of its design and operation, accounting for both thermal constraints and peak dose rate constraints.

> A turning point: the objectives of GENERATION-IV do include P/T (waste minimization). P/T is seen from now on, as consistent with sustainability and nonproliferation objectives: it is the path towards "Advanced Fuel Cycles".

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Geological repositories of High Level Waste: host formations



- The main type of host formations considered at present are:
 - Hard rock formations (mainly granite), studied in Canada, Finland, Japan, Switzerland, Spain, Sweden
 - Argillaceous formations (clay, mudstone) studied in Belgium, France, Japan, the Netherlands, Spain Switzerland, Germany
 - Salt formations (salt layers and salt domes) studied in Germany and the Netherlands
 - Volcanic formations (tuff and basalt) studied in the United States, i.e. Yucca Mountain.

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Nuclear reactions for <u>transmutation</u> of Longlived nuclides



- Long-lived nuclides: Minor Actinides & some of LLFP
 - LLFP: FPs with half-live longer than 30 years such as ⁹⁹Tc (half-life 2x10⁵ y), ¹²⁹I (half-life 1.6x10⁷ y)
- **Neutron reactions** are the only reactions for effective transmutation of **MA** (neutron fission) and **LLFP** (neutron capture). However: for MA, neutron **fission** is always in competition with **capture**.
- Fast neutrons are best for MA transmutation:
 - Most MA have "threshold" fission (i.e.fission only at high neutron energy)
 - Thermal neutrons produce, via neutron capture, more high atomic number MA than cause fission of MA
 - More favourable fission-to-capture probability ratio with fast neutrons
- **Thermal neutrons** better for **LLFP transmutation** (higher capture probabilities) but transmutation rate is very slow. No major benefits, even if LLFP more "mobile" in geological environment.
- No effective means of transmutation of Sr-90, Cs-137 (half-lives ~30 yrs)

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Main features of fast neutron reactor physics



Fast neutron spectrum reactors have a favourable neutron economy with respect to thermal neutron spectrum reactors



Fission-to-Absorption Ratio for PWR and SFR



- Fissile isotopes (e.g. U-235, Pu-239, Pu-241) are likely to fission in both thermal/fast spectrum.
 However, the fission fraction is higher in fast spectrum
- Moreover, significant fission (up to 50%) of fertile isotopes (e.g. U-238, Pu-240) in a fast spectrum

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Scenarios to implement P/T: the three most significant examples



<u>Scenario 1</u>: Sustainable development of nuclear energy with waste minimisation. One type of reactor, one fuel type, one reprocessing process

<u>Scenario 2</u>: "Double strata" fuel cycle: 1) commercial reactors with Pu utilisation 2) separate MA management. Two separate fuel cycles.

<u>Scenario 3</u>: Reduction of TRU stockpiles (e.g. as a legacy from the past operation of power plants)

Scenarios 1 and 2 imply the continuous use of nuclear energy, the stabilisation of the TRU stocks in the fuel cycle and the minimisation of wastes in a repository.

All three scenarios go beyond the strategy of "once-through" ("open") fuel cycle (i.e. the final storage of irradiated fuel), and imply fuel reprocessing.

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Scenario 1: Reference scenario for a sustainable development of nuclear energy with waste minimisation



CR = Conversion ratio = fissile material produced/fissile material destroyed. CR< 1 -> "burner"; CR>1 -> "breeder"

- The multiple recycle of TRU is feasible in a Fast Reactor (FR), whatever its coolant and fuel type: oxide, metal, carbide or nitride
- 2-5% MA in the fuel: close to standard fuel, if homogeneous recycle chosen and CR>0.8
- → Some impact on the fuel cycle, e.g. at fuel fabrication, due to the Cm-244 spontaneous fission neutron emission
- Reprocessing needed to recover notseparated TRU (enhanced proliferation resistance)
- A possible variant: heterogeneous (i.e. target) recycle of MA at the periphery of the core, while Pu recycled as standard fuel in the core. Needs separation of MA from Pu
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Scenario 1: Why not Thermal instead of Fast Reactors?

Consequences on fuel cycle parameters of full TRU recycling in LWRs, e.g. at fuel fabrication:

| Parameter | Multiplying factor ^(a) | |
|----------------|-----------------------------------|-----|
| Activity | ~0.5 | - |
| α-heat | ~ 3 | |
| β-heat | ~ 0.5 | |
| γ-heat | ~ 3.5 | |
| neutron source | ~ 8000 | (a) |

Unacceptably high effect due to high capture cross-sections in thermal spectra, which favour the production of Cf-252 (strong neutron emitter by spontaneous fission)

^{a)} Reference value (=1): Case of Pu-only multirecycling

Cf-252 inventory in the core. Case of full TRU multirecycling in:



<u>Scenario 2</u>: « Double strata »: Pu still a resource. Gen-IV FR deployment delayed





- → The <u>Pu inventory</u> can be stabilized.
- → <u>MA management</u> in dedicated transmuter systems: 1) critical Fast Reactors, or 2) subcritical Accelerator Driven Systems (ADS) with U-free fuels
- → Fuel: New fuel (with high MA content) needs to be developed. In the case of U-free inert matrix is foreseen, thus new fabrication processes.
- → Reprocessing: to be developed in particular for U-free fuels. Choice of support matrix in fuel is relevant. What chemical process: aqueous or pyrochemistry?
- ➔ The "support" ratio, i.e. the ratio of total power of the dedicated systems to the total power of the power generating systems is of the order of 6%

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Scenario 3: Reduction of Pu+MA stockpile (Pu considered as waste)





- →Limited number of dedicated transmuters: need to account for last transmuter in-core inventories
- →Fuel in the dedicated transmuters: Pu/MA ~ 80/20 to be developed
- →New Fabrication processes needed.
- Reprocessing of transmuter fuel: to be developed. What type of chemical process: aqueous or pyrochemical?

If timeframe for reducing stockpile ~100 y, ~20% of initial stockpile is not burnt. This depends on a) transmutation rate (~ 5%/year) and b) from fuel cycle characteristics (e.g. cooling time, reprocessing and refabrication time)

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Potential benefits of P/T



P/T offers significant potential benefits to the fuel cycle:

- Reduction of the potential source of radiotoxicity in a deep geological storage ("intrusion" scenario)
- Reduction of the heat load: larger amount of wastes can be stored in the same repository
- If TRU are not separated (e.g. in the homogeneous recycling in a Fast Neutron Reactor), improved proliferation resistance is expected

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Radiotoxicity reduction





Radiotoxicity reduction is comparable (i.e. higher than a factor 100) in transmutation scenarios 1 and 2, and depends on losses during reprocessing. In the cases presented here a 0.1 % value is taken for all TRU.

However, the impact on the fuel cycle is different. It becomes unacceptably high if all TRU recycled in LWRs, due to the high neutron doses at fuel fabrication (as shown previously).

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Impact of the actinides management strategy on the raditoxicity / Activity of ultimate Waste







Potential Benefits of a Closed Fuel Cycle (P/T) based on Fast Reactors for Waste Management



- Certain elements (plutonium, americium, caesium, strontium, and curium) are primarily responsible for the decay heat that can cause repository temperature limits to be reached
- Large gains in repository space are possible by processing spent nuclear fuel to remove those elements
- The recovered elements must be treated:
 - Caesium and strontium can be stored separately for 200-300 years
 - Plutonium, americium, and curium can be recycled for transmutation and/or fission by irradiation in fast reactors



Part 2: CHALLENGES for P&T development and SELECTED RESULTS



- The <u>Physics</u> of Transmutation is well understood: experiments have been performed irradiating pure TRU isotope samples in power reactors, and transmutation rates have been compared successfully to calculations.
- <u>Chemistry of isotope partitioning</u> and <u>MA-based fuels</u> development are major challenges.
 - Moreover, an industrial deployment implies to upgrade the most promising technologies from the laboratory scale.
- The optimisation (economy, safety, transmutation performance) of <u>innovative fast reactors</u> (in particular ADS) is also a significant challenge.
- **Overall cost** considerations are of course essential.



Technical challenges to Actinide Separations



Requirements

- High separation efficiency (reduction of process losses);
- High selectivity of the extraction agents;
- Compatibility with existing separation processes (e.g. PUREX);
- Minimisation of secondary wastes and effluents.
- Alternative methods to be studied: single separation or "Grouped" separation of TRU
- Cost reductions

Considered processes

- Aqueous process
 - Liquid/liquid extraction
 - Long technical experience
 - Compatible with present reprocessing techniques (e.g. PUREX)
- <u>Dry process (pyrochemistry)</u>
 - Combination of electrolytical processes and extractions
 - Could be suitable for high burn-up fuel and short cooling time
 - Small installations
 - However, at present experience only at laboratory scale

Actinides chemistry is complex: e.g. actinides form multiple valence states, similar to that of lanthanides

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Importance of Processing Loss Fraction Impact of loss fraction 1.E+04 ← 0.1% Loss 1.E+03 **-**0.2% Loss **Foxicity** 1.E+02 Relative 1.E+01 1.E+00 1.E-01

Radiotoxicity goal cannot be achieved if loss fraction increases beyond 0.2% and extends to 10.000 years at 1% losses

100

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Time (years)



1000

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10000



To go towards the full TRU recycle in fast reactors



The Grouped Actinide Extraction «GANEX» developed at CEA



Developments in Europe to be compared to the suite of UREX+ Processes developed in the USA



| Process | Prod #1 | Prod #2 | Prod #3 | Prod #4 | Prod #5 | Prod #6 | Prod #7 |
|---------|---------|---------|---------|---------|----------|---------|---------|
| UREX+1 | U | Тс | Cs/Sr | TRU+Ln | FP | | |
| UREX+1a | U | Тс | Cs/Sr | TRU | All FP | | |
| UREX+2 | U | Тс | Cs/Sr | Pu+Np | Am+Cm+Ln | FP | |
| UREX+3 | U | Тс | Cs/Sr | Pu+Np | Am+Cm | All FP | |
| UREX+4 | U | Тс | Cs/Sr | Pu+Np | Am | Cm | All FP |

Notes: (1) in all cases, iodine is removed as an off-gas from the dissolution process.

(2) processes are designed for the generation of no liquid high-level wastes

U: uranium (removed in order to reduce the mass and volume of high-level waste) Tc: technetium (long-lived fission product, prime contributor to long-term dose at Yucca Mountain) Cs/Sr: caesium and strontium (primary short-term heat generators; repository impact) TRU: transuranic elements (Pu: plutonium, Np: neptunium, Am: americium, Cm: curium) Ln: lanthanide (rare earth) fission products

FP: fission products other than caesium, strontium, technetium, iodine, and the lanthanides

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Technical challenges to Fuel Development



- Large decay heat and high neutron emission of MA give new problems with respect to standard fuel manufacturing
- However problems are smaller if the fuel contains U and small amount of MA (as in the case of <u>Scenario 1</u>) with respect to U-free fuels (as in the case of <u>Scenarios 2 and 3</u>) with large amounts of MA.
- In the case of U-free fuels, the choice of the support/matrix (e.g. for oxide fuels: MgO, ZrO₂, Mo...) is crucial for a good thermal behaviour under irradiation.
- Fabrication processes are challenging (avoid contamination etc.), in particular for a significant content of Cm.
- > In any case, remote handling is needed

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Transmutation fuel development is considerably more challenging than conventional fuels



- Multiple elements in the fuel: U, Pu, Np, Am, Cm
- Varying thermodynamic properties: e.g. High vapor pressure of Am
- Impurities from separation process: e.g. high lanthanide carryover
- High burn-up requirements
- High helium production during irradiation
- Remote fabrication & quality control





Experimental program on Transmutation fuels and targets: the CEA program as an example



Minor Actinides

- Transmutation in Homogeneous mode: SUPERFACT and METAPHIX are first demonstrations of technical feasibility
- Transmutation in **Heterogeneous** mode: by 2008
 - Important experience feedback concerning inert matrices and fissile compounds
 - MgO is today the reference matrix, ZrO₂ (and Mo⁹²) is an alternative
 - MgAl₂O₄ is discarded: not stable enough under irradiation
 - ECRIX (Irradiation including Am) is loaded in Phénix
 - **Demonstration of the technical feasibility** is partially achieved (confirmation with PIE after 2007)
- Between 2008 and 2010
 - Post irradiation examinations will allow design optimisations and improvement of the performances

Long-Lived Fission Products

- ANTICORP 1 (Metallic Technetium) is loaded in Phénix: demonstration of the technical feasibility is partially achieved (confirmation in 2009)
- Transmutations of I and Cs are definitely abandoned

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The role of <u>fast spectrum irradiation</u> reactors is still essential



Irradiations experiments performed in France on fuels with minor actinides



SUPERFACT homogeneous: results



- ➤ Transmutation ratios at the reactor middle plane are ≈ 30 % for ²³⁷Np and ≈ 28 % for ²⁴¹Am confirming the calculated values.
- Same fuel microstructure evolution than standard fuels ones
- U, Pu, Am and Np radial distributions are very flat, i.e. no actinides redistribution
- For low Linear Power, no real influence of the low MA amount up to a BU equal to 6,7 at%, except for the He release of the Am fuel (4 times higher than standard pins ones)



Cross section of SUPERFACT pin after irradiation



SEM image of SUPERFACT pin: strong intergranular porosity due to fission gas release

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Demonstration underway of homogeneous recycle in metal fuel...

Figure 14. Irradiation test pins for METAPHIX (unit : mm) A, C: U-Pu-Zr reference specimen; B: U-Pu-Zr-MA-RE specimen (diameter of pin = 0.55 mm, cladding = 15/15Ti cw)



Transmutation in dedicated systems: the ADS case and the needs for demonstration





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ADS demonstration: The European program as an example

In Europe, a series of demonstrations of separate **ADS** components, has been performed



Figure D.8: Muse ADS experiment with the Masurca reactor and the Genepi accelerator; system power along a reactor section cross line (in blue: critical core without source, in red: subcritical mode with source).

A 1MW liquid **LBE** spallation target: the **MFGAPIF** initiative



Figure D.9: Megapie spallation target (left: global view, right: detail

The physics of the sub critical core: the **MUSE programme**

Some crucial components of a high intensity proton accelerator.....



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MEGAPIE: 1MW liquid LBE neutron spallation target

- 1999-2001: Conceptual design phase and R&D activities
- ≻2001-2004: Component tests (e.g. EMP, Window cooling)
- ➤2004-2005: Construction and out-of-beam tests
- >2005-2006: Out-of-beam tests and

Target irradiation August 2006 – December 2006

Beam power (black, arbitrary units) and temperatures in the target during the start-up phase.





Achievement: Safety, Licensing and Demonstration of technical feasibility

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MEGAPIE: Neutron gain







Neutronenfluss SINQ Strahlkanal 32 NEUTRA

Markus Lüthy Koordination Betrieb Anlagen West 8830

| Preliminary calculations and |
|----------------------------------|
| measurement values of thermal |
| neutron flux measurements |
| (n/cm2/s/mA) with the MEGAPIE |
| target and the solid SINQ target |
| (2005). |
| |

| | | 51032000 |
|--------------------------|--|--|
| Measurement Positions | Ratio MEGAPIE/Solid target Experimental Values | Ratio MEGAPIE/Solid target Calculated Values |
| NEUTRA (30) | 1.85 | 1.57 |
| ICON (50) | 1.81 | 1.68 |
| EIGER (82) | 1.60 | 2.02 |
| NAA | 1.80 | 1.77 |

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ADS demonstration: the Eurotrans Project





Design of an experimental (XT-ADS) and an industrial (EFIT) system (DM1 DESIGN)



CERCER MgO CERMET Mo Liquid metal technology, thermalhydraulics and materials studies

U-free MA Fuel Development

(DM3 AFTRA)





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Cost Evaluations for Advanced Fuel Cycles



- Cost evaluations for advanced fuel cycles (including P/T) have been recently performed in the frame of two different OECD-NEA working groups.
- In particular, cost evaluations have been performed for the two major strategies for implementing P/T, i.e. the "double strata", where ADS multirecycle MA, and the full recycling of not-separated TRU in fast reactors.
- The increase in electricity cost due to advanced fuel cycles, has been found to be "relatively" limited (10% to 20%) compared to the once through fuel cycle
- However the authors of the studies underline the uncertainties associated to these evaluations.
- Moreover technical uncertainties on some innovative techniques, their feasibility and performance should be accounted for.

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Cost Evaluations for Advanced Fuel Cycles



Reference: Advanced Fuel Cycles and Waste Management. OECD/NEA Publication.



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Summary on cost evaluation



- Both the study performed in 2002 and the more recent one (2005-2006) show the same trends in the electricity production costs and its breakdown
- The reactor investment cost dominates the Electricity production costs (56-63%). Reactor investment + operating and maintenance is 80-90% of the overall cost
- In the more recent study the cost of FR is conservatively estimated to be 20% higher than LWR. The cost of ADS is higher of that of FR by 40% to 170%.
- In both studies, the estimated part in electricity costs related to the fuel cycle and the repository are relatively limited.
- The fuel cycle costs uncertainties can be very large (up to factors of 2) and depend on the maturity level of technologies and on local conditions. However, their impact is relatively small.

However, which can be the industry point of view?

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A Regional Approach to the Fuel Cycle with P/T



- Different countries can envisage different policies. According to the strategy, specific fuel cycle facilities have to be deployed.
- Some of these facilities are similar, even if conceived for different strategies.
- The multiplication of such facilities is unlikely, both for nonproliferation and economic reasons
- Can a regional (i.e. with some shared installations and combined resources) approach help? (Consistent with provider/user state concept).
- As an example, consider the case of:
 - A country group « A », which has a spent fuel legacy, no reprocessing installations and no decision yet on final repository.
 - A country group « B », which has an operating power reactor fleet with a waste minimisation objective, has reprocessing capabilities, but looks for an optimisation of resources and investments.

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Scenarios 1 and 2 in a Regional approach



Scenario 1

- This scenario considers the deployment of fast reactors in Group B countries. These fast reactors are deployed with the Plutonium of the two groups and recycle all the minor actinides.
- <u>The main objective</u> of this scenario is to decrease the stock of spent fuel of Group A countries down to 0 at the end of the century and to introduce Gen-IV fast reactors in group B, starting, e.g., in 2035.

<u>Scenario 2</u>

- This scenario considers the deployment of a number of ADS shared by the two groups of countries.
- The ADS will use the Plutonium of the Group A and will transmute the minor actinides of the two groups.
- The Plutonium of the Group B is continuously recycled in PWRs.
- <u>The main objective</u> of this scenario is to decrease the stock of spent fuel of Group A countries down to ~0 at the end of the century, and to stabilize both Pu and MA inventories of Group B countries.

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Scenario 1 in a regional approach





Scenario 2 in a regional approach





Scenario 2 in a regional approach: ADS deployment





Scenarios studies: regional approaches



- Promising results shown in these preliminary studies (i.e. benefits for both groups of countries in terms of investments, use of resources etc.)
- However, there are major issues:
 - Extensive transports of fuels
 - National independence
 - Repository (one "regional" site? How acceptable?)
 - Rationale for funding share

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How to implement the "regional" concept in practice?

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Conclusions (1/2)



- P/T technologies offer the potential for a significant radioactive waste minimisation
- > P/T can be applied to widely different fuel cycle strategies:
 - Sustainable development of nuclear energy
 - Minimisation of the waste arising from a legacy of spent nuclear fuel
- P/T does not eliminate the need for a deep geological storage whatever the strategy but allows to increase its capacity, to reduce drastically the burden on it and improve public acceptance

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Conclusions (2/2)



- Fast Reactors offer the most flexible tool in order to implement P/T: The results of the studies clearly indicate a consensus on the fact that to reach the optimum performances of P/T, <u>fast spectrum reactors and fully</u> <u>closed fuel cycles</u> are needed, together with chemical processes which allow reaching ~99.9% recovery of all TRU.
- Demonstration of P/T implies the demonstration of all the "building blocks": adapted fuels, adapted reprocessing techniques, reactor loaded with significant quantities of MA
- LLFP transmutation is questionable. However the Cs and Sr management is a relevant issue.

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