Development of a GANEX Process

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Waste is what is left when imagination fails
Nuclear Waste Handling

- The used nuclear fuel contains excess uranium, FP and transuranic elements (TRUs)

- Highly radiotoxic
  - short term mostly due to FP
  - long term mostly due to TRUs

- Needs to decay for >100 000 years to equal uranium

- The amount of used nuclear fuel in the world is 350 000 tons

- Different strategies for handling the radioactive waste:
  - Once through option
  - Wait and see – no decisions made
  - Reprocessing
Nuclear Waste handling

Reprocessing

- Recycling of parts of the used fuel
- < 2% of the energy is utilized in the once through fuel cycle
- U and Pu are separated (PUREX process) to make new fuel
- Increases the energy utilisation with 25% (->2.5%)
- 100 000 tons of spent fuel has been reprocessed
  The capacity is ca 4800 tHM/year
## Nuclear Waste handling

<table>
<thead>
<tr>
<th>Country</th>
<th>Facility</th>
<th>App. Capacity (tonnes per year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>France</td>
<td>La Hague</td>
<td>1700</td>
</tr>
<tr>
<td>United Kingdom</td>
<td>Sellafield (THORP, Magnox)</td>
<td>1500+900</td>
</tr>
<tr>
<td>Russia</td>
<td>Ozersk (Mayak)</td>
<td>400</td>
</tr>
<tr>
<td>India</td>
<td>Kalpakkam, Trombay, Tarapur</td>
<td>275</td>
</tr>
<tr>
<td>(Japan)</td>
<td>Rokkasho</td>
<td>800</td>
</tr>
</tbody>
</table>

- More expensive to reprocess than to use fresh fuel
- Proliferation resistance
- Long storage time needed for the reprocessing waste
Partitioning & Transmutation (Gen IV systems)

- Advanced version of reprocessing called: Partitioning and Transmutation (P&T)

- Separation of all the actinides from the fission products

- Radiotoxicity of the used nuclear fuel is mostly due to the transuranic elements Pu, Am and Cm
Partitioning & Transmutation

![Graph showing radiotoxicity over time for different elements and total](Image)
Partitioning & Transmutation

- P&T, a prerequisite for closing the fuel cycle…
Partitioning & Transmutation

- Why is separation of the actinides necessary?
  - E.g. Fission Products with high neutron capture cross section
  - Avoid build-up of new long-lived elements

- Method of choice to achieve the separation: Solvent extraction
  - Already existing technology (PUREX)
  - High purity

- Alternative Method to achieve the separation: Pyro processing
  - Proliferation safety
Solvent Extraction

- Distribution of a solute between two immiscible liquid phases
- Solutes can partition differently between the two phases (organic and aqueous) => Separation
- Molecules that form complexes with the solutes can be employed to enhance the affinity of either phase
The GANEX Concept

- **GANEX= Group ActiNide Extraction (GANEX)**
- A solvent extraction process aimed at P&T

- Previous P&T processes designed to come after PUREX
  - SANEX/DIAMEX, TALSPEAK (Heterogenous)

- **GANEX - aimed at replacing PUREX, more proliferation safe**
  - no pure plutonium stream (Homogenous)

- Extract all the actinides as a group directly from dissolved used fuel, with the removal of bulk U
The GANEX Concept

Homogenous recycling

Spent fuel

Co-conversion Fuel fabr.

Dissolution

U sep.

GANEX

U

εU+Pu+ MA

An sep.

Waste

Several different GANEX processes are under development
The Chalmers GANEX Solvent

Combine two well known extractants with different properties:

**1. bi-Terpyridine bis-Pyridine or BTBP**

Known to: Extract trivalent actinides and separate them from the trivalent lanthanides. Extract pentavalent actinides.

Due to strong acid and irradiation, a stable BTBP is needed: 
CyMe$_4$-BTBP
The Chalmers GANEX Solvent

2. *tri*-butyl phosphate or **TBP**
Known to: Extract tetra- and hexavalent actinides (PUREX process)

1+2 = > No need for redox control

The extractants (BTBP+TBP) should be combined into one solvent
”the GANEX solvent”

The diluent used for this (so far) is cyclohexanone
– solubility
– kinetics
The Development Process

• Screening of the Extraction Behaviour
  – An extraction, kinetics, An/Ln separation
  – Extractant behaviour – e.g. synergistic/antagonistic effects

• Stability
  – Hydrolytic
  – Radiolytic
  – Temperature

• Fission and Corrosion Product Handling
  – Extractions
  – Action plan- solutions

• Towards Process Implementation
  – FP Loading
  – Batch process test
  – Continuous tests
Screening of the Extraction Behaviour

Fast kinetics (< 20 min)

$SF_{\text{Am/Eu}} = 160$
Screening of the Extraction Behaviour

The extractants doesn’t react with each other

Neither affect the extraction behaviour of each other

1=BTBP+TBP
2=BTBP
3=TBP
4=cyclohexanone
1M HNO₃+3M NaNO₃

log(D)

organic phase 1
organic phase 2
organic phase 3
organic phase 4

Eu  U  Np  Pu  Am
Screening of the Extraction Behaviour

Plutonium extracted by BTBP in this solvent!
Important to consider in the context of Plutonium loading…
Pu loading

Original stock, 4.5 g Pu in HCL, add AgNO3, precipitation of AgCl
Pu loading

Three concentrations extracted (pure organic solvent to the left)
Pu loading

Three concentrations extracted

To the left: The solution directly from the precipitation! 37 mM 8.8 g/l

In the middle: 9.5 mM  2.2 g/l

To the right: 3.7 mM 0.88 g/l
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Stability

- Hydrolytic

- Radiolytic

- Temperature

Stable after nitrous acid scavenger addition
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Fission and Corrosion Product Handling
Fission and Corrosion Product Handling

1. Problematic FP/CP
2. complexed FP/CP
3. GANEX extraction

Removal of bulk U

Pre-extraction

GANEX extraction

Scrub
Fission and Corrosion Product Handling

Zr, Mo suppression

Pd, Ag, Zr suppression
The Development Process

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Towards Process Implementation

FP Loading in Jülich= affecting An extraction!  
- Large Iron content
Towards Process Implementation

HAR diluted 1:10, increased aq phase density

Centrifugal contactor test
Towards Process Implementation

<table>
<thead>
<tr>
<th>Element</th>
<th>% of initial in aqueous strip</th>
</tr>
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<tbody>
<tr>
<td>Eu</td>
<td>0.32</td>
</tr>
<tr>
<td>U</td>
<td>93.5</td>
</tr>
<tr>
<td>Np</td>
<td>99.8</td>
</tr>
<tr>
<td>Pu</td>
<td>99.6</td>
</tr>
<tr>
<td>Am</td>
<td>99.8</td>
</tr>
</tbody>
</table>

**Batch Process data**

Θ=0.6  
7 extraction steps  
6 acid scrub 1 steps  
6 acid scrub 2 steps  
1 strip step

**Centrifugal contactor data**

Θ=0.6  
15 extraction steps  
3 acid scrub steps  
1 strip step

<table>
<thead>
<tr>
<th>Element</th>
<th>% of initial extracted</th>
<th>% of initial in aqueous strip</th>
</tr>
</thead>
<tbody>
<tr>
<td>Eu</td>
<td>11</td>
<td>9.1</td>
</tr>
<tr>
<td>U</td>
<td>100.0</td>
<td>95</td>
</tr>
<tr>
<td>Np</td>
<td>100.0</td>
<td>96</td>
</tr>
<tr>
<td>Pu</td>
<td>100.0</td>
<td>58</td>
</tr>
<tr>
<td>Am</td>
<td>100.0</td>
<td>25</td>
</tr>
</tbody>
</table>
A GANEX process based on well known reagents and diluents (TBP, CyMe4-BTBP and cyclohexanone)

Process parameters developed and tested within EU FP7 project ACSEPT

Equipment will be AKUFVE centrifuges run in SISAK mode

Very high throughput but small dead volume = no criticality risk

Optimal storage tanks eliminating criticality issues

Small and flexible unit where different extraction processes can be tested and optimised

Based on a thesis by Emma Aneheim at Chalmers (co funded by the EC and SKB)
Conclusion

- Many aspects are needed to be investigated and understand before any separation process can be used practically

- Experiments are needed. Computer exercises are far from adequate for systems development or even proof of principle

- Proof of concept of a true **Group Actinide Extraction** system has been made, now…

- Further solvent improvements – performance, safety etc.
  - New diluent ?
  - New extractant ?
  - Development now part of EU project SACESS