Development of a GANEX Process

Emma Aneheim and Christian Ekberg

Waste is what is left when imagination fails

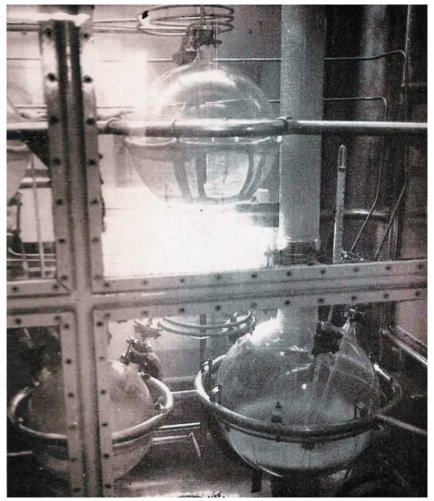
Nuclear Waste Handling

- The used nuclear fuel contains excess uranium, FP and trans uranic 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

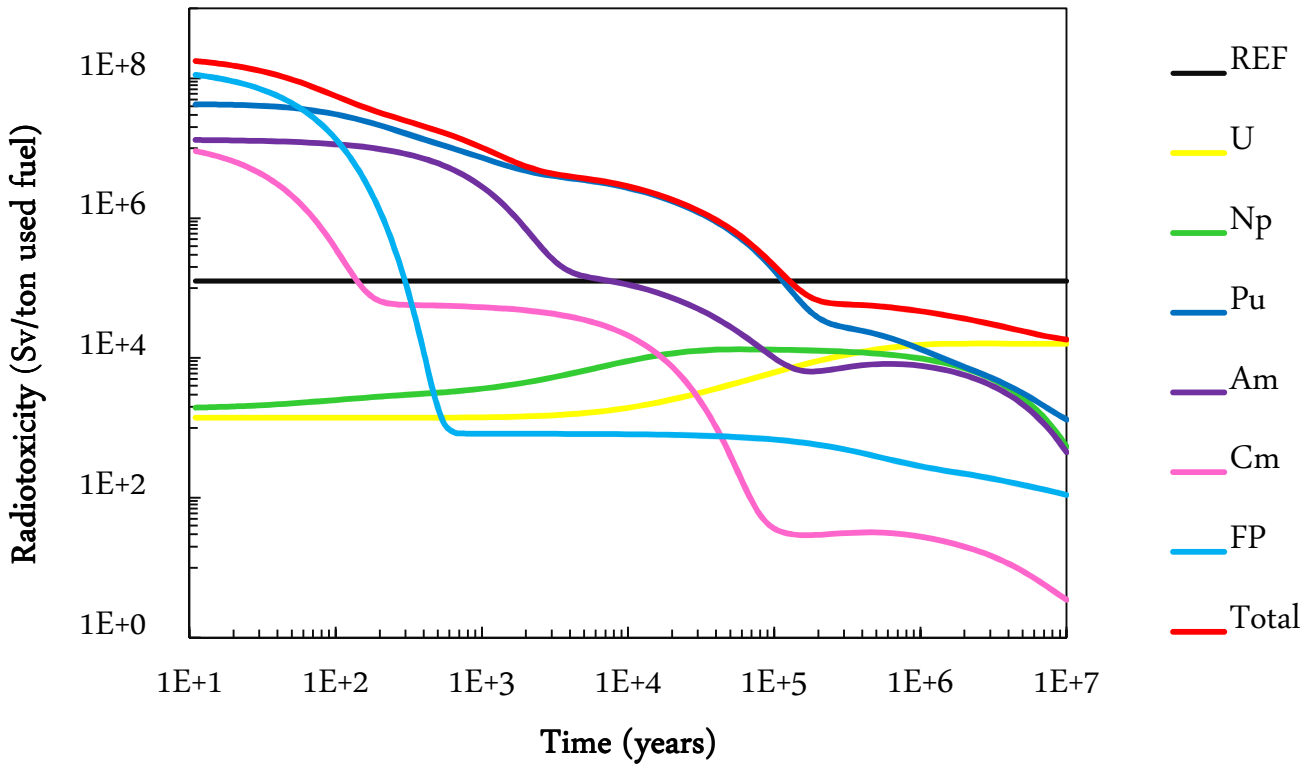
Country	Facility	App. Capacity (tonnes per year)
France	La Hague	1700
United Kingdom	Sellafield (THORP, Magnox)	1500+900
Russia	Ozersk (Mayak)	400
India	Kalpakkam, Trombay, Tarapur	275
(Japan	Rokkasho	800)

- More expensive to reprocess than to use fresh fuel
- Proliferation resistance
- Long storage time needed for the reprocessing waste

Partitioning & Transmutation (Gen IV systems)

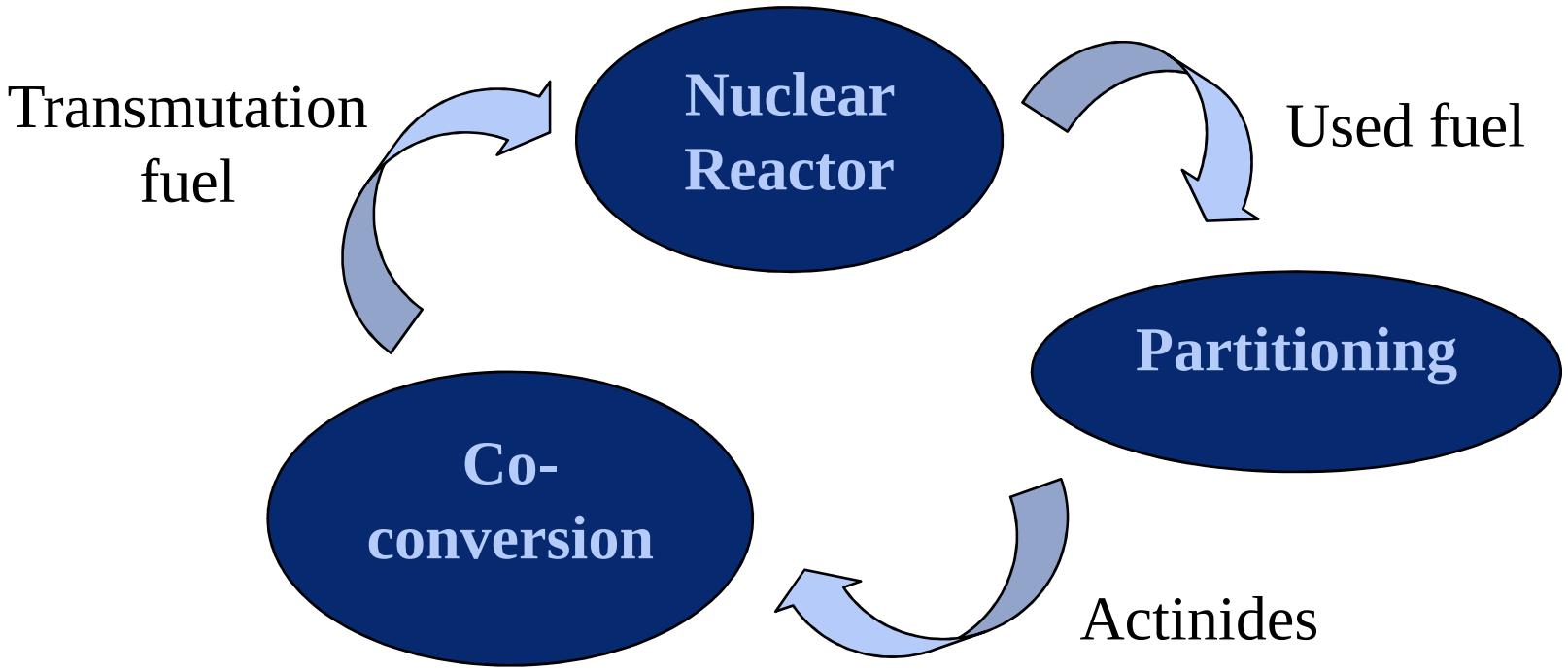
- Advanced version of reprocessing called: Partitioning and Transmutation (P&T)
- Separation of all the actinides from the fission products
- Radiotoxicity of the used nuclear fuel is mostly due to the transuranic elements Pu, Am and Cm

Partitioning & Transmutation



Partitioning & Transmutation

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

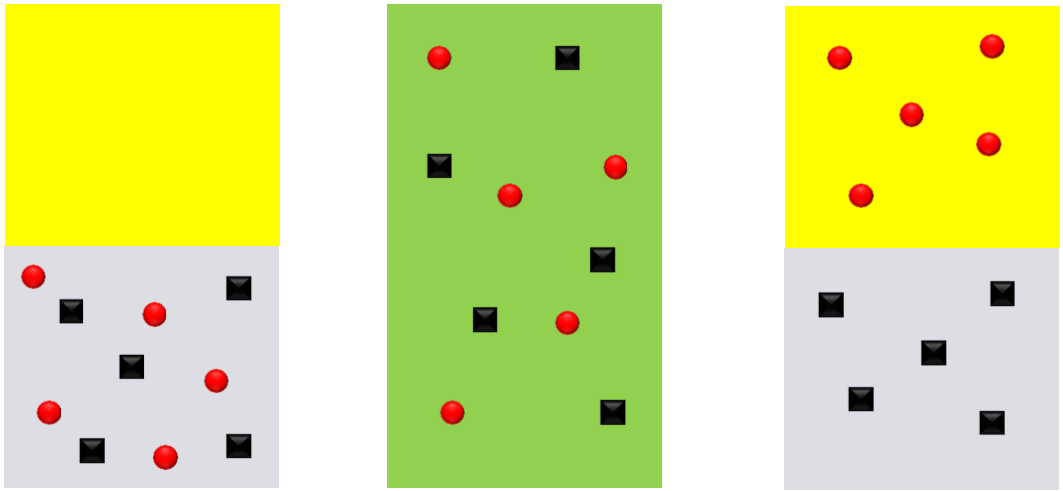


Partitioning & Transmutation

- 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

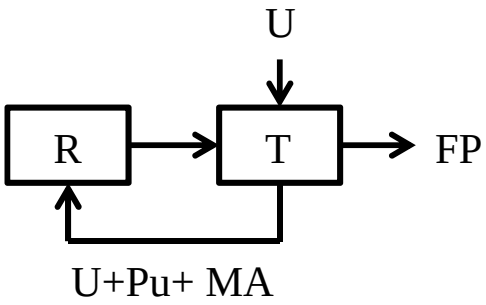
- GANEX= Group ActiNide Extraction (GANEX)
- A solvent extraction process aimed at P&T

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

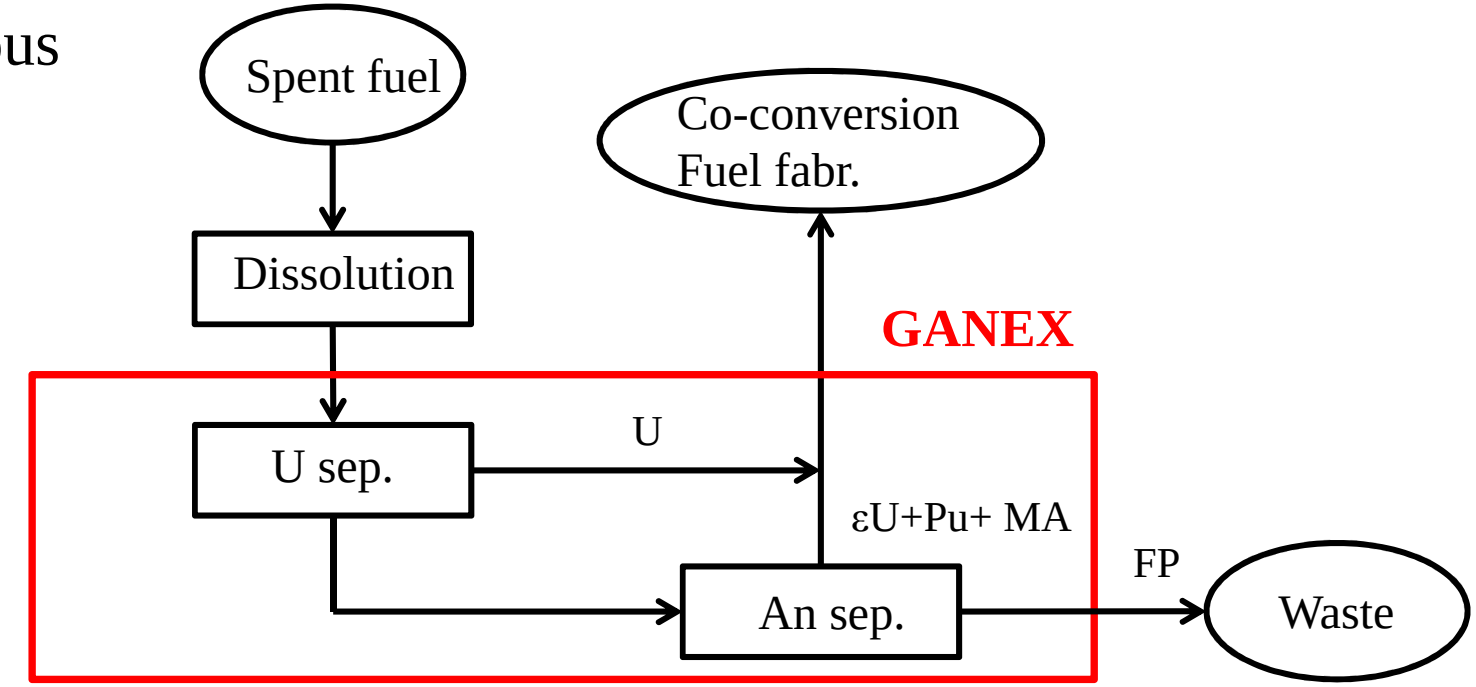
- GANEX - aimed at replacing PUREX, more proliferation safe
  - no pure plutonium stream (Homogenous)

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

The GANEX Concept



Homogenous recycling



Several different GANEX processes are under development

The Chalmers GANEX Solvent

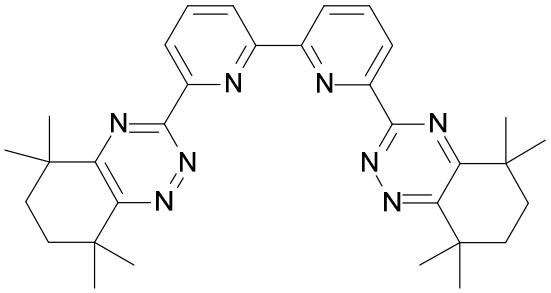
Combine two well known extractants with different properties:

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:

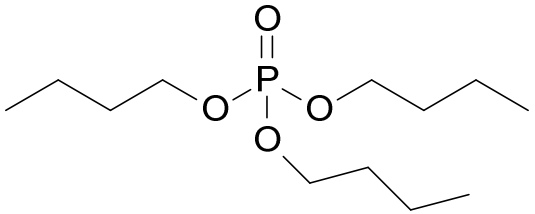
$CyMe_4$-BTBP



The Chalmers GANEX Solvent

2. *tri*-butyl phosphate or **TBP**

Known to: Extract tetra- and hexavalent actinides (PUREX process)

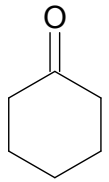


1+2 = > No need for redox control

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

The diluent used for this (so far) is cyclohexanone

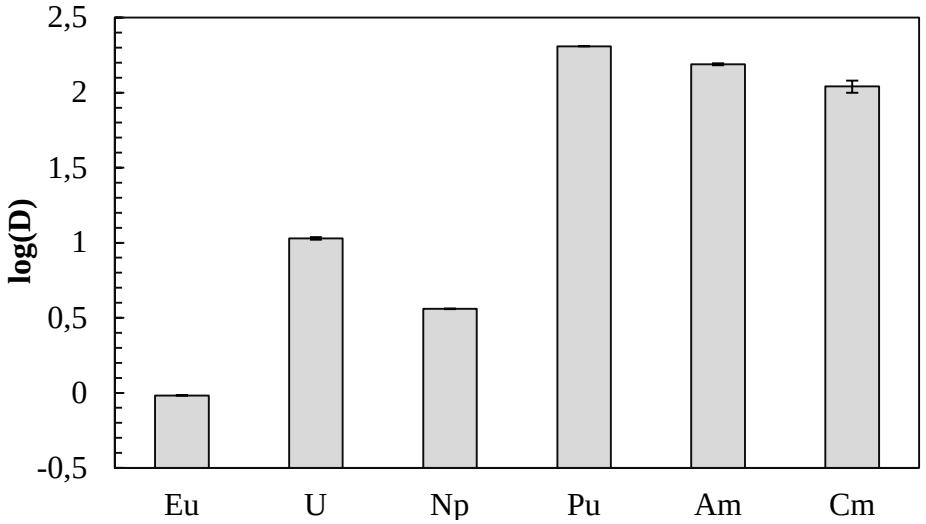
- solubility
- kinetics



The Development Process

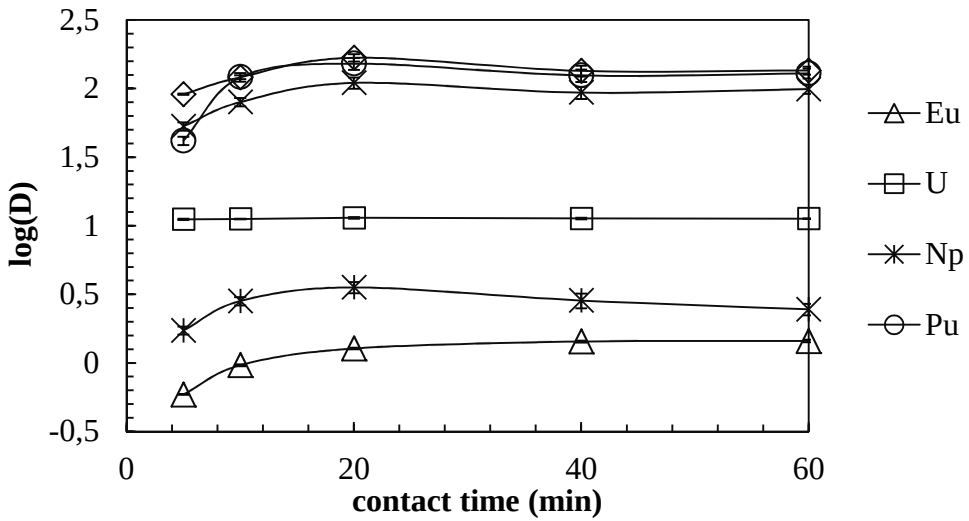
- Screening of the Extraction Behaviour
  - An extraction, kinetics, An/Ln separation
  - Extractant behaviour – e.g. synergistic/antagonistic effects
- Stability
  - Hydrolytic
  - Radiolytic
  - Temperature
- Fission and Corrosion Product Handling
  - Extractions
  - Action plan- solutions
- Towards Process Implementation
  - FP Loading
  - Batch process test
  - Continuous tests

Screening of the Extraction Behaviour

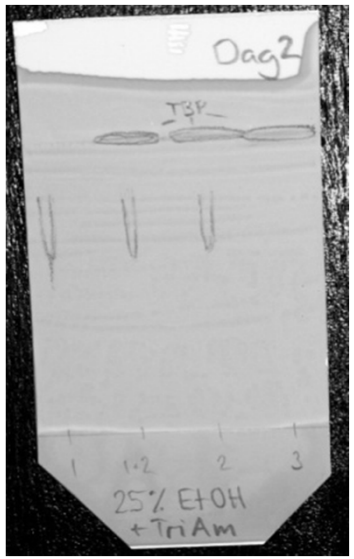


$SF_{Am/Eu}=160$

Fast kinetics (< 20 min)



Screening of the Extraction Behaviour

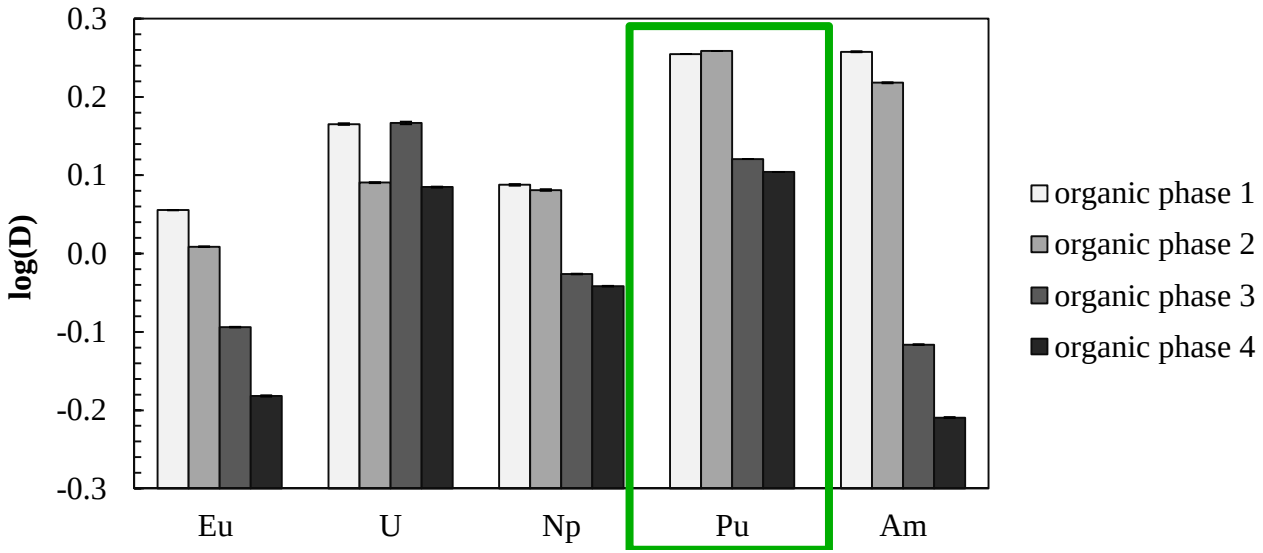


The extractants doesn't react with each other

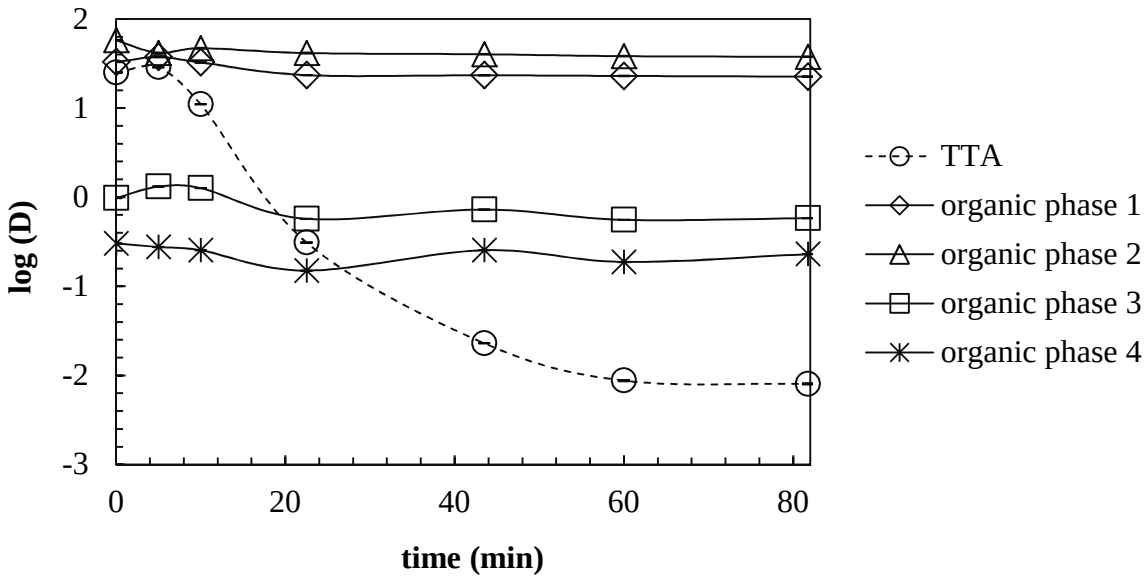
Neither affect the extraction behaviour of each other

1=BTBP+TBP
2=BTBP
3=TBP
4=cyclohexanone

1M HNO_3+3M $NaNO_3$



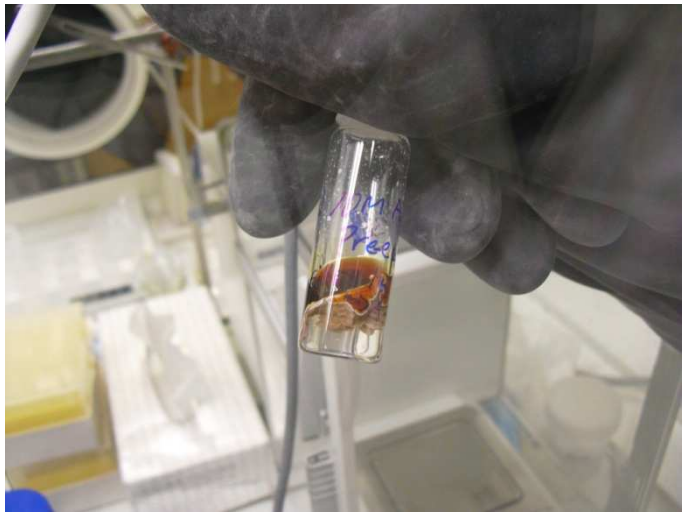
Screening of the Extraction Behaviour

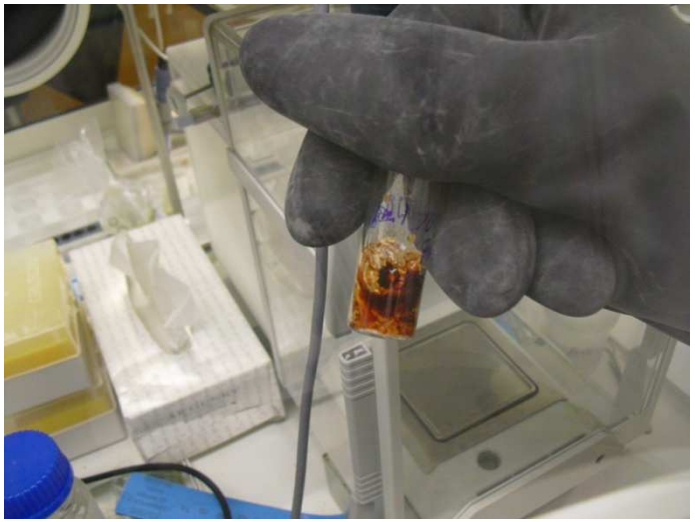


Plutonium extracted by BTBP in this solvent!
Important to consider in the context of Plutonium loading…

Pu loading

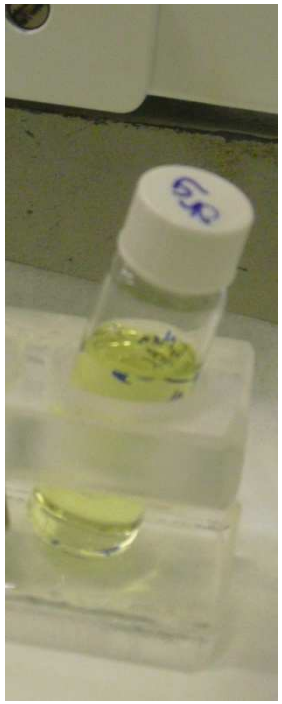
Original stock, 4.5 g Pu in HCL, add AgNO3, precipitation of AgCl

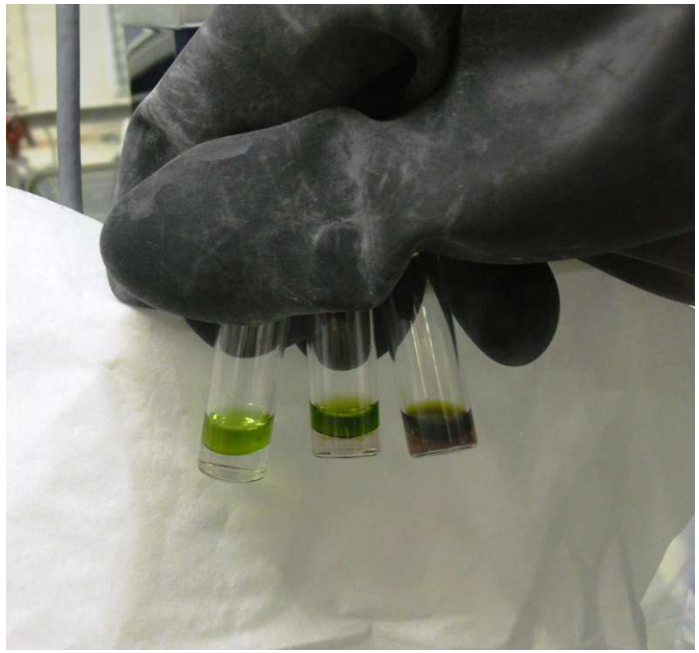




Pu loading

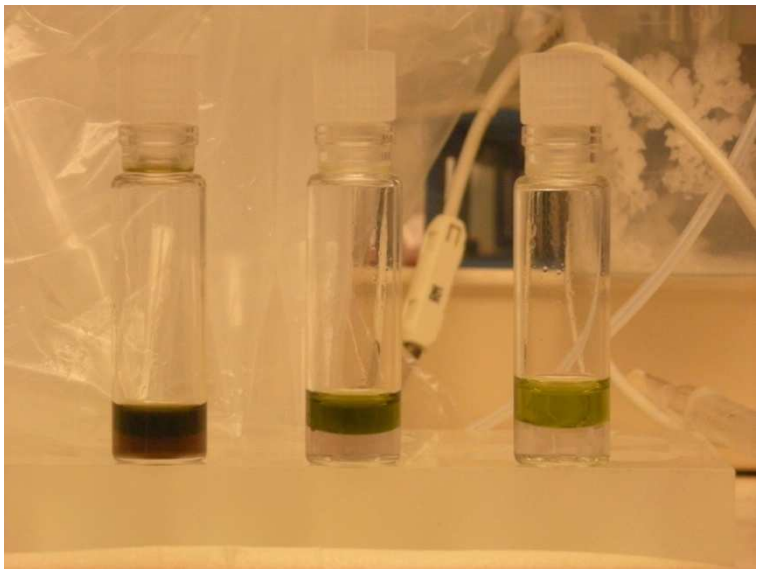
Three concentrations extracted (pure organic solvent to the left)





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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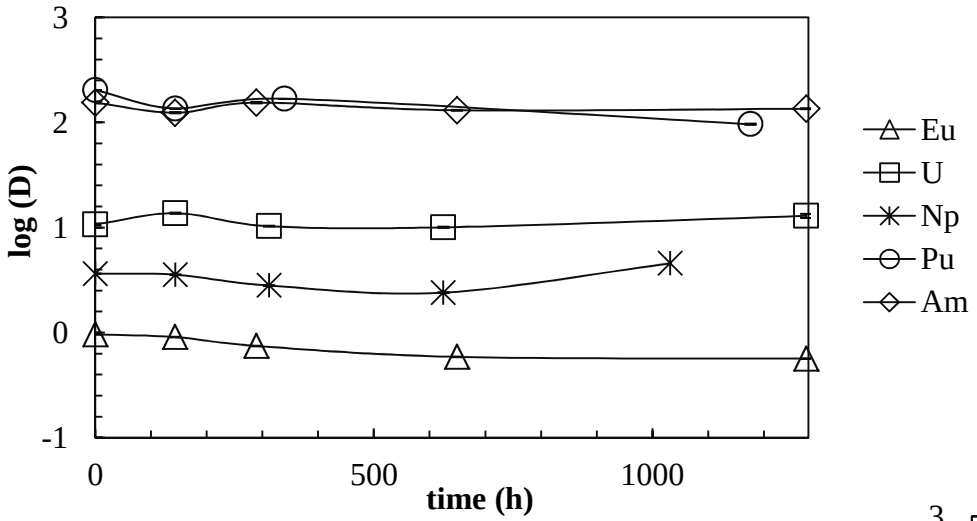
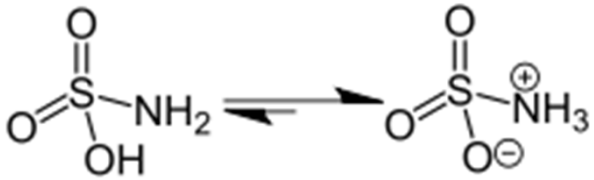
Stability

- Hydrolytic

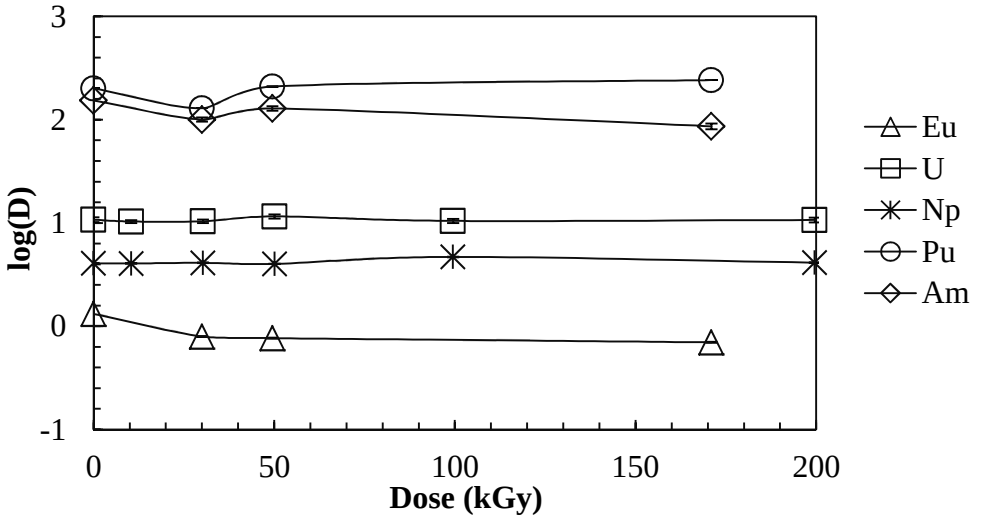


- Temperature



Stable after nitrous acid scavenger addition

- Radiolytic



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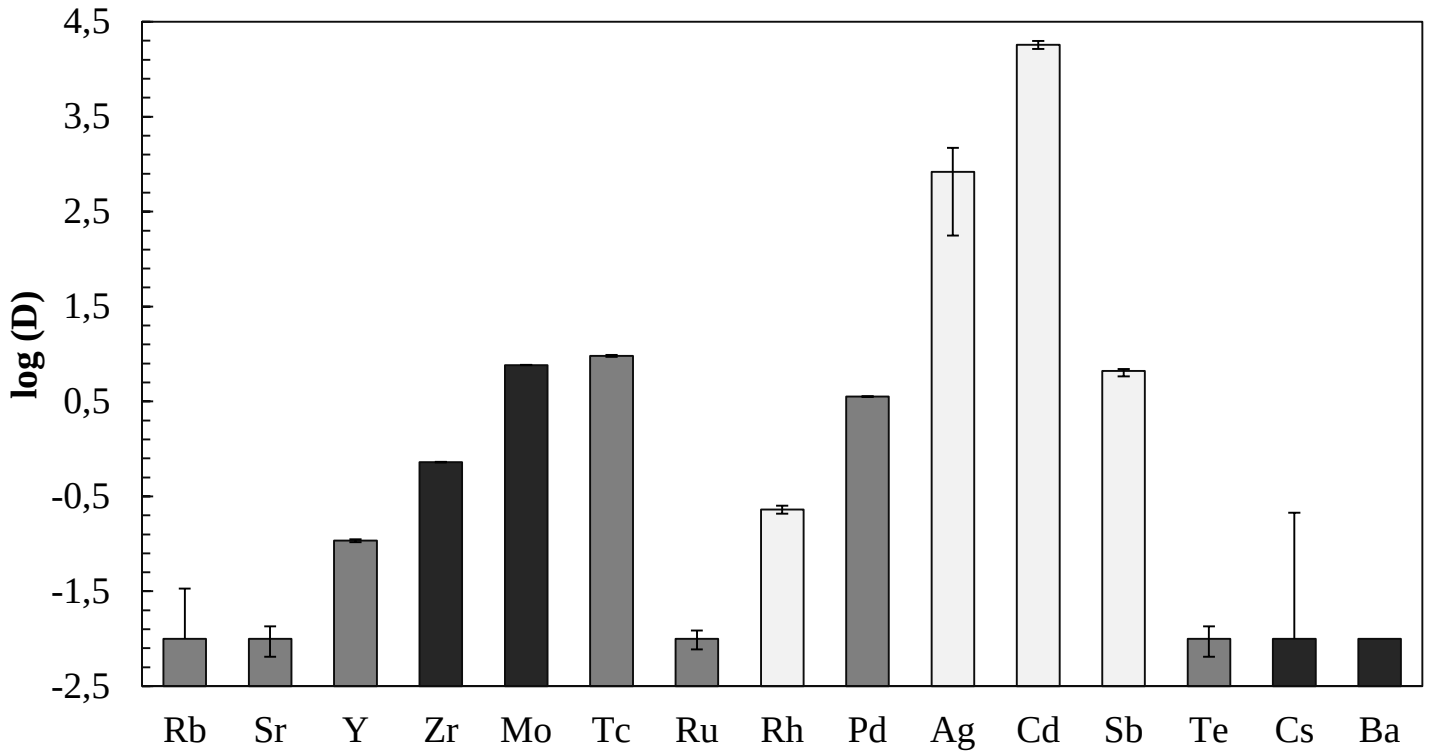
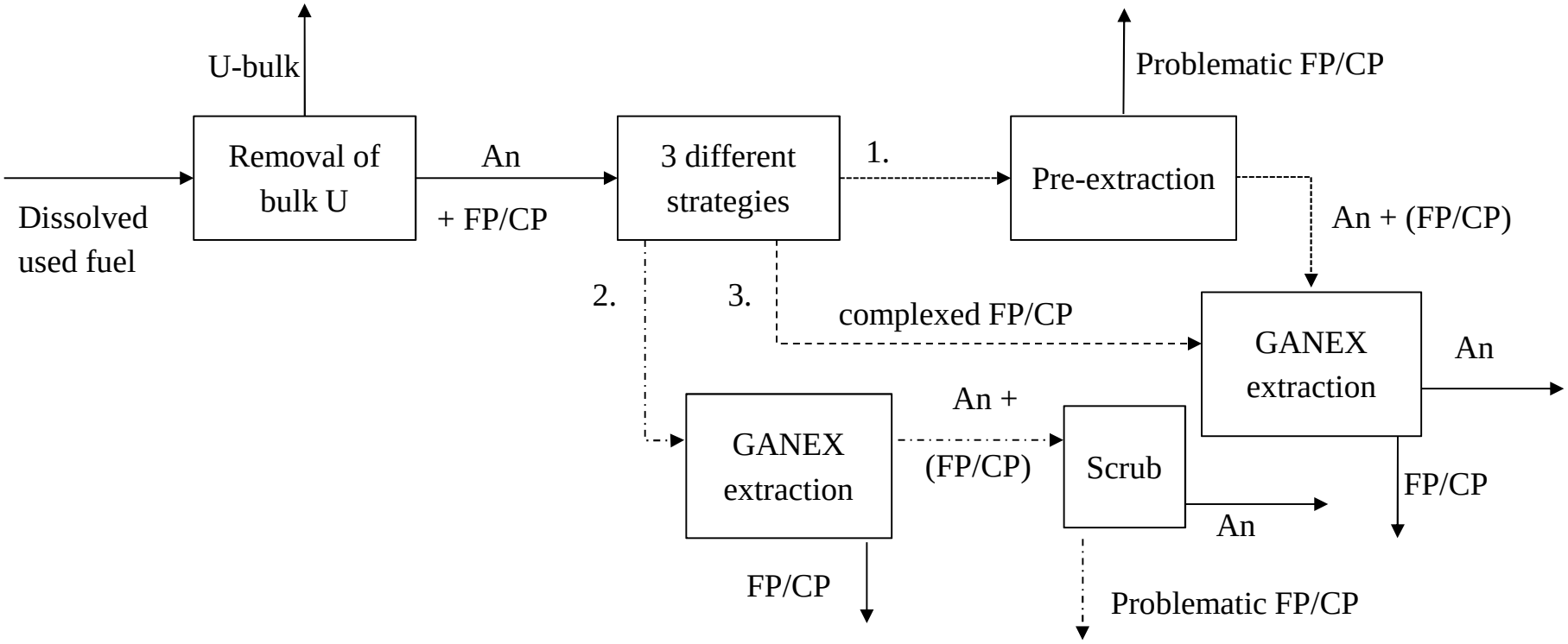
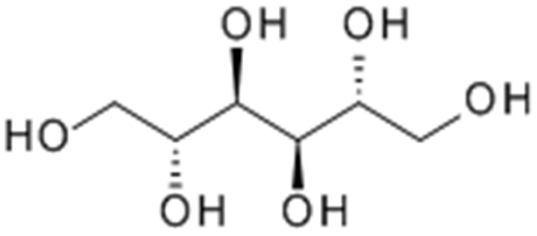
Fission and Corrosion Product Handling

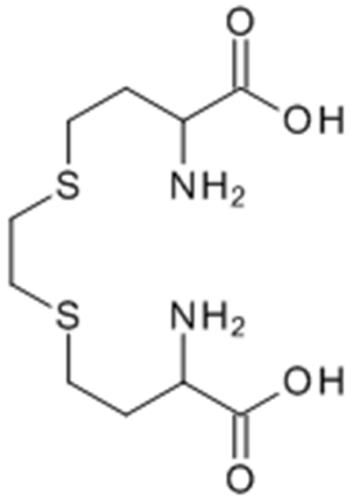


Fission and Corrosion Product Handling

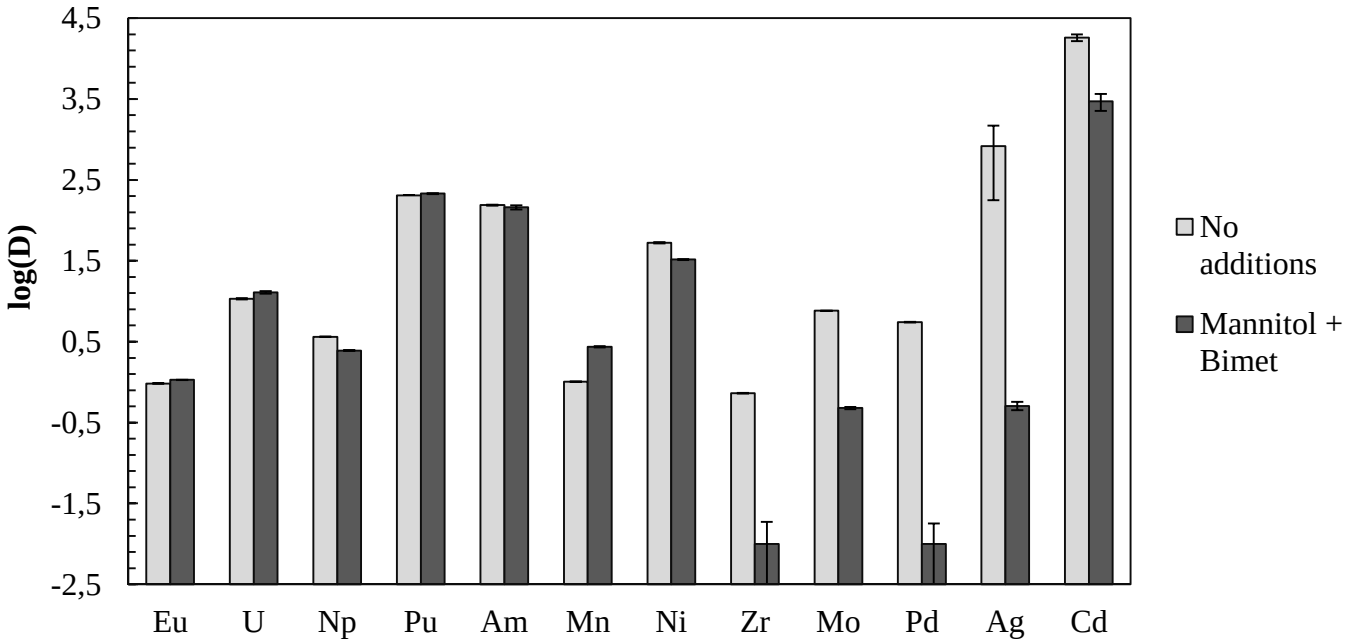


Fission and Corrosion Product Handling





Zr, Mo suppression



Pd, Ag, Zr suppression

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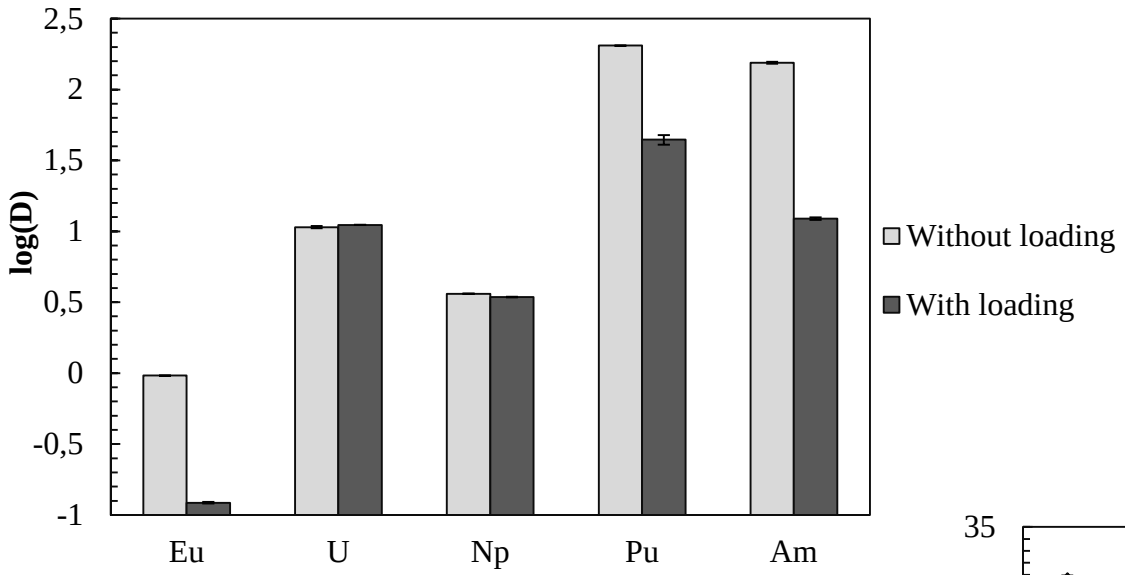
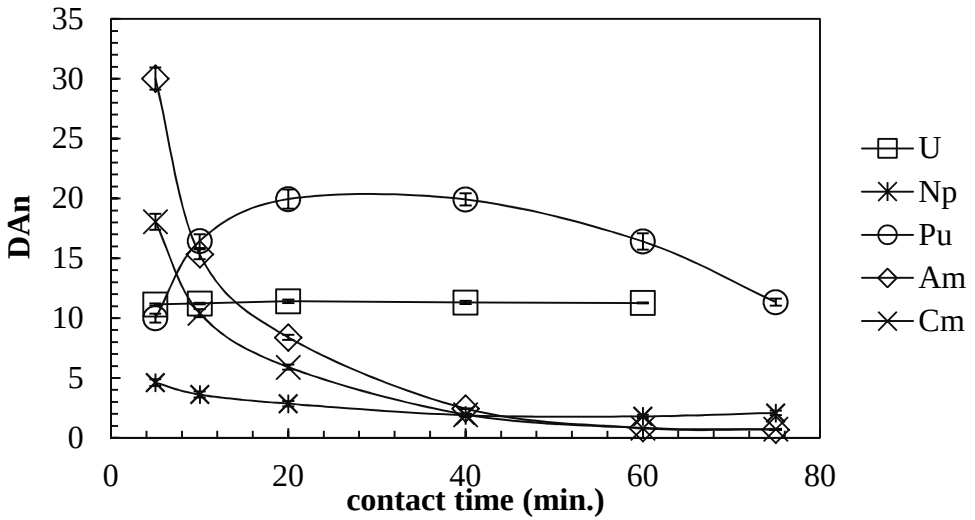
Towards Process Implementation



FP Loading at Chalmers = OK

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



Towards Process Implementation

Centrifugal contactor test

HAR diluted 1:10, increased aq phase density

Towards Process Implementation

Element	% of initial in aqueous strip
Eu	0.32
U	93.5
Np	99.8
Pu	99.6
Am	99.8

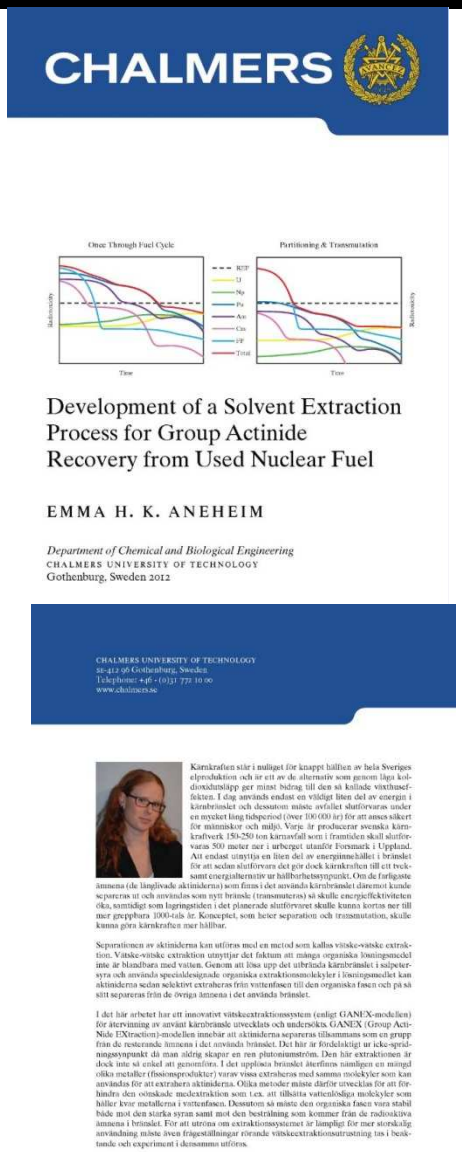
Centrifugal contactor data

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

Batch Process data

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

Element	% of initial extracted	% of initial in aqueous strip
Eu	11	9.1
U	100.0	95
Np	100.0	96
Pu	100.0	58
Am	100.0	25



ELECTRA-CHALMEX

- 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 andy separation process can be used practically
- Experiments are needed. Computer excercises are far from adequat 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