Development of advanced aqueous recycling options for future closed fuel cycles

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UK roadmap => 2050
- UK moves to open cycle >2018
- HLW/SF repository >2070
- New build 0-16 GWe

Why keep options open?
- Low carbon energy needs
- Expanded programme >>16 GWe
- Transition to fast reactors
- Pressures on storage & disposal
- International opportunities
- Next generation skills

=> Retain closed cycle option!
Why/why not recycle SF?

- Improved sustainability
- Reduced heat loading in GDF
- Reduced plutonium inventory
- Increased public acceptability
- Increased proliferation risks
- Increased volumes of secondary wastes
- Cost
- Environmental impacts
- Process safety

=> R&D to justify benefits & remove barriers => Recycle is viable option

"Optimise recycling processes so the option to close the fuel cycle by ~2050 is competitive with other spent fuel management options"

=> PROCESS SIMPLIFICATION / INTENSIFICATION / INNOVATION
Recycle technology development

- **2035 – 2060**
  - Near
  - Medium
  - Long

- **2010**
  - Evolutionary changes
- **2035 – 2060**
  - Radical improvements
- **2060 – 2080**
  - Transformative

**Countries:**

- **Korea**
  - Pyro-processing

**Timeline:**

- **2010**
- **2035 – 2060**
- **2110**

**Types of Changes:**

- **Evolutionary changes**
- **Radical improvements**
- **Transformative**
NNL reprocessing R&D Capability

- **Head end**
  - Dissolution
  - Oxidation

- **Solvent Extraction**
  - U, Pu
  - Np, Am, Cm

- **Finishing**
  - U
  - Pu, MA

- **Testing**
  - Lab scale
  - Pilot scale

- **Engineering**
  - Industrial experience

- **Modelling**
  - Simulation

- **Chemistry**
  - Experiments

- **Theory**

**NNL PuMA Laboratory, Sellafield**
SANEX SF Recycling: Processing schemes

Heterogeneous recycle LWR/MOX fuel

Dissolution

Advanced PUREX

SANEX

U oxide

(U, Pu, Np)O₂

(Am, Cm)O₂

GANEX-1

EURO-GANEX

(.TRU)O₂

U oxide

Homogeneous recycle MOX/FR fuel
Current focus areas

- Basic chemistry of separation processes
- Flowsheet development & testing
- Developing process models
- Underpinning process safety (malops, H2)
- Enhanced Dissolution processes (AgII, carbides)
- Co-conversion processes (oxalate)
Advanced PUREX process

- Single cycle
- Centrifugal contactors
- No separated Pu
- No U(IV)/hydrazine
- UOX & MOX reprocessing
- Np management
Complexant based separation

Some ideal properties

- Bidentate O,O donor ligands
- Selective for An(IV) ions
- Strong hydrophilic complexes
- Hydrolyse in acid solution
- Redox active
- Fast reductant for Np(VI)
- CHON – decomposable

Demonstration

- Flowsheets tested up to FR [Pu]
- Pu-active surrogate feeds
- High DFs (U stream) – up to 1.5E6
- Can leak δU with Pu
- Np routed with Pu
- Widespread international interest
Full Np extraction

- U/Np surrogate feed test (‘cold’ test)
- Adjust HNO3 and temperature only
- >99 % extraction of Np
- Centrifugal contactors used
- Process model developed (with UoM)
- Kinetics & mechanism modelled (with UoL)
- (Adapted by ORNL for $^{237}$Np/$^{238}$Pu separation)

$$NpO_2^+ + \frac{1}{2} NO_3^- + \frac{3}{2} H^+ \Leftrightarrow NpO_2^{2+} + \frac{1}{2} HNO_2 + \frac{1}{2} H_2O$$

Development of a process model

- gPROMS based model
- Based on open source – collaboration with UoM
- Includes mass transfer in centrifugal contactors
- Good agreement with flowsheet profiles
GANEX (grouped actinide extraction)

- Aimed at homogeneous recycle of actinides in FR fuel cycle
- 2 SX cycles
  - GANEX-1 = separation of bulk uranium
  - EURO-GANEX = co-separation of transuranic actinides
- From concept to hot test in 3 years (at ITU on legacy Dounreay fuels)
- Successful
- Leading alternative non-PUREX aqueous separation process

See later talk for more details!
Comparing processes

PUREX (Thorp)

1. Developed 1940-1980
2. TRL = 9
3. 3 cycles SX
4. Pure separate U & Pu products
5. Can not recover minor actinides (Am, Cm)
6. Large process footprint: pulsed columns & mixer-settlers
7. LWR fuels only
8. Uses tributyl phosphate

Advanced PUREX & GANEX

1. Developing now
2. TRL~4
3. 2 cycles SX
4. Pure U product & mixed TRU products
5. Recovers minor actinides (Am, Cm)
6. Compact, intense process: centrifugal contactors
7. MOX & FR fuels (high Pu content)
8. New extractants
TRU-SANEX flowsheet

- Variant to EURO-GANEX & i-SANEX (same chemistry)
- Tested by NNL
- Separate Np+Pu and Am+Cm products
Conclusions & outlook

• Closed fuel cycles may be needed 2030-2070
• New separation processes will be required
• Range of options needed
• Aqueous recycle can meet future fuel cycle demands
• NNL collaborates nationally & internationally
• Integrates chemistry, modelling, safety & engineering
• UK/European focus on:
  • Advanced PUREX-SANEX route
  • GANEX route
• TRU-SANEX process could bridge these routes
• Starting to integrate across the cycle
Selective Actinide Extraction (SANEX)

- European Minor Actinide separations
- Am+Cm recovery from PUREX HLW stream
- Innovative-SANEX developed by Julich
- Successful spiked test in centrifugal contactors
- High recoveries & DFs
- Uses new reagents

SACSESS
- NNL: developing a process model
- NNL: process safety studies & optimisation
**Reprocessing R&D**

**Head end**
- Prepares fuels
- Must provide homogeneous solution in HNO3 for SX
- High Pu fuels challenging
- Non-oxide fuel matrices managed in HE
- Volatile FPs routed to DOG

**Chemical Separation**
- Uses SX
- Advanced PUREX – option for LWR and FR reprocessing
- Minor actinide separations require new SX processes
- Grouped actinide extraction option for FR recycle
- Solvent recycle & treatment

**Conversion (Finishing) processes**
- Convert nitrate to oxide products ready for fuel fab
- Trend towards co-conversion (U,Pu) or (U,Pu,MA)
- TRU finishing is key area for R&D
- Oxalate / MDD / sol-gel are options