Study of reaction kinetics in continuous solvent extraction processes for actinide separations

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Methods & Equipment

Solvent extraction equipment: modified lab-scale short residence time annular centrifugal contactors (ACC)

- PMMA translucent surrounding of annular mixing zone
  - Adaption of annular gap size → mixing zone volume
- Increase rotor length → higher residence time in specific parts of the process
- 3D printing of stator to increase overall performance
- Use of high-speed camera to visualize dispersion in the mixing zone & determine specific surface area for kinetic studies

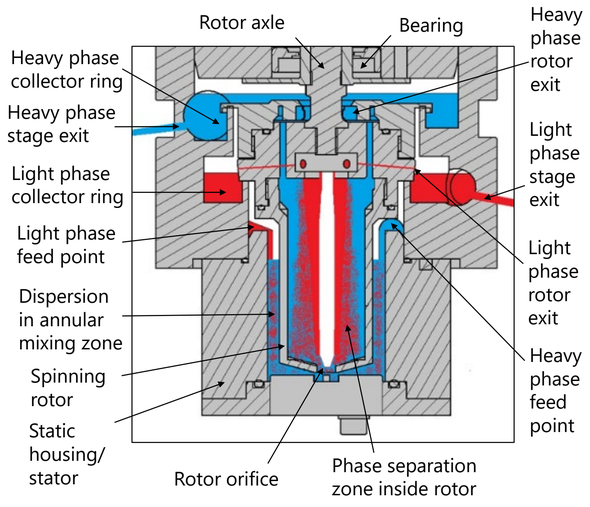

Figure 1: Cross section of ACC rotor & casing

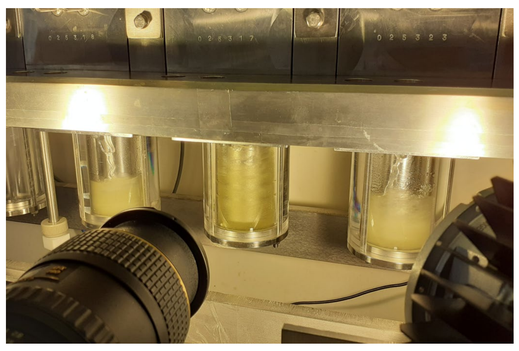

Figure 2: High-speed camera setup

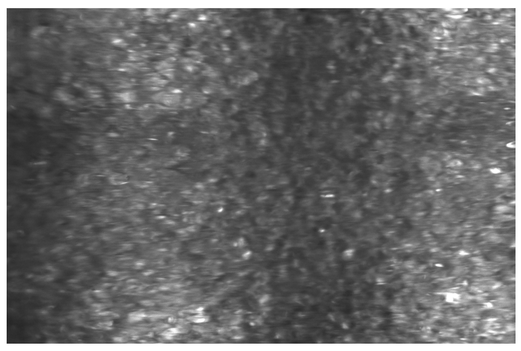

Figure 3: Dispersion at 6250 fps

Introduction

Scope of PhD is to investigate innovative multi-stage solvent extraction processes for separation of actinides

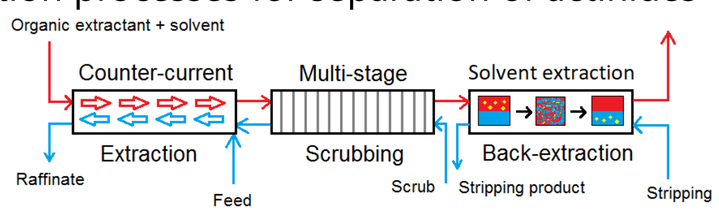

Figure 4: Principle of counter-current multi-stage solvent extraction

- CHON principle → increase sustainability of (future) nuclear fuel cycles by decreasing secondary waste streams
- Separation & purification at short residence times: throughput ↑ & footprint of process equipment ↓
- N,N-dialkylamides D2EHBA & D2EHiBA investigated as alternative for TBP for U extraction
- TBP process used as benchmark

Objectives

- Provide CHON alternative for PUREX at short residence times
- Optimize ACCs

Results

In-house lab-scale ACC redesign

- 3D printing of stator in polyamide for increased flexibility & lowering production costs
- Manufacturing tolerances to be compared with CNC machining → adapt design towards 3D printing
- Chemical compatibility limited in high HNO_3 concentration – test 3D printed stainless steel

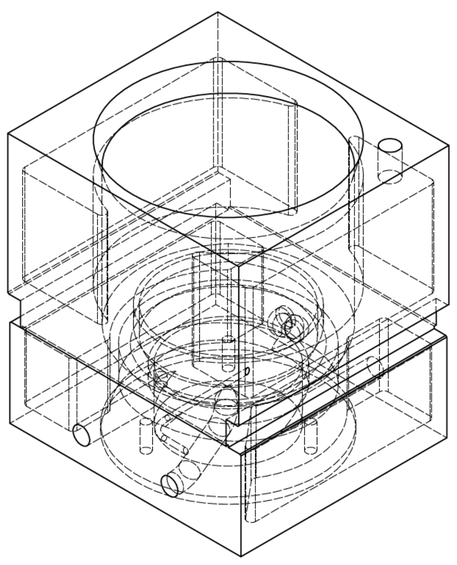

Figure 5: 3D drawing of improved stator

Figure 6: 3D printed stator test piece

Multi-stage experiments

- Repeatability of multi-stage experiments has been demonstrated with TBP and D2EHiBA
- Benchmark experiment with TBP performed

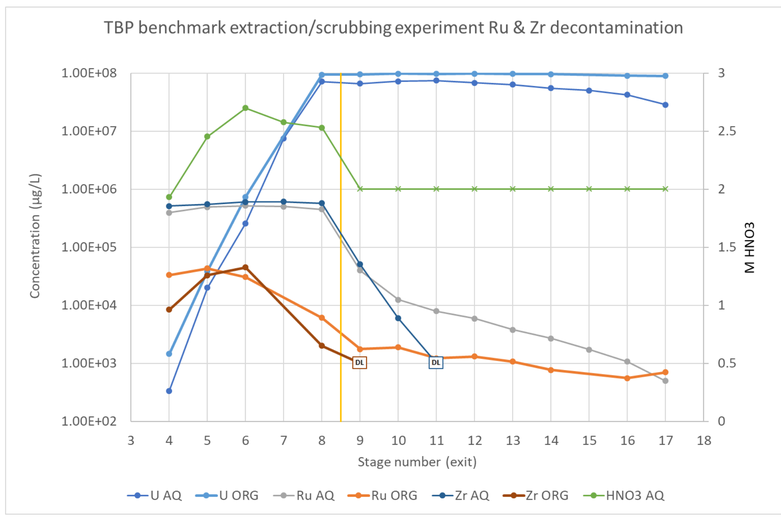

Figure 7: TBP benchmark experiment for U extraction and Zr/Ru decontamination

Process modelling & batch experiments

- Model of TBP in SX Process simulation software → Description of D_U in function of $[U]_{aq}$ and $[HNO_3]_{aq}$ by Langmuir second-order non-linear model
- Distribution isotherms being created for D2EHBA & D2EHiBA → to be implemented in SX Process model

Discussion

Main R&D challenges

- Visualizing and measurements of mixing zone with high light intensity → obtain sufficient focus and contrast
- Measurement of trace quantities (<1 mg/L) of chemical impurities in high U concentrations (50-100 g/L) → Improve DL

Innovative aspects

- D2EHiBA based process already successfully performed at unprecedented short residence times in ACCs
- Exploration of 3D printing possibilities for lab-scale ACCs

Future work

- Hydrodynamic testing of 3D printed stator
- Complete D2EHBA & D2EHiBA extraction isotherms on different $[HNO_3]_{aq}$ & integrate in SX Process model

  → Perform 1st D2EHBA based process in ACCs
- Working with Np, Pu & spent fuel spikes in a newly designed glove box installation

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