ACSEPT
PROGRESSES IN ADVANCED PARTITIONING
AND CHALLENGES FOR THE FUTURE

Actinide reCycling by SEParation and Transmutation
FP7 EURATOM CP 2007-211267 - (March 2008 – February 2012)

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ACSEPT at a glance

34 partners from National Nuclear Research laboratories, Nuclear Industrial Companies, Universities and/or National Fundamental Research Laboratories, European Nuclear Research Laboratory, Small and Medium Size Enterprises.

12 European countries + Japan & Australia

4 years (2008-2012)

130 men.years

Total budget 24M€

EC Grant 9M€

The biggest FP7 Euratom project

Links with Korea in Pyro (via KAERI)

Links with Russia in Pyro (via ISTC, RIAR)

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Assessment of reprocessing flowsheets for different fuel cycle scenarios with a view to their future demonstration at the pilot level.
Hot Facilities in ACSEPT

ATALANTE

+ FZJ, CIEMAT, CRIEPI, NRI

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The process development iceberg…

- Hot tests
- Flow sheeting
- Process modelling
- Coordination chemistry
- Extraction Performance Tests
- Analysis
- Organic synthesis
- Computational chemistry
- Radio chemistry
- …
Progres in Hydrometallurgy
Dissolution studies

- (U,Pu)O₂ and PuO₂ dissolution studies
  - In warm nitric acid - effect of high and low HNO₂
  - In nitric acid with Ce(IV)/Ag(II) with simulated fission products
- Surface studies of UOₓ
- U(VI), Pu(IV,VI) – nitrate complexation studies under dissolver conditions

Heterogeneous recycling

"regular" SANEX (on a DIAMEX raffinate), selective actinide extraction

Two SANEX processes based on BTBP/DMDOHEMA and BTBP/TODGA systems were developed within EUROPART and successfully tested.

Flow-sheet:
- Low flow-rates due to slow kinetics
- High feed acidity (2 M) for a more efficient extraction

Results:
- >99.9% of the An in the product
- Ln remained in the raffinate
- Organic phase clean

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• “innovative” (stripping) SANEX (on a PUREX raffinate), extraction of An, Ln and selective stripping of An.

One process based on a TODGA system was developed and successfully tested.

- Extraction – FP scrubbing: 16 mixer-settlers
- Ln scrubbing: 12 M.S.
- An Stripping: 16 M.S.
- Ln/Y stripping: 8 M.S.
- 9 rotating piston pumps coupled with flowmeters
- 7 on-line spectrophotometric measurements for Am – Nd concentrations
- 2 in-line pH controls

Heterogeneous recycling

D. Warin’s lecture at 5:00 PM

Improvements under studies at FZJ and KIT

A. Wliiden’s lecture at 5:30 PM

A. Geist’s poster IV-1

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GANEX, grouped actinide extraction

Two cycles:
1st cycle: quantitative Uranium extraction,
2nd cycle: grouped actinide extraction

Only the second cycle is under studies within ACSEPT

An alternative to the CEA GANEX (D. Warin’s lecture at 5:00 PM) based on NEWPART-PARTNEW-EUROPART Systems
Homogenous recycling - CHALMERS

A TBP/BTBP-based GANEX Separation Process

Figure 5. Extraction of actinides (\(^{239}\text{U}, ^{237}\text{Np}, ^{239+241}\text{Pu}\)) from Am, Pu, U, Np with 0.01 M Cymethyl-BTBP and 30% TBP in cyclohexanone.

Figure 6. Extraction of lanthanides (\(^{152}\text{Eu}, \text{the rest as non-radioactive metal salts}\)) from 4 M nitric acid with 0.01 M Cymethyl-BTBP and 30% TBP in cyclohexanone.
A TODGA/DMDOHEMA-based GANEX Separation Process

Solvent feed
TODGA
DMDOHEMA
Kerosene

Feed to GANEX Cycle

TRU, Ln co-extract
FP scrub

Pu, Np strip

MA strip

Strip (Np+Pu)
HNO3
AHA

Strip (MA)
HNO3

HAR

Pu, Np, Am, Cm

Ln/FP strip

Residual FP

Solvent recycle

For more information, see R. Taylor at al., Pu Futures 2010

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Homogenous recycling – Next Step

Select (at least) one concept for hot-test demonstration by the end of ACSEPT
Design the flowsheet
Implement the Hot-Test (ITU?)

Challenge
HYDRO - Fuel refabrication

From polyactinide containing solutions to polyactinide containing solids
Basic studies and process orientated studies

Sol gel routes (in Continuation of FP6 EUROPART project)
•External gelation: some tests, not concluding
  •Difficulties on Sol-creation (corrosion of stirrer)
  •Kernels stick at phase interface in gelation column
  •Collapsing of many Kernels during drying at RT
  •Sintering-tests not very promising
•Internal gelation: more efficient, promising results, even if still a lot of work to improve the technique

Alternative routes
•Direct thermal denitration/ solid extractants
Progress in Pyrometallurgy
Direct electrochemical reduction of oxides in molten fluorides

Direct electrochemical reduction of oxides (SnO₂, TiO₂, Fe₃O₄ and UO₂) have been tested in molten fluorides: LiF-LiF-NaF and/or LiF-CaF₂ at 750-850°C.

Thermal treatment

Analytical performance of the Heated Laser Ablation Cell in combination with a mass spectrometric detector was tested. Released of Cs measured with IPC-MS from CsNO₃ (sol.) and CsCl/NaCl powder allows the determination of the detection limit for this element.
Electrorefining of actinides onto solid aluminium cathode in molten chloride salts

Conversion of fuel to metals

Metallic spent fuel: An + FP

Nuclear reactor

Electrorefining in molten LiCl-KCl on Al cathode

LiCl-KCl + FP

Cleaning of salt from FP

Exhaustive electrolysis

LiCl-KCl + AnCl₃ + FP

Cathodes processing: Chlorination

An-Al alloy

An alloy

Al recovery

An - actinides, FP - fission products, Al - aluminium

Processes under development in ITU

C. Nourry’s poster IV-12

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Liquid-liquid reductive extraction in molten fluoride salts/liquid aluminium

**Diagram**

1. **Fluorination** at 450-500°C
2. **Digestion** at 830°C
3. **Extraction** at 830°C
4. **Distillation**
5. **Back Extraction** at 700°C
6. **Chloride salt**
7. **Oxidizing agent**
8. **Gas**
9. **Precipitation**
10. **AnO₂**

**Materials**
- Cs, Rb
- ZrF₄
- LIF·AlF₃ (15-35%)
- Zn
- Al

**Processes**
- Thermal treatment at 1100°C
- Metallic waste
- Platinumides

**Concepts**
- CEA Work
- EURATOM

**Event**
- 11th IEMPT, San Francisco, 1-4 November 2010

**Author**
- S. Bourg
TRU recovery from genuine HLLW prepared by SF dissolution

Concentrate
Denitration
Crucible

CRIEPI Work

Chlorination
Crucible in heating system (650°C)
Calcined (7.3g)

Extraction into Cd

An
Liquid Cd

Li

F\(^{n+}\)

A\(^{n+}\)

Actinide chemistry of reductive extraction
AnCl + Li (in Cd) \(\rightarrow\) LiCL + An(in Cd)

Li; reductant

520g of HLLW
U: 8400 \(\mu\)g/g
TRU: 800 \(\mu\)g/g
FP: 2000 \(\mu\)g/g

Recovery of each TRU, ca. 100%, was achieved in liquid Cd.

Through this experiment, distribution coefficients of U, Pu, Np, Am, Cm and lanthanides were acquired, which are well coincident with those thermodynamically measured.

K. Uozumi’s lecture, tomorrow, 9:30 AM

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Salt recycling - Waste conditioning

FP decontamination in LiCl-KCl molten salt
- Precipitation of FP under solid oxide
- Zeolite Ion-Exchange for Salt Clean-Up

FP decontamination in fluoride molten salt
- Distillation of molten fluoride
- Decontamination by oxide precipitation

Ceramic waste form for chloride salt

Metallic waste form for the conditioning of metallic FP

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Cross-cutting activities
“Brainstorming” Workshops

Two workshops organized in September 2008 during the Prague Meeting.
- Flowsheet calculation requirements
- Cross-Fertilization Seminar In Pyro-Chemistry

Two workshops organized in September 2009 during the Bologna Meeting.
- Requirements to implement a GANEX flowsheet
- Improvement of the electrorefining process flowsheet

-Two Workshops organized in September 2010 during the Petten Meeting
- Status on the GANEX Studies
- How to optimize the use of the organic synthesis manpower

Introductive or illustrative presentation(s) followed by open discussions and exchanges

Helps the Project Coordination Committee to take decisions and to reorient/refocus the research program
Engineering studies

Scale-up issues
- Development of a device for drop-size measurement in centrifugal extractor in order to prepare the scale-up of this device
- Corrosion studies in molten salts

On line monitoring issues in molten salts
- Development of the laser induced breakdown spectroscopy in molten fluoride
- Development of an electrochemical sensor in molten chloride
Definition and design of the MARIOS experiment, now implemented within the FP7 FAIRFUELS Project

Actinide burn-up for \((U_{0.92}(Am,Cm)_{0.08})O_x\)

Production of an orientation document on assessment and ranking of inert matrices in term of reprocessing capabilities and waste management.

- Towards an experimental program?
Expected outcome of ACSEPT in 2012

Exploratory Research

Concepts selected and validated

Demonstration experiments

Simplification « Consolidation »

Towards potential industrialization

Several hundreds of molecules assessed

Scale 1/10000

Scale 1/1000

integration, long-lasting performance representativeness,

here In pyro

here In hydro

The next step?

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Education and training
Fostering visibility/dissemination

on the 2nd year of the project:

Attending International Conferences
around 40 oral contributions

Publishing in journals
more than 10 papers (in addition to proceedings)

Two patents
Investing in people

Attribution of ACSEPT Post-doctoral grants (2 up to now)
(50k€ grant)

Funding mobility of students between Partners (7 up to now)
1 to 3 month periods
(2000€/month)
Dissemination of knowledge

• Promote the participation of students to seminars, scientific workshops or summer schools (contribution to travel and accommodation costs)

• Invite lecturers during ACSEPT meetings.

• Organise specific scientific workshops

Solvent extraction (Jan Olov Liljenzin)
Radiolysis (Bruce Mincher, Steve Mezyk)
Actinide materials (Joe Sommers)
Analysis (Melissa Denecke)
Computational chemistry (Bernd Schimmelpfennig, Enrique Sanchez Marcos)
1st AIWO, March 2010 Lisbon

More than 120 participants
44 oral communications, among them:
12 invited lectures (international experts)
18 contributions from ACSEPT Young Scientists
7 contributions in the ISTC-ACSEPT pyro session
All the sessions chaired by the young generation

All the presentations on our website!

Organized in cooperation with the International Atomic Energy Agency
ACTINET-I3, a tool for networking in actinide sciences

www.actinet-i3.eu

At this occasion:

1st exchanges with J-Actinet

A panel discussion on hot facilities and experimental needs in Europe
Thank you for your attention