Development of the UREX+3 Flowsheet—An Advanced Separations Process for Spent Fuel Processing

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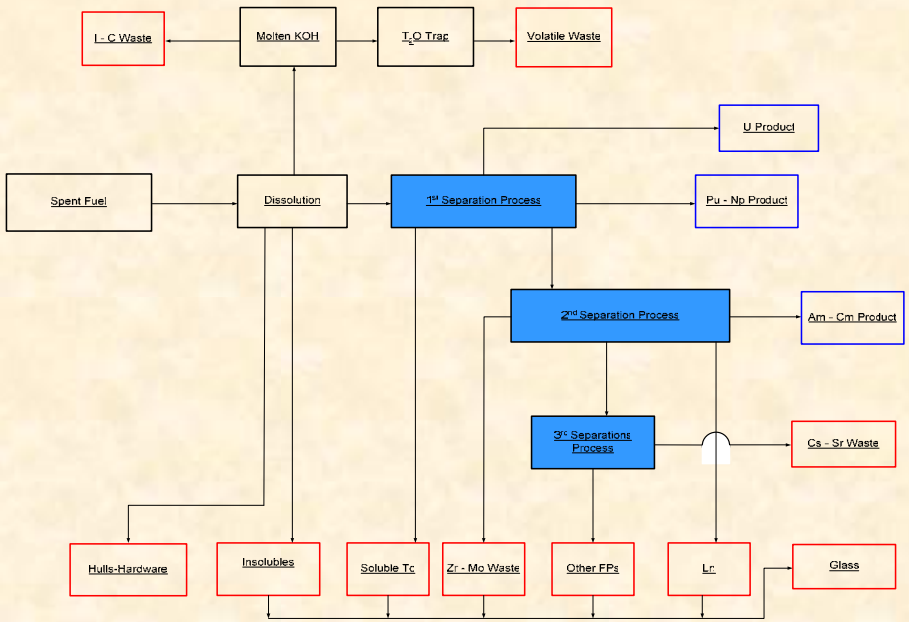
Suite of UREX+ Processes

Process	Prod #1	Prod #2	Prod #3	Prod #4	Prod #5	Prod #6	Prod #7
UREX+1	U	Tc	Cs/Sr	TRU+Ln	FP		
UREX+1a	U	Tc	Cs/Sr	TRU	All FP		
UREX+2	U	Tc	Cs/Sr	Pu+Np	Am+Cm+Ln	FP	
UREX+3	U	Tc	Cs/Sr	Pu+Np	Am+Cm	All FP	
UREX+4	U	Tc	Cs/Sr	Pu+Np	Am	Cm	All FP

Notes: (1) In all cases, iodine is removed as an off-gas from the dissolution process.
(2) Processes are designed for the generation of no liquid high-level wastes.

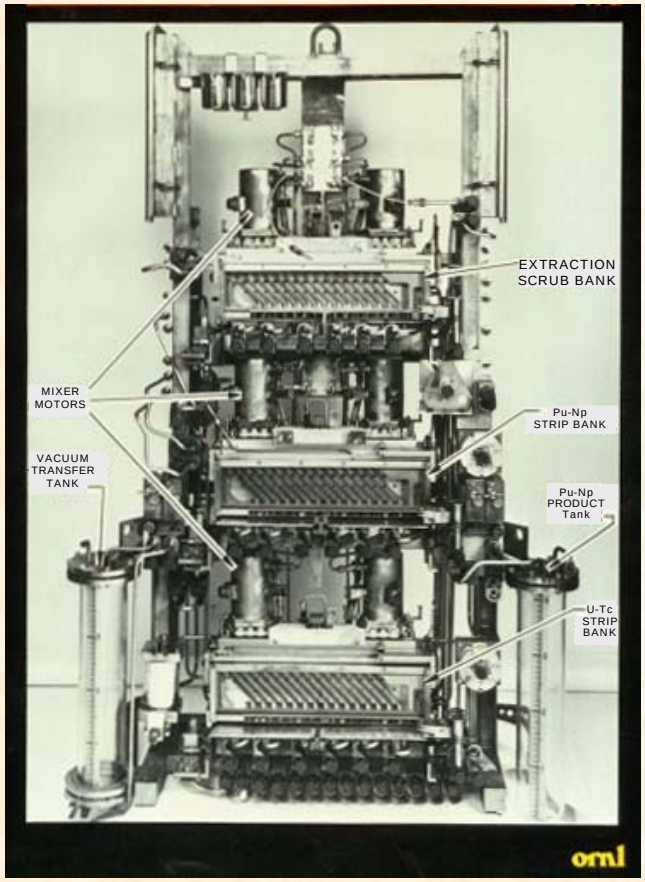
U: uranium (removed in order to reduce the mass and volume of high-level waste)
Tc: technetium (long-lived fission product, prime contributor to long-term dose at Yucca Mountain)
Cs/Sr: cesium and strontium (primary short-term heat generators; repository impact)
TRU: transuranic elements (Pu: plutonium, Np: neptunium, Am: americium, Cm: curium)
Ln: lanthanide (rare earth) fission products
FP: fission products other than cesium, strontium, technetium, iodine, and the lanthanides

Development of the UREX+3 Flowsheet



- **The UREX+3 flowsheet consists of three primary separations processes**
- **The first is a modification of the industrially used continuous solvent extraction process using tributyl phosphate**
  - **Largest-mass component, uranium, is removed**
  - **Plutonium and neptunium (possibly with some uranium) are collected together**
- **The second process, Reverse TALSPEAK, is a continuous solvent extraction, using di-2-ethyl-hexyl phosphoric acid, to separate and recover the minor actinides, americium and curium**
- **The third process, also a solvent extraction, is designed to partition the fission product wastes to separate the high-heat generators, cesium and strontium, for managed storage**

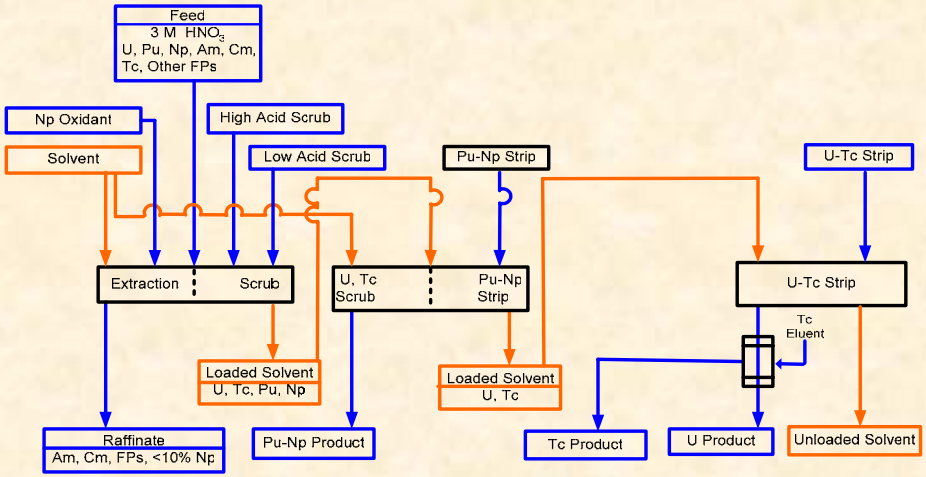
Continuous Solvent Extraction Test Facility at REDC



SETF Mixer-Settler

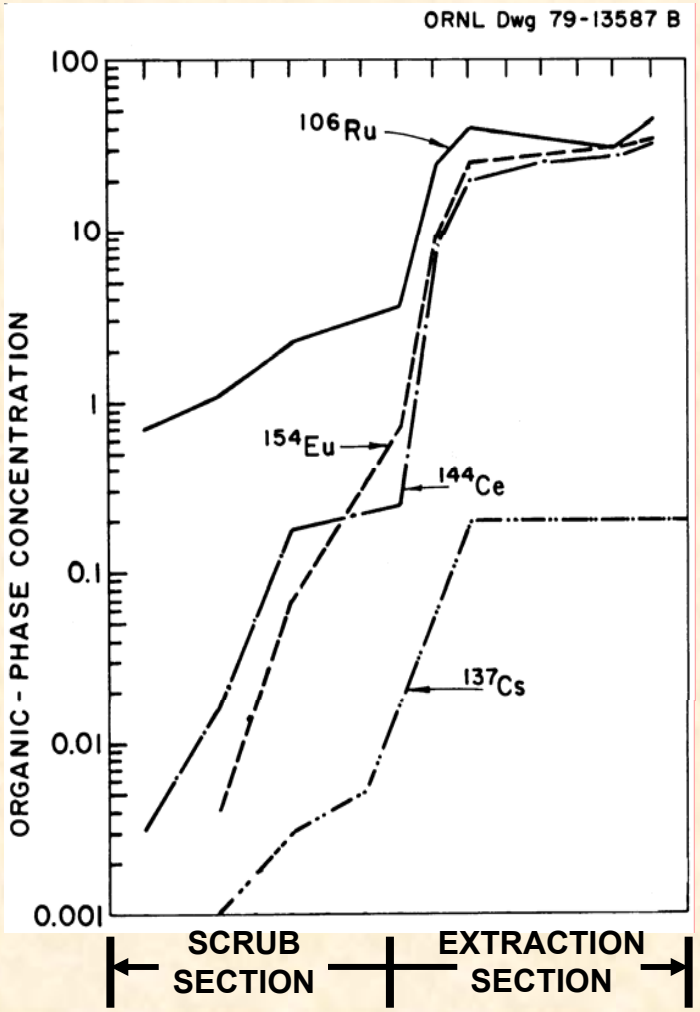
- **Spent fuel dissolver with perforated basket for leaching sheared hulls—2-kg HM capacity**
- **Feed adjustment evaporator and clarification by high-efficiency filtration**
- **Continuous countercurrent solvent extraction in three 16-stage mixer settlers**
- **Reagent tanks and flow controllers**
- **Effluent collection tanks**

Demonstration of the UREX+ Co-Decontamination Flowsheet



- **This process was successfully demonstrated in the Solvent Extraction Test Facility during 2003**
- **Uranium, plutonium, neptunium, and technetium are co-extracted using tributyl phosphate**
- **Americium, curium, and most fission products are not extracted and become feed to the second separations process**
- **Uranium (the largest-mass component) and technetium (the smallest) are co-stripped, and the technetium is recovered by means of anion exchange**

Typical Fission Product Concentration Profiles in the Extraction/Scrub Bank



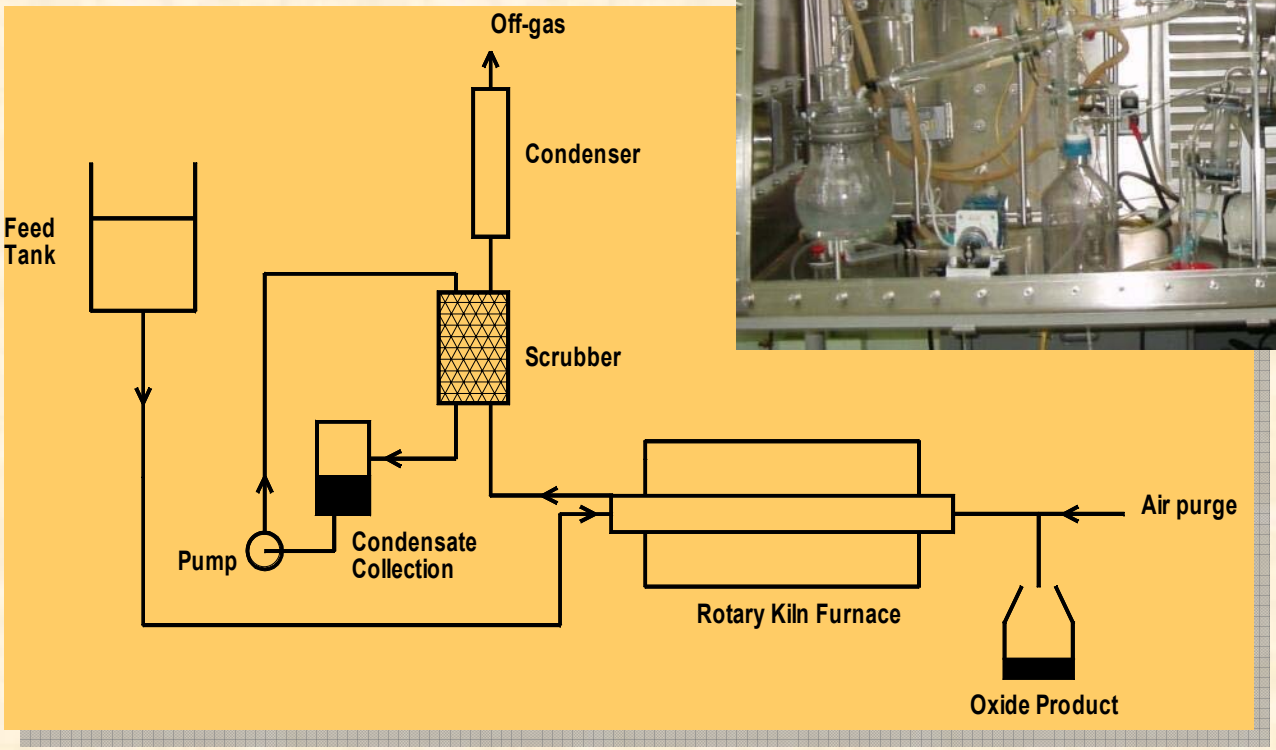
Gamma–Emitting Components in Typical Pu-Np Product Solutions

Component	Activity Level (μCi/g Pu)
^{106}Ru	141
^{110m}Ag	>0.3
^{125}Sb	>1.4
^{134}Cs	4.6
^{137}Cs	16
^{144}Cd	>14
^{154}Eu	>0.7
^{239}Np	1873

- The highest radioactivity in the plutonium-neptunium product for the first ~2 weeks is ^{239}Np
- The ^{239}Np is the decay daughter of ^{243}Am, which was separated during the process
- ^{239}Np emits a 280-keV gamma ray
- During aging of the plutonium-neptunium product, the ^{233}Pa decay daughter of ^{237}Np grows in
- ^{233}Pa emits a 310-keV gamma ray
- The Pu-Np product emits an easily detectable gamma signal, which can be used for monitoring

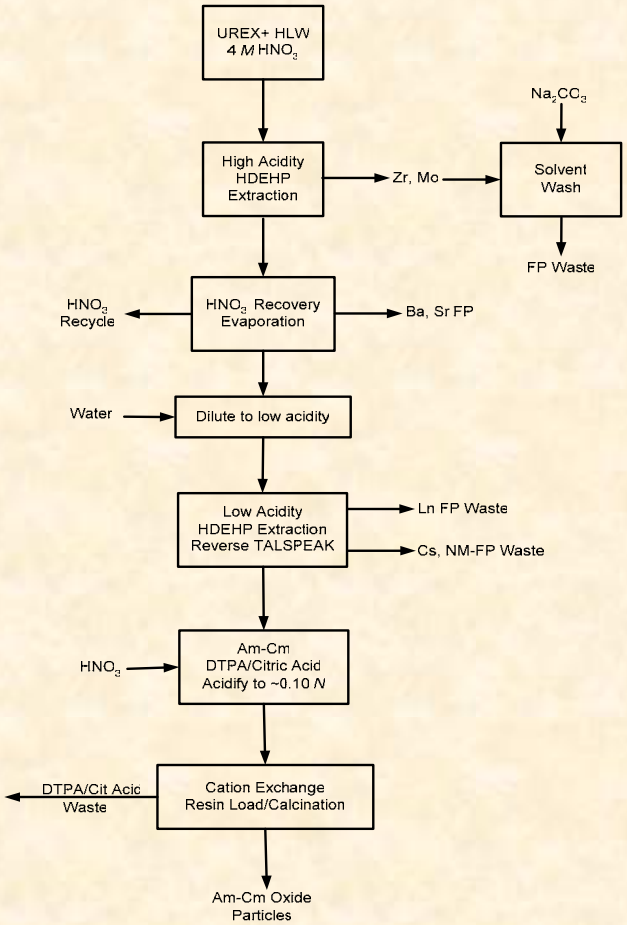
Modified Direct Denitration (MDD) Can Co-Convert Pu-Np to Oxide Powder

- **Heart of MDD process equipment is the rotary furnace**
- **Metal nitrate feed solution enters furnace and exits as converted oxide powder**



- **Condensate is used as scrubbing liquid for off-gas**
- **Air purge sweeps off-gas vapors from the furnace**

Current Reverse TALSPEAK Process Flowsheet



Current Reverse TALSPEAK Flowsheet

- **Intended to recover minor actinides, americium and curium, from fission products in UREX+ co-decontamination process raffinate stream**
- **Uses a single solvent system (HDEHP) in appropriate diluent**
- **Uses multiple extraction steps to avoid hydrolytic problems with fission products, zirconium and molybdenum**

Results from Batch Tests of Reverse TALSPEAK

	Distribution Coefficients					Separation Factors		
	Nd	Ce	^{154}Eu	^{241}Am	^{244}Cm	Nd/Am	Ce/Am	Eu/Am
Extraction								
Raffinate	8.0	3.7	150	4.4	6.0	1.8	0.70	34
Actinide Stripping								
1st DTPA	7.3	4.9	22	0.30	0.25	25	17	65
2nd DTPA	7.1	18	19	0.29	0.26	25	63	65
3rd DTPA	14.1	31	21	0.49	0.22	29	64	44
Lanthanide Stripping								
6 *M* HNO_3	0.01	0.01	0.03	0.09	0.04			

Development of Am-Cm Transmutation Targets Using Sphere-Pac Process

Conventional Fab. Fissile Spheres

Concentrated nitrate solution

↓

Gel-precipitation Co-conversion

↓

~98% dense oxide spheres

(300-1200 microns)

↓

$UO_3 \cdot 2H_2O$ → Sintered UO_2

Remote Fab. Minor Actinide Microspheres

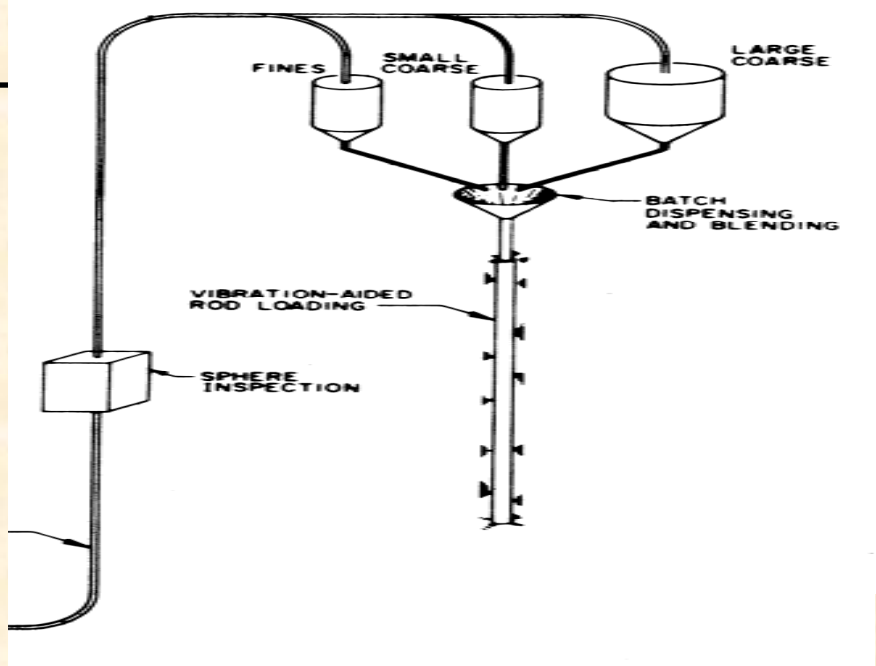
Dilute nitrate solution from UREX

↓

Resin loading Co-conversion

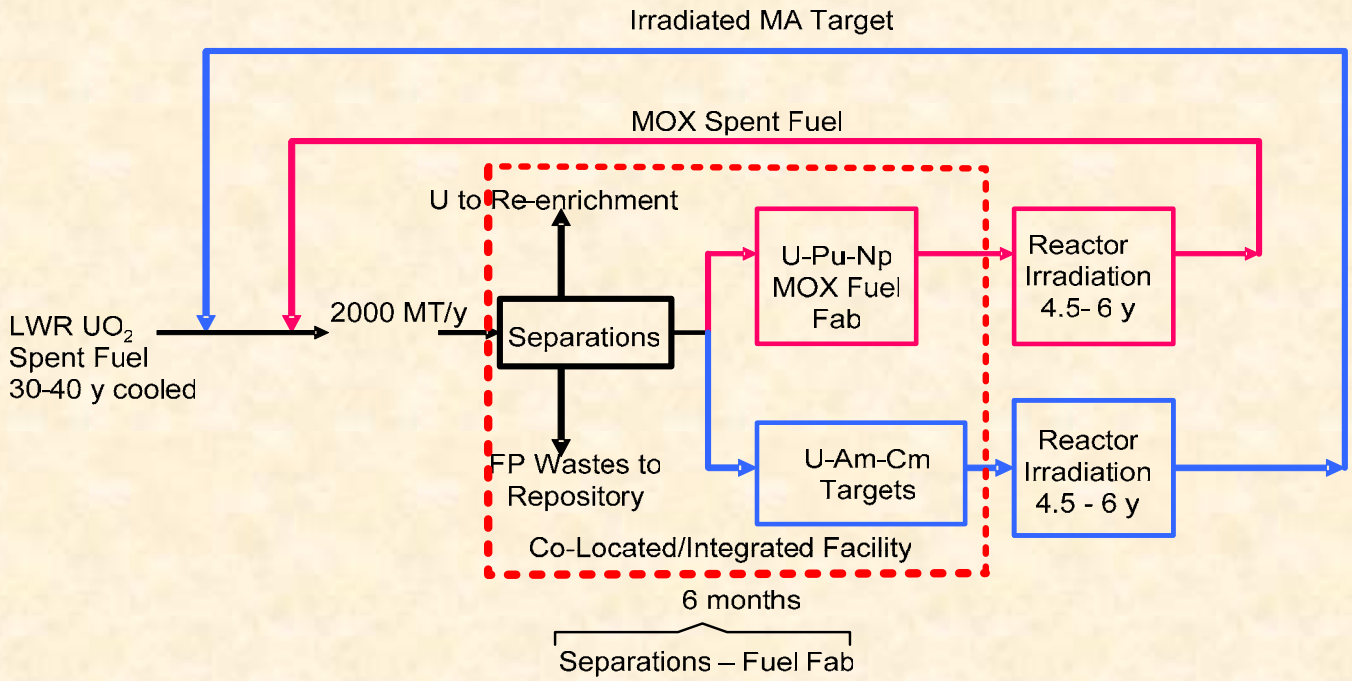
↓

~50% dense oxide spheres (30-80 microns)



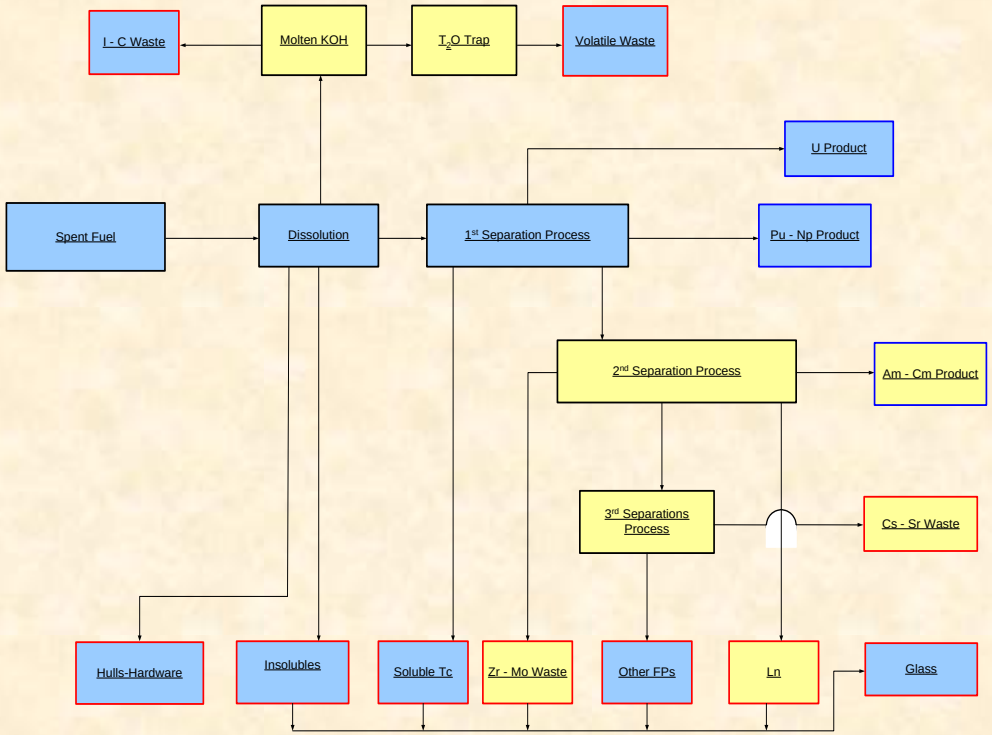
- **Process requires a minimum of mechanical operations**
- **Process is well-suited to remote fabrication**
- **Process produces a dustless fuel form**
- **Process is likely to accept the large helium release from ^{241}Am transmutation with less difficulty than pellet forms**

Use of UREX+3 in Partitioning-Transmutation Scenario



- **The UREX+3 flowsheet is an attractive candidate for deployment in the separations, or partitioning, phase of the overall partitioning-transmutation (P-T) process**
- **The P-T process maximizes lifetime of the HLW repository by minimizing input of heat-generating wastes**
- **The P-T process provides increased proliferation resistance by minimizing "attractiveness" and utilization of co-located, physically protected facilities**
- **The P-T process minimizes mass of wastes by recycling**

Summary—Status of Development of UREX+3



- **Head-end operations and 1st separation process are modifications of existing industrial-scale facilities**
  - **These could be used for an early start to spent fuel treatment**
- **AFCI projects are prioritized on the basis of these factors**
  - **Selection/optimization of Am-Cm recovery process**
  - **Fabrication of Am-Cm transmutation targets**
  - **Development of Cs-Sr waste form for managed storage**
  - **Development of advanced methods for process improvement, simplification, and cost reduction**