GNEP Spent Fuel Processing; Waste Streams and Disposition Options

Nuclear Waste Technical Review Board
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Argonne National Laboratory
National Technical Director for Separations Technology Development
Reprocessing Plants in Operation or Planned Today

<table>
<thead>
<tr>
<th>Country</th>
<th>Location</th>
<th>Capacity, t/y</th>
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<tbody>
<tr>
<td>China</td>
<td>Jiuquan (Planned, 2020-2025)</td>
<td>800</td>
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<tr>
<td></td>
<td></td>
<td>25</td>
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<tr>
<td>France</td>
<td>LaHague (UP2-800, UP-3)</td>
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<td>India</td>
<td>Trombay</td>
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<td></td>
<td>Tarapur</td>
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</tr>
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<td></td>
<td>Kalpakam</td>
<td>300</td>
</tr>
<tr>
<td>Japan</td>
<td>Tokai-mura</td>
<td>100</td>
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<tr>
<td></td>
<td>Rokkasho-mura</td>
<td>800</td>
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<tr>
<td>Russia</td>
<td>Chelyabinsk (Mayak, RT-1)</td>
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<tr>
<td></td>
<td>(Planned, 2025)</td>
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</tr>
<tr>
<td>United Kingdom</td>
<td>Sellafield B205</td>
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<tr>
<td></td>
<td>Sellafield THORP</td>
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<tr>
<td>United States</td>
<td>CFTC (Planned, 2020-2025)</td>
<td>2,500</td>
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</table>
Contemporary Plants use the PUREX Process

LWR Spent Fuel → Chopping and Nitric Acid Dissolution → PUREX Process

Hull Compaction

Cladding Hulls → Iodine → \(^{3}
\text{H}, \text{Kr}, \text{CO}_2\) Released to Atmosphere

Oxalate Pptn. & Product Conversion

Pu Oxide → Geologic Repository

Pu Nitrate Solution → Geologic Repository

MOX Fuel Fabrication

Makeup Uranium → Uranium Storage or Disposal

Packaged Waste Form

Vitrification

Uranyl Nitrate Solution → Vitrification

Np, Am, Cm, Remaining Fission Products

Packaged Waste Form
Issues with the PUREX Process

- Process is well-understood and proven to be commercially viable
- Pure plutonium stream is separated; civil use of this material is against national policy of the United States – mixed oxide fuel will be used for disposition of excess weapons plutonium in commercial reactors, however
- Minor actinides are sent to waste, greatly increasing its radiotoxicity and volume
- Major heat-generating radionuclides go into the high-level waste stream; no benefits to heat management in a geologic repository
- Minor modifications to the process (e.g., recombining uranium and plutonium streams) are easily subverted for clandestine production of plutonium
Design of GNEP Process for Treatment of LWR Spent Fuel

- Generation of no high-level liquid wastes requiring extended underground tank storage
- “Limited emissions” goal
  - Recovery of I, Kr, $^3$H, $^{14}$CO$_2$
- Added fuel cycle costs to amount to minimal increase in the busbar cost of electricity; efficient operation at high throughput
- Efficient removal, > 95%, and immobilization of long-lived fission products (specifically iodine and technetium) to reduce repository dose rate
- Ten-fold or greater reduction in high-level waste volume relative to direct disposal of spent fuel
- Integrated process: ≥99.9% removal of transuranics and short-lived fission products (Cs, Sr) to reduce radiotoxicity and heat load; no separation of pure plutonium
UREX+1a Process (Current GNEP Reference)

LWR Spent Fuel → Voloxidation/Tritium Recovery → Nitric Acid Dissolution and Clarification → UREX Process → Product Conversion

Clarified Dissolver Solution

UREX Raffinate

UREX Process

Product Conversion

Product at Desired Fuel Composition

U/TRU Blend

FPEX Process

Uranium Storage or Disposal as U₃O₈

FPEX Process

TRUEX Process

TALSPEAK Process

Liquid Product Blending

Fuel Fabrication

TRUs (oxide or metal)

Lanthanide Fission Products

Transuranics in Lactic Acid Solution

Geologic Repository

Packaged Waste Form

Decay Storage of Cs & Sr

All Remaining Fission Products except Lanthanides

Transuranics plus Lanthanide Fission Products

Transuranics plus Remaining Fission Products

Cs/Sr Aluminosilicate

Iodine

Lanthanide Fission Products

Alloying / Compaction (hulls + Tc + sludge / balance of hulls)

Washed Cladding Hulls and Sludge

Technetium

Uranyl Nitrate Solution

Uranyl Nitrate Solution

Xe, Kr, CO₂

Washed Clarifier Hulls

Uranyl Nitrate Solution

U₃O₈

Geologic Repository
Laboratory-Scale Testing of the UREX+1a Process
(July 2006, 1 kg LWR spent fuel; solvent extraction process segment only; feed material: Cooper [BWR, 34 GWd/t] and H.B. Robinson [PWR, 76 GWd/t])

<table>
<thead>
<tr>
<th>Element</th>
<th>Recovery Eff.</th>
<th>Remarks</th>
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<tbody>
<tr>
<td>Uranium</td>
<td>99.9992%</td>
<td>Non-TRU (&lt;100 nCi/g)</td>
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<tr>
<td>Technetium</td>
<td>95.5%</td>
<td>Soluble Tc</td>
</tr>
<tr>
<td>Cesium</td>
<td>&gt;99.85%</td>
<td></td>
</tr>
<tr>
<td>Strontium</td>
<td>99.1%</td>
<td></td>
</tr>
<tr>
<td>Plutonium</td>
<td>&gt;99.998%</td>
<td>Total lanthanide content of transuranics &lt;0.05% (DF&gt;2,000)</td>
</tr>
<tr>
<td>Neptunium</td>
<td>&gt;99.992%</td>
<td></td>
</tr>
<tr>
<td>Americium</td>
<td>&gt;99.97%</td>
<td></td>
</tr>
<tr>
<td>Curium</td>
<td>&gt;99.9993%</td>
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### UREX+ Process: Possible Variations

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<tr>
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<tbody>
<tr>
<td>UREX+1</td>
<td>U</td>
<td>Tc, I</td>
<td>Cs, Sr</td>
<td>Other FPs</td>
<td>TRU+Ln</td>
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<tr>
<td></td>
<td>(highly purified)</td>
<td>(LLFPs, dose issue)</td>
<td>(short-term heat mgmt.)</td>
<td></td>
<td>(temporary storage)</td>
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<td>UREX+1a</td>
<td>U</td>
<td>Tc, I</td>
<td>Cs, Sr</td>
<td>FPs</td>
<td>TRU</td>
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<td>(highly purified)</td>
<td>(LLFPs, dose issue)</td>
<td>(short-term heat mgmt.)</td>
<td>(including lanthanides)</td>
<td>(group extraction)</td>
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<td>UREX+2</td>
<td>U</td>
<td>Tc, I</td>
<td>Cs, Sr</td>
<td>Other FPs</td>
<td>Pu+Np</td>
<td>Am+Cm +Ln</td>
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<td>(highly purified)</td>
<td>(LLFPs, dose issue)</td>
<td>(short-term heat mgmt.)</td>
<td></td>
<td>(for FR recycle fuel)</td>
<td>(for FR recycle fuel)</td>
<td>(temp. storage)</td>
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<td>UREX+3</td>
<td>U</td>
<td>Tc, I</td>
<td>Cs, Sr</td>
<td>FPs</td>
<td>Pu+Np</td>
<td>Am+Cm</td>
<td></td>
</tr>
<tr>
<td></td>
<td>(highly purified)</td>
<td>(LLFPs, dose issue)</td>
<td>(short-term heat mgmt.)</td>
<td>(including lanthanides)</td>
<td>(for FR recycle fuel)</td>
<td>(heterogeneous targets)</td>
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<tr>
<td>UREX+4</td>
<td>U</td>
<td>Tc, I</td>
<td>Cs, Sr</td>
<td>FPs</td>
<td>Pu+Np</td>
<td>Am</td>
<td>Cm</td>
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<tr>
<td></td>
<td>(highly purified)</td>
<td>(LLFPs, dose issue)</td>
<td>(short-term heat mgmt.)</td>
<td>(including lanthanides)</td>
<td>(for FR recycle fuel)</td>
<td>(heterogeneous targets)</td>
<td>(storage)</td>
</tr>
</tbody>
</table>

- All processes provide the same repository benefits
- UREX+1 and UREX+1a are designed for homogeneous recycle of all transuranics to fast spectrum reactors
- UREX+2, +3 and +4 are designed for heterogeneous recycling, possibly as an evolutionary step, to preclude the need for remote fabrication of fuel
UREX+1a Process: Waste and Storage Products

LWR Spent Fuel

Voloxidation/ Tritium Recovery

Nitric Acid Dissolution and Clarification

UREX Process

Product Conversion

U₃O₈

Uranium Storage or Disposal as U₃O₈

Decay Storage of Cs & Sr

Geologic Repository

High-Level Waste Form Production

FPEX Process

UREX Raffinate

Uranyl Nitrate Solution

Cs/Sr Aluminosilicate

TRUs (oxide or metal)

TALSPEAK Process

Lanthanide Fission Products

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Washed Cladding Hulls and Sludge

Technetium

All Remaining Fission Products except Lanthanides

Iodine

Transuranics plus Lanthanide Fission Products

Transuranics in Lactic Acid Solution

Transuranics plus Remaining Fission Products

Packaged Waste Form

Alloying / Compaction (hulls + Tc + sludge / balance of hulls)
**Alternative Process**

- UREX+1a process is a group TRU extraction process that requires remote fabrication of the TRU recycle fuel.
- Remote fuel fabrication will almost certainly result in higher fuel fabrication costs (relative to glovebox fabrication of U-Pu MOX fuel).
- Technologies for remote fabrication will not be available at a high level of technological maturity for a number of years.
- Therefore, an alternative process (UREX+3) is being considered that would recycle U-Pu-Np as fast reactor fuel, with Am being transmuted in dedicated target assemblies; Cm can be separated for storage and decay to Pu/Am.
Waste and Product Streams: Present Plans for Disposition

- **Tritium**: collect as tritiated water, incorporate in grout and encapsulate

- **Cladding hulls** (greatest volume contributor to HLW):
  - Largest fraction: wash (target: non-TRU), compact and encapsulate
  - Portion: use as matrix material for technetium/UDS alloy

- **Technetium**: recover in metallic form, combine with undissolved solids and a fraction of the cladding hulls, dispose as a metallic HLW form

- **Xenon/krypton**: immobilize in zeolite or clathrates, dispose as HLW; potential for xenon-krypton separation is being studied

- **Carbon-14**: recover as CO₂, convert to carbonate and dispose as HLW
Waste and Product Streams: Present Plans for Disposition (continued)

- **Iodine**: trap in silver-coated zeolite, convert to potassium iodate and dispose as HLW
- **Uranium**: store in drums for future use (re-enrichment, recycle to fast reactors) or disposal as LLW
- **Cesium/strontium**: recover at high level of purity, immobilize in an aluminosilicate mineral matrix and store until radionuclides have decayed to levels acceptable for disposal as LLW
- **Residual fission products** (lanthanides and transition metals): most have decayed to stable isotopes; lanthanides are good glass-formers and can be vitrified at high level of waste loading; transition metals may be better combined with the recovered technetium and alloyed with Zircaloy cladding hulls
**UREX+1a/+3 Processes: Projected Waste Generation for Every 100 Metric Tons of Spent Fuel Processed**

<table>
<thead>
<tr>
<th>Waste Stream</th>
<th>Waste Composition</th>
<th>Category</th>
<th>Volume, m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium</td>
<td>U₃O₈ powder</td>
<td>(Storage)</td>
<td>18</td>
</tr>
<tr>
<td>Cesium/strontium</td>
<td>Cs/Sr aluminosilicate</td>
<td>(Storage)</td>
<td>1.1</td>
</tr>
<tr>
<td>Hulls + Tc, sludge</td>
<td>Zr-Fe based alloy</td>
<td>HLW</td>
<td>0.6</td>
</tr>
<tr>
<td>Compacted hulls</td>
<td>Non-TRU Zr</td>
<td>HLW</td>
<td>6.1</td>
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<tr>
<td>U losses</td>
<td>Borosilicate glass</td>
<td>HLW</td>
<td>1.0 - 3.4</td>
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<tr>
<td>TRU losses</td>
<td>Borosilicate glass</td>
<td>HLW</td>
<td>0.06</td>
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<tr>
<td>Iodine</td>
<td>Potassium Iodate</td>
<td>HLW</td>
<td>0.018</td>
</tr>
<tr>
<td>Krypton</td>
<td>Zeolite/aluminosilicate</td>
<td>HLW</td>
<td>0.014</td>
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<td>Tritium</td>
<td>Grout</td>
<td>HLW</td>
<td>&lt;0.01</td>
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<td>Lanthanide FPs</td>
<td>LABS glass</td>
<td>HLW</td>
<td>0.31</td>
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<tr>
<td>Carbon-14</td>
<td>Sodium carbonate</td>
<td>HLW</td>
<td>0.034</td>
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</tbody>
</table>

For comparison, 100 tons of untreated spent fuel has an unpackaged volume of 45 m³
Future Improvements

- Cladding hulls comprise the largest part of the estimated high-level waste volume.

- Studies are in progress to evaluate the potential for recycling zirconium for production of LWR fuel cladding.

- Industrial suppliers have indicated the feasibility of fabricating cladding with small content of $^{93}\text{Zr}$.

- Other activation products are removed in the chloride volatility process used for Zr recovery.

- On the other hand, a ten-fold reduction in high-level waste volume may be more than adequate; the compacted hulls and hardware are not significant contributors to the repository heat load.