SRNL Tritium Development Activities for Accelerator Mo-99 Production

Savannah River National Laboratory, Aiken, SC 29808

Abstract: SRNL has support accelerator production of Mo-99 since 2012. Use of deuterium accelerators using a tritium target require additional study and development for gas handling, processing, storage, confinement, and waste disposal. A brief description of some SRNL tritium-related studies to support Mo-99 production are presented here.

Future Work: SRNL continues to evaluate tritium removal from SF6 in FY23. In addition to current SRNL work, future Mo-99 support work will focus on actinide processing, radiological facility issues, and waste disposal activities.

Bellows Sealed Valve Cycle Testing
- Nupro B-series bellows sealed valve for tritium service
- 3 rotates closed pressure test sizes: 1C, 3C, and 5C
- Three tip seal for tritium (no heat-PI for tritium service)
- Copper, bellows fail; actuator failure Copper tests shown here (1C dual actuated)

Aluminum Scroll Pump Development
- Normetex 9 cfm all-metal pump no longer made
- Stainless steel replacement heavy, $$$
- Aluminum coated pump developed
- Stainless steel replacement heavy, $$$

Aluminum Scroll Pump Development
- Copper tests shown here (1C dual actuated)
- 3 failure mechanisms: stem wear, bellows failure, actuator failure
- What is smallest actuator to give reliable valve closure: 150k cycles

Accelerator Mo-99 Production

KOH-Based Process
- Mo-99 as feed for accelerator scc/s He
- KOH-based process
- Small & F ions react with SF6 confinement materials and hold tritium
- Goal is to remove tritiated compounds from SF6 for reuse
- Numerous chemical species can be formed

Sulfur Hexafluoride (SF6) Disposition Strategies
- SF6 commonly used as electrical insulator for accelerators
- Electrical current dissociates SF6 to S and F ions
- Most S and F ions recombine to form SF6
- S & F ions react with SF6 confinement materials and hold tritium
- Numerous chemical species can be formed
- Goal is to remove tritiated compounds from SF6 for reuse

Separations/clean-up method based on species present
- Data collection planned in FY23

Nominal Stripped Volume (L)
- Stripped volume specifications
- Stripped volume specifications

Nominal Stripped Volume (L)
- Stripped volume specifications
- Stripped volume specifications

Mo-99 Production
- Mo-99 production for 2021
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Abstract: The SRNL is supported by the DOE National Nuclear Security Administration to provide R&D assistance to potential new US Mo-99 producers. The assistance includes the development of flowsheets for the purification of U from dissolved targets following Mo-99 recovery and the removal of high specific activity fission products from waste streams to lower the classification (e.g., Class C to Class A or B) of the waste form. A modified PUREX process was demonstrated for nominally 90% U recovery allowing the purge of a small amount of U from the inventory each cycle to minimize the production of Pu during subsequent target irradiations. In small column tests using ammonium molybdophosphate and crystalline silicotitanate (R9120-B) with a simulated acidic waste solution containing U, Pu, Np and non-rad fission product, both ion exchange materials were highly specific for cesium. Neither material removed significant Sr or U.

U Recovery from Mo-99 Targets

• Solvent extraction modeling combined with experimental validation to support the design of a UREX solvent extraction process for approximately 90% U recovery
• Surrogate solution prepared for U purification experiment based on:
  - Target irradiation for 10 days at 30 kW
  - Inventory of fission and activation products calculated using Origin 2.2 following two-days of decay
  - Concentrations scaled based on 275 g/L U in the feed solution
• Feed to solvent extraction process adjusted to 1 M HNO₃ following Mo-99 recovery

Concentrations of U and Selected Activation and Fission Products in Surrogate Solution

<table>
<thead>
<tr>
<th>Element</th>
<th>Concentration (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>1.05</td>
</tr>
<tr>
<td>Np</td>
<td>6.78E-05</td>
</tr>
<tr>
<td>Pu</td>
<td>1.34E-04</td>
</tr>
<tr>
<td>Ce</td>
<td>5.06E-04</td>
</tr>
<tr>
<td>Cs</td>
<td>2.64E-04</td>
</tr>
<tr>
<td>Eu</td>
<td>4.10E-05</td>
</tr>
<tr>
<td>Nd</td>
<td>3.70E-04</td>
</tr>
<tr>
<td>Ru</td>
<td>5.25E-04</td>
</tr>
<tr>
<td>Sr</td>
<td>4.27E-04</td>
</tr>
<tr>
<td>Np(Sr)</td>
<td>1.53E-04</td>
</tr>
<tr>
<td>Zr</td>
<td>8.74E-04</td>
</tr>
</tbody>
</table>

• Argonne Model for Universal Solvent Extraction (AMUSE) used for flowsheet design
  - Number of extraction stages and flow rates were adjusted until the predicted U recovery was nominally 90%

• UREX Process Design
  - Feed solution containing 275-100 g/L, adjusted to 1 M HNO₃
  - 30 vol % tributylphosphosphate (TBP) solvent
  - 0.25 M acetylated acidic acid (AAA) in 0.5 M HNO₃, used to scrub fission and activation products from loaded solvent
  - UMR Colex 300 TBP/300/AAC/300 in 0.5 M HNO₃
  - UMR Colex 300 TBP/300/AAC/300 in 0.5 M HNO₃

• U recovery achieved was 85.7 ± 1.9%
  - Recovery, predicted by AMUSE was 90.8%
  - AMUSE model was not completely optimized to match the SRNL equipment and could be adjusted to more closely predict the measured recovery
  - Measured concentration profiles were generally in good agreement with profiles predicted by AMUSE

Decontamination Factors (DF)

<table>
<thead>
<tr>
<th>Speciation</th>
<th>Measured</th>
<th>Predicted</th>
</tr>
</thead>
<tbody>
<tr>
<td>Np</td>
<td>2.67E10</td>
<td>1.8E10</td>
</tr>
<tr>
<td>Pu</td>
<td>3.90E-17</td>
<td>1.8E-17</td>
</tr>
<tr>
<td>Ce</td>
<td>7.08E-17</td>
<td>3.9E-17</td>
</tr>
<tr>
<td>Cs</td>
<td>3.99E-17</td>
<td>3.9E-17</td>
</tr>
<tr>
<td>Eu</td>
<td>3.99E-16</td>
<td>3.9E-16</td>
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<tr>
<td>Nd</td>
<td>2.10E-15</td>
<td>2.1E-15</td>
</tr>
<tr>
<td>Pr</td>
<td>1.20E-15</td>
<td>1.2E-15</td>
</tr>
<tr>
<td>Sr</td>
<td>3.30E-15</td>
<td>3.3E-15</td>
</tr>
<tr>
<td>Np(Sr)</td>
<td>6.90E-17</td>
<td>6.9E-17</td>
</tr>
<tr>
<td>Pu</td>
<td>1.80E-17</td>
<td>1.8E-17</td>
</tr>
</tbody>
</table>

• Predicted DF were much higher than measured values
  - Pu, Eu, Sr, Np, Sr and Ce concentrations below minimum detection of the analysis

• Waste stream from irradiated LSU saline solution, pH 1
  - Waste to be neutralized and grafted
  - Goal is to minimize amount of Class A and C waste generated by capturing and segregating limiting isotopes

High Specific Activity Fission Product Separation

• Waste stream from irradiated LSU saline solution, pH 1
  - Waste to be neutralized and grafted
  - Goal is to minimize amount of Class A and C waste generated by capturing and segregating limiting isotopes

• Surrogate waste solution prepared based on prior results from Mo titanate column testing, 238U primary metal ion by mass, 127I, 90Sr, 144Ce limiting by regulations/active

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<tr>
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<tbody>
<tr>
<td>238U</td>
<td>58E16</td>
</tr>
<tr>
<td>90Sr</td>
<td>2.3</td>
</tr>
<tr>
<td>90Zr</td>
<td>4.6</td>
</tr>
<tr>
<td>90Nb</td>
<td>5.1</td>
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<tr>
<td>232Th</td>
<td>21.5</td>
</tr>
<tr>
<td>235U</td>
<td>24.0</td>
</tr>
<tr>
<td>249Pu</td>
<td>7.3</td>
</tr>
<tr>
<td>241Am</td>
<td>2.0</td>
</tr>
<tr>
<td>208Tl</td>
<td>0.1</td>
</tr>
<tr>
<td>237Np</td>
<td>1.3</td>
</tr>
<tr>
<td>239Pu</td>
<td>2.0</td>
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<tr>
<td>240Pu</td>
<td>12.0</td>
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<tr>
<td>238U</td>
<td>3.8</td>
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<tr>
<td>236U</td>
<td>1.3</td>
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<tr>
<td>237Np</td>
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<tr>
<td>235U</td>
<td>4.4</td>
</tr>
<tr>
<td>239Pu</td>
<td>0.9</td>
</tr>
<tr>
<td>237Np</td>
<td>0.3</td>
</tr>
<tr>
<td>237Np</td>
<td>0.2</td>
</tr>
</tbody>
</table>

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Aqueous Phase U Concentration Profile

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• Recovery predicted by AMUSE was 90.8%
• AMUSE model was not completely optimized to match the SRNL equipment and could be adjusted to more closely predict the measured recovery
• Measured concentration profiles were generally in good agreement with profiles predicted by AMUSE

• Experiment demonstrated that AMUSE can be used to support the design of a UREX process to control the U recovery efficiency for recycle of 10 to 99 targets

CST R9120-B more specific for Cs. Rp from ph 1 suitable
• Also absorbed significant Mo, little in waste
• Little retention of Sr, Ce and lanthanides
• Some retention of Pu
• Little retention of U

• Retested CST at pH 9
  - Adjusted with carbonate to written pH standard
  - Adjusted to pH 9

• Capacity likely affected by low Cs concentration (below 4E-5 M)
  - CST R9120-B
  - Retention of Cs at pH 9
  - Lowered by carbonate exchange
Management and categorization of wastes resulting from Mo-99 production

J.W. Amoroso, Savannah River National Laboratory, Aiken, SC 29808

Abstract

Appropriate waste management strategies are critical to the production of Mo-99 using low enriched uranium (LEU). There are various Mo-99 production processes for which unique methods and strategies for treatment and disposal of the waste streams is needed. Product specifications, acceptance criteria, facility design, and process operations must all be considered as part of the overall waste management strategy.

Waste Stream Projections

- Contact handled waste
- Process system components
- Excessed equipment & large items
- Process raffinate
  • Compute estimated radionuclide inventory from irradiation and decay based on projected processing steps and volumes

Waste Solidification

- Reduce the long-term environmental burden through efficient disposal of waste materials
- Chemically bind the radioactive and hazardous components into a solid, durable material that will withstand degradation for thousands of years
- The choice of material is dependent on the application and often is a compromise between factors such as cost, performance, and suitability
  • Crystalline ceramic
  • Vitriﬁed glass
  • Cementitious materials
  • Composite

Hydrogen Generation

- Published characteristics (i.e. deposited energy (W/Ci), decay mode) and published G values (molecules $H_2$ / 100 eV) for beta/gamma and alpha irradiation of pure water were used to calculate hydrogen generation rates for various scenarios
- Incorporate best ﬁt exponential and linear functions to integrate the hydrogen generation (moles) from most radioisotopes

Future Work

- Continue to evaluate process system ﬂowsheets to compute waste stream estimates
- Evaluate waste management strategies to ensure available disposition path
- Provide waste form design and processing experience to facilitate efﬁcient waste disposition strategies

Uranium Lease and Take Back (ULTB) Program

- U.S. program to use lease contracts to make LEU available for the domestic production of Mo-99 for medical uses
- U.S. program to use take-back contracts for the final disposition of spent nuclear fuel created by the irradiation, processing, or purification of leased LEU for which there is no commercial disposition path
  • Includes radioactive waste created by the irradiation, processing, or purification of leased LEU, for which the producer does not have access to a disposal path