Inventory-Based Computational Analysis of Hanford Tank Waste
Tank Farms Overview
HANFORD TANK WASTE

- Product of more than 40 years of plutonium production
- 3 chemical separations processes
- Many distinct waste streams and compositions
- 56 million gallons in 177 tanks, including:
  - Various metals
  - Fission products
  - Uranium (~600 metric tons)
  - **Plutonium** (670 kg)
- Criticality safety analysis based on presence of absorber metals:
  - Aluminum
  - Chromium
  - Iron
  - Manganese
  - Nickel
  - Silicon
TANK WASTE PHASES

- **Supernatant**
  - Liquid phase
  - Minimal Pu (~3.5 kg, less than 0.5% of total)

- **Saltcake**
  - Crystallized liquids from Evaporator concentration
  - Majority sodium, much lower Pu masses

- **Sludge**
  - Non-water soluble compounds
  - Large amounts of iron, manganese, aluminum
  - Holds majority of tank plutonium
THE BEST-BASIS INVENTORY (BBI)

- Database of best-estimate tank inventories for:
  - 46 radionuclides
  - 25+ chemical analytes
- Estimates from tank history, sample data, fuel depletion calculations
- Many distinct purposes:
  - Retrieval / transfer planning
  - Chemical compatibility analysis
  - Safety basis requirements (H₂ generation)
  - Criticality safety (Pu & absorber masses)
- Tank contents split into “layers” [currently: 566]
  - Often represent one origin / composition
Tank-Specific Calculations
CALCULATION DESIGN AND METHOD

- One calculation input per tank layer:
  - Infinite-geometry, homogenous MCNP model
  - Element / isotope mass ratios derived BBI inventory
- Inner search on water fraction (wt%) → find maximum $k_{eff}$ (optimal moderation)
  - Water fraction is a BBI parameter
  - Realistic waste contains H in compounds (mainly hydroxides)
    - Highly overmoderated – analysis assumptions very conservative
- Outer search on plutonium mass multiplier → max $k_{eff}$ in target range
  - Relative increase of Pu mass (versus other solids)
  - Calculations performed for all sludge, saltcake layers
Include only Pu and credited absorbers
- Ignores other, large-mass waste constituents (Na)
- Absorbers modelled as oxides

No layer with Pu multiplier < 1.0
Five layers < 2.5x
- All associated with Plutonium Finishing Plant
- Already known to criticality safety
  - Controls on mixing tank solids
Five layers between 2.5 and 5.0
Large margin on most tank farms Pu
- 75% of Pu located in layers > 5x
- 50% of Pu located in layers > 10x
MODEL #2 – REDUCTION BY SOLUBILITY FACTORS

- Current evaluation applies element-specific reduction factors
  - Bounds Pu/absorber separation due to dissolution
  - Largest reductions on aluminum content
- General reduction in calculated Pu multipliers
  - 88% of plutonium in layers > 2.5 x
  - 75% of plutonium in layers > 5 x
- Largest change in high-aluminum layers
  - Mainly cladding waste → low Pu content

### Absorber Mass Reduction

<table>
<thead>
<tr>
<th>Element</th>
<th>Reduction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>98 %</td>
</tr>
<tr>
<td>Cr</td>
<td>70 %</td>
</tr>
<tr>
<td>Fe</td>
<td>10 %</td>
</tr>
<tr>
<td>Mn</td>
<td>12 %</td>
</tr>
<tr>
<td>Ni</td>
<td>50 %</td>
</tr>
<tr>
<td>Si</td>
<td>70 %</td>
</tr>
<tr>
<td>Zr</td>
<td>18 %</td>
</tr>
</tbody>
</table>
MODEL #3 – REDUCTION BY BBI WASH FACTORS

- **Tank-specific estimates for removal fractions**
  - Developed for retrieval process modelling
  - Mainly based on experimental data from tank samples
  - Accounts for different chemical components

- **NCS solubility assumptions nearly always individually bounding for each element**
  - Small fraction of tank/absorber combinations have BBI predict more individual removal
  - Assumption was: solubility factors were conservative taken together, over all absorbers

- **Confirmed criticality safety assumptions bound tank-specific removal fractions.**
  - Only 6 layers had the BBI values giving a more conservative final composition
  - All had very small Pu masses (< 10 g) or high Pu mass multipliers (> 200x)
    - No criticality safety significance
    - Mainly Al cladding waste (98% vs. 100% removal of Al)
MODEL #4 – ADDING URANIUM INVENTORIES

- **600 metric tons** uranium in tank waste
  - Most (75%) at or just below natural enrichment
  - Maximum enrichment – 1.02% $^{235}$U
  - Single primarily-$^{233}$U layer
    - Likely mixed with other waste during C Farm retrieval
- Previous analysis discussed U and Pu separately
- Adding U into calculations already applying solubility factors (absorber reduction):
  - 70% of plutonium in layers > 10 x
  - 80% of plutonium in layers > 5 x
  - 92% of plutonium in layers > 2.5 x
  - Only 4 layers still between 1x and 2x → all PFP-related (minimal U content)
Previous Tank Waste Models
“CARTER MODEL” (1979)

- Created from the four available tank samples
  - 2 from AX-104, one each from A-106 and C-106
  - Took bounding values for each absorber

- Composition modified to due to code limitations
  - Mercury cross-sections not available
  - Limit of 10 isotopes in one calculation
    - Hg, Cr, Ni proportionally re-assigned to Mn

- Pu concentration varied until $k_\infty < 1.0$ for all H-to-X

- 1979 calculated value was 3 g Pu/L, limits based on 1 g Pu/L
  - Using MCNP 6.2 and ENDF/B-VII.1 (without isotope substitutions):
    - 2.46 g Pu / L at $k = 0.935$
    - 2.83 g Pu / L at $k = 1.0$

<table>
<thead>
<tr>
<th>Composition (g/L)</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>100</td>
</tr>
<tr>
<td>Fe</td>
<td>100</td>
</tr>
<tr>
<td>Na</td>
<td>50</td>
</tr>
<tr>
<td>Mn</td>
<td>5</td>
</tr>
<tr>
<td>Si</td>
<td>35</td>
</tr>
<tr>
<td>Cr</td>
<td>3</td>
</tr>
<tr>
<td>Hg</td>
<td>0 or 10</td>
</tr>
<tr>
<td>Ni</td>
<td>4</td>
</tr>
<tr>
<td>NO₃</td>
<td>13 or 130</td>
</tr>
<tr>
<td>O (compounds)</td>
<td>200</td>
</tr>
</tbody>
</table>

CARTER MODEL (1979)

Composition (g/L)
- Al 100
- Fe 100
- Na 50
- Mn 5
- Si 35
- Cr 3
- Hg 0 or 10
- Ni 4
- NO₃ 13 or 130
- O (compounds) 200
“CONSERVATIVE WASTE MODEL” (1993)

- Primarily derived from sample data
  - 28 sample analyses, covering 16 tanks
  - Some input from overall tank inventory estimates
- Developed to produce smaller macroscopic absorption cross-section than actual waste
  - 2002 report compared with against inventory data for all tanks with more than 20 kg Pu
- Calculated subcritical limit of 2.6 g Pu / L.
  - Part of criticality safety evaluation until 2015

**Composition (wt%)**

<table>
<thead>
<tr>
<th>Element</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>O</td>
<td>40.7</td>
</tr>
<tr>
<td>P</td>
<td>6.9</td>
</tr>
<tr>
<td>Si</td>
<td>3.8</td>
</tr>
<tr>
<td>Na</td>
<td>21.5</td>
</tr>
<tr>
<td>Al</td>
<td>7.2</td>
</tr>
<tr>
<td>Fe</td>
<td>19.9</td>
</tr>
</tbody>
</table>

Solids density: 1200 g/L
Search results give a layer-specific measure of neutron absorption in solids:

How do the assumptions from older waste models compare?

<table>
<thead>
<tr>
<th>Waste Solids Model</th>
<th>Subcritical Pu Concentration</th>
<th>Fraction of Pu Mass Bounded by Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carter (1979)</td>
<td>1.0 g/L [derived operating limit]</td>
<td>100%</td>
</tr>
<tr>
<td></td>
<td>2.8 g/L [MCNP 6.2 – 2018]</td>
<td>92%</td>
</tr>
<tr>
<td></td>
<td>3 g/L [GAMTEC II – 1979]</td>
<td>85%</td>
</tr>
<tr>
<td>CWM (1993)</td>
<td>2.6 g/L</td>
<td>97%</td>
</tr>
</tbody>
</table>
CONCLUSIONS

• With modern computer speeds, a new tool to look at criticality safety in tank waste:
  - Pu multiplier gives a more definitive assessment of subcritical margin
  - Compare specific effects of different modelling assumptions
  - Identify any additional layers of potential interest

• Assumptions used to generate previous sets of absorber models shown to bound nearly all tank farms Pu

• Calculations are part of larger effort to focus analysis more onto specific tanks of greatest concern
Questions?