Immobilization of High Level Radioactive Waste
(An American Perspective)

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Nuclear Fuel Cycles and Waste Streams

- Uranium ore
- NORM waste
- Contaminated ware, equipment
- Encapsulation, disposal
- Fuel fabrication
- Power generation
- Nuclear Power
- Spent fuel
- Reprocessing
- Milling, enrichment
- Volatiles?
- Vitrification, storage or disposal
NF and SNF Compositions [Ref. 1]

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half Life (yrs)</th>
<th>In NF (kg)</th>
<th>In SNF (kg)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-234</td>
<td>2.46E+5</td>
<td>0.05</td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td>U-235</td>
<td>2.04E+8</td>
<td>37.5</td>
<td>8.6</td>
<td></td>
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<tr>
<td>U-236</td>
<td>2.36E+7</td>
<td></td>
<td>5.1</td>
<td></td>
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<tr>
<td>U-238</td>
<td>4.47E+9</td>
<td>962.4</td>
<td>934.4</td>
<td></td>
</tr>
<tr>
<td>Total U</td>
<td></td>
<td>1000</td>
<td>948.2</td>
<td>Depletion of U</td>
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<tr>
<td>Np-237</td>
<td>2.14E+6</td>
<td></td>
<td>0.57</td>
<td></td>
</tr>
<tr>
<td>Pu-239</td>
<td>2.4E+4</td>
<td></td>
<td>5.56</td>
<td></td>
</tr>
<tr>
<td>Pu-240</td>
<td>6564</td>
<td></td>
<td>2.46</td>
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</tr>
<tr>
<td>Pu-242</td>
<td>3.7E+5</td>
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<td>0.66</td>
<td></td>
</tr>
<tr>
<td>Total Pu</td>
<td></td>
<td></td>
<td>10.4</td>
<td>Proliferation concern, Incentive to recycle</td>
</tr>
<tr>
<td>Am-241</td>
<td>432.2</td>
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<td>0.05</td>
<td></td>
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<tr>
<td>Am-243</td>
<td>7370</td>
<td></td>
<td>0.16</td>
<td></td>
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<tr>
<td>Total actinide fission products</td>
<td></td>
<td></td>
<td>19.86</td>
<td></td>
</tr>
<tr>
<td>Other fission products (Cs, Sr, Se, I, Tc, Zr etc.) and mass lost in energy conversion</td>
<td></td>
<td></td>
<td>31.94</td>
<td>Major concern in glass immobilization</td>
</tr>
</tbody>
</table>
## Fission products requiring long term isolation

<table>
<thead>
<tr>
<th>FP</th>
<th>Half life (yrs)</th>
<th>Activity discharged (Ci/yr)</th>
<th>Specific activity (Ci/g)</th>
<th>Boiling point (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sr-90</td>
<td>28</td>
<td>2.1E+6</td>
<td>140</td>
<td>1384</td>
</tr>
<tr>
<td>Cs-137</td>
<td>30</td>
<td>2.9E+6</td>
<td>870</td>
<td>678.4</td>
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<tr>
<td>Se-79</td>
<td>6E+4</td>
<td>11</td>
<td>0.07</td>
<td>684.9</td>
</tr>
<tr>
<td>Sn-126</td>
<td>1E+5</td>
<td>15</td>
<td>0.028</td>
<td>2270</td>
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<tr>
<td>Tc-99</td>
<td>2.1E+5</td>
<td>390</td>
<td>0.017</td>
<td>4877</td>
</tr>
<tr>
<td>Zr-93</td>
<td>1.5E+6</td>
<td>50</td>
<td>0.0025</td>
<td>4377</td>
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<tr>
<td>Cs-135</td>
<td>3E+6</td>
<td>8</td>
<td>0.0012</td>
<td>678.4</td>
</tr>
<tr>
<td>Pd-107</td>
<td>7E+6</td>
<td>3</td>
<td>0.00051</td>
<td>2977</td>
</tr>
<tr>
<td>I-129</td>
<td>1.57E+7</td>
<td>1.0</td>
<td>0.00018</td>
<td>184</td>
</tr>
</tbody>
</table>
Current strategy of storage or disposal and some concerns

- Short lived isotopes (Sr-90 and Cs-137):
  Current Solution: Allow cooling by interim storage (need 500-1000 years to meet NRC disposal guideline), or use for medical and other uses
  Issue: Aqueous solubility is high. Proliferation concern.
  Solution: Stabilization for safe storage or use?

- Low boiling point of Cs-137, Se-79, Cs-135, and I-129. Tc-99 (as pertechnatate)
  Solution: Vitrification
  Issue: Volatility will lead to evaporation of these isotopes. Need low temperature stabilization.
  Note: Experimental evidence is that all I-129 and a small part of Tc volatalizes (Ref: Bagassen et al.) the latter as pertechnatate.

- Formation of radium (Ra-225 and Ra-226) in the canisters.
  Current solution: Storage in safe canisters or after stabilization.
  Issue: Ra aqueous solubility is high, may get into environment. Become ingestion hazard in 10,000 years range.
  Solution: No solution is in horizon.
Solution for volatiles

• Stabilize condensed Tc-99 and I-129 at room temperature by other methods.
  - Convert pertechnatate into cation by chemical reduction.
  - Use chemical immobilizers for I-129.
  - Then stabilize in low temperature matrices.

• Convert Cs-127 into insoluble compounds and matrices, and use as safe radioactive sources.
Test criteria after immobilization

1. **Toxicity Leaching Procedure (TCLP)**
   This is a Land Disposal Requirement of the Environmental Protection Agency (EPA) for any hazardous contaminants

   Waste form sample is crushed to <0.8 mm size and put in circulating water for 29 hours. The leachates are analyzed for the hazardous contaminants. The Universal Testing Standard (UTS) limits in mg/L are

   \[
   \begin{align*}
   \text{Cd} & (0.11), \quad \text{Ba} (21), \quad \text{Cr} (0.6), \quad \text{Ni}, \quad \text{Ag} (11), \quad \text{Pb} (0.75), \quad \text{and Zn} \ (4.3). \\
   \end{align*}
   \]

   Note: These limits keep changing, so look for the latest limits while using.
2. Product Consistency Test (PCT) (ASTM C1285-02)

- Used for integrity of the waste form
- Measure the matrix components leached and determine loss in weight.
- Seven day immersion in water kept at 90 C.
- For vitrified glass waste form, mass loss of Na, Si, B < 2 g/square meter of the surface area of the sample.
3. **ANS 16.1 90-Day Leaching Test**

Leachability Index (LI)

\[ LI = \log (\text{diffusion constant of the rad contaminant in water}) \]

in ANS 16.1 test.

Test is run by immersing identical samples for 3, 7, 24, 24, 24, 45, ... hours in 20 times water volume. Level of contaminants is determined and diffusion constant is calculated.

2 inch x 4 inch cylinder

2 inch x 2 inch cube

Requirement: 500 psi
5. Vapor Hydration Test
   ASTM WK84

   • Test method similar to PCT but at 200 C.
   • Designed for waste glass durability
   • Glass alteration rate shall be <50 g/sq.m-day
   • Measured for seven days.
Thermal, Radiation, and Bio-Degradation

- 6. Thirty thermal cycles between 60 and -40 C (ASTM B553-79)
- 7. No evidence of culture growth as per tests ASTM G21-96 and G22-76.
- 8. Exposure to > 1 x E8 rads or maximum level expected within the waste form.
- 9. Immersion degradation: 90 days as per ANS 16.1

In all cases the compressive strength at the end should be > 500 psi as per ASTM C39/C39M-01.
Problems with established tests

• Most tests are designed for boro silicate glass or glass crystalline materials. No established tests exist for alternative waste forms.

• Fission products may need to be stabilized in alternate waste forms. Tests need to be yet established.

• For short lived isotopes (and hence high activity), such as I-131, 90-day immersion test does not provide adequate results. After certain period (456 hours for I-131) increments in leaching level are too low to be measured. Thus one cannot make a long term determination using ANS 16.1 test.

• Acceptability criteria for glass components in tests such as PCT are too liberal (2g/m2). Other waste forms perform much better. American Academy is pushing for alternative waste forms.
Conclusions

• Glass vitrification has been accepted for immobilization and maximum evaluation has been done for this technology in the U.S. and Europe. Russian Federation uses phosphate glass.

• Power plant waste has not been vitrified in the US for safe storage, but plans are under way to vitrify defense waste (at Hanford) and waste acceptance criteria have been develop for storage in Yucca Mountain.

• Defense waste is a result of recycling spent fuel and hence this study will be ideal for Korea in case plans are developed for recycling spent fuel.

• Most contaminants may go in glass, but issue exists about soluble and volatile isotopes for which other waste forms may be needed.

• Ambient temperature technologies are being developed for the volatile contaminants, leading research has been in phosphate cement (Ceramicrete) technology developed in Argonne National Laboratory and tested under DOE and Russian Federation collaboration.
Relevant Standards


- Vapor hydration test (Full standard for the test is yet to be developed).
References

1. Nuclear Fuel Cycle, Chapter 9
Acknowledgement

Thanks to Korea Institute of Nuclear Safety for inviting me to this symposium and Dr. Sueng Young Jeong for the hospitality in Korea.

Dr. Jeong is one of the original inventors of phosphate cements.