Delivering science and technology to protect our nation and promote world stability.
Chlorine Worth Study in Support of PF-4 Operations

Theresa Cutler, Travis Grove, Jesson Hutchinson, Kelsey Amundson, Nick Wynne, Travis Smith

February 2022

LA-UR-22-21223
Motivation

• Aqueous Chloride Operations at PF-4 are important:
  – Recover Pu from pyrochemical residues
  – Reduces waste sent to WIPP
  – Increased throughput for Am production

• Aqueous Chloride Operations have very conservative mass limits (~520 grams Pu)
  – Significant amounts of Chlorine but calculations not crediting Cl-35 neutron absorption
  – Accounting for Cl-35 absorption leads to higher mass limits

• Can experiments be designed/conducted to provide technical justification to NCS in order to increase mass limits?

This is in direct support of 30 pits per year mission, one of the top priorities for NNSA
Timeline (ACCELERATED)

• Initial tasking: June 2020
• Preliminary Design Complete (CED-1 equivalent): November 2020
• Final Design Complete (CED-2 equivalent): March 2021
• Part Receipt and Inspection Complete (CED-3a equivalent): October 2021
• Experiment Execution (CED-3b equivalent): December 2021
• Experiment Documentation Write-up Complete (start CED-4a equivalent): January 2022
• ICSBEP Target (CED-4a equivalent): Fall 2022
Design Process

• Experiment Design Process related to the 2019 ARCHIMEDES LDRD project
  - See the current LDRD EUCLID project for in-depth information

• Design Process:
  - Examine application Pu concentration ranges and associated Cl-35 (n,γ) sensitivities
    ▪ Determine specific Pu concentration applications that cover concentration ranges
  - Develop multiple generic experimental benchmarks
    ▪ Materials, geometries
    ▪ Compare experimental benchmark designs to these applications (Cl-35 (n,γ) sensitivities, i.e. $c_k$ and partial $c_k$)
    ▪Iterate on benchmark design

• Note: $c_k$ and partial $c_k$ are similarity coefficients that utilize model sensitivities to nuclear data and uncertainties associated with that nuclear data
  - What is the best way to determine if two models are “similar”?
Gap Analysis (Comparison to Existing Benchmarks)

- A comparison to existing experiments was performed.
- Very few benchmarks sensitive to Cl-35 \((n,\gamma)\) exist.
- The sensitivity of these benchmarks is much lower than the application.
Final Designs

- Reflector: 3” HDPE (top, bottom, sides)
- Fuel: 5x4 (20 total per unit) ZPPR plates, ~105 g Pu per plate
- Moderator: HDPE (varying geometry)
- Cl Material: PVC or CPVC (varying geometry)

- Configurations:
  1. optimized for 30 g/L application (covers 20-100 g/L range)
     - Stack of HDPE-PVC-HDPE on ZPPR plates
  2. optimized for 300 g/L application (covers 300-400 g/L range)
     - ~7.9” diameter PVC cylinder inside HDPE on ZPPR plates
  3. optimized for 600 g/L application (covers 500-600 g/L range)
     - ~7.9” diameter CPVC cylinder inside HDPE on ZPPR plates
Final Designs

- Note that Al Tray and Al frame (shown in grey) is used to conduct heat out of assembly
  - For the top partial stack, “bottom” ZPPR tray (and shroud) sits directly on membrane/top stationary platform
  - For the bottom partial stack, the shroud goes through the bottom reflector and directly touches the platen
Sensitivity Plots: Cl-35 (n,γ)

Configuration 1
20-100 g/L

Configuration 2
300-400 g/L

Configuration 3
500-600 g/L
Internal Heat Generation Consideration

Final Heat Transfer Model
Initial model without heat removal system:
max temp > 60 °C

Base frame going to platen

Interlocking frame connection to tray and base frame
Gap Considerations

• Compression rods screw into membrane (very small thread length)
  - Compression spacers provide additional surface and separation distance of steel nut from assembly

• Compression plate, studs, spacers and nuts reduce gaps which cause significant neutron streaming paths in assembly
  - Residual gaps measured with shim gauges during experiment
Lateral Gap Considerations

- For lateral gaps, the goal is to align the top and bottom stacks together with known uncertainties
  - Membrane has slight divot in top and bottom sides that the top and bottom stacks align into
  - Assured alignment by operating in LOCAL mode before starting remote approach
  - Imperfect alignment will cause reactivity loss, which is accentuated by the divot
  - Provides a visual indication of alignment from control room (through cameras)
Results
## Measured Results

<table>
<thead>
<tr>
<th>Configuration</th>
<th>Measured Reactor Period (seconds)</th>
<th>Associated Excess Reactivity (cents)</th>
<th>Maximum Observed ΔT (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>57.6</td>
<td>16.5</td>
<td>7.4</td>
</tr>
<tr>
<td>2</td>
<td>58.1</td>
<td>16.4</td>
<td>14.4</td>
</tr>
<tr>
<td>3</td>
<td>85.8</td>
<td>12.4</td>
<td>16.1</td>
</tr>
</tbody>
</table>
Configuration 1: 30 g/L

- Final configuration: 8 units
  - 4 full units on bottom
  - 3 full units on top
  - Partial unit on top of top (Al is aluminum plates nearly matching Pu plate dimensions)
- RTDs
  - Every unit on bottom
  - Top three units on top
Configuration 2: 300 g/L

- Final configuration: 14 units
  - 7 full units on bottom
  - 7 full units on top
  - 0.875” top reflector (reduced from 3.000”)
  - RTDs
    - 5 of 7 units
    - 5 of 7 units
Configuration 3: 600 g/L

- Final configuration: 18 units
  - 10 full units on bottom
  - 7 full units on top
  - Partial unit on top of top (Al is aluminum plates nearly matching Pu plate dimensions)
- RTDs
  - Top: 6 of 8 units
  - Bottom: 5 of 10 units
Physical Measurements

- Previous benchmark experiments have taught us a lot!
  - Many physical measurements required
  - Heights, gaps rotation, levelness
  - Samples of plastics received and sent out for detailed chemical analysis
Preliminary Results

• Simplified MCNP geometry
  – Includes detailed Pu plates
  – Does not include gaps
  – Does not include detailed compositions

<table>
<thead>
<tr>
<th>Configuration</th>
<th>Experiment</th>
<th>Calculated</th>
<th>Partial $c_k$ Cl-35 (n,γ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.00035</td>
<td>1.00613</td>
<td>0.94 (30g/L)</td>
</tr>
<tr>
<td>2</td>
<td>1.00034</td>
<td>1.01211</td>
<td>0.99 (300 g/L)</td>
</tr>
<tr>
<td>3</td>
<td>1.00026</td>
<td>1.00199</td>
<td>0.99 (600 g/L)</td>
</tr>
</tbody>
</table>
Future Work

- Draft ICSBEP benchmark
  - Target Fall 2022 Review Group Meeting, which means full completion by August 2022
  - Poly, PVC, CPVC chemical analysis
    - Received samples with parts
    - Requests out to companies for quotes; usual company no longer doing full analysis
  - Evaluate measurement uncertainty
  - Detailed (very detailed) MCNP model
  - Section 1 rough draft complete
PF-4 Operations Team Engagement

• Aqueous Chloride Operations Personnel Attended 2\textsuperscript{nd} week
  − Supervisor, process engineers, operators
  − Participated in 1/M process
  − Loaded fuel
  − Felt PERSONALLY connected to the work
Acknowledgements

• This work was funded under the Material Recycle and Recovery Program, NNSA Plutonium Program Office (NA-191), under Office of Production Modernization (NA-19), funded and managed by the National Nuclear Security Administration for the Department of Energy

• NCERC is supported by the DOE Nuclear Criticality Safety Program, funded and managed by the National Nuclear Security Administration for the Department of Energy

• This work was supported by the US Department of Energy through the Los Alamos National Laboratory. Los Alamos National Laboratory is operated by Triad National Security, LLC, for the National Nuclear Security Administration of the US Department of Energy under Contract No. 89233218C