# Chlorine Validation through Critical Integral Experiments

Eric Aboud[1],* Allan Krass,* Ruby Araj,* Catherine Percher,* Daniel Siefman,* David Heinrichs,* and Theresa Cutler†

** Lawrence Livermore National Laboratory, 7000 East Ave., Livermore, CA, 94551, aboud3@llnl.gov*
*† Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM*

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## INTRODUCTION

An increased interest in the validation of chlorine nuclear data, specifically $^{35}Cl$, in recent years has led to the prioritization of conducting chlorine critical experiments for nuclear data validation and criticality safety evaluations. A previous experiment was conducted with PVC and CPVC (chlorinated polyvinyl chloride) [1] to reduce the margin of subcriticality for PF-4 operations with aqueous plutonium chloride solutions. Challenges in determining the composition of specific polymers, namely the CPVC, made it difficult to characterize, and therefore benchmark [2].

Additional materials that were utilizable for chlorine validation were not immediately clear, as form, strength, purity, and composition are all important. A series of possible absorber materials were identified and studied, but ultimately granular sodium chloride was chosen.

In addition to the needs of LANL's PF-4 other industry collaborators have brought attention to the need for chlorine validation. Specifically, the need for HEU electrorefining with LiCl salts at Y-12 was taken into consideration for this experiment, with the experimental configurations being finely tuned to best meet their needs. Needs for validation were also presented by Idaho National Lab (INL) and TerraPower for validation of their Molten Chloride Reactor Experiment (MCRE) and Molten Chloride Fast Reactor (MCFR) and have also been considered.

## EXPERIMENTAL CONFIGURATIONS

Five experimental configurations were identified [3], but later down selected to three experiments to be executed. These configurations build upon the TEX-HEU design [4]. Two of the five experiments utilized a *sandwich* configuration which places the absorber material between two layers of moderator to create a flux trap. The three other configurations utilized a *standard* configuration which placed the absorber material directly between the moderator material and the HEU.

All five configurations were finely tuned to the Y-12 sensitivity profiles and utilized the G-parameter [5] as the similarity metric. Essentially the two sensitivity profiles are maximally similar as G approaches zero. Comparisons to the INL/Terrapower upset conditions were made, but the spectra were not tuned to meet these needs.

The three down selected configurations were 6, 8, and 18 layers which have interstitial moderation of 27/16", 7/4", and 1/8", respectively. The 6-layer and 8-layer configurations are similarly thermal, with each having roughly 63% thermal fission fractions. The 6-layer configuration has a G value of 0.009 in comparison to the Y-12 7e case, and the 8-layer configuration has a G value of 0.036 in comparison to the Y-12 8a case (see Figure 1).

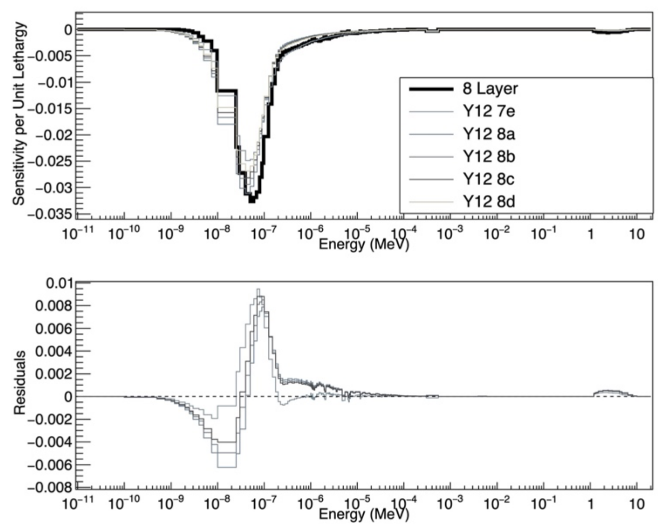


*Figure 1: Sensitivity profile for the 8-layer standard configuration compared to the Y-12 upset case sensitivity profiles.*

The 18-layer configuration, which has a 51.78% intermediate fission fraction and 34.56% fast fission fraction, has a poor similarity metric to the Y-12 sensitivity profiles but maximizes the sensitivity to the $^{35}Cl(n,p)$ reaction, which is the predominant cross section at higher energies (see Figure 2). This is intended to address the nuclear data need for validating this reaction as well as possible for this system. More sensitive configurations to this reaction have been identified and are in the works.

[1] Email: aboud3@llnl.gov

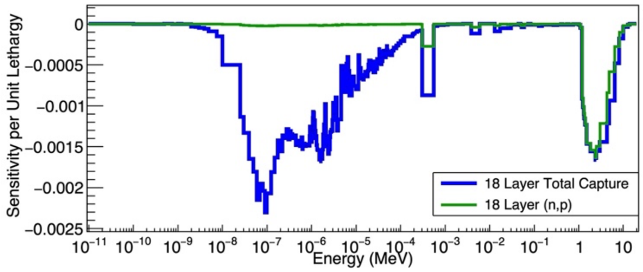

*Figure 2: Total capture sensitivity profile compared to the contribution from the (n,p) reaction for the 18-layer configuration.*

### The Needs of TerraPower and INL

Five sensitivity profiles for upset conditions for MCRE were provided by the criticality safety group at INL. While the main goal of this project was to address the needs of Y-12, the configurations were compared to the needs of INL. Using the same G parameter, the 6-layer configuration had a similarity metric value of 0.211 and the 8-layer configuration a value of 0.134 (see Figure 3). While not ideal, these will provide some value to INL and TerraPower. It should be noted that these needs are largely dependent on the $^{35}Cl(n,p)$ reaction at the higher neutron energies.

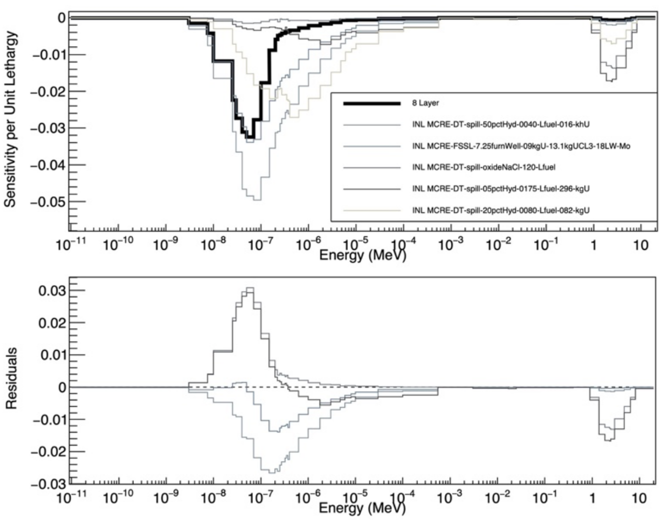

*Figure 3: 8-layer standard configuration compared to the INL upset sensitivity profiles.*

## SODIUM CHLORIDE PLATES

The experiment hinges on the fabrication of characterizable sodium chloride plates. The form of the absorber is granular high-purity, lab-grade NaCl salt. Novel containers have been designed and fabricated to facilitate the absorber materials. Thirty-six containers have been fabricated and filled with the salt. This is because the 6- and 18-layer configurations use a 3/16"-thick NaCl absorber while the 8-layer configuration uses a 1/4"-thick NaCl absorber, plus extras in the event of fabrication issues. The general procedure for filling simply involved using a vibrational packer to forcibly minimize the space between the granular salts, effectively maximizing the packing fraction and therefore maximizing the density relative to the theoretical density. Of course, in granular form the density cannot converge to the theoretical density of 2.16 g/cc, but an average density of about 1.4 g/cc was achieved, almost identical to the density assumed possible and used for the experimental design calculations. To account for possible variability in the density from fabrication, estimates vs actuality, the experimental design report estimated the effect on the neutron multiplication factor if the density was anywhere from 1.217 and 2.16 g/cc, so any differences in the real density were already understood.

Density variability within the plate was also studied. Assuming the plate would have a gradient in the salt densities, since it is vibrationally packed on its side, planar density gradients were formed within the plate in the model, conserving total mass. This analysis showed that even in the event of a very steep density gradient in all of the plates, one end has a density of 1g/cc and the other end a density of 1.9 g/cc, the neutron multiplication factor does not have a significant enough change to warrant any safety concerns. While a very shallow gradient in the density may exist in the plates, it is not expected that it would be great enough to observe or have any impact on the experimental results.

### Fabrication

The NaCl plates were fabricated via a subcontractor by LANL and were filled by staff at LLNL. The filling procedure required that the salt be baked overnight before the filling took place, this ensured that any moisture would be driven off. The seal of the plates is a metal-on-metal contact, which does not preclude moisture accumulation, but was deemed necessary in order to prevent bowing due to the change of pressure between Livermore and the Nevada National Security Sites. All plates were filled using the same procedure and almost all densities fall within the average plus 1 sigma band, see Figure 4 for one set of plates. As anticipated, there was a small accumulation of mass between the date they were fabricated and the date they were shipped to Nevada, this is due to moisture uptake. Figure 5 shows the change in density over that time period for the same set of plates shown in the previous figure. Two plates have noticeable changes to their density over the period of time and will not be used in the experiment. A previous study on a prototype encapsulation showed that moisture uptake was a small effect over a period of months, and this trend is expected for these plates as well.

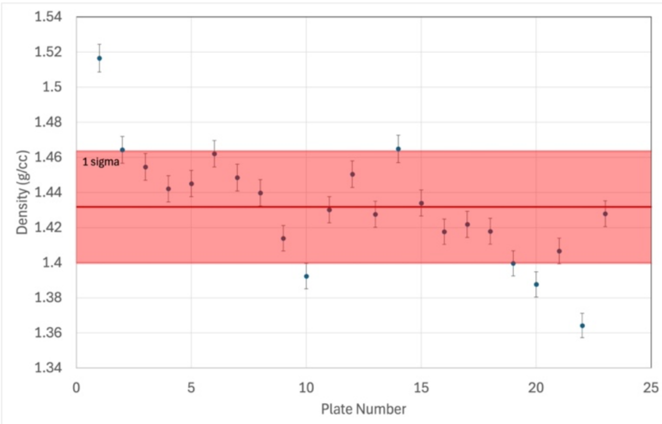


*Figure 4: Density variation for one set of filled NaCl plates.*

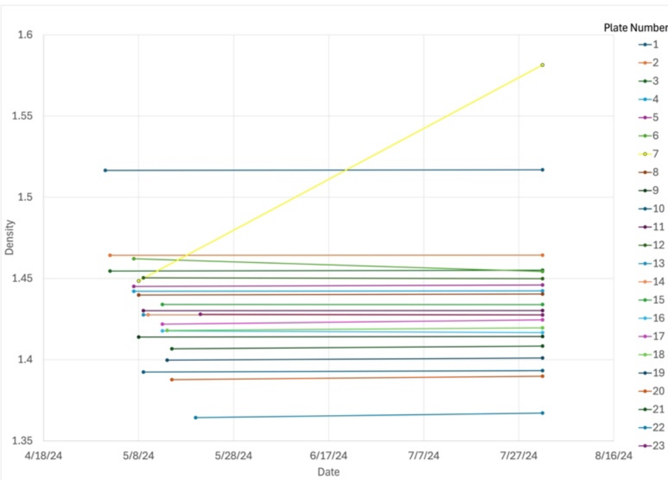


*Figure 5: Density change between fabrication and the time of shipment to Nevada. It is likely that the increase is due to water uptake of the salt. Two noticable outliers exist and will not be used during the experiment.*

Even though the densities were in very good agreement with one another, the density variation was studied further to determine which parameter caused the greatest variability. Figure 6 and Figure 7 show the variability in the salt mass and plate volumes, respectively. It is obvious, based on this analysis, that the small variability is mostly due to the salt mass that was put in each plate. The filling procedure specified that the technician would fill the plates as much as possible, independent of the total salt mass.

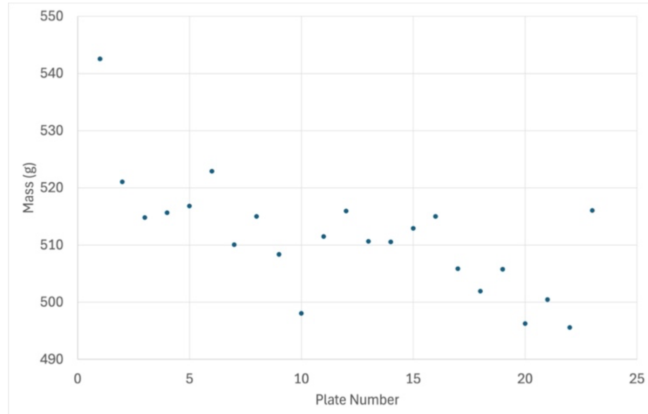


*Figure 6: Variability in the salt mass inserted into the plates for a single set of plates.*

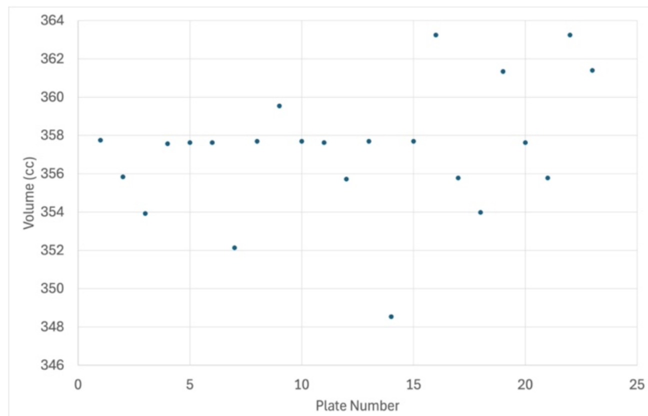


*Figure 7: Variability in the volume of the plates, of the same set, via CMM measurements.*

## MEETING THE NEEDS OF INL/TERRAPOWER

The needs of INL and TerraPower are still left open, for the most part with the completion of this experiment, as well as the mounting nuclear data need for validation of the $^{35}$Cl(n,p) cross section. Substantial work is ongoing, in conjunction with these partners, to meet these needs. Additional configurations with a novel approach to absorber design have been identified and are the process of approval for execution. With these comes additional configurations that can provide great validation of the $^{35}$Cl(n,p) cross section.

## CONCLUSIONS

The TEX with chlorine absorbers variation is a highly anticipated set of experiments set to meet the mounting needs of our industry and laboratory partners for a variety of fabrication, processing, and advanced reactor needs. Three configurations are planned for experimental execution between the time of this summary and the ANS winter meeting. These configurations directly address the needs presented by Y-12 while also providing validation to INL/TerraPower for their needs and touches on validation of the $^{35}Cl(n,p)$ cross section. This benchmark, if accepted into the ICSBEP, would be only the second chlorine validation benchmark, and be the first with well characterizable materials.

The necessary components, the most crucial the new absorber plates, have been fabricated and shipped to NCERC. The fabrication process for these plates was highly successful with little variability in the final salt densities. Experiment execution is planned in the near future, and it is expected to be ready for ICSBEP submission in the next fiscal year.

TEX with chlorine absorbers is an ongoing effort to meet the many needs of the community. Additional configurations have already been identified, using novel absorber designs, to not only meet the needs of INL/TerraPower but provide the crucial validation of the $^{35}Cl(n,p)$ cross section that our nuclear data partners need.

## ACKNOWLEDGEMENTS

This work was supported by the United States Department of Energy (DOE) Nuclear Criticality Safety Program (NCSP), which is funded and managed by the National Nuclear Security Administration for the DOE. This work was performed under the auspices of the DOE by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. This document has been released as External (Unlimited) under LLNL-ABS-866009.

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