Molten Salt Reactor Campaign FY25 Report

Analysis of Retained Voids in Molten Salts by Novel Image Processing Techniques

S. Scott Parker

Hannah K. Patenaude

Alexander M. Long

Marisa J. Monreal

March 2025

Los Alamos
NATIONAL LABORATORY

Prepared for: U.S. Department of Energy/National Nuclear Security Administration, Molten Salt Reactor (MSR) Campaign

Author Affiations: S. Scott Parker, Ph.D., Scientist, LANL, Sigma Division
Hannah K. Patenaude, Ph.D., Post Doctoral Reasearch Associate, LANL, Chemistry Division
Alexander M. Long, Ph.D., Scientist, LANL, Materials Science and Technology Division
Marisa J. Monreal, Ph.D., Scientist, LANL, Chemistry Division

Derivative Classifier Review
Reviewed and determined to be UNCLASSIFIED.
LA-UR-25-23337

Los Alamos NATIONAL LABORATORY

Contents

Figures

1 Executive Summary

The mission of the U.S. Department of Energy – Nuclear Energy (DOE-NE) Molten Salt Reactor (MSR) Campaign is to develop the technological foundations to enable MSRs for safe and economical operations while maintaining a high level of proliferation resistance. The DOE-NE MSR Campaign serves as the hub for efficiently and effectively addressing, in partnership with other stakeholders, the technology challenges for MSRs to enter the commercial market. The MSR Campaign supports experimental and computational efforts to characterize molten salt systems relevant to reactor operation, safety, and maintenance, including, but not limited to, thermophysical properties measurements of actinide (An)-bearing salts. The inherent challenges of measurements in high temperature, corrosive molten salt environments require meticulous preparation for carefully planned experiments to provide reliable data that can support MSR design, computational models, and licensing.

A key aspect of this work is the evaluation of data quality and overall measurement uncertainty reported with new measurements. In recent LANL neutron radiography experiments it was found that the retention of large, irregular void complexes (bubbles) in plutonium-bearing liquid chloride salts introduced significant measurement uncertainty. This report summarizes work to apply novel image processing techniques to quantify these voids so they can be accounted for in the accurate calculation of liquid density. More broadly, this work enables accurate calculation of the density of liquid salts occupying complex geometries. This analysis significantly improves the confidence and reliability of property measurements on liquid salts which contain a significant volume of entrained gas. This result is useful to the MSR Campaign and the advanced reactor community as it enables certification of material performance data relevant to the regulation and licensing of nuclear reactors.

2 Introduction

The thermophysical properties and high-temperature stability of molten salts have been the subject of intermittent study and review, with renewed interest in recent years in fluorides and chlorides for use in molten salt nuclear reactors or as a primary heat transfer fluid. Los Alamos National Laboratory supports the MSR Campaign by targeting specific property measurements relevant to the design and licensing of molten salt reactors. Research in this area is especially important given the limited availability of reliable data in the published literature. During a recent experiment at the Los Alamos Neutron Science Center (LANSCE), the density of binary and ternary plutonium-bearing liquid chlorides ($NaCl$-$MgCl_2$-$PuCl_3$ and $NaCl$-UCl_3-$PuCl_3$) was determined by measuring volumetric expansion as a function of temperature and composition, via neutron radiography of a sealed volumetric dilatometer (containing a column of liquid salt). The result of this work demonstrated that elevated concentrations of $PuCl_3$ in these ternary systems can lead to significant retention of void spaces (bubbles) in the liquid column, even after repeated cycles of melt and solidification. Although gas bubbles were observed in our previous work on uranium-bearing salts, none were observed in our measurements of the relevant binary salts ($NaCl$-$MgCl_2$, $NaCl$-UCl_3, and $MgCl_2$-UCl_3). While this finding has interesting implications for fluid flow characteristics of fuel salt systems (as well as the transport of volatile fission products), it complicated image analysis and increased overall uncertainty. In order to address these issues, we developed a novel image reconstruction approach and applied it to these data sets in order to (1) determine the change in volume of complex liquid geometries, and (2) establish the density of the liquid salt as a function of temperature and composition.

Typical radiographic methods to visualize 3D objects, such as computed tomography (CT) or computed stack-projections, were not possible due to the lack of multi-axis imaging capabilities at the time of this experiment. The advantage of multi-axis imaging is that the exact location of the feature may be determined, whereas depth information is limited in 2D projections. However, when the bubble size approaches the resolution limit, or when the relative contrast difference across a boundary is subtle, significant error propagates through discrete void volume calculations. This effect is especially pronounced in samples with numerous small, networked features. The void space may be estimated directly from the change in grey scale values across 2D images, although this requires calibration of the grey scale values with a feature of known geometry. Fortunately, the position data afforded by CT is not actually needed for volumetric measurements such as ours. This is because, as long as a *portion* of the liquid column is *completely filled*, the *total void space* can be determined by <u>image subtraction of any portion which contains voids from any portion that does not.</u> A completely filled segment was defined by comparing the grey scale values across a horizonal line of pixels with that of the rows above and below. Given that the filled region was represented by a few thousand rows, the typical distribution in gray value as a function of x-position could be determined. Once the standard deviation in the grey scale value at a specific x-position was known, a reduction in grey scale value across the y-column was apparent. In sum, an image with artifacts and bubbles was subtracted from an equivalent reconstruction which was completely filled. The result of this process is that the total volume and density of the liquid was determined. This process is depicted in Section 4 in a step-by-step explanation using an example sample as a case study.

3 Density Measurement of Molten Salts by Neutron Radiography

Building on the success of previous work on the neutron radiography of uranium-bearing molten salts at LANSCE, an improved apparatus was developed for plutonium-bearing molten salts, which minimized sample volume, optimized the necessary attenuation through the sample, and reduced neutron scatter on the furnace components. The apparatus was capable of measurements up to 1250 K with an imaging resolution of $\sim 50\mu m$. Sealed inner crucibles enabled measurement on air-sensitive, hazardous materials, and the use of secondary containment enabled rapid sample changes. The large, multi-zone furnace provided consistent heat across the sample assembly and doubled as a tertiary barrier. The volumetric expansion and density of ternary liquid chlorides ($NaCl$-$MgCl_2$-$PuCl_3$ and $NaCl$-UCl_3-$PuCl_3$) was measured as a function of temperature and composition by neutron radiography of a sealed volumetric dilatometer (containing a column of liquid salt). The apparatus consisted of sets of two orbitally welded 304 stainless steel Swagelok tubes held in place by a central reference guide, all of which was encapsulated in a conflat-sealed secondary container. This double containment strategy provided the safety basis for shipment and testing of plutonium-containing salts. A new imaging setup was designed and build using a $200\ \mu m$ ZnS:LiF scintillator, 50mm lens (Milvus, ZEISS, Germany), and a high-speed high-resolution charge coupled device (CCD) astronomy camera (ASI2600mm, ZWO, Suzhou, China). In this configuration, the imaging assembly could produce $\sim 50\ \mu m$ resolution images in <1 min exposures. To obtain high quality neutron attenuation images for each sample pair, radiographs without the samples in the beam, known as "flat images", were recorded before and after samples were heated. Additionally, images without the beam, known as "dark images", were recorded periodically throughout the measurements to account for thermal back-ground noise in the CCD readout chip.

3.1 Identification of Retained Voids (Bubbles)

Radiography of various binary molten salts in previous measurements demonstrated occasional bubble formation in the liquid. This could usually be resolved by increasing the temperature in order to reduce surface tension, thereby releasing the bubble into the plenum. In samples with >10% $PuCl_3$, voids formed during melt remained stable up to 900 ºC. This is shown in **Figure 1**, where the $PuCl_3$-NaCl eutectic salt mixture exhibited voids throughout the liquid column at 825 ºC. Retained voids in the $PuCl_3$-NaCl salt sample are circled in red. Analysis of the sample volume is further complicated by overlapping heater coils and circular retaining clips. These features complicated image analysis and required a different approach to determine the sample volume. In *Section 4*, a new image processing method is described and applied in a step-by-step explanation in the case of this eutectic sample.

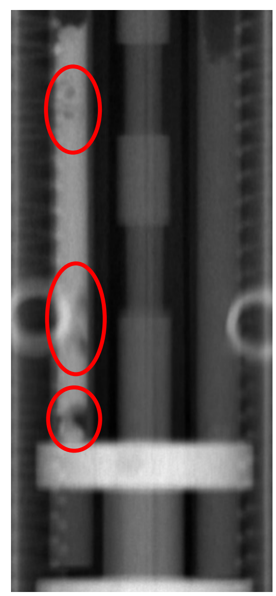

Figure 1: Neutron radiograph of $PuCl_3$-NaCl salt samples. Significant void space is observed in the liquid column in the left sample.

4 Analysis of Retained Voids (Bubbles) in Molten Salts by Novel Image Processing Techniques

The gray scale value of each pixel across the sample corresponds with the total neutron interaction volume, which in turn is related to the volume, composition, and density of the sample. Mapping an exact relationship between gray value and salt depth would require calibration with known volumes. However, such depth information is not necessary in order to calculate the void volume. What follows is a step-by-step explanation of a method for the determination of void volume by image subtraction. In effect, any completely filled portion of the sample may be used to set the relationship between gray value and depth for a given x-position. So long as any void region is subtracted from an equivalent filled region, artifacts such as the tube wall through thickness are removed. **Figure 2** shows a general overview of this process, with the original image on the left, and an example of an equivalent reconstruction on the right. In the following sections, this process is broken down into:

1. Create sample image stack
2. Perform artifact subtraction via radial symmetry
3. Create equivalent height reconstruction
4. Image subtraction of the artifact subtracted sample image from the equivalent height reconstruction

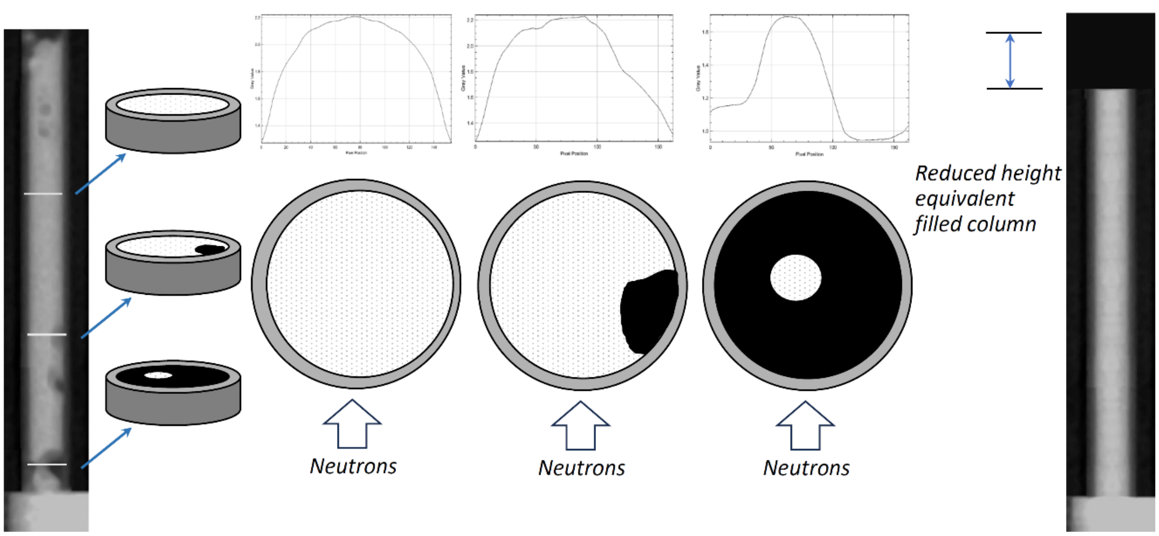

Figure 2: Schematic illustration of image reconstruction based on gray scale distribution. Three rows of pixels are shown as examples to illustrate how the void space translates to a reduction in gray value. The reconstruction on the right is based on the gray scale distribution as measured in the filled portion of the sample tube.

4.1 Image Stacking

To obtain high quality neutron attenuation images for each sample pair, radiographs without the samples in the beam, known as "flat images", were recorded before and after samples were heated. Additionally, images without the beam, known as "dark images", were recorded periodically throughout the measurements to account for thermal background noise in the CCD readout chip. To produce a final image of the sample, 50 images (1-minute exposures) were summed in ImageJ. ***The advantage of summing a stack of images is that it improves boundary resolution and produces consistent contrast across the sample***. The summed flat and dark images were subtracted from the sample image to produce the low noise radiographs shown in **Figure 3**. **Figure 3** consists of a set of 4 radiographs taken at different heights to cover the entire sample volume. Stitched images were taken at all isotherms to verify integrity of the inner container and to account for any residual salt which was not incorporated in the liquid column.

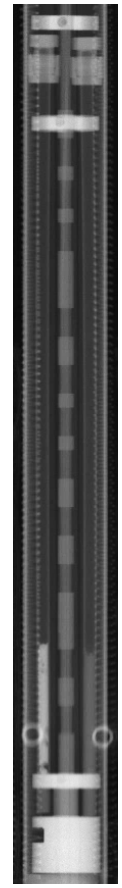

Figure 3: Stacked and stitched radiographs of two $PuCl_3$-NaCl samples at 825 ºC.

4.2 Artifact Subtraction

Image artifacts, such heating elements, complicate the analysis and must be subtracted from the original image. Artifact subtraction relies on the radial symmetry of the sample, and is only possible because the total occlusion is small and isolated to the left side. The color heat map shown in **Figure 4** emphasizes how the coils blur the sample tube boundary. Given the uniform distribution in gray values along any portion of the filled column, the left boundary may be reconstructed by mirroring the right portion of the image across the vertical centerline of the sample (red dashed line). The effect of this transformation is shown in the heat map on the right, where the voids are preserved but the coil features are removed.

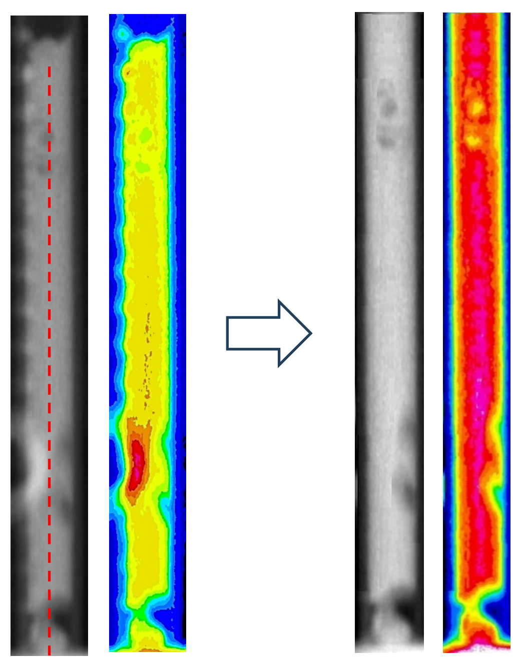

Figure 4: Sample radiograph before and after artifact subtraction. Color heat maps serve as a visual aid to highlight artifacts and voids.

4.3 Equivalent Reconstruction

An equivalent reconstruction is a hypothetical sample that represents a consistent distribution of gray values across any row, as derived from any portion of the sample which is completely filled. In **Figure 5**, this region is highlighted by a dashed red box. The purpose of this image ghost is to allow comparison of every row of the artifact subtracted image to be compared to, and subtracted from, a filled row. The variation in gray value is taken into account from the standard deviation of gray values of all pixels in a given column (i.e. at a specific pixel number across the x-axis)**. By this method, the 2σ is approximately 1.6%.** That is to say, 95% of all gray values in a column are within 1.6% of the average gray value of that column. This small difference is likely due to minor imperfections in the wall thickness of the sample tube.

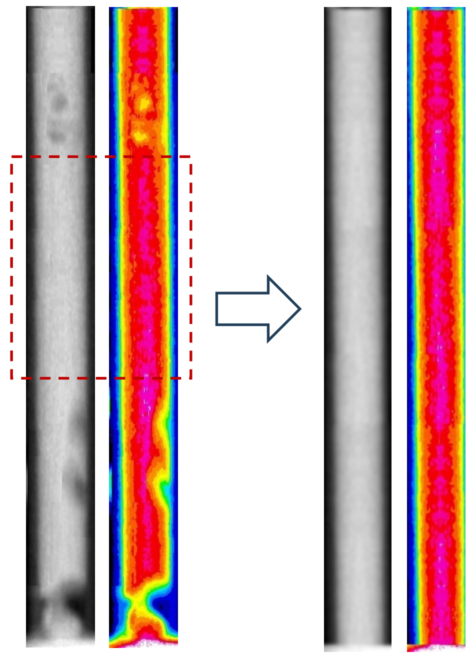

Figure 5: Sample reconstruction on the right is produced from the distribution in gray value from the filled portion of the artifact subtracted image (boxed region). Color heat maps serve as a visual aid to highlight artifacts and voids.

4.4 Image Subtraction

Using the image subtraction feature in ImageJ, the volume of the liquid may be determined. This process is shown with color heat maps in **Figure 6**. Since the mass of the sample was measured prior to the experiment, the density may be determined as a function of temperature and composition.

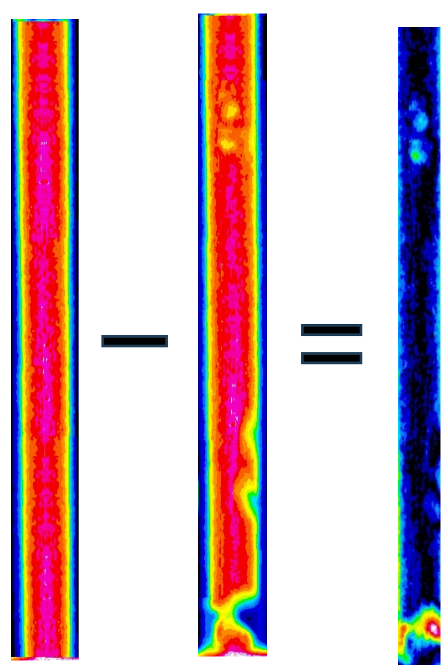

Figure 6: Subtracting the artifact corrected image from the equivalent reconstruction reveals the void volume. Color heat maps serve as a visual aid to highlight voids.

5 Conclusions

The major advantage to this novel technique is that no essential volumetric information is lost. This is a lower complexity strategy to determine density of complex shapes with high fidelity. While it is possible to get even more information from neutron computed tomography (CT) (such as the location in terms of depth into or out of a 2D radiograph, and exact shape of the bubble), this is not essential for the material property of current interest, density of molten salt. This method will be applied to all plutonium-molten salt radiography data sets that contain complex features, and a peer-reviewed manuscript summarizing that work is expected to be submitted for publication in a peer-reviewed journal in 2025.

6 Acknowledgements

The authors would like to acknowledge support from the Chemistry and Sigma Divisions at the Los Alamos National Laboratory. The experimental work described in this report was performed, in part, at the Los Alamos Neutron Science Center (LANSCE), a NNSA User Facility operated for the U.S. Department of Energy (DOE) by Los Alamos National Laboratory (Contract 89233218CNA000001). This work was funded by the US Department of Energy – Nuclear Energy (DOE-NE) Molten Salt Reactor (MSR) Campaign.