Technical Challenges in the Implementation of

Particle Image Velocimetry Measurements for Molten Chloride Salts

N. LeFrançois¹, V. Lamenta¹, M. Tetrault-Friend¹

¹McGill University, Montreal, Quebec, Canada (noah.lefrancois@mail.mcgill.ca)

A Master's Level Submission

Molten salts have garnered interest as heat transfer fluids in Gen IV nuclear reactor designs, concentrated solar power, and thermal energy storage due to their high operating temperatures and low cost. However, there is a lack of experimental data for radiation-induced convection mechanisms in these fluids, introducing a significant source of uncertainty into designs. Particle image velocimetry (PIV) has recently been implemented for flow visualization in nitrates, which are of interest in intermediate heat transport systems and thermal energy storage. Several challenges still need to be addressed in order to extend this technique to molten chlorides, which are more chemically aggressive but are of greater interest as fuel carrier and coolant salts. Chlorides are optically semi-transparent, resulting in a strong coupling of radiation and convection at high temperature. This paper outlines ongoing work in the implementation of this technique, including optical window material compatibility and salt selection.

1. Introduction

Understanding and quantifying the effects of turbulence and radiative heat transfer in molten salt is essential to the design of molten salt reactors (MSRs). A major obstacle in this field is obtaining accurate and reliable measurements of thermal hydraulic data such as flow velocity and temperature profiles. Standard methods for making these measurements rely on invasive techniques like hot-wire anemometry and thermocouple probes, which are challenging to implement in molten salt experiments due to the high operating temperatures and corrosive chemical environment.

Particle Image Velocimetry (PIV) is a non-invasive optical technique for two-dimensional velocity profile measurements. PIV is widely used in the field of experimental fluid mechanics [1] and recent work has even produced simultaneous velocity and temperature measurements in high-temperature gas flows [2].

However, the technique has largely been avoided in past studies of molten salts due to concerns about the difficulty of optical measurements in a high-temperature molten salt environment [3,4]. These studies have sought to validate CFD models using analogous experiments with water as the working fluid and then simulating the flow behaviour of interest numerically by replacing the thermophysical properties of water with those of molten salt. While this technique has had success and greatly reduces experimental complexity, it introduces significant uncertainty into models of molten salt nuclear reactors because water does not exhibit the complex interactions between turbulent natural convection and radiative heat transfer that is present in high-temperature semi-transparent media [5].

PIV measurements were recently implemented for the first time in molten salt, using a binary nitrate mixture of NaNO3-KNO3 (60-40 wt%) [6]. This mixture is known commercially as Solar Salt and is widely used in concentrated solar power and thermal energy storage applications [7], although it has also been employed as a heat transfer fluid in intermediate heat transport systems for molten salt reactors [8]. Solar Salt has relatively low operating temperatures (220°C-550°C) and low corrosion rates compared to the chloride and fluoride salts commonly used as fuel carrier and coolant salts in molten salt nuclear reactor designs [9]. These properties were favourable for the development of an initial proof-of concept experiment, but further work is required to extend the technique to a wider range of salt mixtures.

2. Optical Properties of Molten Salt

Although the optical thickness of molten salt is a key parameter in in design problems involving coupled radiation-convection flow, the accurate measurement of absorption and scattering coefficients for molten salt mixtures of interest is still an area of work in its early stages of development. Existing techniques often require small sample thicknesses (<=10mm) or do not provide accurate measurements at high temperatures [10]; both constraints are problematic for the application of this data to nuclear technology.

Tetreault-Friend et al. [10] developed an optical measurement technique capable of measuring absorption coefficients in Solar Salt and in a binary chloride mixture (NaCl-KCl 50-50 wt%) over a broad spectral range (400nm-8um). The Solar Salt mixture was found to be optically thick in its re-emission spectrum, while the NaCl-KCl mixture is optically semi-transparent; both mixtures have negligible scattering coefficients. For comparison, the re-emission weighted absorption coefficient of each mixture was calculated from this data by Lamenta et al., finding values of 3700m⁻¹ for Solar Salt at 400°C, 0.7m⁻¹ for NaCl-KCl at 800°C, and 8x10⁵m⁻¹ for water at 25°C [6].

Kim & Yun [11] also developed an optical technique for measuring absorption coefficients in molten salt in order to study the effect of lanthanide concentration on the absorption spectra of a LiCl-KCl eutectic mixture. While their data for the absorption coefficient of pure LiCl-KCl (44-56 wt%) covers a narrower spectral range and is not as precise, it provides a valuable point of comparison between the optical properties of the LiCl-KCl and NaCl-KCl mixtures. Qualitatively these salts have similar optical properties, with an absorption edge around 200nm below which the salt is optically thick, and above which the optical thickness is semi-transparent. As will be discussed in the next section, these two mixtures have been identified as candidates for PIV experiments and this optical data will be essential in designing and analysing these experiments.

3. Chloride Salt Selection

Molten chloride salts have been considered in several recent molten salt reactor designs, as both a fuel carrier and coolant [12]. They are also of interest as an ionic liquid electrolyte in pyrochemical processes for recycling used nuclear fuel [11].

NaCl-KCl mixtures are often favoured in concentrated solar power applications, while a variety of mixtures are used in nuclear applications including fuel salts such as uranium or plutonium chloride and "clean" (i.e., non-fissile) salts such as LiCl-KCl [12]. In view of the wide range of relevant salt mixtures, the selection of an initial candidate for the implementation of PIV experiments is an open-ended question.

As discussed in Section 2, optical measurements have been successfully conducted in both NaCl-KCl and LiCl-KCl salts using quartz viewing windows. These candidates have similar optical properties and are therefore expected to display similar flow behaviour in regimes where coupled radiation-convection heat transfer is important. Additionally, both mixtures have negligible vapor pressure below temperatures of approximately 1100°C [9], which is well above the expected operating temperatures in these PIV experiments and in most nuclear and solar applications. Low vapor pressure is a desirable attribute for PIV measurements as well as CSP applications in general, as both cases favour an open-air design in which the top surface of the salt is exposed to the ambient atmosphere instead of the inert atmospheres used in most nuclear applications and thermophysical property experiments to control unwanted reactions of the salt with air and water.

The main factors influencing the decision between these two candidates are operating temperature range, availability of optical property data, and optical window compatibility. The lithium chloride mixture has a melting point of 353°C, while the sodium chloride mixture has a much higher melting point of 657°C; both mixtures have a boiling temperature over 1000°C [9]. The sodium chloride mixture's higher melting point increases experimental complexity since more heating power is required and the losses through an optical window will be more significant, perturbing the natural flow we are trying to observe. The lithium chloride mixture would simplify the thermal design of a PIV experiment and reduce uncertainty in the flow boundary conditions, while still being able to operate at temperatures high enough for radiative heat transfer to be significant.

Conversely, the optical property data available for the sodium chloride mixture covers a wider spectral range with higher precision than the data available for the lithium chloride mixture; this will likely have an impact on the uncertainty introduced into simulations of radiative heat transfer in the salt by optical data. The third major factor in salt selection, optical window compatibility in open-air environments, is a topic which has not been thoroughly addressed in the literature. In Section 4, existing knowledge on the subject will be summarized and ongoing experimental tests seeking to compare the compatibility of quartz optical windows with each of the two candidate salt mixtures will be presented.

4. Optical Window Compatibility

Several experiments requiring optical access have been conducted with molten salts, mostly for spectroscopy measurements [13,14]. In order to meet this requirement, small windows made of quartz, diamond, or sapphire have been used. While this existing work provides some insight into the challenges of optical experiments with molten salt, the scale of our proposed PIV measurements is much larger than the window diameters required for spectroscopy experiments. The characteristic scales in the natural convection flows of interest are on the order of 10cm [6], while the spectroscopy measurements only require windows on the order of 10mm [14]. Increased window cost and manufacturing limitations with increasing scale favour the use of quartz windows, but the exposure of quartz to molten chlorides under ambient atmosphere instead of inert gas must be further investigated in order to inform the design of a molten chloride PIV experiment.

Fogging in quartz windows exposed to NaCl-KCl salt has been observed in the works of Tetreault-Friend et al. [10] and Passerini [15]; this damage limited the number of uses for a single window but was not rapid enough to prevent high-quality optical measurements before the fogging became too severe. Li and Dasgupta [16] had previously observed this phenomenon, proposing that the damage is due to the physical diffusion of alkali ions into the silica matrix of the quartz.

In order to inform design choices in salt selection and window material for open-air PIV experiments, immersion testing of quartz samples is being conducted in both chloride mixtures of interest (LiCl-KCl and NaCl-KCl binary eutectic mixtures). Figure 1 shows the immersion testing setup. A 25mm square slide of fused quartz was fully submerged in a 100g sample of each salt for 2 hours in order to simulate the length of exposure required for PIV measurements. The lithium mixture was held at 500°C, while the sodium mixture was held at 800°C. After 2 hours, the slide was removed using metal tongs. The lithium mixture testing was conducted on a benchtop hot plate, while the sodium mixture required a larger tube furnace configuration similar to the setup of Passerini [15] in order to reach the higher melting temperature of NaCl-KCl. This setup and the higher temperatures made removal of the sodium sample more challenging, and in our initial attempt the slide suffered a stress fracture due to thermal shock during removal. To compare the effects of these chloride mixtures to the acceptable window performance observed by Lamenta et al. [6] with nitrate salts, this same test was repeated using Solar Salt at a temperature of 500°C.



Figure 1: Immersion testing of quartz slide in NaCl-KCl at 800° C (left) and LiCl-KCl at 500° C (right).

Figure 2 shows the results of initial immersion testing. While some loss of clarity was observed in the sodium sample, significantly more fogging as well as small cracks were observed in the lithium sample after removal from the salt. Despite the observed damage after removal from the salt, the lithium sample displayed no visible signs of loss of clarity or other damage during the immersion; in Figure 1 the clarity of the sample while immersed can be easily observed. This visual inspection was not possible in the sodium test, as the visible radiation emitted by the salt at 800°C made the quartz sample difficult to observe clearly.

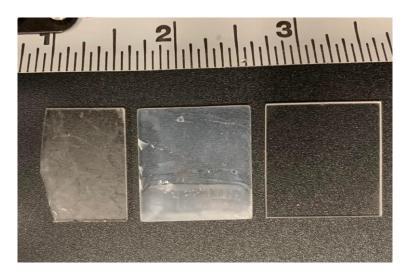


Figure 2: Comparison of 2cm square quartz slides after immersion for 2 hours in NaCl-KCl at 800°C (left), LiCl-KCl at 500°C (centre), and NaNO3-KNO3 (right).

This contrast in the lithium sample between initial clarity during immersion and severe fogging after removal and cleaning suggests that much of the damage may be due to secondary causes such as thermal shock during salt freezing or exposure to HCl formed when solid LiCl interacts with water from the atmosphere due to its strong deliquescence [17]. Replication of these immersion tests and further investigation of sample clarity before and after removal are ongoing in order to better understand the repeatability of optical measurements in hostile chloride salt environments. These tests will aid in the design of full-scale PIV measurements which avoid the need for large and prohibitively expensive diamond or sapphire windows.

5. Conclusion

Particle image velocimetry is a promising technique for non-invasive 2D velocity profile measurements in molten salts and has recently been demonstrated in molten nitrate salts [6]. The extension of this technique to molten chloride salts at higher temperatures is of great relevance to the design of molten salt nuclear reactors, but also presents increased design constraints due to the harsh chemical and thermal environment to which optical windows must be exposed. This material compatibility constraint is one of the main factors in selecting the candidate salt mixture for extension of PIV to chlorides, along with operating temperature. Initial tests indicate that NaCl-KCl is less hostile towards quartz optical windows than LiCl-KCl, but further work is required to corroborate this observation and better inform design choices in the design of full-scale PIV measurements.

6. References

- [1] M. Raffel, C.E. Willert, F. Scarano, C.J. Kahler, S.T. Wereley, J. Kompenhans, "Particle Image Velocimetry: A Practical Guide", 3rd Edition. Springer, 2018.
- [2] Fan, L. "Thermographic Particle Image Velocimetry: From Phosphorescence to Incandescence." Doctor of Philosophy, University of Cambridge, 2018.
- [3] C. Shao, J. Zhou, W. Cheng, "Experimental and Numerical Study of External Performance and Internal Flow of a Molten Salt Pump That Transports Fluids with Different Viscosities. International Journal of Heat and Mass Transfer 89, 2015.

- [4] P.R. Rubiolo, M.T. Retamales, V. Ghetta, J. Giraud, "High Temperature Thermal Hydraulics Modeling of a Molten Salt: Application to a Molten Salt Fast Reactor (MSFR)." ESAIM: Proceedings and Surveys 58, 2017.
- [5] A. Ibrahim, D. Saury, D. Lemonnier, "Coupling of Turbulent Natural Convection with Radiation in an Air-Filled Differentially-Heated Cavity at Ra=1.5×109." Computers & Fluids 88, 2013.
- [6] V. Lamenta, N. LeFrançois, J. Nedic, M. Tetreault-Friend, "Analysis of Convection-Radiation Interactions in Molten Salts Using Particle Image Velocimetry." 19th International Topical Meeting on Nuclear Reactor Thermal Hydraulics, Brussels, Belgium, 2022.
- [7] Sociedad Química y Minera de Chile, "Thermo-Solar Salts Data Sheet", 2010.
- [8] Hastings, P. Design Overview for the Kairos Power Fluoride Salt-Cooled, High Temperature Reactor, 2018.
- [9] W. Ding, A. Bonk, T. Bauer, "Molten Chloride Salts for Next Generation CSP Plants: Selection of Promising Chloride Salts & Study on Corrosion of Alloys in Molten Chloride Salts." AIP Conference Proceedings 2126:1, 2019.
- [10] M. Tetreault-Friend, L.A. Gray, S. Berdibek, T. McKrell, A.H. Slocum, "Optical Properties of High Temperature Molten Salt Mixtures for Volumetrically Absorbing Solar Thermal Receiver Applications." Solar Energy 153, 2017.
- [11] B.Y. Kim, J.I. Yun, "Optical Absorption and Fluorescence Properties of Trivalent Lanthanide Chlorides in High Temperature Molten LiCl–KCl Eutectic." Journal of Luminescence 178, 2016.
- [12] Z. Mausolff, M. DeHart, S. Goluoglu, "Design and Assessment of a Molten Chloride Fast Reactor." Nuclear Engineering and Design 379, 2021.
- [13] J. Zhang, C.W. Forsberg, M.F. Simpson, S. Guo, S.T. Lam, R.O. Scarlat, F. Carotti, K.J. Chan, P.M. Singh, W. Doniger, K. Sridharan, J.R. Keiser, "Redox Potential Control in Molten Salt Systems for Corrosion Mitigation." Review. Corrosion Science 144, 2018.
- [14] T. Nagai, T. Fujii, O. Shirai, H. Yamana, "Study on Redox Equilibrium of UO2 2+/U2 + in Molten NaCl-2CsCl by UV-Vis Spectrophotometry." Journal of Nuclear Science and Technology 41, 2004.
- [15] Passerini, S. "Optical and Chemical Properties of Molten Salt Mixtures for Use in High Temperature Power Systems." M.Sc., Massachusetts Institute of Technology, 2010.
- [16] J. Li, P.K. Dasgupta, "A Simple Instrument for Ultraviolet-Visible Absorption Spectrophotometry in High Temperature Molten Salt Media." Review of Scientific Instruments 71, 6, 2000.
- [17] E. Mullen, R. Harris, D. Graham, C. Rhodes, Z. Hodgson, "Transfer Characteristics of a Lithium Chloride-Potassium Chloride Molten Salt." *Nuclear Engineering and Technology* 49:8, 2017.