

# EXPERIMENTAL AND MODELING STUDIES OF OVER-COOLING TRANSIENTS IN FLUORIDE-SALT COOLED HIGH-TEMPERATURE REACTORS

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## ABSTRACT

The prototypical primary coolant for Fluoride-Salt Cooled High-Temperature reactor (FHR), flibe ( $2\text{LiF-BeF}_2$ ), has a freezing point of  $459^\circ\text{C}$  and a boiling point greater than  $1400^\circ\text{C}$  at atmospheric pressure. FHRs operate in the temperature range of  $600^\circ\text{C}$  to  $700^\circ\text{C}$ , so they have a significant margin to coolant boiling and a relatively smaller margin to freezing. Furthermore, the ultimate heat sink during accident scenarios is ambient air or water, both of which are far below the freezing point of salt. Therefore, overcooling transients that lead to freezing of the salt merit investigation.

The use of simulant fluids, already successfully applied in thermal-hydraulics experiments, needs the definition of the flibe change of phase phenomenology to be extended to study transients that involve freezing. In this view, the eventual distortions that the different simulants introduce into the thermal-hydraulic behavior of the system during overcooling transient have to be identified.

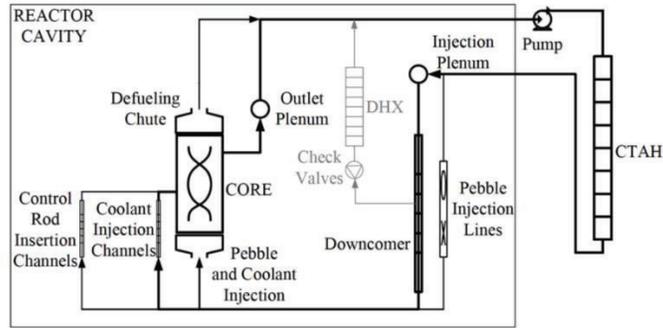
This paper provides a discussion of the up-to-date flibe equilibrium phase diagram and preliminary experimental results of non-equilibrium phenomena in flibe and simulant fluids freezing. These studies will be object of future work at University of Wisconsin Madison, together with the investigation on the impact of overcooling phenomena on FHR salt-to-air heat exchanger and on the stability of the natural and forced circulation loops, which is introduced in this paper.

## KEYWORDS

Overcooling Transients, Freezing, Flibe, FHR, natural circulation.

## 1. INTRODUCTION

Fluoride-Salt Cooled High Temperature reactor (FHR) is a reactor design that combines a high temperature and low pressure fluoride salt coolant with solid fuel elements formed of ceramic encapsulated fuel (TRISO particles) dispersed in a graphite material that provides the geometry of the fuel element [1]. The use of fluoride coolant salt allows the reactor to be coupled to Brayton thermal cycles, more efficient than steam cycles employed by conventional reactors. An air Brayton cycle employs gas turbines that are conventional technology in the natural gas power industry, and permits co-firing with natural gas for power peaking [2]. Fluoride salt coolants have excellent performance as heat transport fluids, leading to several inherent safety features for FHR and enabling economic design of passive safety systems. For example, the passive and passively-initiated emergency decay heat removal system is natural-circulation driven (Figure 1 shows the salt-to-air heat exchanger, CTAH, and the DHX, heat exchanger for the natural circulation system). In the FHR, coolant salt operates in the temperature range of  $600^\circ\text{C}$  to  $700^\circ\text{C}$ , and the freezing of the salt ( $459^\circ\text{C}$  melting point for flibe) has to be considered in specific overcooling transients.



**Figure 1. Primary coolant flow paths under normal power operation in FHR [1].**

The use of simulant fluids has already been successfully applied to study thermal-hydraulic transients of flibe ( $2\text{LiF}-\text{BeF}_2$ ) cooled reactors. Simulant fluids, allow the construction experimental facilities that operate at reduced temperature, reduced geometric scale, and reduced power with little distortion in the prototypical non-dimensional numbers [3]–[5]. The use non-beryllium-surrogate fluoride salts in experiments at prototypical scale and temperatures is also of interest, because beryllium toxicity significantly adds to the cost and complexity of experimental facilities; flinak ( $\text{LiF}-\text{BeF}-\text{NaF}$ ) is one such surrogate salt that has been used in thermal-hydraulic loops [6]. The use of simulant fluids for the study of overcooling transients has not yet been investigated. To properly assess the choice of simulant fluids and the possible distortions that they introduce in experimental systems an understanding of the equilibrium and non-equilibrium freezing phenomenology of flibe salt is necessary.

This paper summarizes initial work on the study of FHR overcooling transients, with a focus on freezing phenomenology for flibe and identification of potential simulant fluids. Section 2 reviews flibe equilibrium solidification through the phase diagram, considering the techniques used to obtain it and the uncertainty connected to the salt composition. The section continues describing non-equilibrium freezing phenomena and particularly to the effect of supercooling, seen in many fluids and expected in flibe. Section 3 presents studies performed to evaluate freezing of salt in pipes while Section 4 reports the implication of freezing in terms of natural circulation stability. Those two section are intended to introduce the possible impact that freezing can have on a component (salt-to-air heat exchanger tubes) and system scale (stability of forced and natural circulation loops).

## 2. FLUORIDE SALT FREEZING PHENOMENA

Flibe is a binary mixture of two fluoride salts,  $\text{LiF}$  and  $\text{BeF}_2$ . Two main regimes have been identified to be characteristic of the solidification process. The equilibrium freezing, described in Section 2.1, which represents the ideal case in which the solidification happens as a chain of infinite equilibrium steps and can be considered accurate to describe very slow real case change of phase. However, in many real case scenarios, freezing is influenced by other parameters, like cooling rate and availability of nucleation sites, that modify the equilibrium conditions, leading for example to supercooling effects. Some preliminary experimental evidences of these effects will be reported in Section 2.2.

### 2.1. Equilibrium Freezing

In order to characterize equilibrium freezing behavior of flibe, we review here (1) the current understanding of the phase diagram for  $\text{LiF}-\text{BeF}_2$  together with a discussion related to the discrepancy between real mixtures and the ideal flibe composition, and (2) the freezing phenomenology based on the phase diagram is then presented. We subsequently provide a discussion of the evolution of experimental methods used to generate the data for the phase diagrams.

### 2.1.1 Equilibrium phase diagram and chemical composition

Phase diagrams provide a wealth of information as a function of the composition of the salt mixture. From the point of view of flibe as coolant for nuclear reactors, the pertinent information is (1) the freezing temperature (in the case of congruent freezing) or the freezing temperature envelope between the liquidus and the solidus curves, and (2) the equilibrium solid phases that the salt forms. Figure 2 shows the LiF-BeF<sub>2</sub> phase diagram, derived by Romberger et al. at ORNL in 1972 [7].

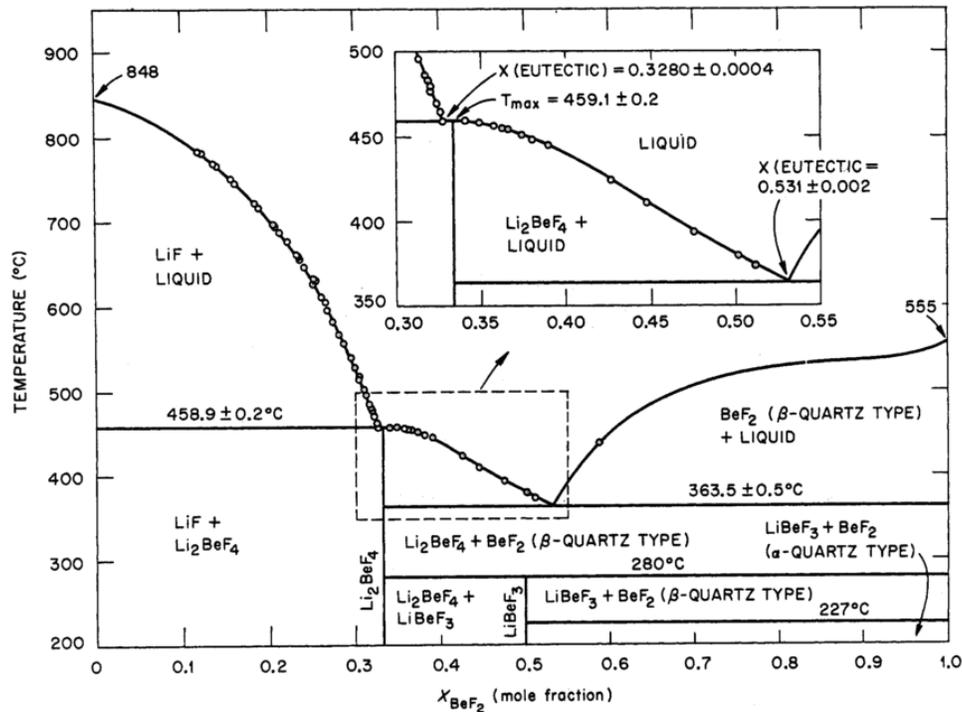


Figure 2. LiF-BeF<sub>2</sub> phase diagram [7].

The nominal composition chosen for FHR is the 2:1 mol ratio of LiF-BeF<sub>2</sub> (0.33% mol fraction BeF<sub>2</sub>). The mixture with this composition is referred to as flibe. This was the theoretical composition used at MSRE for the coolant salt used in the secondary loop and it was selected because it has a sufficiently low freezing point, 459°C, with low enough viscosity and vapor pressure. Increasing the BeF<sub>2</sub> molar fraction, the freezing temperature decreases as we approach the eutectic composition at 0.55% BeF<sub>2</sub> molar fraction; unfortunately BeF<sub>2</sub>-rich compositions have a higher viscosity and higher vapor pressure, that make those mixtures less suitable to be used as a coolant fluid [8].

Production of large-scale quantities of salt with the nominal composition is not always easily achievable. During MSRE project every batch produced had a LiF-BeF<sub>2</sub> ratio that was different from the theoretical 2:1 mol ratio. Figure 3 reports the mole fraction of BeF<sub>2</sub> measured in the different 120 kg batches produced for MSRE operations [9], the measurements performed at UWM on a batch of MSRE coolant salt, and two flibe batches (<100 g) produced at UWM from raw materials [10]. To determine lithium and beryllium content in the salt MSRE researchers used chemical techniques, and UWM used Inductively Coupled Plasma Optical Emission Spectrum. This data shows that the salt composition can differ considerably from the nominal one. UWM melts show that it is possible to produce a mixture within 1%

mol of the nominal 33 mol% BeF<sub>2</sub> composition, at the 10s kg scale, from material that is commercially available today.

The salt composition can change over the long term duration of reactor operations, due to Li and Be neutron transmutation, and dissolution in the melt of elements employed for salt chemistry and corrosion control, such as Be metal. Therefore the salt composition for an operational FHR must be considered with significant uncertainty, or as a non-constant parameter. This is an important observation, since the solidification phenomenology is sensitive to composition and the thermal-physical properties of the melt and the fused salt are likewise composition-dependent.

The presence of impurities is not expected to be sufficiently high to change the LiF/BeF<sub>2</sub> ratio of the melt. However, their presence even in trace amounts may affect the phase change behavior of the salt and its properties. The impurities (Cr, Fe, Ni) that were present in the MSRE salt were at the tens of ppm level [9]. The purification process recently reproduced at University of Wisconsin Madison (UWM) shows that is it possible to decrease impurities to the ppm level [10].

### 2.1.2 Freezing behavior of flibe

Figure 2 shows that at the 2:1 molar ratio (33% BeF<sub>2</sub>), the liquid will freeze congruently at 459.1±0.2°C, with the temperature remaining constant upon heat removal, until all the liquid has frozen. From a solidification point of view the melt at this composition behaves as a single-component salt.

With BeF<sub>2</sub> concentrations slightly lower than the nominal flibe composition, included between 32.8% and 33.3% of BeF<sub>2</sub> (Zone B in Figure 3) the solidification also happens congruently and isothermally, but the solid will form a mixture of LiF crystals and flibe crystals. The melting point in this composition region drops less than 1°C, to 458.9 ±0.2°C.

At molar concentrations of BeF<sub>2</sub> lower than the 32.8% eutectic composition (Zone A), the solidification temperature changes considerably with composition. In this region, the solidification occurs through a liquid + solid transition. Once the melt intercepts the curve LiF crystals start to form. As heat removal continues past the liquidus line, more LiF crystals are formed and the remaining liquid becomes increasingly rich in BeF<sub>2</sub>. The cooling process continues until the temperature of 458.9±0.2°C is reached; at this point the liquid has the composition of flibe and flibe crystal can be formed at constant temperature.

At molar concentrations of BeF<sub>2</sub> higher than 33.3% (Zone C), solidification occurs through a liquid + solid region too. In this case solidification begins at a lower temperature (included between 459.1°C and 363.5°C depending on the composition), as indicated from the liquidus curve. As this temperature is approached the formation of flibe crystals begin. As the cooling continues, more flibe crystals are formed and the remaining liquid becomes increasingly rich in BeF<sub>2</sub>. At ~360°C, the remaining liquid solidifies to BeF<sub>2</sub> of the beta quartz type, maintaining a constant temperature until all of the liquid is solidified. Continuing to remove heat from the solid mixture, its temperature will drop until 280 °C, at which point one last isothermal phase change occurs, with the solid restructuring to a mixture of flibe crystals, and LiF-BeF<sub>3</sub>. This freezing behavior is valid for mixture up to 50% BeF<sub>2</sub> composition.

Freezing of mixtures with lower ratio is not described since it is not interesting from a flibe composition point of view. It is worthy to remark that BeF<sub>2</sub>-rich composition results to be highly viscous and the tendency of BeF<sub>2</sub> to form glass makes the equilibrium condition difficult to be reached. This evidence was observed in different attempts to experimentally obtain phase diagrams.

Thus, in the very close vicinity of the nominal 33.3% composition, we observe four different possible types of behavior for the equilibrium solidification process, four different types of solid crystal structures, and two different kinds of liquid-solid mixtures. The implications of these observations on the thermo-physical properties, and the time-evolution of thermal hydraulic transients involving phase change will need to be investigated. Likewise, it would be relevant to understand the uncertainties associated with the liquidus and solidus lines of the phase diagram. The following section describes the experimental methods employed in generating this phase diagram, and the history of the development of this phase diagram during the MSRE project from the 1950s through 1970s, and more recent re-evaluations of the historical data.

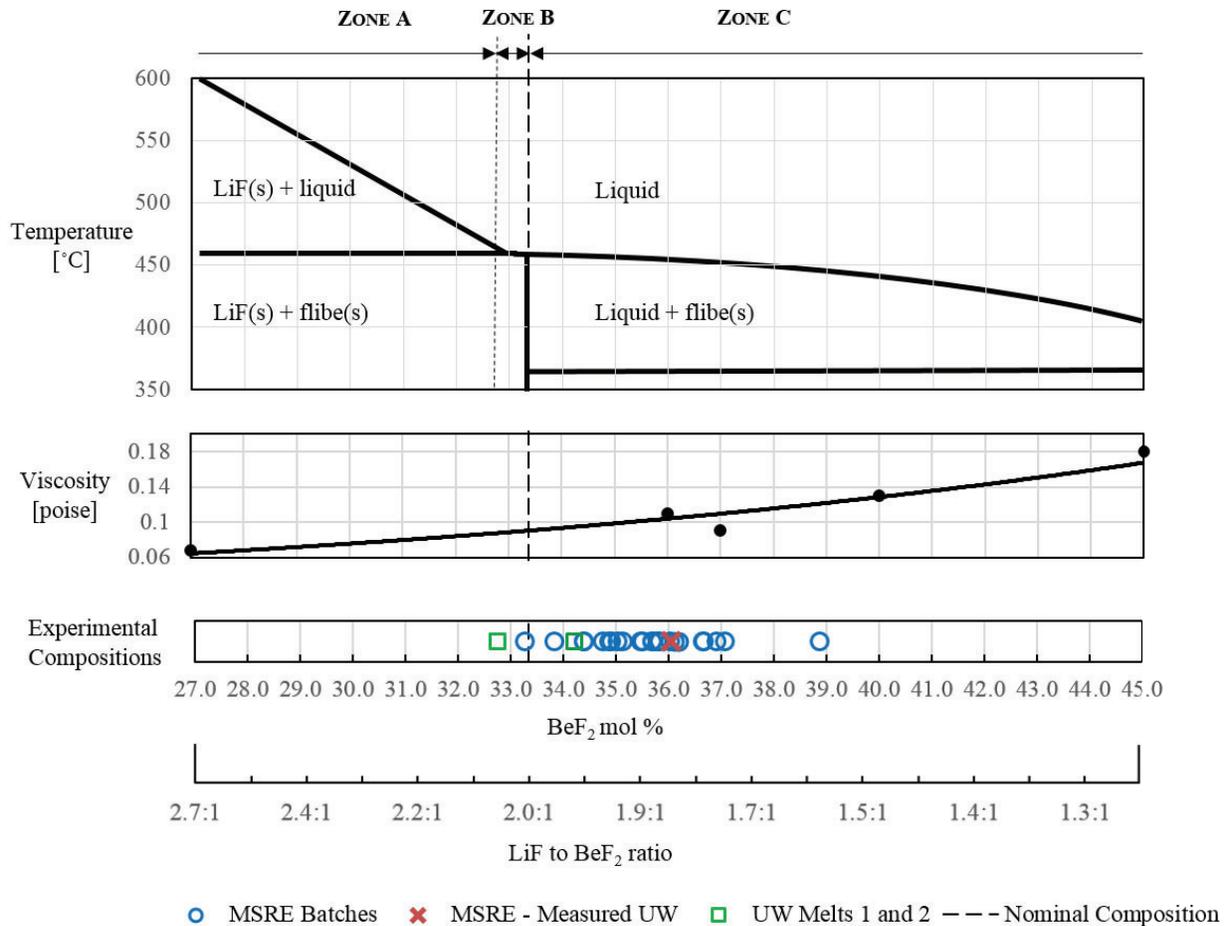


Figure 3. Liquidus curve and viscosity [11] for different salts composition with the scatterplot of different measured  $\text{BeF}_2$  molar fraction in flibe used in MSRE and UWM.

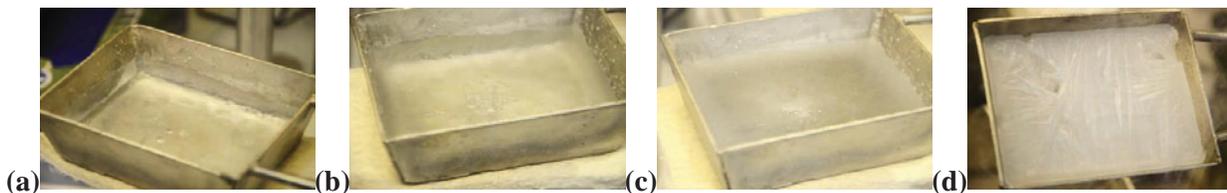


Figure 4. Flibe freezing process; liquid purified flibe with the typical clear appearance (a), first crystals formation (b)-(c); solidified flibe where large crystals are visible (d). [10]

### 2.1.3 Experimental Research at MSRE

The need to define a freezing temperature for a range of different compositions present in the reactor make the definition of an accurate phase diagram important. Since 1950s and 1960s different groups separately tried to study the equilibrium chart of the LiF-BeF<sub>2</sub> mixtures. The definition of a detailed equilibrium phase diagram and liquidus curve has resulted to be a challenging task. Before the start of the MSRE program at ORNL most of the phase diagram studies were focused on the definition of the solid crystalline phases more than an accurate definition of the liquidus curve for the LiF-BeF<sub>2</sub> mixtures. Figure 5(a) gives a summary of the different literature produced in those years. This figure clearly shows important differences among the diagrams, so marked that that only a general trend can be seen and an accurate definition of freezing temperatures for many salt compositions is impossible.

The appreciable differences among the diagrams can be attributed to two distinct kind of factors: first, water, oxygen and impurities in general considerably affect the salt phase properties; second, the phase diagram ideally represents equilibrium phases while many of the experimental study techniques require the deduction of equilibrium information from non-equilibrium experimental data.

Aware of those possible perturbations to phase diagrams, the High Temperature Equilibrium Group of the Reactor Chemistry Division at ORNL studied a large number of binary, ternary and quaternary salt mixtures to define the optimal salt to be used as a coolant in a reactor and as fuel salt to perform thermal breeding as a final goal. The generation of phase diagrams for different mixtures was performed in the 1950s and 1960s combining two experimental techniques: (1) the study of the salt heating and cooling curves and (2) the analysis of the phases obtained after a prompt quenching of the mixture. Thanks to these experimental setups the researcher at ORNL obtained a first phase diagram reported in Figure 5(b) and used for design and operations at MSRE. (3) A third more accurate technique was used in the early '70s to study specifically flibe equilibrium freezing and freezing temperature; it involves the study of electrochemical force measurements in the salt.

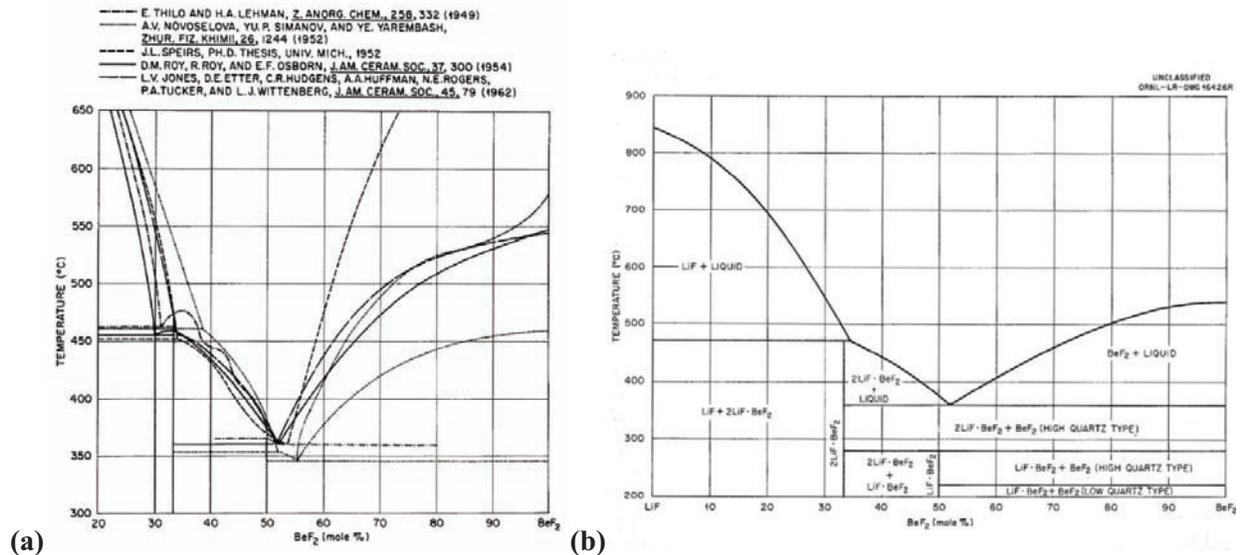


Figure 5. Comparison of phase diagrams of LiF-BeF<sub>2</sub> as reported in previous literature (a) [12]; the system LiF-BeF<sub>2</sub> as reported by Thoma in 1959 (b) [13].

Reference [14] describes the experimental setup that was used at ORNL to obtain phase diagrams from thermal analysis data. The setup, shown in Figure 6 is designed to examine at the same time four samples placed in four different nickel crucibles. A Chromel-Alumel thermocouple, embedded in a Nickel well, is placed into every crucible, and a Nickel stirrer provides the mixing of each salt mixture. The furnace that surrounds the system starts the cooling transients once a temperature of roughly 100°C above the highest estimated freezing point of the salt is reached. The cooling rate is regulated by an automatic control of the voltage of the furnace. The samples were on average 50g for every run. A special procedure, consisting of performing the whole process into a vacuum dry-box, was developed for molten mixtures that showed hygroscopic behavior. The point of phase change was determined from the the slope of the temperature time curves. This technique is reported to be very effective except in the case when the liquidus curve is particularly steep.

This second technique is based on rapidly cooling of tubes containing several samples of salt mixtures. Figure 6 shows the experimental set-up. Samples are maintained at slightly different temperatures around the presumed liquidus curve, and then rapidly quenched. Assuming an instantaneous freezing transient, microscopic observation and x-ray analysis of the phases present in the frozen salt indicates the phases present at the initial condition, before the rapid quench.

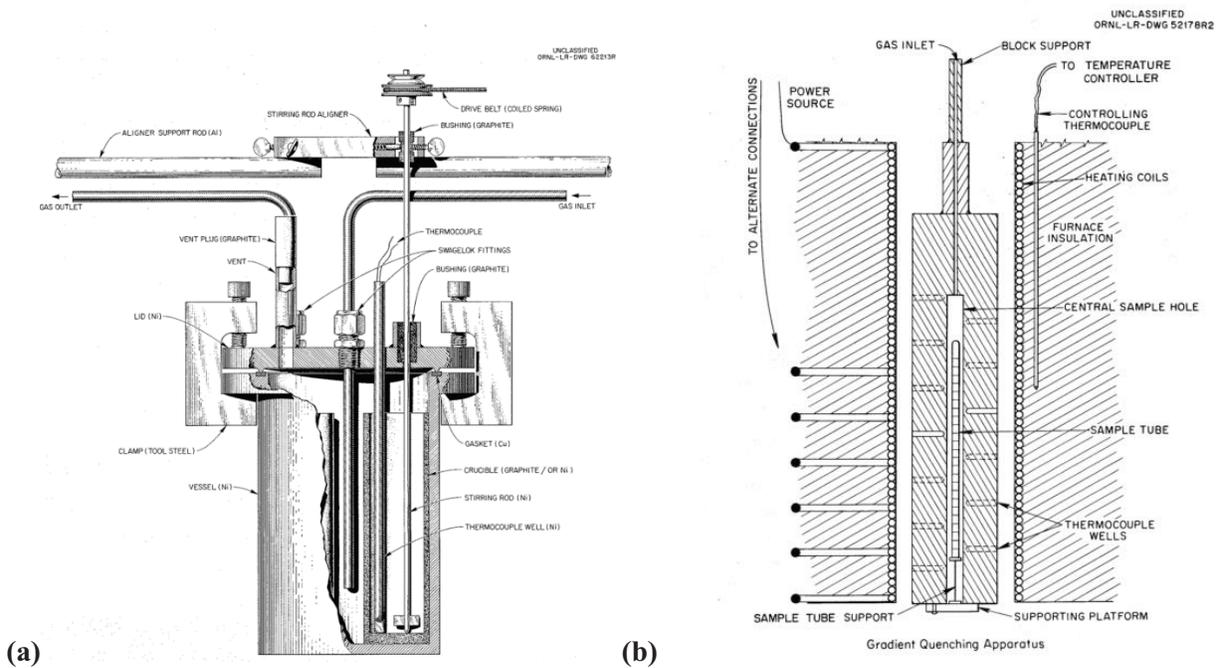
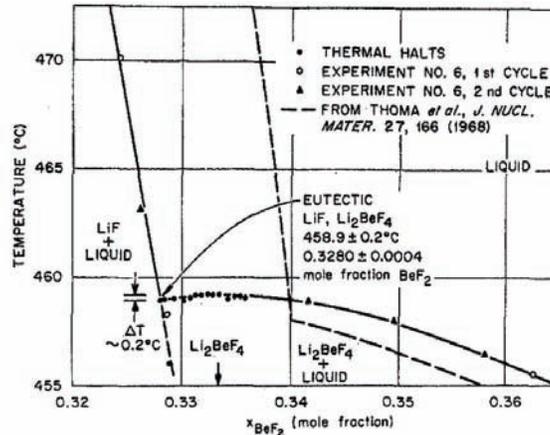


Figure 6. Direct Thermal Analysis Apparatus (a) and quenching apparatus from MSRE (b)[14]

The definition of an exact freezing temperature value for the flibe composition (33% mol BeF<sub>2</sub>), and whether the salt freezes congruently or incongruently has been challenging. In 1968 Thoma and other ORNL researchers stated that flibe was melting incongruently; their results were showing, as reported in Figure 5b, how flibe starts melting at around 455°C and the crystals decompose to form a mixture of liquid and LiF until the temperature reaches 480°C, when all the mixture is in a fully liquid state [12]. However, these considerations were finally revised and updated by the same group 5 years later [7].

Electrochemical measurements were used to conduct a more accurate determination of flibe liquidus curve; the main advantages of this method respect to all the previous experimental experiences was that the measurements were conducted in a real equilibrium status. The experiment was performed in an

electrochemical transference cell, where the emf between two cells containing LiF-BeF<sub>2</sub> molten salts was measured. With this system it was possible to detect changes of phase comparing the change of slope in the emf-composition curve in the case of two cells with different compositions kept isothermal or the discontinuity in slope in the emf-temperature curve in the case of two cells with the same composition but at two distinct temperatures. In these measurements the time was not a variable and the system was in equilibrium during every measurement. The effect of supercooling was reported as a possible issue to affect other experimental measurements; this was completely avoided by approaching the liquidus curve from both sides, increasing and decreasing the temperature [7]. Figure 7 reports the liquidus curve with the data points determined with emf measurements; the reported uncertainty is +/- 0.2°C.



**Figure 7. Experimental points measured by Romberger et al. [7] around the nominal flibe composition. The dotted line represent the previous version of the phase diagram reported in Figure 5(b).**

Additional research was needed in recent years because of the mixtures with high BeF<sub>2</sub> concentration tends to form glasses, with such a high viscosity that makes challenging to determine equilibrium transitions through experimental measurements. New techniques involve the development of a theoretical model, based on the definition of the Gibbs energy functions for all the phases of the system, and the solution of it to obtain a diagram that fit the existing experimental data [8]. Recent papers showed some updates in the phase diagram for mixtures >80% BeF<sub>2</sub>, but for what concerns flibe compositions of interest for nuclear reactor applications, Figure 2 is the most complete phase diagram to date.

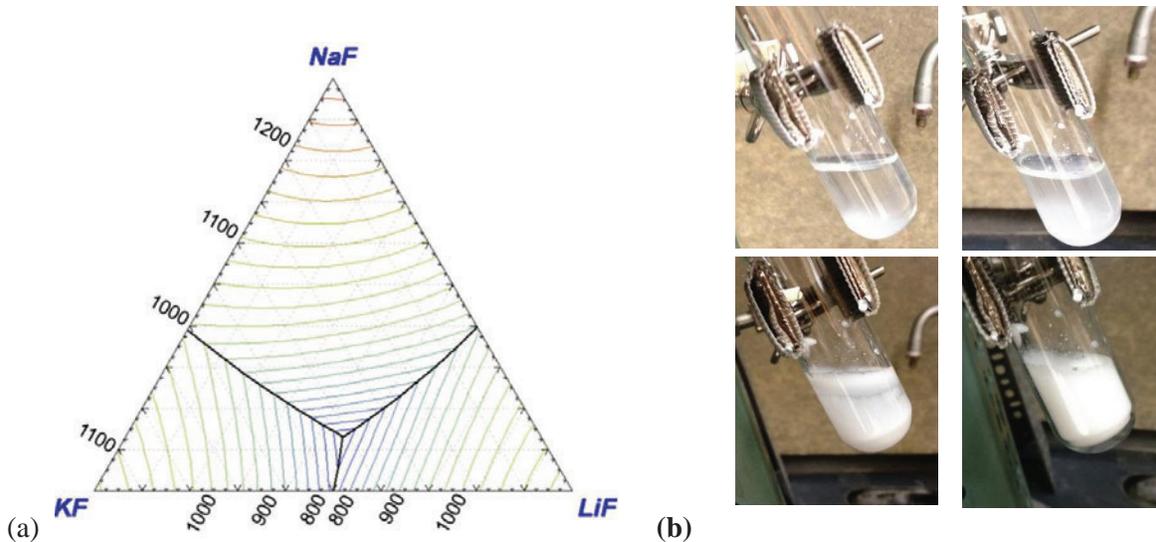
#### 2.1.4 Flinak phase diagram

The LiF-KF-NaF eutectic composition (respectively 46.5,42 and 11.5 mol%), commonly known as flinak, is a heat transfer salt of interest as a non-beryllium surrogate salt for flibe.

The Chinese Academy of Sciences, in the TMSR-SF project, an experimental reactor with solid fuel and flibe molten salt as a primary coolant, is developing a preliminary comprehensive experimental platform called TMSR-SF0. This integral facility will replicate the key materials, technical scheme and equipment design of the final TMSR-SF1 experimental reactor, but it will use flinak as the primary coolant and it will be electrically heated.

Figure 8 shows the flinak phase diagram with projections of liquidus curves at different temperatures; the flinak composition is visible in the center of the diagram that corresponds to the lowest freezing temperature for these three components. As can be seen from the phase diagram, perturbation of the flinak

composition from the eutectic, causes the freezing temperature to increase and leads to incongruent freezing. Differently from flibe, flinak freezes forming a polycrystalline mixture, where every one of the three species in the system forms its own crystals.



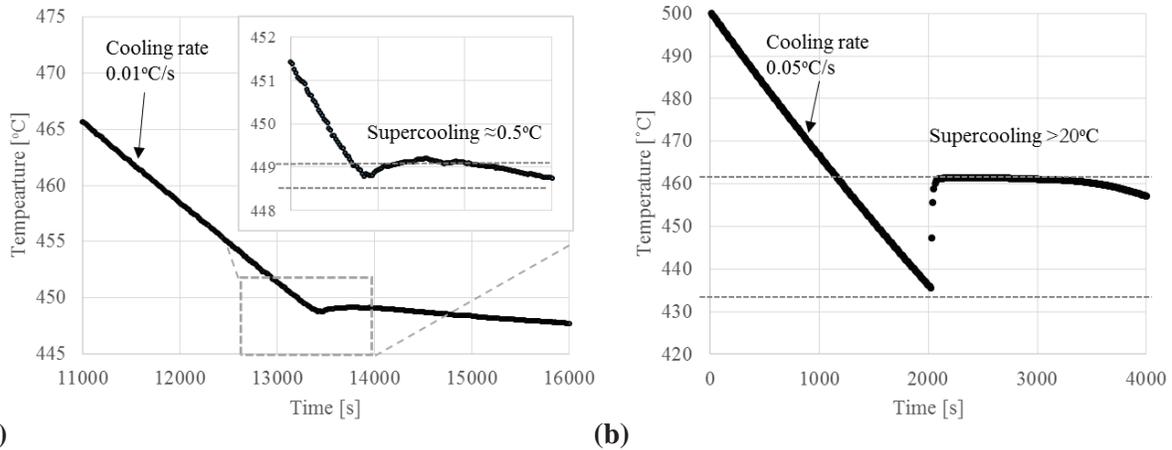
**Figure 8. Liquidus curves for the LiF-NaF-KF mixtures of salts [8] (left), and flinak freezing in a test tube (right).**

## 2.2. Non Equilibrium Freezing - Supercooling

Phase diagrams report equilibrium transitions. In real case transients however, solidification can occur in much different conditions like rapid quenching, lack of nucleation sites and local temperature gradients. In these different conditions the solid phases formed and the freezing temperatures can considerably differ from the one expected from the phase diagrams.

Supercooling is the effect of a material conserving its liquid condition at temperatures below its solidification temperature. No specific research has been performed on supercooling of flibe, but this phenomenon can have relevant implications in view of the possibility of a tube plugging in the salt-to-air heat-exchanger of FHR or the stability of flow loops, particularly natural circulation loops. Some preliminary experimental investigation of flibe supercooling is reported here.

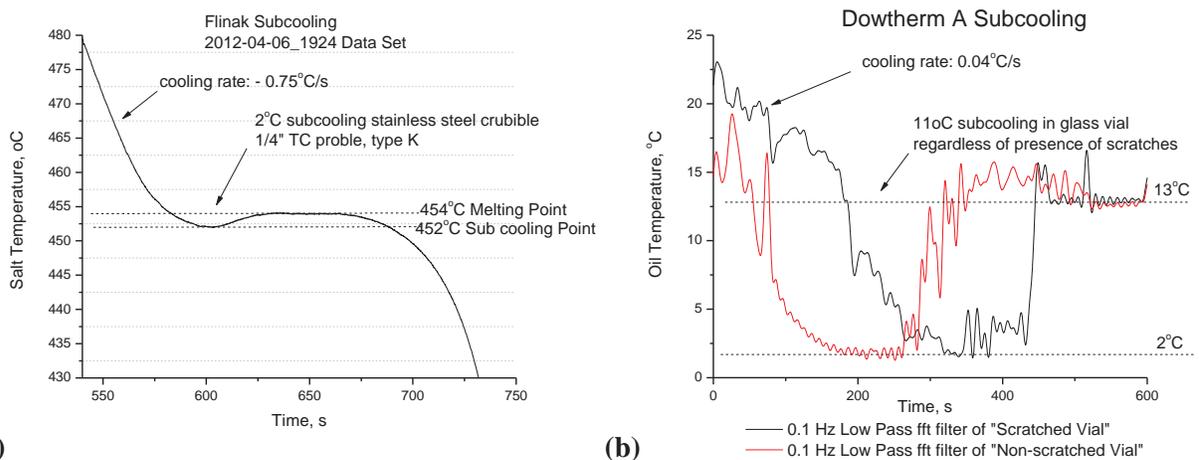
No experiments specifically designed for the study of flibe supercooling have yet been performed. However, temperature evolution data during freezing transitions is available from salt handling operations related to salt purification and corrosion studies performed at UW Madison [10]. Figure 9 shows two such examples of thermocouple data during a phase transition. The data represent the cooling of the salt inside two nickel vessels of different sizes, cooling at different rates. The heat is removed from the external surface of the nickel vessel in contact with air at room temperature. In Figure 9(a), the thermocouple was inserted in a thermowell inserted in the salt; the air layer between the thermocouple and the thermowell may have contributed to the shift of the measured freezing temperature. In Figure 9(b) data was acquired from a thermocouple in the surface of the nickel vessel. This data is an indication that under some conditions flibe may exhibit supercooling at high as 20°C.



**Figure 9. Flibe cooling data observed in different conditions and cooling rates. [10]**

A preliminary set of investigations on the freezing phenomena was also done with the surrogate salt flinak and with the heat transfer simulant fluid Dowtherm A. If these fluids are to be used to reproduce flibe freezing phenomenology in FHR overcooling transients, then their freezing phenomenology must also be understood in order to characterize the experimental distortion arising from using these fluids.

A preliminary flinak freezing observation were done in a stainless steel cylindrical crucible, half-filled with salt; a stainless-steel sheath type K thermocouple inserted in the bulk of the liquid salt. The salt was heated above its melting point in a furnace, then placed at ambient temperature for cooling. Experimental results shows supercooling of 4 °C with a thin thermocouple probe (0.5 mm diameter, type K) and 2 °C with a thick thermocouple probe (6 mm diameter, type K) (Figure 10(a)). The cooling rate for both cases was 0.7°C/s. The salt was not stirred while cooling and relevant temperature gradients were probably present radially and axially in the coolant. Freezing initiation was observed at the stainless-steel thermocouple and crucible surface, and then propagated to the center. The impurities visible in the salt might also affect the supercooling effect, giving possible nucleation sites for the salt.



**Figure 10. Flinak temperature evolution upon exposure to 20°C ambient temperature in a stainless steel vial (a) and Dowtherm A temperature evolution after insertion in a 0°C ice bath (b).**

Dowtherm A freezing observations were done in 20mL glass scintillation vials, filled with approximately 5 mL of Dowtherm A oil. A thermocouple was inserted in the vial and the vial was immersed in an ice-bath that maintained a temperature between 0-0.5°C during the experiments. The measured temperature of the oil is plotted in Figure 10(b), for two vials. The inside of one of the vials (black graph line) was vigorously scratched with a steel brush, to provide nucleation sites for solidification. The thermocouples seems to behave as nucleation sites. The same experiment, in an un-scratched vial, with no thermocouple, will not lead to solidification of Dowtherm A in an ice-bath at 0°C. With thermocouples present, the scratching of the glass vials does not appear to change the temperature to which the fluid supercools before solidification occurs. The melting point of Dowtherm A was measured at 13°C, and the literature melting point is 12°C. Experimental results shows supercooling of 11°C below the freezing point. Dowtherm A remains liquid down to 2°C and upon initiation of freezing its temperature rises to 13°C. The cooling rate was 0.04°C/s.

Therefore, flibe, flinak and Dowtherm A all exhibit supercooling behavior, in preliminary experimental observations. Future experimental work at UW Madison will investigate supercooling phenomenology in flibe and its potential surrogate and simulant fluids, with the objective of identifying the cooling regimes in which it occurs and the parameters that affect the degree of supercooling.

### 3. FREEZING FRONT PROPAGATION IN A PIPE

Characterizing pressure drop evolution in freezing pipe-flow will be an important input for the models of transient response of overcooling transients in FHRs. In order to do so, a model of the freezing front propagation in a pipe needs to be developed. This model will need to incorporate the equilibrium and transient phase change phenomena that occur in flibe, such as supercooling, which were discussed above.

Previous studies on freezing in a pipe include significant work on cold-filling pipes, since there is interest in the field of solar thermal energy storage. For example, Jianfeng et al. built a FLUENT model of salt freezing as it fills an empty pipe. Figure 11 shows a frozen layer that initially develops from the walls; this layer re-melts as the established salt flow continues to provide heat. The solid layer reduces the flow area, so pressure loss increases significantly. For 453K salt flowing at 1.0 m/s through 1.6 cm pipe,  $Re = 5291$ , this freezing and melting process evolves in less than 10 seconds [15]. The salt melts at 410 K, and its heat capacity is  $1510 \text{ Jkg}^{-1}\text{K}^{-1}$ .

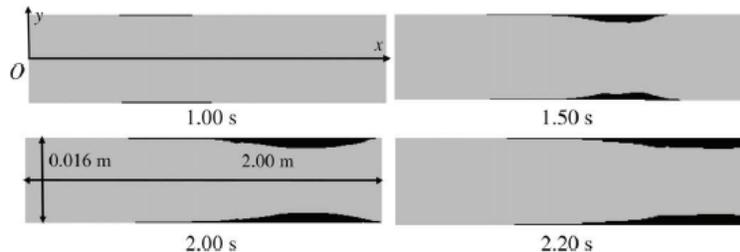


Figure 11. Solidification in the pipe during the freezing process. Adapted from [15].

In another study, Pacheco et al. constructed a molten salt loop and investigated the temperature response of the outside surface of the insulated tubes as they filled with a nitrate salt. They observed similar freezing and melting behavior, as shown in Figure 12. Pipe near the source of salt (“1<sup>st</sup> pass”) heated up quickly and reached equilibrium, while pipe further downstream (where the salt would get cooler) experienced some freezing. This is most obvious in the “4<sup>th</sup> pass” where the temperature evolution is depressed, then the temperature spikes rapidly again at around 690 seconds after melting. The study

shows that the frozen layer acts as an insulating boundary. They estimated significantly lower heat transfer from the frozen layer through this experiment; along the first section of pipe they found  $h = 2700 \text{ W/m}^2\text{K}$  vs  $h = 1000 \text{ W/m}^2\text{K}$  for the fourth section [16]. In this experiment, the pipe inner diameter is 2.37 cm, and the bulk salt temperature is 561 K with a freezing point of 494 K, an approximate  $Re = 7513$ , and an approximate heat capacity of  $1482 \text{ Jkg}^{-1}\text{K}^{-1}$ . For similar heat capacities, the higher Reynolds flow is not as susceptible to freezing front propagation.

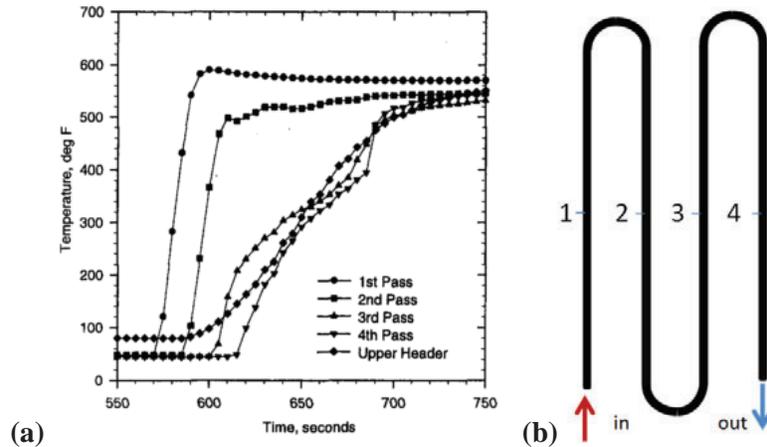


Figure 12. Temperature response of tubes as filled with salt. Adapted from [16].

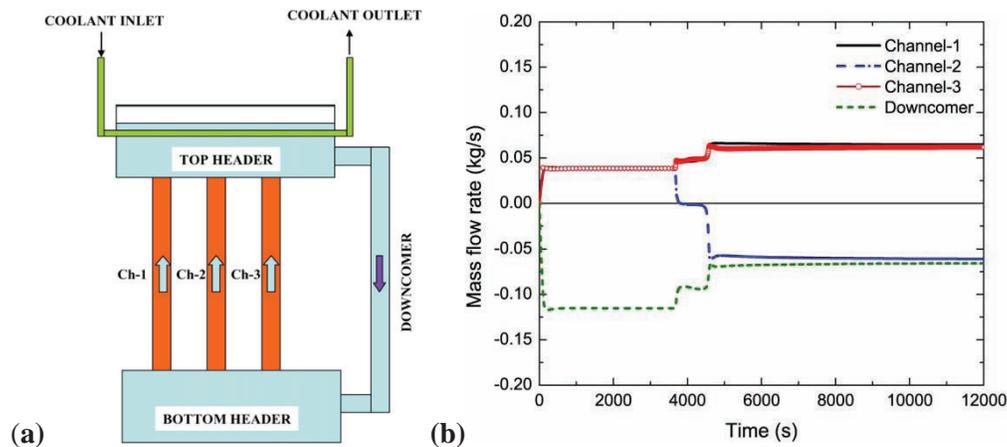
Future experimental studies on salt freezing must eventually investigate frozen front propagation in pipes with flowing salt. The freezing process of flibe and flinak in a pipe will be object of experimental and numerical investigation at UWM. We are interested to study the system replicating the conditions in a salt-to-air heat exchanger tube during different possible transient in FHR. Understanding how much the salt can travel in a cold pipe before plug it and the conditions for re-melt behavior will be the focus of this research. Finally, demonstrating instrumentation that identifies onset of freezing will also be a desired outcome of the experimental studies.

#### 4. STABILITY AND FREEZING OF A NATURAL CIRCULATION LOOP WITH MULTIPLE BRANCHES.

The development of instabilities and flow reversal, generated by an initial power perturbation is in general a concern in natural circulation loops. A case of interest for the FHR natural circulation loop is the study of multiple channel natural circulation, because of the multiple branches that the heat exchanger presents in the FHR system in the use of a direct reactor auxiliary cooling system (DRACS) as part of the ultimate heat sink during transients for decay heat removal [1].

For a system with multiple parallel channels operating under steady single-phase natural circulation condition, P.K. Vijyan et al. [17] showed that a temporary disturbance in the form of an increase in the power of one of the channels can lead, under defined critical conditions, to instabilities in the loop. A power fluctuation in a channel impacts temperature, density and pressure drops in the flow in the specific channel; since the same  $\Delta P$  has to be maintained across all the parallel branches of the system all the neighboring channels will experience and react to the original power perturbation. Depending on the perturbation, configuration and working condition of the natural circulation system, the interaction among

the parallel channels can result in a return to the initial stable condition or in the development of fluctuation and instabilities of different magnitude that can lead to unstable flow conditions or flow reversal in channels. M.R. Gartia et al. studied this kind of instabilities in multiple channel water natural circulation systems with non-uniform heat inputs. A three parallel channel configuration connected to common inlet and outlet headers with a down-comer was investigated (Figure 13(a)). The study showed that multiple steady states with different flow directions may occur for certain perturbations of the heat flux. Figure 13(b) shows the mass flow rate variations with time at different channels in the case where all three channels have equal initial heat fluxes and the power in Channel-2 was gradually decreased during transient conditions. From the figure, it appears that the reduction in power has resulted in reversal of flow in the disturbed channel from upward to downward. This flow reversal phenomenon have been proved to happen after the power of the channel is decreased below a critical value, shown to be function of the operating procedure and starting power [18].



**Figure 13. (a) Schematic of the natural circulation loop and (b) mass flow rate variations at different channels [18].**

Furthermore, the use of molten salt coolant introduces some additional factors in this kind of instability studies, not considered in water loops, related to variation of the salt viscosity with temperature and the possibility for the salt to freeze on the walls of the pipe, impacting heat transfer efficiency and pressure drop, or to completely plug a section of the heat exchanger. Research at UWM is focused to perform natural circulation stabilities studies for FHR loops designs, taking into account these salt related phenomena in terms of feedback coefficients for the dynamic response of the system to perturbations.

## 5. CONCLUSIONS

The phenomenon of flibe freezing and its possible impacts on FHR reactors thermal-hydraulics has been presented. It has been shown how the presence of uncertainty in the composition of the salt can introduce additional uncertainties in the equilibrium freezing temperature and can change the freezing equilibrium phenomenology. Moreover the non-equilibrium freezing phenomena have been considered and it has been presented, through preliminary experimental observations, that flibe, flinak and Dowtherm A all exhibit supercooling behavior.

Future work at UWM will be focused on develop experimental studies to clarify the phenomenology of flibe freezing and the possibility to use simulant fluids to study overcooling transients without major

distortions. The phenomenology of flibe freezing in a pipe will be addressed from both an experimental and modeling approach. The final goal will be to clarify the conditions under which overcooling transients in FHR can lead to instabilities in the natural circulation or forced convection flow loops.

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