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AN EXPERIMENTAL STUDY OF NATURAL CONVECTION MELTING OF ICE IN SALT SOLUTIONS

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by

L. J. Fang, F. B. Cheung,* J. H. Linehan,** and D. R. Pedersen

Reactor Analysis and Safety Division
Argonne National Laboratory
9700 So. Cass Avenue
Argonne, Illinois 60439

* To whom correspondence concerning this paper should be addressed.

** Present Address: Mechanical Engineering Department, Marquette University, Milwaukee, Wisconsin 53233.

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Abstract

The solid-liquid interface morphology and the micro-physical process near the moving phase boundary during natural convection melting of a horizontal layer of ice by an overlying pool of salt solution were studied experimentally. A cathetometer which amplifies the interface region was used to measure the ice melting rate. Also measured were the temperature transients of the liquid pool. Within the temperature and the density ratio ranges explored, the ice melting rate was found to be very sensitive to the ratio of pool-to-ice melt density but independent of pool-to-ice temperature difference. By varying the density ratio, three different flow regimes and morphologies of the solid-liquid interface were observed, with melt streamers emanating from the crests of the wavy interface into the pool in all three cases. The measured wavelengths (spacing) between the streamers for four different pairs of materials were correlated with the density ratio and found to agree favorably with the predictions of Taylor instability theory.

1. INTRODUCTION

Downward penetration of a horizontal solid substrate by an overlying hot liquid pool has recently received special attention [1-4] mainly because of its relevance to the studies of decay heat removal in nuclear reactors. The situation arises following a postulated severe reactor core meltdown accident when a pool of core melt forms on top of an ex-vessel structure such as a concrete basemat [5] or a MgO delay bed [6]. In general the structural or sacrificial bed material, when molten, is miscible with and lighter than the core melt so that the rate of penetration is strongly dependent upon the motion of natural convection in the melt layer driven by the density difference between the core melt and the molten substrate. Understanding of the mechanism of the natural convection melting process is essential to the design of a core-retention system for post-accident heat removal.

Farhadieh and Baker [1] were the first to study the melting rates and the associated convective motions in a heated pool above a melting miscible substrate. They used water soluble wax as the solid material and an aqueous salt solution heated by a suspending planar heater as the overlying pool to make the exploratory experiment. Needle-like streamers or fingers of melt material were observed which extended into the overlying heavier solution from discrete but random sites at the interface. These fingers were essentially buoyant plumes that injected melt material into the liquid pool. Large and small vortices were seen at the outer edge of the fingers, which produced vigorous mixing between the melt and pool materials. The downward melting rate was quite sensitive to the density ratio of the liquid pool and the molten substrate, which was varied in the experiment by changing the salt concentration in the liquid pool. At density ratios between 1.09 and 1.25, the observed natural convection melting rates were consistent with a pool flow regime of ordinary turbulent convection. When the density ratio exceeded about 1.25, there was a sudden increase in the slope of the melting curve which they assumed to be due to a very vigorous turbulent regime, defined as the upper turbulent regime†. There are many questions raised by Farhadieh and Baker's [1] seemingly simple experiment. The existence of the upper turbulent regime, the kinetics of needle-like streamers and the melt layer, and the sites or the population of the streamers at the interface are not quite understood to date.

Eck and Werle [2] conducted a series of experiments similar to those of Farhadieh and Baker [1] by using the same simulant materials with the difference that the grid heater was replaced by a flat plate. They found that the measured downward melting velocity increased with the density ratio and that a transition occurred when the density ratio was about 1.1. They also found that the melting rate increased with the pool-to-substrate temperature difference which agreed with the results of Farhadieh and Baker [1]. Nevertheless, some quantitative discrepancies in the measured melting rate of up to a factor of five were observed. Eck and Werle [2] attributed these discrepancies to the differences in the setup of heater and thermal conductivities of test-section materials.

Based on the assumption that there is a melt layer separating the solid from the overlying liquid pool such that the conditions for Taylor instability at the melt layer-pool interface are met, Catton et al. [3] were the first to model the heat transfer from the liquid pool to the melting miscible substrate. The melting rate was also measured in [3] and was found to be sensitive to the pool-to-melt-layer density ratio as reported by Farhadieh and

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† The existence of this regime was not observed directly but was postulated based on the fact that the measured melting rate changed abruptly with the density ratio beyond the value of 1.25.

Baker [1] and Eck and Werle [2]. However, the results regarding the dependence of the melting rate on the pool-to-substrate temperature difference were not consistent among themselves. While Farhadieh and Baker [1] and Eck and Werle [2] reported that the melting rate was very sensitive to the temperature difference, Catton et al. [3] reported that the melting rate was essentially independent of the temperature difference. Based on the assumption that the spacing between the melt streamers is determined by Taylor instability, Epstein et al. [4] suggested that the viscosity of melting solid could be the resolution of this discrepancy. Unfortunately, there is insufficient data based on which a definitive conclusion can be made. It should be noted that the work of Catton, et al. [3] was the only study that reported the spacing between streamers (wavelength) based on the observation of striations on the surface of the melting material (Benzene). However, the dependence of the wavelength on the density ratio parameter has not been quantitatively studied and its relation with the melt flow is not known.

The existence of an upper turbulent regime, as postulated by Farhadieh and Baker [1] based on their measured melting rate vs. density ratio data, was not confirmed by the experimental evidence given in [2-4]. Questions concerning the occurrence of flow transition and its effect on melt penetration remain to be answered. Thus far, no melting-rate correlation applicable over the entire range of density ratios envisioned in hypothetical situations has been established. In view of these, experiments were conducted in the present study to promote our understanding of the melting phenomena and to strengthen the data base of melt penetration. Experimental technique was developed for the purpose of direct observations of the melting rate and the solid-liquid interface morphology as well as the behavior of the melt streamers. A wide range of pool-to-substrate density ratios were explored which covered the laminar, transition, and turbulent flow regimes.

2. EXPERIMENTAL METHOD

The natural convection melting phenomena were studied experimentally using various salt (KI, NaCl, $MgCl_2$, or $CaCl_2$) solutions as the liquid pool and an ice slab as the solid substrate. To eliminate the possible disturbance of the flow field in the liquid phase as well as the melting interface morphology due to the release of air bubbles from the ice slab, air-bubble-free ice samples were used in the experiments. The test chamber, designed for growing an air-bubble-free ice, was made by fitting together two separate parts (see Fig. 1). The upper part is a single-walled cylinder and the lower part is a double-walled cylinder with inner and outer diameters of 50.8 and 70 mm, respectively. The space between the two cylindrical walls was evacuated to reduce heat transfer in the radial direction. Before each run, air-bubble-free ice was prepared† on top of the aluminum plate inside the test chamber (Fig. 2). To do this, a mechanical stirrer and an electrical heating pad (~ 15 W) were placed through the cylinder at the top. Distilled water was then introduced into the cylinder and the entire system was placed in a freezer held at a desired subzero temperature for about 48 hours. With the space between the double side walls put under vacuum, the water layer was cooled from below by the cold aluminum plate and heated above by the electrical heater. In this way, the ice was able to grow upward from the bottom of the water layer. Meanwhile, water motion created by the stirrer swept away air bubbles that formed at the ice-water interface. After the "clear" ice grew to a thickness of about 0.10 m during a two-day period, the remaining water on

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† A number of different methods for preparing such an ice sample have been reported in the literature. This particular method is chosen according to the one described in [7].

top of the clear ice was removed. The ice surface was made smooth and flat by melting a small part of the top portion of the ice slab. Then, the ice was conditioned to a uniform temperature at ~ -2°C by placing the test chamber containing the ice in a freezer for more than 15 hours.

To prepare for a run, the upper cylinder which was kept at room temperature was assembled onto the top of the lower, double-walled cylinder and the test chamber was removed from the freezer. Then, 250 ml (pool height equals to 123.3 mm) of salt solution (KI, NaCl, $MgCl_2$ or $CaCl_2$ solutions) at a premixed concentration and a prescribed temperature was introduced on the top of the ice slab to initiate the melting process. Special care was taken as the solution was poured over the ice substrate to prevent non-uniform erosion of the initial ice surface. Two thermocouples (K type chromel-alumel), with one junction located at the center of the test chamber and 2.5 cm above the initial ice surface, and the other located at the bottom of the ice slab were used to measure the bulk temperature responses of the liquid pool and the ice temperature, respectively. Only one thermocouple for the bulk pool temperature measurement was found to be necessary because of the uniformity of pool temperature due to the natural convection motion within the pool. The ice melting rate was measured by a cathetometer with 0.01 mm resolution. Since the temperature of the upper cylinder was very close to the initial solution temperature (~ 25°C) in most runs, heat loss from the solution to the side wall was effectively minimized. This provided us an alternate way to determine the melting rate by calculating the time rate of change of the solution temperature. Attempts to measure the height of the melt streamers were also made by using the cathetometer. To limit the pool density change as a result of dilution during ice melting, 240 to 300 second melting periods, which were found to be sufficient to provide quasi-steady melting data, were employed in the experiments (the solution initial depth was 123.3 mm, thus, the maximum pool density change was less than 1.5%). At 320 seconds after melting was initiated, the solution on top of the ice was removed and the roughness on the surface of the ice slab was photographically recorded.

3. OBSERVATIONS OF THE MELT STREAMERS AND MELTING FRONT

In all of the experiments (52 runs and several preliminary runs), erosion of ice commenced immediately after the pouring of the warm solution on top of the ice slab. A very strong disturbance due to mixing between the ice melt (i.e., the pure water) and the solution layers near the melting interface was observed as a result of initial transients due to pouring. The time for this disturbance to die out was about 10-20 seconds. After the initial transient period, melt fingers or streamers, were observed near the melt front, penetrating upward into the overlying solution pool. The melt-ice interface was found to be rough and wavy. The photographic records and visual observations for some preliminary runs indicated that the time for developing a "regular" or "quasi-steady" size of melt interface roughness was at least 2.5 minutes. This is the reason that all of the melting experiments were extended between 4 to 5.5 minutes instead of terminating after approximately 2 minutes as in [3].

By varying the solution concentration, different convection patterns in the liquid pool and different morphologies of the melt-ice interface were observed. When the pool-to-melt density ratio ρ/ρ_m* was between 1.006 and 1.05, the vertical motion of the melt fingers appeared to be "laminar" since the flow of the melt streamers was constantly vertical with no eddies developing in the vicinity. The average height of the streamers was found to be a

* The subscript m refers to the properties of the ice melt layer, the unsubscripted variables refer to the pool properties.

little larger than 10 mm. A photographic study of the melting interface roughness showed that the average "unit" size of the roughness structure (wave length) was between 2 and 5 mm, decreasing with increasing density ratio. Figure 3 presents the top view of the roughness of the melting interface resulting from a typical case of a KI solution with density of 1.012 initially at 24.7°C and an ice substrate initially at ~ -2°C. The picture was taken immediately when the solution was poured out at 5 minutes and 20 seconds after the onset of ice melting. In the picture, the striations represent the crest portion. The average roughness height (wave height) between the crest and trough was about 1 mm measured by using a cathetometer.

When the density ratio was in the range between 1.05 and 1.08, there exists a flow transition region where the melting interface morphology changed considerably. As a result, the sharp and regular structure of melting front roughness disappeared and a radically different melting interface with some irregular striations was present. Figure 4 shows the top view of the melting interface for the case of a KI solution with a density of 1.077 initially at 24°C and an ice substrate initially at -2°C. The picture was taken immediately when the solution was poured out at 5 minutes and 20 seconds after the onset of ice melting. Because of the irregular striation distribution on the melting interface, the height between the crest and trough was difficult to determine.

When the density ratio was larger than 1.08, the flow regime appeared to be highly "turbulent". The strong convective motion in the pool swept away the top portion of the streamers and, consequently, the streamers lost their identities as soon as they left the melting surface. The average height of the streamers was much shorter than that of low density ratio cases. Unlike the laminer case in which the flow of the melt streamers was constantly vertical, here the streamers were twisting in the horizontal direction as they penetrated upward into the solution pool. Due to the turbulent mixing motion and the rapid melting process, the variation of the streamer height with respect to the density ratio was very difficult to determine. From the photographic records, the population of melt streamers per unit area is much higher than the laminar case. The average wave length of the interface roughness elements was about 1 mm in this high density ratio regime. Figure 5 presents the top view of the melting interface for the case of a KI solution with density of 1.376 initially at 24.5°C and an ice substrate initially at ~ -2°C. The picture was again taken immediately when the solution was poured out at 5 minutes and 20 seconds after the onset of ice melting. The striations in the picture represent the crest portion. The average wave height between the crest and trough was much less than 1 mm.

4. TAYLOR INSTABILITY AND THE STREAMER SPACING

In 1950, Taylor [8] discussed the instability of a horizontal interface between two immiscible fluids of infinite depth and showed that disturbances at the interface will grow with time if the upper fluid is heavier (more dense) than the lower fluid. The result of his analysis was an expression for the rate of growth of interfacial waves as a function of the fluid densities, gravitational acceleration constant, and the wavelength. Bellman and Pennington [9] later extended the Taylor analysis to include the effects of surface tension and viscosity of two semi-infinite fluids. The most interesting result of the analysis is that the growth rate of the interfacial wave has a maximum when the wavelength achieves the "most dangerous" value. A great deal of study on this type of instability followed. For example, it has been used to model the film boiling process very successfully [10,11]. Unlike film boiling, in the application of Taylor instability to miscible fluids, interfacial tension is of little importance, instead, the viscosities of the fluids may be the controlling factors. Taghavi-Tafreshi and Dhir [12] studied the combined effect of interfacial tension, liquid viscosity and liquid layer thickness on Taylor instability. Their numerical solution indicated that for the case without interfacial tension, the most dangerous wavelength goes to zero as the melt layer thickness approaches zero. The effect of increased pool-to-melt density ratio is to shorten the wavelength. They also pointed out that a 1 mm thick water layer would act as if it were a semi-infinite body. Recently, Catton, et al. [3] and Epstein, et al. [4] also proposed that the process of melting of a miscible substrate by an overlying heavier liquid pool is governed by the Taylor instability theory for the case of zero surface tension.

To examine the applicability of the Taylor instability theory to predict the average wavelength between streamers, it is necessary to measure the streamer spacing experimentally. To this end, direct observations by a cathetometer were made of the melt fingers or streamers which issued from the crests of the melt ice interface roughness elements and penetrated the overlying pool. Measurements of the crest spacing or wavelength was made by determining the unit size of the melting interface roughness element from the photographical records of the troughs and crests at the conclusion of each experiment. In order to get an average value of wavelength from each photograph, the following procedure was adopted. First, the photograph for each run was enlarged and developed. Then an average unit cell dimension (wavelength) was obtained by measuring twenty-five individual, representative unit cells selected at random locations. The accuracy of each measurement was approximately within plus or minus 0.05 mm. By this method, the 95%

confidence intervals for these measurements can be calculated by assuming the sampling distribution to be approximately normal [13].

Accordingly, the measured average wavelengths are plotted in Figs. 6 and 7 with respect to the density ratio. The maximum deviation of the 95% confidence limits from the average wavelength is approximately plus or minus 8%. The data clearly show that the wavelength decreases with increasing density ratio in the low density ratio region where the flow is laminar. In the high density ratio region where the flow is turbulent, the average wavelength appears to be a constant independent of the density ratio. The equation of Bellman and Pennington [9] is also shown in these figures. This equation was originally derived from the Taylor instability analysis of interfacial waves for two fluids of infinite thickness with the viscosities of the fluids dominating the wavelength as follows:

$$\lambda = \frac{4\pi(\mu + \mu_m)^{2/3}}{[g(\rho - \rho_m)]^{1/3}(\rho + \rho_m)^{1/3}} \qquad (1)$$

where

λ = wavelength,

g = acceleration of gravity,

μ = absolute viscosity of the pool,

μ_m = absolute viscosity of the melt material,

ρ = pool density.

It must be pointed out that the thickness of the melt layer was not infinite in the experiments. However, according to the work of Taghavi-Tafreshi and Dhir [12], a 1 mm thick water layer at room temperature would act as infinite according to the Taylor instability analysis. It is, therefore, proper to use equation (1) for comparison. In the calculation of wavelengths from equation (1), the numerical data for the pool viscosity (a function of solution concentration) and melt viscosity were taken at 20°C and 0°C, respectively [14]. As a result of significant pool viscosity increase with the pool density, the theoretical curves on Figs. 6 and 7 in the high density ratio region are curved into positive slopes. Nevertheless, the Taylor instability prediction compare favorably with the measured wavelength data. This indicates that the streamer spacing may be predicted by Taylor instability theory.

5. THE TEMPERATURE TRANSIENTS

The prime purpose of this part of the melting study is to clarify the role of temperature difference on the melting rate. Therefore, preliminary experiments using solutions with a given pool density but various initial temperatures and various pool depths were conducted. Results from these preliminary experiments indicated that the melting rate was essentially independent of the temperature difference between the pool temperature and melting point of the substrate when the pool depth exceeded ~ 100 mm and the pool temperature was higher than the melting point of the substrate. One of the typical results is described in the following. Two runs (A) and (B) using NaCl solutions (pool depth: 123 mm) with the same initial density at 1.07 but at different initial temperatures (58°C and 24°C) were conducted. Figures 8 and 9 show the transient bulk temperature of the pool and the penetration distance vs. time of these two runs, respectively. After the initial transients due to pouring had died out, the bulk pool temperatures of these two runs remained almost constant as shown in Fig. 8. From Fig. 9, it can be seen that the quasi-steady melting rates (the slopes of the penetration distance versus time curves) of the ice substrate were nearly identical in spite of the large pool temperature difference between the runs (see Fig. 8). The only effect of the higher initial pool temperature was that the transient melting rate in the initial period following pouring was higher (as evidenced by the difference in the zero time intercepts of the quasi-steady curves). Additional evidence for the insensitivity of the melting rate on the pool-to-substrate temperature difference is illustrated in Fig. 10, where the bulk pool temperature and penetration depth are plotted as a function of time. Although the bulk temperature is clearly decreasing during the course of the run, the experimental values of penetration depth and time fall on a straight line, the slope of which is the constant melting rate. Thus, based on the insensitivity of the quasi-steady melting rate on either initial pool temperature from run to run or on bulk pool temperature variations within a run, it is concluded for these low melt layer viscosity experiments that the melting rate is independent of the temperature difference between the pool and ice substrate. These findings are in agreement with the results of Catton et.al. [3], for benzene melting beneath pools of CCl_4 or Ch_2I_2 wherein the density ratio was above 1.5, from which they concluded that the melting rate of benzene was independent of the pool-to-benzene temperature difference. Physically this implies that melting was dictated by mass transfer. It should be mentioned, however, that this finding needs further examination for viscous melt materials such as the poly 1500 (absolute viscosity ~ 80 cp) employed by Farhadieh and Baker [1] and Eck and Werle [2] since both the ice and benzene which were used in this work and Catton's work [3], respectively, are materials with low viscosities (ranging from 0.8 to 9 cp).

6. CORRELATION OF THE MELTING RATE

The measured melting rates for various kinds of salt solution are shown in Fig. 11. Different flow regimes are also indicated on this figure. Three density ratio regions, $\rho/\rho_m < 1.05$, $1.05 < \rho/\rho_m < 1.08$, and $\rho/\rho_m > 1.08$ are designated as laminar, transition, and turbulent flow regime, respectively, according to the observed melt streamer behavior in the interface region. No upper turbulent regime which was proposed by Farhadieh and Baker [1] was found when density ratio was greater than 1.25 since neither a convective flow behavior change within the pool was observed nor an abrupt change in the melting rate with density ratio was measured.

For the purpose of providing a comparison with the results obtained herein and for obtaining a correlation for the downward melting rate, the experimental results obtained by Farhadieh and Baker [1], Catton et. al. [3], and this study are shown in Fig. 12. The data shown in the figure follow the same general trend except for the higher density ratios where the the data of Catton et. al. are located at higher positions. Although there is flow transition in the pool at the density ratios between 1.05 and 1.08, it does not signficantly alter the overall rate of mass flux. In fact the data show a tendency of constant slope over the entire range of density ratios so far explored. It is, therefore, adequate to develop a correlation for the mass flux rate by a linear regression analysis.
This gives

$$\dot{m} = \rho \frac{dz}{dt} = 0.15 \, (\rho/\rho_m - 1)^{0.776} \; (\mathrm{kg/m^2 \cdot s}), \quad 1.006 < \rho/\rho_m < 3.3 \tag{2}$$

where $\dot{m}$ is the mass flux rate, z the interface location, and t the time variable. This correlation fits almost all of the data to within ± 50% over the entire region for 1.006 $\leq \rho/\rho_m \leq$ 3.3. In the higher density ration region, i.e., ρ/ρ_m > 1.5, part of the data which were reproduced from the work of Catton et. al. [3] (one data point was from Farhadieh and Baker's work [1]) indicate about + 60% deviation from the correlation. The spread of the experimental data about the correlation may be explained by the property variations such as the viscosities of the pool and the melted substrate materials and by the mass diffusivity variation for different material pairs should mass transfer dominate the melting process. The above correlation, which covers almost the entire range of density ratios envisioned in hypothetical nuclear reactor accidents, may be quite useful for practical purposes.

7. FINAL REMARKS

It is of interest to re-examine the role of temperature difference on the melting process. We have noted that when the pool depth exceeds a certain limit and the pool temperature is higher than the melting point of the substrate, the melting rate is essentially independent on the pool-to-substrate temperature difference. Two questions could be raised. The first one is "Why does the pool depth need to exceed a certain limit for this to be true?". The answer is very straightforward since the melting rate is very sensitive to the pool density. If the pool depth is not large enough, the melting rate will change according to the variation of pool density as a result of pool dilution due to ice melting. Six preliminary experiments of varying pool depth indicated that when the pool depth was larger than 100 mm, there was no appreciable change in the melting rate. Thus, in all of the melting experiments, a constant pool depth of 123 mm was used. The second question is "Does melting take place when the pool initial temperature is lower than the normal melting point of the substrate?". The answer is yes, if the pool-substrate contact temperature is higher than the liquidus temperature of the pool solution at the corresponding concentration, since the pool is not in thermodynamic equilibrium with the substrate material under such a condition [6,7]. Preliminary experiments showed that a 15 wt. % NaCl solution with initial temperature at -5°C penetrated the ice at the same initial temperature of -5°C although the temperature difference in this case is identically zero. The eutectic phase diagram shows that they are not in thermodynamic equilibrium (the liquidus temperature of 15 wt. % NaCl solution is ~ -11°C). In this case ice melting is mainly dictated by mass transfer. The temperatures of the solution and the ice measured at locations approximately 5 mm above or below the melting interface decreased as melting proceeded. (In the period of 5 to 10 minutes after the contact of the solution and ice, these temperatures actually decreased from -5°C to about -8°C.) This indicated that the interface temperature was lower than the temperatures of both liquid and solid phases, indicating an unconventional heat transfer mode for melting as a result of heat flow from both liquid and solid phases to the melting interface region. Further experiments involving careful measurements of temperature and concentration distributions are required in order to quantify the foregoing effect. An experimental study of the effect of viscosity on the melting process is also required, as this may lead to the resolution of the pool temperature effect on the melting process for different materials.

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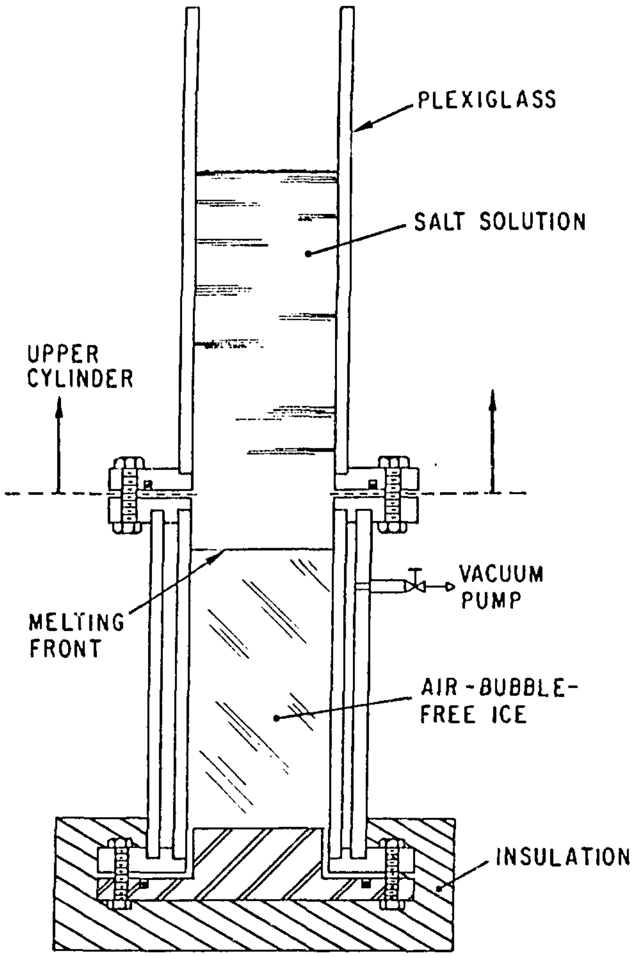

Figure 1. Schematic of the Test Chamber for Natural Convection Melting Study.

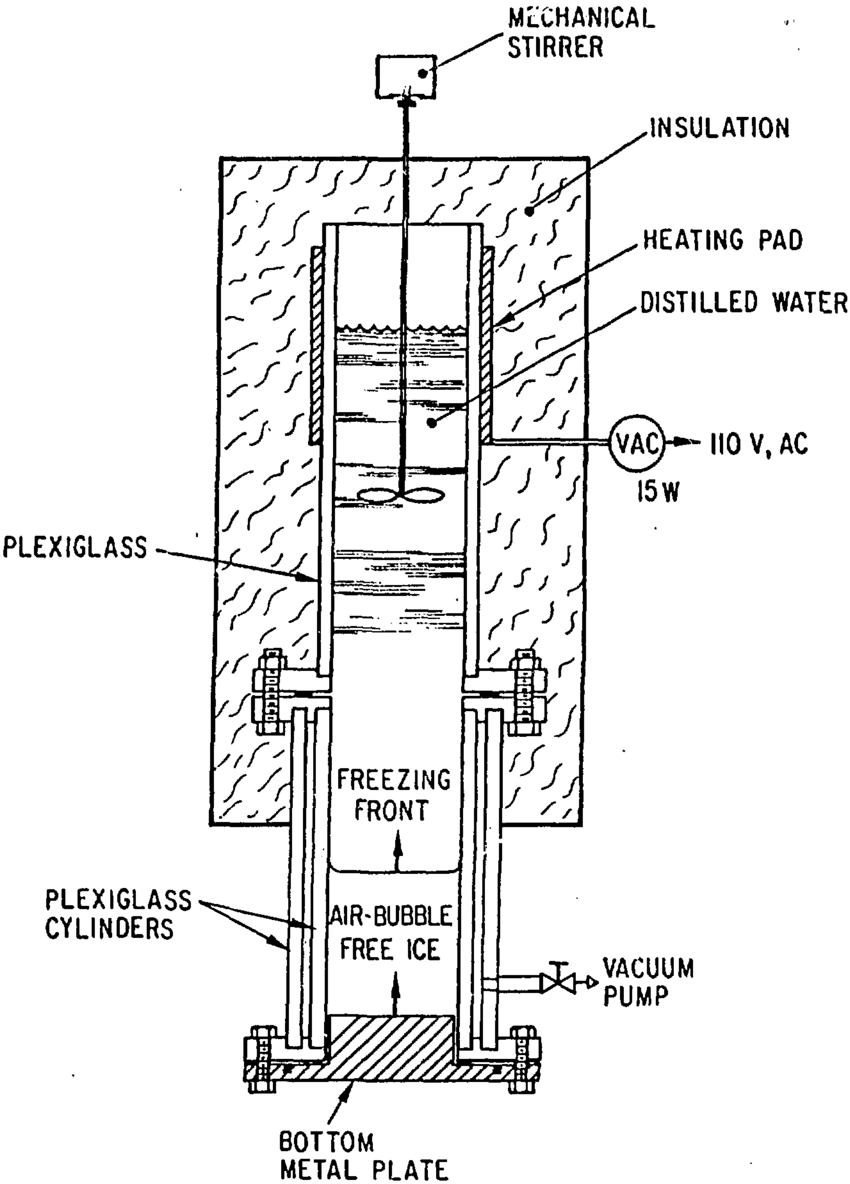

Figure 2. Schematic of the Experimental Arrangement for Air-bubble-free Ice Preparation.

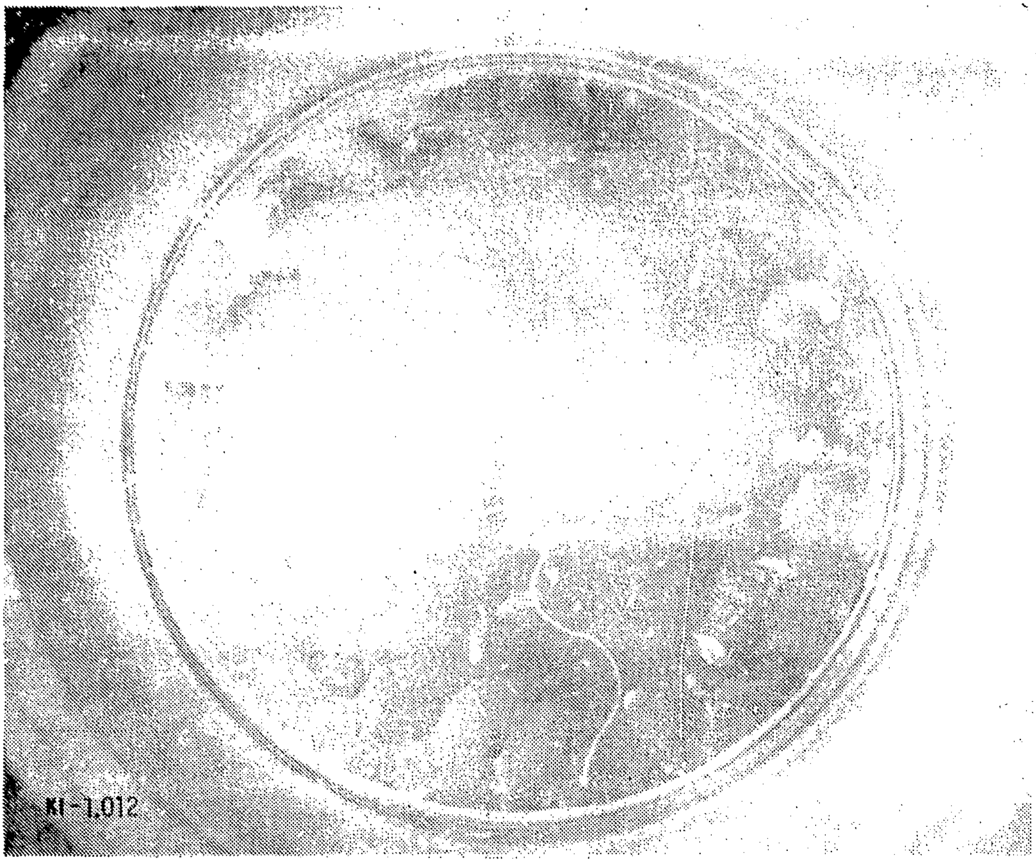

Figure 3. Cusped Ice Interface for Melting Underneath KI Solution; Laminar Region

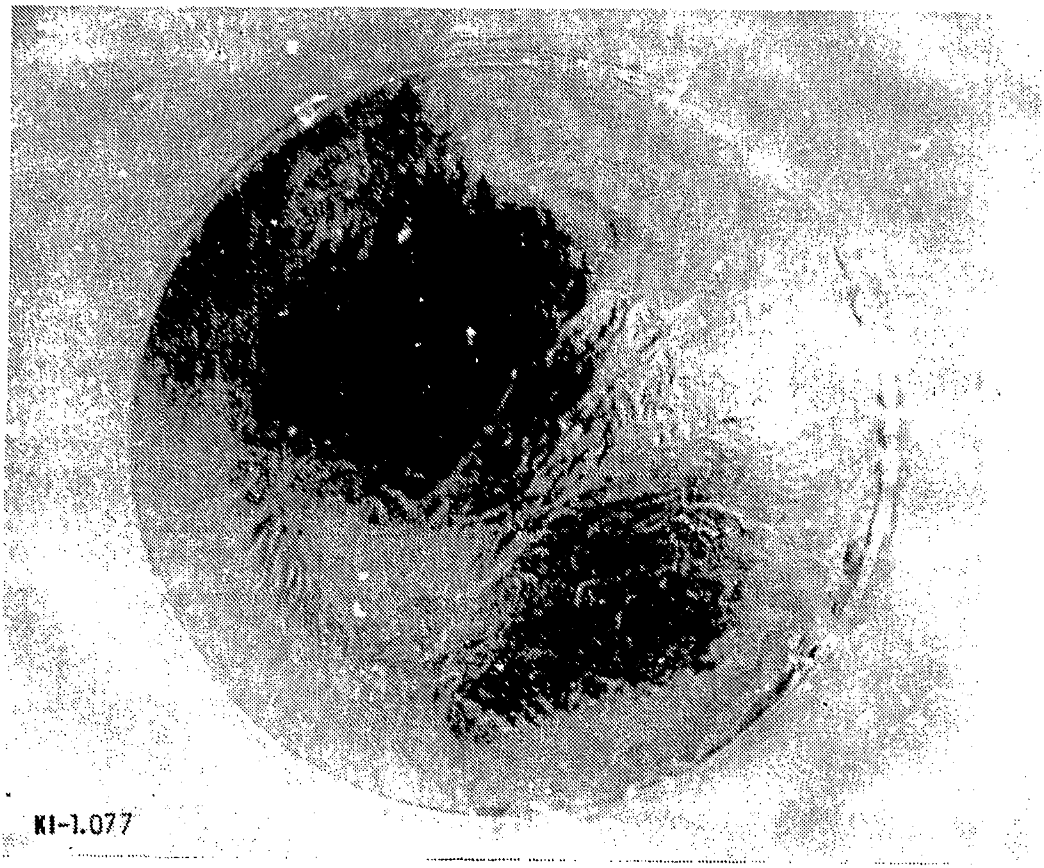

Figure 4. Cusped Ice Interface for Melting Underneath KI Solution; Transition Region

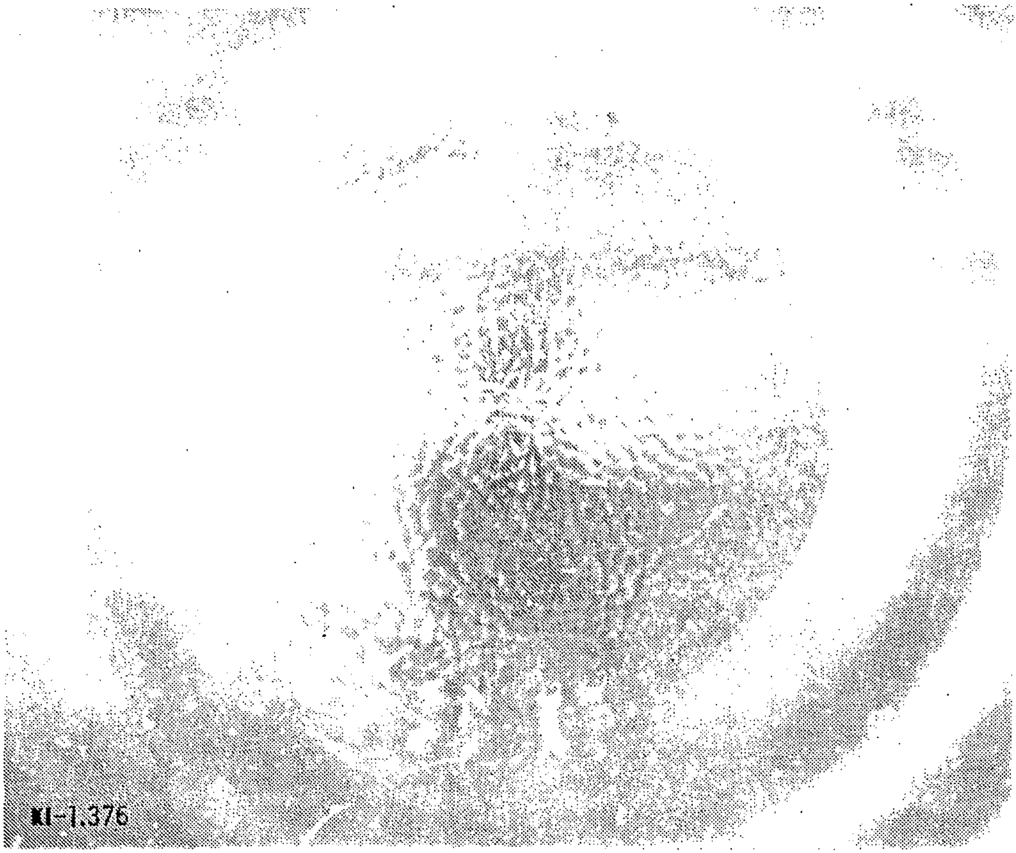

Figure 5. Cusped Ice Interface for Melting Underneath KI Solution; Turbulent Region

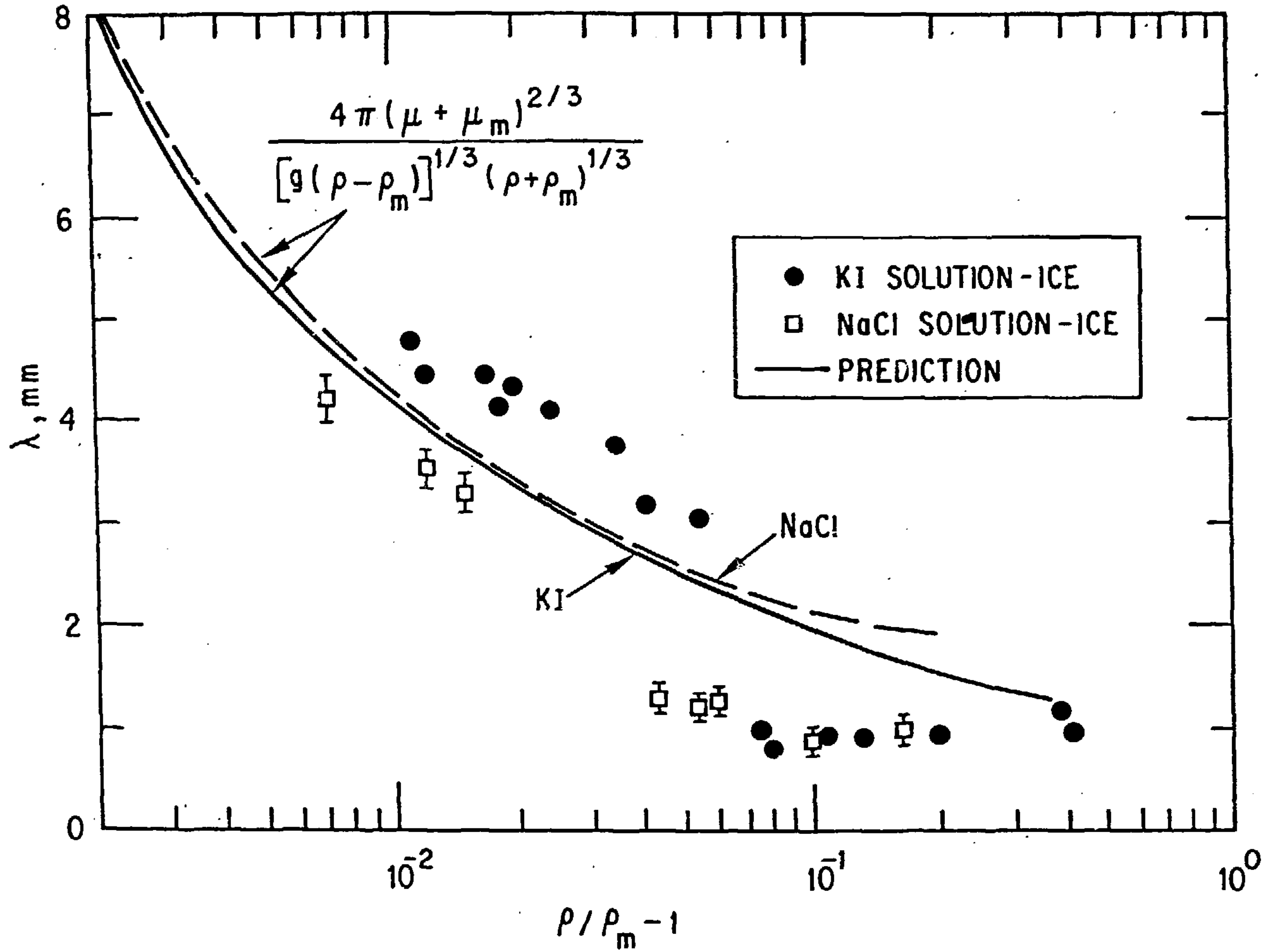

Figure 6. Comparison of the Predicted Wavelengths with Experiments for KI or NaCl Solution on top of Ice.

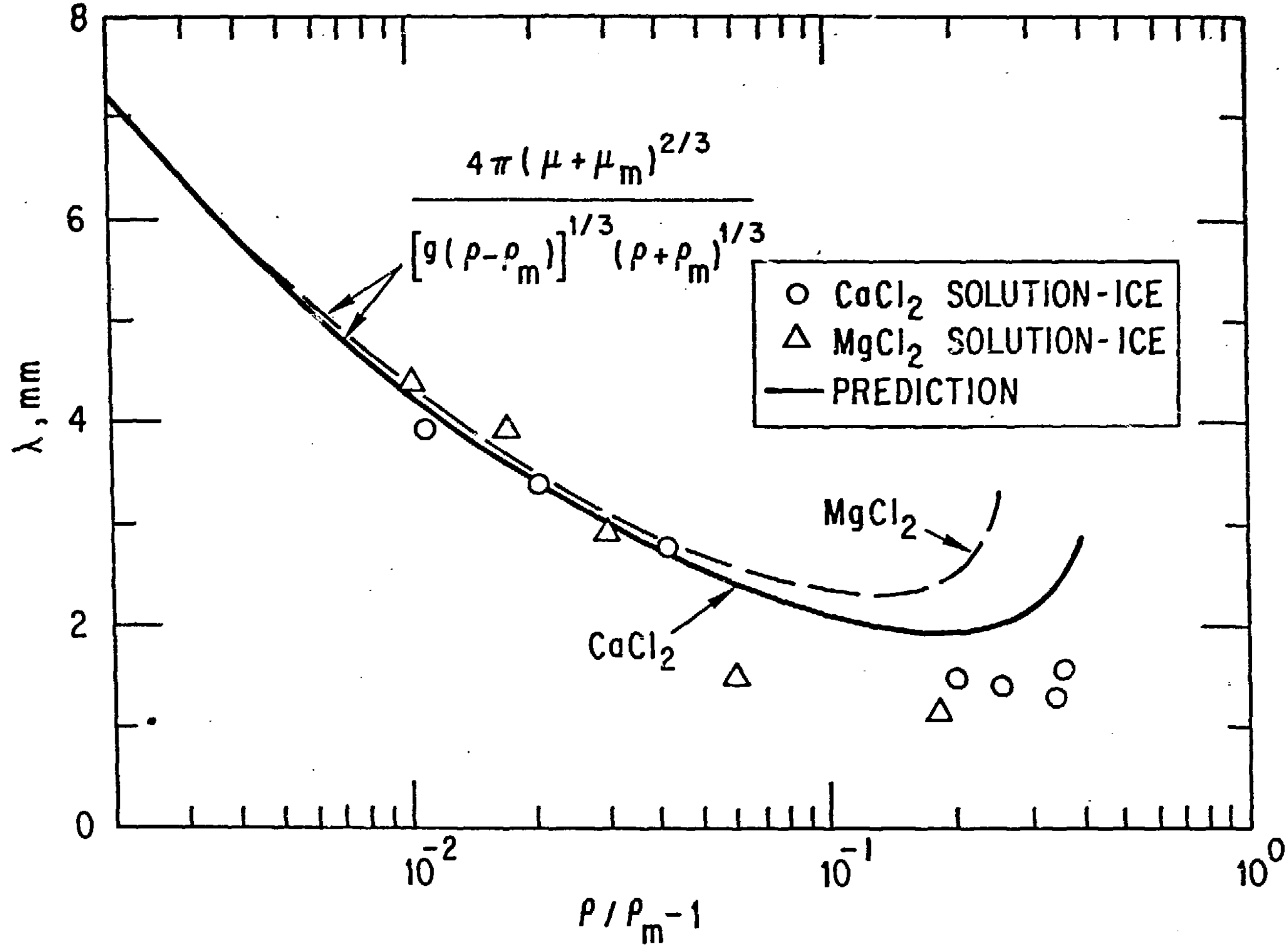

Figure 7. Comparison of the Predicted Wavelengths with Experiments for $MgCl_2$ or $CaCl_2$ Solution on top of Ice.

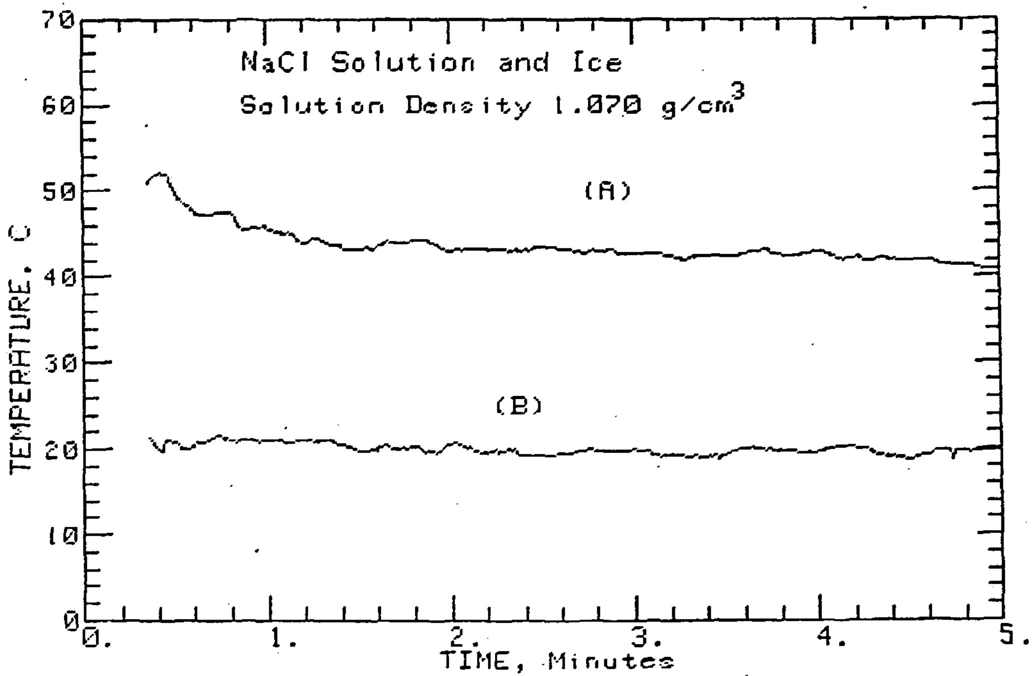

Figure 8. Temperature Transient of the Pool During the Melting process; (20 s. ≤ time ≤ 300 s.)

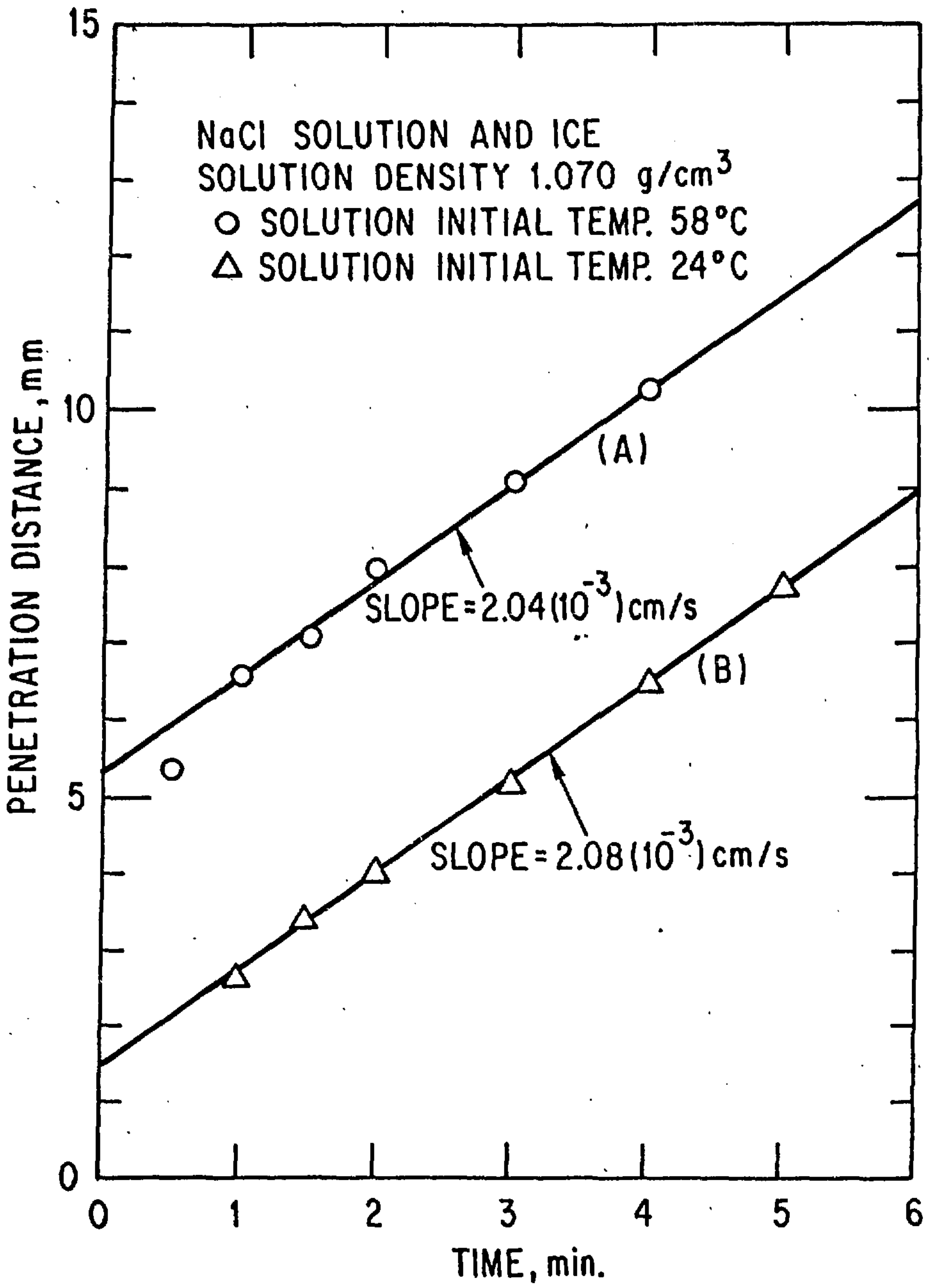

Figure 9. Penetration Distance vs. Time Curve for Two Runs with Same Initial Pool Density.

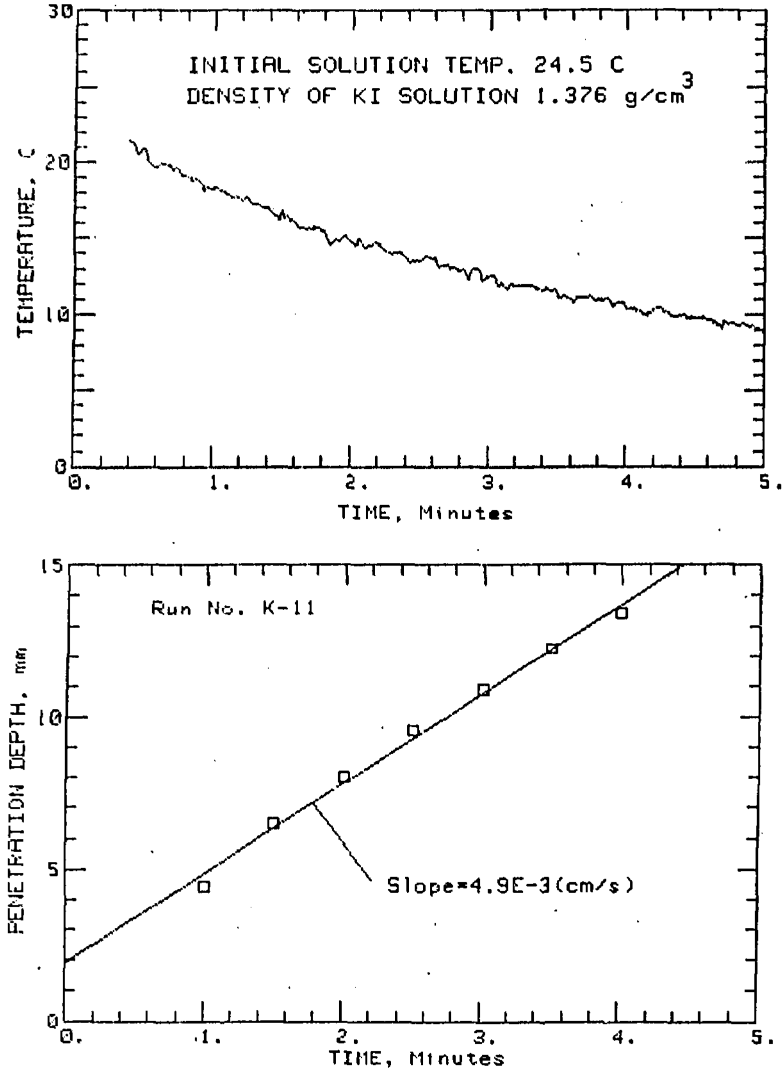

Figure 10. The Bulk Pool Temperature and Penetration Depth vs. Time Curves for Run No. k-11.

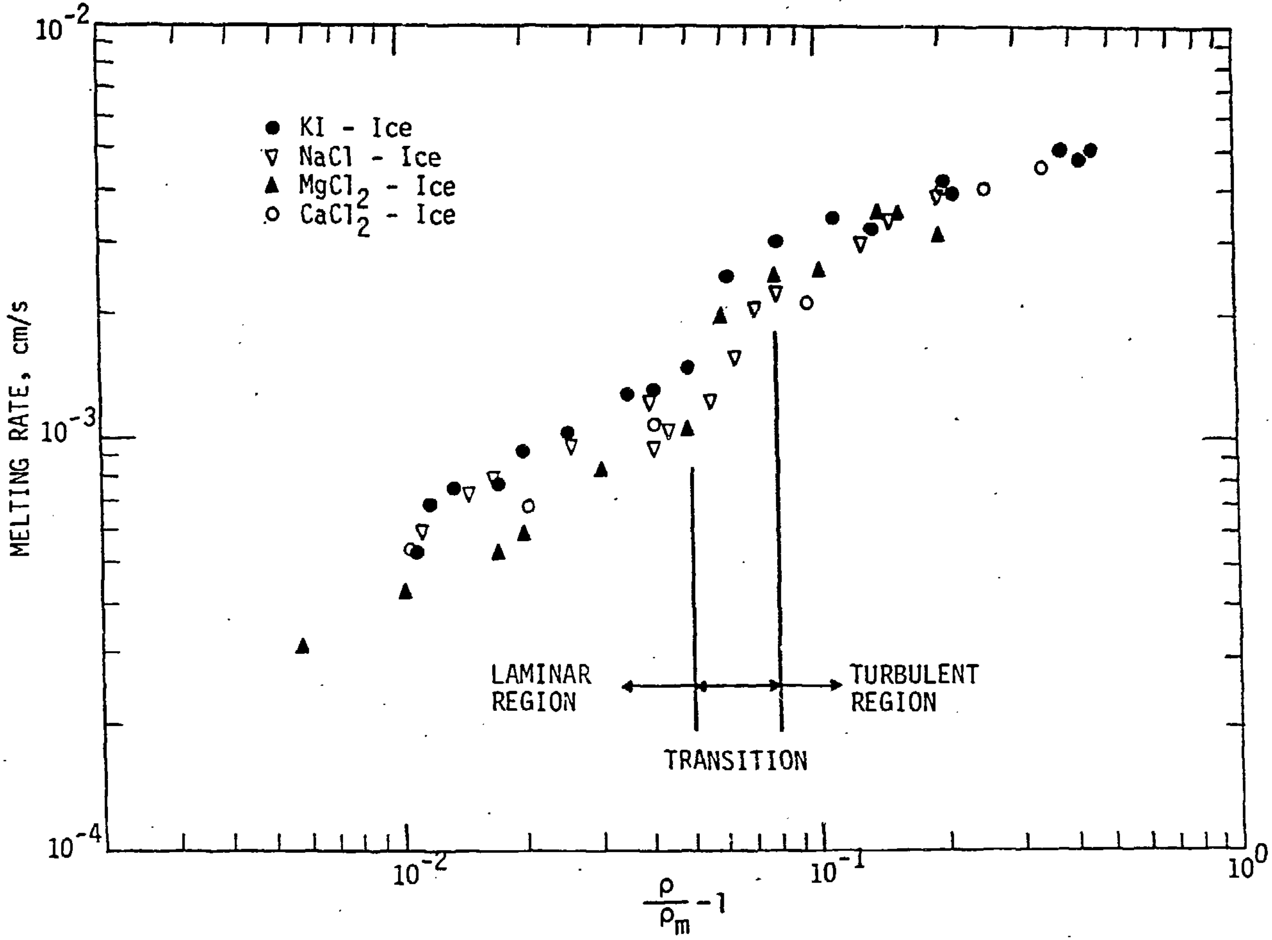

Figure 11. The Observed Melting Rate Dependence on Pool Density in the Three Different Flow Regimes.

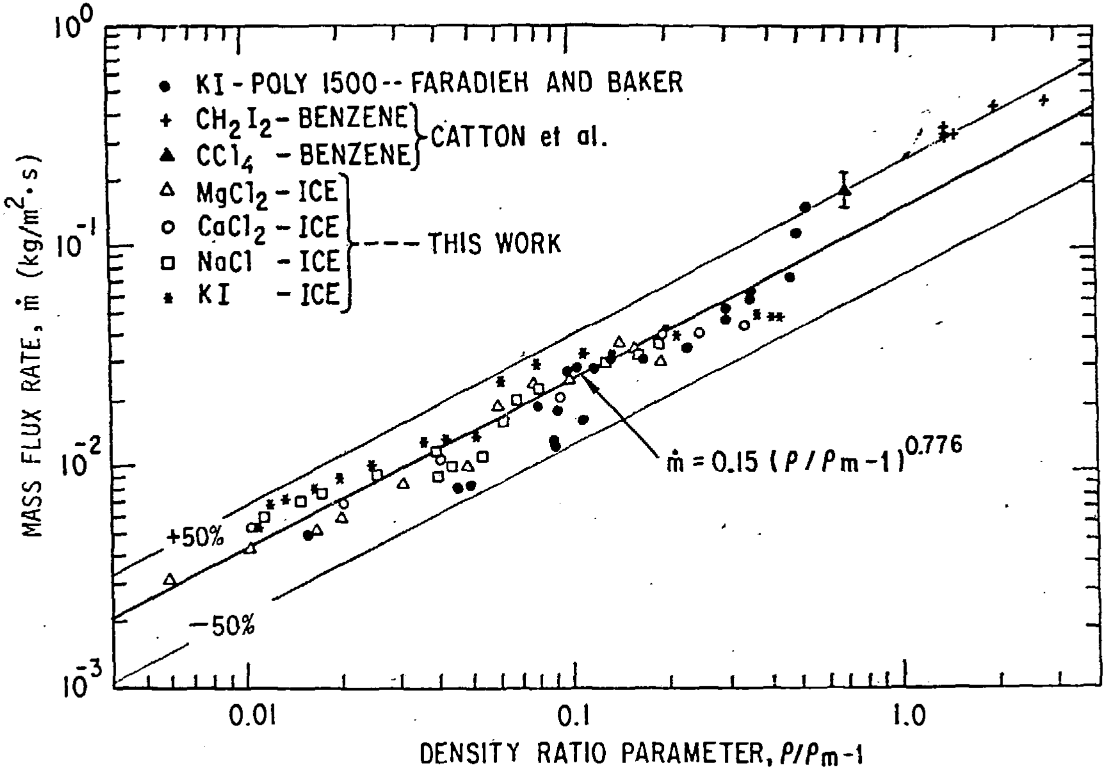

Figure 12. Correlation of the Mass Flux Rate of Melting as a Function of the Pool Density.