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AN INVESTIGATION OF THE FREEZING OF SUPERCOOLED LIQUID IN FORCED TURBULENT FLOW INSIDE CIRCULAR TUBES

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Abstract—A mathematical model is developed to predict the maximum supercooling that can be obtained in a liquid in forced flow inside a circular tube as a function of local tube wall temperature, tube inside diameter, Reynolds number, and a dimensional constant that has been determined from experimental data. An experiment has been carried out to record the maximum supercooling that can be obtained in water. The experimental data have verified the analytical predictions to a good extent. An attempt has also been made to predict the conditions under which flowing supercooled water will freeze.

NOMENCLATURE

C, dimensional constant [deg C/cm];
D, i.d. of tube [cm];
f, Blasius friction factor;
N_{Re}, $\frac{VD}{\nu}$, Reynolds number [dimensionless];
Q, volumetric flow rate;
T_b, bulk liquid temperature [deg C];
T_f, freezing temperature of liquid [deg C];
T_i, initial bulk temperature of liquid flowing through coil, before it is submerged in cooling bath [deg C];
T_{bmin}, minimum bulk temperature of liquid after coil is submerged in cooling bath [deg C];
T_n, effective nucleation temperature [deg C];
T_w, local tube wall temperature [deg C];
ΔT, $T_b - T_f$ [deg C];
ΔT_{bath}, $T_f - T_w$ [deg C];
ΔT_{sup}, degree of supercooling, $T_f - T_b$ [deg C];
V, mean velocity of liquid [cm/s];
δ_b, laminar boundary-layer thickness [cm];
δ_f, boundary-layer thickness at which T_f occurs [cm];
δ_n^*, boundary-layer thickness at which T_n occurs [cm];
δ_n, minimum boundary-layer thickness for nucleation to occur [cm];
ν, kinematic viscosity [cm²/s].

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INTRODUCTION

MANY researchers have investigated the solidification of a liquid flowing through a tube cooled from outside [1–4] and attempts have been made to predict the rate of heat transfer, radius of liquid–solid interface and pressure drop. A common assumption is that the temperature at the liquid–solid interface is equal to the liquid freezing temperature. However, Savino and Siegel's [5] experimental results have revealed the existence of a layer of supercooled water at a temperature as low as −3C in contact with the wall of a chilled flat plate, prior to nucleation. Fernandez and Barduhn [6] have also measured the growth rate of ice crystals in flowing supercooled water in a tube.

A survey of the literature reveals that little research has been done to predict the supercooling that can be obtained in a liquid before it solidifies when under forced flow inside circular tubes. The purpose of this work was to obtain such information using tap water as the fluid and to determine the effect of various parameters on maintaining a constant degree of supercooling. A mathematical model has been developed to predict the maximum supercooling for a given coolant temperature, tube inside diameter, and Reynolds number. The experimental data has been compared with the theoretical predictions and an attempt has been made to explain the deviation.

ANALYSIS

A one-dimensional model of the boundary layer and temperature profile of a liquid flowing through a circular tube is shown in Fig. 1. Liquid

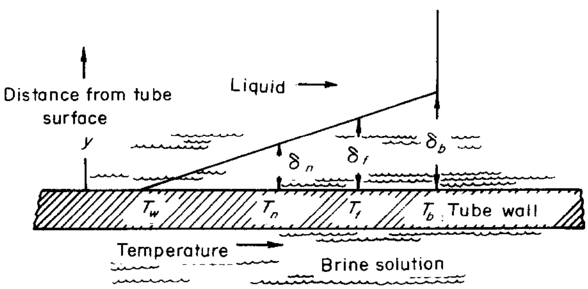

FIG. 1. One-dimensional thermal layer profile for liquid flowing through a tube.

is shown flowing over the tube surface and the tube is cooled convectively from outside by a continuously stirred brine solution. Linear temperature and velocity profile are assumed in the laminar boundary layer.

Assume that ice nucleation will ordinarily occur first at the wall and further crystal growth will occur within the relatively quiescent laminar boundary layer rather than within the turbulent core. The nucleus will then grow along established molecular chains into the flowing fluid.

For ice crystals to begin formation, assume that the temperature within a distance δ_n from the wall must be below a critical nucleation temperature T_n. The value of δ_n may be determined by a minimum critical molecular chain length which will allow crystallization to follow the chain or by some other microscopic phenomenon. If δ_n is less than this value, the crystals will not be nucleated.

Since linear temperature and velocity profiles have been assumed in the laminar boundary layer, the effective boundary-layer thickness, δ_n^*, at which T_n occurs is given by

$$\delta_n^* = \delta_b\left(\frac{T_n - T_w}{T_b - T_w}\right) > \delta_n. \quad (1)$$

From Knudsen and Katz [7], the laminar boundary-layer thickness, δ_b, for flow in a circular tube is given by

$$\delta_b = \frac{5D}{N_{Re}}\left(\frac{2}{f}\right)^{\frac{1}{2}}. \quad (2)$$

The Blasius friction factor equation relates the nondimensional friction factor to the Reynolds number by

$$f = \frac{0{\cdot}079}{(N_{Re})^{\frac{1}{4}}} \text{ for } 3000 < N_{Re} < 100000. \quad (3)$$

Then substituting (3) into (2), the laminar boundary layer thickness, δ_b, is given by

$$\delta_b = \frac{5D}{N_{Re}}\left(\frac{2}{0{\cdot}079}\right)^{\frac{1}{2}}(N_{Re})^{\frac{1}{8}}\ \delta_b = \frac{25{\cdot}2D}{(N_{Re})^{\frac{7}{8}}}. \quad (4)$$

Substituting (4) into (1), it follows that, for ice formation and growth to occur,

$$\delta_n^* = \frac{25{\cdot}2D}{(N_{Re})^{\frac{7}{8}}}\left(\frac{T_n - T_w}{T_b - T_w}\right) > \delta_n. \quad (4a)$$

At this point, let us consider 2 cases.

Case I. Supercooled bulk fluid

Rearranging equation (4a) and defining

$$T_f - T_w = \Delta T_{\text{bath}}$$

and the degree of supercooling of the bulk fluid

$$T_f - T_b = \Delta T_{sup}$$

gives

$$(T_n - T_w) > \frac{\delta_n (N_{Re})^{\frac{7}{8}}}{25\cdot2D}(\Delta T_{bath} - \Delta T_{sup}). \quad (5)$$

The degree of supercooling of the bulk fluid, ΔT_{sup}, is thus given by

$$\Delta T_{sup} > \Delta T_{bath} - \frac{(T_n - T_w)\, 25\cdot2D}{\delta_n (N_{Re})^{\frac{7}{8}}}. \quad (6)$$

For a given wall temperature and liquid; T_n, T_w and δ_n are all constants. Therefore, define a dimensional constant C as

$$C = \frac{25\cdot2\,(T_n - T_w)}{\delta_n}.$$

It can be seen that C is proportional to a critical temperature gradient, $(T_n - T_w)/\delta_n$, that must be exceeded for the nucleation of ice to begin.

Then equation (6) gives

$$\Delta T_{sup} > \Delta T_{bath} - \frac{CD}{(N_{Re})^{\frac{7}{8}}}. \quad (7)$$

The relationship given by equation (7) applies for $3000 < N_{Re} < 100000$. It predicts the degree supercooling required in the bulk fluid for ice formation to occur.

The survey of literature has revealed no attempt to determine the effective nucleation temperature, T_n, and boundary layer thickness, δ_n, at which T_n occurs during initial ice crystal nucleation. Also, T_w is a variable and is a function of cooling bath temperature and other factors.

The constant C must at present be determined from experimental data.

Case 2. Bulk fluid above the freezing temperature

Another form of the equations results for the case in which the bulk fluid temperature is above the nominal freezing temperature ($T_b > T_f$). In this case, supercooling may still exist within the boundary layer; however, the bulk fluid will not be supercooled. For supercooling to occur, $\delta_n < \delta_n^* < \delta_f$.

The boundary-layer thickness, δ_f at which T_f is reached, is given by

$$\delta_f = \delta_b \left(\frac{T_f - T_w}{T_b - T_w}\right)$$

or

$$\delta_f = \delta_b \left[\frac{T_f - T_w}{(T_f - T_w) + (T_b - T_f)}\right]. \quad (8)$$

Substitute

$$T_f - T_w = \Delta T_{bath}$$

$$T_b - T_f = \Delta T.$$

Equation (8) gives,

$$\delta_f = \delta_b \left[\frac{\Delta T_{bath}}{\Delta T_{bath} + \Delta T}\right]. \quad (9)$$

Substituting the value of δ_b from equation (4), the point within the boundary layer at which the freezing temperature is reached, δ_f, is given by

$$\delta_f = \frac{25\cdot2D}{(N_{Re})^{\frac{7}{8}}}\left(\frac{\Delta T_{bath}}{\Delta T_{bath} + \Delta T}\right). \quad (10)$$

Applying the supercooling condition that $\delta_f > \delta_n$ gives

$$25\cdot2\,(\Delta T_{bath}) > \delta_n (N_{Re})^{\frac{7}{8}} (\Delta T_{batn} + \Delta T)$$

or

$$\Delta T < \Delta T_{bath}\left[\frac{25\cdot2D}{\delta_n (N_{Re})^{\frac{7}{8}}} - 1\right] \quad (11)$$

or

$$(T_b - T_f) < (T_f - T_w)\left[\frac{25\cdot2D}{\delta_n (N_{Re})^{\frac{7}{8}}} - 1\right]. \quad (12)$$

Equation (12) predicts that the bulk fluid temperature must differ from the freezing temperature of the fluid by less than the amount predicted by the right-hand side of the equation, or ice will not form even though the tube wall itself is below the freezing point. No experimental verification of this case is presented here, but such experiments appear attractive for determining the value of δ_n.

Experimental data on the degree of supercooling versus Reynolds number are necessary to further study the applicability of the analytical model and to evaluate the constant C. Such data were obtained, and will now be discussed.

EXPERIMENTAL APPARATUS AND RESULTS

The experimental apparatus consisted of a copper tube (0·96 or 0·79 cm o.d.) which could be submerged in a cooling bath. A schematic diagram of the apparatus is shown in Fig. 2.

A pump was used to recirculate melted ice or unprepared tap water. The flow rate was obtained before the coil was submerged in the cooling bath by using a stop watch and a measuring flask. The cooling bath consisted of a mixture of brine solution and dry ice.

Usually the temperature of a liquid flowing inside a tube is measured by a thermocouple placed inside the tube, but in this experiment, placing a thermocouple inside the copper tube would have merely provided a surface nucleation site on which supercooled water would have frozen. Such a thermocouple is not expected to record a temperature below 0°C even when supercooled water is flowing in the tube. Therefore, the temperature measuring arrangement for this experiment consisted of a thermocouple soldered outside a 36 cm long piece of the same copper tube as was used for the main coil. This tube was thermally isolated from the main coil by a small length of the teflon tube of the same i.d. and wall thickness as the copper tube. Swagelok ferrule type fittings were used for all tube connections to minimize the presence of surface nucleation sites. The copper tube with soldered thermocouple was connected within 60 cm of the outlet of the coil from the cooling bath. The entire length of tube from outlet to 60 cm downstream from the thermocouple was

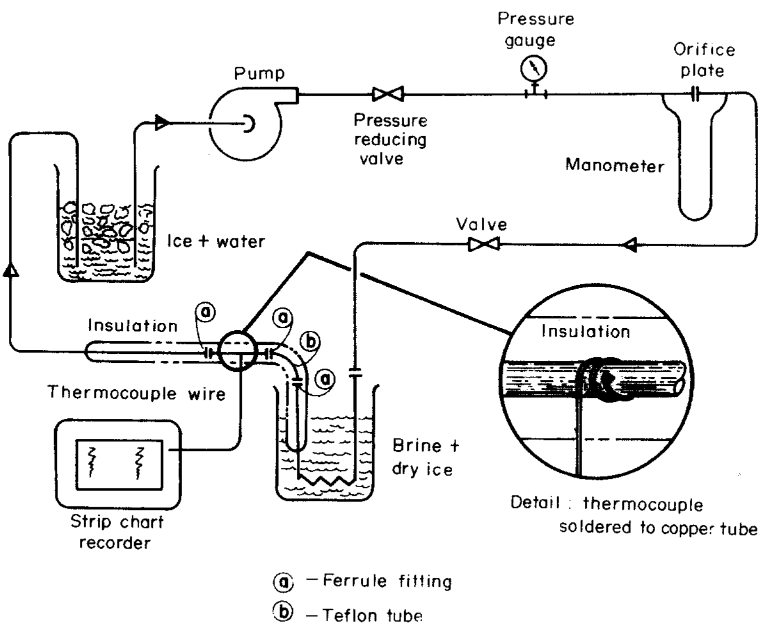

FIG. 2. Schematic diagram of experimental apparatus.

well-insulated using glass wool so as to minimize the effect of ambient temperature on the temperature reading of supercooled water. The temperature was recorded on a strip chart recorder. The Reynolds number was varied by varying the flow rate. An attempt was made to obtain the maximum supercooling for the same flow rate (i.e. approximately the same Reynolds number) by varying the temperature of brine solution and by continuous agitation of brine solution. An attempt was also made to maintain a constant degree of supercooling in the water flowing through the tube but it was not very successful.

Ice and water mixture was used as the reference temperature for the copper–constantan thermocouple. The mixture was stirred every few minutes and was observed to be at 0°C. But, still, there is a possibility of an error up to ±0·1°C in the temperature readings. The Reynolds number is estimated to be correct within ±2·5 per cent.

The diameter of the tube was measured and assumed constant. The flow rate of water flowing through the tube was measured by an orifice meter and assumed constant. For the calculation of supercooling, the freezing temperature of unprepared salt-free tap water, T_f, is considered as 0°C.

It has been assumed that the minimum temperature measured by the thermocouple after the coil is submerged in the brine solution, $T_{b_{min}}$, is the bulk water temperature. To support this assumption, an analysis was performed for the data shown in Fig. 3c. In this case the flow rate, and therefore, the flow velocity is small. The differential temperature between the initial bulk water temperature, T_i (which is also the initial tube wall temperature) and the minimum bulk water temperature after the coil is submerged in the cooling bath, $T_{b_{min}}$, is large. In this particular case the calculated time for this differential temperature to come within 0·1°C is about 5 s. However, most of the experimental data is for larger flow rates and smaller differential temperatures. Therefore, the time taken for the wall and bulk temperatures to become nearly equal will be shorter.

The calculations have demonstrated that $T_{b_{min}}$ measured by the thermocouple is very close to the actual bulk water temperature at the time of maximum supercooling.

A few typical readings are shown in Fig. 3. The

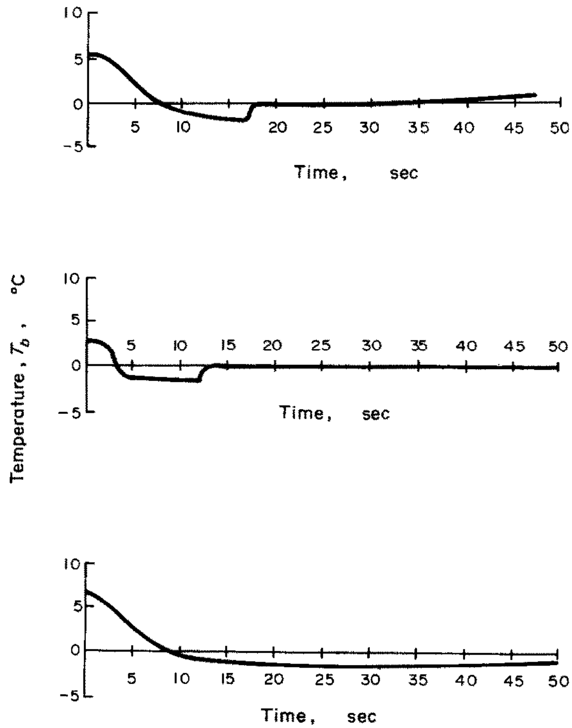

FIG. 3. Bulk liquid temperatures at outlet of cooled tube.
(a) Inlet temperature, 6·2°C; tube diameter, 0·79 cm; flow rate, 2·3 l/min.
(b) Inlet temperature, 3·3°C; tube diameter, 0·79 cm; flow rate, 7·8 l/min.
(c) Inlet temperature, 7·5°C; tube diameter, 0·79 cm; flow rate, 2·0 l/min.

effect of the heat capacity of the tube wall on the water temperature at the thermocouple position was calculated for the data shown in Fig. 3c and is shown in Fig. 4.

COMPARISON OF ANALYSIS AND EXPERIMENT

The experimental data of Arora [8] is

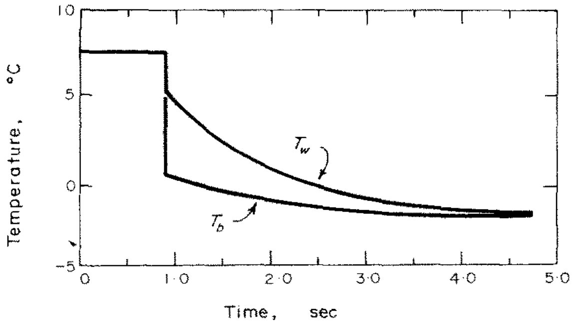

FIG. 4. Comparison of predicted wall temperature, T_w, as affected by a step change in bulk liquid temperature T_b, at entrance to measuring section.

presented in graphical form in Fig. 5. The data points of greatest supercooling for a given Reynolds number for two tube diameters are joined to form the limiting region curves. For comparison the theoretical curves of required supercooling are also shown in Fig. 5. The theoretical curves are for cooling bath temperature held constant at −7°C. Zero degree supercooling is assumed for Reynolds number 3000 and for 0·79 cm o.d. tube. The Reynolds number is based on the water properties at the temperature T_i.

It was observed during the experiment that supercooling did not occur when the Reynolds number was less than 3000. It was also observed that to obtain any supercooling for Reynolds number between 3000 and 3500, the cooling bath temperature was about -7 ± 1°C.

To evaluate the dimensional constant C for the analysis (equation (7)) it was assumed that for 0·79 cm dia tube, supercooling ΔT_{sup} is zero at Reynolds number 3000 and cooling bath bulk temperature −7°C. Assuming that the cooling bath bulk temperature and the local tube wall temperature, T_w, are equal.

$$\Delta T_{bath} = T_f - T_w = 0 - (-7) = +7°\text{C}.$$

Inside diameter of tube = 0·792 cm − 0·157 cm = 0·635 cm. Substituting $\Delta T_{sup} = 0$, $N_{Re} = 3000$, $D = 0{\cdot}635$ cm and $\Delta T_{bath} = 7{\cdot}0$°C in equation (7), we obtain

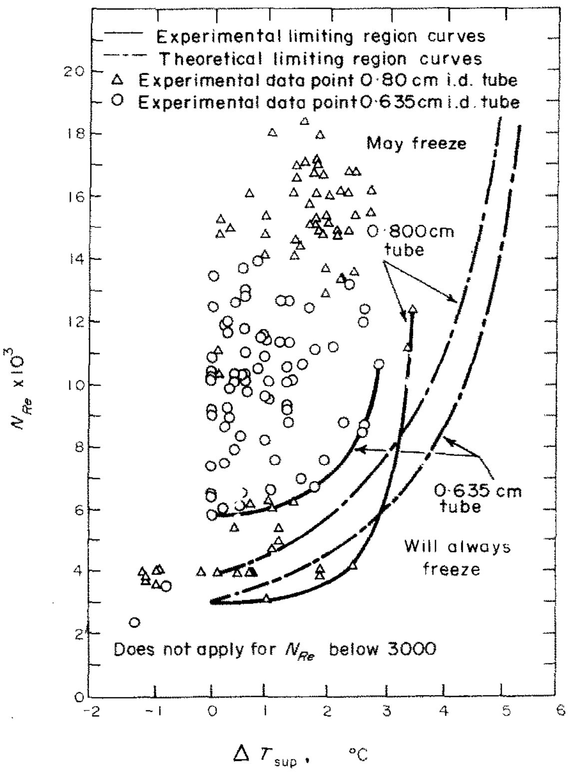

FIG. 5. Comparison of experiment and theory for ice formation from a supercooled liquid.

$$0 = 7{\cdot}0 - \left[\frac{C(0{\cdot}635)}{(3000)^{\frac{7}{8}}}\right]$$

$$C \approx \frac{7{\cdot}0(3000)^{\frac{7}{8}}}{0{\cdot}635}$$

$$= 1{\cdot}21 \times 10^4 \text{ deg C/cm}.$$

Based on this value of $C = 1{\cdot}21 \times 10^4$, the critical temperature gradient is found to be

$$\frac{T_n - T_w}{\delta_n} = \frac{1{\cdot}21 \times 10^4}{25{\cdot}2} \text{°C/cm} = 480\text{°C/cm}. \quad (13)$$

This value, of course, applies only to the particular wall temperature and geometry studied here. If either T_n or δ_n can be determined from other studies, then the other can be determined from equation (13).

The predicted supercooling for this particular value of C can be calculated as follows:

At $N_{Re} = 6000$

$$\Delta T_{\text{sup}} = +7{\cdot}0 - \left[\frac{1{\cdot}21 \times 10^4 \times 0{\cdot}635}{(6000)^{\frac{7}{8}}}\right]$$

$$= +7{\cdot}0 - 3{\cdot}9 = 3{\cdot}1°\text{C}.$$

Similarly for 0·96 cm outside diameter tube and 0·08 cm wall

$$D = 0{\cdot}80 \text{ cm}$$

$$\Delta T_{\text{sup}} > 7{\cdot}0 - \left[\frac{1{\cdot}21 \times 10^4 \times 0{\cdot}80}{(6000)^{\frac{7}{8}}}\right]$$

$$> 7{\cdot}0 - 4{\cdot}8 = 2{\cdot}2°\text{C}.$$

The theoretical required supercooling for this value of C can be predicted as a function of Re and D as shown in Fig. 5.

The data points fall generally below the prediction. This may be due to the fluctuations in boundary-layer thickness that generally occur in highly turbulent flows. Such fluctuations would provide the δ_n values necessary for ice growth to occur at random intervals for N_{Re} below those predicted. Also, other mechanisms than the one postulated here may trigger the freezing process at smaller subcoolings leaving the predictions of this paper as a maximum supercooling that can be attained.

DISCUSSION AND RECOMMENDATIONS

An experimental investigation of water supercooling in forced flow inside a circular tube performed in this work agrees relatively well with the analytical predictions. The required degree of supercooling, ΔT_{sup}, is found to be a function of Reynolds number, N_{Re}, tube i.d., D, and the local tube wall temperature, T_w. As predicted, more supercooling was obtained at higher Reynolds numbers, but contrary to expectations more supercooling was observed for the 0·96 cm tube than for the 0·79 cm tube. With the present state of the art no satisfactory explanation is available. In any case, it has been confirmed that significant supercooling can be obtained and recorded in a forced turbulent flow inside a circular tube. The shape of the curve compares favorably with the predicted one.

Care was taken to obtain a flow path free of nucleation sites within the limits of the commercial fittings and tubes used. The maximum degree of supercooling obtained experimentally is shown in Fig. 5. It was obtained by varying the path temperature between $-12°$C and $-7{\cdot}0°$C for Reynolds numbers below 10000; and between $-18°$C and $-12°$C for Reynolds numbers above 10000. In any case, the maximum degree of supercooling obtained for Reynolds numbers above 6000 is less than the degree of supercooling predicted theoretically for a cooling bath bulk temperature of $-7{\cdot}0°$C. One of the factors contributing to this deviation could be the assumption made to evaluate the dimensional constant, C. It was assumed that local tube wall temperature, T_w, is $-7{\cdot}0°$C for the cooling bath bulk temperature of $-7{\cdot}0°$C whereas in reality there may be a significant thermal gradient between the two. Such a difference in T_w will significantly affect the analytical prediction of equation (7).

It was also observed that under seemingly the same experimental conditions a different degree of supercooling was obtained for the same Reynolds number and for the same cooling bath bulk temperature. This could be the effect of fluctuating boundary layer thickness, or of numerous nucleation sites existing in the flow path of supercooled water, any one of which could trigger the complex phenomenon of freezing. In most cases, the freezing occurred very abruptly. Two typical examples are shown in Figs. 3a and 3b. The readings such as shown in Fig. 3c where the supercooling was maintained for a length of time, were very few.

Another observation was that in most cases, supercooling was obtained within a few seconds of submerging the coil in the brine solution. The flow rate then dropped, the bulk fluid temperature suddenly increased and ice pieces were observed coming out with water. These observations agree well with those of Savino and Seigel [5].

No appreciable success was achieved in maintaining a constant degree of supercooling in water flowing through the cooled tube. Also, from the experimental data obtained, it is rather difficult to predict exactly when the flowing supercooled water will freeze. However, there is a distinct demarcation of the "may freeze" and "will always freeze" zones. As shown in Fig. 5, above the curve, the water may freeze; but below the curve, it will always freeze. That is why these curves have been termed as limiting region curves.

The effects of various solutes such as air entrapped in water are not known. It is quite possible that supercooling occurs in laminar flows but the equipment used for this work was not sufficiently sensitive to record the small supercooling temperature for those small flow rates.

This work has amply demonstrated the necessity of developing more elaborate equipment to study this problem. An attempt is being made to develop the apparatus that will permit visual observation of the phenomenon. The present work is an initial effort on a very complex problem which needs to be studied in detail. Besides using distilled water as the fluid, future work will be done to study the effects of various solutes and impurities on the degree of supercooling obtained for water in turbulent flow. Various liquids will be used.

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ETUDE DE LA CONGELATION D'UN LIQUIDE SUR-REFROIDI EN ECOULEMENT TURBULENT FORCE A L'INTERIEUR DE TUBES CIRCULAIRES

Résumé—Un modèle mathématique permet d'estimer le sur-refroidissement maximal possible dans un liquide en écoulement forcé à l'intérieur d'un tube circulaire, fonction de la température locale de la paroi du tube, du diamètre intérieur de celui-ci, du nombre de Reynolds et d'une constante empirique sans dimension. On a mené des expériences pour déterminer le sur-refroidissement maximal qui peut être obtenu dans l'eau. Les résultats expérimentaux s'accordent avec les estimations analytiques. En outre un essai a été fait pour estimer les conditions dans lesquelles gélera un courant d'eau sur-refroidie.

UNTERSUCHUNGEN DES GEFRIERENS VON UNTERKÜHLTER FLÜSSIGKEIT BEI ERZWUNGENER STRÖMUNG IN KREISROHREN.

Zusammenfassung—Es wurde ein mathematisches Modell entwickelt, um die maximale Unterkühlung vorauszusagen, die man in einer Flüssigkeit mit erzwungener Strömung in einem Kreisrohr erhalten kann als Funktion einer örtlichen Rohrwandtemperatur, des Rohrinnendurchmessers, der Reynoldszahl und einer dimensionsbehafteten Konstanten, die experimentell zu bestimmen ist. In einem Versuch wurde die maximale Unterkühlung aufgezeichnet, die man mit Wasser erhalten kann. Die experimentellen Meßwerte haben die analytischen Voraussagen gut erfüllt. Es wurde auch versucht, die Bedingungen vorauszusagen, unter denen fließendes unterkühltes Wasser gefrieren wird.

ИССЛЕДОВАНИЕ ЗАМЕРЗАНИЯ ПЕРЕОХЛАЖДЕННОЙ ЖИДКОСТИ ПРИ ВЫНУЖДЕННОМ ТЕРБУЛЕНТНОМ ТЕЧЕНИИ В КРУГЛОЙ ТРУБЕ

Аннотация—Разработана математическая модель для расчета максимально возможного переохлаждения жидкости при вынужденном течении в круглой трубе в зависимости от локальной температуры стенки трубы, внутреннего диаметра трубы, числа Рейнольдса и безразмерной константы, определяемой по экспериментальным данным. Эксперимент проводился с целью измерения максимально возможного переохлаждения воды. Теоретический расчет довольно хорошо подтверждается экспериментальными данными. Была сделана попытка определить заранее, при каких условиях замерзает поток переохлажденной жидкости.