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HEAT TRANSFER BETWEEN IMMISCIBLE LIQUIDS ENHANCED BY GAS BUBBLING

MASTER

by

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ABSTRACT

The phenomena of core-concrete interactions impact upon containment integrity of light water reactors (LWR) following postulated complete meltdown of the core by containment pressurization, production of combustible gases, and basemat penetration. Experiments have been performed with non-reactor materials to investigate one aspect of this problem, heat transfer between overlying immiscible liquids whose interface is disturbed by a transverse non-condensable gas flux emanating from below. Hydrodynamic studies have been performed to test a criterion for onset of entrainment due to bubbling through the interface and subsequent heat transfer studies were performed to assess the effect of bubbling on interfacial heat transfer rates, both with and without bubble induced entrainment. Non-entraining interfacial heat transfer data with mercury-water/oil fluid pairs were observed to be bounded from below within a factor of two to three by the Szekeley surface renewal heat transfer model. However heat transfer data for fluid pairs which are found to entrain (water-oil), believed to be characteristic of molten reactor core-concrete conditions, were measured to be up to two orders of magnitude greater than surface renewal predictions and are calculated by a simple entrainment heat transfer model.

BACKGROUND

The phenomena of core-concrete interactions impact upon containment integrity of a light water reactor (LWR) following postulated complete meltdown of the core by containment pressurization due to condensable and non-condensable gas generation, possible ignition of combustible gases, and concrete basemat penetration. In order to develop a predictive capability to analyze such complicated interactions, the CORCON code [1] has been developed at Sandia Laboratory under USNRC sponsorship.

Modeling of core-concrete interactions involves many poorly understood and complicated heat transfer phenomena for which there exists a sparse data base. In support of the CORCON development effort, one heat transfer aspect of core-concrete interactions has been investigated which had been found to have significant impact upon the results of generic code calculations, namely the phenomenon of heat transfer between overlying immiscible liquid layers whose interface is agitated by gases liberated from the underlying concrete.

The model used in CORCON to characterize liquid-liquid heat transfer to an interface agitated by transverse gas flow is given by a correlation developed by Konsetov

[2] and modified by Blottner [3] as

$$h = k\left(\frac{\Pr g}{\nu^2}\right)^{1/3} (.00274\ \beta\Delta T + .4\alpha^2)^{1/3} \qquad (1)$$

There had been only one heat transfer data point for comparison to this correlation for a slag-metal system [4] and this was found to be greater in magnitude than the calculated heat transfer coefficient by a factor of three. However, a limited amount of data with an oil-water fluid pair were available from KFK [5] and these data are shown along with the predictions of the modified Konsetov model as well as a model developed by Grief [6] and the surface renewal heat transfer model of Szekeley [4] in Figure 1. In this figure, it is clear that the data greatly exceed the Konsetov model in magnitude even at modest superficial gas velocity of less than 1 cm/s by as much as two orders of magnitude. A more realistic larger estimate of the bubble radius would tend to increase the disagreement between the Szekeley model and the data as well.

This apparent modeling deficiency was chosen for study and a sensitivity analysis was performed. Generic CORCON code calculations were run for the Zion plant input data deck. The effect of interfacial heat transfer was examined parametrically, by increasing the heat transfer coefficient by a factor of 10 and 100, chosen on the basis of comparison of the heat transfer models to the limited KFK data. It was found that the integrated results of the core-concrete interactions were significantly affected by these parametric variations on the interfacial heat transfer coefficient. It was found that the heat transfer model given by Equation (1) always gave an upper bound to the generation rates of H_2, CO, H_2O and CO_2 gases; however, increasing the magnitude of the coefficient by factors of 10 and 100 would reduce these gas liberation rates by as much as a factor of from two to five. The reason for this effect on the gas release rates from the concrete is that the downward heat flux into the concrete from the heavy core oxide layer is reduced due to the increased upward heat flux into the overlying lighter metallic layer. This reduced downward heat flux similarly reduces the concrete ablation rate and, for the parametric calculations performed with the Zion plant input deck, reduced the rate of dilution of the lower oxide layer by concrete slag. Accompanying the calculation of reduced gas generation rates and reduced concrete ablation rate by increasing the interfacial liquid-liquid heat transfer coefficient, it was found that the layer temperatures themselves would cool significantly faster with the increase in the magnitude of the interfacial heat transfer coefficient, so fast in fact that layer inversion may be postponed until solidification of the corium melt occurred. An example of the effect of the magnitude of the interfacial liquid-liquid heat transfer coefficient on the temperature history of the core oxide layer for the Zion input deck calculations is shown in Figure 2. On the basis of these observations, the experimental and analytical program about to be described was begun.

DESCRIPTION OF EXPERIMENT

An experimental apparatus to investigate heat transfer between immiscible liquid layers with gas agitation was constructed. A schematic diagram of the apparatus is shown in Figure 3. The apparatus is a liquid pool constructed of plexiglass and lexan. It is approximately 13 cm by 22 cm in cross section and 40 cm deep. A porous frit is mounted in the base to provide a spatially uniform distributed gas flux. The porous frit is installed in such a manner that it may be easily interchanged with another of a different pore size. Electrodes are installed in the side walls to provide internal heating of the lower liquid layer. The electrodes are connected to a calibrated watt meter for accurate determination of power dissipation in the pool. For the cases in which the lower fluid is a liquid metal, the electrodes can be replaced by a submerged cartridge heater. The porous frit is connected to a bank of air rotameters and a turbine flow meter for accurate and duplicate measurement of the gas superficial velocity. A vertical traversable thermocouple rake is installed along the center line axis of the pool for determination of the temperature distribution in the pool. Twelve thermocouples are installed at a nominal separation of one inch. All thermocouples were calibrated from the ice to steam point with an accuracy of 0.1 C. All instrumentation is interfaced to the Hewlett Packard series 1000 mini computer-based data acquisition system. The entire apparatus, with the exception of the surface, is insulated with one inch thick polystyrene sheet to insure one dimensional heat transfer and minimize boundary heat losses, which have been estimated to be 2-3%.

Experiments were performed with silicone oil-water and water-mercury fluid pairs. Thermophysical properties were measured whenever possible (density, viscosity, surface tension); when not feasible, vendor supplied or literature values were utilized. The apparatus was charged with the fluids and power supplied via the electrodes. The temperature distribution in the liquids was monitored until steady state conditions were achieved at a prescribed gas flux. The overall interfacial heat transfer coefficient was calculated as the electrical power supplied divided by the overall temperature difference between the liquid layers and the cross sectional area. The superficial gas velocity was the volume gas flux divided by the cross sectional area of the pool. Tests were performed with zinc sulfate-silicone oil (Series 100) fluid pairs with a density ratio of 0.80, copper sulfate-silicone oil (Series 200) fluid pairs with a density ratio of 0.65, and water-mercury (Series 300, 400) fluid pairs with a density ratio of approximately 0.1. In the discussion that follows, these are compared to water-oil and Wood's metal-oil data of Werle [5, 7, 8] with density ratios of approximately 0.9 and 0.1, respectively. The data to be presented indicate an average of up to 20 separate experiments at nominally the same conditions of power and gas flux.

EXPERIMENTAL RESULTS AND COMPARISON TO MODELS

Mercury-Water Bubbling Interfacial Heat Transfer

Two sets of bubbling heat transfer data were taken with mercury-water fluid pairs, Series 300 and Series 400 data. The bubble radii were in the range 0.3 to 0.5 cm and the superficial gas velocity was varied over the range from zero to 1.4 cm/sec. These data are presented in Figure 4 along with the Wood's metal-oil data of Werle. In the limit of zero gas flow rate, these data converged asymptotically to a lower limit calculated by the conducting-sheet model of Haberstroh [9] as would be expected. A brief description of the conducting-sheet heat transfer model is given below. At zero gas flux, the liquid-liquid interface is not disturbed and the heat transfer is controlled by turbulent natural convection in each layer. The layers are observed to be nearly isothermal and the temperature gradients are restricted to the interface region. For both liquid pairs the data were found to agree with the conducting-sheet model of Haberstroh within an uncertainty band characteristic of the uncertainty in the thermophysical properties.

As the superficial gas velocity increased, the heat transfer coefficient similarly increased due to the periodic bubble-induced disturbances at the liquid-liquid interface. The vertical temperature distribution demonstrated a sharp gradient in the vicinity of the fluid-fluid interface, suggesting that the interface did maintain its approximate spatial integrity and that mixing and entrainment were absent. These observations were further supported by visual and photographic evidence of the absence of entrainment of mercury even under intense interfacial disturbance.

The mercury-water heat transfer data were found to be greater in magnitude than the Wood's metal-oil data (KFK) by a significant margin. The mercury and Wood's metal layers have negligible thermal resistance to heat transfer in comparison to the water and oil layers and for calculational purposes may, thus, be neglected. The observed superiority of the water layer to the oil layer in transferring heat is evident from the data in Figure 4, and the ratio is roughly a factor of five increasing to as much as ten. On the basis of the surface renewal formulation shown in Figure 4, this ratio should be approximately four, however, as will become evident in the discussion, there are factors absent from this formulation which, when included, may account for this discrepancy.

The regime of heat transfer between two fluid layers enhanced by interfacial disturbances generated at their interface by rising bubbles with the absence of entrainment is referred to as the surface renewal regime. A brief description of the surface renewal model is given. When the gas flux is initiated, the interfacial heat transfer coefficient is found to increase above the value characteristic of pure steady natural convection. For the mercury-water case, no entrainment of the mercury is observed into the overlying water layer. When the bubble breaks the interface, a droplet forms behind the bubble from the metal film that previously surrounded it [10]. For this liquid pair, the density difference between the liquids is great enough that the droplet remains at the interface while the bubble rises through the water. Evidently, the buoyancy of the bubble is not sufficient to carry the drop of mercury upward. For this case, the bubble acts only to disrupt the temperature gradients at the interface and transient conduction acts to renew the gradients until the arrival of a subsequent bubble. This procedure of thermal gradient destruction by discrete bubble agitation with no accompanying mass transfer or significant physical alteration of the interface is referred to as surface renewal. The mercury-water and Wood's metal-oil data are characterized by this model.

The major assumptions of the surface renewal model are that a rising bubble totally destroys the temperature gradients on both sides of the interface only in the area of impact projected by the bubble, no influence is felt outside the bubble area, and surface disturbances do not enhance the transport mechanisms or the interfacial surface area. As is evident from Figure 4, the surface renewal model of Szekeley [4], modified by Blottner [3],

$$h_{SZE} = 1.69\ k\ (j_g/\kappa r_b)^{1/2} \tag{2}$$

represents a lower bound to both the mercury-water data as well as the Wood's metal-oil data. In both cases the deviation between the measured and calculated heat transfer coefficient increases with increasing superficial gas velocity, indicating the effect of the increasing disturbance intensity and interfacial wave propagation on the magnitude of the heat transfer. The fact that the discrepancy is greater for the water-mercury data than for the oil-Wood's metal data may indicate the presence of a Prandtl number effect in addition to the hydrodynamic interfacial stretching mechanism due to instability formation.

Nevertheless, for fluid pairs that do not mix or entrain even under the influence of transverse gas bubbling through their interface, the simple transient conduction surface renewal model is found to predict a lower limit to the magnitude of the interfacial heat transfer coefficient, differing from the measured data by up to a factor of four over the range of conditions covered by these experiments.

Water-Oil Bubbling Interfacial Heat Transfer

In addition to the liquid metal-oil/water interfacial heat transfer experiments which did not exhibit entrainment over the entire range of gas velocity covered, experiments were also performed with zinc sulfate-silicone oil (Series 100) and copper sulfate-silicone oil (Series 200) fluid pairs which did demonstrate entrainment and mixing when their interface was agitated by rising bubbles from below. The bubble radii and superficial gas velocity were in the same range as for the liquid metal-oil/water experiments. The data for the Series 100 and 200 oil-water experiments are presented in Figure 5 along with the oil-water data of Werle. These experiments, all experiencing liquid-liquid entrainment effects, are compared to the Wood's metal-oil data previously discussed. Since all these data sets, entraining or non-entraining, involving silicone oil have the resistance to heat transfer concentrated in the oil layer, this comparison is a test of the contribution to the net heat transfer of the entrainment process versus the surface renewal process.

Once again it is evident that, in the limit that the superficial gas velocity asymptotically goes to zero, these data converge to the natural convection conducting-sheet limit represented by the Haberstroh model within an uncertainty band characteristic of the uncertainty in the thermophysical properties.

As the superficial gas velocity was increased, a dramatically different behavior was observed than for the fluid pairs which did not exhibit entrainment. Instead of a gradual increase in magnitude, the heat transfer coefficient is seen to suddenly jump almost a factor of ten at the onset of bubbling and steeply increase until, at a gas velocity of 1 cm/s, it is greater than the silicone oil-Wood's metal data by more than two orders of magnitude. The measured vertical temperature distribution exhibited characteristics of an intermediate mixing zone in which the temperature gradually changed from one layer to the other. This is in contrast to the sharp temperature gradient measured with non-entraining fluids previously. These observations were further supported by visual and photographic evidence of the severe mass entrainment rate even at the modest superficial gas velocities below 1 cm/sec.

The significant increase in interfacial heat transfer for the silicone oil-water fluid pair over that measured for the silicone oil-Wood's metal fluid pair (KFK) is attributed directly to the effect of mass entrainment of the hot lower fluid across the interface into the cold upper fluid. This regime of heat transfer is referred to as the entrainment heat transfer regime [12]. A detailed discussion of the modeling of entrainment heat transfer is given in the Appendix and will only be briefly mentioned here.

As the bubble penetrates the liquid-liquid interface, a finger of the lower heavy fluid is sucked upward into the upper layer in the bubble wake region. At some location, this liquid finger is observed to pinch off; the fluid below the point of the break returns downward through the interface, while the fluid above this point continues to entrain upwards in the wake region of the bubble. In the case of large entrained drops, they are also observed to fragment in the vortex region behind the bubble into smaller droplets, greatly increasing the surface area for heat transfer. On the basis of simple analysis of transient convective heat transfer around a sphere [11], it can be shown that for the conditions of these experiments, the droplets essentially transfer all their excess enthalpy to the upper fluid prior to settling back to the lower fluid layer from whence they came. On the basis of these observations, it is argued that one only need to know the liquid entrainment rate in order to calculate the entrainment heat transfer rate. In this fashion, the overall heat transfer coefficient can be written as the sum of the interfacial surface renewal contribution and the entrainment contribution as

$$h_{eff} = h_{SZE} + j_2 \rho_2 C_{p2} \tag{3}$$

where j_2 is the volumetric entrainment rate of the lower fluid per unit cross sectional area. At present, calculation of the liquid entrainment rate, j_2, is treated parametrically as a function of the gas superficial velocity, $j_2 = C_2 j_g$. In reality it is recognized that C_2 is not a constant but is a function of j_g itself. For this discussion, C_2 is assigned the values 0.3 and 1.0 awaiting further attempts to improve the entrainment rate model which are currently underway in recognition of the obvious non-linear relationship between j_g and j_2.

The results of the comparison of Equation (3) to the oil-water entrainment data are shown in Figure 5. Note that the choice of C_2 in the range 0.3 to 1.0 appears to bracket the available data. The development of a more refined entrainment rate model will enable a more mechanistic calculation of entrainment heat transfer rate.

Nevertheless, for fluid pairs that exhibit interfacial mixing and entrainment under the influence of transverse gas bubbling through their interface, a simple entrainment rate heat transfer model is seen to reasonably bracket the available experimental data when appropriate assumptions regarding the mass transfer rate are incorporated. This limitation is expected to be relaxed when a mechanistic entrainment rate model for j_2 is available.

DISCUSSION

In the case of several immiscible overlying liquid layers agitated at their interface by gases rising from below, there are two regimes of interfacial heat transfer that may occur.

One is the surface renewal regime in which the gas flux serves to enhance the heat transfer by disrupting the steady state temperature gradients at the interface. This mode of heat transfer is characterized by the absence of liquid entrainment and mixing, and presently the mathematical model that appears to do the best job of predicting the available experiment data is the Szekeley surface renewal heat transfer model given by Equation (2). This model consistently underpredicts the available experimental data and the degree of disagreement increases to as much as a factor of four at the highest superficial gas velocities examined. It is expected that this disagreement would increase with a further increase in gas flux. Accounting for interfacial area enhancement due to surface waves and enhanced radial heat transfer would be expected to improve the model predictions and bring them into significantly better agreement with the data. Efforts to do this are currently underway. At this point, the Szekeley model is recommended over either the Konsetov or Grief models for calculation of interfacial heat transfer with bubbling for conditions under which liquid entrainment is negligible.

The other mode of heat transfer that has been observed is the entrainment heat transfer regime. This regime is characterized by entrainment of the lower fluid into the upper layer in the form of droplets driven by bubble-induced hydrodynamic instabilities at the interface. The magnitude of the entrainment heat transfer rate has been measured to be as much as two orders of magnitude greater than for the case without entrainment. An entrainment heat transfer model (Equation (3)) has been developed which, at this time, requires some estimate of the volumetric entrainment rate. Realistic parametric estimates of the entrainment rate for the oil-water experiments described have done an encouraging job of bracketing the available heat transfer data. Parallel efforts are underway to refine the entrainment model which appears to predict the potential for onset of entrainment well but severely overpredicts the rate of entrainment. Under conditions which the entrainment onset model predicts entrainment, this model is recommended instead of the surface renewal model.

Parametric calculations have been performed with the CORCON code for the Zion plant input deck for the case of a molten reactor core attacking underlying concrete. At the initial debris temperature of 2500K, the density ratio of the metal layer (6.9 gm/cm^3) to the heavy oxide layer (8.0 gm/cm^3) is calculated to be approximately 0.86 and approaches unity as concrete slag is ablated and dissolved in the oxide layer. This represents a case similar to the oil-water experiments which had density ratios in the range 0.65 to 0.80 and were found to be dominated by liquid entrainment. Such behavior is also expected for the case of core oxide-metal interfacial heat transfer during core-concrete interactions. Under such conditions and assumptions, it is clear that the surface renewal heat transfer model will significantly underpredict the upward heat transfer rate. Interfacial liquid-liquid heat transfer must be calculated with the entrainment heat transfer model under these conditions. From the non-reactor materials experiments performed, it was found that the entrainment model would predict a heat transfer coefficient at least two orders of magnitude greater than the surface renewal-type model currently in CORCON. This is identical to the parametric code calculations referenced earlier in this paper. Relying on those parametric code results with enhanced heat transfer, it is clear that modeling of liquid-liquid heat transfer with bubble-induced entrainment represents a mitigating influence on the severity of the consequences of core-concrete interactions from the point of view of containment response. Calculations have shown that increasing the interfacial liquid-liquid heat transfer coefficient by a factor of ten to one hundred would reduce the overall mass of decomposition gases generated by a factor of two as well as accelerate the cooling rates of the oxide and metal layers by as much as 380K and 100K, respectively, over the first two hours of core-concrete interaction. Improvement of the surface renewal model would also result in severely mitigated consequences should calculations not be based upon the presence of entrainment.

NOMENCLATURE

A	cross sectional area
C_1	constant (defined in Equation A.2)
C_2	constant (defined in Equation A.5)
C_p	specific heat
g	gravitational acceleration
h	heat transfer coefficient
j	superficial velocity
k	thermal conductivity
Pr	Prandtl number
r_b	bubble radius
ΔT	temperature difference
α	void fraction
β	coefficient of thermal expansion
κ	thermal diffusivity
ν	kinematic viscosity
ρ	density

Subscripts

2	entrained phase
g	gas phase
SZE	Szekeley model value

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APPENDIX: ENTRAINMENT HEAT TRANSFER MODELING

Current modeling of liquid-liquid entrainment due to gas bubbling is crude at best. Although a model has been developed based upon a static force balance which appears to describe the conditions for the onset of entrainment, calculated entrainment rates have been greater than measured rates in single bubble entrainment tests by approximately a factor of ten. For the purposes of this development, therefore, the entrainment will be treated in a parametric fashion, subject to future refinement in the entrainment rate model.

For the entrainment heat transfer model, it is assumed that the heat transfer is composed of a component due to the interfacial agitation (surface renewal) and a component due to the mass entrainment itself. The Szekeley heat transfer coefficient is given by

$$h_{SZE} = 1.69\, k(j_g/\kappa r_b)^{1/2} \tag{2}$$

The interfacial heat transfer rate (surface renewal) is therefore

$$q_{interface} = h_{SZE}\, A\, \Delta T_{12} \tag{A.1}$$

where A is the interfacial cross-sectional area and ΔT_{12} is the temperature difference between the upper (1) and lower (2) layers. The interfacial area was demonstrated to be enhanced by disturbance formation and wave propagation and, as a result, $q_{interface}$ represents a lower bound to the interfacial heat transfer rate as previously discussed.

The component of the heat transfer due to entrainment can be expressed as the product of the mass rate of entrainment times the excess enthalpy of the entrained phase (2) transferred to the continuous phase (1), that is

$$q_{entrain} = \text{(mass entrainment rate)x(excess enthalpy transferred/unit mass)}$$

or

$$q_{entrain} = (j_2\, A\rho_2)\,(C_1\, C_{p2}\, \Delta T_{12}) \tag{A.2}$$

where j_2 is the superficial velocity of the entraining fluid, ρ_2 is the density, C_{p2} is the specific heat, and C_1 is the fraction of the excess enthalpy of fluid 2 that is transferred to fluid 1. It can be easily shown by analysis of transient convective heat transfer around a sphere, under conditions of the entrainment heat transfer data, that $C_1 \sim 1$ [11].

Combining Equations (A.1) and (A.2), we have

$$q_{total} = h_{SZE}\, A\Delta T_{12} + j_2 A\rho_2\, C_{p2}\, \Delta T_{12} \tag{A.3}$$

If the effective heat transfer coefficient is defined as $h_{eff} = q_{total}/A\Delta T_{12}$, then this reduces to

$$h_{eff} = h_{SZE} + j_2\rho_2\, C_{p2} \tag{A.4}$$

Let $j_2 = C_2 j_g$, where C_2 is the ratio of the volumetric entrainment rate of fluid 2 to the volumetric gas flux. Then Equation (A.4) becomes

$$h_{eff} = h_{SZE} + C_2 j_g \rho_2 C_{p2} \tag{A.5}$$

In the oil-water experiments, the component of the interfacial heat transfer resistance on the water side of the interface can be shown to be negligible with respect to that on the oil side of the interface. As a result the overall interface heat transfer coefficient from the surface renewal model is simply that calculated for the oil. In the general case, where both sides of the interface must be considered, the overall interfacial heat transfer coefficient due to interfacial disturbances (not entrainment) would be

$$\frac{1}{h_{overall}} = \frac{1}{h_{SZE1}} + \frac{1}{h_{SZE2}} \tag{A6}$$

At present, the major uncertainty in applying Equation (A.5) to the case of heat transfer with entrainment is the determination of the coefficient $C_2(j_g)$. Comparison of Equation (A.5) to the available BNL and KFK entrainment heat transfer data suggests that the data are bracketed by the choice of C_2 in the range (0.3, 1.0). Further work is underway to improve this aspect of the entrainment heat transfer model.

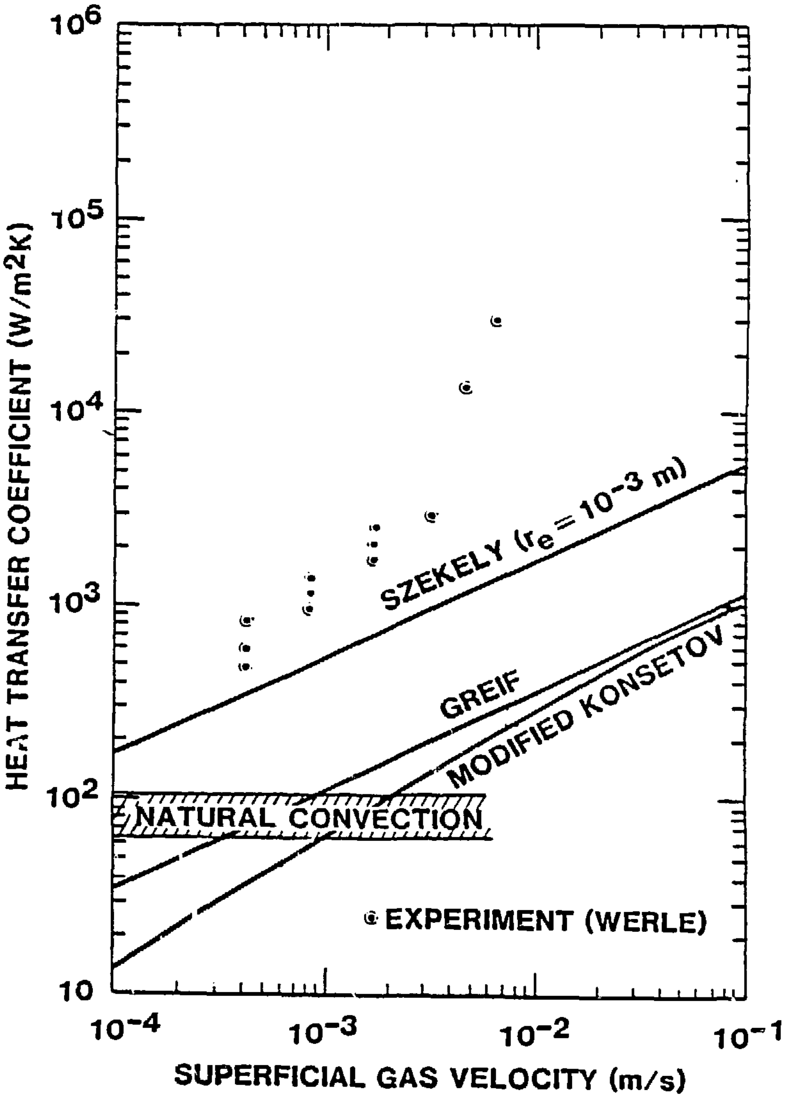

FIGURE 1 - Liquid-Liquid Interface Heat Transfer With Gas Injection-Oil Water (Reference 1)

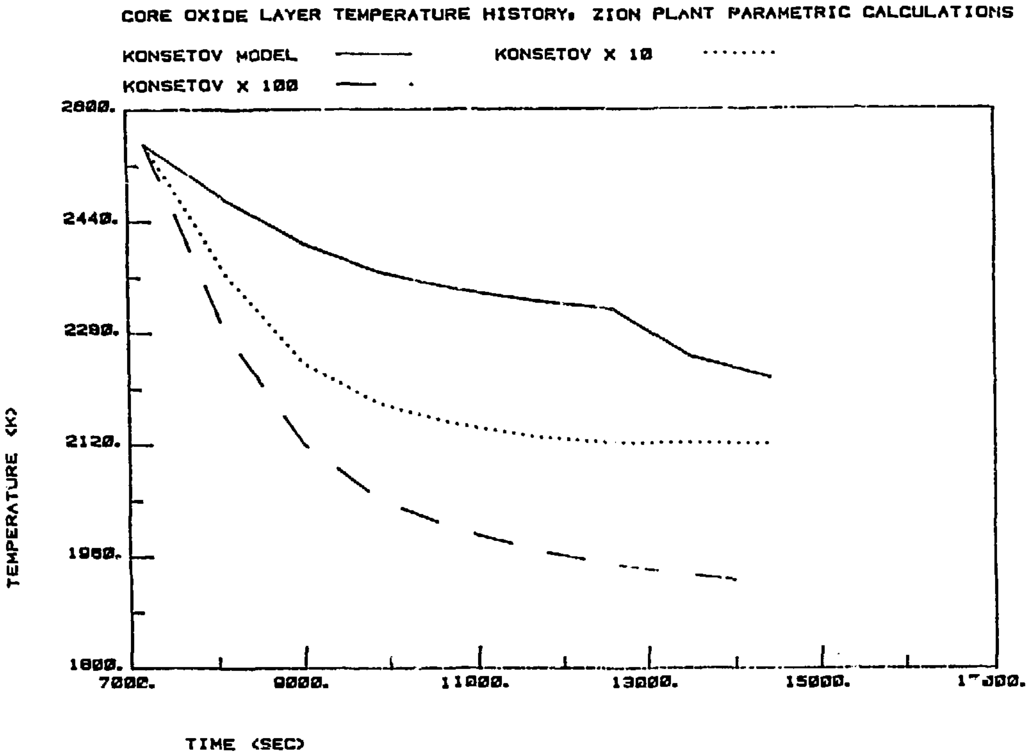

FIGURE 2-Calculated Molten Oxide Layer Temperature vs. Time for Parametric Values of Liquid-Liquid Interfacial Heat Transfer Coefficient

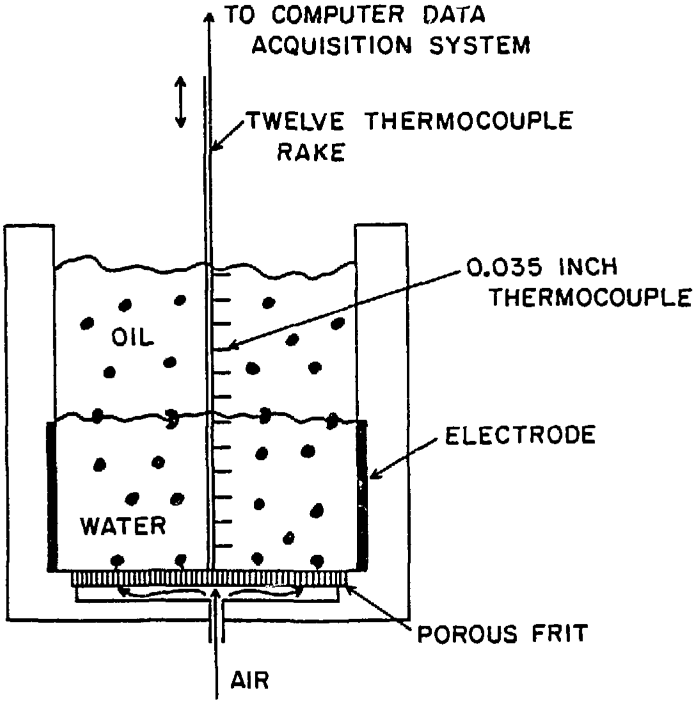

FIGURE 3-Schematic Diagram of Experimental Apparatus

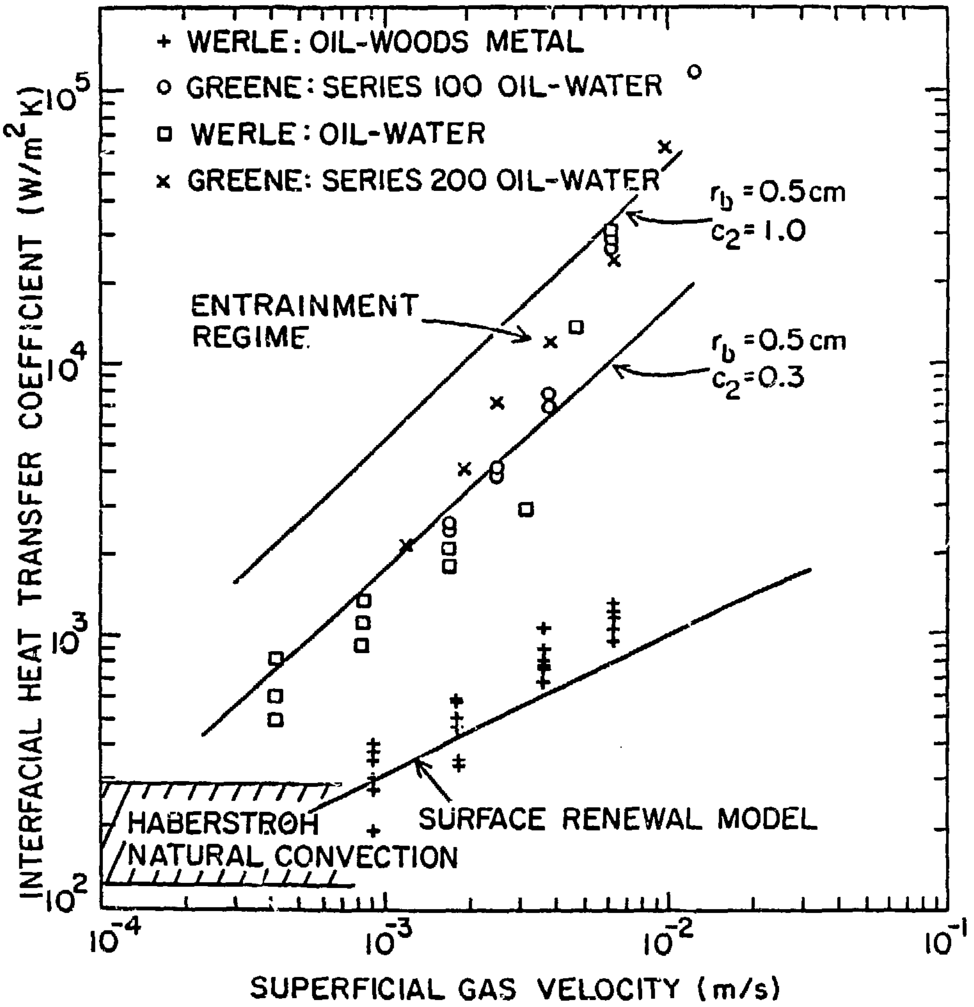

FIGURE 5-Liquid-Liquid Interfacial Heat Transfer Coefficient vs. Superficial Gas Velocity-Entrainment Regime