This paper describes a study of two-phase gas-liquid vertical annular climbing film flow. Using a high speed Fastax movie camera, motion was slowed sufficiently to facilitate detailed analysis of the climbing film structure, based on single frame projections. The dynamic characteristics of the liquid film could likewise be obtained, though these were necessarily of a qualitative nature.

A technique based on a diffusion controlled electrochemical reaction was employed to monitor the local instantaneous mass transfer coefficient at the solid-liquid interface. The resultant fluctuating electrical signal was amplified and presented for direct comparison with the climbing film structure, using split image photography. The signal was also subjected to a spectral density analysis to further understanding of the nature of the turbulence produced at the interface.

The vertical annular column was constructed of a three-inch
inside diameter clear plexiglass tube, with a one-inch outside
diameter concentric core, supported laterally by a series of stream-
lined centering pins. The column extended some 35 feet from the
base with all tests carried out approximately midway up the column
to minimize end effects. The particular construction of the column
permitted an unobstructed view of the free surface of the climbing
film. Determination of the pressure gradients was likewise sim-
plified as the inner surface of the outer tube remained essentially
dry throughout the experimental program.

The air flow rates varied from 172 cfm to 616 cfm at one
atmosphere pressure and 68°F temperature. Liquid flow rates were
confined to a range from .19 lbm/min to 1.5 lbm/min so as to stay
within the climbing film regime.

From the movie film analysis, values for the average film
thickness, wave frequency and phase velocity were determined with
their respective standard deviations. Two empirical models have
been proposed which fit the average film thickness data to within 6%.
The film thickness results were also compared with the values pre-
dicted by several mathematical models, two of which were based on
a modification of Kapitza's original theory.
Motion Picture Study of a Vertical Climbing Film in an Annular Duct

by

Bruce Douglas Scott

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INTRODUCTION

Two-phase gas-liquid flow can be broken down into several sub-categories or flow regimes including, bubble, slug, semi-annular flow and mist. The normal practice in research has been to study each flow regime individually due to the otherwise unwieldy scope of such an undertaking. Film flow encompasses but one of these flow regimes, and is in itself a formidable challenge to the researcher. Though a common phenomenon in the process industry, and more recently as applied to reactor cooling systems, its basic characteristics are as yet poorly understood.

In climbing film flow, a liquid film on a vertical surface is transported up the surface due to interaction with a gas stream, moving tangentially to the liquid. The essential obstacle to a basic understanding of this rather complex phase interaction is in the nature of the waves produced at the free surface of the liquid film, in addition to the specific mechanism involved in momentum transfer from the gas stream to the liquid. Even in falling film flow, this characteristic wave motion has been found (1, 2, 6), thus indicating a possible film interaction at both the solid-liquid and the gas-liquid phase boundaries. As shown by Ruckenstein and Berbente (40), a
considerable increase in the rates of heat and mass transfer may
be expected with the onset of wave motion in the film. Thus knowledge
of the specific nature of the wave flow is necessary for accurate pre-
diction of transfer properties in process equipment.

The study of both climbing and falling film flow has been con-
ducted by previous investigators, variously employing flat vertical
plates, inclined planes, rectangular ducts, and smooth vertical pipes,
the latter alternative being particularly suitable for climbing film
flow. In order to facilitate observation of the climbing film charac-
teristics, a vertical annular column was used in the present study.
This arrangement was such as to permit the liquid to flow up the
inner core of the annulus, while the outer transparent tube wall re-
mained essentially dry. The measurement of pressure gradients was
likewise simplified, as the pressure taps, installed flush with the
outer tube wall were consequently free from error due to the pres-
ence of liquid.

This study was principally concerned with the pictoral record-
ing of a climbing liquid film under the influence of a restricted range
of gas-liquid flow rates. High speed motion picture photography,
up to 2400 frames per second, was used, employing split-image
photography to simultaneously record the apparent local mass trans-
fer rate at the solid-liquid interface, as displayed on an oscilloscope.
Single frame projections, taken individually or in series, were then
analysed in detail to better determine the characteristics of the liquid film.

The determination of the local mass transfer rate at the solid-liquid interface was based on an electrochemical technique, using a ferro-ferricyanide redox couple. The resultant fluctuating signal, being of an electrical nature, was easily displayed for the purposes of simultaneous photography with the liquid film. This electrical signal was further subjected to a spectral density analysis in an attempt to better understand the mechanics of liquid turbulence at the surface of the annular core.

This study is a portion of a continuing basic investigation into annular film flow. Previous findings by Kim (24) and Sutey (42) in this area have been utilized and extended. The results of this study of a quantitative nature have further been compared to theoretical predictions based on several models for film flow.
1.0 PREVIOUS WORK AND THEORY

1.1 Models for Falling Film Flow

Nusselt's solution for a vertically falling liquid film in rectilinear motion (36), considered the simplified equation of motion,

\[ g + \nu \frac{d^2 u}{dy^2} = 0 \]

\[ \frac{du}{dy} \bigg|_{y=0} = 0 \]

\[ \frac{du}{dy} \bigg|_{y=h_0} = 0 \]

Solving for the film thickness,

\[ h_0 = \left( \frac{3 \nu u h_0}{g} \right)^{\frac{1}{3}} \]

Due to the assumption of steady-state, this theory does not hold for wavy motion, but serves as a satisfactory first approximation to the average film thickness.

In order to handle the more complex versions of the equations of motion, numerical techniques have been utilized. Nakaya and Takaki (33) thus dealt with the stability of the falling film in wave motion on an inclined plane. The most comprehensive theoretical study to date appears to be that of Massot, et al. (31). They
essentially extended Kapitza's theory, including the term, $\frac{\partial^2 u}{\partial x^2}$, in the equations of motion, which Kapitza had previously neglected. The solution of the linearized Navier-Stokes equations in three dimensions was carried out numerically in order to compute the instantaneous stream lines of the falling film. Thus, the surface contour as well as the instantaneous velocity profiles could be determined directly.

1.2 Models for Climbing Film Flow

Willis (43) proposed a mathematical model of the vertical climbing film assuming laminar flow through a tube of circular cross section. Working from a force balance on the liquid film,

$$\frac{1}{r} \frac{\partial}{\partial r} (r \tau_r) = \rho_L \frac{\partial P}{\partial x}$$

(1.2-1)

and considering the assumed laminar shear stress distribution in the liquid, he derived the following relationship between the liquid flow rate and the film thickness,

$$Q = \frac{\pi g h_o^2}{12 \mu_L} (6 \frac{dP}{dx} r_1 (r_1 - h_o) - \rho_L (8 r_1 - 5 h_o) h_o)$$

(1.2-2)

Hewitt, Lacey and Nicholls (18) similarly derived an expression involving the film thickness to the fourth power. Their estimation of the interfacial shear stress was somewhat more refined,
as it included a term covering the static weight of the gas stream in
the force balance. The main concern of these authors however was
the prediction of the conditions necessary for the inception of down-
flow. Experiments carried out by this group confirmed the Willis
theory for a flow regime in which low interfacial shear stresses
existed. Under these conditions, the liquid film flows up the tube
with more nearly laminar characteristics, thus approaching the
conditions assumed in the laminar models.

The well known Lockhart-Martinelli correlation has been found
to be reasonably successful in dealing with annular flow in smooth
tubes (10, 12, 14, 17). Kim (24) however noted a marked deviation
from the predicted trend when considering the climbing film regime.
From his data, it appeared that a transition occurred near a liquid
holdup of 1%.

1.3 Wave Motion in the Liquid Film

In view of the fact that the various climbing film theories break
down under a more turbulent flow pattern, the next phase in analysis
might well account for the wavy motion observed on the film surface.
In dealing with a wavy surface, the problem arises of how to ap-
proach the interfacial momentum transfer from the air stream to the
liquid film.

Several theories have been postulated on the generation and
growth of gravity waves. Jeffreys (22) suggested that the fluid was
in essence pulled along by the air stream as a result of pressure
variation developing across the wave crests. Phillips (37) based his
model on the assumption of random pressure variations due to tur-
bulence in the air stream occurring at the interface. The develop-
ment of these random waves was attributed to natural resonance ef-
facts, where a component of the surface pressure moved in phase
with the free surface wave. Miles (32) postulated a mechanism by
which an existing surface wave of a given phase velocity might grow
exponentially. This model was based on a parallel shear flow from
the air stream. His treatment however required a power series ap-
proximation to the air velocity profile near the interface.

Evaluation of the gas phase forces acting on the liquid surface
is only part of the picture, as the liquid phase response should also
be considered in terms of surface tension, geometry, physical
properties and turbulence characteristics.

The various models for the generation of gravity waves through
wind action are not directly applicable to the prediction of capillary
wave characteristics. In a theoretical development covering the
capillary wave, the influence of a finite solid boundary must be con-
sidered. Surface effects appear to be quite significant from the
results of experiments on the falling film. Waves were generated
in each case without any appreciable air-liquid interaction (1, 2, 6).
Kapitza (23) proposed a theoretical model for flow of thin layers of viscous liquid down a smooth surface, under the influence of gravity. His development appears to have been the first attempt to describe the wavy flow by assuming an oscillatory steady state solution to the equations of motion. The film surface tension was also considered as a contributing factor, being roughly of the same order of magnitude as the other forces involved.

Levich (29) expanded on Kapitza's derivation somewhat, and arrived at an expression for the average film thickness closely resembling Nusselt's result for laminar film flow,

\[ h_o^3 = \frac{3\nu Q}{g} \Phi \]

where

\[ \Phi = \frac{1}{\lambda} \int_{0}^{\lambda} \frac{(1 + \frac{c}{u_o} \phi)^2}{(1 + \phi)^2} \, dx \]  \hspace{1cm} (1.3-1)

The model further predicted the frequency, amplitude, phase velocity and velocity profile for the falling film.

Kim (24) extended this development to climbing film flow by adding a constant term to the equations of motion, describing the air-liquid interfacial shear stress. A minor error has since been found in the latter portion of this derivation, thus negating the final equations reached.

In the present analysis, Kim's development will be corrected,
and the resulting equations mechanized for numerical solution.

1.4 Approximate Solution to Climbing Film Flow

With fluid flow in a thin film up the inner core of an annulus, the curvature may be reasonably neglected in constructing a theoretical model of the liquid film. Thus, the liquid motion can be considered to be quasi-two dimensional. Choosing the x-axis to be in the direction of flow and the y-axis to be perpendicular to the core surface, the equations of motion simplify to the following,

$$\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} = -\frac{1}{\rho} \frac{\partial P}{\partial x} + v \frac{\partial^2 u}{\partial x^2} - g \quad (1.4-1)$$

$$0 = \frac{\partial P}{\partial y} \quad (1.4-2)$$

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \quad (1.4-3)$$

with the four boundary conditions required for solution,

$$\begin{align*}
at \ y = 0, \ u &= v = 0 \\
at \ y = h(x, t), \quad \frac{\partial u}{\partial y} &= \frac{F}{\mu} \\
\end{align*} \quad (1.4-4)$$

$$P = P_\sigma(x, t) + P_\tau(x)$$

The interfacial conditions are unknown at this point, and a constant shear stress at the air-liquid interface is postulated, which causes
the liquid film to climb the inner core. This shear stress can be
considered analogous to momentum loss due to friction in dealing
with fluid flow in rough pipes. The interfacial pressure exerted by
the liquid film must balance the sum of the air pressure, $P_g$, and
the surface pressure, $P_{\sigma}$, due to surface tension. For small sur-
face deformations, curved in only one direction, the surface pres-
sure can be expressed (1) as,

$$P_{\sigma} = -\sigma \frac{\partial^2 h}{\partial x^2}$$  \hspace{1cm} (1.4-5)

As pressure is assumed to be independent of $y$ by Equation 1.4-2,
the pressure term in Equation 1.4-1 may be replaced by the boundary
condition at $y = h(x, t)$. From Equation 1.4-3,

$$v = -\int_0^y \frac{\partial y'}{\partial x} \, dy' = \frac{\partial h}{\partial t}$$  \hspace{1cm} (1.4-6)

combining this with Equation 1.4-5, Equation 1.4-1 becomes,

$$\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} - u \frac{\partial}{\partial y} \int_0^y \frac{\partial u}{\partial x} \, dy' = -\frac{\sigma}{\rho} \frac{\partial^3 h}{\partial x^3} - \frac{1}{\rho} \frac{\partial P_g}{\partial x} - \nu \frac{\partial^2 u}{\partial y^2} - g$$  \hspace{1cm} (1.4-7)

where

$$\frac{\partial h}{\partial t} = -\frac{\partial}{\partial x} \int_0^y u \, dy' \hspace{1cm} (1.4-8)$$

at $y = 0, \ u = v = 0$

at $y = h(x, t), \ \frac{\partial u}{\partial t} = \frac{F}{\mu}$
In order to solve the non-linear differential equation, some estimate of the velocity profile is required. By initially neglecting convective forces relative to the viscous forces, Equation 1.4-7 simplifies to

\[
\nu \frac{\partial^2 u}{\partial y^2} = g - \frac{1}{\rho} \frac{\partial P}{\partial x} = 0
\]

where

\[
\frac{\partial u}{\partial y} \bigg|_{y = h(x, t)} = \frac{F}{\mu}
\]

where

\[
\frac{\partial P}{\partial x} \approx \frac{\Delta P}{\Delta x}
\]

is assumed constant and measurable experimentally. Solution of the above equation yields a first approximation to the velocity profile,

\[
u = \frac{3}{h^2} \left( \frac{h}{2} \frac{F}{\mu} - \overline{u} \right) \left( \frac{y^2}{2} - hy \right) + \frac{F}{\mu} y
\]

where

\[
\overline{u} = \frac{1}{h} \int_0^h u dy
\]

Substituting into Equation 1.4-7, and with considerable simplification yields,
\[
\frac{\partial \ddot{u}}{\partial t} + \frac{\partial \ddot{u}}{\partial x} \left( \frac{9}{10} \ddot{u} - \frac{1}{5} \frac{Fh}{\mu} \right) = \frac{\sigma}{\rho} \frac{\partial^3 h}{\partial x^3} + \frac{3 \nu h \frac{F}{\mu} - \ddot{u})}{\frac{212}{h}} - \frac{1}{\rho} \frac{\partial P}{\partial x} - g \quad (1.4-11)
\]

and

\[
\frac{\partial h}{\partial t} = \frac{\partial}{\partial x} (h \ddot{u}) \quad (1.4-12)
\]

Considering the film thickness in terms of a deviation from an average value,

\[
h = h_0 (1 + \phi) \quad (1.4-13)
\]

As the film thickness and the average velocity over the film cross section are both functions of the argument, (x-ct), where c is the phase velocity,

\[
\frac{\partial \ddot{u}}{\partial t} = - c \frac{\partial \ddot{u}}{\partial x} \quad (1.4-14)
\]

\[
\frac{\partial h}{\partial t} = - h_0 c \frac{\partial \phi}{\partial x} \quad (1.4-15)
\]

Substituting into Equation 1.4-12 and integrating yields,

\[
\ddot{u} = c - u_0 \frac{c - u_0}{1 + \phi} \quad (1.4-16)
\]

where \( u_0 \) is the average cross sectional film velocity when the film thickness is \( h_0 \). Expanding in a binomial series,
\[ \bar{u} = u_0 + (c - u_0)\phi - (c - u_0)^2 + \ldots \quad (1.4-17) \]

\[ \frac{\partial \bar{u}}{\partial t} = (c - u_0)(1 - 2\phi) \frac{\partial \phi}{\partial x} \quad (1.4-18) \]

Upon substitution into Equation 1.4-11, neglecting the terms involving \( \phi^2 \) and \( \phi^3 \), and simplifying,

\[
\frac{\sigma h_0}{\rho} \frac{\partial^3 \phi}{\partial x^3} + (c - u_0)(c + \frac{F h_0}{\mu} - \frac{9}{10} u_0) \frac{\partial \phi}{\partial x} - \frac{3\nu}{h_0^2} (c - u_0 + \frac{h_0 F}{2\mu}) \phi + \left( \frac{3\nu}{2\mu} \frac{F}{h_0} - \frac{3\nu u_0}{h_0^2} - \frac{1}{\rho} \frac{\partial P}{\partial x} - g \right) = 0 \quad (1.4-19)
\]

For an undamped periodic solution, the constant term and the coefficient on \( \phi \) must be equated to zero.

\[
\frac{3\nu}{2\mu} \frac{F}{h_0} - \frac{3\nu}{2} u_0 - \frac{1}{\rho} \frac{\partial P}{\partial x} - g = 0 \quad (1.4-20)
\]

\[
\frac{3\nu}{h_0} (c - u_0 + \frac{h_0 F}{2\mu}) = 0 \quad (1.4-21)
\]

From Equation 1.4-20,

\[
h_0 = \frac{\frac{3F}{2\rho L}}{\sqrt{\frac{9F^2}{4\rho L^2} - 12u_0\left(\frac{1}{\rho} \frac{\partial P}{\partial x} + g\right)}} \quad (1.4-22)
\]
With the assumptions on the nature of the solution, Equation 1.4-19 simplifies to,

\[ \frac{\sigma h_0}{\rho} \frac{\partial^3 \phi}{\partial x^3} + (c - u_0)(c + \frac{1}{5} \frac{Fh_0}{\mu} - \frac{9}{10} u_0) \frac{\partial \phi}{\partial x} = 0 \]  

(1.4-23)

solving,

\[ \phi = a \sin k(x - ct) \]

where

\[ k^2 = \frac{\rho (c - u_0)}{\sigma h_0}(c + \frac{1}{5} \frac{Fh_0}{\mu} - \frac{9}{10} u_0) \]  

(1.4-24)

substitution of Equation 1.4-21 into Equation 1.4-24 gives,

\[ k = \sqrt{\frac{\rho F}{20\pi \mu} \left( \frac{3Fh_0}{\mu} - u_0 \right)} \]  

(1.4-25)

combining Equations 1.4-10 and 1.4-17,

\[ u = \frac{3}{h} \left( \frac{h F}{2 \mu} - u_0 \right) - (c - u_0) \phi \left( \frac{\mu}{2} - hy \right) + \frac{F \gamma}{\mu} \]  

(1.4-26)

The approximate value of the amplitude, \( a \), can be obtained from an energy balance on the film, setting \( \phi = a \), as a first approximation.

The energy transferred from the air stream is balanced by the gain in potential energy and the viscous energy dissipation, \( E_d \), in the liquid film.
From Equation 1.4-26, assuming $\phi = a$,

$$E_d = \frac{\mu}{g_c} \int_0^h (\frac{\partial u}{\partial y})^2 dy$$  (1.4-27)

expanding,

$$E_d = \frac{\mu}{g_c} \left( \frac{3}{h} \right) \left( \frac{h F}{2 \mu} - u_0 - (c - u_0) a \right)^2 + \frac{F^2 h_0}{g_c \mu}$$

$$- \frac{3F}{g_c} \left( \frac{h_0 F}{2\mu} - u_0 - (c - u_0) a \right)$$  (1.4-28)

$$E_d = a^2 \left( \frac{3(c - u_0)^2 \mu}{g_c h_0} \right) + a \left( \frac{3(c - u_0)}{g_c} \right) \left( F - \frac{2F_0 h}{h_0} \right)$$

$$+ \frac{1}{g_c} \left( \frac{3\mu^2}{h_0} + \frac{F^2 h_0}{\mu} - 3F \xi \right)$$  (1.4-29)

where

$$\xi = \frac{h_0 F}{2\mu} - u_0$$

The energy transferred from the air stream is a resultant of the normal and tangential stresses,

$$\frac{\Delta P}{\Delta x} u_0 h_0 + F \cdot c$$

The potential energy gained by the film is,
Combining into the energy balance and solving,

\[ a = \frac{-\beta \pm \sqrt{\beta^2 - 4\alpha\gamma}}{2\alpha} \]  

where

\[ \alpha = \frac{3(c - u_0)\mu}{g_c h_0} \]
\[ \beta = \frac{6(c - u_0)\mu u_0}{g_c h_0} \]
\[ \gamma = \frac{1}{g_c} \left( \frac{3\mu \xi^2}{h_0^2} + \frac{F^2 h_0}{\mu} - 3F\xi \right) - \frac{\rho Q}{2\pi r_1 g} \]
\[ - \frac{\Delta P}{\Delta x} u_0 h_0 - F \cdot c \]
\[ \xi = \frac{h_0 F}{2\mu} - u_0 \]

The only quantity as yet unobtainable experimentally is the interfacial shear stress, \( F \).

The equation of motion in regards to the air stream in cylindrical coordinates is,
Kim (24) concluded that the constant shear stress at the air-liquid interface on the annular core could be expressed as

$$\rho u \frac{\partial u}{\partial x} = - \frac{\partial P}{\partial x} + \frac{1}{r} \frac{\partial}{\partial r} (r T_r) - \rho g$$

(1.4-30)

Thus, the film thickness, $h(x, t)$, can be predicted for steady state oscillatory motion of a climbing film.

$$h(x, t) = h_0 (1 + a \sin k(x - ct))$$

(1.4-32)

1.5 Electrochemical Measurement of the Local Mass Transfer Coefficient in Climbing Film Flow

Simple electrochemical reactions generally involve two major steps; diffusion of the ions in question from the bulk fluid or stream to the electrode surface, followed by the actual chemical reaction at the surface. With relatively low ion concentration, diffusion of the reaction products away from the surface presents a negligible impediment to ion diffusion towards the surface, and is therefore ignored in this discussion. The ion diffusion towards the electrode surface can be effected by several means including diffusion due to a concentration gradient, natural convection, forced convection, and
ionic migration. The latter possibility is due to the potential field established between the electrodes. Through the use of an indifferent electrolyte, such as sodium hydroxide, in relatively high concentration, such ionic migration can be virtually eliminated (30). Under these conditions, the conductivity of the solution is greatly enhanced, thus minimizing the potential gradient near the electrode surface. Natural convection due to density variation need not be considered here.

In the case of the vertical climbing film, the two remaining possibilities for ion transport may be considered dominant over different portions of the fluid cross section. At the electrode surface, a laminar sublayer is thought to exist, across which a concentration gradient is established. Within this region, diffusion can be considered to proceed, due solely to the concentration gradient. Over the outer turbulent region, the fluid is assumed to be thoroughly mixed. With only a slight degree of electrochemical reaction, it is further assumed that this outer region is held at an essentially constant ion concentration throughout. This diffusion due to a concentration gradient is the limiting factor in the overall ion migration. The thickness of the laminar sublayer is however affected by the turbulent eddies produced in the outer turbulent region. Thus, the degree of turbulence also has a pronounced effect on the mass transfer rate.
In dealing with a relatively fast reaction, the rate of diffusion to the reacting site limits the rate of reaction. Ignoring the migration contribution, a charge balance over the electrode yields the following:

\[
\frac{i}{\text{An} \cdot \mathcal{F}} = \bar{k}_c (c_B - c_W)
\]  

(1.5-1)

Thus the current passing through an electrochemical cell is an indication of the rate of reaction. With a diffusion limiting reaction, the current is further indicative of the rate of diffusion across the laminar sublayer and subsequently of the rate of mass transfer at the electrode surface. The current resulting from the applied potential can be increased until the ion concentration at the surface becomes negligible. Beyond this point, further gain with potential can be brought about only by additional reactions, such as the evolution of hydrogen. At the limiting current, indicated by the flat portion of the polarization curve, a maximum concentration gradient exists. With the ion concentration in the bulk stream known, the mass transfer coefficient at the solid-liquid interface can be determined directly from Equation 1.5-1.

By using a large anode and a relatively small cathode, the reaction rate is determined by the conditions found at the cathode. With tangential flow over the cathode, the concentration gradient begins at the point where the fluid first contacts the electrode, and
continues to develop along the length of the electrode. The limiting current realized, and hence the surface mass transfer coefficient, is actually the average value over the surface. As the cathode becomes smaller, the measured mass transfer coefficient approaches the value of the local mass transfer coefficient.

The electrochemical reaction employed, involved the reduction of the ferricyanide ion to a ferrocyanide ion at the cathode, with subsequent oxidation at the anode.

$$\text{Fe(CN)}_6^{3-} + e^{-} \rightarrow \text{Fe(CN)}_6^{4-} \quad (1.5-2)$$

This reaction proceeds rapidly at room temperature, that is, rapidly enough so as not to exceed the critical flow rate in this study. This latter point has been substantiated by the nature of the polarization curves (42).
2.0 DESCRIPTION OF APPARATUS

2.1 Vertical Annular Column

The outer tube of the vertical annular column was assembled in four foot sections of three inch inside diameter flanged plexiglass tubing, totaling about 20 feet. The inner core of the annulus was composed primarily of one inch outside diameter stainless steel tubing, extending some 22 feet from the base. The column base rested on a 15 foot, three inch inside diameter galvanized pipe, which also served as the entrance for the air stream and included a sharp-edged orifice for air flow rate determination.

The main advantage of the annular construction in this study was in the ease of visual observation afforded by the clear outer plexiglass tube. An unobstructed tangential view of the film cross section was thus permitted for photographic purposes as well as an overall view of the developing wave train, at low air flow rates.

Vertical support for the outer shell was provided at the base, 8 feet and 20 feet along the length of the column, whereas the full weight of the inner core rested on the base. With the outer tube mounted and aligned, the inner core was made concentric and was supported by a series of streamlined stainless steel centering pins spaced along the column. A minimum number of these pins were used so that the resultant liquid entrainment, due to the climbing
Figure 1. Schematic flow diagram.
film impinging on the pins, was minimized. In spite of this precau-
tion, the arrangement left much to be desired with respect to entrain-
ment at the higher liquid flow rates.

The electrochemical probe was mounted as an integral part of
the annular core, 13 feet from the base. This midway mounting
served to reduce end effects, and allowed about 13 feet of core over
which steady state film conditions could be approached. Wire leads
from the probe were routed out the top of the hollow core to the elec-
trical system supplying power to the probe.

At the extremities of the test section were located the auxilliary
equipment including the air source, the calming section and the liquid
injection system at the base, with the air-liquid separator at the top
of the column.

2.2 Air Source

Air injected at the base of the column was provided by a Sutorbilt
8 HB rotary blower, driven by a 30 hp, 1760 rpm General Electric a-c
induction motor. The rated capacity was given as 550 cfm (1 atm,
68°F) at 9 psig output pressure. As the system was in essence a
constant volume device, flow regulation was accomplished by means
of a baffled bypass, regulated by a gate valve. Intake for the blower
was through a cloth filter so as to ensure a particle-free air source.
Air from the blower passed through a copper, water-cooled, finned
Figure 2. Annular column.
heat exchanger, and subsequently through a calming section, prior to the point of liquid injection. The calming section consisted of seven thin-walled copper tubes, 15 inches long and 7/8 inch in diameter, placed around and parallel to the annular core. Transference of blower vibration to the column was reduced through the use of flexible tubes installed between the blower and the heat exchanger, and between the heat exchanger and the base of the column.

2.3 Liquid Supply System

A 20 gallon, stainless steel, steam jacketed kettle was used as the liquid supply tank. Liquid temperatures were controlled by regulating the flow of cooling water through the jacket. Because of the possible side reactions in the electrochemical system due to the presence of dissolved oxygen, the kettle was maintained under a slight nitrogen pressure, thus ensuring nitrogen saturation of the solution. This resulted in a negligible oxygen concentration (at 68° F and 1 atm) as opposed to .0005% when saturated with air. The liquid was pumped from the supply vessel by means of a gear-type rotary pump, driven by a 1/4 hp a-c induction motor, through the rotameter, and subsequently onto the annular core through a porous section. Flow was regulated by a valved recycle stream. In addition, as the assumption of constant volume output does not hold with back-leakage due to worn gears, etc., gross regulation was realized with
Figure 3. Liquid injection system.
a valve installed at the rotameter inlet. Continuous mixing of the solution in the supply vessel was maintained by the recycle stream.

The porous injection system of the annular core consisted of two lengths of porous material. The first was a one inch outside diameter stainless steel porous tube (rolled and seamed) two inches long and 3/16 inches thick. This was followed by six inches of Bendix-Poroloy rolled wire mesh tubing, having a somewhat larger effective pore size which served to offset the larger hydraulic head operating in the lower section. With the considerable area involved, liquid appeared to evolve evenly over the surface of the porous tubing, with no gross discontinuities in the flow leading to initial entrainment.

2.4 Air-Liquid Separator

The corrosive liquid passing up the column was separated from the air for more convenient handling. This was accomplished by discharge of the column effluent through a reducing section, followed by a facsimile of a cyclone separator made from a 55 gallon oil drum. The air escaped to the atmosphere from the top of the drum, while the liquid from the reducing section and from the base of the drum drained to waste. Due to the impurities
resulting from corrosion of the drum separator, variable salt concentrations from residuals in the system, and absorption of oxygen upon passing through the column, the liquid was not considered suitable for recycle.

2.5 Electrochemical System

The electrochemical system used in this study can be broken down into several categories including the electrical circuit, the electrolyte, and the electrode assembly.

Electrical Circuit

A diagram of the electrical circuit is provided in Figure 9. The potential required for the operation of the electrochemical probe was supplied by three 2.2 volt lead-acid storage batteries connected in series. Current and voltage measurements were made with Simpson and Weston meters respectively, both having suitable ranges. System response to electrochemical activity was monitored across coil wound resistors by high impedance devices so as to have negligible effect on the character of the response.
Figure 4. Pressure measurement and purge system.

Figure 5. Swivel adapter for column alignment.
Electrolyte

The liquid used throughout this study was a 0.00625 equimolar solution of potassium ferricyanide and potassium ferrocyanoide in 0.5N sodium hydroxide. The sodium hydroxide was added in order to reduce the tendency of the solution to decompose. In addition, it served as an indifferent electrolyte, thus eliminating mass (ion) transfer due to potential gradients established near the electrodes. The solutions were prepared by weighing the reagent grade chemicals on a double pan balance, followed by dilution to the proper volume with distilled water. Resultant concentrations were estimated to be accurate to within 0.5%. Potassium ferrocyanoide has been found to gradually decompose in the presence of light to form hydrogen cyanide (26). Thus, to avoid possible poisoning of the electrodes with HCN, the solutions were prepared immediately prior to the experimental runs, and kept in darkness until they were used.

Electrode Assembly

In order to determine the local mass transfer coefficients, a 1/16-inch nickel wire cathode was employed along with a one-inch long nickel ring serving as the anode, Figure 8. Both the cathode
and the anode were mounted on a hollow plexiglass core and machined flush to the one inch diameter surface. The wire was mounted about 1/2-inch below the ring and was electrically insulated from the anode by the intervening plexiglass. A 3.5-inch long section of smooth plexiglass preceded the wire electrode (cathode) in order to reduce any film disturbance initiated by the core surface transition from stainless steel tubing to the plexiglass body of the probe. The anode was also insulated from the upper portion of the annular core with a plexiglass spacer. The probe assembly was installed about midway on the column so as to minimize end effects. Each end of the probe was threaded to match suitably threaded plexiglass plugs fitted and glued to the stainless steel core with epoxy cement.

2.6 Instrumentation for the Recording and Analysis of the Fluctuations in the Mass Transfer Rate

In order to record and save the system response for future analysis and reproduction, an Ampex FR1300 tape deck was used. With an FM amplifier for recorder input, a DC signal could be modulated and stored on tape as a frequency. Subsequent demodulation through a suitable output amplifier would then reproduce the DC signal. This combination allowed a possible frequency response varying from zero hertz upwards, according to tape speed.
Figure 6. Partial section of annulus.

Figure 7. Mass transfer at the solid-liquid interface.
In order to use the full linear capacity of the recorder of + 1.5 volts DC, a Tektronix type 122 preamplifier was used with an adjustable maximum bandwidth of .2 - 40k hertz, and an approximate voltage gain of 1000. An alternative gain of 100 was also provided by means of a frequency compensated attenuator. Further adjustment of the signal amplitude was effected by using a variable resistance across which the signal was monitored. A compensating impedance was used elsewhere in the circuit so as to keep the average DC voltage constant. The preamplifier was self-balancing within the indicated response range, thus eliminating the DC component of the signal. As the recorder had a high input impedance (20k resistive in parallel with 150 μF unbalanced to ground), a power amplifier was not required.

In order to clearly reproduce the fluctuating signal, the recorder was driven at high speed (60 IPS with frequency response 0-20k hertz ± 1 dB), with slow speed playback (1 7/8 IPS) through an X-Y plotter. The time scale was simulated on a TR20 analog computer, manufactured by Electronic Associates, Inc. of N.J. Using an amplifier and variable potentiometer from the analog computer, the signal amplitude was further modified such that the amplitudes of the various inputs signals could be matched for visual comparison. For purposes of a spectral density analysis, the signal was recorded at low speed (frequency
Figure 8. Electrochemical Probe
Figure 9. Electrical circuit for recording and display of electrochemical mass transfer fluctuations.
response 0 - 625 hertz ± 1 dB), and played back at high speed in order to spread out the dominant frequency range, thus facilitating analysis.

The wave analyser used was Type 1900-A, in conjunction with a graphic level recorder type 1521-B, both manufactured by the General Radio Company. The analyser was essentially a heterodyne voltmeter with a variable quartz-crystal filter in series for increased and adjustable specificity. The former portion included a high frequency adjustable oscillator used to isolate any particular narrow range desired, by interference and resonance. The frequency range of the system extended from 10 hertz (1 dB down) or 5 hertz (2 dB down) to 54k hertz. The major frequency contributions were first indicated by a preliminary scan with a Tektronix Type RM32 plug-in wave analyser and oscilloscope, and were found to lie primarily between zero and 1k hertz, with a very pronounced response tending toward zero. In order to better utilize the greater frequency response range provided in the Type 1900-A analyser, in addition to partially circumventing poor response characteristics in the lower frequency ranges, the signal was played back at a speed 32 times that at which it was recorded. Thus a real time frequency of .5 hertz would be
considered as a contribution to the analysed signal at 16 hertz, well within the effective range of the instrument. As the signal was found to be quite noisy, the largest available bandwidth was used (50 hertz or 1.56 hertz real time) in order to better determine the dominant frequencies contained within the signal. Although the 1900-A analyser did provide some degree of averaging, it was found to be somewhat lacking for analysis of this particular signal. Thus several passes through the dominant frequency range were required before distinct frequency peaks could be ascertained.

The analyser measured the absolute instantaneous rms voltage (in dB), having been passed through the band pass filter and averaged by an RC smoothing circuit. In order to obtain an estimation of the root-mean-square-voltage density spectrum, the equivalent voltage response from the analyser for a one-cycle bandwidth was required. This was then corrected for the ratio of rms to overall average voltage for Gaussian distribution. Due to several severe limitations in the approach, the spectral plots have been included essentially as they were received from the analyser. As the absolute power spectrum is of secondary importance to the relative relationships between the various plots, this approach seems to be satisfactory.
3.0 EXPERIMENTAL PROGRAM

3.1 Liquid Flow Measurement

Liquid flow rates from 0.2 to 1.47 lb/min were determined with a rotameter, No. FM1048B by the Manostat Corporation of New York. This meter had been calibrated by the manufacturers for water, and was guaranteed accurate to within 2% of full scale. It had since been recalibrated for a 0.025 equimolar potassium ferri-ferrocyanide solution in 2N sodium hydroxide (42). This resulted in a maximum variance of only 1.5%, which is of the order of the expected accuracy.

As the physical properties of the electrolyte were more nearly approximated by water, the former calibration was used in this study. As this investigation was limited to the climbing film regime of two-phase flow, the lower limit indicated above was found to be prerequisite to the formation of a film over the inner core of the annulus. Below this flow rate, the liquid proceeded up the inner core in several rivulets, with the film breakdown downstream due to the core supporting and centering pins. The upper limit of the rotameter was found to be more than adequate. With increased liquid flow, the problem of entrained droplets forming, with subsequent deposit on the outer tube wall, became more serious. This reaction was also chiefly initiated by the centering pins. The relatively
slight amount of liquid rising as a film on the outer wall of the annulus would not be expected to significantly alter the character of the electrochemical reaction proceeding at the surface of the inner core. It did however hamper clear observation and attempts at photography.

3.2 Air Flow Measurement

The air flow rate was determined by use of a two-inch sharp-edged orifice of 1/16-inch brass. Pressure taps installed on either side of the orifice were connected to a differential manometer filled with a liquid having a specific gravity of 2.92. The orifice was calibrated using a Meteric Orifice Flow Prover Type CR size 2, manufactured by the American Meter Company. Calibration was also checked by integration of the velocity profiles with a resultant estimate on the accuracy of calibration of ± 1% (24).

An additional static pressure tap was installed at a point prior to liquid injection. The overall column pressure for each set of flow conditions, was determined by use of a Crosby pressure gauge (range, 0-15 psig).

3.3 Recording of the Local Mass Transfer Rate

Recordings were made of the local mass transfer fluctuations, as represented by a variable emf with the DC component eliminated
from the signal. A high-gain, high impedance preamplifier was placed across a variable wire wound resistor in the probe circuit, the output of which was run to a tape recorder with an oscilloscope wired in parallel to monitor the recorded signal. Such measurements were taken at limiting current so as to give consistent results.

The probe was cleaned electrolytically with a current density of about 20 ma for 15 minutes every two hours of operation, to eliminate gross chemical polarization. There was noted a pronounced decrease in sensitivity with polarization, when the probe was allowed to operate as long as four hours without cleaning. No problems with corrosion or with deposits forming at the cathode were apparent throughout the experimental program.

3.4 High Speed Photography of the Climbing Film

A high speed motion picture study of the climbing film was conducted using a Fastax camera, Model No. WFl4S, manufactured by the Revere-Wollensak Division of the Minnesota Mining and Manufacturing Company. Two features of this camera lent itself particularly to this study. Camera operation was based on a rotating prism rather than the conventional shutter. This allowed for a considerable improvement in image sharpness, without the necessity for a high intensity strobe of short duration, synchronized with the camera, as would be required in normal high speed shutter operation. Clear
distinct single frames were imperative for accurate mean film thickness determinations. The other added feature of the camera was a second lens mounted so as to allow an oscilloscope streak to be recorded on film.

With a two-inch extension tube in conjunction with a 50 mm Fastax-Raptar lens, the minimum distance from the film to the focus plane was reduced from 28 inches to about five inches, with commensurate magnification. All motion photography was carried out using a single 150 watt photo spot light to backlight the subject.

By means of an adjustable internal screen, 1/2 of the film was blocked off from the principle image. This unused portion of the film received the oscilloscope streak trace from the auxiliary lens. With this split image technique, a simultaneous record of the film character and the electrochemical response was obtained for direct comparison. Due to the construction of the camera, it should be noted that the electrochemical response as monitored by the oscilloscope, was so recorded as to lag behind the image of the climbing film by five frames.

A tektronix oscilloscope, Model No. RM43 was used to obtain a sharp, dense oscilloscope trace. This unit had a .01 microsecond rise time and an image density found to be adequate for high speed filming. The plug-in Tektronix preamplifier had a useful frequency range of 0-30k hertz, well beyond that required. A slow internal
sweep speed was used in order that the recorded trace image not deviate from center frame more than 15% during a single run, lasting about four seconds.

As the various air flow rates resulted in widely varied average liquid film velocities, several camera speeds were required in order that a similar sampling could be obtained throughout. To this end, two film types were used, Plus-X type 7276 and Tri-X type 7278, both 16 mm black and white Reversal by Kodak. Although the higher film speed tended to be somewhat grainy when observed in single frames, the effect was not serious.

3.5 Liquid Film Thickness Determination

Several experimental techniques have been used to measure the thickness of a liquid film in wavy motion. A good survey of the methods presently in use has been compiled by Collier and Hewitt (5). The methods regarded in the present study limit the range of possibilities to those applicable to local film thickness determination.

Hewitt and others (13, 14, 15, 17) made use of conductance probes which, with the associated equipment, enabled them to monitor the local average film thickness, as it was presupposed to be directly related to the conductance of the liquid film. As the probes were mounted flush with the tube wall, there is some question as to the character of the potential gradients involved. In purely laminar
flow for example, the film depth beyond the diffusion layer would have little effect on the realized conductance. Thus the degree of turbulence and its effect on the conductance should have been considered for more meaningful results.

The contact needle technique (17), where the liquid film is approached from the free surface side, eliminates these diffusional problems, since the method is purely mechanical as to probe stimulation. The role the needle plays in the development of interfacial air turbulence in the vicinity of the measurement, and hence on the character of the free surface of the film, is difficult to specify. This approach otherwise appears quite good for the accumulation of average, if not instantaneous data on wave characteristics of the film surface.

Light absorption methods (4, 19, 39, 41) have proved relatively successful in dealing with smooth liquid films. Some mechanical difficulty is to be expected in the setup, as the light source and light analyser must be located on opposite sides of the film. Further, the interpretation of the light absorption results becomes complex when the free surface of the film is roughened, as in the case of ripples appearing in the climbing film regime. The lens effect of the wavy film likewise tends to distort the results, as the intensity of light transmitted no longer follows Beer's Law directly, but is also a consequence of the free surface curvature.
Fluorescence techniques (18, 20) have been found to be somewhat more successful, as the free surface of the film is no longer a distorting influence on the results, at least in dealing with annular tube flow.

The most direct approach to recording the film surface characteristics is through the use of photographic methods. Although statistical interpretation is somewhat more tedious, as the quantitative data observed on the film must first be tabulated prior to processing, there are no problems with mechanical interference of the testing equipment, or with the interpretation of instrument data.

Filming of horizontal channel flow (4, 21) allowed for such direct visual cross sectional wave information, with some problems however arising from channel wall interference. Photography, as attempted by Cooper and others (6, 27, 35) of vertical annular tube flow, lead to difficulties similar in nature to those encountered with the light absorption techniques.

A modification of this system was used in the present study so as to observe more directly, the free surface of the upward flowing liquid film. By forcing the liquid to flow up the inner core of an annulus, the film could be photographed tangentially, thus eliminating many of the obstacles met in previous arrangements.

Kim (24) based his film thickness determination on single-frame photography, using the annular column. The method used in
this study, as described previously, was essentially identical with regard to the average as well as instantaneous liquid film thickness. An advantage of the movie film over the single-frame approach lies in the increased accuracy of interpretation permitted, and in sorting out the push wave fronts on either side of the cross section considered.

The average film thicknesses were obtained for each set of flow rates recorded, by tracing single frame projections of random frames from a given run. These tracings were then subjected to an area determination with a planimeter, from which the average film thicknesses could be determined. For statistical significance, a standard deviation was calculated and tabulated with each average film thickness. Two spring steel reference wires (gage 19) were mounted normal to the liquid film and in the same plane as the recorded image, at about .3 centimeters from the annular core surface. The actual distance was determined by difference measurement through an additional hole drilled near the base of the two wires. The distance was also checked by means of a transit fitted with an extension tube. During a dry run, the relationship between the core and the reference wires in the projected image could be established. Thereafter, as all equipment remained clamped in position throughout the experimental runs and analysis, this relationship was taken to remain constant.
3.6 Wave Velocity and Frequency Determination

The speed at which a particular liquid wave traveled up the column could easily be ascertained from the high speed film recordings with respect to the 60 cycle timing mark, registered at one edge of the film. This time reference was necessary due to the fact that in attaining a preset maximum camera speed, the film endured a relatively gradual acceleration throughout each four second run due to the inertia of the mechanism. The timing marks were provided by a 1/2 watt Xenon light mounted within the camera housing, the terminals of which were connected to a 220 volt AC power line.

As the waves were rather unevenly spaced, the dominant average wave frequency had to be determined separately from the average wave velocity. A similar procedure was used here as in the wave velocity determination, with reference to the 60 cycle timing mark. In both cases, the average values were calculated and tabulated with their respective standard deviations.
4.0 EXPERIMENTAL RESULTS

4.1 Empirical Correlations for Average Climbing Film Thickness

Intuitively, the average film thickness can be thought to vary approximately inversely with air velocity. The average liquid film thicknesses, as empirical functions of the inverse average air velocity and the inverse air flow rate, Figures 12 and 13 respectively, were obtained by cross-plotting the information in Figure 10. As this resulted in approximately linear relationships between the film thickness and the inverse quantities, at a given liquid flow rate, the original assumption appears to hold a good approximation to an empirical data fit. Figure 10 suggested a first order dependence of the film thickness on the liquid flow rate. By trial, however, a quadratic with empirically determined constants was found to give the best empirical correlations, over the range of flow rates investigated.

\[ h_0 = \frac{(AR^2 + BR + C)}{V} \]  
\[ (4.4-1) \]

For zero liquid flow rate, the film is non-existent, thus eliminating the constant, C. The constants A and B were then determined to three places so as to give the best over-all data fit. With the correlation involving air flow rate, the expression,

\[ h_0 = \frac{R(5.1 - 2.6R)}{M} \]  
\[ (4.4-2) \]

was found to give the best fit, with an average deviation of 6.8% and a standard deviation of 4.2%. The correlation with average air velocity gave somewhat better results,
Figure 10. Average film thickness with liquid flow rate.
Figure 11. Average film thickness with air flow rate.
Figure 12. Average film thickness with inverse air velocity.
Figure 13. Average film thickness with inverse air flow rate.
with an average deviation of 5.8% and a standard deviation of 3.3%.

Both correlations are presented graphically in Figures 14 and 15.

As the liquid film used in this experiment was held relatively invariant as to liquid properties, no reasonable attempt could be made to include the effects of these various properties in the correlation. Nusselt (13) suggested a square root dependence on the kinetic viscosity coefficient,

\[ h_0 \propto \sqrt{\nu} \]

A comparison with the data of Kim (24) (see Figure 11), who used the same apparatus but a slightly less viscous liquid (water), indicated that the empirical film thickness dependence might more likely be of the form

\[ h_0 \propto \nu^4 \]

though no theoretical basis for this suggestion has been found. A liquid series of varying properties must first be subjected to a similar analysis as to film thickness before a more general empirical correlation can be developed.
Figure 14. Consistency of empirical correlation with air flow rate.
Figure 15. Consistency of empirical correlation with air velocity.
4.2 Fluctuations in the Rate of Mass Transfer

Electrode signal traces of the fluctuating rate of mass transfer occurring at the solid-liquid interface have been presented in Figures 16-20, for the various flow conditions investigated in this study. Although the amplitudes of the signals have been modified for more convenient comparison as to frequency characteristics, it should be noted that as the air flow rate was decreased, the resultant signal amplitudes were found to increase considerably. This tendency could have been predicted in terms of the approach of incipient downflow. Under these conditions, the outer surface of the film rises with the air stream, while the portion closer to the wall is thought to fall down the column, in spite of a net upward liquid flow. This characteristic turbulence can be observed on a large scale during rip tide, where two interacting masses of water, one below the other, move in different directions, creating a noticeable turbulence at the surface, and presumably throughout the depth of both liquid masses.

The high speed photographic study permitted direct comparison of the instantaneous mass transfer rate with the wave character of the climbing film. From this was indicated that the nature of the free surface was indeed related to the rate of mass transfer at the solid-liquid interface. Fluctuations in the electrochemical system
appear to bear a direct correspondence to the wave character of the film, with the maximum rate of mass transfer approached in the wake of each major liquid wave, presumably as a consequence of an increased eddy flow in this region.

It would appear that this approach to monitoring the instantaneous mass transfer coefficient might well be extended to a general method of classifying the various regimes in two-phase flow. A similar undertaking has been attempted by Hubbard and Dukler (9), based on air pressure fluctuations in tube flow. The advantage of such an analysis, utilizing an electrochemical technique, lies in the essential focus of the research into two-phase flow. In terms of heat transfer between the solid and liquid phases, as found in a great variety of heat exchangers, information as to the nature of the flow in the immediate neighborhood of this interface is obviously of greater value.

4.3 Spectral Density Determination

The spectral density determinations on the interfacial mass transfer rate fluctuations, though not absolute, may be interpreted relatively. The various plots of the relative energy content versus frequency (Figures 21 through 25) were drawn in such a way that they all share a common point, that of 50 dB at a frequency of 125 cps. As would be expected, the plots for the lower air flow
rates indicate a relatively low mean wave frequency. Although several rather dominant nodes are apparent in the higher frequencies scanned, no reasonable explanation can be offered at this time, except possibly in terms of harmonics.

Two major frequencies were apparent from the movie film for the lower air flow rates. The relatively large push waves were found visually to have a fairly constant average frequency as indicated in Table 1. Superimposed on these larger waves were seen ripples, similar in relative magnitude to those observed on the large gravity waves of the ocean, driven by a moderate wind. As the air rate increased, the frequency range spread considerably for waves of a given amplitude, thus resulting in a broadening of the energy content with frequency, while lowering the mean value.

As was previously indicated, mechanical vibration of the column was reduced through the use of flexible tubing between the blower and the base of the column. Although dry runs showed no adverse affects from possible mechanically induced variations on the internal resistance of the electrochemical probe, the possibility cannot be eliminated that the higher modes of mechanical vibration were nonetheless transmitted to the inner core, thus affecting the wave characteristics of the climbing film. This supposition is based on the fact that the column was mounted on heavy springs under tension, spaced between the column flanges and metal braces,
Figure 16. Mass transfer fluctuations on annular core surface.
Figure 17. Mass transfer fluctuations on annular core surface.
Figure 18. Mass transfer fluctuations on annular core surface.
Figure 19. Mass transfer fluctuations on annular core surface.
Figure 20. Mass transfer fluctuations on annular core surface.
connected directly to the building structure. The springs of course are quite capable of transmitting the higher mechanical vibrations induced in the building structure by the blower, to the column proper. Should further work be attempted in this area, some improvement in the blower mounting is called for, which would insulate the building structure as well as the column from vibration.

As the electrical currents involved in the electrochemical circuit were of the order of 400 microamperes, electrical noise was a potential problem. This was solved through the use of extensive grounded shielding, and, as indicated by the spectral density plots, the residual noise contributed negligibly to the energy content of the signal at 60 cps.

4.4 Motion Picture Study of the Climbing Film

Two types of waves could be seen in the projected film sequences. At low air flow rates, liquid was carried up the column primarily by large push waves. This form of wave motion was found to be relatively stable, forming approximately symmetrical rings around the annular core and proceeding up the column at a relatively constant speed. The liquid film analysis described in this study was based on this wave type, in view of its continuity and coherence.

The second principle wave type noted was of the ripple
variety, superimposed on the larger push waves. Little wave continuity was discernable, but rather a characteristic randomness existed in wave fronts. This latter observation might lend support to Phillips' theory (37) on wave generation based on random fluctuations in the air stream at the air-liquid interface.

A continuum of wave types emerged as the air flow rate was increased. The mean wave velocity in this grouping was found to have an amplitude of about 30% that of the larger push waves. With a variety of phase velocities present, the larger waves were observed to overtake and engulf the lower amplitude waves, regenerating them in their wake. Thus the intermediate variety did not exhibit a continuous presence along the length of the column.

Entrainment at the higher liquid flow rates was estimated to be less than 2% of the total liquid flow for the relative flow rates investigated. Droplet formation and subsequent deposit on the outer wall of the annulus was principally due to the presence of the core centering pins, rather than as a consequence of the turbulent nature of the gas-liquid interface. In the one recorded sequence showing the formation of a droplet from this source, the liquid film appears rather tenacious in gathering it back, all within a time and space duration of .02 seconds and 0.35 centimeters respectively.
Figure 21. Frequency domain for 172 CFM.
Figure 22. Frequency domain for 300 CFM.
Figure 23. Frequency domain for 411 CFM.
Figure 24. Frequency domain for 518 CFM.
Figure 25. Frequency domain for 609 CFM.
4.5 Numerical Determination of the Average Climbing Film Characteristics

An attempt was made to determine the validity of the equations developed in this paper, pertaining to the average climbing film thickness. The Nusselt equation (Equation 1.1-2), though not directly applicable to the problem, was included to determine its usefulness as a first approximation.

The Nusselt relationship, as derived for a falling film in laminar rectilinear motion down a vertical flat plate, would at best be expected to give results differing from the experimental values by a constant coefficient due to the differences in geometry. A slight modification was made in the original equation so as to yield an explicit equation for the film thickness,

\[ h_0 = \left( \frac{3v \cdot h \cdot u_0}{g} \right)^{1/3} \]  
\[ \text{where} \]
\[ u_0 = \frac{R}{(2\pi r_1 h_0 \rho)} \]  

Substituting,

\[ h_0 = \left( \frac{3vR}{g\rho 2\pi r_1} \right)^{1/3} \]  

In view of the limitations imposed on the derivation, this relatively simple equation gives remarkably good results over the flow rate
ranges covered in the present work.

An adaptation of the Willis model, Equation 1.2-2, was also investigated.

\[ Q = \frac{\pi g h_0^2}{12 \nu} (h_0 (8r_1 + 5h_0) - 6 \frac{\partial P}{\partial x} \frac{r_1}{\rho_L} (r_1 + h_0)) \] (4.5-3)

As this relationship was based on a vertical annular climbing liquid film, it was much better suited to the present study than was the Nusselt relation. Table 3 indicates the correspondence between the predicted and experimental flow rates as a function of average film thickness and pressure loss. Variance in the predicted liquid flow rates ranged from about 60% to 250% of the experimental values. As Equation 4.5-3 involved the average film thickness to the fourth power, extracting the converse relationship was somewhat more complex. The results of this latter root finding attempt is also presented in Table 3, with variance in the predicted liquid film thickness from about 70% to 135% of the experimentally determined values.

A somewhat simplified form of the final algebraic equations derived herein for the average thickness of the climbing film was also considered,

\[ u_0 = \frac{1}{h} \int_0^h u \, dy \] (4.5-4)
Combining Equations 1.4-10 and 1.4-16,

\[ u_0 = \frac{1}{h} \int_0^h \left[ \frac{3}{2} \frac{h^2}{\mu} - \frac{h^2}{2} (u - \Delta \phi) \left( \frac{\gamma}{2} - hy \right) + \frac{Fy}{\mu} \right] \, dy \]  
(4.5-5)

Integrating and simplifying,

\[ u_0 = \frac{3Fh}{2\mu} - \left( \frac{h^2}{2\mu} - u_0 - \Delta \phi \right) \]  
(4.5-6)

By definition of \( u_0, h = h_0 \) and \( \mu = 0 \), thus,

\[ u_0 = \frac{3Fh_0}{2\mu} - (u_0 - \frac{h_0 F}{2\mu}) \]  
(4.5-7)

Solving for the film thickness,

\[ h_0 = \frac{\mu u_0}{F} \]  
(4.5-8)

Substituting for \( u_0 \), the final equation becomes,

\[ h_0 = (\mu R / (2\pi r_i L^2))^{1/2} \]  
(4.5-9)

Where \( F \) is the term describing the interfacial shear flow,

\[ F = \left( \frac{r_i^2 - r_m^2}{2r_i^2} \right) \left( P \frac{\partial P}{\partial x} + \rho \frac{g}{g_c} \right) \]  
(1.4-31)

Convergence of the implicit function for the average film thickness.
was found to be adequate, based on a modified method of successive substitutions of the form,

\[ h_n = f(\frac{h_n \cdot 1 + h_n \cdot 2}{2}) \] (4.5-10)

Good agreement with experimental values is shown for this film thickness model, as indicated in Table 2. The predicted values vary from about 50% to 80% of the experimental film thicknesses. It appears that this model is somewhat insensitive however to liquid flow, and is at its best at low liquid flow rate.

The final equations reached in the theoretical section regarding the average film thickness, though somewhat more complex, were found to require about half the number of iterations as in the previous model for convergence.

\[ h_0 = \frac{\frac{3F}{2\rho \cdot L}}{\sqrt{\frac{9F^2}{4\rho \cdot L^2} - 12u_0 \left(\frac{1}{\rho \cdot g} \frac{\partial P}{\partial x} + g\right)}} \] (1.4-22)

Convergence of this implicit relation was based on the same method of successive substitutions as used above, Equation 4.5-9. In reference to Table 2, this model appears to be the most successful of those tested, with variance in the predicted liquid film thicknesses from about 50% to 90% of the experimental values. It can further be noted that, whereas with the former model, Equation 4.5-9,
accuracy of prediction falls off at the higher air flow rates, the present model is more consistent over the air flow rate range investigated.

With the assumptions made as to the sinusoidal cross sectional form of the free surface of the climbing liquid film, an estimation on the expected frequencies of the first order waves was also attempted. Combining Equations 1.4-13 and 1.4-24,

\[ h = h_0(1 + a \sin k(x - ct)) \]  \hspace{1cm} (4.5-11)

For a fixed axial position, \( x' \), the frequency of the wave fronts follows directly,

\[ \text{Freq} = k' \cdot \frac{c}{2\pi} \]

where \( c \) is the experimental phase velocity and \( k \), as defined previously,

\[ k = \left( \frac{\rho_F}{20\sigma\mu} \left( \frac{3Fh_0}{\mu} - u_0 \right) \right)^{1/2} \]  \hspace{1cm} (1.4-25)

In reference to Table 4, the predicted frequencies were found to be on the order of 0.7 to 12 times those observed experimentally. As the assumed sinusoidal nature of the film free surface was not realized, such deviation could be expected.

From the photographic study, the large push waves proceeded up the annular column with no apparent interaction, as implied by
the sinusoidal model. The spacing of the major wave fronts was found to increase, relative to average amplitude, with air flow rates, thus accounting for the decrease in accuracy as the air flow rate increased. Conversely, at the lower air flow rates, the observed wave spacing, relative to amplitude, diminished. As an approximate sinusoid was thus approached by the free surface, the predicted frequencies tended to a closer correspondence with experimental findings.
BIBLIOGRAPHY


Physical Properties of the Electrolyte

Specific Gravity

Specific gravities of the solutions used in this study were determined with the aid of a chainomatic balance, manufactured by Christian Becker Inc. of New York. The specific gravity of the 0.00625 equimolar solution of potassium ferricyanide and potassium ferrocyanide in 0.5 N sodium hydroxide as a function of temperature, is presented in Figure 26.

Viscosity

Viscosities of the redox solutions used in this study were determined by means of a "Cannon-Fenske" (size 150, No. V138) viscometer. Calibration was based on the Hagen - Poiseuille equation, with a correction for end effects (8),

\[ \nu = At - \frac{B}{t} \]

where

\[ A = \frac{\pi r^4 gh^3}{8Vf} \]

\[ B = \frac{V}{8\pi f} \]

The quantities \( V \) and \( f \) were measured directly, thus permitting an evaluation of \( B \). The viscometer characterization factor, \( A \),
was determined as a function of temperature, using distilled water as the known liquid standard. The kinetic viscosity of the 0.00625 equimolar solution of potassium ferricyanide and potassium ferrocyanide in 0.5 N sodium hydroxide is presented as a function of temperature in Figure 27.
Figure 26. Specific gravity of electrolyte.

Figure 27. Viscosity of electrolyte.
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* at 1 atm. and 68° F.
Table 2. Theoretically predicted average film characteristics

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Table 4. Liquid wave frequency prediction

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* at 1 atm. and 68° F.
PREDICTION OF AVERAGE CLIMBING FILM CHARACTERISTICS

REAL K

31 FORMAT(2F5.2,3F6.1,3F7.5,5F6.1)
32 FORMAT(/,5X,F6.2,F6.0,D8.5,2E10.3,13,/,10X,2E10.3,13,12E10.3,/,15X,F7.1,2E10.3)

G=32.172
RL=64.0
SG=.000417
DO 7 I=1,10
READ(7,31) GL,RM,QA,C,DPDX,FHO,RG,V,FRQ
V=V/100.
DPDX=-DPDX/10000.
U0=144.*GL/(3.1416*RL*FHO)
FRQC=K*C/(120.*3.1416)
DFQ=FRQC*100./FRQ

DO 2 L1=1,30
N1=L1
F=-(RM**2-RI**2)*(DPDX+RG/(12.*144.))/(2.*RI)
B1=3.*144.*F/(2.*RL)
B2=2.25*(144.*F)**2/RL**2-V*U0*(1728.*DPDX/RG+1.)/(5.*G)
B3=144.*DPDX/RG+1./12.
FH1=(B1-SQRT(B2))/133
D=(FH1-X)/FH1
IF (ABS(D).LT.0.0001) 3,1
1 Y=(FH1+X)/2.
RI=.5+Y
UO=U0*X/Y
2 X=Y
3 DH1=FHO*100./FHO
RI=.5+FHO
X=FHO
DO 5 L2=1,30
N2=L2
F=-(RM**2-RI**2)*(DPDX+RG/(12.*144.))/(2.*RI)
FH2=SQRT(2.*V*GL/(3.1416*G*F))
D=(FH2-X)/FH2
IF (ABS(D).LT.0.0001) 6,4
4 Y=(FH2+X)/2.
RI=.5+Y
5 X=Y
6 DH2=FHO*100./FHO
FH3=(3.6*288.*V*GL/(3.1416*G*RL))**(1./3.)
DH3=FHO*100./FHO
7 PRINT 32,GL,QA,FHO,FH1,DH1,N1,FH2,DH2,N2,FH3,DH3,
FRQC,DFQ
END
Figure 28. Sequence of motion picture frames, run no. 1

R = 0.20 lb./min.  M = 172 c.f.m.
Figure 29. Sequence of motion picture frames, run no. 3

$R = 0.47 \text{ lb./min.} \quad M = 172 \text{ c.f.m.}$
Figure 30. Sequence of motion picture frames, run no. 4

R = .79 lb./min.   M = 176 c.f.m.
Figure 31. Sequence of motion picture frames, run no. 5

*R = 0.20 lb./min.  M = 378 c.f.m.*
Figure 32. Sequence of motion picture frames, run no. 6

Direction of flow

Air  Liquid  Annular Core

Frame 1
.0000 sec.

Frame 3
.0042 sec.

Frame 5
.0083 sec.

Frame 7
.0125 sec.

Frame 9
.0166 sec.

Frame II
.0208 sec.

R = .47 lb./min.  M = 378 cfm.
Figure 33. Sequence of motion picture frames, run no. 7
The fundamental dimensions are represented by the following letters: $F = \text{force}$, $L = \text{length}$, $m = \text{mass}$, $t = \text{time}$.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Dimensions</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$</td>
<td>amplitude of the liquid wave</td>
<td>$L$</td>
</tr>
<tr>
<td>$A$</td>
<td>Surface area of working electrode</td>
<td>$L^2$</td>
</tr>
<tr>
<td>$c$</td>
<td>phase velocity</td>
<td>$L/t$</td>
</tr>
<tr>
<td>$c_B$</td>
<td>bulk concentration of ferricyanide ions</td>
<td>moles/$L^3$</td>
</tr>
<tr>
<td>$c_W$</td>
<td>interfacial (solid-liquid) concentration</td>
<td>moles/$L^3$</td>
</tr>
<tr>
<td>$E_d$</td>
<td>rate of energy dissipation per unit area</td>
<td>$F/Lt$</td>
</tr>
<tr>
<td>$F$</td>
<td>momentum transfer at air-liquid interface</td>
<td>$F/L^2$</td>
</tr>
<tr>
<td>$F$</td>
<td>Faraday (96, 500 coulombs)</td>
<td>---</td>
</tr>
<tr>
<td>$g$</td>
<td>acceleration of gravity</td>
<td>$L/t^2$</td>
</tr>
<tr>
<td>$g_c$</td>
<td>force-mass conversion factor</td>
<td>$mL/Ft^2$</td>
</tr>
<tr>
<td>$h$</td>
<td>instantaneous film thickness</td>
<td>$L$</td>
</tr>
<tr>
<td>$h'$</td>
<td>hydrostatic head on viscometer</td>
<td>$L$</td>
</tr>
<tr>
<td>$h_0$</td>
<td>average liquid film thickness</td>
<td>$L$</td>
</tr>
<tr>
<td>$i$</td>
<td>current</td>
<td>amperes</td>
</tr>
<tr>
<td>$k$</td>
<td>wave number</td>
<td>$1/L$</td>
</tr>
<tr>
<td>$K_c$</td>
<td>average mass transfer coefficient</td>
<td>$L/t$</td>
</tr>
<tr>
<td>$L$</td>
<td>length of viscometer capillary tube</td>
<td>$L$</td>
</tr>
<tr>
<td>Symbol</td>
<td>Definition</td>
<td>Dimensions</td>
</tr>
<tr>
<td>--------</td>
<td>---------------------------------------------------------------------------</td>
<td>-------------</td>
</tr>
<tr>
<td>M</td>
<td>standard volumetric air flow rate (1 atm 68 deg F)</td>
<td>L³/t</td>
</tr>
<tr>
<td>n</td>
<td>number of electrons involved in electrode reaction</td>
<td>----</td>
</tr>
<tr>
<td>P</td>
<td>pressure</td>
<td>F/L²</td>
</tr>
<tr>
<td>Q</td>
<td>volumetric liquid flow rate</td>
<td>L³/t</td>
</tr>
<tr>
<td>r</td>
<td>radial distance co-ordinate</td>
<td>L</td>
</tr>
<tr>
<td>r₁</td>
<td>radius of viscometer capillary tube</td>
<td>L</td>
</tr>
<tr>
<td>rₗ</td>
<td>interface (liquid-air) radius</td>
<td>L</td>
</tr>
<tr>
<td>rₘ</td>
<td>radius of maximum air velocity</td>
<td>L</td>
</tr>
<tr>
<td>rₒ</td>
<td>inner core radius</td>
<td>L</td>
</tr>
<tr>
<td>r₂</td>
<td>outer tube radius</td>
<td>L</td>
</tr>
<tr>
<td>R</td>
<td>liquid flow rate</td>
<td>m/t</td>
</tr>
<tr>
<td>u</td>
<td>local liquid velocity in the x-direction</td>
<td>L/t</td>
</tr>
<tr>
<td>ʋ</td>
<td>average film velocity</td>
<td>L/t</td>
</tr>
<tr>
<td>u₀ₘ</td>
<td>liquid stream velocity at average film cross section, h₀</td>
<td>L/t</td>
</tr>
<tr>
<td>v</td>
<td>local velocity in r- (or y-) direction</td>
<td>L/t</td>
</tr>
<tr>
<td>V</td>
<td>average air stream velocity</td>
<td>L/t</td>
</tr>
<tr>
<td>V₀ₘ</td>
<td>volume of liquid passing down the viscometer capillary tube</td>
<td>L³</td>
</tr>
<tr>
<td>x</td>
<td>axial co-ordinate in direction of flow</td>
<td>L</td>
</tr>
<tr>
<td>y</td>
<td>horizontal distance co-ordinate</td>
<td>L</td>
</tr>
<tr>
<td>Symbol</td>
<td>Definition</td>
<td>Dimensions</td>
</tr>
<tr>
<td>--------</td>
<td>----------------------------------------------------------</td>
<td>------------</td>
</tr>
<tr>
<td>( \delta_c )</td>
<td>concentration boundary layer thickness</td>
<td>L</td>
</tr>
<tr>
<td>( \delta_m )</td>
<td>laminar layer thickness</td>
<td>L</td>
</tr>
<tr>
<td>( \Delta )</td>
<td>( (c - u_0) )</td>
<td>L/t</td>
</tr>
<tr>
<td>( \lambda )</td>
<td>wave length</td>
<td>L</td>
</tr>
<tr>
<td>( \mu )</td>
<td>viscosity</td>
<td>m/Lt</td>
</tr>
<tr>
<td>( \nu )</td>
<td>kinematic viscosity</td>
<td>L^2/t</td>
</tr>
<tr>
<td>( \rho_g )</td>
<td>density of gas phase (air)</td>
<td>m/L^3</td>
</tr>
<tr>
<td>( \rho_L )</td>
<td>density of liquid (electrolyte)</td>
<td>m/L^3</td>
</tr>
<tr>
<td>( \sigma )</td>
<td>coefficient of liquid surface tension</td>
<td>F/L</td>
</tr>
<tr>
<td>( \tau )</td>
<td>shear stress</td>
<td>F/L^2</td>
</tr>
<tr>
<td>( \chi )</td>
<td>quality (weight fraction) of the gas phase</td>
<td>---</td>
</tr>
</tbody>
</table>

**Greek Symbols**
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Dimensions</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>phase velocity</td>
<td>ft/min</td>
</tr>
<tr>
<td>DPDX</td>
<td>vertical pressure gradient in annular column</td>
<td>#f/in(^3)</td>
</tr>
<tr>
<td>F</td>
<td>momentum transfer at air-water interface</td>
<td>#f/in(^2)</td>
</tr>
<tr>
<td>FH0</td>
<td>average liquid film thickness</td>
<td>inch</td>
</tr>
<tr>
<td>FH1</td>
<td>liquid film thickness (Eq. 1. 4-22)</td>
<td>inch</td>
</tr>
<tr>
<td>FH2</td>
<td>liquid film thickness (Eq. 4. 5-9)</td>
<td>inch</td>
</tr>
<tr>
<td>FH3</td>
<td>liquid film thickness (Eq. 4. 5-2)</td>
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</tr>
<tr>
<td>FH4</td>
<td>liquid film thickness (Eq. 4. 5-3)</td>
<td>inch</td>
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<tr>
<td>FRQ</td>
<td>frequency of liquid surface waves</td>
<td>cps</td>
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<tr>
<td>FRQC</td>
<td>wave frequency (Eq. 4. 5-12)</td>
<td>cps</td>
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<tr>
<td>G</td>
<td>acceleration of gravity</td>
<td>ft/sec(^2)</td>
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<tr>
<td>GL</td>
<td>liquid flow rate</td>
<td>#m/min</td>
</tr>
<tr>
<td>K</td>
<td>wave number</td>
<td>1/ft</td>
</tr>
<tr>
<td>Q</td>
<td>volumetric liquid flow rate</td>
<td>ft(^3)/min</td>
</tr>
<tr>
<td>QA</td>
<td>air flow rate (1 atm, 68 deg F)</td>
<td>ft(^3)/min</td>
</tr>
<tr>
<td>Q1</td>
<td>liquid flow rate (Eq. 4. 5-3)</td>
<td>ft(^3)/min</td>
</tr>
<tr>
<td>RI</td>
<td>interface (liquid-air) radius</td>
<td>inch</td>
</tr>
<tr>
<td>RG</td>
<td>density of air</td>
<td>#m/ft(^3)</td>
</tr>
<tr>
<td>RL</td>
<td>density of liquid (electrolyte)</td>
<td>#m/ft(^3)</td>
</tr>
<tr>
<td>RM</td>
<td>radius of maximum air velocity</td>
<td>inch</td>
</tr>
<tr>
<td>SG</td>
<td>liquid surface tension</td>
<td>#f/ft</td>
</tr>
<tr>
<td>Symbol</td>
<td>Definition</td>
<td>Dimensions</td>
</tr>
<tr>
<td>--------</td>
<td>-----------------------------------------------------</td>
<td>------------</td>
</tr>
<tr>
<td>U₀</td>
<td>liquid stream velocity at average film cross section</td>
<td>ft/min</td>
</tr>
<tr>
<td>V</td>
<td>kinematic viscosity</td>
<td>ft²/sec</td>
</tr>
</tbody>
</table>