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The Effect of Composition on Heat Transfer Coefficients for Condensing Binary Vapors With a Single Liquid Phase.

James Berwick Todd
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THE EFFECT OF COMPOSITION ON HEAT TRANSFER COEFFICIENTS FOR CONDENSING BINARY VAPORS WITH A SINGLE LIQUID PHASE

A Dissertation

Submitted to the Graduate Faculty of the Louisiana State University and Agricultural and Mechanical College in partial fulfillment of the requirements for the degree of Doctor of Philosophy in The Department of Chemical Engineering

by

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B.S., Louisiana State University, 1948
M.S., Louisiana State University, 1950
June, 1953
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ABSTRACT

Much work has been done on the determination of film coefficients of heat transfer for pure components, but for binary systems little work has been done. Data for the ethanol-water system only, and that over a narrow range of composition, were found in a search of the literature. Considerable disagreement between these experimental results and predicted values from the Nusselt Equation was noted.

A study of the theoretical background of the problem indicates that the Nusselt Equation should apply in the prediction of film coefficients of heat transfer for binary vapors that form a single liquid phase if the operating conditions approximate those for which the equation was developed, and if special care is taken in evaluating the temperature drop across the film of condensate. However, the practical use of the equation is nullified by the lack of necessary physical data for solutions. This fact emphasizes the desirability of being able to predict the value of the film coefficient for the condensation of binary vapors from a knowledge of the values for the single components, which are easily obtainable. A study of fragmentary data from the literature indicates that it may be possible to predict coefficients for ideal solutions from the simple weighted
average of the properties of the single components but not necessarily for non-ideal solutions.

An apparatus previously designed by the author was adapted to the conditions met in measurement of film coefficients of heat transfer of binary solutions. The noteworthy features of this apparatus are the ability to maintain isothermal conditions the entire length of the condensing surface and the technique of mounting the surface thermocouples to give correct temperature readings. The apparatus was designed in such a fashion as to allow a stream of vapor of constant composition to be fed to the condensing surface. Other design features insured the maintenance of steady state conditions.

Data for three binary systems are included here as well as that for the single components: the acetone-methanol and benzene-methanol data collected by the author and the isopropanol-methanol data collected subsequently by Trachtenberg on the same apparatus.

The results indicate that a workable apparatus has been designed which will give consistent results. For binary systems with a single liquid phase that approach ideality, the experimental results indicate that the film coefficient for condensation of the vapor is a linear function of the composition at a constant temperature drop across the film. For binary systems with a single liquid phase which do not approach ideality, no such simple relationship is apparent.

Under the conditions which Nusselt's Equation is applicable, it appears
that the correct temperature drop is the difference between the boiling point of the condensate and the surface temperature.
CHAPTER I

INTRODUCTION

The proper design of heat transfer equipment necessitates the accurate evaluation of the overall coefficient of heat transfer for use in Newton's Equation.

\[ q = U A \Delta t \]  \hspace{1cm} (1)

- \( q \) = rate of heat transfer, BTU/hr.
- \( U \) = overall coefficient of heat transfer, BTU/hr \cdot \text{ft.}^2 \cdot ^\circ\text{F}.
- \( A \) = area of heat transfer, ft.\(^2\)
- \( \Delta t \) = temperature difference, \(^\circ\text{F}\)

The conventional approach to the evaluation of the overall heat transfer coefficient is to make use of the resistance concept—that the total resistance to the flow of heat is the sum of the various resistances in series.

In the case of transfer of heat from a vapor to a cooling medium through a containing wall wherein the vapor is condensed, the total resistance, \( R_t \), is made up of the resistance of the film of condensate, \( R_v \), the resistance of any dirt or scale covering the vapor side of the containing wall, \( R_{dv} \), the resistance of the containing wall, \( R_w \), the resistance of any dirt or scale on the opposite of the containing wall, \( R_{dL} \), and the resistance of a film of the cooling medium, \( R_L \).

\[ R_t = R_v + R_{dv} + R_w + R_{dL} + R_L \]  \hspace{1cm} (2)
Now the total resistance is defined as the reciprocal of the overall coefficient and the individual resistances as the reciprocal of the individual coefficients. In the specific case of the condensation of a vapor on the outside of a pipe, the overall coefficient of heat transfer may be represented by the following equation wherein the terms for diameter represent the area corrections:

\[
\frac{1}{U_o} = \frac{1}{h_v} + \frac{1}{h_{dv}} + \frac{L}{k} \frac{D_o}{D_{avg}} + \frac{1}{h_{dL}} \frac{D_o}{D_{avg}} + \frac{1}{h_{L}} \frac{D_o}{D_{avg}} \]  

(3)

Of these coefficients, perhaps the most uncertainty exists with respect to the evaluation of the individual coefficient of the condensing vapor. The coefficients for the dirt and scale formations depend on the time elapsed since cleaning the surface and the nature of the vapor and the cooling medium and must be determined experimentally or estimated from experience. The resistance of the wall is easily determined since its thickness and thermal conductivity in most cases is readily available. The coefficient for the cooling medium may be determined from semi-empirical relationships which have been substantiated by many investigators.\(^{(12)}\)

A theoretical treatment of the problem of predicting the value of the film coefficient for the condensation of vapor on a colder surface was published by Nusselt\(^{(13)}\) in 1916. This treatment is dependent on several assumptions which will be discussed in a later section. This treatment applies only for a condensate forming a
continuous, homogeneous film on the condensing surface. For the cases of "two phase" and "dropwise" condensation, no completely theoretical relationships have been found.

Operating under the conditions assumed by Nusselt in the development of his equation, many investigators have found good agreement between the predicted values and the experimental values of film coefficients for pure compounds. In many cases, the magnitude of the predicted and the experimental coefficients were somewhat different; but in most cases the relation of physical properties as predicted by Nusselt has been substantiated. Several investigators have studied the condensation of binary vapors forming a two-phase condensate, but no general theory has been proposed. It has been said that the Nusselt Equation should apply in the condensation of binary vapors forming a single liquid phase\(^{(11)}\).

A search of the literature revealed that only one system forming a single liquid phase has been studied previous to this project and that for a limited range of compositions.\(^{(24)}(9)\) This meagre data shows considerable deviation between the experimental values and theoretical expectations\(^{(12)}\). The present work was undertaken to collect additional data on other systems in the hope of aiding in the establishment of some general system of behavior. As a necessary adjunct, a previously designed apparatus was adapted to the conditions encountered in dealing with binary vapors and was
tested to determine its reliability.
CHAPTER II

THEORETICAL CONSIDERATIONS

The Nusselt Equation

The conditions encountered in the study of film coefficients of heat transfer for binary vapors condensing to form a single liquid phase are in many ways similar to those met in a like study of pure component vapor condensation. Since an excellent analysis and solution of the problem for single components has been achieved by Nusselt, it is necessary that his basic work be included here.

Nusselt (13) found it necessary to impose the following conditions in order to solve the problem of calculating a film coefficient of heat transfer for the condensation of a vapor on a colder surface.

1) The temperature of the liquid must be the same as the wall temperature at the wall surface and the temperature at the surface in contact with the vapor the same as the vapor temperature. 2) The velocity of the film at the wall surface must be zero and the film must cling to the metallic wall; and the surface of the film in contact with the vapor must be under the same pressure as the vapor. 3) The heat passing through the film is that of the latent heat of condensation. 4) The velocity of the vapor parallel to the surface of condensation is less than one meter per second. 5) The curvature of the film and
the velocity component perpendicular to the wall can be neglected.

6) The temperature distribution through the film can be represented by a straight line relationship. 7) The temperature drop across the film is constant. 8) The physical constants to be used in the equation may be evaluated at average film temperature.

Let \( u \) be the downward velocity component of the film a distance \( a \) from the wall and \( \mu_F \) the coefficient of viscosity. With a velocity change \( \frac{du}{da} \) at the position \( a \), there results a shearing stress \( S \):

\[
S = \mu_F \frac{du}{da}
\]  

(1)

At a distance \( da \) from this position,

\[
\int S = \mu_F \frac{du}{da} da
\]  

(2)

is the change in shearing stress. This shearing stress change is equal to the downward component of the liquid due to the force of gravity. If the surface is at an angle \( \varphi \) with the horizontal plane, and if \( \rho \) designates the weight per unit volume of condensate, then

\[
\int S + \rho \sin \varphi da = 0
\]  

(3)

or

\[
\frac{d^2u}{da^2} = \frac{\rho}{\mu_F} \sin \varphi
\]  

(4)

Upon integration

\[
u = -\frac{\rho \sin \varphi}{2 \mu_F} a^2 + C_a + C_z
\]  

(5)
If the liquid clings to the wall, then \(a = 0, u = 0,\) and \(C_2 = 0,\) then

\[
U = C_1 a - \frac{\rho \sin \theta}{K M_F} a^2
\]  

(6)

The value \(C_1\) depends on the flow conditions of the vapor, for if the vapor strikes the film's shearing stress will be set up on the film. Since the velocity of the vapor with respect to the film has been assumed so small as to be negligible, this stress was neglected.

If \(a_o\) is the thickness of the film at a spot and correspondingly \(du/da = 0,\) then

\[
C_1 = \frac{\rho \sin \theta}{M_F} a_o
\]  

(7)

and

\[
U = \frac{\rho \sin \theta}{M_F} a_o a - \frac{\rho \sin \theta}{2 M_F} a^2
\]  

(8)

\[
= \frac{\rho \sin \theta}{2 M_F} \left(2 a_o a - a^3\right)
\]  

(9)

Then

\[
U_m = \frac{1}{a_o} \int_0^a u \, da = \frac{1}{a_o} \int_0^{a_o} \frac{\rho \sin \theta}{M_F} a_o \, da_o = \frac{1}{a_o} \int_0^{a_o} \frac{\rho \sin \theta}{2 M_F} a^2 \, da
\]  

(10)

\[
U_m = \frac{\rho \sin \theta}{M_F} \left(\frac{a_o^2}{2} - \frac{a^2}{6}\right) = \frac{\rho \sin \theta}{3 M_F} a_o^2
\]  

(11)

In a tube length of unity and an arc element \(r \, d \phi\)

\[
d\phi = \frac{k r (v - w)}{a_o} \, d \phi = \rho \, d \left(U_m a_o\right)
\]  

(12)
Based on the equations

\[ q = \frac{k}{\alpha} (t_v - t_w) \]  
\[ w = \frac{k}{\lambda} \frac{t_v - t_w}{\alpha} \]  

where \( q = \) rate of heat transfer

\( k = \) thermal conductivity

\( t_v = \) temperature of the vapor side of the film

\( t_w = \) temperature of the surface side of the film

\( w = \) the mass rate of transfer

\( \lambda = \) the latent heat of vaporization

From Equation (11)

\[ d_\phi \frac{d}{d_\phi} \sin(\phi) = \frac{\rho^2 \lambda}{3 \mu_f} \sin(\phi) \]  
(15)

and if

\[ B = \frac{3 \mu_f k_f \lambda (t_v - t_w)}{q r_f^2 \lambda} \]

From (12) and (15)

\[ B d_\phi \sin(\phi) = \frac{\rho^2 \lambda}{3 \mu_f} \sin(\phi) + \frac{\rho^4}{B} \sin(\phi) \cos(\phi) d_\phi \]  
(16)

Now let

\[ z_1 = \psi^4 = \frac{\rho^4}{B} \]  
(17)

Then from (16)

\[ 3/4 \sin(\phi) \frac{d^2}{d_\phi} + z_1 \cos(\phi) - 1 = 0 \]  
(18)

Equation (17) is a linear differential equation whose solution is

\[ z_1 = \frac{1}{\sin^{4/3} \phi} \left( \frac{4}{3} \int \sin^{4/3} \phi d_\phi + C \right) \]  
(19)

If no liquid drops on top of the tube, when \( \phi = 0 \), the film thickness
\( a_0 \) is finite and hence \( z_1 \) is finite. Then the constant \( C \) must be zero, and

\[
z_i = \frac{1}{\sin \frac{\pi}{3} \psi} \left( \frac{4}{3} \int_0^{\frac{\pi}{3}} \sin \frac{\psi}{3} \varphi \, d\varphi \right)
\]

(20)

Graphical integration of the integral \( \frac{4}{3} \int_0^{\frac{\pi}{3}} \sin \frac{\psi}{3} \varphi \, d\varphi \) between limits \( \psi = 0 \) and \( \psi = \pi \) gives the value 3.428.

(Recent evaluation \(^1\) by gamma functions has shown the value to be 3.4495 \( \pm 0.0002 \), but this makes only a slight difference in the resulting equation giving a constant of 0.728 as compared to Nusselt's 0.725.)

From (17)

\[
d_0 = \psi \sqrt{B} = (z, B)^{\frac{1}{4}}
\]

(21)

The local coefficient at angle \( \psi \) is

\[
h = \frac{\delta}{d_0} = \frac{1}{\psi} \sqrt{\frac{\delta^2 \lambda \kappa^2 \varphi}{3 \mu L (t_v - t_w)}} = \frac{\delta}{\psi \sqrt{B}}
\]

(22)

By integrating from \( \psi \) to \( \psi_2 \), the mean coefficient is obtained.

\[
\left[ h_m \right]_{\psi_{\psi^{1/4}}}^{\psi_{\psi^{1/4}}} = \frac{\delta}{\sqrt{B} (\psi_{\psi^{1/4}} - \psi_{\psi^{1/4}})} \int_{\psi_{\psi^{1/4}}}^{\psi_{\psi^{1/4}}} \frac{\psi \, d\psi}{\psi}
\]

(23)

Substituting the value for \( \psi \) from (21) and integrating graphically

\[
h_m = 0.725 \sqrt{\frac{\delta^2 \lambda \kappa^2 \varphi}{3 \mu L (t_v - t_w)}}
\]

(24)

The physical properties are the average properties for the film and should be evaluated at the film temperature, \( t_f \).

\[
t_f = t_v - \frac{3}{4} \Delta t
\]

(25)
as shown by Drew, \( t_v \) is the saturated vapor temperature and \( \Delta t \) is the temperature drop across the film. It is to be noted that this equation takes into consideration only the heat of condensation, ignoring that heat which may be given up by the cooling of the film of condensate or any superheat of the vapor. A correction for this fault may be made, but in most cases the correction is of such small magnitude as to be negligible. Another point to be kept in mind is that the film of condensate must flow downward in streamline motion if the equation is to be strictly applicable. The above equations can be put into dimensionless form including the Reynolds Number \( \frac{\mu \Gamma}{
abla_f} \)

where \( \Gamma = \frac{w}{L} \); \( w \) being one half the mass rate of flow and \( L \) the length of the horizontal tube where the equivalent diameter is equal to four times the film thickness. This equation is

\[
\frac{h_m \left( \frac{\mu f^2}{K_f g} \right)^{\frac{1}{3}}}{1.51 \left( \frac{\mu \Gamma}{M_f} \right)^{\frac{1}{3}}} = 26
\]

Each of the conditions imposed by Nusselt in his derivation must now be studied with reference to a binary system to see whether his equation can be expected to hold true for calculations of film coefficients of heat transfer for binary systems forming a single liquid phase. Condition Number 1 "that the temperature of the liquid must be the same as the wall temperature at the wall surface and the temperature at the surface in contact with the vapor the same as the vapor temperature" requires careful study. This assumption was
made simply to obtain the temperature drop across the film. The derivation does not depend on the identity of the vapor side temperature of the film with the vapor temperature. This assumption merely served to establish the vaporside film temperature easily for pure compounds. Hence the assumption may be modified to "the temperature drop across the film must be determinable." Certainly the temperature on the wall side of the film is very near that of the outer surface of the wall. However, the evaluation of the temperature on the wall side of the film is much more complicated in the case of binary vapors. Colburn and Drew\textsuperscript{(4)} have studied the problem by relating mass and heat transfer. Their work is summarized in the following paragraphs with the nomenclature changed to the generally accepted symbols as appear in more recent literature.\textsuperscript{(20)} The theory infers that between the interface and the main vapor stream there exists a layer of vapor that is stagnant or in streamline flow. (Fig. No. 1). At points within the film or layer of vapor, the molal rate of transfer of the more volatile component toward the interface in the case of equimolal counter diffusion is given per unit area as

\[
N_A = -D_V \frac{dc_A}{dz} = -D_V \frac{dp}{RT} \frac{dz}{dz} \tag{27}
\]

- \(c\) = concentration, (mols/ft.\(^3\))
- \(D_V\) = molecular diffusivity in gas (ft.\(^2\)/hr)
- \(N_A\) = rate of diffusion (mols/hr)
- \(p\) = partial pressure of more volatile component (atm)
- \(R\) = Gas Constant (ft.\(^3\))(atm)/(lb mol)(\(^\circ\)R)
- \(T\) = Absolute temperature, (\(^\circ\)R)
- \(z\) = distance in direction of diffusion, ft
FIGURE 1
TEMPERATURE AND CONCENTRATION
GRADIENTS FOR A CONDENSING
MIXED VAPOR
Equation (27) may be integrated to give

\[ N_A = \frac{D_v (p_{AI} - p_{A2})}{RT_2} \]  

(28)

This equation expresses the resistance to mass transfer due to molecular diffusion alone. However, another factor, eddy diffusivity, is involved which is also proportional to the partial pressure gradient. Since the effect of molecular diffusion and eddy diffusion cannot be separated at the present time, a coefficient lumping them together is commonly used as defined by the following equation.

\[ N_A = k_g (p_1 - p_8) \]  

(29)

where \( k_g \) is called the mass transfer coefficient. In the case where eddy diffusivity is very large as compared to the molecular diffusivity then \( k_g \) is seen to approach the value \( D_v / RT_2 \).

In the general case of condensation of binary vapors the mass transfer to the interface between vapor and liquid is brought about by diffusion through the "so called" stagnant film and bodily transport due to condensation. The above may be expressed mathematically by the approximate equation

\[ N_A = -k_g \frac{dA}{dz} + (N_A + N_B)y_A \]  

(30)

Where the first term on the right represents the rate of diffusion and the second the bodily transport. The fact that thermal diffusion is neglected as well as the effect of component B on the mass diffusion is a limitation of the above equation.
Equation (30) can be integrated between the limits \( z = 0 \) and \( z = 2 \), \( P_a = P_v \) and \( P_a = P_i \) to give

\[
(N_A + N_B) = \frac{N_a}{N_a + N_b} P \ln \left( \frac{N_a - y_A}{N_a + N_b - y_B} \right)
\]

At the condensate vapor interface, \( N_A \) and \( N_B \) moles are at a temperature \( t_i \). Assuming that the enthalpies of the pure components in both the liquid and vapor phases are additive, the latent heat of the mixture is

\[
f_L = N_A (h_g^A - h_l^A) + N_B (h_g^B - h_l^B)
\]

where \( f_L \) is the heat of condensation and \( h_g \) is the enthalpy of the saturated vapor and \( h_l \) is the enthalpy of saturated liquid. The molal enthalpies are evaluated at \( t_i \). Colburn and Drew neglected the heat of mixing for non-ideal solutions to simplify the equation.

If it is further approximated for simplification that the molal latent heats of the components are equal then \( \lambda_m = (N_A + N_B) \lambda_m \) where \( \lambda_m \) is the mean molal latent heat of the components.

The heat flux which arrives at the interface as sensible heat is the result of the cooling of the main stream of vapor and the cooling of the \( N_A + N_B \) moles per hour which are condensing from the temperature of the vapor stream to the temperature at the interface. The sum of these two is expressed in terms of the heat transfer coefficient \( h \). Since the flow of matter across the film will convey heat bodily, it must be considered in arriving at an equation to express the
amount of sensible heat arriving at the interface. Therefore

\[ q_s = h \frac{d}{dz} (N_A C_{pA} + N_B C_{pB}) (t - t_i) \]  \hspace{1cm} (33)

In the above equation it is assumed that no condensation takes place within the film. The first term takes into consideration the heat flux across the vapor layer arising from the local temperature gradient. The second term in the equation takes into consideration the heat carried by mass transfer. Of the quantities expressed in Equation (35) above, only \( t \) varies with \( z \) and integration results in

\[ q_s = \frac{C_o}{1 - e^{-C_o}} h (t_v - t_i) \] \hspace{1cm} (34)

where \( C_o = \frac{1}{h} (N_A C_{pA} + N_B C_{pB})/C_{pA} \) and \( C_{pB} \) are molal heat capacities.

From Equations (31), (32), and (36)

\[ q_r = \frac{C_o}{1 - e^{-C_o}} h (t_v - t_i) + \lambda \frac{K_g \rho}{\rho_{m}} P \ln \left( \frac{N_A}{N_A + N_B} \frac{N_A}{N_A + N_B} - \frac{n_i}{n_v} \right) \] \hspace{1cm} (35)

where \( q_r \) is the total heat effect.

The values \( h \) and \( k_g \) may be evaluated from the following relations

\[ j_h = h \left( \frac{C_p u}{h} \right)^{2/3} = \frac{f}{2} \] \hspace{1cm} (36)

\[ j_d = \frac{k_g \rho_{m}}{G_m} \left( \frac{u}{\rho_{Dv}} \right)^{2/3} = \frac{f}{2} \] \hspace{1cm} (37)
where \( G \) and \( G_m \) are the mass rate of flow and molal rate of flow respectively and \( f \), the dimensionless friction factor. The \( j \) factors are found to approximate \( f/2 \) experimentally for certain simple streamline shapes.

Johnston and Pigford \(^{(11)}\) report the following equation to give excellent results when working with binary mixtures in a wetted wall column.

\[
J_D = 0.033 \text{Re}^{-0.23} \tag{38}
\]

Since the molar rate of condensation is usually known, the value of \( \frac{N_A}{N_A + N_B} \) may be determined by solving Equations (31) and (37). If we let \( X_a = \frac{N_A}{N_A + N_B} \) and express Equation (31) in exponential form we have

\[
\frac{\chi - y_v}{\chi - y_i} = e^{-\frac{(N_A + N_B)}{k_g P}} = e^{-\left(\frac{\left(N_A + N_B\right) \rho B m}{J_D G_m \rho_p \gamma_p \gamma \rho}\right)} = E \tag{39}
\]

or

\[
\chi = \frac{y_v - E y_i}{1 - E} \tag{40}
\]

The value of \( y_v \) and \( y_i \) lies between zero and unity. Since \( (N_A + N_B) \) is positive if condensation is taking place, \( E \) must also have a value between zero and unity.

If the value of \( E \) is one as it is when no condensation takes place, then Equation (39) shows that \( y_i \) would equal \( y_v \) and if there were any condensate, it would be in static equilibrium with the main vapor stream. This condition would be approached if only a very
small amount of the total flow of vapor was allowed to condense. At the other extreme where $E$ is zero or approaches zero due either to the large amount of condensate or to the operating conditions which make the factor $k_g$ very small, Equation (40) shows that $x_1$ would equal $y_v$, or in short, bulk flow would explain the mass transfer.

In this work, the real question is the establishment of the temperature at the interface. In the case where $E$ has a value of one, then the condensate is in static equilibrium with the vapor and the temperature of the interface must approach or equal that of the main vapor stream provided there is no superheat. In the other case, where $E$ is equal to or approaches zero and the composition of the condensate approaches that of the main vapor stream, the temperature at the interface would be that at which the solution of that composition would exist at the given pressure or, in short, its normal boiling point. The limits of the temperature of the interface would then be the boiling point of a solution and the dew point of the vapor of the same composition depending upon the method of operation. Between the limits described above, the temperature of the interface could be established making use of mass transfer data. With the temperature at the interface known, the temperature drop across the film of condensate could be established for use in Nusselt's equation.

The second condition set forth by Nusselt "That the velocity
of the film at the wall surface must be zero, and the film must cling to the wall; and the surface of the film in contact with the vapor must be under the same pressure as the vapor** certainly applies as well to binary liquid solutions as to pure liquids.

The third condition "that the heat passing through the film is that of the latent heat of condensation" applies as well to binary solutions as to pure liquids provided there is no appreciable superheat and that the latent heat of condensation is evaluated from an enthalpy-concentration diagram. This diagram shows the enthalpy of a mixture as a function of temperature and composition. (See Fig. Nos. 27 and 28 in Appendix). Thus, if we know the composition of a liquid and the initial conditions of temperature and phase at a constant pressure and the final conditions of temperature, the change in enthalpy can be read easily from this diagram. The accurate construction of this diagram calls for data which is not too prevalent, but approximations can be made to alleviate this shortage of data. This change in enthalpy may be thought of as a "latent heat of condensation" although it is not actually such. As far as the Nusselt Equation is concerned, the use of latent heats is to simplify the determination of the total heat transferred and the strict usage of the latent heat is not a limitation so long as a correct value for the total heat transferred is used.

If the operating conditions are such that the velocity of the
vapor past the film of condensate is within the bounds established by Nusselt and the rate of condensation is such that streamline motion prevails, then the remainder of Nusselt's stipulations should apply equally well to binary solutions as to pure compounds.

It appears then that Nusselt's Equation should apply in the prediction of heat transfer coefficients for binary vapors that form a single liquid phase since the soundness of the equation for pure compounds has been proven by many investigators.

Other Considerations

It has been shown that the Nusselt Equation should be usable in the prediction of the film coefficients of heat transfer for the condensation of binary vapors that form a single liquid phase. However, the obstacle to its use is the absence of the necessary physical data needed for its solution. There is a shortage of the necessary physical data for solutions, but there is adequate data available for the pure components of most solutions. The obvious hope then is to apply some weighted average to the properties of the pure components.

Using the best data available, Flitcraft and Younger found that the group \( \frac{K^3 \rho^2 \lambda}{h} \) was very nearly independent of temperature over short temperature ranges which tends to improve the hope of determining the film coefficient of heat transfer for condensing binary vapors that form a single liquid
phase since there is usually a wide variation of temperature with composition of a binary system. Latent heat of condensation need not be considered since it is practically a linear function of composition.

The question now arises as to what can be done to determine an average value for the aforementioned group. Obviously, the problem can be separated into two parts, properties for ideal solutions and properties for non-ideal solutions.

There is a limited amount of data on thermal conductivity as a function of composition of binary solutions. It must be pointed out that most of this meagre data is of doubtful accuracy as pointed out by Sakiadis and Coates\(^{19}\). However, it may serve to illustrate the problems at hand.

Before going further, a thorough understanding of thermal conductivity is necessary since it plays such an important part in determining the value for coefficients of heat transfer as shown in Nusselt's Equation. Thermal conductivity may be thought of as one of the transport properties of a substance. These properties include diffusivity and viscosity. Thermal conductivity is a property which is temperature dependent. Numerous attempts have been made to relate thermal conductivity to the more commonly used and easily obtained physical properties. This approach has not been entirely satisfactory up to the present time for the equations which have been
developed though rigorous are not useful for they are so complicated. However, Palmer\(^{17}\) has improved a semi-empirical relation to tie in thermal conductivity to other physical properties.

\[
 k = 0.0947 \rho C_p \sqrt{\frac{\lambda}{T}}
\]  

\( (41) \)

- \(c_p\) = specific heat (g. cal. /gram-\(\degree\)C)
- \(k\) = thermal conductivity(Cal./sec.-cm\(^2\) -\(\degree\)C/cm.)
- \(m\) = molecular weight
- \(T\) = boiling point (\(\degree\)K)
- \(\lambda\) = latent heat of vaporization(Cal./mol)
- \(\rho\) = density (g/cc)

This equation was found to hold for 48 liquids within \(\pm 8.8\%\).

The last part of the equation (\(\lambda /T\)) warrants discussion here since it was introduced by Palmer to show the effect of hydrogen bonding on thermal conductivity. He noted that for liquids that were hydrogen bonded, such as water and methyl alcohol, the thermal conductivity was abnormally high compared to that expected on comparison with substances of like molecular weight. The entropy of vaporization was chosen to show the effect of hydrogen bonding. The validity of this belief was enhanced by application of this term for it improved the accuracy of the equation from \(\pm 25\%\) to the present \(\pm 8.8\%\) for the 48 substances for which experimental data were available.

Palmer went further in the study of the effect of hydrogen bonding on thermal conductivity and by studying a homologous series proposed that for water 80% of the heat conducted was due to the presence of
hydrogen bonds and for methyl alcohol 32%.

Perhaps, the reasoning in the foregoing paragraphs may be applied to the case of binary solutions. It would seem reasonable to expect that on mixing two substances which form an ideal solution, that each would contribute an additive part to the thermal conductivity of the solution. Furthermore it appears that a plot of thermal conductivity versus composition gives more nearly linear relationships if the composition is handled on a weight per cent basis rather than on a mole per cent basis. Data for the three systems studied in this work are not available, but data for similar systems are available over a limited temperature range. Also, it must be strongly emphasized that the values for thermal conductivities herein used are highly questionable but it is hoped that the relative effect of composition is not too greatly in error.

From the foregoing discussion, it might be expected that thermal conductivity for an ideal binary solution may be determined from averaging values for the pure components on a weight basis. In order to check this assumption, data for a system such as benzene-toluene should be used. Unfortunately, that data is not at present existant, but there is some data for the ethanol-methanol system, which is a very nearly ideal system. (For a discussion of the reasons for non-ideality in solutions see Gautreau' "A Thermodynamic Analysis of Correlations for Vapor-Liquid Equilibria in
FIGURE 2

THERMAL CONDUCTIVITY (BTU/HR-FT²-°F/FT)

versus

COMPOSITION

- Ethanol-Water 68°F
- Ethanol-Methanol 68°F
- Acetic Acid-Benzene 68.8°F

WEIGHT PER CENT ETHANOL, ACETIC ACID
Non-Ideal Solutions. A plot of this data at 68°F shows only a very slight deviation from linearity (Fig. No. 2). However, the plot for ethanol-water shows a greater departure from linearity, and with benzene-acetic acid, the departure is extremely great, and furthermore a minimum is reached at approximately 40 weight per cent acetic acid. The plot for acetone-water shows the same general tendency of departure from linearity with the indication that at high weight concentrations of acetone, the effect of the acetone on the thermal conductivity tends to lessen. It seems then that the thermal conductivity of a solution is dependent on the degree of ideality of the solution. Since it has been proposed that thermal conductivity is to some extent dependent on hydrogen bonding and since hydrogen bonding has been found to be a very great factor in the behaviour of solutions as far as ideality is concerned, it is not surprising that the above noted behavior of thermal conductivity in solutions is the case. Qualitatively, this behavior might be explained as follows. The thermal conductivity of associated liquids is found to be abnormally high. Now if some substance is introduced which causes a decrease in the association of the molecules of the substance, then not only should the overall thermal conductivity for the solution be changed due to the presence of the foreign molecules as such but the additional effect of this dissociation should be apparent. Certainly this might explain the case for the system including water discussed above; and
FIGURE 4

VISCOSEITY (\text{cP})

versus

COMPOSITION

- Ethanol - Water 77°F
- Ethanol - Methanol 77°F
- Acetic Acid - Benzene 77°F

WEIGHT PER CENT ETHANOL, ACETIC ACID
FIGURE 5

THE GROUP \( \left( \frac{A^2 C^2}{kL} \right)^{1/4} \)

versus COMPOSITION

ETHANOL-WATER 68°F
ETHANOL-METHANOL 68°F
ACETIC ACID-BENZENE 69.8°F

WEIGHT PER CENT ETHANOL, ACETIC ACID
by analogy it should certainly hold for the systems used in this work substituting methyl alcohol, an associated liquid, though not to such a high degree as water, for water. Similar plots for densities and viscosities of the above solutions (Figs. 3 and 4) show the same general tendency as that noted for the thermal conductivities.

For the three systems for which data are available, a plot was made of \( \left( \frac{\mu^3 \rho^2}{\kappa} \right)^{1/4} \) versus Composition (Fig. No. 5). This plot shows that the group \( \left( \frac{\mu^3 \rho^2}{\kappa} \right)^{1/4} \) is very nearly linear for the systems which do not contain a highly associated liquid, but for the water-ethanol system, a great deviation from linearity is noted. To be sure this computation was carried out at a low temperature but lack of sufficient data was the limitation.

The systems used in this work are isopropanol-methanol, reported by Trachtenberg\(^{(23)}\), acetone-methanol, and benzene-methanol. The choice of these components allows the effect of various classes of substances on an associated substance to be studied as far as heat transfer coefficients are concerned. The class of the substance referred to is that given by Ewell et al.\(^{(6)}\) in his qualitative discussion of ideality of solutions.

Class I - Liquids capable of forming 3-dimensional networks of strong hydrogen bonds e.g. water, glycol.

Class II - Liquids composed of molecules containing both active hydrogen atoms and donor atoms (O, N, F) e.g. alcohols, acids.
Class III - Liquids composed of molecules containing donor atoms but no active hydrogen atoms e.g. ether, ketones, aldehydes.

Class IV - Liquids composed of molecules containing active hydrogen atoms but no donor atoms e.g. CHCl₃, CH₂Cl₂.

Class V - All other liquids having no hydrogen bond forming capabilities.

According to this grouping, methanol is a Class II compound, acetone a Class III compound, benzene a Class V compound, and isopropanol a Class II compound. The significance of this classification is to determine the effect upon mixing. When benzene and methanol are mixed, according to Ewell, the predominant effect is to break hydrogen bonds of the associated liquid and it would seem to decrease the value of the thermal conductivity. When acetone and methanol are mixed, hydrogen bonds of the associated liquid are broken, but additional hydrogen bonds between the unlike molecules are formed. The outcome is not predictable. It depends upon which effect is predominant. When the two alcohols, methanol and isopropanol, are mixed the outcome is again unpredictable.
FIGURE 6
FILM COEFFICIENTS OF HEAT TRANSFER FOR CONDENSATION OF ETHANOL-WATER VAPORS AT CONSTANT TEMPERATURE DIFFERENCE BASED ON THE BOILING POINT (Data of Wallace and Davison) (25)
VERSUS COMPOSITION

\[ \text{MOL} \% \text{ ETHANOL} \]

\( \Delta T = 10^\circ F \)
\( \Delta T = 15^\circ F \)
\( \Delta T = 20^\circ F \)
\( \Delta T = 25^\circ F \)
\( \Delta T = 30^\circ F \)
CHAPTER III

PREVIOUS EXPERIMENTAL WORK

A search of the literature has revealed that very little work has been published on heat transfer coefficients of binary vapors forming a single liquid phase. Evidence of only two investigations was found, that of Wallace and Davison\(^\text{24}\) and Hagenbuch \(^\text{9}\) who have reported a scanty amount of data on the ethanol-water system. The former's data was collected for condensation on a horizontal cylindrical surface, and Hagenbuch's data is for condensation on a vertical cylindrical surface. The data of these two investigators has been consolidated by McAdams\(^\text{12}\) and compared to a semi-theoretical plot based on the weighted average of the film coefficients for the pure components. The agreement is far from good. Actual computation of the theoretical values for the film coefficients cannot be made since the data for the various physical properties as a function of composition and temperature is not available. However, data at one low temperature (25°C) has been found in the literature and the variation of the group \(\left(\frac{k^3 \rho^2}{\mu}\right)^{1/4}\) as a function of composition has been made and discussed (See Theory). The general shape of this curve is just about the same as that obtained when Wallace and Davison's data is plotted at constant
temperature drop across the film. (Fig. No. 6) Unfortunately, the data is for too narrow a concentration range to draw any sound conclusions. Hagenbuch's data is even more scanty than Wallace and Davison's, but the two are in general agreement when converted to a common basis. Wallace and Davison's data indicated to them a general agreement with the conclusions reached by Colburn and Drew in their theoretical study of the problem.

The condensing surface for Wallace and Davidson's work consisted of a brass tube 30 inches long having an outside diameter of 0.34 inches. The tube was placed inside a steel jacket 3 inches in diameter. Thermocouples were imbedded in the surface of the tube and thermometers were placed in the vapor stream. The vapor was introduced at one end of the jacket and withdrawn at the opposite end. Cooling water flowed inside the tube and thermometers measured the inlet and outlet water temperature. The liquid was heated and vaporized in a boiler which was a steel drum provided with a steam coil. Provisions were made to collect the condensate from the tube. A heat balance was made based on the temperature rise and the amount of water flowing through the tube.

In calculating the film coefficient, it was assumed that the composition of the condensate was equal to the composition of the liquid at the vapor-liquid interface. By referring to the boiling point diagram for the ethanol-water system, the temperature at the inter-
face was assumed to be equal to the boiling point of the composition of the condensate. By evaluating the temperature at the interface, it was possible to determine the film coefficient of the mixture.

Initially, the composition of the condensate was kept constant, but later work was performed allowing the composition of the condensate to vary, keeping the cooling water rate constant. The composition of the entering vapor approximated the composition of the condensate with the variation seldom exceeding 2% and in most instances less than 1%.

"Apparent" and "True" film coefficients were calculated. The former were based on the temperature difference between the vapor and the surface while the latter were based on the temperature difference between the estimated interface temperature and the surface temperature.
CHAPTER IV

APPARATUS

In previous work, the author\(^{(22)}\) designed an apparatus for the determination of film coefficients of heat transfer. Since part of the present work was to complete the testing and to adapt the apparatus to the determination of the film coefficients of heat transfer for condensing binary vapors, a short discussion of the equipment will be given here. Details of the apparatus are shown in Figures 7, 8, 9, and 10. Figures 11 and 12 are photographs of the apparatus.

Briefly, the apparatus consisted of a steam heated boiler connected to a vapor jacket surrounding a 3.908" copper bar 36" long which acted as the condensing surface. The vapor jacket was connected to a condenser to take care of any vapor not condensed on the primary condensing surface. Cooling water was introduced into both ends of the copper bar through specially drilled holes and removed at the opposite ends. A system was provided for the collection of condensate from the condensing surface. The details will be discussed in the following section.

As it has been pointed out previously, any apparatus for the determination of film coefficients of heat transfer must provide an accurate means for evaluating the amount of heat transferred, \(q\).
and the temperature drop across the film of condensate, $\Delta t$, for a
given heat transfer area. Of these terms, the most difficult to evaluate accurately is the temperature drop across the film of condensate, for this term depends on the accurate determination of the temperature of the condensing surface. For pure liquids, the temperature drop across the film is simply the temperature of the saturated vapor minus the temperature of the condensing surface, but with binary mixtures it is the difference between the equilibrium temperature of the condensate and the surface temperature. After a careful study of the means available for measuring surface temperature\(^{(22)}\), it was decided to use thermocouples.

In deciding on the exact technique of installation of the surface thermocouples for this particular project, the following considerations were kept in mind.

1. The junction should be as small as possible.

2. The junction should be as near the surface as possible.

3. The condensing surface at the junction should be as smooth as possible.

4. The leads in the immediate vicinity of the junction should pass through an isothermal zone to prevent the conduction of heat to or from the junction.

5. The insulation of the leads must be of a material that resists deterioration due to the action of the material being condensed.
FIGURE 7
THERMOCOUPLE INSTALLATION
In order to obtain a very small bead, 30 Gauge (B & S) constantan and copper wires were selected. The insulation was of woven fiber glass with silicone varnish. This small wire allows the formation of a very small bead and still gives sufficient strength.

Each thermocouple was then calibrated, determining the electromotive force corresponding to a known temperature. The value of the temperature was established by the boiling points of pure ethyl alcohol, distilled water, and pure hexane.

As mentioned previously, the condensing surface is a copper bar 3.908" in diameter and 36" long mounted horizontally. At the lowest point on the circumference, a slot 1/4" wide and 3/8" deep was milled longitudinally along the surface and terminated 2" from either end of the bar. Using a blunt pipe cutting wheel in a lathe, a groove was rolled 0.142" on either side of the slot its full length. This roller pushed up the metal at its edges and left the metal 1/32" higher than its original position. Then the metal between the groove and the slot wall was milled out 1/8" deep to form a shoulder to serve as a base for the cover plate. (See Fig. 7). At one end, a hole 5/16" in diameter was drilled through the remaining 2" of metal between the end of the slot and the end of the bar at such a depth that the bottom of the slot coincided with the edge of the hole nearest the center of the bar. Thus the surface of the bar was unmolested between the end of the slot and the end of the bar. From each point where it was desired
to locate a thermocouple, a groove was rolled in the surface of the bar with the blunt cutting wheel along the circumference to a point 1/2" from the slot. The depth of this groove was 1/16" with a ridge of metal pushed up to either side of the wheel 3/64" high. The width of the bottom of the groove was 0.040". At the point where the junction was to be located, the groove was allowed to become more shallow until the 1/32" bulb barely remained below the surface. A hole 5/64" in diameter was drilled through the remaining 1/2" of metal between the end of the groove and the slot in such a manner that the very bottom of the groove was connected to the very bottom of the slot.

The next operation was to imbed the thermocouples.

The thermocouples were then laced through the slot into the groove. A thoroughly cleaned piece of copper wire was then laid over the thermocouple wire and the metal forced up by the wheel was peened over the thermocouple junction and lead wire. The additional copper wire served as a filler. The surface was somewhat roughened by the peening but the thermocouple wire and the additional copper wire left the peened surface higher than the unmolested surface allowing this roughness to be smoothed with a file and fine emery paper. This operation was repeated with each thermocouple. When the smoothing operation was completed, each thermocouple could be barely detected in the copper surface due to the contrast in appearance of constantan and copper. Thus the thermocouples were located at or
FIGURE 9
COOLING WATER SYSTEM

1/2" SS Pipe Screwed Into Holes 2, 4, 6 of Plate "F".

Fittings of Copper Tubing Screwed Into Holes 1, 3, 5 of Plate "F".

1/2" Copper Tubing Soldered To Inside Of Ring "A" As Shown In Fig. A.

1/2" SS Pipe Welded As Shown, 6" Long.

1/2" SS Pipe Mounted Concentrically With The Ends Sealed By 1/8" Steel Plate Welded On.

Well "B" End View Of Well "B".

Cover Plate 3/4" Spacer (3" x 8")

3/8" Pyrex Plate Glass

Gaskets Plate Welded To Vapor Jacket

End View Of Vapor Jacket Showing Location Of Sight Glasses

End View Showing Position Of Trough And Bar

Vapor Line From Boiler

V-Groove For Separating Condensate

Condensate Outlet

Copper Bar

To Condenser

Cooling Water Stream

End View Showing Position Of Trough And Bar

DEPARTMENT OF CHEMICAL ENGINEERING
LOUISIANA STATE UNIVERSITY
APPARATUS FOR MEASURING FLM COEFFICIENTS
Drawn By: J B TODD 20 JAN 1950
Scale: 1/4" To 1"  Drawing No 5A
near the surface, the leads departed through an isothermal zone, and the surface at the installation was smooth and visibly undisturbed. It should be mentioned here that in the makeup of the thermocouples care was taken to insure that the insulation on the copper and the constantan wires was intact right up to the 1/32" bulb forming the junction. This insulation was fiber glass covered with silicone varnish. Thus the temperature of the bulb is the temperature measured as the surface temperature.

In order to complete the isolation of the thermocouple leads from the vapor, the slot was sealed. This was accomplished by fitting a 1/8" copper strip snugly over the slot on the bearing surface previously prepared. This operation was completed by peening down the metal pushed up by the cutting wheel in preparing the slot.

To test the effectiveness of the seal obtained, the bar was submerged in water while maintaining the slot under air pressure of 4 PSIG. Some bubbles appeared but were readily stopped by further peening. It can be said then that the seal was effective.

The wire emerging from the bar was left free for connection to a 10 point switch and thence to a vacuum bottle to contain an ice bath for the cold junctions. The exposed portion of the wire was protected by encasing it in protective tubing leading to the terminal block of the multi-point switch. The cold junctions were insulated from each other by covering with fiber glass tubing and then were
FIGURE 10
THE VAPOR JACKET

JACKET TO BE MADE FROM A 36" SECTION OF 6" SS PIPE

Vapor Line To Condenser

1" Standard Brass Gate Valves

1 1/2" Vapor Ducts From Boiler Welded To Jacket As Shown

PLATE "F"
1/4" Steel Plate

PLATE "E"
3/4" Steel Plate Welded To Edge Of 6" SS Pipe And Machined Smooth

PLATE "C"
3/4" Thickness Of Jacket

PLATE "B"
1/4" Steel Plate Standard Brass Gate Valves

PLATE "A"
3/8" Holes

3/16" Holes

PLATE "G"
GASKET

INDICATES HOLES TAPPED TO RECEIVE 5/16" BOLTS

INDICATES 11/32" HOLES

1/2" Steel Plate To Be Welded Over Slots In Jacket As Shown

Steel Plates Welded Flush With Slots

Location Of Slots For Sight Glasses
Plates To Be Welded On As Shown Above

Right Head To Be Constructed Identical To Left Head

1/4" To 1" DRAWING NO. 3A

DEPARTMENT OF CHEMICAL ENGINEERING
LOUISIANA STATE UNIVERSITY
APPARATUS FOR MEASURING FILM COEFFICIENTS
Drawn By: J B Todd 8 May 1949
Scale: 1/4" To 1"
wrapped together with rubber tape as a waterproof protection. The cold junctions could then be introduced into the ice bath.

In order to obtain the vapor temperature, a thermocouple installation was made in the boiler. A thermocouple well was made by brazing a 9" sealed length of 3/16" copper tubing to a 1/2" pipe plug which was in turn screwed into the vapor space of the boiler. The tubing was bent downward so that oil might be retained in the well although it was mounted horizontally. The length of the well was such that it is felt that the conduction of heat from the location of the junction to the boiler wall is negligible. The thermocouple was connected to the multi-point switch and its cold junction assembled with those of the surface thermocouples.

A mercury-filled thermometer was installed in the vapor jacket as an aid in maintaining a constant composition when working with binary systems, for a constant vapor temperature indicates a mixture of constant composition, pressure remaining constant.

Two means are available for determining the amount of heat transferred from the vapor to the cooling fluid. One is to base the determination on the change in temperature of the cooling medium at a known rate of flow if specific heat data are available, and the other is to base the determination on the amount of condensate collected from a given heat transfer area if latent heat data are available. In the former method, with low fluid rates and small temperature
changes, very precise measurements of temperature are necessary if any degree of accuracy is to be obtained. For example, for a change of temperature of 10°F, for accuracy of 1%, readings accurate to 0.1°F must be made. This in itself is not an obstacle, but experience with this particular apparatus shows that even with the care taken to maintain a constant rate of flow of cooling fluid, variations of 0.4°F were noted within a period of approximately a minute. However, by measuring the weight of condensate collected from a given area and making use of latent heat data very accurate results can be obtained. The condensate can be weighed within 1 gram in 1000 grams, which is an error of 0.1%. The problem then is to collect the condensate accurately from a given area. Due to the fact that the copper bar must be suspended from either end there is the possibility that heat may be conducted to the surroundings and that condensate from the suspension system may mingle with that from the desired area. To prevent this undesirable effect, it was decided to collect the condensate from the central 27 15/16" of the 36" bar allowing the condensate from the 4" end sections to drip back to the boiler. To achieve this result, special rings were constructed to separate the areas. These rings were made from 1/4" brass stock. The outer surface of the ring was grooved to a depth of 3/16" in a "V" shape. This ring was placed on the bar with the center of the ring over the point 4" from the end of the bar. A trough constructed of thin copper sheets was
placed in the bottom of the vapor jacket so that the ends of the trough were located just below the deepest portion of the "V"-cut of the ring. Thus a split of the condensate was obtained between the end sections and the central section. A visual check through the sight glasses of the vapor jacket shows that the split does occur sharply and that all the condensate from the desired section is collected in the trough. The condensate was seen to follow the surface of the bar to the lowest section of the bar and not to drop off before this point was reached. A piping system allows this condensate to be collected and weighed or to recycle to the boiler. Speculation that the vapor might be of a quality less than 100% was discounted by checking the heat transfer by measuring the condensate against that of the cooling water and finding agreement within approximately 2% with that of the water being the higher. To protect the bar against impingement of the vapor from the boiler and therewith any droplets of liquid, baffles were placed over the vapor inlet ports for the end ducts. The central duct was unimpeded since it was felt that the presence of the condensate trough served the same purpose for that particular duct. It might be noted that the placement of the end ducts was such that the area contacted by the direct stream of vapor from the boiler was that area excluded from collection of condensate. Similarly for the ducts to the condenser, the placement was such that any condensate dripping back would fall on the area excluded from collection of condensate in the case of
the end ducts. However, for the middle duct, a copper baffle 3" wide supported half way between the top of the vapor jacket and the bar on a brass wire frame was constructed. This shield extended downward far enough to insure that any drip fell free of the copper bar. As for condensate on the inside of the vapor jacket, no difficulty was encountered. In warm-up periods, the condensate followed the surface of the jacket to its lowest point before dropping off. After insulation was applied to the jacket, very little condensate was seen to collect on the inside of the vapor jacket wall.

When the condensate was removed for weighing it had to be cooled to room temperature to prevent evaporation losses. This cooling was accomplished by use of a small heat exchanger constructed of 1/2" copper tubing with water as the cooling medium.

The condensate was collected for timed periods in half-gallon jugs. The outlet from the cooler was connected to a loosely fitting 3/8" tee. To each end of this tee, about 2" above the mouths of the jugs, a 6" nipple was screwed. Thus by pushing downward on the nipple, the flow of condensate could be changed instantly to either of the jugs.

The jugs were fitted with loosely fitting copper covers to prevent excessive evaporation losses. Thus an accurate means for collecting the condensate was provided.

The boiler was constructed from a 36" section of 12" standard steel pipe. One end was sealed completely by welding on a 3/4" steel plate. The other end was provided with a flange and a removable
head. Vapor ducts were installed as shown (Fig. 8). The heating element was originally composed of three sections of 1" standard steel pipe fabricated as shown. However, using the liquids with low latent heats of vaporization, the minimum rate of heat transfer to the liquid in the boiler proved too great, and one of the sections had to be removed. Even this did not obtain the desired results, and the system of maintaining a pressure slightly above atmospheric pressure by means of a pressure reducing valve and trap had to be abandoned. Instead, the trap was taken off, and a globe valve put in the line just downstream from the pressure reducing valve. The outlet from the coils was put under a liquid seal provided by the condenser water and by adjusting the valve, the proper amount of steam was bled in to give the desired heat transfer. The use of the pressure reducing valve just ahead of the adjusting valve proved to be a very desirable feature in maintaining a steady rate of heat transfer.

For this particular work, it is most important that steady state conditions be maintained. Certain features were incorporated into this apparatus to attain this desired end. First, of course, a constant flow of vapor to the condensing surface is necessary. As mentioned above, this constant flow of vapor was attained by maintaining a constant heat input to the boiler. Othmer and White\(^{(16)}\) devised a scheme for obtaining isothermal conditions over the length of a cylindrical condensing surface by using the counter flow of a
coolant through the cylinder. Features for obtaining this counterflow are evident in the accompanying drawings (Fig. No. 9) and further discussion may be found in a previous work of the author (22). Not only must there be a counterflow of the coolant, but the flow in each leg of the system must be uniform and equal. This constant flow was obtained by use of a constant head box to overcome the fluctuations in the pressure of the city mains. In order to be able to check this equality of flow, manometers were installed in each leg of the system. The use of the city water supply with a constant head box obviously limits to a rather narrow degree the variation in the quantity of heat which may be removed. This limitation was undesirable when working with organic compounds of widely different latent heats since approximately equal temperature drops across the film of condensate were desired and this could only be controlled by variation of the heat removed by the coolant.

Therefore it was necessary to vary the temperature of the coolant. A means was provided for both heating and cooling the water before it was introduced to the constant head box. The water was heated and maintained at the desired temperature by use of a Sarco "Blender." This "Blender", a commercial device, allowed the temperature to be controlled within ± 1°F and mostly even better than that. For obtaining temperatures lower than those prevailing in the city mains, a refrigeration unit was used. This unit was not
FIGURE 11

VIEWS OF THE APPARATUS

Above: The Complete Apparatus

The Condensate Collecting System
FIGURE 12

A VIEW OF THE APPARATUS BEFORE INSULATING
quite large enough to maintain a constant temperature of the coolant during an entire run, but the drift upward in temperature was only about 4\(^\circ\)F.

To show that the desirable design features were attained the following experimental evidence is offered. First, the reasons for using the standard Temperature-Electromotive Force relationships for copper-constantan thermocouples as published by Leeds and Northrup will be discussed. All the wire from the thermocouples used was cut from the same spool which should make for uniformity among thermocouples. Six of the thermocouples were tested in cooperation with Orr\(^{(14)}\) according to his procedure using normal hexane, ethyl alcohol, and distilled water to establish the primary temperature. The average deviation was -0.3\(^\circ\)F for the six thermocouples. It must be pointed out that the insulating well used in this testing was a thick walled glass tube which could have a bearing on this low temperature finding although it would not in the least affect Orr's uses. The remaining thermocouples were tested against one of these six and the deviation was found to be zero when averaging the values for all the thermocouples. All of this testing was done prior to the installation of the thermocouples on the condensing surface. To be accurate, the thermocouples, of course, should be calibrated in place, but no means were available to carry out this calibration. Since the results of this quasi-calibration
showed results only slightly lower than that expected in temperature determinations (about 0.1°F) and since there was a possibility of a slight change in characteristics of the thermocouples upon installation and since the temperatures obtained by the thermocouples would be averaged to obtain the surface temperature, it was decided to use the standard Temperature-Electromotive Force relationship rather than a calibration curve of doubtful value. In order to check the temperature measured by the various thermocouples one against the other, the following scheme was used. With the "blender", the temperature of the coolant could be made the same as that of the vapor surrounding the condensing surface. With the apparatus in operation, the temperature of the coolant was raised gradually until the last signs of condensation were observed. The temperature of the coolant was then lowered slightly until a very thin film of condensate appeared. This film was so thin that only a very few drops of condensate were seen to fall from the surface. The temperature measured by each couple was then recorded after allowing a reasonable time for equilibrium to be established. The results are here recorded.
<table>
<thead>
<tr>
<th>Thermocouple No.</th>
<th>Electromotive Force</th>
<th>Temperature (°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>2.370</td>
<td>136.0</td>
</tr>
<tr>
<td>1</td>
<td>2.373</td>
<td>136.1</td>
</tr>
<tr>
<td>2</td>
<td>2.374</td>
<td>136.1</td>
</tr>
<tr>
<td>3</td>
<td>2.369</td>
<td>136.0</td>
</tr>
<tr>
<td>4</td>
<td>2.376</td>
<td>136.2</td>
</tr>
<tr>
<td>5</td>
<td>2.371</td>
<td>136.0</td>
</tr>
<tr>
<td>6</td>
<td>2.370</td>
<td>136.0</td>
</tr>
<tr>
<td>7</td>
<td>2.376</td>
<td>136.2</td>
</tr>
<tr>
<td>8</td>
<td>2.377</td>
<td>136.2</td>
</tr>
<tr>
<td>9</td>
<td>2.372</td>
<td>136.1</td>
</tr>
<tr>
<td>10 (Vapor Space)</td>
<td>2.374</td>
<td>136.2</td>
</tr>
</tbody>
</table>

(For Thermocouple locations see Figure No. 7)

The consistency of the readings further verifies the validity of using the standard Temperature-EMF relations.

To show that steady-state conditions were obtained, a typical "run" is cited. This "run" was carried out over a period of 25 minutes.

<table>
<thead>
<tr>
<th>Thermocouple No.</th>
<th>Temperature (°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>At start of operation</td>
</tr>
<tr>
<td>0</td>
<td>154.8</td>
</tr>
<tr>
<td>1</td>
<td>156.8</td>
</tr>
<tr>
<td>2</td>
<td>157.6</td>
</tr>
<tr>
<td>3</td>
<td>158.1</td>
</tr>
<tr>
<td>4</td>
<td>158.1</td>
</tr>
<tr>
<td>5</td>
<td>159.2</td>
</tr>
<tr>
<td>6</td>
<td>159.6</td>
</tr>
<tr>
<td>7</td>
<td>158.2</td>
</tr>
<tr>
<td>8</td>
<td>159.0</td>
</tr>
<tr>
<td>9</td>
<td>159.0</td>
</tr>
<tr>
<td>10 (Boiler Temp.)</td>
<td>176.7</td>
</tr>
<tr>
<td>Average Surface Temperature</td>
<td>158.0</td>
</tr>
</tbody>
</table>

This constancy of surface temperature indicates the uniformity of
operation over the timed period. As described so far the equipment was for use with pure compounds. The use of mixed vapors, however, introduced one other problem.

This problem came about as a result of the fact that as a solution is boiled, the composition is changed (azeotrope excepted, of course). This change in composition brings about a change in condensing temperature which is undesirable in this work. So, the following adaptation of the equipment was made. It was thought that steady-state conditions could be attained if a constant holdup of condensate was made and as much solution fed back to the boiler as was removed from the boiler. This called for a metering device for establishing the feed rate through trial and error. For this purpose a rotameter was placed in the feed line. The use of this rotameter along with the proper technique of operation to be described later brought about satisfactory results.
CHAPTER V

EXPERIMENTAL WORK

The choice of the systems to be used in this work was dictated by two primary considerations. First, the components of the system had to have widely different heat transfer coefficients. Secondly, systems were chosen for which vapor-liquid equilibrium data were available, for obvious reasons. A contributing factor to the choice was the desirability of having refractive indices widely different for the pure components of a system in order to use the refractive index of a solution for establishing its composition. Furthermore, the systems chosen should have no adverse effects on the experimental equipment and should cause a minimum permanent deposit on the condensing surface. Other considerations discussed in the Theory Section along with the above points led to the choice of methanol-benzene and methanol-acetone solutions.

Reagents

Obviously, the purest reagents obtainable should be used in any experimental work. However, due to the large quantity necessary to operate this apparatus successfully, 10 gallons, it was decided to use commercial grade substances. The comparison of the various refractive indices with the literature values for pure compounds as
shown in the following tables indicates that the difference is only slight.

<table>
<thead>
<tr>
<th>Substance</th>
<th>Refractive Index $n_D^{25\text{oC}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Literature</td>
</tr>
<tr>
<td>Methanol</td>
<td>1.3267</td>
</tr>
<tr>
<td>Benzene</td>
<td>1.4967</td>
</tr>
<tr>
<td>Acetone</td>
<td>1.3566</td>
</tr>
<tr>
<td>Isopropanol</td>
<td>1.3758</td>
</tr>
</tbody>
</table>

Operating Procedure

Before any series of determinations was carried out, the apparatus was thoroughly cleaned. This cleaning included the condensing surface. It was necessary to dismantle one end of the apparatus to clean the "bar" or condensing surface. This cleaning was accomplished through the use of crocus cloth and soft rags. It was not necessary to remove the "bar" from the vapor jacket. This cleaning was repeated when the "bar" appeared to be tarnished. Only when using the acetone-methanol system did appreciable deposits occur. This deposit was a fine, whitish powder, and it was removed as soon as it became visible, about every three days. The remainder of the cleaning was accomplished by rinsing with small amounts of the substance to be used and purging with air.

The substance to be used then was poured into the boiler in the amount of approximately 6 gallons. This amount was enough to cover the steam coils during operation, which could be checked by the
sight glass on the boiler. It is important that the coils be covered to prevent any superheating of the vapor.

To begin the determination, the flow of water through each branch of the cooling system of the condensing surface was equalized at the desired rate to obtain any particular rate of condensation. If a temperature of the coolant higher than that of the city mains was desired, the "blender" was adjusted properly. If colder water was desired, the refrigeration system was adjusted to the proper level. Also, the flow of water through the auxiliary condenser and the condensate cooler was regulated.

All valves in the vapor stream from the boiler to the auxiliary condenser were opened and particularly the vent in the auxiliary condenser. The valves in the condensate line were adjusted to allow recycling to the boiler. Steam was then admitted to the heating coils to bring about boiling. During this initial warmup period, considerable vapor was fed to the auxiliary condenser to force out any air in the system. The steam flow then was adjusted manually to give only a slight amount of condensate from the auxiliary condenser. The amount of condensate was checked visually during warmup and was adjusted to about 50 ml. per 20 minute period. This rate was maintained at such a low value to assure almost total condensation in the vapor space and yet to maintain the system free from air. Rigid control of the steam flow was necessary to maintain
a constant flow of vapor to the condensing surface. This control of
the steam was difficult and modifications were necessary to get
satisfactory results.

It was necessary to prepare the ice bath for the thermocouples at least one hour before a "run" was made. After about an
hour's "warm up" period, checks of the surface temperature showed
that steady-state conditions had been reached. Runs of 20-25 minutes
then were made. For the pure components, the total amount of con-
densate for the timed period was collected and then weighed. How-
ever, for the binary mixtures of non-azeotropic composition, a
different technique was used due to the change in composition of the
solution during prolonged boiling. In this case, during both the warm-
up and during the regular run about 1/2 gallon of the solution was
maintained as holdup. By maintaining this hold-up constant and feed-
ing back to the boiler the same amount of liquid as was boiled off per
unit time, the composition of the vapors entering the vapor jacket was
maintained constant. This procedure called for continual collecting,
weighing, and dumping of the condensate. The feed-back rate could
be measured by the rotameter in the line from the feed tank to the
boiler. The rate was arrived at through trial and error until the
condensate collection was just equal to the feed-back. The success of
this technique was proven by the analysis of small amounts of con-
densate collected periodically. The variation shown by analysis was
of the order of 0.5 mol per cent. This technique requires considerable effort on the part of the operator. The condensate then could be weighed over any timed period.

During the timed run, the electromotive force generated by each thermocouple was read at least twice by use of the Leeds and Northrup Type K Potentiometer. Three samples of the condensate, a collective sample of the condensate from the auxiliary condenser, and a sample from the boiler were taken during the run. Analysis was carried out by use of the Abbe Refractometer making use of a previously prepared refractive index-composition curve. Analysis for composition in this manner gives a high degree of accuracy as shown by Gautreaux.

The temperature indicated by each thermocouple was obtained from a plot of electromotive force versus temperature. These temperatures for the ten surface thermocouples were then averaged algebraically to determine the average surface temperature. During the same time, periodic determinations of the vapor temperature were made through use of the thermocouple mounted in the boiler. Thus the basic data were obtained.

In order to determine the amount of heat transferred per unit time, the amount of condensate was weighed. This weight of condensate then was converted to heat units by use of the latent heat of condensation. For the pure components, this heat of condensation
was simply the latent heat of vaporization from the literature. For
the solutions, however, the latent heat of condensation was determined
by use of an enthalpy-concentration diagram. These diagrams were
constructed according to the method outlined by Dodge\(^5\) using data
from the International Critical Tables\(^{10}\) and the Table of Physico-
Chemical Constants\(^{21}\). There is a lack of specific heat data for both
solutions, consequently an average value was calculated using mol
percentage as the basis. In getting this specific heat, the effect of
temperature on the specific heat of the pure liquid was taken into
consideration. The heat of condensation then was read from the dia-
gram using a line of constant composition. This point is further
considered in the Results Section. Thus the necessary bits of informa-
tion were determined for solution of Newton's Equation.
CHAPTER VI

RESULTS

It must be remembered that the object of this work is to study the effect of composition on the heat transfer coefficients for the condensation of binary vapors forming a single liquid phase. The problem was attacked in the following manner. The film coefficients of heat transfer for the pure components were first determined. Then, the film coefficients of heat transfer for various condensate compositions were measured. A series of determinations was carried out under such conditions as to maintain a constant condensate composition and almost total condensation. Various series were then run for each system, methanol-benzene and acetone-methanol. The data of Trachtenberg\(^{(23)}\) for isopropanol and methanol is included in this discussion for it was determined on the same apparatus and under the same conditions at a subsequent date. The data determined for the pure components will be discussed first.

Table No. 1 gives the experimental results for methanol, acetone, and benzene. The film coefficients of heat transfer were determined in the following manner. The temperature drop across the film was determined by subtracting the average surface temperature from the vapor temperature as measured. The heat transferred through a given area \(2.38 \text{ ft}^2\) was measured by collecting the

61
condensate and making use of heat of vaporization data. The known values were then substituted in Newton's Equation to determine \( h \), the heat transfer coefficient. Since the group \( \left( \frac{k^2 \rho^2 \lambda}{M_f} \right)^{1/4} \) has been found to be almost constant as a function of temperature, a plot of \( h \) versus \( \Delta t \), the temperature drop across the film should give a straight line with a slope of -0.25 on log-log paper. This plot was made for each pure component (Fig. No. 13). The lines shown are the result of the calculation to determine the best straight line to represent the experimental data by the method of least squares. In addition, the significance of the data was tested by statistical methods as outlined by Brownlee \(^{(2)} \). The confidence limits show that 95% of the determinations should fall within these bounds or in short we have the consistency of the data. This does not establish absolute accuracy but only the consistency of the determinations with this equipment. The confidence limits show that the consistency of the data is approximately \( \pm 4.5\% \). This value would seem to imply that the equipment gives reproducible results and hence accurate results based upon the findings discussed in the Apparatus Section.

Since Nusselt's equation has been found generally to be in agreement with experimental results determined by many investigators, a comparison of experimental data here determined and theoretical results is in order. Lines representing theoretical film coefficients are included on Fig. No. 13. The very good agreement between the
### TABLE I

**EXPERIMENTAL RESULTS**

**FLM COEFFICIENTS FOR CONDENSING VAPORS**

(Single Components)

#### A. Benzene

<table>
<thead>
<tr>
<th>Run No.</th>
<th>Vapor Temp. (°F)</th>
<th>Avg. Wall Temp. (°F)</th>
<th>Condensate (#/hr)</th>
<th>Temp. Drop Across Film (°F)</th>
<th>Film Coefficient h, (BTU/(hr) ft²) (°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>176.9</td>
<td>146.9</td>
<td>93.2</td>
<td>30.0</td>
<td>221</td>
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<tr>
<td>2</td>
<td>176.9</td>
<td>153.6</td>
<td>73.5</td>
<td>23.3</td>
<td>225</td>
</tr>
<tr>
<td>3</td>
<td>176.9</td>
<td>158.5</td>
<td>60.2</td>
<td>18.4</td>
<td>233</td>
</tr>
<tr>
<td>4</td>
<td>176.9</td>
<td>159.0</td>
<td>60.4</td>
<td>17.9</td>
<td>239</td>
</tr>
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<td>5</td>
<td>176.5</td>
<td>155.0</td>
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<td>229</td>
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<td>6</td>
<td>176.8</td>
<td>151.9</td>
<td>82.2</td>
<td>24.9</td>
<td>234</td>
</tr>
<tr>
<td>7</td>
<td>176.8</td>
<td>155.7</td>
<td>68.4</td>
<td>21.1</td>
<td>232</td>
</tr>
<tr>
<td>8</td>
<td>176.5</td>
<td>159.9</td>
<td>57.5</td>
<td>16.6</td>
<td>248</td>
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<tr>
<td>9</td>
<td>176.9</td>
<td>168.1</td>
<td>35.5</td>
<td>8.8</td>
<td>287</td>
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<tr>
<td>10</td>
<td>176.9</td>
<td>165.1</td>
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<td>11.8</td>
<td>270</td>
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<td>176.9</td>
<td>162.4</td>
<td>51.9</td>
<td>14.5</td>
<td>256</td>
</tr>
<tr>
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<td>175.9</td>
<td>159.7</td>
<td>58.1</td>
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<td>13</td>
<td>175.8</td>
<td>152.9</td>
<td>63.1</td>
<td>17.9</td>
<td>251</td>
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<td>176.3</td>
<td>157.3</td>
<td>64.5</td>
<td>19.0</td>
<td>241</td>
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</tbody>
</table>
### TABLE I (Cont.)

**B. Methanol**

<table>
<thead>
<tr>
<th>Run No.</th>
<th>Vapor Temp. (°F)</th>
<th>Avg. Wall Temp. (°F)</th>
<th>Condensate Temp Drop (°F)</th>
<th>Across Film Temp Drop (°F)</th>
<th>Film Coefficient $h_f$ (BTU/hr)(ft²)(°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>147.8</td>
<td>135.6</td>
<td>29.7</td>
<td>12.2</td>
<td>487</td>
</tr>
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<td>2</td>
<td>147.9</td>
<td>135.1</td>
<td>30.0</td>
<td>12.8</td>
<td>467</td>
</tr>
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<td>3</td>
<td>147.5</td>
<td>136.8</td>
<td>26.3</td>
<td>10.7</td>
<td>491</td>
</tr>
<tr>
<td>4</td>
<td>147.3</td>
<td>136.9</td>
<td>27.1</td>
<td>10.4</td>
<td>516</td>
</tr>
<tr>
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<td>147.4</td>
<td>136.9</td>
<td>27.4</td>
<td>10.5</td>
<td>517</td>
</tr>
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<td>6</td>
<td>147.7</td>
<td>133.1</td>
<td>33.6</td>
<td>14.6</td>
<td>457</td>
</tr>
<tr>
<td>7</td>
<td>147.6</td>
<td>133.0</td>
<td>33.5</td>
<td>14.6</td>
<td>456</td>
</tr>
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<td>8</td>
<td>147.6</td>
<td>130.5</td>
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<td>17.1</td>
<td>421</td>
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<td>9</td>
<td>147.4</td>
<td>130.1</td>
<td>35.6</td>
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<td>411</td>
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<td>147.3</td>
<td>128.9</td>
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<td>421</td>
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<td>147.4</td>
<td>129.3</td>
<td>39.8</td>
<td>18.1</td>
<td>437</td>
</tr>
<tr>
<td>12</td>
<td>147.4</td>
<td>128.6</td>
<td>39.0</td>
<td>18.8</td>
<td>414</td>
</tr>
<tr>
<td>13</td>
<td>147.7</td>
<td>139.4</td>
<td>21.0</td>
<td>8.3</td>
<td>505</td>
</tr>
<tr>
<td>14</td>
<td>147.6</td>
<td>127.3</td>
<td>41.5</td>
<td>20.4</td>
<td>404</td>
</tr>
<tr>
<td>15</td>
<td>147.3</td>
<td>127.2</td>
<td>40.4</td>
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<td>400</td>
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<td>16</td>
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<td>126.7</td>
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<td>21.0</td>
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<td>147.7</td>
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<td>433</td>
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<td>19</td>
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<td>20</td>
<td>148.0</td>
<td>116.6</td>
<td>57.5</td>
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</table>
### C. Acetone

<table>
<thead>
<tr>
<th>Run No.</th>
<th>Vapor Temp. (°F)</th>
<th>Avg. Wall Temp. (°F)</th>
<th>Condensate Temp Drop (°F)</th>
<th>Temp Drop Across Film (°F)</th>
<th>Film Coefficient $h_c$ (BTU/ (hr)(ft$^2$)(°F))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>133.0</td>
<td>116.3</td>
<td>52.1</td>
<td>16.7</td>
<td>293</td>
</tr>
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<td>2</td>
<td>132.4</td>
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<td>318</td>
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FIGURE 13
FILM COEFFICIENTS OF HEAT TRANSFER
OF SINGLE COMPONENTS
versus
TEMPERATURE DROP ACROSS THE FILM

- Experimental Data
- Theoretical Prediction
- 95% Confidence Limits

TEMPERATURE DROP (°F)
### TABLE II
EXPERIMENTAL RESULTS
FILM COEFFICIENTS OF HEAT TRANSFER OF BINARY VAPORS

#### A. 17.2 Mol % Acetone in Methanol

<table>
<thead>
<tr>
<th>Run No.</th>
<th>Avg. Wall Temp. (°F)</th>
<th>Condensate (#/hr)</th>
<th>Δ(T_v)</th>
<th>Δ(T_{BP})</th>
<th>Δ(T_{DP})</th>
<th>Film Coefficients (BTU/hr-ft(^2)-°F)</th>
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<td>266 329 274</td>
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TABLE II (Cont.)

B. 42.3 Mol % Acetone in Methanol

\[ T_v = 137.1 ^\circ F \text{ (AVG)} \quad T_{DP} = 135.7 ^\circ F \]

\[ T_{BP} = 132.6 ^\circ F \]

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<th>(\Delta T_{DP})</th>
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<th>(h_{BP})</th>
<th>(h_{DP})</th>
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### TABLE II (Cont.)

**C. 57.3 Mol % Acetone in Methanol**

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<th>ΔT&lt;sub&gt;BP&lt;/sub&gt;</th>
<th>ΔT&lt;sub&gt;DP&lt;/sub&gt;</th>
<th>h&lt;sub&gt;v&lt;/sub&gt;</th>
<th>h&lt;sub&gt;BP&lt;/sub&gt;</th>
<th>h&lt;sub&gt;DP&lt;/sub&gt;</th>
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### TABLE II (Cont.)

#### D. 80 Mol % Acetone in Methanol (Azeotrope)

\[
T_v = T_{BP} = T_{DP} = 131.3^\circ F \text{ (Avg)} \quad h_v = h_{BP} = h_{DP}
\]

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<th>Run No.</th>
<th>Avg. Wall Temp (°F)</th>
<th>Condensate Flow (# / hr)</th>
<th>Temperature Drop (°F), ( \Delta T_v )</th>
<th>Film Coefficients, ( h ) (BTU/(hr)(ft²)(°F))</th>
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<td>335</td>
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</table>
TABLE II (Cont.)

E. 17.7 Mol % Methanol in Benzene

\[ T_v = 165.8^\circ F \text{ (Avg.)} \]
\[ T_{BP} = 137.1^\circ F \text{ (Avg.)} \]
\[ T_{DP} = 165.2^\circ F \text{ (Avg.)} \]

<table>
<thead>
<tr>
<th>Run No.</th>
<th>Avg. Wall Temp (°F)</th>
<th>Condensate (#/hr)</th>
<th>Temperature Drop (°F)</th>
<th>Film Coefficients (BTU/(hr)(ft²)(°F))</th>
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<td>( \Delta T_v )</td>
<td>( h_v )</td>
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TABLE II (Cont.)

F. 46.5 Mol % Methanol in Benzene

\[ T_v = 143.7^\circ F \text{ (Avg)} \quad T_{BP} = 136.0^\circ F \text{ (Avg)} \quad T_{DP} = 145.9^\circ F \text{ (Avg)} \]

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<th>Run No.</th>
<th>Avg. Wall Temp (°F)</th>
<th>Condensate (# / hr)</th>
<th>Condensate Temperature Drop (°F)</th>
<th>Condensate Temperature Drop (°F)</th>
<th>Condensate Temperature Drop (°F)</th>
<th>Film Coefficients (BTU/ hr) (ft²) (°F)</th>
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TABLE II (Cont.)

G. 61.2 Mol % Methanol in Benzene (Azeotrope)

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<th>Condensate Rate (#/hr)</th>
<th>Temperature Drop (°F)</th>
<th>Film Coefficients, ( h_v ), (BTU/(hr)(ft²)(°F))</th>
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<td>Run No.</td>
<td>Avg. Wall Temp (°F)</td>
<td>Condensate (# / hr)</td>
<td>Temperature Drop (°F)</td>
<td>Film Coefficients (BTU/(hr)(Ft²)(°F))</td>
</tr>
<tr>
<td>---------</td>
<td>---------------------</td>
<td>---------------------</td>
<td>-----------------------</td>
<td>-------------------------------------</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>ΔT&lt;sub&gt;v&lt;/sub&gt;</td>
<td>h&lt;sub&gt;v&lt;/sub&gt;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>ΔT&lt;sub&gt;BP&lt;/sub&gt;</td>
<td>h&lt;sub&gt;BP&lt;/sub&gt;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>ΔT&lt;sub&gt;DP&lt;/sub&gt;</td>
<td>h&lt;sub&gt;DP&lt;/sub&gt;</td>
</tr>
<tr>
<td>1</td>
<td>126.3</td>
<td>24.3</td>
<td>16.0</td>
<td>277</td>
</tr>
<tr>
<td>2</td>
<td>125.8</td>
<td>24.1</td>
<td>16.5</td>
<td>10.6</td>
</tr>
<tr>
<td>3</td>
<td>119.8</td>
<td>34.9</td>
<td>22.5</td>
<td>277</td>
</tr>
<tr>
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<td>121.7</td>
<td>32.4</td>
<td>20.5</td>
<td>10.6</td>
</tr>
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<td>5</td>
<td>121.8</td>
<td>31.9</td>
<td>20.2</td>
<td>20.7</td>
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<td>6</td>
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<td>23.2</td>
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</tr>
<tr>
<td>7</td>
<td>118.4</td>
<td>39.0</td>
<td>24.2</td>
<td>293</td>
</tr>
<tr>
<td>8</td>
<td>114.3</td>
<td>46.1</td>
<td>28.3</td>
<td>288</td>
</tr>
<tr>
<td>9</td>
<td>114.5</td>
<td>44.6</td>
<td>28.3</td>
<td>288</td>
</tr>
<tr>
<td>10</td>
<td>113.4</td>
<td>43.4</td>
<td>28.9</td>
<td>288</td>
</tr>
<tr>
<td>11</td>
<td>129.9</td>
<td>18.5</td>
<td>12.9</td>
<td>12.5</td>
</tr>
<tr>
<td>12</td>
<td>129.1</td>
<td>18.6</td>
<td>13.7</td>
<td>7.8</td>
</tr>
</tbody>
</table>

H. 83 Mol % Methanol in Bensene

T<sub>v</sub> = 142.4° F (Avg)  T<sub>BP</sub> = 136.9° F  T<sub>DP</sub> = 142.4° F
Theoretical and experimental values for methanol is very evident. However, the values for acetone and benzene are greatly in variance, although the slopes of the lines are in good agreement with theory. These slopes from experimental data are -0.27, -0.23, and -0.22 for methanol, acetone, and benzene respectively. The fact that the methanol data is so good discounts the effect of a faulty apparatus. The only reason that is evident for the discrepancy noted in the benzene and acetone data is the impurity of the reagents as pointed out previously. This purity, along with questionable data for the physical properties, could cause this disagreement. It is believed, on the strength of the above discussion, that the data presented here represents accurate determinations for the substances used in this work.

Since a calculation of the operating conditions necessary to give a Reynolds Number of 2100 showed that all experimental determinations were well into the streamline flow region, turbulence in the film could not appreciably affect these results.

Surface temperatures measured in this work show agreement with previous work as far as variation of the temperature around the circumference of the cylinder is concerned. The highest temperature occurred at the top of the cylinder with the lowest temperature indicated by the thermocouple on the lowest portion of the cylinder. This variation amounted to approximately 2.5°F on the average determination.
It is thought that the placement of the thermocouples on the condensing surface allowed algebraic averaging of the surface temperatures to determine a usable surface temperature.

At all times, with the pure components, the film of condensate appeared to be uniform and to flow evenly over the surface. Very little tarnishing of the surface was noted. The drops fell from the condensing surface at uniform intervals showing that the bar was in a perfectly horizontal position.

Extreme care had to be exercised in preventing an excessive amount of vapor to flow to the auxiliary condenser. The effects of this vapor flow on the coefficients was evident from several runs made with extremely adverse conditions prevailing. The coefficients so determined were abnormally high. This velocity effect was probably accentuated by the design of the jacket surrounding the condensing surface, an annular space of slightly less than an inch. This feature was incorporated into the design for possible future use in the study of velocity effects on heat transfer coefficients.

Experimental data for the benzene-methanol and acetone-methanol systems are presented in Table No. II. Operating conditions were controlled in such a manner as to give almost total condensation of vapors and a condensate of constant composition as previously described. The condensate was weighed for a timed period and the heat transferred was calculated using an enthalpy-concentration
The value of the temperature drop across the film however could be reckoned in several possible ways.

The theoretical discussion has shown the limiting values of the composition at the interface depending on the operating conditions. A calculation was made to determine the theoretical relationship between the composition of the vapor and the condensate. Using Equations (37 & 38), \( k_g \) was calculated for a typical run.

In order to make this calculation, some necessary approximations were made. Since the total flow past the surface was almost zero, values for \( G_{in} \) and the Reynolds Number were obtained by dividing the total rate into the vapor jacket by 4 since the flow was split equally on each side of the bar and the rate at the top of the bar was practically zero. From Equation (38), \( j_D \) was calculated to be approximately 0.008. A value for \( \frac{\mu}{\rho_D v} \) \( z \) .86 was taken at an average of those reported by Johnston and Pigford for binary mixtures. The value of \( p_{BM} \) was assumed as one atmosphere. The value for \( k_g \) was then found to be 0.009 # mols/(hr)(ft\(^2\))(atmos). The value of \( E \) was then calculated from Equation 39 to be \( 1/e^{35} \) which is practically zero.

This finding substantiates theoretically the fact that the composition of the condensate should be practically that of the main vapor stream as shown by Equation (40).

\[
x_i = \frac{y_v - Ey_i}{1 - E}
\]
The experimental findings were in close agreement with this conclusion, for analysis of the sample from the auxiliary condenser showed it to be only slightly richer in the more volatile component than the condensate. This enrichment might well have been increased by partial condensation in the pipe leading to the auxiliary condenser.

It seems reasonable to expect that the temperature of the liquid at the interface should be the normal boiling point of the solution at the given pressure for these operating conditions. However, the experimental results are presented with the heat transfer coefficient based upon a temperature drop across the film of condensate using the boiling point corresponding to the condensate composition, the dew point, and the temperature of the entering vapor. The results are listed in Table No. III.

The boiler temperature was determined directly from the thermocouple located in the boiler. The dew points and boiling points were obtained from vapor-liquid equilibrium diagrams (Fig Nos. 29, 30, and 31). There was some difficulty in choosing the proper set of vapor-liquid equilibrium data for the acetone-methanol system from the four sets available. The thermodynamic consistency of each set of data was tested and that of Othmer\textsuperscript{(15)} was chosen based on the consistency test and the fact that this was the only data that gave the temperature of the saturated vapor equal to or lower than the temperature
measured in the vapor space in this work. This data, though not entirely consistent thermodynamically, was smoothed according to the Gibbs-Duhem Equation. This smoothing caused only slight temperature changes. The data of Chu\(^{(3)}\) and Gautreaux\(^{(8)}\) for the benzene-methanol were in fairly good agreement. The data determined by Gautreaux were extremely accurate and were taken on very pure substances. It has already been pointed out that the purity of the substances used in the present experimental work is not too great. At the azeotropic composition, the temperatures measured in the boiler were in good agreement with Chu’s azeotropic temperature but somewhat lower (1\(^\circ\)F) than that of Gautreaux. Also between the azeotropic composition for the system and that of pure methanol, the temperature of the saturated vapor was higher for Gautreaux’ data than that measured in the boiler, a case which seems to be unreasonable. Chu’s data, however, gives the expected agreement. This is not a criticism of Gautreaux’ data but it is believed the difference noted is due to the purity of the substances used, that of Chu’s being closer to that used here. For this reason Chu’s vapor-liquid equilibrium data was used. From the discrepancies noted here it would seem that the vapor-liquid equilibrium data should be obtained using the very same quality substances as used in the heat transfer determinations, or extremely pure substances should be used in the heat transfer determinations.

A plot of the heat transfer coefficient versus the temperature
### TABLE III

FILM COEFFICIENTS AT A CONSTANT TEMPERATURE DROP

A. Film Coefficients of Single Components as a Function of Temperature Drop Across the Film

\( (T_v - T_w) \)

\( T_v = \) Vapor Temperature, °F  \( T_w = \) Surface Temperature, °F

<table>
<thead>
<tr>
<th>Temperature Drop °F</th>
<th>Film Coefficient (BTU/hr-ft²-°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Benzene</td>
</tr>
<tr>
<td>10</td>
<td>278</td>
</tr>
<tr>
<td>15</td>
<td>252</td>
</tr>
<tr>
<td>20</td>
<td>238</td>
</tr>
<tr>
<td>25</td>
<td>227</td>
</tr>
<tr>
<td>30</td>
<td>218</td>
</tr>
</tbody>
</table>
TABLE III (Cont.)

B. Film Coefficients of the Acetone-Methanol System at a Constant Temperature Drop \((T_{BP} - T_w)\) Across the Film as a Function of Composition.

\[T_{BP} = \text{Boiling Point of Condensate (°F)}; \quad T_w = \text{Surface Temperature (°F)}\]

<table>
<thead>
<tr>
<th>Temperature Drop °F</th>
<th>Film Coefficient, (h_{BP}) (BTU/hr-ft(^2)-°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>17.2 Mol % Acetone</td>
</tr>
<tr>
<td>10</td>
<td>435</td>
</tr>
<tr>
<td>15</td>
<td>385</td>
</tr>
<tr>
<td>20</td>
<td>357</td>
</tr>
<tr>
<td>25</td>
<td>335</td>
</tr>
<tr>
<td>30</td>
<td>320</td>
</tr>
</tbody>
</table>
TABLE III (Cont.)

C. Film Coefficients of the Acetone-Methanol System at a Constant Temperature Drop ($T_{DP} - T_w$) as a Function of Composition

$T_{DP}$ = Dew Point of Condensate ($^\circ$F); $T_w$ = Surface Temperature ($^\circ$F)

<table>
<thead>
<tr>
<th>Temperature Drop ($^\circ$F)</th>
<th>Film Coefficient, $h_{DP}$, (BTU/hr-ft$^2$-$^\circ$F)</th>
</tr>
</thead>
<tbody>
<tr>
<td>17.2 Mol % Acetone</td>
<td>42.3 Mol %</td>
</tr>
<tr>
<td>10</td>
<td>310</td>
</tr>
<tr>
<td>15</td>
<td>310</td>
</tr>
<tr>
<td>20</td>
<td>305</td>
</tr>
<tr>
<td>25</td>
<td>292</td>
</tr>
<tr>
<td>30</td>
<td>270</td>
</tr>
</tbody>
</table>
TABLE III (Cont.)

D. Film Coefficients of the Acetone-Methanol System at a Constant Temperature Drop ($T_v - T_w$) as a Function of Composition

$T_v$ = Vapor Temperature °F  
$T_w$ = Surface Temperature °F

<table>
<thead>
<tr>
<th>Temperature Drop (°F)</th>
<th>Film Coefficient, $h_v$, (BTU/hr-ft²-°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>17.2 Mol % Acetone</td>
</tr>
<tr>
<td>10</td>
<td>250</td>
</tr>
<tr>
<td>15</td>
<td>290</td>
</tr>
<tr>
<td>20</td>
<td>300</td>
</tr>
<tr>
<td>25</td>
<td>275</td>
</tr>
<tr>
<td>30</td>
<td>260</td>
</tr>
</tbody>
</table>
E. Film Coefficients of the Methanol-Benzene System at a Constant Temperature Drop Across the Film ($T_{BP} - T_w$) As a Function of Composition

$T_{BP}$ = Boiling Point of Condensate ($^\circ$F); $T_w$ = Surface Temperature ($^\circ$F)

<table>
<thead>
<tr>
<th>Temperature Drop ($^\circ$F)</th>
<th>Film Coefficients, $h_{BP}$ (BTU/hr-ft$^2$-°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>17.7 Mol % Methanol</td>
</tr>
<tr>
<td>10</td>
<td>345</td>
</tr>
<tr>
<td>15</td>
<td>304</td>
</tr>
<tr>
<td>20</td>
<td>280</td>
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<tr>
<td>25</td>
<td>263</td>
</tr>
<tr>
<td>30</td>
<td>250</td>
</tr>
</tbody>
</table>
TABLE III (Cont.)

F. Film Coefficients of the Methanol-Benzene System at a Constant Temperature Drop ($T_{DP} - T_w$) as a Function of Composition

$T_{DP} = \text{Dew Point of Condensate (°F)}$  $T_w = \text{Surface Temperature, °F}$

<table>
<thead>
<tr>
<th>Temperature Drop</th>
<th>Film Coefficients, $h_{DP}$, (BTU/hr-ft$^2$-°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>17.7 mol % Methanol</td>
</tr>
<tr>
<td>10</td>
<td>--</td>
</tr>
<tr>
<td>15</td>
<td>--</td>
</tr>
<tr>
<td>20</td>
<td>--</td>
</tr>
<tr>
<td>25</td>
<td>--</td>
</tr>
<tr>
<td>30</td>
<td>--</td>
</tr>
</tbody>
</table>
TABLE III (Cont.)

G. Film Coefficients of the Methanol-Benzene System at a Constant Temperature Drop ($T_V - T_W$) as a Function of Composition

$T_V = $ Vapor Temperature (°F) \hspace{1cm} T_w = $ Surface Temperature (°F)

<table>
<thead>
<tr>
<th>Temperature Drop (°F)</th>
<th>Film Coefficient, $h_v$, (BTU/hr-ft$^2$-°F)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>17.7 Mol % Methanol</td>
</tr>
<tr>
<td>10</td>
<td>--</td>
</tr>
<tr>
<td>15</td>
<td>--</td>
</tr>
<tr>
<td>20</td>
<td>--</td>
</tr>
<tr>
<td>25</td>
<td>--</td>
</tr>
<tr>
<td>30</td>
<td>--</td>
</tr>
</tbody>
</table>
FIGURE 14
FILM COEFFICIENTS OF HEAT TRANSFER
AT CONSTANT COMPOSITION
VERSUS
TEMPERATURE DROP ACROSS THE FILM
17.2 MOL % ACETONE IN METHANOL

TEMPERATURE DROP (°F)

FILM COEFFICIENT, h, (BTU/HR-F² °F)

- ΔT = (Tbp - Tw)
- ΔT = (Td - Tw)
- ΔT = (Tv - Tw)
- Single Components

Methanol
Acetone
Figure 15

Film coefficients of heat transfer at constant composition versus temperature drop across the film.

42.3 mol% acetone in methanol

Film coefficient, $h$, (BTU/HR•Ft²•°F)

$\Delta T = (T_{bp} - T_w)$

$\Delta T = (T_{dp} - T_w)$

$\Delta T = (T_y - T_w)$

Single Components

Temperature drop (°F)
FILM COEFFICIENTS OF HEAT TRANSFER AT CONSTANT COMPOSITION versus TEMPERATURE DROP ACROSS THE FILM 57.3 MOL% ACETONE IN METHANOL

\[ \Delta T = (T_{bp} - T_w) \]
\[ \Delta T = (T_{dp} - T_w) \]
\[ \Delta T = (T_v - T_w) \]

- Acetone
- Methanol
- Single Components

TEMPERATURE DROP (°F)
FIGURE 17
FILM COEFFICIENTS OF HEAT TRANSFER AT CONSTANT COMPOSITION versus TEMPERATURE DROP ACROSS THE FILM 80 MOL% ACETONE IN METHANOL (AZEOTROPE) 500

\[ T_{bp} = T_{dp} = T_v \]
\[ \frac{d}{d} \Delta T = (T_v - T_w) \]

Single Components

TEMPERATURE DROP (°F)

Methanol
Acetone
FIGURE 18
FILM COEFFICIENTS OF HEAT TRANSFER
AT CONSTANT COMPOSITION

versus
TEMPERATURE DROP ACROSS THE FILM
46.5 MOI % METHANOL IN BENZENE

\[ \Delta T = (T_{bp} - T_w) \]

\[ \Delta T = (T_{bp} - T_w) \]

\[ \Delta T = (T_v - T_w) \]

Single Components
FIGURE 19
FILM COEFFICIENTS OF HEAT TRANSFER
AT CONSTANT COMPOSITION
VERSUS
TEMPERATURE DROP ACROSS THE FILM
61.2 MOL. % METHANOL IN BENZENE
( AZEOTROPE )

\[ T_{bp} = T_{dp} = T_v \]
\[ \Delta T = (T_v - T_w) \]

Methanol
Benzene
Single Components

TEMPERATURE DROP (° F)
FIGURE 20

FILM COEFFICIENTS OF HEAT TRANSFER
AT CONSTANT COMPOSITION
VERSUS
TEMPERATURE DROP ACROSS THE FILM
83 MOL% METHANOL IN BENZENE

TEMPERATURE DROP

FILM COEFFICIENT, \( h \) (BTU/HR-FT²°F)

\[ T = (T_{bp} - T_w) \]

\[ \Delta T = (T_{dp} - T_w) \]

\[ \Delta \Delta T = (T_V - T_w) \]

Single Components
drop across the film based on the three temperatures mentioned above was made for each series of runs at a constant composition (See Fig. Nos. 14-20). Again it seems that the plot should be a straight line with approximately the theoretical slope of -0.25 on log-log paper since the group \( \left( \frac{R_i}{L_i} \right)^{1/4} \) is believed to be almost independent of temperature just as with the pure substances as discussed in the theory section. This was found to be generally the case for the coefficients based on the boiling point. The best line representing the experimental data was then plotted. From this line, the heat transfer coefficients for a temperature drop of 10°, 15°, 20°, 25°, and 30°F. were read and recorded in Table No. III.

Difficulty was encountered at low concentrations of methanol in benzene in getting a smooth film of condensate. At a concentration of 46.5 mol% methanol in benzene a peculiar type of condensation was noted (Figure 21). At low temperature drops across the film, a ring appearance was noted in the film of condensate. These rings of condensate were uniformly spaced as shown. They seemed to move gradually toward the center of the condensing surface. As the rate of condensation was further reduced, the uniform ring formation was disrupted and a random uneven pattern was noted in the film (Fig. No. 22). It is believed that this non-uniform film at the lower rates of condensation accounts for the curve upward in the determination of the heat transfer coefficients for 46.5 mol per cent methanol in benzene. For
FIGURE 21
RING APPEARANCE IN FILM OF CONDENSATE AT LOW CONCENTRATION OF METHANOL IN BENZENE AND INTERMEDIATE RATES OF CONDENSATION
IRREGULAR PATTERN OF CONDENSATE AT LOW CONCENTRATION OF METHANOL IN BENZENE AT LOW CONDENSATION RATES
high rates of condensation, the uniform film again appeared. For 17.2% methanol in benzene, the condition was even worse. A uniform film could be obtained only at very high rates of condensation, consequently determinations at temperature drops less than 20°F were not made. This same sort of film appearance has been recently reported by Bromley\(^{(1)}\) for pure water and for pure acetone, not a mixture; and this observation was noted on an apparatus which was constructed from stainless steel.

Plots were then made of the film coefficient of heat transfer at a constant temperature drop as a function of mol fraction as shown in Figs. 23, 24, and 25. The data of Trachtenberg is here included for comparison. A comparison of the three sets of curves for each system immediately brings out the non-similarity. For the acetone-methanol system and the benzene-methanol system, there are three points in common, the terminals and the azeotropic composition points. For the isopropanol-methanol systems only the terminal points are in common.

In each case the uniformity of the curves with the coefficient based on the boiling point is striking. In each case, the similarity of the curves based on the dew point and the vapor temperature is evident and such should be the case for the two temperatures are not greatly different. In all three systems, a minimum is reached in the curves based on the vapor and dew point temperatures. In the case of the
Figure 23

Film coefficients of heat transfer at constant temperature drop versus composition

(Acetone-Methanol System)

Coefficient based on the boiling point:
\[ \Delta T = (T_{bp} - T_w) \]

Coefficient based on the dew point:
\[ \Delta T = (T_{dp} - T_w) \]

Coefficient based on the vapor temperature:
\[ \Delta T = (T_v - T_w) \]
FIGURE 2.4
FILM COEFFICIENTS OF HEAT TRANSFER
AT CONSTANT TEMPERATURE DROP
VERSUS
COMPOSITION
(Methanol-Benzene System)

COEFFICIENT BASED ON THE
BOILING POINT
$\Delta T = (T_{bp} - T_w)$

COEFFICIENT BASED ON THE
DEW POINT
$\Delta T = (T_{dp} - T_w)$

COEFFICIENT BASED ON THE
VAPOR TEMPERATURE
$\Delta T = (T_v - T_w)$

NOTE: The Dotted Lines Represent
Constructed Points For 17.7 Mol %
FIGURE 25
FILM COEFFICIENTS OF HEAT TRANSFER
AT CONSTANT TEMPERATURE DROP
VERSUS
COMPOSITION
(Methanol-Isopropanol System)

COEFFICIENT BASED ON THE BOILING POINT
\[ \Delta T = (T_{bp} - T_w) \]

COEFFICIENT BASED ON THE DEW POINT
\[ \Delta T = (T_{dp} - T_w) \]

COEFFICIENT BASED ON THE VAPOR TEMPERATURE
\[ \Delta T = (T_v - T_w) \]

FILM COEFFICIENT, \( h \) (BTU/hr-ft\(^2\)-F)

MOL. % METHANOL

\( \Delta T = 10^\circ F \)
\( \Delta T = 15^\circ F \)
\( \Delta T = 20^\circ F \)
\( \Delta T = 25^\circ F \)
\( \Delta T = 30^\circ F \)
isopropanol-methanol system, the curve is uniform and approaches the minimum gradually. A check of the temperature-composition plot for isopropanol shows that there is a relatively small change in temperature between the saturated vapor and liquid compositions. The higher saturated vapor temperature accounts for the dip in the curves plotted. The acetone-methanol temperature-composition plot shows a wider variation in the temperatures of the saturated vapor and liquid at a given composition except near the azeotropic point. There is a pronounced minimum at approximately 80 mol per cent methanol but none at compositions higher than the azeotropic composition. The latter is due to the fact that the difference between the saturated vapor and liquid temperatures between zero and twenty mol per cent methanol is nil. A glance at the plots for the methanol-benzene system shows more pronounced minima on either side of the azeotropic composition for coefficients based on the dew points and boiling temperatures. Again, these minima occur at points of greatest temperature difference between liquid and saturated vapor.

In order to determine the significance of these curves, it must be recalled that a plot of the group \( \left( \frac{R^2}{\mu^2} \right)^{1/4} \) as a function of composition was made for the ethanol-water, acetic acid-benzene, and ethanol-methanol systems (Fig. No. 5). These lines, though not all approaching linearity, did not show in any case more than one point of inflection. It seems that we could expect similarly shaped curves
FIGURE 26
FILM COEFFICIENTS OF HEAT TRANSFER
AT CONSTANT TEMPERATURE DROP
BASED ON THE BOILING POINT
VERSUS
COMPOSITION

ACETONE-METHANOL  BENZENE-METHANOL  ISOPROPANOL-METHANOL

WEIGHT PER CENT METHANOL

FILM COEFFICIENT, h, (BTU/Hr-Ft °F )
for the heat transfer coefficients as a function of composition at a constant temperature drop across the film since only the temperature drop across the film would be appreciably affecting the values. If only the terminal points and the aseotropic composition are considered, such a curve does result. Certainly, the curve based on the boiling point approaches these expected results closer than those obtained using the dew point or vapor temperature as a basis for calculating the heat transfer coefficients.

Since the plots of thermal conductivity, density, and viscosity seemed to approach linearity when plotted as a function of weight percent rather than a function of mol percent, the same scheme was carried out using the film coefficients of heat transfer based on the boiling point (Fig. No. 26). This change did serve to smooth out the dip somewhat except in the case of the acetone-methanol system. In the case of the isopropanol-methanol system we have lines which approach linearity closely, particularly at the higher rates of heat transfer. This system is one that approaches ideality.

It must be pointed out that a small error in the vapor-liquid equilibrium diagram will cause more irregularity in the lines representing heat transfer coefficients for small temperature drops than for the larger ones for obvious reasons. Thus the effect of composition on the coefficients should be better shown for the constant temperature drop of 30°F.
For the benzene-methanol plot, a small upsurge in the line is noted at approximately 20 weight % methanol. This probably stems from the peculiar condensation noted previously wherein the film was not of uniform smoothness resulting in higher values for the coefficients.

The acetone-methanol plot has the same appearance for low methanol concentrations and in addition, a downward bend is noted in the neighborhood of 70 weight per cent. The reason for this is not known, but it must be remembered that the vapor-liquid equilibrium data was somewhat of questionable value. However, these variations are not very far from a straight line connecting the values of the coefficients of the single components.
CHAPTER VII

CONCLUSIONS

It seems clear from the work done on pure compounds and the statistical analysis of the data, that a workable apparatus has been designed. The desirable features of isothermal conditions along the length of the condensing surface and an improved technique of thermocouple installation have been successfully incorporated into the apparatus.

The work with binary solutions has evolved a workable procedure to be followed in determining film coefficients of heat transfer as a function of composition. It appears that for ideal systems, the coefficients at any composition may be determined by plotting that for the pure components at a given temperature drop at zero and 100 weight per cent and connecting the two points by a straight line. The coefficient at a given weight per cent then can be read simply from the plot. This conclusion is not final, rather further evidence from similar investigations on solutions approaching ideality is needed. For non-ideal solutions, true values for the film coefficients do not seem to be so easily obtained. True, in this case the variation from linear relationship is small; and in the absence of experimental data a procedure might be followed similar to that for ideal solutions without too much error. However, it is believed that solutions containing water as one
component should not be handled in this manner, since the variation of the group \( \left( \frac{k^2 \rho^2}{\mu^2} \right)^{\frac{1}{4}} \) with composition calculated from existing data though meager was seen to vary non-uniformly with composition. This seems to be borne out by the findings of Wallace and Davison, scant though they are. Water is singled out here since it is such a highly associated substance as pointed out by Ewell\(^6\).

Furthermore, it appears that the temperature drop across the film of condensate is the difference between the surface temperature and the boiling point of the condensate for these particular operating conditions. It must be remembered that this is a limiting condition as opposed to a case in which only a small fraction of the entering vapor is condensed. For the latter case, the use of the boiling point as the interfacial temperature will not necessarily be correct since other resistances to transfer enter into the picture.

It seems reasonable to expect that the film coefficients of heat transfer could be predicted for binary solutions that form a single liquid phase by the Nusselt Equation when the necessary physical data become available.
CHAPTER VIII

RECOMMENDATIONS

Further work seems to be in order on both ideal and non-ideal solutions, with water as one component particularly of importance.

This work has served to point out the desirability of having values for density, viscosity, and particularly thermal conductivity as a function of composition and temperature. It is believed that this equipment might be used to study the value of the vapor side film temperature as a function of the degree of partial condensation when this data becomes available.

It is believed that vapor-liquid equilibrium data should be determined for the reagents actually used in the heat transfer work, as small temperature errors (even 1 or 2°F) are important particularly at the lower temperature drops across the film.

Slight changes in the apparatus should be made to ease the operating technique as well as to improve the results obtainable. The vapor jacket probably should be enlarged to minimize adverse vapor velocity effects. Steps should be taken to provide a better control of the heat input. Cleaning of the condensing surface should be made easier, or the condensing surface should be made less liable to tarnishing perhaps through plating on a thin film of non-corrosive metal. An easier
method of measuring the condensate should be devised, perhaps a volumetric system which would be almost automatic. The condensate return line should be brought to the center of the boiler to prevent a concentration gradient across the boiler from one end to the other. Perhaps, the temperature measurements may be speeded by connecting all the surface thermocouples in series and reading a total voltage generated, then dividing by the number of thermocouples to get an average value. This would allow installation of thermocouples at other strategic points, since the limitation has been the time necessary to take the readings. Separate readings for each surface thermocouple do not seem to be so important for subsequent work since the variation around the circumference has been found to be rather small.


(19) Sakiadis, B. S., and Coates, J., A Literature Survey of the Thermal Conductivity of Liquids, Engineering Experiment Station Bulletin No. 34, Louisiana State University, Baton Rouge, Louisiana, 1952.


NOMENCLATURE

\(a\) = distance from wall out into film of condensate (ft)

\(A\) = area of heat transfer \((ft^2)\)

\(B\) = assumed a constant

\[ B = \frac{3\mu f \kappa (t_v - t_w)}{\int \rho^2 \lambda \lambda} \]

\(c\) = concentration \((\text{moles/ft}^3)\)

\(C\) = constant of integration

\(C_{pA}, C_{pB}\) = molal specific heat of components A and B respectively \((\text{BTU}/(\#\text{mol})(\degree\text{F}))\)

\(D_{avg}\) = average diameter of pipe (ft)

\(D_i\) = inside diameter of pipe (ft)

\(D_o\) = outside diameter of pipe (ft)

\(D_v\) = molecular diffusivity in gas \((ft^2/hr)\)

\(g\) = acceleration due to gravity \((4.17 \times 10^8 \text{ ft/hr}^2)\)

\(G\) = mass rate of flow \((\#/hr)(ft^2)\)

\(G_m\) = molal rate of flow \((\#\text{mol}/hr)(ft^2)\)

\(h\) = local coefficient of heat transfer \((\text{BTU}/hr)(ft^2)(\degree\text{F})\)

\(h_{ag}, h_{bg}\) = enthalpy of saturated gas of components A and B respectively \((\text{BTU}/\#)\)

\(h_{dL}\) = coefficient of heat transfer of dirt on liquid side of the pipe

\(h_{dL}\) = coefficient of heat transfer of dirt on liquid side of the pipe \((\text{BTU}/hr)(ft^2)(\degree\text{F})\)

\(h_{dv}\) = heat transfer coefficient of dirt on the vapor side of the wall

\(h_{dv}\) = heat transfer coefficient of dirt on the vapor side of the wall \((\text{BTU}/hr)(ft^2)(\degree\text{F})\)
\[ h_L = \text{coefficient of heat transfer of liquid} \]
\[
= \text{(BTU/hr-ft}^2\text{-°F)}
\]

\[ h_{1A}, h_{1B} = \text{enthalpy of saturated liquid of components A and B respectively} \]

\[ h_m = \text{mean coefficient of heat transfer (BTU/hr-ft}^2\text{-°F)} \]

\[ h_v = \text{vapor side film coefficient of heat transfer, (BTU/hr-ft}^2\text{-°F)} \]

\[ j_{H^*J_D} = \text{dimensionless factors for heat and mass transfer respectively} \]

\[ k = \text{thermal conductivity (BTU/hr-ft}^2\text{-°F/ft)} \]

\[ k_g = \text{mass transfer coefficient (#mols)/(hr-ft}^2\text{-atm)} \]

\[ L = \text{thickness of the pipe wall (ft)} \]

\[ M = \text{molecular weight} \]

\[ N_A = \text{rate of mass transfer of more volatile component (#mols/hr)} \]

\[ p = \text{partial pressure (atm)} \]

\[ P = \text{total pressure (atm)} \]

\[ P_{BM} = \text{log mean pressure of less volatile vapor (atm)} \]

\[ p_i = \text{partial pressure at interface (atm)} \]

\[ p_s = \text{partial pressure in main stream (atm)} \]

\[ q = \text{rate of heat transfer (BTU/hr)} \]

\[ R = \text{gas constant (ft}^3\text{-atm)/ (#mol-°R)} \]

\[ r = \text{radius of pipe (ft)} \]

\[ R_t = \text{total resistance} \]

\[ S = \text{shearing stress (# force/ft}^2) \]

\[ T = \text{absolute temperature (°K)} \]

\[ t_f = \text{film temperature} = t_v - 3/4 \Delta t, \text{ where } \Delta t \text{ is temperature} \]
drop across the film ($^\circ$F)

$t_i$ = temperature at interface ($^\circ$F)

t$_v$ = vapor temperature ($^\circ$F)

t$_w$ = temperature of condensing surface ($^\circ$F)

$u$ = downward velocity of the film ($(\text{ft})/\text{hr}$)

$U_o$ = overall coefficient of heat transfer based on outside area of pipe (BTU/(hr)($\text{ft}^2$)($^\circ$F))

$U$ = overall coefficient of heat transfer (BTU/(hr)($\text{ft}^2$)($^\circ$F))

$w$ = mass rate of transfer (#/hr($\text{ft}^2$))

$x_a$ = mol fraction of more volatile component in liquid phase

$y$ = mol fraction of vapor, $y_v$ in main vapor stream $y_1$ at interface

$Y$ = film thickness at a given point (ft)

$z$ = distance in direction of diffusion (ft)

$z_1$ = a function of film thickness

$\Gamma$ = mass rate of flow over a surface per unit length (lb/hr-ft)

$\Delta t$ = temperature difference ($^\circ$F)

$\lambda$ = latent heat of vaporization (BTU/#)

$\lambda_m$ = average molal latent heat for solution (BTU/#mol)

$N$ = coefficient of viscosity (Mass/unit length - unit time) = $M_F$ $f$

$M_F$ = viscosity in force units (Force - Time/Length$^2$)

$\rho$ = density (#/s/ft$^3$) or consistent units

$\phi$ = angle of surface with reference to horizontal plane (radius)

$\psi$ = a function of film thickness in substitution

$f$ = refers to average film properties
Calculation of Confidence Limits According to Brownlee$^2$.

There is reason to expect that heat transfer coefficients can be correlated for a given substance or a given composition of a mixture by the following relation:

$$\log h = \log k - m \log \Delta T$$

where $h$ is the heat transfer coefficient, $k$ and $m$ are constants, and $\Delta T$ is the temperature drop across the film.

To test the significance of this linear relationship, the correlation coefficient, $r_c$ must be calculated as follows

$$r_c = \frac{\sum (\Delta T - \bar{\Delta T})(h - \bar{h})}{\sqrt{\sum (\Delta T - \bar{\Delta T})^2 \sum (h - \bar{h})^2}}$$

where $\Sigma$ denotes summation and

$$\sum (\Delta T - \bar{\Delta T})^2 = \sum (\Delta T^2) - \frac{(\sum \Delta T)^2}{n}$$

$$\sum (h - \bar{h})^2 = \sum (h^2) - \frac{(\sum h)^2}{n}$$

$$\sum (\Delta T - \bar{\Delta T})(h - \bar{h}) = \sum (h \Delta T) - \frac{\sum \Delta T \sum h}{n}$$

By use of Table IV p. 150, the probability of getting such a value is determined. If the probability is high then there is good reason to suspect that the linear relationship exists. For the determination on methanol, the correlation coefficient is 0.9499. The probability
read from the table of getting this value in the absence of the correlation is nil, consequently the use of the relationship is sound.

The equation for the line is calculated according to the method of least squares. The calculation for methanol gives an equation

\[
\log h = 2.994566 - .27235 \log \sqrt{T}.
\]

The standard deviation, \( \sigma_r \), may be calculated as follows

\[
\sigma_r = \sqrt{\frac{1}{n - 2} \sum (h - \bar{h})^2}
\]

This standard deviation may then be used to draw the limits in which a given percentage of the data should fall. For limits in which 95% of the data will fall, use Table I, p. 144, to determine a value for \( t \).

For methanol the standard deviation was found to be 0.00993134.

The value of \( t \) is 2.1 to give a deviation from the line of \( \pm 0.020856 \) logarithmic units, thus establishing the confidence limits.
FIGURE 27
ENTHALPY-CONCENTRATION DIAGRAM
ACETONE - METHANOL

Saturated Vapor

Saturated Liquid

MOL PER CENT ACETONE

( BTU/POUND )

ENTHALPY

560
520
480
440
400
360
320
280
240
200
160
120
80
40
0
0 10 20 30 40 50 60 70 80 90 100
FIGURE 29
BOILING POINT DIAGRAM
Acetone - Methanol
FIGURE 30
BOILING POINT DIAGRAM
Methanol-Benzene (3)
FIGURE 31
BOILING POINT DIAGRAM
Methanol : Isopropanol

TEMPERATURE (°F)

MOL % METHANOL
James B. Todd, Jr., was born January 17, 1924, in Franklin, Louisiana. He attended public schools in East Baton Rouge Parish and graduated from Baton Rouge High School in 1941. He was enrolled at Louisiana State University, Baton Rouge, Louisiana, from June, 1941 to June, 1943 at which date he entered the Army of the United States. Upon discharge from the Army in September, 1946, he again enrolled at Louisiana State University and was graduated in June, 1948, with the Degree of Bachelor of Science in Chemical Engineering. He received the Master's Degree in Chemical Engineering in 1950 from Louisiana State University. He continued graduate studies at the same institution until August, 1951, at which time he re-entered the Army to serve as a Gunnery Instructor in the Army Field Artillery School until November, 1952. He then re-entered the University and at present is a candidate for the Degree of Doctor of Philosophy.
EXAMINATION AND THESIS REPORT

Candidate: James B. Todd, Jr.

Major Field: Chemical Engineering


Approved:

[Signatures]

Major Professor and Chairman

Dean of the Graduate School

EXAMINING COMMITTEE:

[Signatures]

Date of Examination:

May 5, 1953