Heat Transfer Coefficients for Condensing Organic Vapors of Pure Components and Binary Mixtures

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The behavior of the film coefficient of heat transfer for the condensation of organic vapor mixtures was investigated experimentally to establish a satisfactory basis for applying the Nusselt equation to binary systems. Five ideal and nonideal pairs, all of which gave miscible condensates, were studied; the work was carried out under conditions of almost total condensation on a horizontal condensing surface designed to comply as rigidly as possible with the conditions for which Nusselt's equation is valid.

The same behavior was observed for all systems and all concentrations studied: the experimental coefficients fell between those for the pure components and followed the behavior pattern for pure components when the temperature difference was taken as that between the *bubble point* of the condensate and the surface temperature, rather than between the dew point or the measured vapor temperature and the surface temperature. Correlation of the film coefficient showed it to vary approximately linearly with composition if the coefficients were compared at a constant value of the temperature difference, defined as above. This permits determination of the coefficient for a mixture by interpolation between the coefficients for the pure components, which are easily obtained, in preference to making the calculations with the properties of the mixture obtained by laborious and uncertain weighting of the corresponding properties of the pure components.

When results are interpreted in the light of the theories of Colburn and Drew, the presence of a vapor-phase resistance to heat and mass transfer, as postulated by them, is indicated.

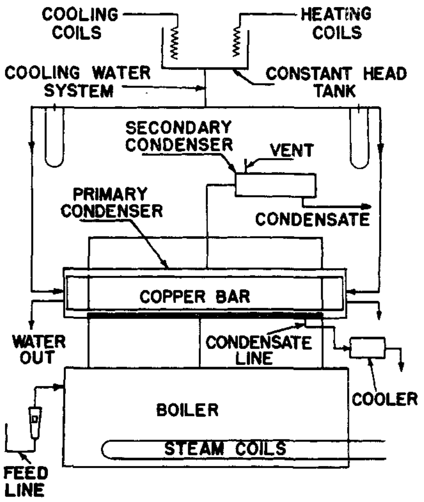


Fig. 1. Test assembly.

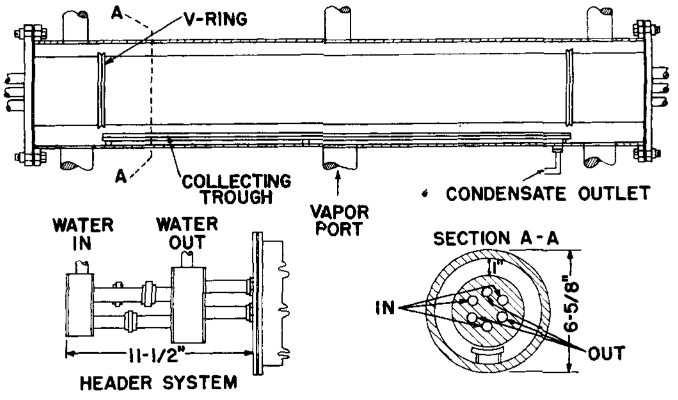
The Nusselt equation (*8*) is the most widely used method for predicting the coefficient of heat transfer for condensing vapors. Its validity for pure materials has been established by many investigators (*9, 10, 13*) and its concepts have been extended to apply to mixtures in which one component is noncondensable and to mixtures in which the condensate consists of two immiscible liquids (*1, 2*). However, the condensation of mixtures which give a condensate with a single liquid phase has received a minimum of attention despite the frequency with which this situation occurs in industry. Colburn and Drew (*3*) analyzed the principles involved and developed theoretical relations predicting the behavior of condensing mixtures; Wallace and Davidson (*18*) reported data for the system ethanol-water which gave limited verification to the Colburn-Drew theories. Accordingly, this research was undertaken to provide additional data which would test the Colburn-Drew theory and to establish an accurate and practical method of applying it to design.

APPARATUS AND PROCEDURE

The test assembly, shown in Figure 1, consisted of a boiler which generated the vapors, a horizontal primary condenser on which the test data were obtained, a secondary condenser into which a small amount of excess vapor was vented, and a water-coolant system.

The primary condensing surface was a 4-in. copper cylinder 3 ft. long, installed concentrically in a 6-in. insulated steel pipe which formed the vapor space as shown by Figure 2. Coolant circulated through six ⅝-in. holes drilled longitudinally through the cylinder, entering three of the channels at one end and three at the other in accordance with the method first used by Othmer and White (*11*) to approach isothermal conditions. As shown by Figure 2, end effects were minimized by collecting the condensate from the center 28 in. and withdrawing this for weighing and analysis.

Fig. 2. Condensing surface.



Surface temperatures were measured by ten embedded thermocouples located at various circumferential and longitudinal positions as shown in Figure 3. Installation and additional mechanical details are given in the Appendix.*

All work was carried out at atmospheric pressure plus the 1 or 2 in. of water pressure necessary to maintain the excess vapor flow to the secondary condenser. For each material and concentration a series of tests was made with the temperature difference across the condensate film as the primary variable. This could be controlled over the

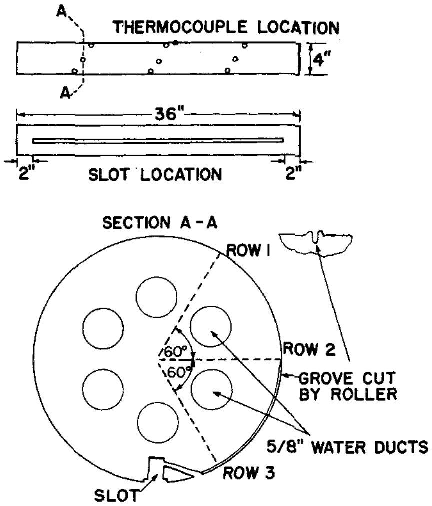


Fig. 3. Thermocouple installation.

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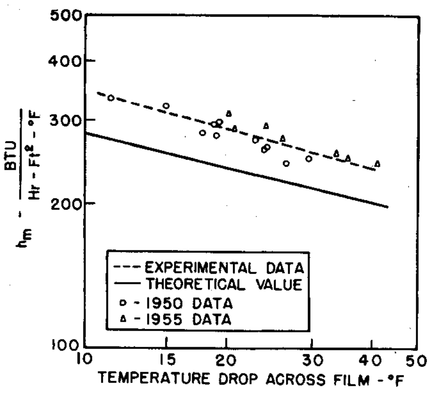
*The Appendix has been deposited as document 5302 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $1.25 for photoprints or 35-mm. microfilm.

range of 7° to 50°F. by regulating the coolant-water temperature and rate. During each test all thermocouple readings and condensation rates were observed at frequent intervals and any tests showing a significant variation between start and finish were discarded.

Seven organic materials and five binary mixtures of these were studied. To minimize the possibility of fouling of the surface with time, the general procedure followed was to test two pure materials, then mixtures of these. The source and properties of the materials used are shown below; because of the quantities required, technical grades were used in preference to reagent grade.



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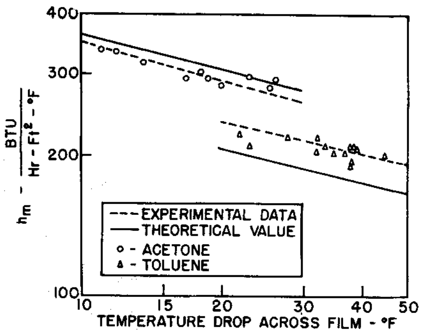

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Figs. 4, 5, 6. Variation of heat transfer coefficient with temperature drop across film: (4) benzene, (5) toluene and acetone, (6) alcohols.

		Physical Properties			
		Density, g./cc. at 25°C.		Refractive index $N_D{}^{25}$	
Material	Source	Literature	As used	Literature (*12, 15*)	As used
Benzene	Matheson Chem. Co.	0.879	0.8773	1.4980	1.4875
Toluene	General Chem. Co.	0.8623	0.8635	1.4941	1.497
Acetone	U. S. I. Chem. Co.	0.7920 at 20°	0.7917 at 20°	1.3566	1.3555
Methanol	Allied Chem. & Dye	0.7866	0.7868	1.3267	1.3268
Ethanol	U. S. I. Chem. Co.	0.7894	0.7897	1.3595	1.3588
i-Propanol	Allied Chem. & Dye	0.7812	0.7814	1.3758	1.3750
n-Butanol	Carbide & Carbon Chem.	0.8096	0.8091	1.3990	1.3966

RESULTS

General Observations

As observed through sight glasses in the vapor jacket, the condensate formed a continuous film which drained from the bar in a smooth, uniform manner in most tests. Exceptions were noted in the case of methyl alcohol–benzene mixtures, which gave an uneven drainage pattern at low condensation rates, and in many tests in which the temperature difference was about 40° to 50°F.; in the latter case the condensate could be seen to swirl and otherwise deviate from smooth flow.

Vapor velocities in the annulus around the condensing surface were calculated and found to have a maximum value less than 1 ft./sec.; the more usual value was about 0.5 ft./sec. at the inlet and must have fallen off rapidly as the vapors condensed. In general, the vapor rate to the secondary condenser was adjusted to be approximately 10% of that on the primary surface to prevent the accumulation of any noncondensables evolved from the boiling liquid and also to provide a slight positive pressure to prevent the entry of air. Thus the velocity of the vapor leaving the primary condenser was about 0.05 ft./sec.

TABLE 1. TYPICAL TEMPERATURE DISTRIBUTION AND TEST DATA

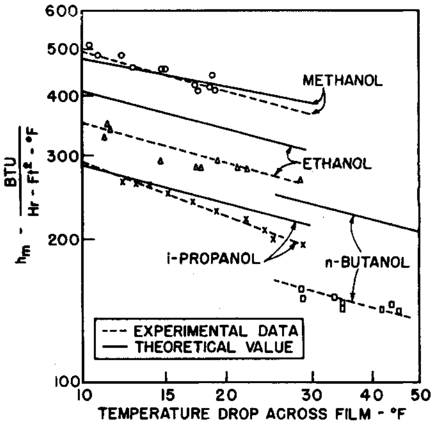
163.6°F.
162.7 162.7 163.8
162.5 162.5 163.2
161.9 160.8 162.3

(Note: temperatures shown above correspond to thermocouple locations shown in Figure 3)

Condensing vapor	Benzene
Average surface temperature	162.6°F.
Observed vapor temperature	176.7°F.
Δt	14.1°F.
Condensation rate	434 g./min. = 57.5 lb./hr.
Reynolds number ($4\Gamma'/\mu$)	121.5
Heat flow	9,750 B.t.u./hr.
h_m (experimental	289 B.t.u./ (hr.)(sq. ft./°F.)
h_m (calculated for same condensation rate)	266 B.t.u./ (hr.)(sq. ft./°F.)

The temperature distribution shown in Table 1 is typical of that found in the course of the work. In ten tests, chosen at random, the highest difference between any two thermocouple readings was 5+°F.; more commonly, the range was less than 3°F. between extremes, or approximately 18% of the average Δt, the standard deviation varying from 0.5° to 3°F., or 3 to 17% of the average Δt. Further analysis of these tests was made to determine the mean deviation of each thermocouple position from the average of all ten embedded thermocouple readings for that test; the results are summarized:

Position	Average deviation, °F.
Top of cylinder (1 thermocouple)	+1.37
First horizontal row (3 thermocouples)	+0.32
Middle horizontal row (3 thermocouples)	+0.06
Bottom horizontal row (3 thermocouples)	−0.79



(6)

This temperature pattern is explainable by the variation in the local coefficient with radial position around the bar. If, as Nusselt predicted, the coefficient is highest at the top, where the cumulative amount of condensate is a minimum, the total thermal resistance between the vapor and an isothermal plane within the copper bar is a minimum, and the local heat flux is a maximum. As the condensate runs down the face of the cylinder, it increases in thickness and thermal resistance, and the heat flux decreases; since the metal resistance is substantially the same at all radial positions, the temperature difference across it decreases and the surface temperature is thus cooler, as indicated.

Since the magnitudes of these deviations was generally small relative to the average temperature difference, and since average, rather than point, coefficients were desired, the arithmetic average of the ten embedded thermocouples was used as the surface temperature in determining the temperature difference for use in Newton's equation:

$$h_m = q/A\Delta t \qquad (1)$$

PURE COMPONENTS

Tests were made with pure components for two reasons: to compare the results

Table 2. Summary of Results—Pure Compounds

Material	Δt, °F.	Range of values $4\Gamma'/\mu$	h_m, B.t.u./(hr.)(sq. ft./°F.)	Correlation of experimental data
Benzene	7.0–29.5	105–203	251–480	$h_m = 289(\Delta t)^{-0.284}$
Toluene	21.9–53.6	146–377	186–239	$h_m = 477(\Delta t)^{-0.235}$
Methanol	8.3–18.8	42–75	409–517	$h_m = 1059(\Delta t)^{-0.336}$
Ethanol	11.2–28.3	35–62	266–350	$h_m = 652(\Delta t)^{-0.270}$
i-Propanol	12.2–28.8	32–48	194–264	$h_m = 684(\Delta t)^{-0.372}$
n-Butanol	28.2–49.9	62–80	133–158	$h_m = 401(\Delta t)^{-0.277}$
Acetone	9.7–26.0	102–208	282–362	$h_m = 659(\Delta t)^{-0.275}$

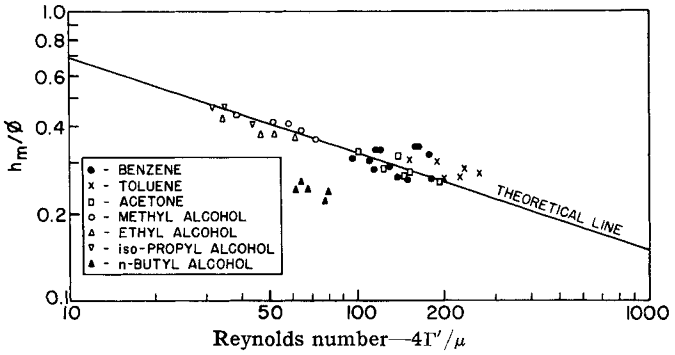


Fig. 7. Generalized correlation of heat transfer coefficient with Reynolds number.

obtained with this apparatus with the theoretical values predicted by the Nusselt equation and to provide experimental values for the pure components against which their mixtures could be compared under the same conditions.

Results are summarized in Table 2; typical test results are shown graphically for the various materials as Figures 4, 5, and 6. Of primary interest is the agreement between the observed and the theoretical values, which can be seen visually in Figures 4 to 6 and is summarized statistically in Table 3.

Figures 4 to 6 test the agreement of the data with one form of Nusselt's equation:

$$h_m = 0.725(k^3\rho^2 g\lambda/D\mu\Delta t)^{0.25} \qquad (2)$$

Since Δt was the controlled variable, for Equation (2) to be valid the logarithmic plot should show a linear variation of h with Δt and should have a slope approximating -0.25. Deviation from 0.25 is to be expected since the physical properties of the condensate vary with temperature; the properties are evaluated at a film temperature, $t_f = t_{sv} - 0.75\,\Delta t$, and would, therefore, vary with the value of Δt.

As shown in Table 2, values of the slope in Figures 4 to 6 varied from -0.24 to -0.37. Although it was essentially parallel to the theoretical line in all cases, the best statistical line fell significantly above the theoretical for benzene and toluene and below the theoretical for ethanol, butanol and acetone. High values may be attributed to turbulence induced by the vapor velocity and low values to the fouling of the condenser surface; however, since both high and low results were obtained and the averages shown in Table 3 are in good quantitative agreement with the corresponding values reported in McAdams (*6*), it is more probable that the difference represents a variance between the physical properties of the actual materials tested and the values reported for these in the literature and used in calculating the theoretical values of the coefficient. Whatever the cause of this discrepancy, the mutual consistency and reproducibility of the results lend confidence to their value and the use of the apparatus for the binary studies.

Figure 7 represents the alternate, generalized form of Nusselt's equation:

$$h_m/\phi = 1.51(4\Gamma'/\mu_f)^{0.333} \qquad (3)$$

where

$$\phi = (k^3\rho^2 g/\mu^2)^{0.333} \qquad (3a)$$

$4\Gamma'/\mu_f$ is the Reynolds number for the drainage of the condensate film from a horizontal tube. Its critical value is generally accepted to be about 4,200, but it will be noted in Figure 7 that there is an indication that turbulence begins at a lower value, possibly as low as 200, and that at Reynolds numbers above this the coefficient increases, rather than decreases, with increasing condensation rates; this behavior has been established for condensation on vertical tubes (*7*) and has been suggested for other transfer phenomena (*14*). Additional data at higher condensation rates, not obtainable with this apparatus, are needed to establish this quantitatively.

Table 3. Statistical Analysis of Results—Pure Compounds

Material	Standard deviation in h_m From experimental correlation, B.t.u./(hr.)(sq.ft.)(°F.)	From theoretical value for same Δt, B.t.u./(hr.)(sq.ft.)(°F.)	$\left[\frac{h_{m\ expt'l.}}{h_{m\ theor.}}\right]_{avg.}$ Experimental	Literature (*9*)
Benzene	17.8	66.2	1.27	0.84–1.22
Toluene	21.3	38.1	1.16	0.76–1.04
Methanol	19.9	24.5	1.00	1.06
Ethanol	11.7	66.1	0.85	1.02
i-Propanol	3.6	15.8	0.95	0.73–0.94
n-Butanol	5.3	85.8	0.65	0.85
Acetone	10.8	17.6	0.97	

BINARY MIXTURES

To apply Equation (1) and determine h_m for the binary mixtures q was evaluated by multiplying the difference between the enthalpies of a saturated vapor and a saturated liquid of the condensate composition by the weight rate at which the condensate was collected; the enthalpy chart and the vapor-liquid equilibrium diagram for each system, needed for this purpose, were prepared from the best available equilibrium and thermal properties data (*12*, *15*). Typical results are given in Table 4.

As shown by Figure 8 and as considered speculatively by Colburn and Drew (*3*) and Wallace and Davidson (*18*), three choices exist as to the correct temperature to be used with the surface temperature to establish Δt for use in Equation (1). These are t_v, the vapor temperature, measured by a thermocouple in the line between the reboiler and the vapor jacket around the bar; t_{DP}, the dew point of the vapor, read from the temperature-composition diagram for the system; and t_{BP}, the bubble point of the condensate, also read from the temperature-composition diagram. By definition these give

$$\Delta t_v = t_v - t_{surface} \qquad (4a)$$

$$\Delta t_{DP} = t_{DP} - t_{surface} \qquad (4b)$$

$$\Delta t_{BP} = t_{BP} - t_{surface} \qquad (4c)$$

Values of the film coefficient were calculated for each set of data, by use of each of these choices of Δt. The results obtained with *n*-butanol–benzene are typical and are shown in Figure 9. If the three lines obtained from the same experimental data are studied in relation to the corresponding lines for the pure components, it will be noted that the definition of Δt by Equation (4*c*) was the only one which gave a slope approximating that of the pure components. Further, the line based on Δt_{BP} is the only one to fall between the pure com-

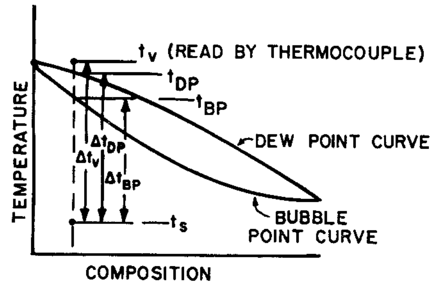


Fig. 8. Definitions of Δt for binary mixtures.

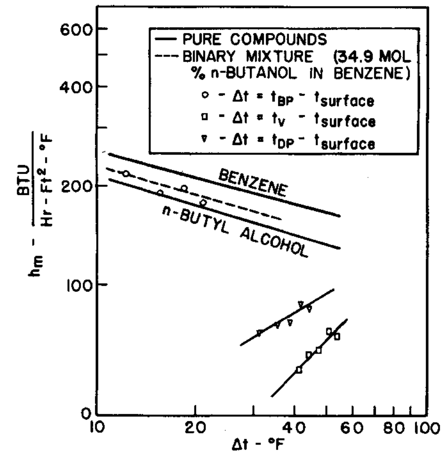


Fig. 9. Correlation of film coefficient with temperature difference for binary mixtures (*n*-butyl alcohol–benzene system).

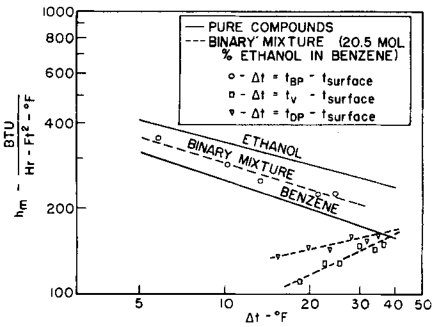


Fig. 10. Correlation of film coefficient with temperature difference for binary mixtures (benzene-ethanol system).

ponents over the entire range of Δt's studied.

The *n*-butanol–benzene system for which the data were plotted in Figure 9 is nonazeotropic, if not ideal. When ideal mixtures are considered, the physical properties of the mixture are the weighted average of those of the pure components and must fall between them; therefore, the heat transfer coefficient based on these should also fall between the limiting values established by the pure components at the same Δt. The conclusion follows that the correct Δt to use is that based on the bubble point of the condensate.

Figure 10 shows the same results for ethanol-benzene, a nonideal mixture. Again, the coefficient calculated with a Δt based on the bubble point was the only one to fall between the values for the pure components and the only one with a slope approximating −0.25. Although the physical properties of a nonideal mixture are not necessarily additive and the coefficient need not fall between those for the pure components, the slope should nevertheless be essentially −0.25 since all tests were made with the same mixture and, within a few degrees, for the same film temperature.

For each of the binary systems studied, the experimental coefficient was correlated with composition. This is shown in Figure 11, for the system ethanol-benzene; temperature difference is used as a parameter to put all mixtures on a comparable basis. The resulting plot approaches linearity, particularly at the higher values at Δt. Even at lower Δt's, where the plot deviated markedly from a straight line, 7% was the maximum difference found between the observed

Table 4. Test Results*

System	Condensate composition	Avg. surface temperature, °F.	Observed vapor temperature, °F.	Bubble point, °F.	Δt_{BP}, °F.	Condensate rate, lb./hr.	Experimental coefficient, B.t.u./(hr.)(sq. ft./°F.)
Methanol-benzene	0.0 mole % alcohol	158.0	176.7	175.3	17.3	77.1	293
	0.0	150.6	176.8	175.3	24.7	100.9	273
	0.0	140.5	177.3	175.3	34.8	125.2	242
	17.7	114.9	165.8	137.1	22.2	60.5	273
	46.5	115.6	143.7	136.0	20.4	46.1	310
	61.2†	117.7	136.7	135.7	17.9	37.3	317
	83.0	129.1	142.4	136.9	13.3	18.6	436
	100.0	128.9	147.3	148.1	19.2	38.9	418
	100.0	136.8	147.5	148.1	11.3	26.4	487
Ethanol-Benzene	20.5 mole % alcohol	138.2	167.9	155.4	17.5	50.3	285
	44.8†	140.8	153.8	153.8	13.0	34.6	289
	57.5	127.5	158.6	154.1	26.7	52.8	239
	81.3	141.2	166.7	158.3	17.5	30.9	250
	100.0	144.5	172.9	173.3	28.8	48.6	266
	100.0	155.2	172.9	173.3	18.1	33.6	284
n-Butanol-benzene	20.9 mole % alcohol	167.0	195.2	178.4	11.2	31.4	224
	34.9	160.4	213.5	181.3	21.0	42.7	179
	58.5	169.8	228.0	189.6	18.6	29.6	158
	83.7	200.6	237.2	210.0	8.9	14.6	179
	100.0	197.8	243.2	243.9	46.1	60.9	143
	100.0	210.0	243.2	243.9	33.9	47.5	153
Methanol-acetone	0.0 mole % alcohol	114.6	132.6	133.0	18.4	58.0	302
	0.0	123.4	133.1	133.0	9.6	37.8	362
	17.7	114.9	165.8	137.1	22.2	60.5	273
	20.0†	115.7	131.3	131.3	15.6	45.9	348
	42.7	124.0	133.5	131.6	7.6	24.9	468
	57.7	108.2	137.1	132.6	24.4	46.3	307
	82.5	120.9	143.4	138.4	17.5	25.4	372
Methanol-isopropanol	0.0 mole %	158.3	180.2	180.5	22.2	40.1	220
	0.0 methanol	168.0	180.2	180.5	12.5	26.8	264
	27.5	161.0	173.5	167.8	6.8	21.1	408
	42.5	140.3	169.9	162.0	21.7	41.8	273
	62.8	142.7	165.5	156.2	13.5	34.9	399
	80.0	144.5	159.6	152.0	7.5	26.5	603

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*Typical results only. Complete data available in references *5, 8, 16, 17,* and/or research records.
†Azeotrope composition.

coefficient and the corresponding value predicted by linear interpolation between the values for the pure components at the same Δt.

In both these regards—the correlation with Δt_{BP} and the linearity of h_m with composition—*all* binary systems investigated behaved similarly. Since these included methanol-isopropanol (ideal), butanol-benzene (nonideal, but nonazeotropic), methanol-benzene, acetone-methanol, and ethanol-benzene (minimum boiling azeotropes), the conclusions appear to apply to all types of mixtures.

These experimental results can be considered in terms of the theoretical relations developed by Colburn and Drew (*3*) to predict interface concentrations and temperatures as the rate of condensation and the conductance of the gas and liquid films varied.

First the fact that the bubble-point temperature exists at the liquid-vapor interface requires that there be a gas-phase resistance to account for the difference in the vapor temperature and the bubble point: were this resistance negligible, the vapor temperature or the dew point would determine the temperature driving force across the condensate film; however, as shown, this did not correlate the results.

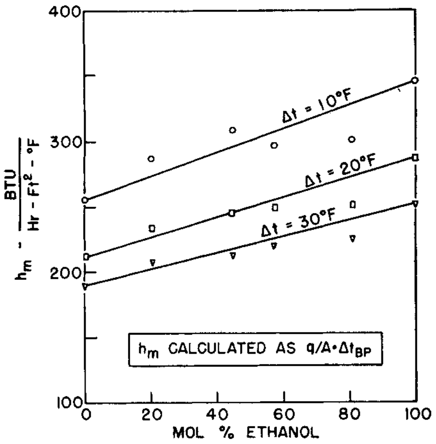

Fig. 11. Variation of coefficient with composition of binary mixtures (ethanol-benzene system).

Colburn and Drew postulated that higher rates of condensation would tend to give total condensation (no fractionation) and lower rates for the same system would tend to give equilibrium condensation, with a corresponding variation in the interface temperature. No such variation was observed; in all tests the use of bubble point gave the most consistent results, with no obvious difference in the deviation from the best correlations at either high or low deltas, as would be expected if the use of the bubble point did not hold throughout the range studied. This, of course, does not repudiate the theory of Colburn and Drew but does indicate that, for the materials studied in this work, a wider range of temperature differences must be investigated to verify it.

Kent and Pigford (*4*) extended the Colburn-Drew work in considering the fractionation effected in condensation. Specifically, they postulated that the interfacial concentration depended on the relative values of the gas- and liquid-phase mass transfer coefficients. The calculations made in the present work assumed that there was no difference in the composition of the condensate from the vapor-liquid interface to the wall. This may be taken as evidence that the gas film controlled in this system. Were there any significant change in composition across the liquid film, the bubble point at the vapor-liquid interface would be different from that based on the average condensate composition; the temperature difference would vary accordingly and would show in the correlations made, particularly at low values of the temperature difference.

In considering these observations, one must keep in mind that the research was carried out at almost total condensation. Since the primary interest was in the liquid film coefficient, the composition of the residual vapor was not ascertained as a routine; however, in the few cases where it was determined, it was consistently richer in the more volatile component than was the condensate obtained at the same time. This enrichment appeared to resemble that predicted by the equilibrium diagram for the condensate composition, but additional study is needed to verify this and ascertain the limits under which it holds true.

ACKNOWLEDGMENT

The authors wish to acknowledge use of data obtained by Jacob J. Trachtenberg (*17*), James A. Malone (*5*), and Habib Labbauf as part of their respective thesis researches while graduate students at Louisiana State University. The latter two were holders of the Dow Research Fellowship in Chemical Engineering during the period of their research. Recognition is due also to Jesse Coates, who directed the research of Dr. Todd (*16*) and Mr. Trachtenberg.

NOTATION

A = heat transfer area, πDL, sq. ft.
D = outside diameter of condensing surface, ft.
g = acceleration of gravity, ft./hr.^{2}
h_m = average coefficient of heat transfer, B.t.u./(hr.)(sq. ft. °F.)
k = thermal conductivity of condensate, B.t.u./(hr.)(sq. ft.)(°F./ft.)
L = length of condensing surface, ft.
q = rate of heat transfer,—B.t.u./hr.
t = temperature, °F.
t_{BP} = bubble point of mixture, as determined by temperature-composition diagram
t_{DP} = dew point of mixture, as determined by temperature-composition diagram
t_f = temperature of condensate film
t_s = temperature of condensing surface as measured experimentally
t_{sv} = temperature of saturated vapor
t_v = temperature of vapor phase as measured experimentally
W = rate of condensate formation, lb./hr.
Δt = temperature difference, °F.

$$\Delta t_{BP} = t_{BP} - t_s$$

$$\Delta t_{DP} = t_{DP} - t_s$$

$$\Delta t_v = t_v - t_s$$

Γ' = rate of condensate drainage, W/L, lb./(hr.)(ft.), for horizontal surfaces
λ = latent heat of vaporization, B.t.u./lb.
μ = viscosity of condensate, lb.(ft.)(hr.)
μ_f = viscosity of condensate, lb./(ft.)(hr.) at t_f
ρ = density of condensate, lb./cu. ft.
ϕ = physical property group, $(k^3\rho^2 g/\mu^2)^{1/3}$, B.t.u./(hr.)(sq. ft./°F.)

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