

EVAPORATION OF BINARY MIXTURES IN UPWARD ANNULAR FLOW

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(Received 24 July 1974)

Abstract—A theoretical study is presented of the evaporation of binary mixtures in upward annular flow. Heat and mass balances are written and the resulting equations solved to give axial and radial variations of concentrations, temperatures and flowrates of ethanol–water mixtures. Mass and heat transfer within the film are calculated by an extension of the Dukler–Hewitt method for heat transfer in single-component films. It is concluded from the worked examples that, for the mixture considered, the film flowrates and wall temperatures are not significantly controlled by mass transfer in either phase and can be calculated by flash vaporization methods.

1. INTRODUCTION

The presence of a second component in a fluid undergoing a change of phase adds to the number of phenomena which govern that change. Nucleate and film pool boiling of binary mixtures have been studied (Van Stralen 1970; Calus & Rice 1972; Stephan & Körner 1969; Van Stralen *et al.* 1972), so too has condensation of binary mixtures (Colburn & Drew 1937; Van Es & Heertjes 1962). This paper presents a theoretical solution to the problem of evaporation in the annular flow regime. Such a flow pattern occurs towards the outlet of many vertical tube boilers. A modified version of this analysis would apply to condensation in annular flow.

The basic problem examined here is that of axial and radial gradients of concentration, and hence of saturation temperature, due to the greater volatility of the light boiling component and to the resistance to the diffusion of matter within the phases. This resistance causes the concentration distribution to be different from that for an equilibrium flash vaporization. In the following sections of this paper, heat- and mass-balance equations are presented and a series of solutions conducted to determine the controlling resistances in a variety of typical cases. The results are compared with those from equilibrium flash calculations.

Solution of the basic equations requires heat- and mass-transfer coefficients. A new method for calculating mass-transfer coefficients in climbing liquid films, based on the Dukler–Hewitt (Hewitt 1961) approach for heat and momentum transfer, is described in section 2. The method could be adapted for falling films. Heat-transfer coefficients in films can be affected by diffusion of matter within them; section 3 describes an extension of the basic Dukler–Hewitt heat-transfer solution to such cases.

Two basic assumptions made in this work are as follows: (1) The presence of liquid droplets in the vapour core and of vapour bubbles in the film can be ignored. (2) The liquid film has a smooth surface and the effect of waves can be neglected.

2. HEAT AND MASS BALANCES

The most far reaching and limiting of the assumptions to be made have been introduced in section 1. Several others are made to simplify the solution of the equations, these are as follows: (a) The two components are miscible in all proportions. (b) The integral specific latent heat (that needed to evaporate completely unit mass of liquid) is linearly related to the concentration. (c) Heat gained or lost by the film due to the axial change in saturation temperature is negligible compared with the latent heat. (d) Equilibrium prevails at the interface. (e) Steady state conditions prevail. (f) Circumferential symmetry exists. (g) In the liquid film axial changes of u_z , the velocity in the axial direction, can be ignored. (h) Radial and axial gradients of liquid density

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can be ignored. (i) The concentration profile in the film is fully developed. (j) The rates of axial diffusion and conduction are negligible.

Consider an elemental length of a circular channel carrying liquid and vapour, each component being present in each phase. A mass balance over the element gives

$$dW_L = -dW_G \quad [1]$$

where W_L and W_G are the mass flowrates of liquid and vapour, respectively. A material balance on the light component gives

$$d(W_L x'_b) = -d(W_G y'_b) \quad [2]$$

where x' and y' are mass fractions of the light boiling component in the liquid and vapour phases respectively. Subscript b indicates the bulk, or mixing-cup, value.

Each side of [2] represents the rate of change of the component flow over the element and can be written as

$$-d(W_L x'_b) = 2\pi R j_{r,L,i} dz - \frac{dW_L}{dz} x'_b dz, \quad [3a]$$

$$d(W_G y'_b) = 2\pi R j_{r,G,i} dz + \frac{dW_G}{dz} y'_b dz, \quad [3b]$$

in which R is radius, z is distance in the axial direction, $j_{r,L}$ and $J_{r,G}$ are radial diffusive mass fluxes in liquid and vapour respectively relative to the mass average radial fluxes, subscript i indicates interface values.

Mass-transfer coefficients k'_L and k'_G in the liquid and vapour phases respectively are defined by

$$j_{r,L,i} = k'_L (x'_b - x'_i), \quad [4a]$$

$$J_{r,G,i} = k'_G (y'_i - y'_b). \quad [4b]$$

The stars on the coefficients indicate that they may differ from those, k_L and k_G which would be found if the mass average radial fluxes were zero. The differences are due to the influence of the net mass fluxes on the concentration profiles. From [1] to [4]:

$$\frac{dx'_b}{dz} = \left(\frac{dW_L}{dz} + 2\pi R_i k'_L \right) (x'_i - x'_b) \frac{1}{W_L}. \quad [5a]$$

$$\frac{dy'_b}{dz} = \left(\frac{dW_L}{dz} - 2\pi R_i k'_G \right) (y'_b - y'_i) \frac{1}{W_G}. \quad [5b]$$

The loss of light component from the film in the length dz is given by [3a]. Since in a binary mixture $j_A = -j_B$ where A and B refer to light and heavy components respectively, we can write for the loss of heavy component

$$-d(W_L \{1 - x'_b\}) = -2\pi R_i dz k'_L (x'_b - x'_i) - (1 - x'_i) \frac{dW_L}{dz} dz. \quad [6]$$

From assumption (b) we obtain that the supply of latent heat in the length dz , Q_{LG} , is given by

$$Q_{LG} = \left[-x'_i \frac{dW_L}{dz} dz + 2\pi R_i dz k'_L (x'_b - x'_i) \right] h_{LG,A} + \left[-(1 - x'_i) \frac{dW_L}{dz} dz - 2\pi R_i dz k'_L (x'_b - x'_i) \right] h_{LG,B}, \quad [7]$$

where h_{LG} is specific latent heat. The flux of this heat flow through the interface is

$$q_{LG} = \frac{Q_{LG}}{2\pi R_i dz} \quad [8]$$

The heat reaching the interface, equal to that passing through the wall, assumption (c), provides the latent heat for the evaporating stream plus the heat removed from the interface by conduction and convection. Hence

$$q_w A_w = q_{LG} A_i + q_c A_i, \quad [9]$$

where q_w is the heat flux through the wall which is of area A_w , A_i is the interfacial area and q_c , the heat removed by conduction and convection:

$$q_c = h \xi (T_i - T_{G,b}), \quad [10]$$

$h \xi$ is the heat-transfer coefficient in the vapour and the * indicates that it may differ from that in the zero mass-transfer case, T_i is the interfacial temperature and $T_{v,b}$ is the bulk vapour temperature.

Combining [7] to [10] and rearranging:

$$\frac{dW_L}{dz} = \frac{-2\pi R_w q_w + 2\pi R_i k_L' (x'_b - x'_i)(h_{LG,A} - h_{LG,B}) + 2\pi R_i h \xi (T_i - T_{G,b})}{x'_i h_{LG,A} + (1 - x'_i) h_{LG,B}}, \quad [11]$$

where R_w is the tube radius.

For a pure coolant [11] reduces to the simple form

$$\frac{dW_L}{dz} = \frac{-2\pi R_w q_w}{h_{LG}}, \quad [12]$$

in which, following the arguments of Lacey (1964), we take $T_i = T_{G,b}$. In common with this simple form the equation for mixtures, [11], has as its numerator the heat available for evaporation and as its denominator the effective specific latent heat.

Not all the heat entering through the wall is available for net evaporation of the two-component film. This is due to (i) the equal and opposite diffusion of the two components through the interface which while not contributing to the net evaporation rate may yet require a net heat flow (which may be in either direction) if $h_{LG,A} \neq h_{LG,B}$; this is reflected in the second term in the numerator of [11] and (ii) the removal of heat from the interface by conduction and convection, reflected in the final term of the numerator of [11].

Assumption (b) explains the replacement of h_{LG} in [12] by the linear relationship in the denominator of [11].

An overall and a component mass balance give respectively

$$W_T = W_L + W_G, \quad [13]$$

and

$$W_T x'_F = W_L x'_b + W_G y'_b, \quad [14]$$

where W_T is the total mass flowrate and x'_F is the mass fraction of light component in the entering stream averaged over both phases.

The interfacial equilibrium, assumption (d), is expressed by

$$y'_i = K' x'_i, \quad [15]$$

where K' the equilibrium ratio, is a function of temperature, composition and pressure.

Differentiation of [14] and insertion of [5a], [5b] and [11] gives

$$x'_i = \frac{2\pi R_i(k'_L x'_b + k'_G y'_b)}{\frac{dW_L}{dz}(1 - K') + 2\pi R_i(k'_L + K'k'_G)} \quad [16]$$

In order to solve these equations suitable expressions must be found for the various transfer coefficients.

3. LIQUID-PHASE MASS-TRANSFER COEFFICIENTS

Many studies have been performed on liquid resistances in interphase mass transfer, mostly in stirred tanks or falling liquid films (Whitman 1923; Danckwerts 1951; Jepsen *et al.* 1966; King 1966; Ciborowski & Rychlicki 1971). A number of these studies are concerned with the penetration theory of Danckwerts (1951), which cannot be used for the prediction of resistances unless the surface renewal factor is known as a function of the hydrodynamic parameters of the system. The necessary data are not yet available for the present problem.

The method of Dukler (1960) for momentum transfer in downward annular flow and its extension by Hewitt (1961) for heat transfer in upward annular flow have been examined by Collier & Pulling (1962) who found reasonable agreement with experimental results. The method derived here is an extension of the Dukler-Hewitt model to encompass the case of mass transfer. The different boundary conditions at the wall preclude the derivation of a solution by analogy.

Noting assumptions (g) and (h) we can assume that the net radial velocity in the liquid is zero, i.e.

$$n_A + n_B = 0 \quad [17]$$

where $n_A + n_B$ are the radial mass average fluxes in the film of the two components relative to stationary axes. Hence

$$k'_L = k'_L \quad [18]$$

Assumption (i) indicates that the mass-transfer coefficient in the liquid film is independent of axial distance. The validity of this assumption is examined in appendix B in which development lengths are calculated for the boundary conditions of constant interface concentration and of constant interface mass flux. A constant-diffusion coefficient is assumed which renders the solutions strictly valid only for the case of laminar flow. For the two boundary conditions dimensionless development lengths given by

$$z_{dev} = \frac{D_L z_{dev}}{\bar{u} a_i^2} \quad [19]$$

are calculated to be 0.182 and 0.245 respectively. D_L is the diffusion coefficient, z the downstream distance, \bar{u} the mean velocity and a_i the film thickness.

Substituting into [19] to calculate z_{dev} (the true development length) using the laminar-diffusion coefficient and \bar{u} and a_i values typical of those examined later in the paper gives excessively large development lengths. However, making a conservative assumption and introducing a more probable value of the overall (turbulent plus laminar) diffusion coefficient gives predicted development lengths of 1.8 and 2.4 cm for the constant x_i and constant j_i cases. The corresponding mass transfer Graetz numbers, Gz_m , are 2300 and 1750 where

$$Gz_m = \frac{W_L}{D_L z} \quad [20]$$

The development lengths are small compared with the tube length and hence the mass-transfer coefficient can truly be considered to be independent of length.

In the following analysis of the mass-transfer coefficient the assumption of a constant interfacial mass flux has been made. Although the true condition will lie somewhere between the two extreme boundary conditions this is permissible in view of the similarity of the asymptotic values of the coefficients predicted from the two boundary conditions. Note also that the conclusions reached in section 7 of this paper show the resistance to mass transfer in the film to be negligible.

With assumptions (i) and (j) a mass balance on the light component taken over the element shown in figure 1 gives

$$\rho_L \frac{\partial x'}{\partial z} \int_{R_w}^r r u_z dr + r j_{r,L} = 0, \tag{21}$$

where ρ_L is the liquid density, and R_w is the tube radius.

For $r = R_i$ (the interfacial radius):

$$\rho_L \frac{\partial x'}{\partial z} \int_{R_w}^{R_i} r u_z dr + R j_{r,L,i} = 0. \tag{22}$$

Fick's Law is extended to turbulent systems to give

$$j_{r,L} = \rho_L (D_L + \epsilon_D) \frac{\partial x'}{\partial r}, \tag{23}$$

where D_L and ϵ_D are the molecular and eddy diffusivities, respectively.

Defining a dimensionless velocity, radial position and concentration

$$u^+ = u_z / u^*, \quad r^+ = \frac{\rho_L u^* r}{\mu_L}, \quad C^+ = \rho_L \frac{u^* (x' - x'_i)}{j_{r,L,i}}, \tag{24}$$

where u^* is the friction velocity and μ_L the liquid viscosity, we obtain from [21] to [24]

$$\frac{R_i^+ \int_{R_w^+}^{r^+} r^+ u^+ dr^+}{r^+ \int_{R_w^+}^{R_i^+} r^+ u^+ dr^+} = \frac{\rho_L}{\mu_L} (D_L + \epsilon_D) \frac{dC^+}{dr^+}. \tag{25}$$

The distance from the wall $a = R_w - r$ is in dimensionless form,

$$a^+ = \frac{a \rho_L u^*}{\mu_L} = Re^* - r^+. \tag{26}$$

where $Re^*(= R_w \rho_L u^* / \mu_L)$ is a curvature Reynolds number (Hewitt 1961). On substitution of [26]

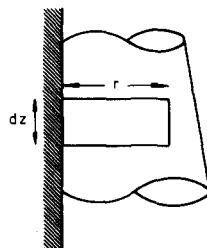


Figure 1. Element of film.

we obtain from [25]

$$\frac{1 - \eta/Re^* \int_0^{a^+} (1 - a^+/Re^*)u^+ da^+}{1 - a^+/Re^* \int_0^{\eta} (1 - a^+/Re^*)u^+ da^+} = - \left(\frac{1}{Sc_L} + \frac{\epsilon_D \rho_L}{\mu_L} \right) \frac{dC^+}{da^+}, \quad [27]$$

in which Sc_L is the liquid phase Schmidt number and $\eta (= a_i \rho_L u^* / \mu_L)$ is the dimensionless film thickness. As $\eta/Re^* \rightarrow 0$, i.e. for thin films in large tubes, [27] gives

$$\frac{\int_0^{a^+} u^+ da^+}{W_L^+} = - \left(\frac{1}{Sc_L} + \frac{\epsilon_D \rho_L}{\mu_L} \right) \frac{dC^+}{da^+}, \quad [28]$$

where $W_L^+ = \int_0^{\eta} u^+ da^+ = W_L / 2\pi R_w \mu_L$ and is the dimensionless film flowrate. Calculation continues using [28] since in most practical cases the "flat plate" approximation is permissible.

Data on eddy diffusivity are sparse. Kalinske & Pien (1944) concluded from open channel experiments that $\epsilon_D \approx \epsilon_M$, the eddy viscosity. It is assumed here that the turbulence structure in the film is related to a and τ , the shear stress, as for purely single-phase flow. For $a^+ < 20$ we use the expression of Deissler (1955). Putting this in dimensionless form and substituting in [28] gives

$$\frac{\int_0^{a^+} u^+ da^+}{W_L} = - \left[\frac{1}{Sc_L} + n^2 u^+ a^+ (1 - e^{-n^2 u^+ a^+}) \right] \frac{dC^+}{da^+}, \quad [29]$$

where $n \approx 0.1$. Hewitt (1961) has derived, from a force balance,

$$\frac{\tau}{\tau_w} = \frac{\epsilon_M \rho_L}{\mu_L} \frac{du^+}{da^+} = 1 + a^+ \sigma^3 / \eta, \quad [30]$$

in which $\sigma^3 / \eta = a_i / (\eta^2 \mu_L^2 / \rho_L g)^{1/3}$ is a parameter characterising the gradient of shear stress through the film. τ_w is the wall shear stress. Hence in $a^+ < 20$ the dimensionless velocity profile can be found by substituting Deissler's expression for ϵ_M into [30] and solving the resultant equation.

In the region $a^+ > 20$ we ignore molecular diffusion since $1/Sc_L \ll \epsilon_D \rho_L / \mu_L$. Hewitt (1961) has solved [30] using a dimensionless form of the equation of von Karman (1930),

$$\frac{\tau}{\tau_w} = \frac{X^2 (du^+ / da^+)^4}{(d^2 u^+ / da^{+2})^2}, \quad [31]$$

where $X = 0.36$, to give the dimensionless velocity gradient

$$\frac{du^+}{da^+} = \frac{1}{X} \frac{\sigma^3 / \eta}{2[(1 + a^+ \sigma^3 / \eta)^{-1/2} - 1]}. \quad [32]$$

Substituting [30] and [31] into [28] and neglecting $1/Sc_L$ gives

$$\frac{dC^+}{da^+} = - \frac{1}{2X} \frac{\sigma^3 / \eta}{(1 + a^+ \sigma^3 / \eta)[(1 + a^+ \sigma^3 / \eta)^{1/2} - 1]} \frac{\int_0^{a^+} u^+ da^+}{W_L^+}. \quad [33]$$

For most practical cases τ is high and $\sigma^3/\eta \rightarrow 0$, in which case, [32] can be simplified to

$$u^+ - u_1^+ = \frac{1}{X} \ln(a^+/a_1^+), \quad [34]$$

where u_1^+ is the dimensionless velocity at $a^+ = a_1^+ = 20$ calculated from [30] with Deissler's expression for ϵ_M . Substituting [34] into [28] gives

$$\frac{dC^+}{da^+} = \frac{1}{Xa^+} \frac{\int_0^{a^+} \left[u_1^+ + \frac{1}{X} \ln(a^+/a_1^+) \right] da^+}{W_L^+}. \quad [35]$$

Equations [29] and [35] have been solved numerically (Shock 1973) to give the dimensionless concentration profile. The boundary conditions are

$$(i) \text{ at } a^+ = 0 \quad u^+ = 0$$

$$(ii) \text{ at } a^+ = \eta \quad C^+ = 0$$

The mass-transfer coefficient is evaluated as follows. The bulk concentration in the liquid film is defined by

$$x'_b = \frac{\int_0^{a_1} x' u da}{\int_0^{a_1} u da}. \quad [36]$$

Substitution of [20], [24] and [36] into [4a] gives

$$\frac{\rho_L u^*}{k'_L} = \frac{\int_0^\eta C^+ u^+ da^+}{\int_0^\eta u^+ da^+}. \quad [37]$$

The R. H. S. of [37] is the bulk value of C^+ , i.e. C_b^+ and is evaluated from the profiles of C^+ and y^+ (Shock 1973).

The Sherwood number, dimensionless mass-transfer coefficient, is defined by

$$Sh_L = \frac{k'_L a_i}{\rho_L D_L}, \quad [38]$$

from which it can be shown that

$$Sh_L = \frac{\eta Sc_L}{C_b^+}. \quad [39]$$

Numerical calculations were performed to give Sh_L at various values of liquid film Reynolds number, $Re_L (= 4W_L^+)$, and Sc_L for two values of σ^3/η . Results are shown in table 1. As $Re_L \rightarrow 0$, Sh_L is asymptotic to 5. An analytical solution for a laminar film (appendix A) confirms that numerically calculated value.

Table I. Values of Sh_L for various values of Re_L , Sc_L and σ^3/η

$\frac{Sc_L}{Re_L}$	$\sigma^3/\eta = 0$					$\frac{Sc_L}{Re_L}$	$\sigma^3/\eta = 0.5$				
	125	250	500	1000	2000		125	250	500	1000	2000
10.12	5.740	6.515	7.969	10.64	15.49	10.24	6.392	7.405	9.279	12.68	18.79
21.12	8.186	11.02	16.18	25.54	42.71	20.56	9.626	13.20	19.62	31.17	52.13
31.16	11.39	16.86	26.81	45.09	79.03	30.68	14.20	21.36	34.27	57.74	100.9
41.32	15.42	24.20	40.30	70.15	126.1	40.56	19.77	31.37	52.45	91.15	163.1
50.80	19.76	32.18	55.11	97.96	178.9	52.16	27.55	45.50	78.39	139.3	253.5
100.7	50.66	90.09	164.8	307.6	582.8	102.2	73.84	131.5	239.9	445.2	837.5
152.1	93.34	171.8	322.5	614.0	1181	153.7	138.2	253.9	474.4	897.1	1713
200.7	141.2	264.8	503.8	969.2	1879	202.7	210.6	393.6	745.1	1424	2743
250.3	195.6	371.2	712.8	1381	2693	250.9	289.1	546.7	1044	2011	3896
502.3	514.1	1003	1967	3875	7657	506.5	760.1	1479	2891	5673	11,170
759.0	977.8	1923	3797	7519	14,920	760.2	1237	2433	4799	9486	18,790
1000	1667	3290	6512	12,920	25,860	1001	1669	3299	6535	12,970	25,760
1501	3065	6078	12,080	24,030	47,850	1507	2516	4998	9943	19,800	39,450
2017	4371	8693	17,310	34,510	68,840	2073	4387	8729	17,390	34,650	69,100
2519	5486	10,930	21,800	43,500	86,840	2547	6709	13,360	26,610	53,060	105,800
5032	9978	19,940	39,840	79,640	159,200	5038	22,510	44,920	89,660	179,000	357,600
7506	13,760	27,500	54,990	109,900	219,900	7522	40,320	80,530	160,900	321,500	642,600
10,010	17,350	34,680	69,360	138,700	277,400	10,030	59,200	118,300	236,400	472,800	945,300
15,010	24,140	48,270	96,530	193,100	386,100	15,060	99,150	198,200	396,200	792,200	1,584,000
20,010	30,650	61,290	122,600	245,100	490,300	20,070	141,700	283,300	566,500	1,133,000	2,265,000
25,040	37,010	74,010	148,000	296,000	592,000	25,090	186,900	373,600	747,100	1,494,000	2,988,000
50,030	67,290	134,600	269,200	538,300	1,077,000	50,070	439,000	877,900	1,756,000	3,511,000	7,022,000
75,040	96,520	193,000	386,100	772,200	1,544,000	75,120	724,600	1,449,000	2,898,000	5,796,000	11,590,000
100,000	125,300	250,500	501,100	1,002,000	2,004,000	100,100	1,032,000	2,065,000	4,130,000	8,260,000	16,520,000

4. LIQUID PHASE HEAT-TRANSFER COEFFICIENTS

The wall temperature of a tube carrying an evaporating film of a binary mixture may differ from that of one carrying a pure fluid of the same physical properties under the same heat flux and hydrodynamic conditions. One reason for this difference lies in the interfacial condition being different from that in the equilibrium flash vaporization, zero mass-transfer resistance, case. The second reason lies in the different temperature drop through the film due to thermal energy transport by mass transfer. An energy balance for a turbulent film on a surface of large radius and where assumptions (c) and (j) and [19] hold gives

$$q = q_w = (\kappa_L + \epsilon_L \rho_L c_L) \frac{dT}{da} + \frac{\bar{H}_A j_{L,A}}{M_A} + \frac{\bar{H}_B j_{L,B}}{M_B}, \tag{40}$$

where q is the heat flux, κ_L is thermal conductivity of the liquid and c_L is its specific heat capacity. ϵ_H is the eddy thermal diffusivity, T is temperature, \bar{H} and M are respectively partial molar enthalpy and molar mass. j_L is mass flux in the liquid normal to the wall. We note that $j_{L,A} = -j_{L,B}$ and approximate

$$\frac{\bar{H}}{M} = c_L (T - T_{ref,\bar{H}}), \tag{41}$$

where $T_{ref,\bar{H}}$ is the reference temperature datum for enthalpy. Hence

$$q_w = -(\kappa_L + \epsilon_H \rho_L c_L) \frac{dT}{da} + (c_{L,A} - c_{L,B}) j_{L,A} (T - T_{ref,\bar{H}}). \tag{42}$$

With the flat plate approximation, already made in [40], [21] becomes

$$j_{L,A} = -\rho_L \frac{\partial x'}{\partial z} \int_0^a u_z da. \tag{43}$$

Substituting [26] and [43] into [41] and choosing $T_{ref,\bar{H}}$, which can take any value, to be T_w we

obtain

$$\left(\frac{1}{Pr_L} + \frac{\epsilon_H \rho_L}{\mu_L}\right) \frac{dT^+}{da^+} - 1 + QT^+ \int_0^{a^+} u^+ da^+ = 0, \quad [44]$$

where

$$T^+ = \frac{C_{L,w} \rho_{L,w}}{q_w} u^* (T_w - T), \quad [45]$$

and

$$Q = \frac{C_{L,A} - C_{L,B}}{C_{L,m}} \frac{\mu_L}{\rho_L u^*} \frac{\partial x'}{\partial z}, \quad [46]$$

where $c_{L,M}$ is the mixture specific heat capacity. Q is a dimensionless constant which characterises the effects of interdiffusion of the two components on heat transfer; it can be positive or negative. An improvement in heat transfer will occur with positive Q .

Equation [44] was solved numerically to give the Nusselt number

$$Nu_L = \frac{q_w a_i}{(T_w - T_i) \eta_L} = \frac{\eta Pr_L}{T_i^+}, \quad [47]$$

for various values of Re_L , σ^3/η , Pr_L and Q . Table 2 shows some of the results. It can be seen that for low σ^3/η , if Q is sufficiently large and negative, Nu_L can fall with increasing Re_L . This is however somewhat academic since it is unlikely that in practice Q would be greater than 10^{-6} .

Since T_w , the reference temperature in [41], is not constant along the tube, the calculation of length mean averages, from local values given by the above methods, is complex.

5. VAPOUR-PHASE MASS-TRANSFER COEFFICIENTS

The Chilton–Colburn analogy is used for vapour-phase mass-transfer and gives

$$j_D = \frac{k_G}{\rho_G u_{G,b}} Sc_G^{0.67} = \frac{f_G}{2}, \quad [48]$$

where j_D is the j -factor for mass transfer, k_G the mass-transfer coefficient based on molar flux and mole fraction driving force, ρ_G and Sc_G are the vapour density and Schmidt numbers respectively, $u_{G,b}$ is the bulk average axial vapour velocity relative to the interface and f_G is the friction factor given by

$$f_G = 0.079 Re_G^{-0.25}, \quad [49]$$

where Re_G is evaluated using $u_{G,b}$, and a diameter equal to that of the vapour core.

Now k_G is the limiting molar diffusion rate as the net molar flux tends to zero. It can be shown (Shock 1973) that k'_G , the limiting mass-diffusion rate as the net mass flux tends to zero, is

$$k'_G = \frac{k_G M_A M_B}{y'_b M_B + (1 - y'_b) M_A}. \quad [50]$$

Bird *et al.* (1960) show the derivation of an equation for $\theta = k'_G/k_G$ based on a film theory model. Their equations can be reworked with concentrations on a mass basis to give

$$\theta = \frac{k'_G}{k_G} = \frac{\phi}{e^\phi - 1}, \quad [51]$$

Table 2. Values of Nu_L for various values of Re_L , Pr_L , Q and σ^3/η

Q Re_L	$\sigma^3/\eta = 0$									
	$Pr_L = 1$			$\sigma^3/\eta = 0$				$Pr_L = 5$		
	10^{-4}	10^{-6}	0	-10^{-6}	-10^{-4}	10^{-4}	10^{-6}	0	-10^{-6}	-10^{-4}
10.12	1.000	1.000	1.000	1.000	1.000	1.003	1.003	1.002	1.002	1.002
21.12	1.003	1.001	1.002	1.002	1.002	1.013	1.011	1.011	1.011	1.008
31.16	1.005	1.005	1.005	1.005	1.004	1.026	1.022	1.022	1.022	1.018
41.32	1.009	1.008	1.008	1.008	1.007	1.043	1.038	1.038	1.038	1.032
50.80	1.013	1.012	1.012	1.012	1.010	1.062	1.054	1.054	1.054	1.046
100.7	1.044	1.040	1.040	1.040	1.035	1.185	1.165	1.165	1.164	1.145
152.1	1.086	1.078	1.078	1.078	1.070	1.325	1.291	1.291	1.290	1.257
200.7	1.131	1.119	1.119	1.119	1.107	1.457	1.409	1.409	1.408	1.362
250.3	1.179	1.162	1.162	1.162	1.146	1.589	1.526	1.525	1.525	1.464
502.3	1.428	1.383	1.383	1.383	1.339	2.206	2.058	2.056	2.055	1.916
759.0	1.701	1.625	1.624	1.624	1.551	2.793	2.556	2.554	2.552	2.334
1000	1.993	1.887	1.886	1.884	1.783	3.359	3.044	3.041	3.037	2.750
1501	2.549	2.364	2.362	2.360	2.187	4.457	3.951	3.947	3.942	3.491
2017	3.115	2.824	2.822	2.819	2.553	5.601	4.847	4.840	4.833	4.176
2519	3.663	3.247	3.243	3.239	2.867	6.728	5.681	5.671	5.662	4.772
5032	6.522	5.131	5.119	5.106	4.000	12.81	9.515	9.486	9.457	7.001
7506	9.722	6.778	6.753	6.728	4.649	19.89	12.97	12.92	12.86	8.336
10,010	13.51	8.328	8.287	8.245	5.003	28.53	16.29	16.20	16.10	9.103
15,010	23.25	11.20	11.12	11.03	5.138	51.60	22.56	22.37	22.18	9.502
20,010	36.77	13.89	13.75	13.61	4.841	85.17	28.53	28.21	27.89	9.030
25,040	55.40	16.46	16.26	16.05	4.343	133.5	34.33	33.85	33.38	8.139
50,030	280.8	28.29	27.56	26.85	1.822	827.5	61.57	59.88	58.22	3.440
75,040	893.2	39.29	37.79	36.33	0.6119	3266	87.52	83.97	80.50	1.156
100,000	1983	49.91	47.39	44.97	0.1874	8591	112.9	106.9	101.1	0.3541
	$\sigma^3/\eta = 0.5$									
10.24	1.000	1.000	1.000	1.000	1.000	1.004	1.003	1.003	1.003	1.002
20.56	1.003	1.002	1.002	1.002	1.002	1.013	1.012	1.012	1.012	1.010
30.68	1.006	1.005	1.005	1.005	1.005	1.028	1.025	1.025	1.025	1.022
40.56	1.010	1.009	1.009	1.009	1.008	1.046	1.042	1.042	1.042	1.038
52.16	1.016	1.014	1.014	1.014	1.013	1.071	1.065	1.065	1.065	1.060
102.2	1.051	1.048	1.048	1.048	1.045	1.197	1.184	1.184	1.184	1.171
153.7	1.096	1.091	1.091	1.091	1.086	1.330	1.310	1.310	1.310	1.290
202.7	1.142	1.135	1.135	1.135	1.127	1.449	1.422	1.422	1.422	1.395
250.9	1.188	1.178	1.178	1.178	1.169	1.559	1.525	1.525	1.525	1.492
506.5	1.419	1.396	1.396	1.396	1.373	2.061	1.991	1.990	1.989	1.921
760.2	1.625	1.586	1.586	1.585	1.547	2.481	2.371	2.370	2.369	2.263
1001	1.802	1.746	1.746	1.745	1.691	2.840	2.689	2.688	2.686	2.543
1507	2.141	2.045	2.044	2.043	1.951	3.521	3.276	3.274	3.271	3.043
2073	2.525	2.393	2.393	2.391	2.266	4.219	3.891	3.888	3.885	3.582
2547	2.852	2.696	2.694	2.693	2.544	4.787	4.401	4.397	4.393	4.038
5038	4.513	4.197	4.194	4.191	3.896	7.682	6.945	6.938	6.931	6.263
7522	6.100	5.591	5.587	5.582	5.114	10.46	9.316	9.305	9.294	8.274
10,030	7.663	6.932	6.925	6.918	6.254	13.20	11.60	11.58	11.57	10.16
15,060	10.70	9.457	9.445	9.433	8.330	18.55	15.91	15.89	15.86	13.60
20,070	13.66	11.83	11.81	11.79	10.20	23.77	19.96	19.93	19.89	16.70
25,090	16.59	14.10	14.07	14.05	11.93	28.94	23.85	23.80	23.76	19.57
50,070	30.89	24.34	24.28	24.22	19.06	54.25	41.42	41.31	41.19	31.42
75,120	45.21	33.53	33.43	33.32	24.68	79.66	57.21	57.03	56.83	40.78
100,100	59.69	42.03	41.87	41.73	29.34	105.4	71.84	71.57	71.29	48.52

where

$$\phi = \frac{1}{2\pi R_i k'_G} \frac{dW_L}{dZ}, \tag{52}$$

hence k'_G* can be evaluated from [48] to [52].

6. VAPOUR-PHASE HEAT-TRANSFER COEFFICIENTS

Again we use the Chilton–Colburn analogy

$$j_H = \frac{h_G}{\rho v C v u_{G,b}} Pr_G^{0.67} = \frac{f_G}{2}, \tag{53}$$

where j_H is the j -factor for heat transfer, h_G is the heat-transfer coefficient in the absence of mass transfer, c_G and Pr_G are the vapour specific heat capacity and Prandtl number respectively. To calculate h_G^* we again use the method derived by Bird *et al.* (1960) which gives

$$\theta = \frac{h_G^*}{h_G} = \frac{\phi}{e^\phi - 1}, \quad [54]$$

where

$$\phi = \frac{n_{A,G,i}c_{G,A} + n_{B,G,i}c_{G,B}}{h_G}, \quad [55]$$

and $n_{A,G,i}$ the total mass flux of component A in the vapour at the interface, is

$$n_{A,G,i} = (n_{A,G,i} + n_{B,G,i})z'_e = G_e z'_e, \quad [56]$$

z'_e is the mass fraction of light component in the evaporating stream, i.e. the evaporation rate of that component divided by the total evaporation rate. G_e the total evaporation rate, is given by $-(1/2\pi R_i)(dW_L/dz)$.

We can write

$$q_{L,G} = G_e z'_e h_{L,G,A} + G_e (1 - z'_e) h_{L,G,B}, \quad [57]$$

hence from [7]

$$G_e z'_e = -x'_i \frac{dW_L}{dz} \frac{1}{2\pi R_i} + k'_L (x'_b - x'_i), \quad [58]$$

$$z'_e = x'_i + \frac{k'_L (x'_b - x'_i)}{G_e}. \quad [59]$$

This equation is solved for z'_e and hence $n_{A,G,i}$ and $n_{B,G,i}$ can be found from [56] and we now have all the terms necessary to evaluate h_G^* .

It can be noted from [58] and [59] that at high G_e , i.e. at high evaporation rates, $x'_b = x'_i$, and no rectification occurs $[(dx'_b/dz) = 0]$. As G_e decreases z'_e becomes greater than x'_i and rectification occurs since $x'_b > x'_i$. Each phase is assumed to be of uniform concentration at the channel inlet. Since at zero G_e no mass transfer occurs and $x'_b = x'_i$ (see [58]) then $x'_b - x'_i$ must go through a maximum as G_e decreases from infinity to zero.

For evaluation of the vapour-phase heat-transfer in [11] we also need T_i and $T_{G,b}$. Since equilibrium is being assumed at the interface T_i can be calculated from the pressure and local concentration. It can be shown (Shock 1973) that, if the dew point line is straight between y_b (bulk vapour mole fraction of light component) at inlet and x_F (mole fraction averaged over entering fluid stream) and furthermore that the Lewis number (Pr_G/Sc_G) is unity, the bulk vapour stream will remain at saturation temperature or tend towards it, depending on the state of the stream entering the channel. If these conditions are fulfilled $T_{G,b}$ is the dew point corresponding to y_b . It is intended in further work to examine the consequences if these conditions are not fulfilled as well as the consequences of heat absorption or release by the liquid film.

7. CASE STUDIES

This section describes the calculation of conditions in a tube and determination of the controlling resistances in a number of cases using the methods described in the previous sections. A computer routine was written (Shock 1973) to solve the equations and also to calculate the conditions expected from an equilibrium flash vaporization. Comparison of the results shows in which, if either, phase there is a resistance to mass or heat transfer which controls the important variables and causes them to differ significantly from those in the equilibrium flash, zero resistance, case. These important variables were taken to be the flowrates of the two phases and

the temperature at the wall; this is convenient for the constant heat flux case examined here. The mixture studied was ethanol-water.

Solution of the equations is carried out in steps of length 0.01 m, the tube radii used were 0.01 and 0.0165 m. In most of the calculations the pressure was assumed not to vary along the channel, hence confining the problem to heat flux induced gradients and ignoring flashing. Furthermore this minimises the error due to assumption (c).

We can decide whether a particular resistance controls an important design parameter by examining whether its absence has any significant effect on that parameter. The runs to be described consider different heat fluxes, tube radii, inlet concentrations and qualities in order to see whether changes in these conditions bring changes in any controlling resistances.

For each set of conditions the calculations were performed four times. In these four modes the mass-transfer coefficients were calculated as already described, and, as shown in table 3, multiplied by a factor F , set equal to 5.

It is shown later that the fivefold reduction in the mass-transfer resistance is equivalent to removal of that resistance. The fourth mode, with the removal of both resistances, approximates to equilibrium flash vaporization with no intraphase concentration gradients.

The table below summarises the conditions used in the runs to be described. It will be seen that in the second and subsequent runs only one condition is different from the basic set used in the first run.

To find a value of F which effectively reduces the resistances to zero a series of calculations was performed with different values of F and Mode = 4, the conditions were those of Run 1. Figure 2 gives a plot of x'_i against F for three values of z . Also shown are the values of x' for an equilibrium flash vaporization.

As the resistance is decreased x'_i tends to an asymptote. The flash method gives values which are close to the asymptote. Due to the stepwise nature of the calculations the component material balance [14] can be disobeyed as F increases. This was found to be unacceptably so for $F \gg 5$. This value however gives results sufficiently close to the asymptotes.

Figures 3, 4 and 5 show the axial variations of flowrate, concentrations and temperatures for Run 1 with Mode = 1. Figure 3 shows that dW_L/dz decreases as z increases in the lower part of the tube and then becomes constant. The second and third terms in the numerator of [11] vary

Table 3. Mass-transfer coefficients used in calculations

Mode	Phase	
	Liquid	Vapour
1	k_L^*	k_G^*
2	$F \times k_L^*$	k_G^*
3	k_L^*	$F \times k_G^*$
4	$F \times k_L^*$	$F \times k_G^*$

Table 4. Summary of conditions in calculations

Run	$W_T/(\text{kg s}^{-1})$	x_b (inlet)	W_L (inlet)/ kg s^{-1}	$q/(\text{Wm})^{-1}$	R_w (m)	Remarks
1	0.1	0.1	0.095	2×10^6	0.0165	
2	0.1	0.1	0.095	2×10^6	0.0165	Allows pressure to drop
3	0.1	0.1	0.095	2×10^5	0.0165	change of q
4	0.1	0.1	0.095	2×10^6	0.010	change of R_w
5	0.1	0.1	0.075	2×10^6	0.0165	change of inlet quality

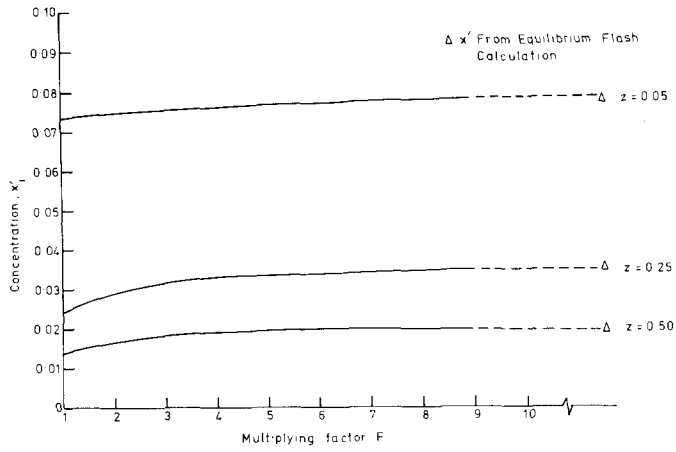


Figure 2. Graph of x'_i vs F for various z .

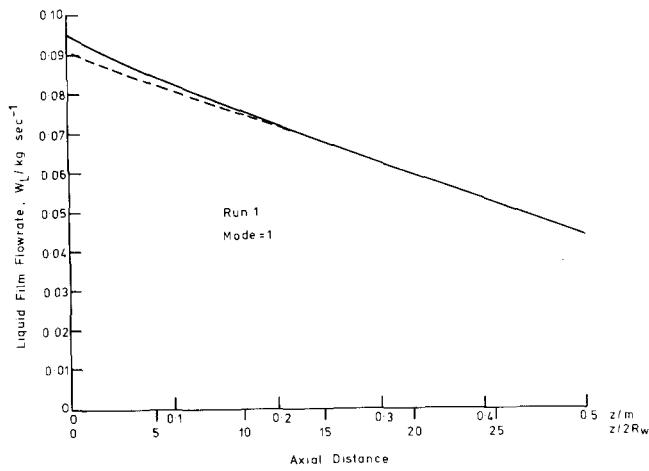


Figure 3. Axial variation of film flowrate for Run 1.

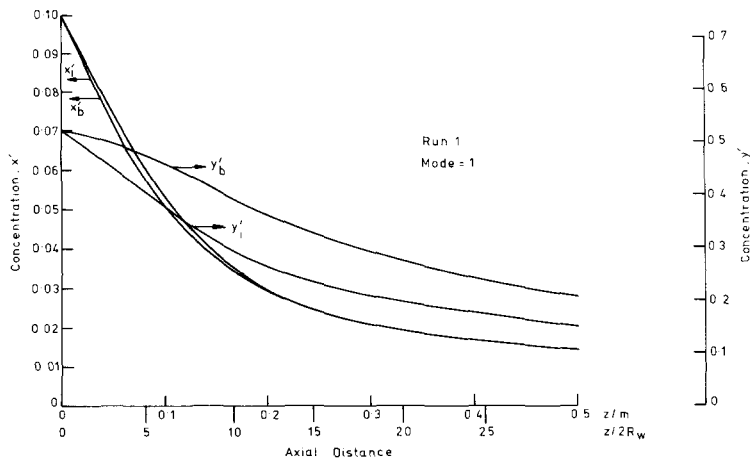


Figure 4. Axial variation of concentrations for Run 1.

with z but are shown later to be negligible everywhere compared with the, constant, first term. The denominator of [11] varies with x'_i and figure 4 shows that (dx'_i/dz) is greatest near the tube inlet.

Throughout the tube the preferential stripping from the film of the light component causes x'_b to be greater than x'_i . The value of $x'_b - x'_i$ is greatest near the inlet (it is zero at $z = 0$) although at

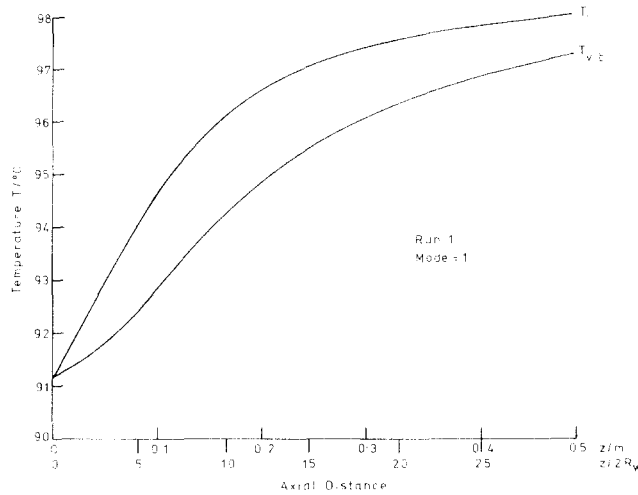


Figure 5. Axial variation of temperatures for Run 1.

$z = 0.05$ m it is only 3.4×10^{-3} . Perhaps unexpectedly we find that $y'_b > y'_i$. Thus although the mass average radial velocity is directed away from the interface the light component is diffusing counter to that flow, towards the interface. The radial concentration gradient in the vapour also at first increases with z' but then decreases.

Table 5 shows some of the subsidiary parameters associated with heat and mass transfer in this run and with Mode = 1.

The table shows that the effect of the mass transfer is to reduce the heat-transfer coefficient by at least three orders of magnitude. The mass-transfer coefficient is reduced by 34% at $z = 0.1$ m, a factor which falls to 17% by the end of the length examined.

From table 2 it can be seen that the values of Q are small enough not to influence the temperature drop across the film. From the methods described in section 4 the temperature difference across the film has been calculated and is also shown in table 5. The wall temperatures are found by adding these values to T_i . The temperature drops are perhaps higher than occur in typical reboilers because the evaporation rate is somewhat higher, at the high wall superheats predicted it is likely that the nucleate boiling neglected here would occur.

The terms in the numerator of [11] were evaluated. The term $-2\pi R_w q_w = -2.07 \times 10^5 \text{ Wm}^{-1}$. The others are listed in table 6 where

$$A = 2\pi R_i k'_L (x'_b - x'_i)(h_{LG,A} - h_{LG,B}), \quad [60]$$

$$B = 2\pi R_i h'_G (T_i - T_{G,b}). \quad [61]$$

The term A is always at least one order of magnitude less than $-2\pi R_w q_w$; the effect, discussed in section 2, is in a direction aiding evaporation. Term B increases along the tube but is always negligible compared with the other two terms in the numerator of [11].

Considering now the same run in the other calculation modes it is found that W_L is altered by no more than 0.7% by the removal of the resistance in either phase. However the plots of x'_i and T_i , figures 6 and 7, do illustrate some effects. Removal of the liquid phase resistance, Mode = 2,

Table 5. Heat- and Mass-transfer parameters

z (m)	$\frac{k'_G}{k_G}$	$\frac{h'_G}{h_G}$	z'_e	$Q \times 10^8$	$(T_w - T_i)$ °C
0.1	0.653	2.22×10^{-9}	0.05376	17.1	81.2
0.2	0.750	1.56×10^{-5}	0.03019	5.99	84.9
0.3	0.789	3.62×10^{-4}	0.02118	2.65	84.2
0.4	0.811	1.79×10^{-3}	0.01685	1.53	84.0
0.5	0.826	4.89×10^{-3}	0.01114	1.04	82.4

Table 6. Terms in (11)

z (m)	A/W (m^{-1})	B/W (m^{-1})
0.1	-4.2×10^4	1.2×10^{-10}
0.2	-1.4×10^4	7.2×10^{-7}
0.3	-0.49×10^4	2.0×10^{-5}
0.4	-0.22×10^4	1.5×10^{-4}
0.5	-0.12×10^4	4.2×10^{-4}

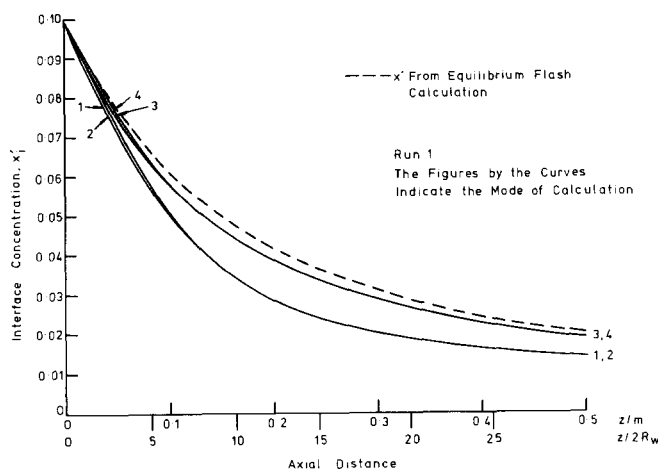


Figure 6. Axial variation of interface concentration for Run 1, effect of Mode.

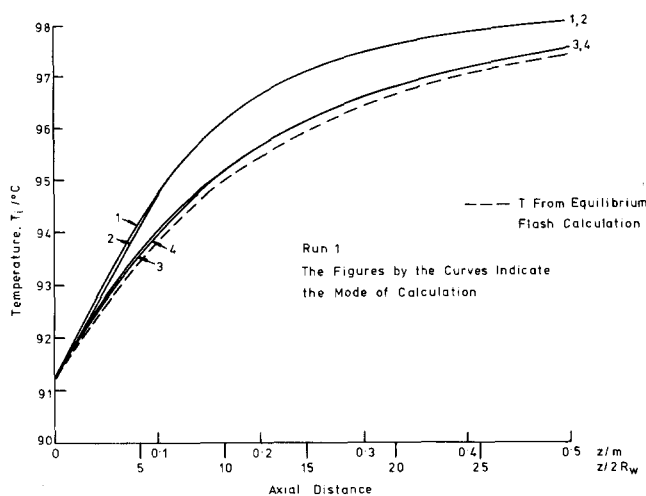


Figure 7. Axial variation of interface temperature for Run 1, effect of Mode.

has little effect on x_i' and the corresponding T_i . Hence there is little effect on the wall temperature. If the vapour phase resistance is removed, Mode = 3, x_i' changes at all z . The maximum increase occurs at $z = 0.2$ m and causes T_i there to decrease by 0.96°C ; due to the small Q , T_w will decrease similarly. Comparison of the Mode = 4 case with the latter shows again that removal of the liquid phase resistance has little effect on the axial profiles of x_i' and T_i . The graphs also show results from equilibrium flash vaporization calculations which differ little from those with Mode = 4.

At $z = 0.2$ m, where the temperature drop through the film is 84.9°C , a change of 0.96°C in T_i and thus T_w shows that, although in absolute terms, the vapour phase resistance is controlling T_w , its effect is not significant compared with errors in calculating film temperature drops which hence control wall temperatures.

It can also be seen in figures 6 and 7 that it is the vapour phase resistance which causes the temperature and concentration profiles to differ from those in the equilibrium flash vaporization case.

The driving force for setting up the concentration gradients is given by $y_b'' - x_b'$, where y_b'' is the mass fraction in the vapour in equilibrium with x_b' . At the feed concentration $x_f' = 0.1$ we are examining the region where this difference is, for ethanol-water, a maximum and the bubble point is most sensitive to concentration. Thus these conditions should show any effects of controlling resistances to their greatest extent.

Run 2 was performed with the same conditions as in Run 1 but the pressure was allowed to drop. The total calculated pressure drop over the 0.5 m was $9 \times 10^3 \text{ Nm}^{-2}$. Comparison between this and the previous run shows the same W_L in all values of Mode. Similarly there is little difference in the axial or radial variations of concentrations. Figure 8 shows the axial profiles of T_i . The axial variation of saturation temperature T_{sat} is given by

$$\frac{dT_{\text{sat}}}{dz} = \left(\frac{\partial T_{\text{sat}}}{\partial x'} \right)_P \frac{dx'}{dz} + \left(\frac{\partial T_{\text{sat}}}{\partial P} \right)_{x'} \frac{dP}{dz}. \quad [62]$$

In Run 1 (dP/dz) is set to 0, hence T_i follows the first term on the R.H.S. of [62] and increases with z . In Run 2 we have, towards the end of the tube,

$$-\left(\frac{\partial T_{\text{sat}}}{\partial P} \right)_{x'} \frac{dP}{dz} > \left(\frac{\partial T_{\text{sat}}}{\partial x'} \right)_P \frac{dx'}{dz},$$

hence T_i decreases with increasing z . The temperatures are still most sensitive to the vapour phase resistance. In succeeding runs (dP/dz) = 0.

The results from Runs 3–5 are shown in figures 9–11 respectively.

As the heat flux is reduced (Run 3) so is the tendency to set up concentration gradients. The film temperature drops are now $\sim 4^\circ\text{C}$ but the differences in T_i caused by the mass-transfer resistances are so small that they are still negligible compared with errors in calculating film temperature drops.

In Runs 4 and 5 control again lies in the vapour-phase mass-transfer resistance but the greatest differences in interfacial temperatures due thereto were respectively 0.5 and 1.1°C .

Hence we can conclude that for the mixture ethanol-water at 1 atm. the concentration and temperature profiles are not significantly affected by the resistance to mass transfer and can be calculated by the methods applicable to flash vaporization processes. Contrast nucleate boiling of that mixture which is significantly affected by liquid phase resistance to mass transfer (Stephan & Körner 1969).

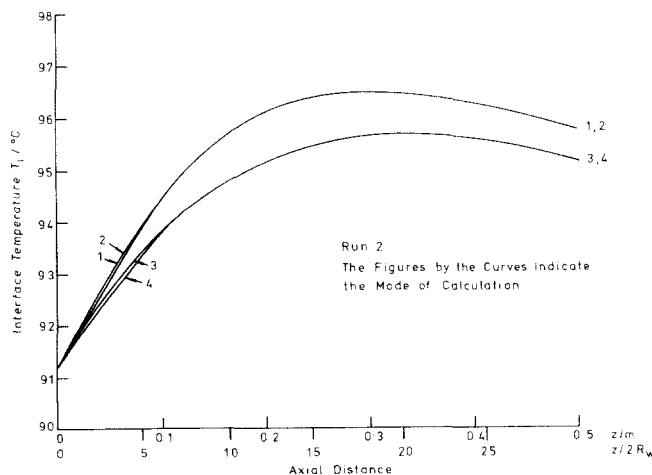


Figure 8. Axial variation of interface temperature for Run 2, effect of Mode.

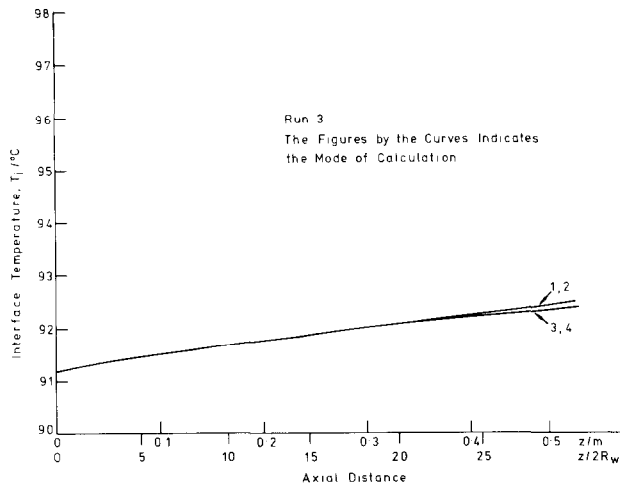


Figure 9. Axial variation of interface temperature for Run 3, effect of Mode.

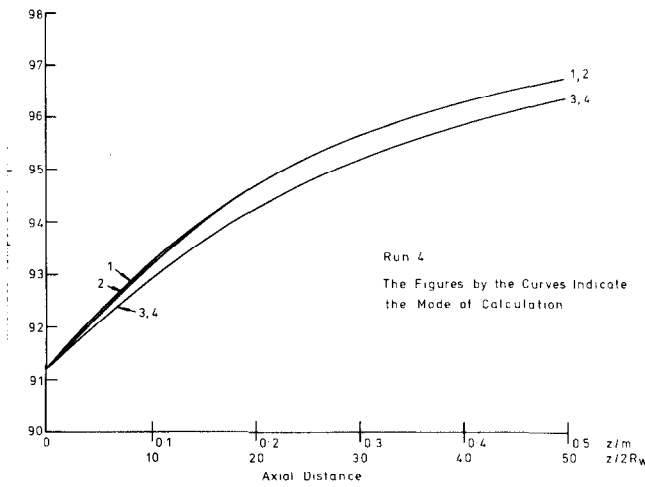


Figure 10. Axial variation of interface temperature for Run 4, effect of Mode.

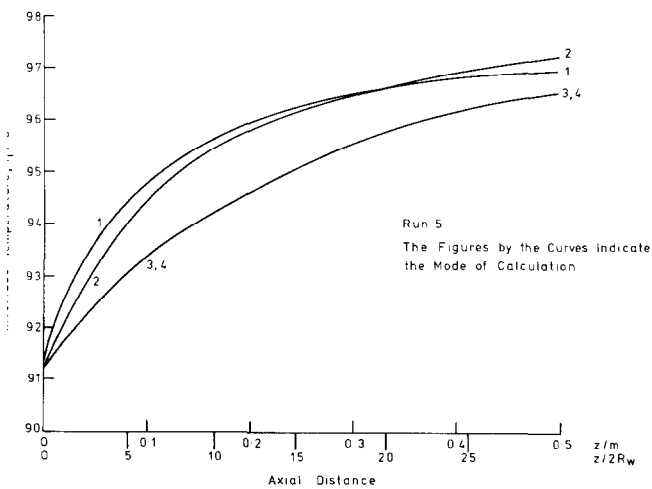


Figure 11. Axial variation of interface temperature for Run 5, effect of Mode.

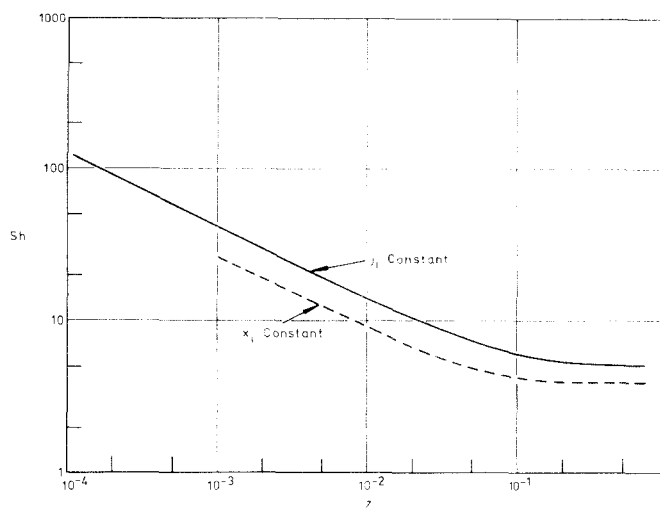


Figure 12. Graph of Sh vs Z .

It is, however, possible that such conclusions would not apply to mixtures with higher boiling ranges where the temperature differences in the various modes of calculation are potentially larger. Heat-transfer coefficients in condensation are known to be significantly affected by the presence of inert gases. Further work is aimed at the study of such cases and comparison with experimental results.

8. CONCLUSIONS

Generalized equations have been presented for the variations of flowrates and concentrations in vertical round tubes with upward flowing evaporating binary mixtures. It is assumed that all the liquid flows in an annular film dragged up the wall by shear stress exerted by the gas core. Mass- and heat-transfer coefficients in the liquid film can be calculated by the new methods presented. The model of the liquid film assumes a smooth film profile.

The equations have been solved for an ethanol-water mixture, with a boiling range of 20°C. In all the cases examined any significant difference in the concentration profiles from the equilibrium flash vaporization case appears to be due to the resistance to mass transfer in the vapour phase. Differences in wall temperatures are, however, small compared with those due to errors in calculating the temperature drop across the film. Such conclusions may not apply to mixtures with higher boiling ranges.

Heat transfer through the film is not significantly affected by the mass transfer occurring within it, but the convective heat-transfer coefficient within the vapour is very much reduced by the mass transfer through the boundary layer. The mass-transfer coefficient is similarly reduced although to a lesser degree.

Acknowledgements—The author's grateful thanks are due to Dr. D. B. R. Kenning, Dr. G. F. Hewitt and Mr. J. G. Collier for many helpful discussions in the course of this study.

Financial support from the United Kingdom Atomic Energy Authority is acknowledged.

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APPENDIX A

Mass transfer in a laminar liquid film

The Sherwood number for a purely laminar film can be derived analytically as follows.

For most cases of climbing film flow the shear stress is high and can be considered constant across the film, i.e. $\sigma^3/\eta = 0$.

Thus, for that case only,

$$\tau = \mu_L \frac{du_z}{da}, \quad [A1]$$

$$u_z = \frac{\tau_w a}{\mu_L}. \quad [A2]$$

Γ the volumetric flowrate per unit wetted perimeter, is given by

$$\Gamma = \frac{\tau_w m^2}{\mu_L 2}, \quad [\text{A3}]$$

where m is the film thickness.

In the case where the flat plate approximation is permissible [21] and [22] become

$$\rho_L \frac{\partial x'}{\partial z} \int_0^a u_z da + j = 0 \quad [\text{A4}]$$

and

$$\rho_L \frac{\partial x'}{\partial z} \int_0^m u_z da + j_i = 0, \quad [\text{A5}]$$

where j is given by

$$j = -\rho_L D_L \frac{\partial x'}{\partial a}. \quad [\text{A6}]$$

We can also write

$$\int_0^m u_z da = \Gamma. \quad [\text{A7}]$$

From [A2] to [A7] we obtain

$$\frac{-j_i a^2}{m^2} = \rho_L D_L \frac{\partial x'}{\partial a}. \quad [\text{A8}]$$

On integration and rearrangement this gives

$$x' = x'_w - \frac{j_i a^3}{3m^2 \rho_L D_L}. \quad [\text{A9}]$$

From [36] and [A7] we obtain

$$x'_b = \frac{\int_0^m x' u_a da}{\Gamma}. \quad [\text{A10}]$$

Substituting [A2], [A3] and [A9] into [A10] and integrating:

$$x'_b = x'_w - \frac{2j_i m}{15\rho_L D_L}. \quad [\text{A11}]$$

If [A8] is written for the interface and rearranged:

$$x'_i = x'_w - \frac{j_i m}{3\rho_L D_L}. \quad [\text{A12}]$$

From the definition of the mass-transfer coefficient

$$j_i = k'_L (x'_b - x'_i), \quad [\text{A13}]$$

results

$$k'_L = \frac{5\rho_L D_L}{m}, \quad [\text{A14}]$$

and from the definition of Sh_L

$$Sh_L = 5. \quad [\text{A15}]$$

APPENDIX B

This appendix outlines the calculation of development lengths for mass transfer in liquid films. The method is described elsewhere in greater detail (Shock 1975).

The equation of continuity for a flat film of constant density and fully developed velocity profile with negligible axial diffusion can be written

$$u \frac{\partial x'}{\partial z} = D_L \frac{\partial^2 x'}{\partial a^2}. \quad [\text{B1}]$$

In upward annular flow, with shear stress constant across the film, the velocity profile can be expressed by

$$u = \frac{2Qa}{a_i^2}, \quad [\text{B2}]$$

where Q is the flowrate per unit wetted perimeter.

The following dimensionless variables are introduced

$$U = u/\bar{u}, \quad [\text{B3}]$$

$$A = a/a_i, \quad [\text{B4}]$$

$$Z = D_{LZ}/\bar{u}a_i^2. \quad [\text{B5}]$$

For the case of constant interfacial mass flux a dimensionless concentration is defined by

$$\theta_j = \frac{\rho_L D_L (x' - x'_0)}{j_i a_i}, \quad [\text{B6}]$$

where x'_0 is the uniform concentration at $Z = 0$. For constant interface concentration we define

$$\theta_{x'} = \frac{x' - x'_i}{x'_i - x'_0}, \quad [\text{B7}]$$

where x'_i is the concentration at the interface at $Z > 0$.

Introducing [B2] to [B6] into [B1]:

$$\frac{U \partial \theta_j}{\partial Z} = \frac{\partial^2 \theta_j}{\partial A^2}, \quad [\text{B8}]$$

and a similar equation with $\theta_{x'}$ in place of θ_j is produced for the constant interface concentration case.

The boundary conditions for the constant j_i case are

$$\theta_j = 0 \text{ at } Z = 0,$$

$$\frac{\partial \theta_j}{\partial A} = 0 \text{ at } A = 0,$$

$$\frac{\partial \theta_j}{\partial A} = -1 \text{ at } A = 1,$$

and for the constant x_i case they are

$$\begin{aligned}\theta_{x'} &= 1 \text{ at } Z = 0, \\ \theta_{x'} &= 0 \text{ at } A = 1, \\ \frac{\partial \theta_{x'}}{\partial A} &= 0 \text{ at } A = 0.\end{aligned}$$

Equations [B8] are solved numerically to give the axial and radial variations of θ_j and $\theta_{x'}$. Using the definition of the Sherwood number together with Fick's Law it can be shown that

$$Sh_j = \frac{1}{\theta_{j,b} - \theta_{j,i}}, \quad [\text{B9}]$$

for the constant j_i case (where $\theta_{j,b}$ is the bulk value of the dimensionless concentrations and $\theta_{j,i}$ is its interface value).

Similarly it can be shown that

$$Sh_{x'} = -\frac{(\partial \theta_{x'}/\partial A)_{A=1}}{\theta_{x',b}}, \quad [\text{B10}]$$

for the constant x'_i case.

Calculated values of Sh are shown plotted against Z in figure 12. The asymptotic values of the Sherwood number for the constant x_i and j_i cases are 3.93 and 5.0 respectively. The appropriate dimensionless development lengths are 0.182 and 0.245. These are the points at which the Sherwood number is within 1% of its asymptotic value.

It should be noted that this analysis is for purely laminar flow (in the solution of the equations the value of D_L is taken to be constant across the film).

A typical set of inlet parameters in the problems considered in the main text of this paper is

$$\begin{aligned}R_w &= 0.0165 \text{ m}, \\ a_i &= 3 \times 10^{-4} \text{ m}, \\ \Gamma &= 9.2 \times 10^{-4} \text{ m}^2 \text{ s}^{-1},\end{aligned}$$

where Γ is the volumetric flowrate per unit wetted perimeter.

Equation [B4] can be transformed to give

$$z = \frac{Z\Gamma a_i}{D_L}, \quad [\text{B11}]$$

and the actual development lengths for the cases of constant x'_i and j_L become 50.2 and 67.5 m. Thus if these films were to be in laminar flow the assumption of constant mass-transfer coefficient would be untenable.

However, the values of a_i^+ are approximately 75 and thus a large degree of turbulent mixing would thus be expected. The total (turbulent plus laminar) diffusivity would no longer be constant across the film and the analysis presented in this section would not be valid.

We can, however, make some estimate of the likely order of magnitude of the development length by the following reasoning.

In his analysis Hewitt assumes the Deissler (1955) relationship for the turbulent viscosity to hold in the region $0 < a^+ < 30$. It is assumed here that the turbulent diffusivity is equal to the

turbulent viscosity at any a^+ (see Shock 1973). Thus at $a^+ = 30$ (at which point $u^+ = 13.3$) the turbulent diffusivity is given by

$$D_L' = 0.01\mu u^+ a^+ [1 - \exp(-0.01 u^+ a^+)] = 2.6 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}. \quad [\text{B12}]$$

Assuming that this value might hold over the entire film we find from [B11] that the development lengths are 2.4 cm and 1.8 cm for the constant j_i and constant x_i' cases respectively. In the region between $a^+ = 30$ and the interface the turbulent diffusivity will be considerably greater than $2.6 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$. Even though the analysis does not hold for a variable diffusivity it can be argued that the development lengths will thus be no greater than 2.0 cm. Hence in tubes of length greater than about 20 cm the assumption of a fully developed concentration profile will cause little error.