Mass Transfer in Falling Films: Influence of Finite-Amplitude Waves

An analytical, theoretical investigation of mass transfer in a Newtonian, isothermal falling film displaying periodic, finite-amplitude waves is presented. The solution derived provides full information on the mass transfer rate as expressed exclusively in terms of physical properties of the liquid and the completely determined flow.

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Introduction

Falling film equipment is widely used in chemical, process and other industries for mass and heat transfer purposes. Under operating conditions these liquid films are in general not planar, unlike the laminar falling film, for which the well-known semiparabolic theoretical solution is due to Nusselt. On the contrary, the film surface is rippled and the liquid film flow is of a convective character. Under such conditions the mass (or heat) transfer rate is increased considerably beyond the rate predicted by a theoretical model based on the laminar falling film solution. The increased transfer rate is due to convection within the liquid film. Such an underestimation of transfer rates is unsatisfactory for dimensioning purposes. These convective effects have been reported in the literature by, for example, Oliver and Atherinos (1968) and Seban and Faghri (1978).

A number of theoretical models for mass transfer in wavy falling films are reported in the literature, for example by Banerjee et al. (1967), Howard and Lightfoot (1968), Rice (1971), and Javdani (1974). These models rely on some modeling of the flow.

In view of the above, an analytical, theoretical study of the mass transfer in a wavy falling film might throw light on the manner in which the transfer rate is affected by the oscillations in the falling film. Thus the object of the present work is to provide a full theoretical solution to the problem regarding the influence of periodic, finite-amplitude waves in a Newtonian, isothermal liquid film flowing along a vertical wall on the mass transfer in such a film.

In a previous work (Barrdahl, 1986), an analytical solution was derived for the fully developed periodic, finite-amplitude wavy flow of a Newtonian, isothermal liquid film flowing along a vertical wall under the action of gravity. The hydrodynamic analysis was restricted to conditions where:

1. The liquid film thickness was much smaller than the characteristic length of the wave structure, which may be expressed as $(\omega\delta_o) \ll 1$, ω being the wave number and δ_o the mean liquid film thickness
2. Diffusive momentum transfer dominated over convective, which may be expressed as $(\omega\delta_o)\, Re < 1$, Re being the Reynolds number

In this model the stream function (and consequently the Cartesian velocity components), the liquid film surface, and the pressure were of the form:

$$\chi(x, y, t) = \sum_{\beta=-N}^{\beta=N} e^{i(\beta\omega)(x+kt)} G_\beta(y)$$

This model, or more generally, a model with this formal structure, will be used below for the fluid dynamical description of the falling film.

On the basis of such a model, the influence of the waves on the mass transfer rate in a mass transfer process, such as the absorption of a species from the gas phase, will be studied. The present work thus constitutes a natural continuation of the previous work (Barrdahl, 1986).

It may be expected that the relative influence on the mass transfer rate of an oscillatory motion in the liquid film will increase with increasing Schmidt number (the ratio of the kinematic viscosity to the mass diffusivity) of the liquid film. For this reason the present analysis will emphasize conditions where the Schmidt number is much larger than unity.

Theory

It will be assumed that it is sufficient to consider only two space coordinates, namely the longitudinal or flow direction coordinate and one transverse coordinate, the film coordinate. Thus the transverse coordinate, which is orthogonal to the normal to the wall, does not enter into the calculations.

The analysis will be carried out in the case of mass transfer although the solution also would be applicable for the case of

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heat transfer provided that any influence of a variation of the temperature on the physical properties can be neglected and that corresponding parameters, such as the Prandtl number, assume values of similar order of magnitude.

The diffusion equation then becomes:

$$\frac{\partial c}{\partial t} + u \cdot \frac{\partial c}{\partial x} + v \cdot \frac{\partial c}{\partial y} = D\left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2}\right) \tag{1}$$

The boundary conditions for absorption of a species from the gas phase are then formulated as follows:

No mass penetrates the wall:

$$\frac{\partial c}{\partial y} = 0; \quad y = 0; \quad \forall x, t \tag{2}$$

The mass concentration at the liquid film surface takes a constant value along the mass transfer section:

$$c = c^*; \quad y = \delta(x, t); \quad \forall x \geq 0 \tag{3}$$

The mass concentration inside the liquid film is homogeneous (as averaged with respect to time) before the mass transfer section:

$$c = c_o; \quad x = 0; \quad \forall y < \delta(x, t) \tag{4}$$

We now seek a solution of the following form:

$$c(x, y, t) = C_o(y) + \sum_{\beta=0}^{\beta=N} \cdot \left[\zeta_\beta(y) + \sum_{\substack{\alpha=-N \\ \alpha\neq 0}}^{\alpha=N} \xi_{\beta,\alpha}(y)\Xi_\alpha(x, t)\right] \cdot \chi_\beta(x) \tag{5}$$

with the liquid film motion described by

$$\psi(x, y, t) = \psi_o(y) + \sum_{\substack{\alpha=-N \\ \alpha\neq 0}}^{\alpha=N} \psi_\alpha(y)\Xi_\alpha(x, t) \tag{6}$$

$$\delta(x, t) = \delta_o + \sum_{\substack{\alpha=-N \\ \alpha\neq 0}}^{\alpha=N} \epsilon_\alpha \Xi_\alpha(x, t) \tag{7}$$

ψ being the stream function and δ the film thickness, where

$$u(x, y, t) = u_o(y) + \sum_{\substack{\alpha=-N \\ \alpha\neq 0}}^{\alpha=N} \frac{\partial \psi_{\alpha(y)}}{\partial y} \Xi_\alpha(x, t) \tag{8}$$

$$v(x, y, t) = -\sum_{\substack{\alpha=-N \\ \alpha\neq 0}}^{\alpha=N} \psi_\alpha(y) \frac{\partial \Xi_\alpha(x, t)}{\partial x} \tag{9}$$

Here N is an arbitrarily large number and

$$\chi_\beta(x) = e^{-\lambda_\beta x} \tag{10}$$

$$\Xi_\alpha(x, t) = e^{i(\alpha\omega)(x+kt)} \tag{11}$$

Insertion of Eqs. 5, 6, and 8–11 into Eq. 1 and suppression of χ_β and Ξ_α gives:

$$C_o''(y) = 0 \tag{12}$$

$$\zeta_\beta''(y) + \left[\frac{\lambda_\beta u_o(y)}{D} + \lambda_\beta^2\right]\zeta_\beta(y) = -\sum_{\substack{\alpha=-N \\ \alpha\neq 0}}^{\alpha=N} \left\{\frac{[\lambda_\beta + i(\alpha\omega)]\psi'_{-\alpha}(y)}{D}\xi_{\beta,\alpha}(y) - \frac{i(\alpha\omega)\psi_{-\alpha}(y)}{D}\xi'_{\beta,\alpha}(y)\right\}; \beta = 0, \ldots, N \tag{13}$$

$$\xi''_{\beta,\alpha}(y) + \left\{\frac{\lambda_\beta u_o(y)}{D} - \frac{i(\alpha\omega)}{D}[u_o(y) + k] + [\lambda_\beta - i(\alpha\omega)]^2\right\} \cdot \xi_{\beta,\alpha}(y) = -\left[\frac{\lambda_\beta \psi'_\alpha(y)}{D}\zeta_\beta(y) + \frac{i(\alpha\omega)\psi_\alpha(y)}{D}\zeta'_\beta(y)\right]; \quad \beta = 0, \ldots, N; \ \alpha = -N, \ldots, N; \ \alpha \neq 0 \tag{14}$$

where primes denote differentiation with respect to y.

Equations 13 and 14 are second-order coupled differential equations for the concentration functions $\zeta_\beta(y)$ and $\xi_{\beta,\alpha}(y)$.

Next a solution is sought for the systems of Eqs. 12–14 on the assumption that contributions containing third- and higher order terms in ψ_α, ψ'_α, ϵ_α, etc. may be neglected. In addition, the solutions sought will be approximated in that u_o, ψ_α, ψ'_α are evaluated at $y = \delta_o$.

Under these circumstances, a solution is:

$$C_o(y) = A_o + B_o y \tag{15}$$

$$\zeta_\beta(y) = \zeta_\beta^o(y) + \eta_\beta(y) \tag{16}$$

$$\zeta_\beta^o(y) = A_\beta \cos(\theta_\beta y) + B_\beta \sin(\theta_\beta y) + 0[(\psi_\alpha/\psi_o)^3] \tag{17}$$

$$\theta_\beta = \left[\frac{\lambda_\beta u_o}{D} + \lambda_\beta^2 + \sum_{\substack{\alpha=-N \\ \alpha\neq 0}}^{\alpha=N} \frac{\psi'_\alpha \psi'_{-\alpha}\lambda_\beta}{D(u_o + k)}\right]^{1/2} \tag{18}$$

$$\eta_\beta(y) = \sum_{\substack{\alpha=-N \\ \alpha\neq 0}}^{\alpha=N} \frac{1}{(\theta_{\beta,\alpha}^2 - \theta_\beta^2)}\left\{\frac{[\lambda_\beta + i(\alpha\omega)]\psi'_{-\alpha}}{D}\xi_{\beta,\alpha}^o(y) - \frac{i(\alpha\omega)\psi_{-\alpha}}{D}\xi_{\beta,\alpha}^{o\prime}(y)\right\} + 0((\psi_\alpha/\psi_o)^3) \tag{19}$$

$$\xi_{\beta,\alpha}(y) = \xi_{\beta,\alpha}^o(y) + \eta_{\beta,\alpha}(y) \tag{20}$$

$$\xi_{\beta,\alpha}^o(y) = A_{\beta,\alpha}\cos(\theta_{\beta,\alpha}y) + B_{\beta,\alpha}\sin(\theta_{\beta,\alpha}y) + O[(\psi_\alpha/\psi_o)^2] \tag{21}$$

$$\eta_{\beta,\alpha}(y) = -\frac{1}{(\theta_{\beta,\alpha}^2 - \theta_\beta^2)}\left[\frac{\lambda_\beta\psi'_\alpha}{D}\zeta_\beta^o(y) + \frac{i(\alpha\omega)\psi_\alpha}{D}\zeta_\beta^{o\prime}(y)\right] + 0[(\psi_\alpha/\psi_o)^2] \tag{22}$$

$$\theta_{\beta,\alpha} = \left\{\frac{\lambda_\beta u_o}{D} - \frac{i(\alpha\omega)}{D}[u_o + k] + [\lambda_\beta - i(\alpha\omega)]^2\right\}^{1/2} \tag{23}$$

where the argument $y = \delta_o$ has been suppressed on u_o, ψ_α, and ψ'_α.

It remains to determine the eigenvalues θ_β, $\theta_{\beta,\alpha}$, λ_β and the amplitudes A_o, B_o, A_β, B_β, $A_{\beta,\alpha}$, and $B_{\beta,\alpha}$. For this purpose the boundary conditions of Eqs. 2 to 4 are required and use will then be made of Eqs. 5–11 and 15–23.

The boundary condition in Eq. 2 gives:

$$B_o = 0 \tag{24}$$

$$\theta_\beta B_\beta + \sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N} \frac{\theta_{\beta,\alpha}}{(\theta_{\beta,\alpha}^2 - \theta_\beta^2)} \cdot \left\{ \frac{[\lambda_\beta + i(\alpha\omega)]\psi'_{-\alpha}}{D} B_{\beta,\alpha} + \frac{i(\alpha\omega)\psi_{-\alpha}}{D} \theta_{\beta,\alpha} A_{\beta,\alpha} \right\} = 0 \tag{25}$$

$$\theta_{\beta,\alpha} B_{\beta,\alpha} - \frac{1}{(\theta_{\beta,\alpha}^2 - \theta_\beta^2)} \left[\frac{\lambda_\beta \psi'_\alpha}{D} \theta_\beta B_\beta - \frac{i(\alpha\omega)\psi_\alpha}{D} \theta_\beta^2 A_\beta \right] = 0 \tag{26}$$

Functional values taken at $y = \delta(x, t)$ are, for use with the boundary conditions in Eqs. 3 and 4, expanded around $y = \delta_o$ in a Taylor series of the form: $Y(\delta_o + \epsilon) = Y(\delta_o) + \epsilon Y'(\delta_o) + \cdot (\epsilon^2/2) Y''(\delta_o) + \cdot \cdot \cdot$, where ϵ represents terms of the form $\Gamma_\alpha \Xi_\alpha$, Γ_α being any function.

The boundary condition in Eq. 3 gives:

$$A_o = c^* \tag{27}$$

$$\begin{aligned} &A_\beta \cos(\theta_\beta \delta_o) + B_\beta \sin(\theta_\beta \delta_o) + \sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N} \frac{1}{(\theta_{\beta,\alpha}^2 - \theta_\beta^2)} \\ &\cdot \left\{ \frac{[\lambda_\beta + i(\alpha\omega)]\psi'_{-\alpha}}{D} [A_{\beta,\alpha}\cos(\theta_{\beta,\alpha}\delta_o) + B_{\beta,\alpha}\sin(\theta_{\beta,\alpha}\delta_o)] \right. \\ &\left. + \frac{i(\alpha\omega)\psi_{-\alpha}}{D} \theta_{\beta,\alpha} [A_{\beta,\alpha}\sin(\theta_{\beta,\alpha}\delta_o) - B_{\beta,\alpha}\cos(\theta_{\beta,\alpha}\delta_o)] \right\} \\ &+ \sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N} \epsilon_{-\alpha} \left(-\theta_{\beta,\alpha} A_{\beta,\alpha} \sin(\theta_{\beta,\alpha}\delta_o) + \theta_{\beta,\alpha} B_{\beta,\alpha} \cos(\theta_{\beta,\alpha}\delta_o) \right. \\ &+ \frac{1}{(\theta_{\beta,\alpha}^2 - \theta_\beta^2)} \left\{ \frac{\lambda_\beta \psi'_\alpha}{D} \theta_\beta [A_\beta \sin(\theta_\beta\delta_o) - B_\beta \cos(\theta_\beta \delta_o)] \right. \\ &\left. + \frac{i(\alpha\omega)\psi_\alpha}{D} \theta_\beta^2 [A_\beta \cos(\theta_\beta\delta_o) + B_\beta \sin(\theta_\beta\delta_o)] \right\} \\ &\left. - \frac{1}{2} \epsilon_\alpha \theta_\beta^2 [A_\beta \cos(\theta_\beta\delta_o) + B_\beta \sin(\theta_\beta \delta_o)] \right) \\ &+ 0[(\psi_\alpha/\psi_o)^3] = 0 \end{aligned} \tag{28}$$

$$\begin{aligned} &A_{\beta,\alpha}\cos(\theta_{\beta,\alpha}\delta_o) + B_{\beta,\alpha}\sin(\theta_{\beta,\alpha}\delta_o) \\ &- \frac{1}{(\theta_{\beta,\alpha}^2 - \theta_\beta^2)} \left\{ \frac{\lambda_\beta\psi'_\alpha}{D} [A_\beta \cos(\theta_\beta\delta_o) + B_\beta\sin(\theta_\beta\delta_o)] \right. \\ &- \frac{i(\alpha\omega)\psi_\alpha}{D} \theta_\beta [A_\beta \sin(\theta_\beta\delta_o) - B_\beta\cos(\theta_\beta\delta_o)] \\ &\left. - \epsilon_\alpha \theta_\beta [A_\beta \sin(\theta_\beta\delta_o) - B_\beta \cos(\theta_\beta\delta_o)] \right\} \\ &+ 0[(\psi_\alpha/\psi_o)^2] = 0 \end{aligned} \tag{29}$$

The boundary condition in Eq. 4 gives the single expression:

$$\begin{aligned} (c_o - c^*) = \sum_{\beta=0}^{\beta=N} \Bigg[&A_\beta \cos(\theta_\beta y) + B_\beta \sin(\theta_\beta y) \\ &+ \sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N} \frac{1}{(\theta_{\beta,\alpha}^2 - \theta_\beta^2)} \left\{ \frac{\psi'_{-\alpha}}{D} [A_{\beta,\alpha}\cos(\theta_{\beta,\alpha} y) \right. \\ &+ B_{\beta,\alpha}\sin(\theta_{\beta,\alpha} y)][\lambda_\beta + i(\alpha\omega)] \\ &\left. + \frac{i(\alpha\omega)\psi_{-\alpha}}{D} \theta_{\beta,\alpha} [A_{\beta,\alpha}\sin(\theta_{\beta,\alpha} y) - B_{\beta,\alpha}\cos(\theta_{\beta,\alpha} y)] \right\} \\ &+ \sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N} \left(A_{\beta,\alpha}\cos(\theta_{\beta,\alpha} y) + B_{\beta,\alpha}\sin(\theta_{\beta,\alpha} y) \right. \\ &- \frac{1}{(\theta_{\beta,\alpha}^2 - \theta_\beta^2)} \left\{ \frac{\lambda_\beta\psi'_\alpha}{D} [A_\beta\cos(\theta_\beta y) + B_\beta \sin(\theta_\beta y)] \right. \\ &\left.\left. - \frac{i(\alpha\omega)\psi_\alpha}{D} \theta_\beta [A_\beta \sin(\theta_\beta y) - B_\beta \cos(\theta_\beta y)] \right\} \right) \\ &\cdot \Xi_\alpha(0, t) \Bigg] + 0[(\psi_\alpha/\psi_o)^3] \end{aligned} \tag{30}$$

Next, a solution involving Eqs. 25, 26, and 27–30 is sought on the following assumptions:

1. The molecular mass diffusion of the absorbed species is a much slower process than the molecular momentum diffusion in the liquid. This may be expressed as $Sc \gg 1$, where $Sc = \nu/D$.
2. The characteristic length of the wave structure is much shorter than that of the mass transfer process. This may be expressed as $|\theta_{\beta,\alpha}| \gg \theta_\beta$ or $(\omega\delta_o)Re(z + 1)Sc \gg 1$.

Under these circumstances Eqs. 25, 26, 28, and 29, after simplification and substitution among them, become:

$$B_\beta = -iA_\beta\theta_\beta \sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N} \theta_{\beta,\alpha} \frac{\psi_\alpha\psi_{-\alpha}}{(u_o + k)^2} [1 + 0(\theta_\beta/\theta_{\beta,\alpha}, \psi_\alpha/\psi_o)] \tag{31}$$

$$B_{\beta,\alpha} = A_\beta \frac{\theta_\beta^2}{\theta_{\beta,\alpha}} \frac{\psi_\alpha}{(u_o + k)} [1 + 0(\theta_\beta/\theta_{\beta,\alpha}, \psi_\alpha/\psi_o)] \tag{32}$$

$$A_{\beta,\alpha} = -iA_\beta \frac{\theta_\beta^2}{\theta_{\beta,\alpha}} \frac{\psi_\alpha}{(u_o + k)} [1 + 0(\theta_\beta/\theta_{\beta,\alpha}, \psi_\alpha/\psi_o)] \tag{33}$$

$$\cos(\theta_\beta\delta_o) - i\theta_\beta \sin(\theta_\beta\delta_o) \sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N} \theta_{\beta,\alpha} \frac{\psi_\alpha\psi_{-\alpha}}{(u_o + k)^2} \cdot [1 + 0(\theta_\beta/\theta_{\beta,\alpha}, \psi_\alpha/\psi_o)] = 0 \tag{34}$$

Here Eq. 34 provides the condition to determine the eigenvalues θ_β and hence also λ_β and $\theta_{\beta,\alpha}$. A solution to Eq. 34 is as follows, upon suppression of the indication of neglected terms of $0(\theta_\beta/\theta_{\beta,\alpha}, \psi_\alpha/\psi_o)$:

$$\theta_\beta = \theta_\beta^o + \kappa_\beta \tag{35}$$

where

$$\theta_\beta^o = \frac{\pi}{\delta_o}\left(\beta + \frac{1}{2}\right) \tag{36}$$

$$\kappa_\beta = -i\frac{\theta^o_\beta}{\delta_o}\sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N}\theta_{\beta,\alpha}\frac{\psi_\alpha\psi_{-\alpha}}{(u_o+k)^2} \tag{37}$$

In order to determine the remaining unknowns, the amplitudes A_β, Eq. 30 is multiplied on both sides with θ^o_β, integrated from $y=0$ to $y=\delta(0,t)$, and subsequently averaged with respect to time. The result is, after similar suppression of terms as above:

$$\begin{aligned}(c_o-c^*)\frac{(-1)^\beta}{\theta^o_\beta} &= \frac{\delta_o}{2}\left(1-\frac{1}{2}\frac{\kappa_\beta}{\theta_\beta}\right)A_\beta+\frac{1}{2}\frac{1}{\theta_\beta}B_\beta \\ &+\sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N}\frac{\theta^2_{\beta,\alpha}}{(\theta^2_{\beta,\alpha}-\theta^2_\beta)^2}A_{\beta,\alpha}\frac{i(\alpha\omega)\psi_{-\alpha}}{D} \\ &+\sum_{\substack{\gamma=0\\ \gamma\neq\beta}}^{\gamma=N}\left(A_\gamma(-1)^{\gamma-\beta}\cdot\frac{\kappa_\gamma\delta_o^2}{2\pi}\left[\frac{1}{(\gamma-\beta)}-\frac{1}{(\beta+\gamma+1)}\right]\right. \\ &+B_\gamma\frac{\delta_o}{2\pi}\left\{\frac{1}{(\gamma-\beta)}+\frac{1}{(\gamma+\beta+1)}-(-1)^{\beta-\gamma}\left[\frac{1}{(\gamma-\beta)}\right.\right. \\ &\left.\left.-\frac{1}{(\gamma+\beta+1)}\right]\right\}+\sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N}A_{\gamma,\alpha}\frac{\theta^2_{\gamma,\alpha}}{(\theta^2_{\gamma,\alpha}-\theta^2_\gamma)}\frac{1}{(\theta^2_{\gamma,\alpha}-\theta^2_\beta)} \\ &\left.\cdot\frac{i(\alpha\omega)\psi_{-\alpha}}{D}\right)=0\end{aligned} \tag{38}$$

After substitution among expressions 31 to 37, Eq. 38 gives:

$$A_\beta=\frac{2}{\pi}(c_o-c^*)\frac{(-1)^\beta}{\left(\beta+\frac{1}{2}\right)}\cdot\left\{1-\frac{1}{2}\frac{\kappa_\beta}{\theta_\beta}\left[1+4\left(\beta+\frac{1}{2}\right)\sum_{\substack{\gamma=0\\ \gamma\neq\beta}}^{\gamma=N}\frac{(-1)^{\beta-\gamma}\left(\gamma+\frac{1}{2}\right)}{(\gamma-\beta)(\gamma+\beta+1)}\right]\right\} \tag{39}$$

By means of Eq. 18, λ_β can be determined as

$$\lambda_\beta=\frac{D}{u_o}\theta^{o^2}_\beta\left(1+2\frac{\kappa_\beta}{\theta^o_\beta}\right) \tag{40}$$

and $\theta_{\beta,\alpha}$ can be expressed as

$$\theta_{\beta,\alpha}=\left[-\frac{i(\alpha\omega)}{D}(u_o+k)\right]^{1/2}[1+0(\theta_\beta/\theta_{\beta,\alpha})] \tag{41}$$

The solution is now complete.

The expression for the rate of mass transfer (of the absorbed species) at the liquid film surface is:

$$\frac{\partial M(x,t)}{\partial t}=-DS\frac{\partial c(x,y,t)}{\partial y}\bigg|_{y=\delta(x,t)} \tag{42}$$

Insertion of Eqs. 5–11, 15–24, 27, 31–33, 35–37, and 39–41 into Eq. 42 and suppression of the indication of neglected terms of $0(\theta_\beta/\theta_{\beta,\alpha},\psi_\alpha/\psi_o)$ gives:

$$\frac{\partial M_o(x)}{\partial t}=2DS\frac{(c_o-c^*)}{\delta_o}\sum_{\beta=0}^{\beta=N}e^{-\lambda^o_\beta(1+2\kappa_\beta/\theta_\beta)x}\cdot\left\{1+\frac{1}{2}\frac{\kappa_\beta}{\theta_\beta}\left[1-4\left(\beta+\frac{1}{2}\right)\sum_{\substack{\gamma=0\\ \gamma\neq\beta}}^{\gamma=N}\frac{(-1)^{\beta-\gamma}\left(\gamma+\frac{1}{2}\right)}{(\gamma-\beta)(\gamma+\beta+1)}\right]\right\} \tag{43}$$

where

$$\lambda^o_\beta=\theta^{o^2}_\beta\frac{D}{u_o} \tag{44}$$

$$\kappa_\beta=\theta^o_\beta(1-i)[(\omega\delta_o)ReSc(-1/2)(z+1)]^{1/2}.$$

$$\sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N}(\alpha)^{1/2}\frac{\psi_\alpha\psi_{-\alpha}}{u_o^2(z+1)^2}\frac{1}{\delta_o^2}[1+0(\theta_\beta/\theta_{\beta,\alpha},\psi_\alpha/\psi_o)] \tag{45}$$

with

$$Re=\frac{u_o\delta_o}{\nu} \tag{46}$$

$$Sc=\frac{\nu}{D} \tag{47}$$

$$k=zu_o \tag{48}$$

Thus the influence on the mass transfer rate of the oscillations in the liquid film is determined. It is interesting to note the dependency of the wave oscillations on the concentration and the mass transfer rate, namely a proportionality to the square root of $(\omega\delta_o)ReSc(z+1)(-1)$ multiplied by a sum containing a quadratic form of the perturbed stream functions, which in their turn are proportional to the amplitude of the oscillations.

An estimate will now be made of κ_β by means of the specific fluid dynamical model derived in Barrdahl (1986), despite the fact that this model is restricted to values of $(\omega\delta_o)$ Re less than about 1/3. The model is expressed in the approximate form:

$$\epsilon_{\pm1}=a\delta_o \tag{49}$$

$$\epsilon_{\pm2}=a\delta_o\left(\frac{7}{6}a\mp\frac{i}{6}\right) \tag{50}$$

$$\psi_{\pm1}=u_oa\delta_o\left(1\mp i\frac{9}{2}a\right) \tag{51}$$

$$\psi_{\pm2}=u_oa\delta_o\left(-20a\mp i\frac{5}{16}\right) \tag{52}$$

with

$$a=\pm\frac{1}{5}(\omega\delta_o)Re;\quad(\omega\delta_o)Re\leq\frac{1}{3} \tag{53}$$

$$z=-2 \tag{54}$$

$$\omega = u_o\left(\frac{\rho}{\gamma\delta_o}\right)^{1/2} \tag{55}$$

where dependency on k or z has been neglected formally.

After insertion of Eqs. 49–55 into Eq. 45, the result is:

$$\kappa_\beta = \theta_\beta^o(\omega\delta_o)^{5/2}Re^{5/2}Sc^{1/2} \cdot \left[\left(1 + 2^{1/2}\cdot\frac{25}{256}\right) + \left(400\cdot 2^{1/2} + \frac{81}{4}\right)\frac{1}{25}(\omega\delta_o)^2Re^2\right] \cdot \frac{2^{1/2}}{25} \tag{56}$$

Results

The complete solution can be expressed in the form:

$$c(x,y,t) = c^* + \sum_{\beta=0}^{\beta=N} e^{-\lambda_\beta x}\Bigg[A_\beta\cos(\theta_\beta y) + B_\beta\sin(\theta_\beta y) + \sum_{\substack{\alpha=-N\\ \alpha\neq 0}}^{\alpha=N}\Bigg(-\frac{\psi'_{-\alpha}}{(u_o+k)}[A_{\beta,\alpha}\cos(\theta_{\beta,\alpha}y) + B_{\beta,\alpha}\sin(\theta_{\beta,\alpha}y)] - \frac{\psi_{-\alpha}}{(u_o+k)}\theta_{\beta,\alpha}[A_{\beta,\alpha}\sin(\theta_{\beta,\alpha}y) - B_{\beta,\alpha}\cos(\theta_{\beta,\alpha}y)] + e^{i(\alpha\omega)(x+kt)}\Bigg\{A_{\beta,\alpha}\cos(\theta_{\beta,\alpha}y) + B_{\beta,\alpha}\sin(\theta_{\beta,\alpha}y) - \frac{\psi_\alpha}{(u_o+k)}\theta_\beta[A_\beta\sin(\theta_\beta y) - B_\beta\cos(\theta_\beta y)]\Bigg\}\Bigg)\Bigg] \tag{57}$$

where

$$A_\beta = \frac{2}{\pi}(c_o - c^*)\frac{(-1)^\beta}{\left(\beta+\frac{1}{2}\right)} \left\{1 - \frac{1}{2}\frac{\kappa_\beta}{\theta_\beta}\left[1 + 4\left(\beta+\frac{1}{2}\right)\sum_{\substack{\gamma=0\\ \gamma\neq\beta}}^{\gamma=N}\frac{(-1)^{\beta-\gamma}}{(\gamma-\beta)}\frac{\left(\gamma+\frac{1}{2}\right)}{\left(\gamma+\beta+\frac{1}{2}\right)}\right]\right\} \tag{39}$$

$$B_\beta = A_\beta\kappa_\beta\delta_o \tag{57}$$

$$A_{\beta,\alpha} = -iA_\beta\frac{\theta_\beta^2}{\theta_{\beta,\alpha}}\frac{\psi_\alpha}{(u_o+k)} \tag{33}$$

$$B_{\beta,\alpha} = iA_{\beta,\alpha} \tag{58}$$

$$\theta_\beta = \theta_\beta^o + \kappa_\beta \tag{35}$$

$$\theta_\beta^o = \frac{\pi}{\delta_o}\left(\beta + \frac{1}{2}\right) \tag{36}$$

$$\kappa_\beta = 2^{1/2}\frac{\theta_\beta^o}{\delta_o}\left[\frac{-\omega(u_o+k)}{D}\right]^{1/2}\cdot\sum_{\alpha=1}^{\alpha=N}(|\alpha|)^{1/2}\frac{\psi_\alpha\psi_{-\alpha}}{(u_o+k)^2} \tag{59}$$

$$\theta_{\beta,\alpha} = \left[-\frac{i(\alpha\omega)}{D}(u_o+k)\right]^{1/2} \tag{41}$$

$$\lambda_\beta = \theta_\beta^{o^2}\frac{D}{u_o}\left(1 + 2\frac{\kappa_\beta}{\theta_\beta^o}\right) \tag{40}$$

where only the lowest order terms have been retained in the expressions. With a value of less than $-u_o$ for k, which is true at least for smaller perturbations to the laminar flow, κ_β becomes completely real.

It is readily apparent that with a sufficiently large value of the Schmidt number the rate of mass transfer will be substantially increased even at low values of the wave amplitude. A dimensional analysis of κ_β reveals that it is essentially dependent on the following group of dimensionless numbers in the region investigated:

$$a^2(\omega\delta_o)^{1/2}Re^{1/2}Sc^{1/2}(z+1)^{-3/2}$$

where a is an amplitude coefficient defined through the first Fourier component of the liquid film thickness as $\epsilon_{\pm 1} = a\delta_o$, $(\omega\delta_o)$ the product of wave number and mean liquid film thickness, z the nondimensional wave velocity, $Re = u_o\delta_o/\nu$ the Reynolds number, and $Sc = \nu/D$ the Schmidt number. a is not to be considered as a constant but depends on the liquid film flow and its properties in an intimate manner.

In the limit, as the film thickness vanishes the above functional expression reduces to:

$$\left(\frac{\rho\nu^2}{\gamma\delta_o}\right)^{5/4}Re^5Sc^{1/2}$$

where, as a consequence of the analytical solution used for the flow, the Reynolds number also vanishes.

Insertion of the hydrodynamical parameters derived in Barrdahl (1986) into the presently derived mass transfer model gives an altogether analytical solution valid at generally low flow rates, or more precisely up to a value of about ⅓ of the dimensionless aggregate $(\omega\delta_o)Re$. Unfortunately, no experimental results for mass transfer rates are available in this region of flow.

At higher values of $(\omega\delta_o)Re$, no full analytical solution to the equations of motion—that is, a unique solution expressed solely in terms of physical constants of the liquid and the flow rate—is presently available. Therefore a comparison with existing experimental data will by necessity be of a restricted character.

Brauer (1985) has reported correlated experimental data for the presently studied type of system, and presents them in the form $Sh = fRe^bSc^{1/2}$, where $Sh = \alpha\delta_o/D$ is the Sherwood number for infinite mass transfer section length; f is a factor the magnitude of which depends on the Reynolds number; b is a magnitude coefficient dependent on the Reynolds number; and where $Re \geqq 12$ and Sc is much larger than unity. α is the mass transfer coefficient.

A comparison of the presently derived qualitative expression of dimensionless entities:

$$a^2(\omega\delta_o)^{1/2}Re^{1/2}Sc^{1/2}(z+1)^{-3/2}$$

with the above correlation formula leads to the following conclusions:

1. The dependencies on the Schmidt number are identical. This is an encouraging result in view of the fact that at these higher flow rates, the convective contribution to the mass trans-

fer process becomes relatively more important than the diffusive one.

2. The Reynolds number raised to a certain power appears in both formulas. Due to the indeterminate flow parameters in these regions of higher flow rates, a direct comparison of the dependency on the Reynolds number cannot be made.

Conclusions

An analytical theoretical model for the mass transfer in falling liquid films at high Schmidt numbers has been presented. The present model constitutes an analytical improvement over previous models employing the completely laminar flow model as input into the diffusion equation, in that account is taken of periodic ripples in the liquid flow.

The mass transfer characteristics are determined completely and solely in terms of physical properties of the liquid and the diffusing species, the parameters of the fully described liquid film flow, and the mass transfer section geometry. These properties and parameters are: the mass diffusivity of the diffusing species D; the mean liquid film thickness δ_o; the surface velocity of the liquid at the mean liquid film thickness u_o; the wave number ω; the Fourier components of the stream function ψ_α; the nondimensional wave velocity z; and Re and Sc, the Reynolds and Schmidt numbers. For the evaluation of a mass transfer coefficient, the mass transfer section width and length are also needed.

The present model predicts a substantial increase in the rate of mass transfer due to the presence of the waves in the falling film. The increase in the rate of mass transfer is essentially proportional to the following aggregate of dimensionless numbers:

$$\mathscr{a}^2(\omega\delta_o)^{1/2}Re^{1/2}Sc^{1/2}(z+1)^{-3/2}$$

where $\mathscr{a}$ is a wave amplitude defined through $\epsilon_1 = \mathscr{a}\delta_o$, the first Fourier component for the liquid film surface.

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Notation

$\mathscr{a}$ = wave amplitude coefficient
A_o = amplitude factor for component o, kg/m^3
A_β = amplitude factor for component β, kg/m^3
$A_{\beta,\alpha}$ = amplitude factor for component with indices β, α, kg/m^3
B_o = amplitude factor for component o, kg/m^4
B_β = amplitude factor for component β, kg/m^3
$B_{\beta,\alpha}$ = amplitude factor for component with indices β, α, kg/m^3
c = mass concentration of diffusing species, kg/m^3
c^* = interfacial mass concentration of diffusing species, kg/m^3
c_o = initial mass concentration of diffusing species, kg/m^3
C_o = mass concentration function for component o, kg/m^3
D = mass diffusivity, m^2/s
g = gravitational coefficient, m/s^2
k = wave velocity in negative x direction, m/s
M = mass, kg
Re = Reynolds number, $u_o\delta_o/\nu$
S = surface area, m^2
Sc = Schmidt number, ν/D
t = time coordinate, s
u = longitudinal velocity, m/s
u_o = mean longitudinal velocity at $y = \delta_o$, m/s
v = transverse velocity, m/s
x = longitudinal coordinate, m
y = transverse coordinate, m
z = nondimensional wave velocity, k/u_o

Greek letters

α = integer summation number
β = integer summation number
γ = integer summation number and surface tension coefficient, kg/s^2
δ = liquid film thickness, m
δ_o = mean liquid film thickness, m
ϵ_α = Fourier component of liquid film thickness, m
λ_β = eigenvalue for component β, 1/m
θ_β = eigenvalue for component β, 1/m
$\theta_{\beta,\alpha}$ = eigenvalue for component with indices β, α, 1/m
ζ_β = mass concentration function for component β, kg/m^3
$\xi_{\beta,\alpha}$ = mass concentration function for component with indices β, α, kg/m^3
Ξ_α = circular function
χ_β = exponential function
ψ = stream function, m^2/s
ψ_o = unperturbed stream function, m^2/s
ψ_α = stream function of component α, m^2/s
ν = kinematic viscosity coefficient, m^2/s
ρ = mass density, kg/m^3
κ_β = eigenvalue for component β, 1/m
ω = wave number, 1/m
η_β = inhomogeneous part of solution to concentration function for component β, kg/m^3
$\eta_{\beta,\alpha}$ = inhomogeneous part of solution to concentration function with indices β, α, kg/m^3

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