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TURBULENT MASS TRANSFER AT FREE, GAS–LIQUID INTERFACES, WITH APPLICATIONS TO FILM FLOWS

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Abstract—The film hydrodynamics determined by Telles and Dukler, together with a turbulence-centred mass-transfer model and an empirical estimate for the macroscale, provide a coherent description of a rather unique mass-transfer mechanism which is found to be in agreement with overall mass-transfer measurements.

NOMENCLATURE

c_f, concentration in film;
c_0, initial concentration;
c_s, interfacial concentration;
c_w, concentration in wave;
c_2, cup-mixing concentration;
C, wave celerity;
C_{IMA}, constant of $k_L = k_L^I$ regime, equation (2);
C_{UET}, constant of $k_L = k_L^D$ regime, equation (1);
D, molecular diffusivity;
$F(\alpha)$, factor for transient effects, given in Fig. 1;
g, acceleration of gravity;
h_w, mean height of wave above base film;
k_L, overall mass-transfer coefficient;
k_L^D, mass-transfer coefficient for dissipative regime;
k_L^I, mass-transfer coefficient for inertial regime;
L, macroscale;
L_S, test-section length;
L_w, base length of large waves;
m_L, base-film thickness;
Q, volumetric flow rate per unit width;
Re_f, base-film Reynolds number;
Re_t, turbulence Reynolds number, LV/ν;
Re_t^*, critical turbulence Reynolds number, see equations (1) and (2);
Re_w, wave Reynolds number, $4C(m_L+h_w)/\nu$;
T, characteristic time of large-scale motions, $L/\sqrt{2}V$;
t_{exp}, exposure time for mass transfer;
V, turbulence intensity;
$\bar{V}$, bulk velocity;
V_0, Levich's characteristic eddy velocity;
V_s, surface velocity;
V_w, wave volume per unit width;
V_*, shear velocity.

Greek symbols

α, t_{exp}/T;
ε, rate of dissipation per unit mass;
λ_p, average wave separation;
π, 3·1416;
ρ, density;
σ, surface tension;
ν, kinematic viscosity;
χ, fraction of film flow in the waves.

1. INTRODUCTION

THE PROBLEM dealt with in this paper is the theoretical prediction of mass-transfer coefficients for gas absorption in turbulent, thin, falling, liquid films. A rather detailed description of the gross fluid dynamics of such flows has recently emerged. It appears clear now that turbulent falling films consist of two distinct flow substructures: a laminar base film flowing next to the wall and turbulent waves which slide at much higher velocities on the top of this base film. These waves have an extremely large base-to-height ratio, move independently of each other, and may better be envisioned as solitary segments of thicker turbulent films. A physically meaningful description of the mass-transfer process must take into account this rather unique flow structure. For the case of absorption with chemical reactions, the benefits of such an approach become of more practical significance, as conversions and selectivities are likely to be affected. These considerations provided the primary motivation for this work.

For the turbulent portion of the flow, the classical problem of turbulence interaction with a "free" gas–liquid interface and the implications to mass transfer must be addressed. This fundamental step of relating the mass-transfer coefficient to the "local" turbulence properties should not be peculiar only to film flows, but generally applicable to flows of all geometries with gas–liquid interfaces. Open-channel flows, bubble flows, and jet flows provide a few such examples. Experience from these systems is also extensive and may be used to an advantage for guiding efforts towards a more unified approach in predicting absorption rates. For a complete formulation, a knowledge or predictive capability of the relevant turbulence properties must be available. For film flows, experimental information of this type is lacking. Specifically, the longitudinal turbulence intensity and integral length scale are needed. The empirical estimates provided in this work are partially supported by the consistency found in the mass-transfer predictions.

2. TURBULENCE–INTERFACE INTERACTIONS

The role of surface tension in affecting the fluid motions within the "interfacial region" and, hence, the mass-transfer rates, remains uncertain. Two widely different, but yet partially successful, approaches exist. The first, due to Levich [1], is based on the concept of a surface tension damped laminar sublayer. The second, due to Fortescue and Pearson [2] and Banerjee *et al.* [3], is based on idealized eddy structures of turbulence unaffected by interfacial forces. These eddy models, themselves, are in difference. Fortescue and Pearson's large-eddy model assumes that energy-containing motions control the transfer process. Banerjee's small-eddy model assumes that the dissipating motions are controlling. In an attempt to better understand the nature of these differences the performance of all these models was investigated [4] against mass-transfer data obtained in a variety of flow systems: open-channel flow with and without turbulence producing grids [5], bubble flow [6], and jet flow [7]. A generalization of the eddy models was also proposed and found capable of a more coherent representation of the available experimental evidence. From this work, the recommended relationship between mass-transfer coefficient and local turbulence properties is broken down in two transfer regimes and may be summarized as follows:

$$k_L = k_L^D = C_{\mathrm{UET}}\sqrt{D}\left(\frac{V^3}{\nu L}\right)^{1/4} \quad \text{for} \quad Re_t > Re_t^* \qquad (1)$$

$$k_L = k_L^I = C_{\mathrm{IMA}}\,F(\alpha)\left(\frac{DV}{L}\right)^{1/2} \quad \text{for} \quad Re_t < Re_t^*. \qquad (2)$$

Where $F(\alpha)$ is given in Fig. 1 and the values of 0·25, 0·7 and 500 were recommended for the C_{UET}, C_{IMA} and Re_t^*, respectively. These relationships will be utilized here to describe mass transfer in the turbulent portions of the film.

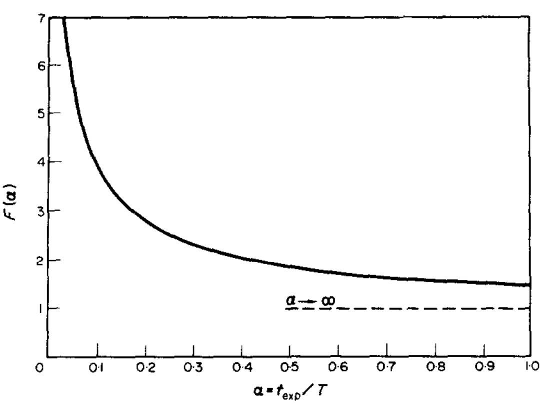

FIG. 1. The function $F(\alpha)$.

3. FILM FLOW CHARACTERISTICS

The recent work of Telles and Dukler [10] employed statistical methods to explore the structure of the gas–liquid interface of thin, vertical, falling, liquid films. The interface was shown to be random and to possess a relatively infrequent large-wave structure and a secondary pattern of small ripples. The large "lumps of liquid" were shown to carry a significant portion of the total flow and to move over the base film with no change in speed or shape. The ripples, however, lost their identity over short distances. From electrical conductivity measurements, time-varying, local film thickness records were obtained, and the resulting data on the probability distribution and spectral and cross-spectral densities were used to determine the average propagation velocity, C, average wave separation, λ_p, and the average wave profile. A representation of the falling film based on the structure of this "average" wave is given in Fig. 2. The thickness of the base film is m_L, and its bulk velocity is $\bar{V}$. The base of the large wave is L_w, its volume per unit width is V_w, and its mean height above the base film is h_w. Values of these wave parameters for film Reynolds numbers in the range of 1150–5750 may be found in the paper by Telles and Dukler [10].

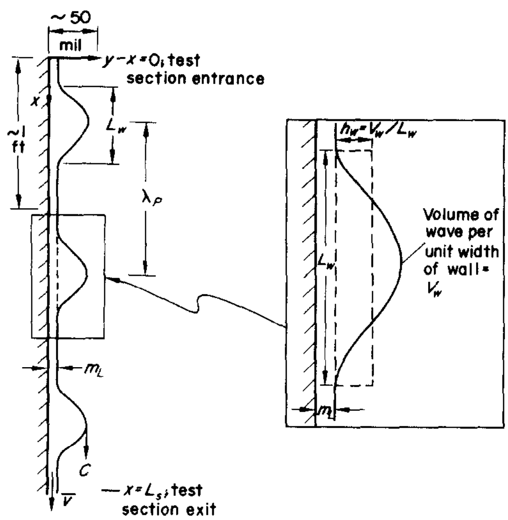

FIG. 2. Representation of a falling film according to Telles and Dukler [10].

The Reynolds number, Re_f, for the base film is $(1-\chi)Re$: for $1150 < Re < 5750$ we obtain $800 < Re_f < 1900$, indicating that the base film is essentially in laminar flow. For the waves, $Re_w = 4C(m_L+h_w)/\nu$, and $3000 < Re_w < 11\,000$, indicating turbulent flow. These aspects of film flow are also in agreement with Jackson [11] and Miya *et al.* [12]. These waves, or turbulent disturbances, are "flat," as graphically illustrated in Brauer's shadowgraphs [13].

For the velocity scale, V, needed in evaluating mass-transfer rates, the shear velocity, V_*, may be utilized. This choice was found satisfactory for the core region in pipe flow and for open-channel flow in the neighborhood of the free interface. For a film of thickness h flowing down a plate at an angle θ with the horizontal, the shear velocity is

$$V_* = (gh\sin\theta)^{1/2}. \qquad (3)$$

For the macroscale, the following empirical equation was formulated:

$$\frac{L}{h} = \frac{2000}{Re}. \tag{4}$$

The mass-transfer data of Davies and Warner [8] and Orridge [9] were obtained with films on inclined plates with large-scale roughness elements. The geometry is illustrated in Fig. 3. These films, flowing in nearly uniform-thickness turbulent flow in the space between the roughness elements, have quite a distinct overall hydrodynamic character from that of falling, vertical films. However, their turbulence structure should not differ greatly from that of the large waves of vertical films. In the absence of direct experimental information, the development of equation (4) was guided by these mass-transfer data. This equation will be shown to be equally capable of estimating the macroscale in the turbulent waves of vertical falling films, for the purpose of mass-transfer prediction.

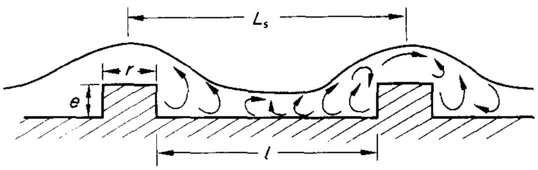

FIG. 3. Representation of turbulent film flow on a plate with large-scale roughness elements.

4. RESULTS AND DISCUSSION

For all film data considered, $Re_t \ll Re_t^*$, indicating that the $k_L = k_L^l$ regime is applicable. Equations (2)–(4) are utilized. Further details on the computations are given below.

For the data of Davies and Warner [8] and Orridge [9], the mean film thickness for each Reynolds number is obtained from the surface velocity, V_s, measurements given by these authors. Upon encountering a roughness element the film is assumed to be completely mixed. An appropriate exposure time for entrance (transient) effects is given by $t_{\exp} = L_S/V_S$. The results for both water and white spirit are compared with the experimental data in Fig. 4. The white spirit data do exhibit stratification and a lower sensitivity. Most likely this is due to its surface tension being about one-third that of water. However, even the surface tension-centred Levich theory results in a similar trend, the white spirit Levich constant being about one-half that required to fit the water data. In conclusion, entrance effects evaluated in the present model represent corrections in the range 3–25 per cent.

For the vertical falling film the classical data of Emmert and Pigford [14] and Kamei and Oishi [15] were analyzed together with the more recent data of Lamourelle and Sandall [16]. The total height $(m_L + h_w)$ of the turbulent wave and the wave Reynolds number were utilized in equations (3) and (4). The penetration theory with exposure time, $t_{\exp} = (\lambda_p - L_w)/(C - \bar{V})$, was utilized to estimate the absorption rate into the laminar

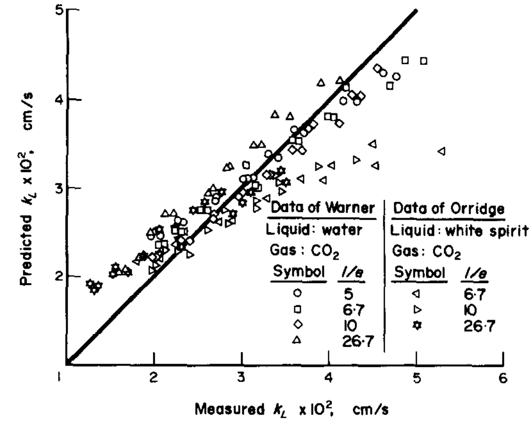

FIG. 4. Comparison of proposed model with experimental results of Davies and Warner [8] and Orridge [9].

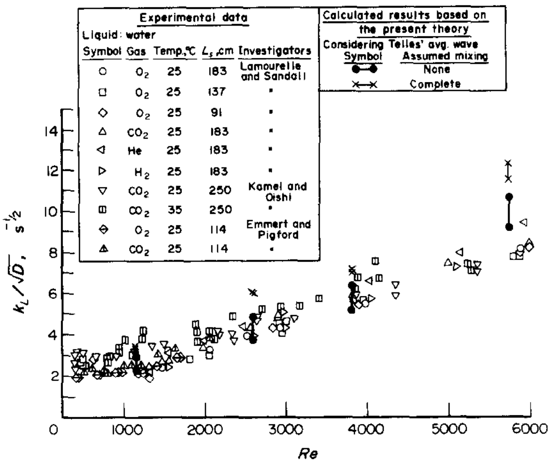

FIG. 5. Comparison of proposed model with experimental data for a turbulent film flowing on a smooth, vertical surface.

base film. Entrance effects for the turbulent waves were negligible. Mass-transfer calculations were carried out for four film Reynolds numbers, covering the range for which wave properties are given by Telles. A number of different gases and lengths of the film and the two limiting cases of no mixing and complete mixing between base film and waves are considered. Further details are given in the Appendix. The resulting mass-transfer coefficients are compared with experimental data in Fig. 5. For the no-mixing extreme, the calculated values of $k_L/\sqrt{D}$ showed some sensitivity to L_S and D. The whole range of these calculated values is indicated by the vertical bars shown in Fig. 5. Less sensitivity is observed for the complete mixing case. Furthermore, a mild sensitivity to the mixing assumption is noted. The no-mixing calculations seem to agree better with the data. The calculations show that the mass-transfer coefficients for the waves are of the order of five times that for the base film. With no mixing, the bulk concentration in the wave increases much more rapidly down the column than that in the film. A net effect of a smaller overall mass-transfer coefficient, as compared to the case of complete mixing, is thus produced.

5. CONCLUSIONS

A turbulence-centred model appears capable of describing mass-transfer rates at gas–liquid interfaces found in open-channel flows, bubble (pipe) flows, jet flows, and film flows. Surface tension effects are not likely to dominate the process. Yet, corrections for such effects would be desirable. Additional mass-transfer data covering wide ranges of surface tension and turbulence Reynolds number are needed for this purpose. The film hydrodynamics determined by Telles and Dukler, together with this mass-transfer model and an empirical estimate for the macroscale, provide a coherent description of a rather unique mass-transfer mechanism. Predictions are found to be in agreement with overall mass-transfer measurements.

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APPENDIX

Derivation of Mass-transfer Expressions for Falling Films

1. *No-mixing case*

The concentration of solute, c_w, in the wave as a function of position, x, along the absorption length is:

$$\frac{c_w(x)-c_s}{c_0-c_s} = \exp\left\{-\frac{L_w}{V_w}C_{\mathrm{IMA}}F\left(\frac{x}{CT}\right)\left[\sqrt{\left(\frac{DV}{L}\right)}\right]\frac{x}{C}\right\}. \quad (A.1)$$

For the base film we have:

$$\frac{c_f(x)-c_s}{c_0-c_s} = \exp\left\{-\frac{\lambda_p-L_w}{m_L\lambda_p}2\left[\sqrt{\left(\frac{D(C-\bar{V})}{\pi(\lambda_p-L_w)}\right)}\right]\frac{x}{\bar{V}}\right\}. \quad (A.2)$$

The cup-mixing solute concentration at the test-section exit, c_2, is related to the overall mass-transfer coefficient, k_L, by:

$$\frac{c_2-c_s}{c_0-c_s} = \exp\left\{-\frac{k_L L_S}{Q}\right\}. \quad (A.3)$$

But c_2 is related to c_w and c_f at the exit by:

$$c_2 = \chi c_w(L_S)+(1-\chi)c_f(L_S). \quad (A.4)$$

Hence:

$$k_L = -\frac{Q}{L_S}\ln\left\{\chi\exp\left[-\frac{L_S L_w C_{\mathrm{IMA}}}{CV_w}\sqrt{\left(\frac{DV}{L}\right)}F(\alpha)\right] +(1-\chi)\exp\left[-\frac{k_f(\lambda_p-L_w)L_S}{m_L\lambda_p\bar{V}}\right]\right\} \quad (A.5)$$

with:

$$\alpha = \frac{\sqrt{(2)}VL_S}{LC}.$$

2. *Complete mixing case*

A derivation similar to the previous case yields the following expression for the overall mass-transfer coefficient:

$$k_L = -\frac{Q}{L_S}\ln\left\{\chi\exp\left[-\frac{L_w L_S C_{\mathrm{IMA}}}{B_1 C}\left[\sqrt{\left(\frac{DV}{L}\right)}\right]F(\alpha) -\frac{B_2 L_S}{B_1 C}\right] + (1-\chi)\exp\left[-\frac{L_w C_{\mathrm{IMA}}}{B_1}\left[\sqrt{\left(\frac{DV}{L}\right)}\right] \times\left(\frac{L_S}{C}-\tau\right)F\left(\alpha-\frac{\tau}{T}\right)-\frac{B_2}{B_1}\left(\frac{L_S}{C}-\tau\right)-\frac{k_f t_{\mathrm{exp}}}{m_L}\right]\right\} \quad (A.6)$$

with:

$$B_1 = V_w+m_L L_w+(C-\bar{V})m_L\tau\exp\left(-\frac{k_f t_{\mathrm{exp}}}{m_L}\right)$$

$$B_2 = (C-\bar{V})m_L\left[1-\exp\left(-\frac{k_f t_{\mathrm{exp}}}{m_L}\right)\right]$$

$$\tau = \frac{\bar{V}t_{\mathrm{exp}}-L_w}{C}, \quad k_f = 2\sqrt{\left(\frac{D}{\pi t_{\mathrm{exp}}}\right)},$$

$$\alpha = \frac{(\sqrt{2})VL_S}{LC}.$$

TRANSFERT DE MASSE TURBULENT AUX INTERFACES LIBRES LIQUIDE–GAZ ET APPLICATION AUX ECOULEMENTS EN FILM

Résumé—L'hydrodynamique du film déterminée par Telles et Dukler, jointe à un modèle de transfert de masse turbulent et à une estimation empirique de l'échelle intégrale, fournit une description cohérente d'un mécanisme de transfert de masse en accord avec les mesures globales de transfert de masse.

TURBULENTER STOFFÜBERGANG AN FREIEN GAS-FLÜSSIGKEITS-TRENNFLÄCHEN MIT ANWENDUNG AUF FILMSTRÖMUNGEN

Zusammenfassung—Die von Telles und Dukler bestimmte Filmhydrodynamik ermöglichte in Verbindung mit einem turbulenten Stoffübergangsmodell und einer Abschätzung der Größenordnung eine einheitliche Beschreibung eines ziemlich einzigartigen Stoffübertragungsmechanismus. Übereinstimmung mit Gesamt-Stoffübertragungsmessungen kann nachgewiesen werden.

ТУРБУЛЕНТНЫЙ ПЕРЕНОС МАССЫ НА ГРАНИЦАХ РАЗДЕЛА ГАЗА И ЖИДКОСТИ ПРИ ПЛЕНОЧНОМ ТЕЧЕНИИ

Аннотация — Гидродинамическая трактовка пленочного течения Теллеза и Даклера вместе с моделью турбулентного переноса массы и эмпирической оценкой макромасштаба позволяет дать описание довольно уникального механизма переноса массы, которое согласуется с экспериментом.