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A numerical study of interfacial transport to a gas-sheared wavy liquid

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Abstract—Interfacial transport of a passive scalar from a gas to a thin liquid film is considered. The base flow in the liquid is laminar, with linear velocity profile produced by gas shear. The velocity disturbances imposed by waves are simulated by an inviscid, constant-vorticity model. Numerically computed roll waves are shown to enhance interfacial transport by modifying the convection pattern below the crest and by inducing a mixing effect on the substrate. It is argued that the entire wave spectrum is active in this enhancement of interfacial transport. © 1998 Elsevier Science Ltd. All rights reserved.

1. INTRODUCTION

Transport processes across gas–liquid interfaces are of considerable significance in engineering applications as well as in natural flows. An example-involving mass transfer—is the dissolution of a sparingly soluble gas into an unsaturated liquid, which has applications in the design of absorption equipment for the chemical process industry and for the abatement of gaseous pollutants. Another example is the direct-contact condensation of a pure vapor on a subcooled liquid, a problem which is of interest in nuclear reactor cooling and in the design of efficient heat-transfer equipment.

One characteristic shared by the above examples is that, in both cases, the transfer rate is controlled by the resistance in the liquid. With respect to this kind of problem, a survey of the literature indicates two things:

- Similar behavior has been observed for the phenomena of heat and mass transfer. As noted by Back and McCready [1], a correlation summarizing the data of many investigations of mass-transfer in cocurrent gas–liquid flow is remarkably similar to the correlation of the data of Jensen and Yuen [2] for steam condensation on subcooled liquid. This result is surprising since the Sc number is ordinarily two orders of magnitude greater than the Pr .
- Although various correlations have been proposed and are used for predicting interfacial transport rates [3, 4], a fundamental understanding of the transport mechanism is presently lacking. This holds true even in the simplest case of a flat and

shear-free horizontal interface and is attributed to our incomplete knowledge of the flow structure near a gas–liquid interface [5, 6].

A major complication arises from the deformable nature of the interface, which—under moderate gas shear—leads to the formation of wave patterns. Experiments have repeatedly demonstrated [7–9] that the occurrence of waves causes dramatic increase in interfacial transfer rates. However, despite extensive theoretical and modelling efforts, a satisfactory explanation of the role of waves is not presently available. There are two major trends among the theories put forward in recent years, one stressing the roll of waves—either capillary [8, 10] or the entire spectrum [9]—and the other the role of turbulence intensity below the interface [6, 11]. The underlying question is whether there exists a unique set of variables suitable for parameterizing the interfacial transport process. Of course, waves and turbulence are not independent entities. They exchange and re-distribute the wind energy—by interacting with each other and with the mean flow—through mechanisms which are poorly understood and are also the subject of intense investigation [12].

Experimental evidence in favour of a distinct role for wave-induced fluctuations in the transfer process has recently been provided by Wolff and Hanratty [13]. They used a laser fluorescence technique to measure local instantaneous rates of oxygen absorption in cocurrent air–water flows. Their measured time-variation in the concentration gradient normal to the interface is too striking to be consistent with the notion that absorption is governed by turbulence.

The role of waves in interfacial transport has also been investigated by a combination of numerical simulation and experimental data. Representative modelling efforts in this direction are the numerical studies of Back and McCready [1] and Wasden and

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NOMENCLATURE

a	wave amplitude
amp	dimensionless wave amplitude
c	passive scalar concentration
d	liquid depth
D	molecular diffusivity
k	wavenumber
L	wavelength
Pr	Prandtl number
r	ratio of gas to liquid density
Sc	Schmidt number
u, v	components of velocity field in the liquid
U	gas velocity

V wave phase velocity.

Greek symbols

ζ	liquid vorticity
$\eta(x)$	shape of the interface
ρ	liquid density
σ	surface tension.

Subscripts

flat	referring to the flat-film solution
ref	in the stationary reference frame
s	at the interface.

Dukler [14]. The first authors predict the velocity field below the waves of a certain experimental realization by using wave spectra measurements and solutions of the Orr–Sommerfeld equation. The last authors numerically calculate the velocity field for a particular, experimentally determined, wave shape and phase velocity. Recently, Jayanti and Hewitt [22] used a CFD code to gain insight into the effect of disturbance waves on heat transfer.

A complementary approach in the investigation of the role of waves is to perform numerical simulations using a laminar wavy flow field. Thus, turbulence is artificially excluded from the phenomenon. What is sought, is to examine the conditions under which waves lead to significant enhancement of interfacial transport rates and the extent of the enhancement. This is the goal of the present work.

Inviscid waves are considered, on a liquid film with constant vorticity. This hydrodynamic model—which corresponds to a base flow with linear velocity profile—is described briefly in the next section. An interesting feature of the model is that—for high enough gas velocities—it predicts roll waves, that is waves with a recirculating closed region below the crest [15]. Roll waves are suspected to play a key roll in the enhancement of mass-transfer [14].

The present work represents an attempt to assess the applicability of the aforementioned model to interfacial transport computations. What is sought, is the construction of a computational tool and a systematic examination of the effect of various parameters pertinent to the problem. Some preliminary results of this effort have already been reported elsewhere [16].

2. THE HYDRODYNAMIC MODEL

The model of the wave-induced flow field in the liquid, which is used in the interfacial transport computation, is an inviscid solution for a liquid with con-

stant vorticity, sheared by cocurrent gas flow [15]. The parameters pertinent to this model are the uniform gas velocity, U , the vorticity, ζ , of the liquid base flow, the depth, d , of the liquid film and the wave length, L , and amplitude, a , of the wave. This flow is sketched in Fig. 1, and—with the exception of the nonzero liquid vorticity—represents the classical Kelvin–Helmholtz configuration.

The model corresponds to a laminar base flow with linear velocity profile and is thus, appropriate when considering thin films. It is also a useful model of the drift layer of a deep liquid, when waves are short enough that the vorticity distribution they experience can be satisfactorily approximated by the surface value [17].

Inviscid flows with initially uniform vorticity distribution are amenable to simplified analysis because Kelvin's circulation theorem guarantees that the vorticity will always remain constant [18]. In physical terms, the constant velocity model implies that the only effect of liquid viscosity is in determining the base flow profile. The disturbances of the flow field induced

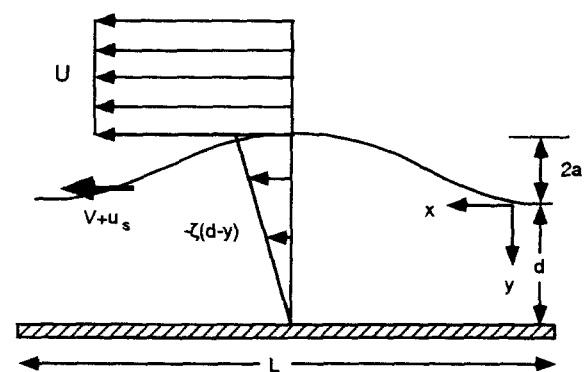


Fig. 1. Sketch of the flow with all the pertinent parameters.

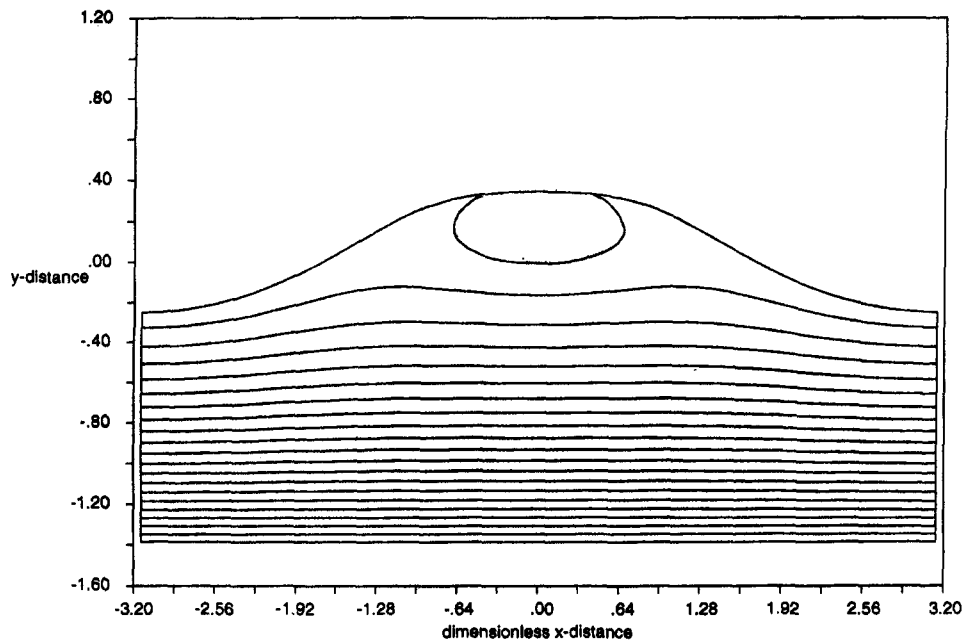


Fig. 2. Typical streamline pattern of an inviscid roll wave. Parameters used in this computation are $L = 0.0628$ m, $a = 0.003$ m, $d = 0.0109$ m, $U = 7.83$ m s⁻¹ and $\zeta = 31.3$ s⁻¹.

by waves have a timescale which is typically much smaller than the timescale associated with the diffusion of vorticity. Therefore, they are treated as essentially inviscid.

An important finding of the above wave model is that, in a reference frame stationary with respect to the wave, the flow field calculated for high gas velocities contains a recirculating eddy below the crest. A typical streamline pattern, corresponding to this case, is shown in Fig. 2. It is interesting to note that a crest eddy has been invoked to model other vorticity-dominated flows, such as turbulent free-falling films [19].

For a constant gas velocity, the closed flow region appears above a certain wave height, and further expands with increasing wave height. The amplitude corresponding to the onset of the phenomenon can be described to first order by the equation [20],

$$a_r = \frac{C_l}{XkC_l - \zeta} \quad (1)$$

where a_r is the amplitude at which closed streamlines first appear, C_l is the linear wave celerity and X is related to the film depth through the expression,

$$X = \frac{1 + \exp(-2kd)}{1 - \exp(-2kd)} \quad (2)$$

The recirculating eddy appears earlier for higher gas velocities and is present in all waves, irrespective of amplitude, for U greater than critical, given by the expression

$$U_{\text{roll}} = \left[\frac{g(1-r) + \sigma k/\rho}{r} \right]^{1/2} \quad (3)$$

Term ρ is the liquid density, r is the gas-to-liquid density ratio and σ is the surface tension.

The aforementioned inviscid solution has certain similarities with experimentally observed roll waves. In particular, the closed eddy is calculated only above a certain, high gas velocity. Also, the circulating fluid moves on a substrate whose velocity field does not significantly deviate from the base flow. Thus, periodic, roll waves are similar to a series of solitary waves.

The solution differs from experimental observations in other aspects, such as the lack of asymmetry in the wave shape. It is speculated that the reason for the appearance of a symmetric (about the crest) circulation zone is the neglect of interfacial shear which would cause the wave to tumble.

We are interested in testing the predictions of this hydrodynamic model with reference to the problem of interfacial transport into liquid films. To this end, the velocity field calculated from the weakly nonlinear solution of the problem [20] will be used in the numerical solution of the transport equation. The weakly nonlinear flow field was preferred from the fully nonlinear numerical results, because the analytical expressions provide greater flexibility. It is evident that, with increasing wave height, the transfer rates computed become less accurate. However, they are deemed sufficiently accurate for useful qualitative conclusions to be drawn.

The physical mechanism investigated involves a wave disturbance, which 'rides' on low-concentration fluid producing a local change in the diffusion process, and then moves downstream to the next portion of

fluid. The hydrodynamic model used involves steady, periodic waves. To this end, the velocity field at the wave trough is assumed to correspond to the undisturbed base flow. This assumption is not strictly correct in general. However, it seems satisfactory for the special case of roll waves considered in the present work. Indeed, as can be observed from Fig. 2, the flow pattern below the wave trough is very similar to the undisturbed base flow.

3. THE TRANSPORT MODEL

3.1. The steady formulation

Definition of the appropriate problem setup for a meaningful simulation of the role of waves in interfacial transport is not trivial. Heat/mass transfer is described in the general case by the two-dimensional convection–diffusion equation,

$$\frac{\partial c}{\partial t} + u \frac{\partial c}{\partial x} + v \frac{\partial c}{\partial y} = D \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} \right). \quad (4)$$

When considering progressive waves of permanent form riding on the flat film, a reference frame moving with the wave celerity can be used to render the flow field steady (frozen field hypothesis). Modelling of the flow field by progressive waves of permanent form seems like a reasonable choice, in particular when the parametric dependence of the transport process on wave properties is investigated. The additional assumption that the passive scalar field is, in such a reference frame, also steady is much more drastic and open to question. If such an approximation is invoked, it is described by the equation

$$\frac{\partial c}{\partial t} = -V \frac{\partial c}{\partial x} \quad (5)$$

where V is the reference velocity rendering the wave motionless. Equation (5) has been used by Wasden and Dukler [14], in their work aiming at the numerical calculation of mass transfer to a free-falling liquid film.

The constant concentration approximation has certain limitations. For a wave moving on a fully developed film, and in the steady reference frame, the liquid entering the waveform from downstream has a concentration distribution dictated by the flat film solution. Constant concentration under the wave cannot be reached, unless this entering distribution remains constant for the time needed for steady-state to develop. Thus, equation (5) implies that the relaxation time of the concentration field below the wave is much smaller than the time necessary to observe significant change in the flat-film concentration, when travelling downstream with the wave velocity. The numerical computations presented next, support the argument that this assumption is not generally justified.

Furthermore, by assuming zero mass accumulation

within the wave, equation (5) sets the transfer rate through the interface equal to the rate of removal from the back of the wave. Thus, the enhancement of mass-transfer is governed by the convection ability of the flat film following the wave. It is reasonable to expect that this assumption represents a lower limit. Because of the limitations of the steady-state assumption, an unsteady formulation will be used in the present work.

3.2. The unsteady formulation

In the present work, the passive scalar field is treated as unsteady in the reference frame moving with the wave celerity. The transport problem simulated in the study can be illustrated by again referring to Fig. 1. In the reference frame rendering the flow stationary, the liquid is entering the wave from the front side and leaving from the back. The wave is considered to appear at the entrance of the gas–liquid contact section and to ride on a fully developed flat film. Therefore, the inlet condition to the wave is provided by the concentration profile of the base flow, at the position where the wave is located. This determination of the boundary condition at the wave front introduces a time dependence of the transport process, as the concentration profile in the base flow varies continuously with distance downstream.

The flat-film concentration profile is calculated numerically by solving equation (4), subject to the following initial and boundary conditions for the scalar concentration $c(x, y, t)$:

$$c(x, y, 0) = 0 \quad \text{for all } x, y \quad (6)$$

$$c(0, y, t) = 0 \quad \text{for all } y, t \quad (7)$$

$$\frac{\partial c}{\partial y} = 0 \quad \text{at } y = d \quad \text{for all } x, t \quad (8)$$

$$c(x, 0, t) = c_s. \quad (9)$$

Position $y = 0$ is the surface of the liquid film and c_s is the saturation concentration. The solution is followed in time until steady-state is achieved. An independent check of accuracy of this solution was provided by the satisfaction of the integral mass balance (less than 0.1% deviation). Values of the flat-film concentration distribution at intermediate distances are computed by three-point Lagrangian interpolation of the above numerical solution.

For the wavy flow, equation (4) is solved using the steady-state velocity field computed from the hydrodynamic model and subject to the following boundary and initial conditions. At the solid boundary, $y = d$, the no-flux condition—equation (8)—is again invoked. The interface concentration is set by the saturation condition

$$c(x, \eta, t) = c_s \quad \text{at } y = \eta(x) \quad (10)$$

where $y = \eta(x)$ describes the interface in the stationary reference frame. At the outlet, $x = L$, behind the

wave, the curvature of the streamwise concentration distribution is taken equal to zero.

$$\left(\frac{\partial^2 c}{\partial x^2}\right)(x=L) = 0 \quad \text{for all } y. \quad (11)$$

The impact of condition (11) was tested by including a variable section of the flat film behind the wave in the computational domain. It was indeed verified that the concentration distribution under the wave crest is insensitive to changes in this tail length and that equation (11) behaves better than the alternative boundary condition that sets the slope of the streamwise concentration equal to zero.

The inlet condition at $x=0$ is dictated by the flat film profile. As noted above, this condition links—through the frozen field hypothesis—the characteristic time of the convection process to the streamwise variation of the flat-film profile. This time-varying inlet boundary condition, which represents a novel feature of the present computations, is implemented in the following way: the reference frame rendering the wave stationary is moving with a horizontal velocity U_{ref} , given by

$$U_{\text{ref}} = V + u_s \quad (12)$$

where V is the wave phase velocity and $u_s = \zeta d$ is the base flow velocity at the interface. The inlet condition to the wave at time t is given by the steady state, flat-film solution at a distance x from the edge of the absorption section

$$c(0, y, t) = c_{\text{nat}}(x, y) \quad (13)$$

where

$$x = U_{\text{ref}} t. \quad (14)$$

Particular attention is paid to the accurate implementation of the interfacial boundary conditions near the entrance section. At that region, the interface condition, $c = c_s$, is applied only to the part of the wave surface which, at the given time, has appeared downstream from the entrance.

The consistent implementation of this time-dependent condition was checked by performing computations for a wave of zero amplitude (linear wave). The solution thus generated corresponds to the flat-film concentration profile, scanned with a horizontal velocity $U_{\text{ref}} = V_0 + u_s$. (V_0 is the phase velocity of the linear wave.) The concentration profiles calculated in this way by the unsteady code were found to agree to three decimal figures with the original flat film distribution.

The solution of the transport equation (4) is generated by an explicit finite-difference algorithm. An orthonormal, curvilinear coordinate system is used, which conforms to the wave profile at the liquid surface and to a flat line at the bottom. The coordinates (ξ, η) are the stream function and velocity potential of the irrotational flow with the same wave profile. Evidently, these terms do not have any physical sig-

nificance; they are just convenient tools because they describe the flow domain in a smooth way and they automatically satisfy the Cauchy–Riemann conditions.

Grid refinement studies showed that a grid mesh of 40×40 provides sufficient detail for transport modelling. Runs were executed on an Apollo 9000 HP/735 workstation.

4. RESULTS AND DISCUSSION

The main goal of the present work is to investigate the effect of the roll wave transition on the intensity of the interfacial transfer process. Therefore, no attempt is made to cover the multi-dimensional parameter space relevant to the problem. The numerical results presented are limited to a single value of film thickness, wavelength and gas velocity. The first-order estimates, provided by equations (1)–(3), are used to investigate the effect of wavelength and gas velocity on the predicted phenomena.

The wave related parameters are made dimensionless using the inverse wavenumber k^{-1} as the characteristic length and $(g/k)^{1/2}$ as the characteristic velocity. For example, the dimensionless wave amplitude, amp , is defined as $amp = ka$. For a wavelength of 0.0628 m—representative of the region where both gravity and surface tension are significant—the dimensional variables that have been used in the present simulation are, liquid depth $d = 0.011$ m, gas velocity $U = 7.83$ m s⁻¹, vorticity $\zeta = 31.3$ s⁻¹ and wave amplitudes 0.001, 0.0015 and 0.003 m.

It should be noted that inviscid theory treats gas velocity and liquid vorticity as independent input parameters. However, when liquid vorticity is induced by gas shear, the two variables are related through the interfacial stress balance. The above given values correspond to a laminar liquid with viscosity equal to 0.018 kg m s⁻¹ (18 cp).

The Pr in this computation has the value $Pr = 5.4$ and is more relevant to heat than to mass transfer. However, as noted in the introduction, the interfacial transport of heat and mass seem to follow similar behaviour and the present results are expected to provide useful insights for both processes.

The main result of the simulation is the concentration distribution in the liquid. From this, the interfacial transport flux and other parameters of interest can be calculated. The overall effect of waves in the transport process is quantified by an enhancement term, which is defined as the mean flux below the wave divided by the corresponding value for the flat substrate. The variation of the wave enhancement as the wave moves downstream is presented for three different waves in Fig. 3. The two lower curves represent waves where the recirculating region either does not exist ($amp = 0.10$) or is starting to appear ($amp = 0.15$). The upper curve corresponds to a wave with a well developed closed eddy. It is evident that the roll wave is the only one which provides significant

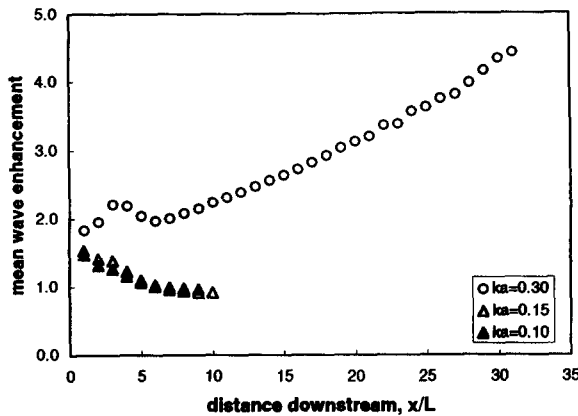


Fig. 3. The average wave-induced enhancement of the transport rate, as the wave moves downstream from the entrance.

enhancement of the transfer rate (actually, the two other waves appear to have a negative effect on interfacial transport after a distance of 6–7 wavelengths downstream). Furthermore, the enhancement steadily increases with distance downstream, at least for the distance of thirty wavelengths considered in the computation. The above observations support the notion that roll waves play a distinct role in interfacial transport.

To investigate the mechanisms through which roll waves influence the transport process, the evolution of the concentration distribution in the liquid is examined in detail. Concentration patterns below the roll wave ($ka = 0.30$) are presented in Figs. 4(a)–(d), corresponding to the wave located at a distance equal to L , $2L$, $5L$ and $10L$ from the entrance, respectively. Two regions of high interfacial flux are identified along the free surface: one is near the back stagnation point and the other somewhat above the front stagnation point. The front stagnation point itself defines a region of high-concentration fluid which is entrained by the flow towards the interior of the liquid. One striking characteristic is the creation and persistence of a low-concentration region in the recirculating zone.

The physical mechanism which emerges from these pictures involves the engulfment of clear liquid by the crest eddy during the initial stages of the wave travel [Fig. 4(a)–(b)]. A low-concentration region is thus created and provides the driving force for enhanced interfacial transport. This structure seems to persist because the recirculating motion induces a convection pattern leading to the entrainment of high-concentration liquid from the wave front, below the eddy, and towards the interior. The active region associated with the back stagnation point become gradually elongated by the flow, tentatively contributing to increased transport rates at the wave tail.

The above described effect of the higher wave has strong qualitative similarities with the results com-

puted by Wasden and Dukler [14] for roll waves riding on a free-falling film. As was concluded by these authors, the increase in interfacial transport results from the interaction of the roll wave with the slowly moving substrate. It is interesting to note that, despite the difference in the orientation of the flows, the two problems share a common feature. They involve vorticity in the liquid, which is created in one case by the gas shear and in the other by the effect of gravity. Therefore, it could be argued that liquid vorticity is an appropriate variable for parameterizing interfacial transport in thin films.

It is of interest to consider the condition at which the substrate film is left after the passage of a wave. To this end, the local transport rate (based on the concentration distribution right after the passage of a wave) is computed and compared to the steady-state transport rate of the flat film at the same location. The results are presented—again in terms of an enhancement factor—in Fig. 5. It is noted that all the waves have a nonzero effect. However, the effect of the small (nonrolling) waves is limited to the entrance section and diminishes after a distance of a few wavelengths. On the contrary, the enhancement provided by the roll wave is distinctively higher and increases with distance downstream. This result leads to the notion that roll waves may contribute to higher interfacial transport also through a mixing effect imposed on the substrate. It should be clear that the results of Fig. 5 correspond to ‘instantaneous’ enhancement at a certain location, right after the passage of the wave. The long-term persistence of this phenomenon is indicated by the computed result that roughly 20% of this enhancement remains after the wave has moved away from the location a distance equal to one wavelength. The above computations indicate that the substrate mixing mechanism—though not dominant—is not negligible.

One final investigation concerns the significance of the size of the waves. The discussion so far is based on numerical results for a wave of length equal to 0.0628 m, that is a wave influenced by both gravity and surface tension. In view of the different theories that have appeared in the literature (see Introduction), it is of interest to consider whether the present approach argues in favour of a distinct role for a certain region of the wave spectrum. Given the significance of the roll-wave transition, what we need to know is whether roll waves preferably develop only from short or long waves. To this end, equation (1)—which is an estimate of how high waves need to get to develop a recirculating flow structure—is used. Results are presented in Fig. 6 for a water layer, 1 cm thick, sheared by a gas velocity $U = 3 \text{ m s}^{-1}$. It turns out, however, that the effect of gas velocity is minor and the results are valid for all subcritical gas velocities.

A qualitative conclusion is that roll waves are equally possible throughout the entire wave spectrum. According to Fig. 6, capillary waves need to reach higher steepness for the recirculating eddy to develop.

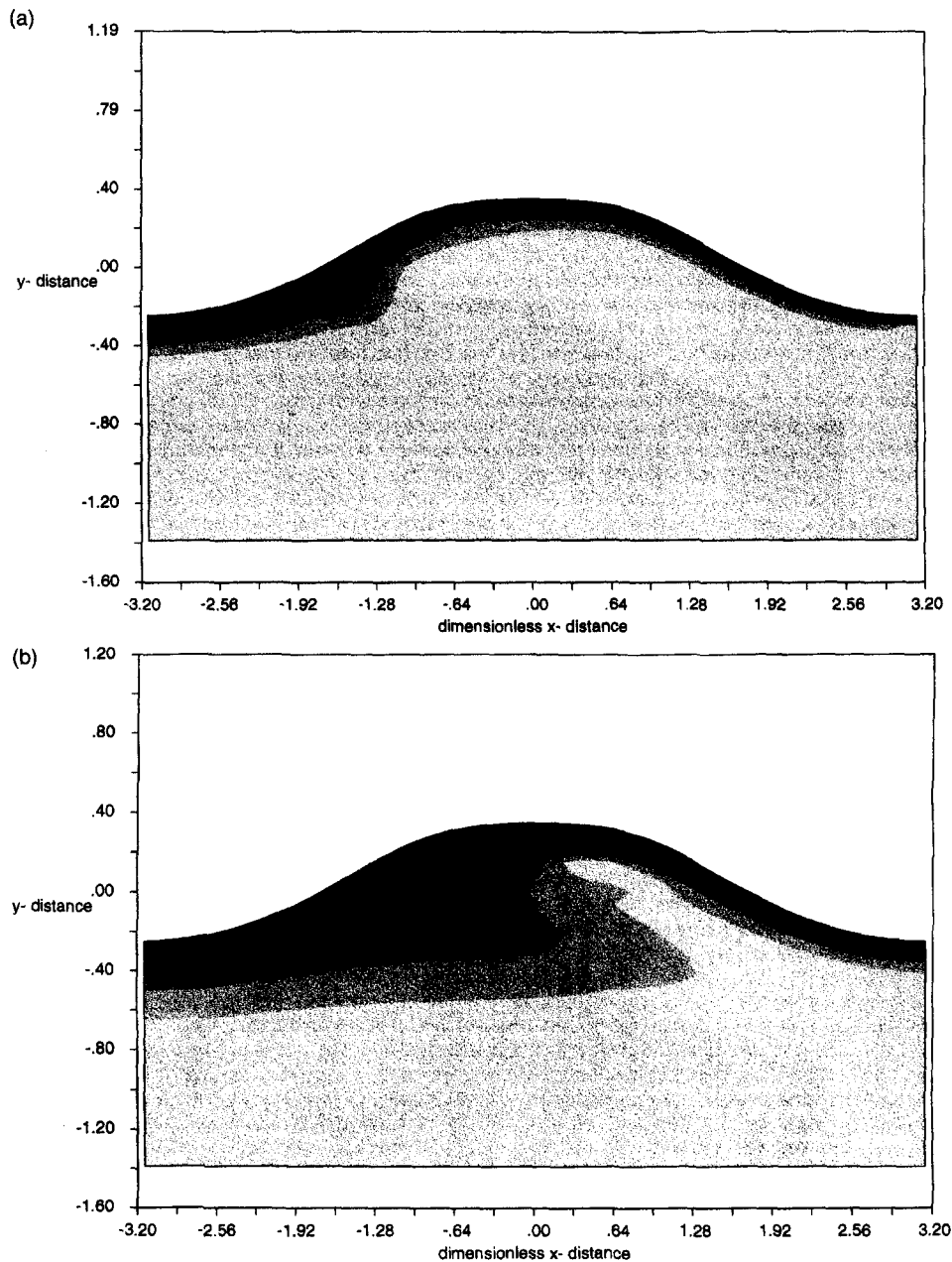


Fig. 4. (a)–(d) Iso-concentration contours below the wave, at distances L , $2L$, $5L$ and $10L$ away from the entrance.

However, it is well documented (by experimental observations and computation) that capillary waves are steeper than their gravity counterparts roughly by the order of magnitude appearing in the results of Fig. 6. Therefore, according to the present model, the entire wave spectrum is responsible for the enhancement in interfacial transport. This conclusion is in agreement with the findings of Jähne *et al.* [9], who best correlated their mass transfer measurements with the mean, rather than the capillary, wave steepness.

5. CONCLUSIONS

Interfacial transport to a wavy liquid film with linear base velocity profile, produced by gas shear, is considered. The problem is formulated in a reference frame moving with the wave celerity, using a steady velocity profile but a time-dependent concentration. It is argued that this setup is useful in the description of the interaction of progressive waves of permanent form with a fully developed flat substrate.

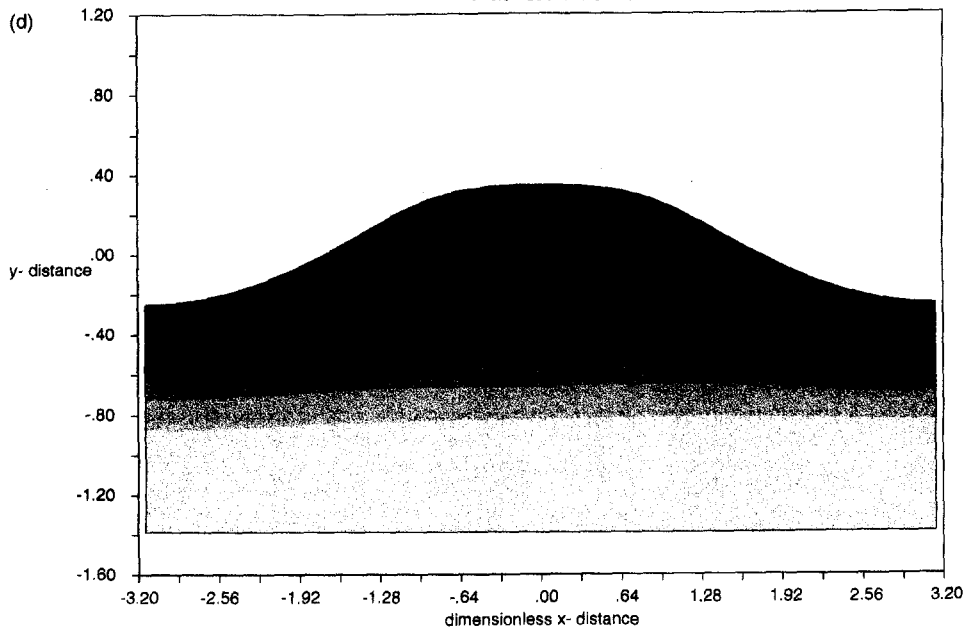
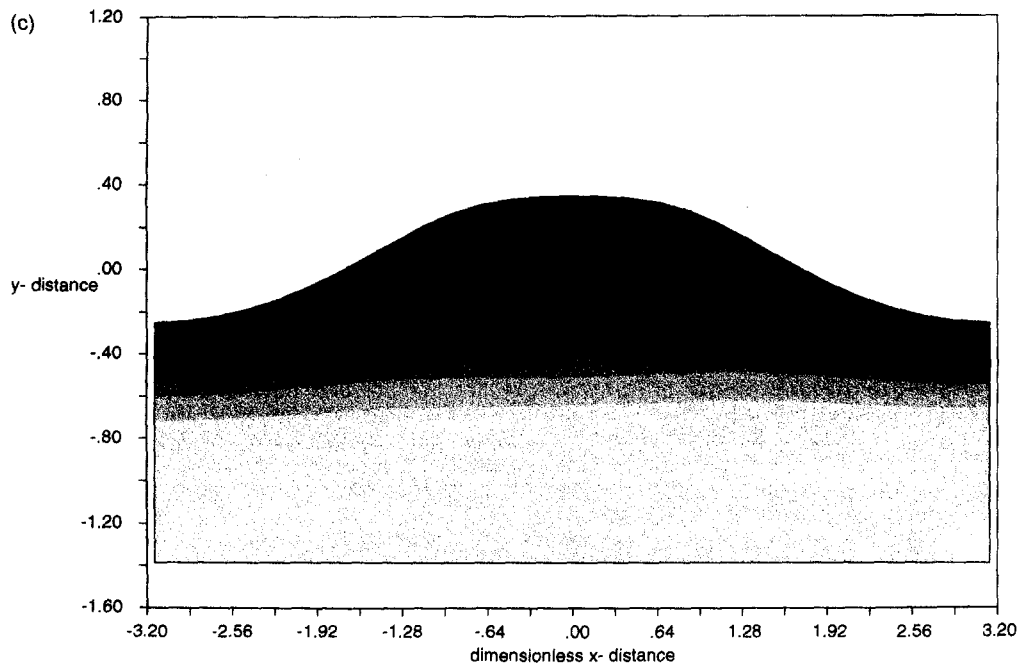


Fig. 4—continued.

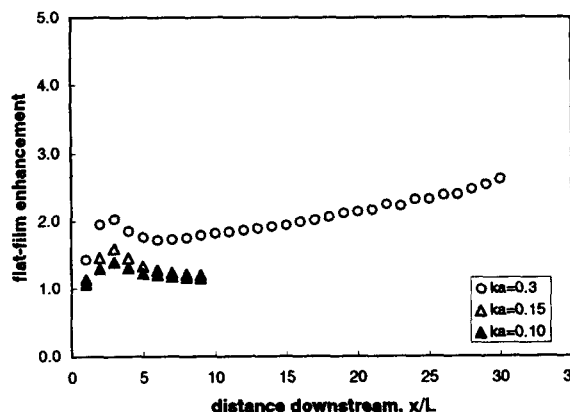


Fig. 5. The enhancement in the flat-film transport rate, right after the passage of a wave, as a function of distance downstream from the entrance.

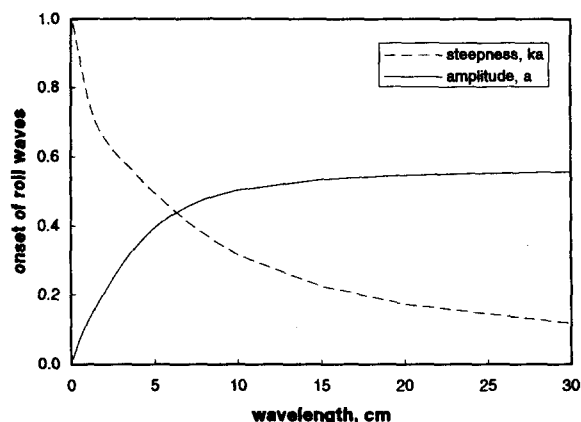


Fig. 6. First-order estimate of the wave amplitude and steepness at which roll waves appear.

The numerical results indicate that roll waves drastically alter the interfacial transport rate. This is attributed to the convection patterns generated by the recirculating eddy below the crest, but also to the mixing induced on the substrate by the passage of the wave. It is finally argued that the entire wave spectrum is active in the enhancement of interfacial transport. The present approach, though of limited quantitative value at this stage, could contribute to the development of improved conceptual models of interfacial transport processes.

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