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Surface divergence models for scalar exchange between turbulent streams ☆

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Abstract

Surface divergence models for prediction of scalar exchange at fluid–fluid interfaces are investigated. The models, based on the Hunt–Graham blocking theory, are shown to predict experimental data at unsheared interfaces, and new results of direct numerical simulation for deformable, nonbreaking sheared interfaces. The parameterization is in terms of the turbulent Reynolds number defined by the integral velocity and length scales in the bulk flow, which makes it useful for practical purposes.

Keywords: Deformable interfaces; Surface divergence; Gas transfer; DNS

1. Introduction

Scalar exchange between turbulent streams separated by a deformable fluid–fluid interface plays an important role in the performance of equipment like evaporators, condensers, gas–liquid and liquid–liquid contactors, and in environmental systems. There has been an intense and renewed interest in the subject due to its central role in the uptake of greenhouse gases and release of moisture by terrestrial water bodies. To put the greenhouse gas uptake problem in context, it

☆ This paper is dedicated to Prof. George Yadigaroglu on the occasion of his 65th birthday. George is truly a renaissance man having worked (and lived) with distinction in a number of milieus. This paper relates to one of his most recent interests, viz. direct numerical simulations of multiphase systems—particularly of interphase transport processes. As all the authors have interacted with him closely in parts of this work, we feel it appropriate to contribute a paper in a field which has benefited so greatly from his presence.

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should be noted that approximately 30–40% of man-made CO_2 (the most persistent greenhouse gas) is taken up by the oceans. However, the uncertainty in the correlations used to estimate CO_2 uptake is such that they range from 1.1 PgC/year (Liss and Merlivat, 1986, correlation) to 3.3 PgC/year (Wanninkhof and McGillis, 1999, correlation) according to Donelan and Wanninkhof (2002).

Clearly, there are considerable incentives to improve our understanding of scalar exchange phenomena and reduce such uncertainties, which have major impacts on policy, related, for example, to the utilization of fossil fuels. Gas exchange problems also occur in numerous other environmental settings, such as desorption of dissolved substances, like PCBs, from inland and coastal water bodies, that can be of significant air quality concern.

Be that as it may, a comprehensive review of turbulence and scalar exchange is available in Banerjee and MacIntyre (in press), hence-forward called BM, and should be referred to for a discussion of the published literature. The purpose of this paper is to focus on a particular scalar exchange model, viz. the surface divergence model of Banerjee (1990). This is also briefly discussed in the BM review, which considered its application to a limited set of laboratory and field data, but not the direct numerical simulations (DNS) discussed here.

We will proceed as follows. First, we will briefly review the so-called surface divergence models and their derivation from the Hunt and Graham (1978) "blocking" theory. Second, we will review applications of these models to laboratory data for scalar exchange across unsheared gas–liquid interfaces (also done in BM) to set the stage for what follows. Third, we will consider their application to recent DNS of coupled gas–liquid turbulence and scalar exchange across deformable surfaces (De Angelis, 1998; Fulgosi et al., 2003; Lakehal et al., 2003). New direct numerical simulations performed by the ETH Zurich group with variations in shear velocities have been used to validate the models.

The DNS allows direct calculation of the surface divergence field, which was predicted on the basis of the Hunt and Graham (1978) "blocking" theory by Banerjee (1990). The DNS data also allows calculation of the relationship between scalar exchange and the surface divergence field, and thus tests the exchange model directly. The direct simulation studies referred to here consider situations with a range of gas shear at the surface, with turbulence being generated at the interface itself, rather than elsewhere, e.g. at the bottom boundary of the flowing stream.

The flow configuration studied in these simulations have previously helped clarify the transfer mechanisms at deformable interfaces (De Angelis, 1998; Fulgosi et al., 2003; Lakehal et al., 2003). In these references the waves fall within the gravity-capillary range, with waveslope $ak = 0.01$ (wave amplitude a times wavenumber k), and very small phase-speed to friction-velocity ratio, $C/u_\star$. In the new simulations presented here, the friction velocities, $u_\star$, were considerably increased so as to generate surface deformations of higher waveslopes (up to $ak = 0.12$), but without leading to wave breaking.

2. Surface divergence (SD) models

2.1. The basic idea

The surface divergence model is now briefly discussed before making comparisons with experiments and DNS. Banerjee (1990) derived a general form of the expression for the mass transfer coefficient for the case where there is no gas shear at the interface and the far-field turbulence is homogeneous and isotropic, based on the blocking theory of Hunt and Graham (1978). Using a result from McCready et al. (1986), Banerjee (1990) showed that for unsheared interfaces at which high Sc gas transfer occurs, the gas transfer velocity, β, is given by

$$\frac{\beta Sc^{1/2}}{u} \approx Re_t^{-1/2}\left[\left(\frac{\partial u'}{\partial x}+\frac{\partial v'}{\partial y}\right)^2\right]_{\rm int}^{1/4} \tag{1}$$

where the subscript "int" denotes the interface, and all quantities on the RHS of (1) have been normalized by u and Λ, the integral velocity and length scales in the far field, and $Re_t = u\Lambda/\nu$ is the turbulent Reynolds number based on these scales.

The quantity in square brackets in (1) is the square of the surface divergence field, or the divergence of the 2D velocity vector tangential to the interface (u' and v' are the fluctuating velocities in the streamwise and spanwise directions, respectively), due to the fluctuating motions. Indeed, on a free water surface tangential velocity fluctuations are possible, meaning that the 2D continuity equation at the interface is not satisfied. Physically, this is the signature of surface convergence/divergence and renewal caused by turbulence events that bring bulk fluid to the interface, known as "sweeps". On the liquid side these are sometimes termed "upwellings" and on the gas side as "downdrafts".

Equation (1) arises in a straightforward way by noting that in contrast to rigid walls, at a free surface the velocity fluctuations normal to the interface are given by (where z being the distance from the interface)

$$w' \propto \left.\frac{\partial w'}{\partial z}\right|_{\rm int} z + {\rm HOT} \tag{2}$$

as a result of the boundary conditions, whereas at rigid surfaces it scales as

$$w' \propto \frac{\partial^2 w'}{\partial z^2}\Big|_{\rm wall} z^2/2 + {\rm HOT} \tag{3}$$

This has of course been pointed out by many authors, notably McCready et al. (1986). If the interface deforms then its curvature, $\kappa = -\nabla\cdot\mathbf{n}$, where $\mathbf{n}$ is the normal vector to the interface, should also enter the definition of (1), i.e.

$$\frac{\beta Sc^{1/2}}{u} \approx \frac{C}{Re_t^{1/2}}\left[\left(\frac{\partial u'}{\partial x}+\frac{\partial v'}{\partial y}-2w'\nabla\cdot\mathbf{n}\right)^2\right]_{\rm int}^{1/4} \tag{4}$$

where we have now introduced a proportionality coefficient $C \sim {\rm O}(1)$. While we will refer to the quantity between the parentheses on the RHS as the surface divergence, the last term is actually a surface dilation. If (1) and (4) are written in dimensional terms then

$$\beta \approx C(D\gamma)^{1/2} \tag{5}$$

where γ is the dimensional surface divergence, which has the dimensions of an inverse time scale ($1/s$), and D is the molecular diffusivity. In a sense γ takes the place of the renewal parameter—the mean time between surface renewals—$\bar{\tau}$ in Danckwerts (1951), who extended the Higbie (1935) penetration theory to turbulence-dominated situations. Note that the Higbie–Danckwerts surface renewal model predicted the dependence of the liquid-side scalar transfer rate on D, as $D^{1/2}$, which was also found in laboratory experiments for high Schmidt numbers.

However, application of the Higbie–Danckwerts surface renewal models is difficult since the time between renewals $\bar{\tau}$ remains unspecified. This led a number of researchers to propose various models for this quantity, notably the "large-eddy model" (LE) of Fortescue and Pearson (1967) and the "small-eddy model" (SE) of Banerjee et al. (1968), viz. $\bar{\tau} \approx \Lambda/u$ and $\bar{\tau} \approx (\nu/\varepsilon)^{1/2}$, respectively. The main advantage of transfer models involving the surface divergence instead of the time between renewals is that γ is more easily measured than $\bar{\tau}$—usually by scattering particles on the liquid surface and measuring their trajectories (see, for example, Kumar et al., 1998).

As explained previously, expressions (1) and (4) apply strictly for far-field turbulence that is homogeneous and isotropic, and at unsheared interfaces. If gas shear is imposed, then the turbulence structure near the interface has characteristics somewhat similar to that of wall turbulence. The surface divergence scaling is still expected to apply for liquid-controlled transport processes, but the turbulence structure is now controlled by generation in the near-interface region. The appropriate scaling variables are now related to the gas stress imposed on the water surface and the fluid kinematic viscosity (the so-called inner variables), i.e. $\beta^+ = \beta/u_{\star,\text{frict}}$ and $u_i'^+ = u_i'/u_{\star,\text{frict}}$, in which case expression (4) should be recast in the form

$$\beta^+ Sc^{1/2} \approx C\left[\left(\frac{\partial u'^+}{\partial x^+} + \frac{\partial v'^+}{\partial y^+} - 2w'^+\nabla^+\cdot\mathbf{n}\right)^2\right]_{\text{int}}^{1/4} \tag{6}$$

with the same proportionality coefficient $C \sim O(1)$ as in (4), and all quantities on the RHS of (6) are now normalized by $u_{\star,\text{frict}}$ and ν.

2.2. Blocking-theory based SD model

The surface divergence cannot be predicted without a theory, and therefore the Hunt and Graham (1978) blocking theory was used to relate it to the far-field turbulence characteristics when they are homogeneous and isotropic. To proceed, for an unsheared interface with homogeneous isotropic far-field turbulence, Banerjee (1990) derived, reworking a result of Brumley and Jirka (1987), the spectrum for the surface divergence term $\gamma = [\partial u'/\partial x + \partial v'/\partial y]_{\text{int}}$ in the form

$$S(\Omega) = 0.3[12\Omega^{1/2} - 7.2\Omega^{1/3}] \tag{7}$$

where Ω is the nondimensional frequency ($\Lambda\omega/u$), and the spectrum is valid for $\Omega > 5$. Banerjee then integrated the spectrum from the integral length scale Λ to the viscous cut-off $(\nu/\varepsilon)^{1/2}$ and used the relationship between the integral and Kolmogorov scales as

$$\Lambda/\eta \approx 0.5Re_t^{3/4} \tag{8}$$

with $\eta = (\nu^3/\varepsilon)^{1/4}$ to obtain the mass transfer coefficient for high Sc as

$$\frac{\beta Sc^{1/2}}{u} \approx \frac{C}{Re_t^{1/2}}\left[0.3\left(2.83Re_t^{3/4} - 2.14Re_t^{2/3}\right)\right]^{1/4} \tag{9}$$

Table 1
Various correlations for mass transfer velocity (β or k) for liquid side at high Schmidt number

Model	$\beta Sc^{1/2}$	Reference
Large-eddy (LE)	$C_1 u Re_t^{-1/2}$	Fortescue and Pearson (1967)
Small-eddy (SE)	$C_2 u Re_t^{-1/4}$	Banerjee et al. (1968)
Surface divergence (SD)	$C_3 Sc^{1/2}(D\gamma)^{1/2}$	Banerjee (1990)
SD no shear	$C_3 u[0.3(2.83Re_t^{3/4} - 2.14Re_t^{2/3})]^{1/4} Re_t^{-1/2}$	Banerjee (1990)
Interfacial shear	$0.108 - 0.158u_{\star,\mathrm{frict}}$	Banerjee (1990)
Eddy resolving analytical	$C_4 p_1 u_{\star,\mathrm{frict}}$	Csanady (1990)
Surface processes	$C_5 p_2 u_{\star,\mathrm{frict}}(1 + Rf/Rf_{\mathrm{cr}})^{1/4}(1 + Ke/Ke_{\mathrm{cr}})^{-1/2}$	Soloviev and Schluessel (1994)

C_1, C_2, C_3, C_4 are constants. p_1 is the fraction of the surface undergoing intense renewal. p_2 is the probability distribution of renewal events, Rf is the flux Richardson number, i.e. $gH\nu/\rho c_p u^4_{\star,\mathrm{frict}}$, and Rf_{cr} is the critical value $\approx 1.5 \times 10^{-4}$. Ke is the Keulegan number, i.e. $u^3_{\star,\mathrm{frict}}/g\nu$, and $Ke_{\mathrm{cr}} \approx 0.18$. H is the surface heat flux obtained by summing latent, sensible and long wave radiation fluxes. (Adapted from Banerjee and MacIntyre (in press)).

The proportionality constant $C \approx \mathrm{O}(1)$ is as in (4) and (6). This is sometimes called the surface divergence (SD) model. The quantity within the first set of parentheses is the square of the non-dimensional surface divergence. The expression was asymptotic to $Re_t^{-1/2}$ at small turbulent Reynolds numbers and to $Re_t^{-1/4}$ at large turbulent Reynolds numbers, which was in line with Fortescue and Pearson (1967) LE model, and Banerjee et al. (1968) SE model, respectively, and supported Theofanous' (1984) predictions for the asymptotic forms of the mass transfer rate with regard to Re_t. Note that this expression applies only to clean, unsheared rigid interfaces, with no effects due to surfactants or natural convection. Note, too, that the expression for surface divergence can be directly checked by DNS—a procedure we will follow when assessing the model.

The various expressions for the transfer rate of sparingly soluble gases are summarized in Table 1, which also includes expressions from Csanady (1990) and Soloviev and Schluessel (1994). Several other forms of parameterization were suggested, e.g. by Caussade et al. (1990) and Coantic (1986), so Table 1 does not list all the parameterizations proposed.

3. Unsheared interfaces: comparisons

In this section we will consider turbulence phenomena and scalar exchange when gas shear at the interface is relatively small or nonexistent. We review here for a more complete picture of the SD model some results of comparisons with experiments, which also appear in BM. In laboratory studies, the unsheared interface situation is reproduced using either a stirred tank or open-channel flow, where turbulence is generated by the shear at the bottom. In most of these experiments, it is difficult to keep the liquid surface free of surfactants, and therefore the results should be treated with caution, unless precautions have been explicitly taken to keep the surface clean. This is especially true for stirred-vessel experiments, where the liquid surface is quite stagnant.

Turning now to gas transfer data, Chu and Jirka (1992) measured the gas flux at the air–water interface of a grid-stirred tank. There is some concern that their data may have been affected by accumulation of surfactants at the water surface. This is discussed in more detail by McKenna et al. (1999), who also made simultaneous mass transfer and surface divergence measurements using DPIV. They found that the particles used for DPIV gave rise to surface-active effects unless they

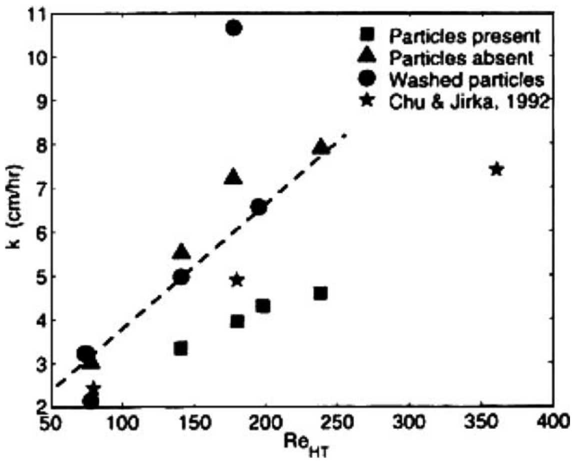

Fig. 1. Mass transfer coefficients at the unsheared surface of a grid-stirred vessel. Data from McKenna et al. (1999). Particles evidently give rise to surfactant-like effects. The data with the particles absent is considered the most reliable. The line is from Eq. (9) with $C = 0.20$ (also called C_3 in Table 1). (Adapted from Banerjee and MacIntyre (in press)).

were thoroughly cleaned. In fact, the mass transfer rate data with particles that had not been washed fell below data without particles or with washed particles. The data with and without particles are compared with Chu and Jirka's data, and the predictions of the parameterization in Eq. (9) as shown in Fig. 1. As evident from the figure, the particles reduce the gas transfer velocity, and Chu and Jirka's data lies somewhere between the cases in McKenna et al. (1999), with and without particles.

The parameterization in Eq. (9) also gives a reasonable fit to the data, which is encouraging, since it was developed before the data were taken. The constant $C \approx 0.20$, might be expected since Eq. (9) is based on a "rigid lid" approximation for the free surface, and in reality there will be some give which would reduce the surface divergence. Note also that the turbulent Reynolds number, Re_t, in Eq. (9) is half the turbulent Reynolds number, Re_{HT}, used by McKenna et al. (1999). Also, the point in their data shown in Fig. 1, where k (also called β) ≈ 11 cm/h at $Re_{HT} \approx 180$, is out of line with all the other data and may be an outlier.

Knowlton et al. (1999) compared predictions to Komori et al. (1989) open-channel mass transfer data and attempted to validate the surface divergence theory by calculating surface divergence directly from the velocity field measured by Kumar et al. (1998). They then used it to solve the 3D concentration field equation from which they calculated the mass transfer coefficient. The only case for which Kumar et al. (1998) data coincided with one of Komori's cases was for depth-based $Re = 2800$. Knowlton et al. (1999) found remarkable agreement with Komori et al. (1989) data for this case. However, when Knowlton et al. (1999) calculated the gas transfer data based on a "rigid lid" direct numerical simulation, they found gas transfer velocities that were about 2–3 times higher. These results are shown in Fig. 2. In Table 2, we show the predictions of Eq. (9) with $C = 0.20$ (which was the value that agreed with the stirred vessel gas transfer data of McKenna et al. (1999)). It is evident that the agreement with these predictions is quite good. The velocity scale for Eq. (9) is taken to be the wall friction velocity, and the length scale was the depth. While these scales are reasonable, it is likely that the length scale is a weak function of the depth-based Reynolds number, i.e. (Λ/depth) varies as $Re^{-1/8}$, which is what would be expected in

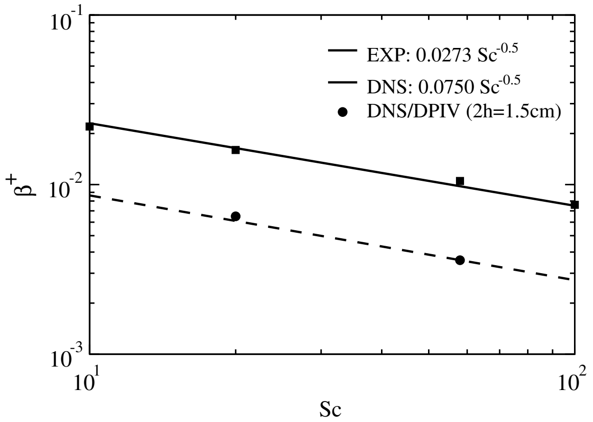

Fig. 2. Comparison of DNS with rigid slip surface as the interface in channel flow with experiments (Komori et al., 1989) and calculations based on experimentally determined surface divergence field. Note that the rigid surface gives higher values for the mass transfer velocity. (Adapted from Knowlton et al. (1999)).

Table 2
Comparison of Komori et al. (1989) experimental data with Eq. (9) with $C = 0.20$ (Adapted from Banerjee and MacIntyre (in press).)

Run no.	δ [cm]	U_{av} [cm/s]	$u_\star$ [cm/s]	Exp. $\beta \times 10^5$ [m/s]	Eq. (9) $\beta \times 10^5$ [m/s]
I	1.1	23.5	1.48	1.65	1.97
II	2.9	9.7	0.61	0.75	0.81
III	3.1	18.3	1.06	1.60	1.22
IV	5.0	5.9	0.37	0.45	0.49
V	5.1	11.9	0.69	0.90	0.78
VI	6.4	19.9	1.01	1.20	1.01
VII	7.0	13.8	0.75	1.30	0.80
VIII	10.0	10.5	0.58	0.70	0.56
IX	11.2	10.9	0.59	0.80	0.55

the core region of pipe flow. Making such an assumption would improve the agreement between Eq. (9) and the experimental data, which is rather over predicted at low Reynolds numbers and under predicted at high Reynolds numbers.

In any case, the SD model with $C = 0.20$ appears to predict gas transfer rates reasonably for unsheared interfaces. Note that experimental data are at high Schmidt numbers.

4. Sheared interfaces: comparisons

4.1. Simulations

De Angelis and Banerjee (1999) have reported DNS with a nonbreaking deformable interface between turbulent air and water streams. The details are available in De Angelis (1998). Their

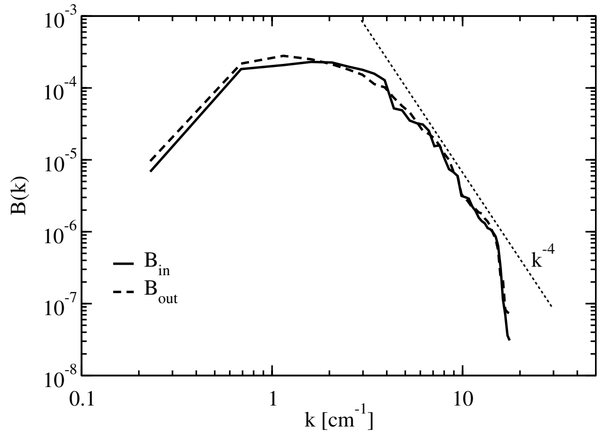

Fig. 3. Saturation spectra of the wave fields at the beginning (B_{in}) and at the end (B_{out}) of the sampling period. (Adapted from Lakehal et al. (2003)).

results show that waves exert significant effects on the mean flow and turbulence characteristics. The turbulence intensities and other qualitative features, e.g. streak spacing and burst frequency, on both gas and liquid sides of the interface, were found to scale with $u_{\star,\text{frict}}$ and ν, the kinematic viscosity.

More recently, Fulgosi et al. (2003) conducted a similar DNS with shear Reynolds number of 171, in which they were interested in the turbulence structure at the gas side of the interface as this deforms under the actions of the imposed shear. Fig. 3 shows the wave spectra obtained after statistical steadiness was reached. The wave saturation spectrum is defined by

$$B(\mathbf{k}) = |\mathbf{k}|^2(2\pi)^{-2}\int Z(\mathbf{r})\,\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{r}}\,\mathrm{d}\mathbf{r} \tag{10}$$

where $Z(\mathbf{r}) = \overline{f(\mathbf{x}+\mathbf{r},t_0)f(\mathbf{x},t_0)}$ is the covariance of the instantaneous nondimensional surface displacement $f(\mathbf{x},t)$. The figure clearly indicates that the wave properties did not change significantly within this time interval, confirming the existence of the saturation or equilibrium range, which is synonymous to convergence in this context. The comparisons reported with the data are when these "equilibrium" conditions are reached.

The initial study of Fulgosi et al. (2003) has been extended for the present contribution by performing new simulations with higher liquid shear velocities $u_{\star_L}$, although at the same shear Reynolds number (by varying the domain size). The purpose is to study the effect of surface dilation on the transfer mechanism for variable waveslopes. These new simulations were performed for shear velocities $u_{\star_L}$ equal to 0.000685, 0.002013, and 0.004 m/s, respectively. For the smallest shear velocity, the gas–liquid interface was almost flat. Note that $u_{\star_L}$ was set to 0.001 m/s in Fulgosi et al. (2003).

For all shear velocities studied, the difference between the gas and the liquid phase turbulence intensities in the near-interface region followed the same trend, as shown in Fig. 4. Gas-side turbulence displayed in the left panel behaves much like flow over a solid wall. If the distance is measured from the wavy interface, then there is little effect of wave deformations on intensity. The

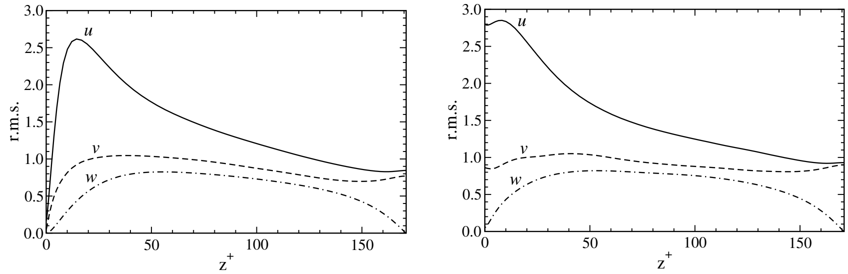

Fig. 4. Turbulence intensities on the gas side (left panel) and liquid side (right panel) as a function of the nondimensional distance z^+ from the interface, in the wavy interface conditions ($u_{\star_L} = 0.002013$ m/s). All quantities are nondimensionalized by the friction velocity.

liquid, as evident from the neighboring panel, has the largest fluctuations in the streamwise and spanwise direction right at the interface itself. The interface is perceived as a free slip boundary, except for the mean shear. The wave effect is somewhat more pronounced, but still rather small in the nondimensionalized form shown.

The simulations of De Angelis and Banerjee (1999) and Fulgosi et al. (2003) both proved useful in clarifying aspects of turbulence structure near deformable (nonbreaking) air–water interfaces, and they have been extended to studies of scalar exchange by De Angelis (1998) for high *Sc* numbers (up to $Sc = 200$), and recently by Lakehal et al. (2003) for low-to-moderate *Pr* or *Sc* numbers, up to $Sc = 10$. The calculations for $Sc \approx O(1)$ are straightforward once the velocity field has been calculated. However, for higher *Sc*, De Angelis (1998), who did not solve explicitly the coupled momentum and scalar equations, reduced the interface-normal mesh spacing in order to resolve concentration fluctuations using interpolated velocity flow fields (on the refined grid). In contrast, Lakehal et al. (2003), who were interested in comparing the entire scalar flux and variance balance equations solved the coupled equations for $Pr(Sc) = 1$, 5 and 10, using two grid resolutions.

The results presented below are new and differ from those presented in Lakehal et al. (2003), in that they concern the two shear velocities resulting in a flat interface ($u_{\star_L} = 0.000685$ m/s) and a wavy interface ($u_{\star_L} = 0.002013$ m/s). Contours of the instantaneous scalar fluxes at flat (left panels) and deformable (right panels) interfaces from these direct simulations are shown in Fig. 5, for the gas (first row) and the liquid side (second row), respectively. These results are compared in Fig. 6 with the shear stress at the interface, for flat and deformable interfaces. It is immediately evident that the gas-side fluxes correlate well with the shear stress, which has been independently found by De Angelis et al. (1999). This suggests that sweeps give rise to higher scalar exchange rates, as they also produce regions of high shear stress. On the other hand, the flux field on the liquid side shows a much finer structure and no such correlation exists. De Angelis et al. (1999) showed that this occurred because liquid-side sweeps did not give rise to the high shear stress regions at the surface, but they did give rise to regions of high mass transfer.

4.2. Surface divergence model comparisons

Before comparing the results of the new simulations with the surface divergence models, we first examine the surface divergence term γ^+ with and without curvature contribution for

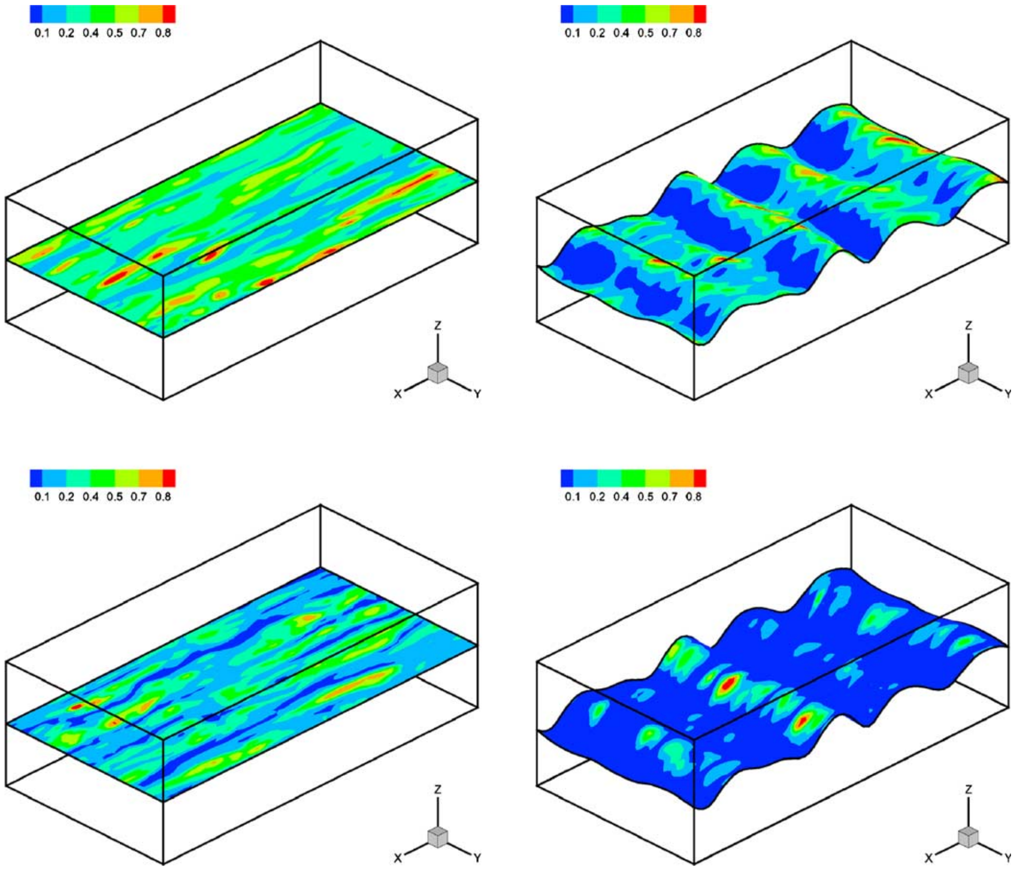

Fig. 5. Instantaneous patterns of the interfacial heat transfer coefficient at the flat and wavy interface. (upper panels) gas side, (lower panels) liquid side.

$u_{\star_L} = 0.002013$ m/s. The isocontours of the normalized (by inner variables) surface divergence is shown in Fig. 7, with (panel c) and without (panel a) the surface dilation contribution (shown in panel b). The dilation term is clearly shown to play an important role in estimating γ^+; panel (b) highlights indeed the crests and the troughs marked by positive and negative isocontours of the nondimensionalized dilation term κ^+. The wavelengths are also clearly visible from the figure. The lower panel corresponding to the modified surface divergence term including the dilation term exhibits differences with panel (a), which excludes the dilation term from the surface divergence.

Next, we assess the Banerjee (1990) derivation of the surface divergence field based on Hunt–Graham blocking theory (term in parenthesis in Eq. (9)) against the DNS data, before turning to the parameterization of the scalar exchange based on the SD transfer models. Note that in estimating the integral velocity scale Λ appearing in the definition of the turbulence Reynolds number, use was made of expression $\varepsilon \approx u^3/\Lambda$, where ε is the turbulent energy dissipation rate in the bulk flow. For the purpose of comparison, we have plotted in Fig. 8 the DNS-calculated values of γ (term between parentheses in Eq. (4)) against the term between brackets in Eq. (9), for various shear velocities (note though that the figure compares the entire RHS terms of these two

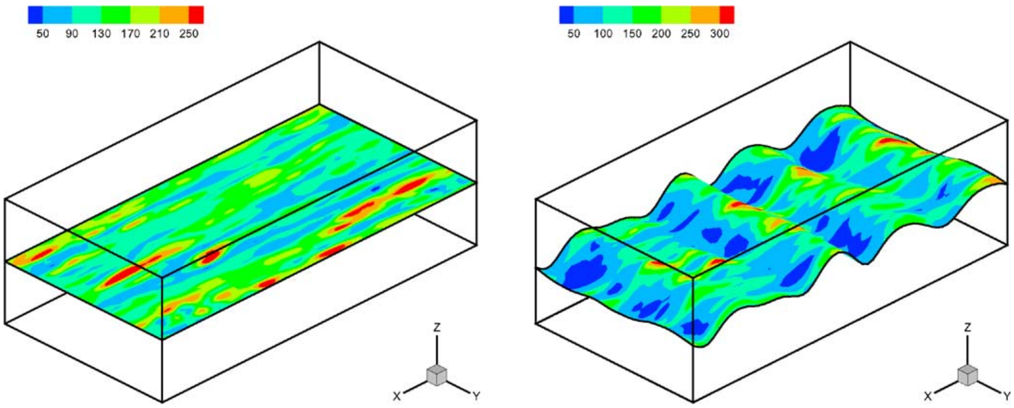

Fig. 6. Instantaneous patterns of the nondimensional interfacial shear stress, at the flat and wavy interface.

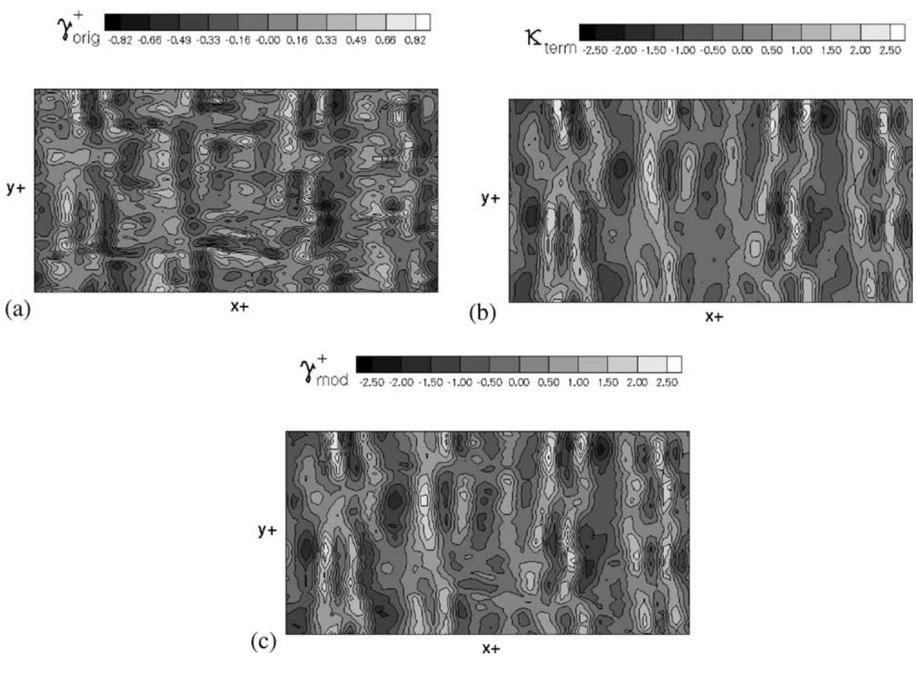

Fig. 7. (a) Isocontours of the surface term without the dilation term for $u_{\star_L} = 0.002013$ m/s. (b) Isocontours of the curvature or dilation term. (c) Isocontours of the surface term with the dilation term.

equations). The comparison shows very good agreement between the two quantities. This is a remarkable result, confirming the validity of the derivation proposed by Banerjee (1990) starting from blocking theory. Why it works for sheared interfaces is not clear, but the surface divergence expression derived from the blocking theory in (9) is certainly accurate.

Turning now to the scalar exchange parameterization, we consider the case where gas stress is imposed on the liquid interface, which is the context of the DNS studies discussed here. In these

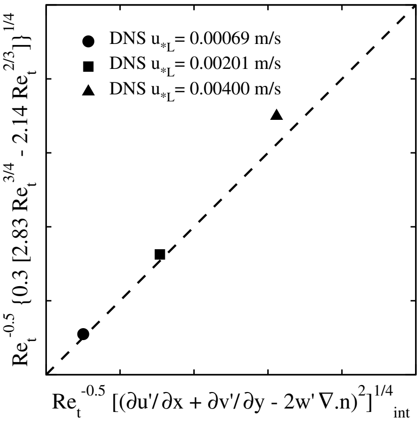

Fig. 8. Comparison of the surface term with the dilation contribution to the Hunt–Graham blocking theory, using DNS data for various shear velocities.

circumstances, the parameterization of the scalar exchange should be first examined with reference to Eq. (6). The Schmidt number dependencies in Eq. (6) are compared with simulation results for different values of *Sc* and for $u_{\star_L} = 0.002013$ m/s in Fig. 9(a). The best fit to the DNS data is obtained with the proportionality constant $C \approx 0.45$. It is clear that the dependence is well predicted up to $Sc = 200$, but there is some deviation at $Sc = 1.0$. In Fig. 9(b) the DNS results of the liquid-side mass transfer coefficient are compared with the SD model (Eq. (6)) for $Sc = 1.0$–1.2. The value of the constant $C \approx 0.35$ fits the SD model to the DNS results for various shear velocities quite well. Here the surface divergence is calculated from the DNS directly. This result is in conformity with what has been speculated before: for $Sc \approx 1$ the proportionality coefficient C in Eq. (6) should be somewhat lower. A similar trend has already been observed by De Angelis et al. (1999) (and confirmed by the present DNS simulations) in their parameterization of the scalar transfer by reference to the Banerjee et al. (1968) SE model, i.e. $\beta^+ \approx 0.108Sc^{-0.5}$, as also shown in Fig. 9(a). It is evident now that in sheared interface situations, both the SE and SD models (using inner-variables scalings) predict the gas transfer rate less accurately for $Sc \approx 1$ than for higher Schmidt numbers.

It is understandable that from an engineering point of view, the surface divergence term γ^+ with the dilation contribution cannot be easily determined; the alternative approximation to it is expression (9) using the far-field turbulence quantities, i.e. Re_t. According to the results in Fig. 8, the derivation is likely to hold for scalar transfer.

Let us then proceed to test whether even in gas-shear conditions, the SD model still applies for liquid-controlled transport processes. The question to which we seek an answer is as to whether the far-field turbulent Reynolds number could be employed for prediction, albeit the turbulence is actually generated in the near-interface region. It is therefore interesting to see how Eqs. (4) and (9) compare with simulation results. The Schmidt number dependencies in both equations are compared with simulation results for Schmidt numbers up to 10 in Fig. 10. The *Sc* dependence of the dimensional liquid-side mass transfer coefficient can be well predicted by both equations only with the proportionality coefficient set equal to $C = 0.35$. Figure 10 shows the SD model (Eqs. (4) and (9) with $C = 0.35$) to compare surprisingly well with simulation results for different values of

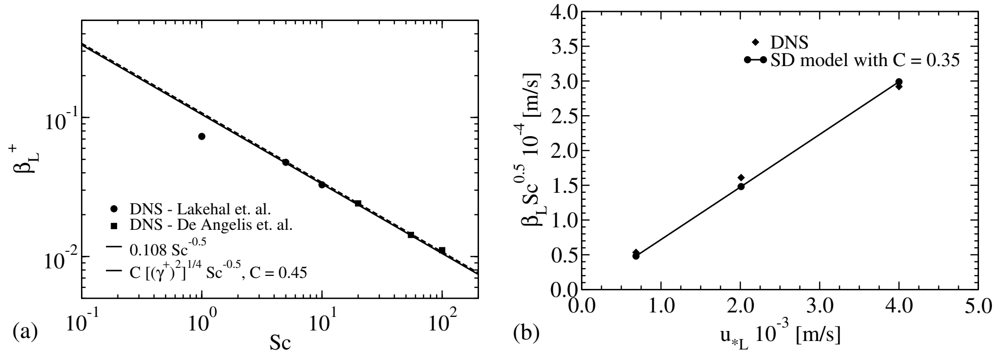

Fig. 9. (a) The scaling of the liquid-side mass transfer velocity (nondimensionalize by inner variables) with the *Sc* number using the SE model parameterization ($\beta^+ \approx 0.108Sc^{-0.5}$; c.f. De Angelis et al., 1999) and (6) with $C = 0.45$, for $u_{\star_L} = 0.002013$ m/s. (b) Dimensional mass transfer velocity versus frictional velocity for $Sc = 1.0$–1.2. The SD model Eq. (6) is clearly seen to fit the DNS data with $C = 0.35$.

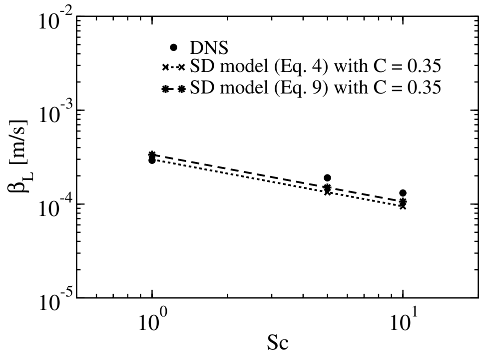

Fig. 10. The scaling of the liquid-side mass transfer velocity versus the the *Sc* number using the SD models under Eqs. (4) and (9) forms, for $u_{\star_L} = 0.002013$ m/s.

the shear velocity. However, the value of the constant should not be taken outside the interval $1 < Sc < 10$, as for higher *Sc* it may be different, as suggested by the results in Fig. 9(a).

5. Conclusions

In the absence of shear, and when the far-field turbulence approximates the homogeneous isotropic case, it has been found that the Hunt–Graham blocking theory applies, and predictions of the near-interface damping of the normal component of turbulence, and enhancement of the tangential components, are well predicted (Brumley and Jirka, 1987). Banerjee (1990) has applied the Hunt–Graham theory to calculate the surface divergence and developed the so-called surface divergence model, which predicts gas transfer across unsheared interfaces. It has also been shown that this model predicts at low turbulent Reynolds number the same behavior as the Fortescue and Pearson (1967) large-eddy model, and the Banerjee et al. (1968) small-eddy model at high turbulent Reynolds numbers. The model also agrees with gas transfer experiments across unsheared interfaces.

In situations in which the shear rate imposed by the gas is high, turbulence is generated in the vicinity of the interface, much like near solid boundaries. Models for gas transfer based on scaling of active events (such as sweeps and ejections) with interfacial frictional shear, have proved to be successful in predicting laboratory data, before microbreaking of waves in the capillary/gravity range commences, typically at 10 m wind velocities (U_{10}) of 4–5 m/s (De Angelis et al., 1999; Lakehal et al., 2003).

We have also tested the surface divergence model against DNS for sheared interfaces. What is the most surprising finding is that the expression for the surface divergence derived by Banerjee (1990) and shown in Eq. (9) applies not only for predictions of gas transfer at unsheared interfaces, but also in cases with wind shear. In fact, the predictions of the surface divergence in terms of Re_t on the RHS of Eq. (9) appears to hold even at sheared interfaces. This is a remarkable, and difficult to explain, finding. The value of the constant in Eq. (9) apparently changes somewhat between the sheared and unsheared cases with regard to scalar exchange, but the surface divergence itself is directly and accurately predicted. As the SD model is widely used to predict field experiments with high wind shear (Banerjee and MacIntyre, in press), this finding supports such usage.

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