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Systematic analysis of high Schmidt number turbulent mass transfer across clean, contaminated and solid interfaces

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ABSTRACT

A series of numerical simulation is carried out for high Schmidt number turbulent mass transfer across interfaces of different dynamical conditions, i.e., a clean or contaminated free surface and also a solid surface. A distinct feature of free surface turbulence close to a contaminated interface is drastic damping of the surface divergence fluctuations at low frequencies, which play a critical role in the interfacial mass transfer. Various concentration statistics reveal that the transport mechanism at a highly contaminated interface becomes dynamically equivalent to that at a solid surface. Consequently, the interfacial mass transfer rate falls down to the value on a solid surface, so that the Schmidt number dependency of the mass transfer rate switches from $Sc^{-0.5}$ to $Sc^{-0.7}$. Based on a one-dimensional advection–diffusion equation, it is demonstrated that the ratio between typical intensity and frequency of the fluctuating surface divergence is a critical parameter for the transition of the turbulent mass transfer mode.

1. Introduction

It is well-known that the absorption of surface-active agents, i.e., surfactants drastically retards interfacial mass transfer. For example, Frew (1997) conducted laboratory experiments under an identical wind velocity with seawaters sampled at different areas. They showed that the reduction rate of the mass transfer rate was as high as 90% in the case of coastal waters containing the highest level of surfactants. This suggests that the unique relationship between the wind velocity and the mass transfer rate, which is usually assumed in estimating the atmosphere–ocean gas flux, is unlikely to exist for natural waters.

In natural waters, surfactants are commonly present due to marine exudates, excretion and degradation products of phytoplankton, machine lubricants and so forth. In general, surfactants influence air–water gas exchange through static and dynamic mechanisms. The static effect represents the additional resistance to mass transfer due to physical barrier provided by a surfactant layer. This effect is considered to be minor in the real ocean (Frew, 1997; Donelan and Wanninkhof, 2002). In contrast, the dynamic effect is pronounced even at a slightly contaminated interface. The viscoelastic properties of a contaminated interface modify interfacial boundary conditions so as to oppose eddy motions close to the interface (Davies, 1966). As a result, free surface turbulence and associated gas exchange are strongly damped. In the present study, we focus on the hydrodynamic effect, which is also referred to Marangoni effect.

Up to now, numerous studies of gas transfer across clean and contaminated air–water interfaces have been carried out in stirred vessels and wind-wave facilities, e.g., Asher and Pankow (1986) and Jähne et al. (1987). In these studies, a common trend has been observed. Specifically, the mass transfer rate Q increases only slowly below a critical wind velocity, while it suddenly increases beyond the critical wind velocity. It was also confirmed that the Schmidt number dependence of Q switches from $Q \propto Sc^{-0.7}$ to $Q \propto Sc^{-0.5}$ above the critical wind velocity (Jähne et al., 1987). Since the Schmidt number of slightly soluble gases such as CO_2 is generally high ($Sc = O(10^3)$), the transition of the mass transfer mode causes a tenfold difference in the mass transfer rate.

So far, most researchers have been attributing the transition of the mass transfer mode to capillary-gravity waves, which start to grow beyond the critical wind velocity. In contrast, Jähne et al. (1987) argued that the change in the Schmidt number dependency of the mass transfer rate indicates a change in the transport mechanism. It should be a transition from a rigid to a free surface regime rather than from a smooth to a rough regime. Actually, direct measurement of concentration fluctuations close to an interface (Lee et al., 1980; Asher and Pankow, 1986) indicate that surface renewal eddies are retarded due to surface contamination, and therefore the concentration fluctuations are drastically decreased inside the viscous sublayer. Despite considerable researches, interaction between underlying eddies and a contaminated

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Nomenclature

Notations

c	concentration of solute
C	mean concentration
C_B	bulk mean concentration
C_I	mean concentration at an interface
c_τ	friction concentration
D	molecular diffusivity
E_d	eddy diffusivity
k	kinetic energy
k_x, k_y, k_z	number of modes in streamwise, interface-normal and spanwise directions
L	integral length scale
p	pressure
q	local mass transfer rate (dimensionless local mass flux)
Q	global mass transfer rate (dimensionless global mass flux)
$R_{\alpha\beta}$	correlation coefficient between α and β
Re_T	Reynolds number based on macroscopic velocity scale u_T and integral length scale L
Re_τ	Reynolds number based on friction velocity u_τ and depth δ
n	Schmidt number exponent of mass transfer rate
N	number of grid points
Sc	Schmidt number
t	time
U	mean streamwise velocity
u, v, w	velocity components in the x, y, and z directions
u_T	macroscopic velocity scale
u_τ	friction velocity
W_β	frequency spectrum of the surface divergence β
x, y, z	streamwise, interface-normal and spanwise directions
y_p	peak location of concentration fluctuation

Greeks

β	surface divergence
β_E	effective surface divergence
ΔC	difference between mean concentrations at an interface and a bottom boundary
ΔC_B	difference between interfacial and bulk mean concentrations
Δt	time step
Δx, Δy, Δz	grid spacings in streamwise, interface-normal and spanwise directions
δ	depth of computational domain
δ_c	thickness of diffusive sublayer
γ	surfactant concentration
ν	kinematic viscosity
ρ	fluid density
σ	surface tension
τ	interfacial shear stress
ω	frequency

Superscripts

$()^*$	dimensional value
$()^+$	value non-dimensionalized by the shear unit
$()'$	fluctuating component
$\overline{()}$	mean component

Subscripts

$()_a$	value in the air phase
$()_w$	value in the water phase
$()_0$	value at equilibrium

interface, and associated turbulent mass transfer have not been well understood due to various obstacles in simultaneous measurement of the velocity and concentration fields close to a free surface.

Numerical simulation is quite useful in clarifying such microscopic transport phenomena close to an interface. For example, Tsai and Yue (1995) studied laminar interactions between a contaminated free surface and a vortical flow below. Shen et al. (2004) studied turbulence structures close to a contaminated shear-free surface and observed drastic damping of up- and downwelling motions, which play a critical role in near-surface turbulent transport. Handler et al. (2003) clarified the relationship between an interfacial thermal field and underlying free surface turbulence close to a contaminated interface by direct numerical simulations (DNS). However, no numerical study focuses on the transition of the mass transfer mode so far.

In the present study, we conduct numerical simulation of high Schmidt number turbulent mass transfer across clean, contaminated and solid interfaces in order to understand how an interfacial dynamical condition influences the interfacial mass transfer. We pay particular attention to the transition mechanisms of the turbulent mass transfer mode by systematically changing the Marangoni number representing a degree of surface contamination. We will proceed as follows. First, we show fundamental statistics inside a concentration boundary layer close to clean, contaminated and solid interfaces. Then, we study the transition mechanisms of the turbulent mass transfer mode at a contaminated interface. Finally, a critical parameter for the transition is discussed.

2. Computational model and numerical method

2.1. Numerical conditions

In this study, we consider a countercurrent air–water flow driven by constant pressure gradient as shown in Fig. 1, where x, y and z are the streamwise, interface-normal and spanwise directions, respectively. This configuration is advantageous for highlighting the effects of interfacial shear stress, since the free-slip boundary condition can be imposed at the bottom boundary to minimize its effects. It should be also noted that this configuration is nearly equivalent to a co-current air–water flow in which a coordinate system moves at the same speed as that of the interface, because the mean pressure gradient plays only a minor role compared with the interfacial shear in the near-interface region.

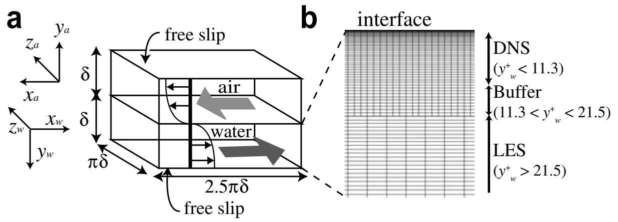

Fig. 1. (a) Computational domain and coordinate system and (b) grid system in hybrid DNS/LES.