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Perspective

# Transport Phenomena at Interfaces Between Turbulent Fluids

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## Introduction

Transport phenomena at interfaces between turbulent fluids play a central role in many industrial and environmental processes. The subject is crucial for the design of gas-liquid reactors,[1,2] phase-change heat exchangers, and steam reformers.[3] The issue has received renewed attention with the need to scrub pollutants from flue gas,[4] and in the design of bubble columns for air-sparging[5] and bioreactors.[6] Prediction of gas and mass transfer to and from water bodies, such as rivers, lakes, and the oceans is also of considerable interest for environmental systems. For instance, the transfer of greenhouse gases, e.g., methane and $CO_2$, in surface waters is important because of its impact on global warming.[7] Other examples include the atmospheric exchange of oxygen with hypoxic water bodies, and the desorption of dissolved toxics, such as PCBs, from inland and coastal water bodies.[8]

The broad interest in this topic is evident from the diversity of experimental and numerical studies published in recent monographs.[9,10] The research teams include engineers, physicists, oceanographers and atmospheric scientists. Experiments range from microscopic-scale laboratory studies to large field-scale studies done using satellites and ships.

Until recently, estimates of the rate of interfacial mass-transfer for these processes were based on experimental measurements and semiempirical expressions. Scale-up was costly and often unreliable. The advent of large-scale and affordable computational resources has started to change this, through the development of computational fluid dynamics (CFD) tools.[11] However, these computations are not yet able to directly simulate interfacial transfer due to the extremely fine resolution necessary for the task. Therefore, even with the aid of CFD, the interfacial transport rate must be calculated from empirical parameterizations.

The focus of this Perspective article is to review our current understanding of scalar exchange processes at gas-liquid interfaces. There is certainly room for improvement. For example, a recent comparison of empirical measurements to computational predictions for interfacial heat transfer in a steam condensation pipeline found disagreement of about two-orders of magnitude.[12] Likewise, recent attempts to predict the annual transfer of atmospheric $CO_2$ to the Earth's oceans, using various air-water gas-transfer parameterizations, encounter disagreements of factors of three.[13,14] In other applications these same air-water gas-transfer parameterizations produce even larger discrepancies.[15]

The solution of the usual advection-diffusion equation should, of course, accurately predict these phenomena. However, in most cases the flow field near the interface is poorly known, and, consequently, the results of advection-diffusion calculations are uncertain. Improvement in understanding of the near-interface flow field has been slow because the dynamics are complex, and measurements near the deforming interface are difficult. To provide some perspective, note that understanding of turbulence near solid boundaries, like wall turbulence, still eludes us despite the vast effort devoted to this end.

Nonetheless, progress is being made and will be discussed in this Perspective. The most significant advances in understanding have considered simple gas-liquid systems with a continuous interface between the fluids, e.g., the air-water interface of surface waters, the gas-liquid interface of stirred tanks, or the steam-water interface of film condensing equipment. This Perspective will focus on these types of systems. Excellent general reviews on the subject exist in the literature.[15–17] Recently, much has been learned using direct numerical simulations (DNS)[18,19] and experimental studies using particle imaging velocimetry (PIV),[20,21] both of which will be discussed in this article.

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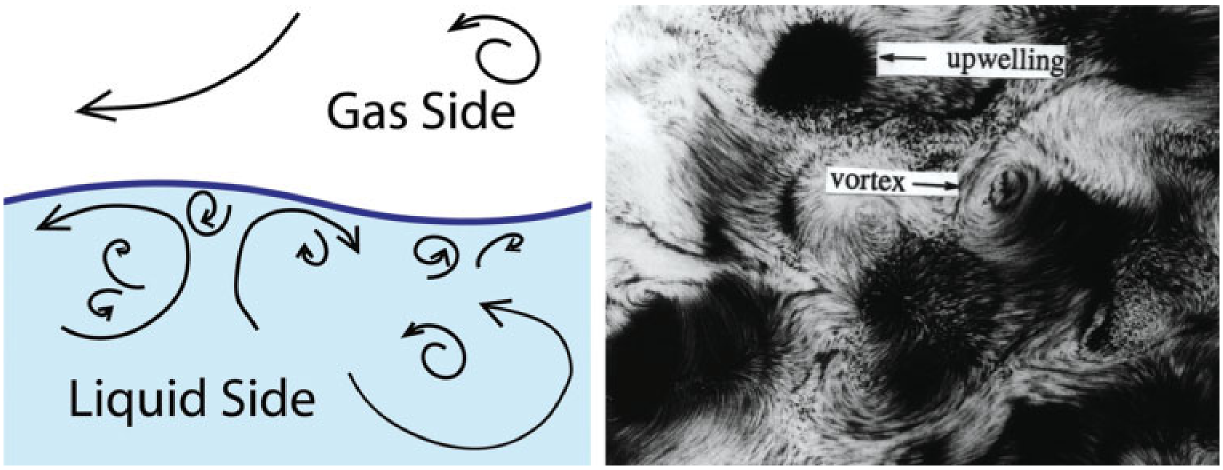


**Figure 1. A conceptual picture of the problem considered here.**

On the left is an illustration of a gas-liquid interface. Small eddies, large eddies and deformations all interact near the interface. On the right is a plan-view laboratory photograph of an air-water interface under turbulent conditions.[22] Silvered floating particles, which have been sprinkled on the air-water interface, show the presence of upwellings, downwellings, and vortices.

## Near-Interface Flow Field

Scalar transfer at interfaces is usually described with a transfer coefficient, here termed $\beta$, which describes the ratio of the flux across the interface $F$, to the difference in scalar concentration between the bulk fluid of the two phases, i.e., $\Delta c$ for a concentration difference, or $\Delta T$ for a temperature difference. For example, this gives $F = \beta \Delta c$, where $\Delta c$ is usually better known than $\beta$. The value of $\beta$ depends on the flow conditions near the interface, and the diffusivity of the scalar of interest. This indicates the importance of the near-interface flow field, especially since it is an aspect that is poorly understood, and can vary greatly from system to system.

Consider Figure 1, which shows a sketch of the canonical gas-liquid system considered in this article. In this system, some soluble gas or other scalar quantity, e.g., heat, is transferred across the interface due to nonequilibrium concentration conditions between the bulk gas and bulk liquid. Far from the interface, advection is able to transfer and mix the scalar quickly, and, therefore, the concentration fields are relatively uniform. Near the interface, however, the turbulence is damped and the effectiveness of mixing is reduced. Consequently, diffusional processes account for the final transfer of scalar quantities across the interface. In this region the scalar concentration will have strong gradients and it is, therefore, called the concentration boundary layer.

For many gas-liquid systems it is the liquid-side motions that have the greatest effect on the interfacial transfer coefficient $\beta$. This is due to a combination of reasons. First, molecular diffusivity in liquids is three-to four-orders of magnitude less than in gases, which causes the concentration gradient to be much greater on the liquid side. Further, many systems involve the transfer of a species that is poorly soluble in the liquid phase, e.g., $CO_2$, $O_2$, $H_2$, or $CH_4$ in water. All this causes the concentration boundary layer to lie mainly on the liquid side, causing the liquid side motions to affect the transfer rates much more than those of the gas side. This is also true for condensation or evaporation systems, so long as a single component is involved and there are no noncondensables.

As a side note, there are situations in which the gas-side motions can control the transfer. Absorption or desorption of mixed species, where one or more components have high liquid-side solubility, but the others do not, can lead to situations in which gas-side motions control transfer rates. Also, chemicals with high-solubility in the liquid phase force the concentration boundary layer into the gas side. This occurs, for example, with $SO_2$ gas transferring to and from water. A more detailed discussion of the controlling side for gas-liquid systems is available in the literature.[17]

In most situations the gas-side motions behave as if, to a first approximation, the liquid were a relatively slowly deforming solid. Interfacial transfer in systems where the gas-side motions determine the transfer rate is very similar to transfer from a solid wall. The dynamics of the interfacial transfer are well understood compared to the case for which liquid-side motions control the transfer rate. This article, therefore, focuses on the less understood, liquid-side controlled, systems. These systems introduce phenomena different from what is seen for transfer from a solid boundary.

We turn now to the interaction between the turbulent flow field and the interface. A useful approach was proposed by Hunt and Graham[23] who considered the case of homogeneous turbulence incident on a flat interface. Using the "method of images" to determine the interaction between the flow field and the interface, they were able to reach the following conclusions: (a) beyond a distance of one integral length scale $L$, which is the size of the larger turbulent eddies, the interface does not affect the turbulence significantly, (b) within a distance of $L$ the interface distorts the turbulence such that flows directed toward, or away from, the interface are reduced and redirected parallel to the interface, and (c) within a viscous boundary layer, which for turbulent flows is much thinner than $L$, the interface affects the flow through viscosity. The second region, the one within a distance of $L$, is termed the "surface-influenced region". Hunt and Graham's calculations found that within this region the magnitude of the interface-normal turbulence intensity decreases as $1.2Y^{1/3}$, where $Y$ is the distance from the interface normalized by $L$. They further were able to calculate one-dimensional (1-D) turbulent power spectra at varying distances from the interface, finding that, for a particular location, the interface damps eddies with wavelengths greater than the distance to the interface. Subsequent laboratory measurements and CFD have confirmed the Hunt-Graham theory.[24,25] Later theoretical work by Brumley and Jirka[24] and Banerjee[26] extended this theory to allow calculation of interfacial transfer rates for homogeneous turbulence. This will be discussed further in the section on Surface Divergence Theories.

A separate development concerned the kinematics within the concentration boundary layer. Due to the fact that the Schmidt number *Sc,* the ratio of kinematic viscosity to scalar diffusivity, is large, the concentration boundary layer is thinner than both the viscous boundary layer and the smallest turbulent eddies. This allows a simplification, first utilized by Chan and Scriven,[27] that the flow in the concentration boundary layer may be considered a "stagnation" type of flow. For these stagnation flows the interface-normal velocity is proportional to the distance from the interface, i.e., $v \propto y$, where $v$ is the velocity normal to the interface, and $y$ is the distance to the interface. The slope of this proportionality is termed $\gamma$ and its sign, negative or positive, indicates upwelling or downwelling, respectively. Due to fluid continuity the interface-parallel velocity is divergent under upwelling conditions, and convergent under downwelling conditions. Using this kinematic simplification the advection-diffusion equation may then be analyzed explicitly. This type of approach is termed a surface divergence model and will be discussed later. These models remained largely out of the mainstream research until the 1980s. Before this, the focus was largely on surface renewal models.

## Surface Renewal Models

Early work by Lewis and Whitman[28] hypothesized that a film of laminar fluid existed adjacent to the interface. The bulk fluid was obviously well mixed, and, therefore, this laminar film provided the main resistance to mass transfer. This model took no account of the turbulence reaching the interface. Indeed, very little was known about turbulence at the time. Lewis and Whitman's model predicted interfacial transfer coefficients proportional to the scalar diffusivity, i.e., $\beta \propto D$. Subsequent experiments in stirred tanks made it clear that these coefficients were actually proportional to $D^{1/2}$, and, therefore, the Lewis and Whitman model needed refinement. This led to the speculation that episodic diffusive absorption mediated the mass-transfer process. Transfer was hypothesized to proceed into an essentially laminar fluid for some time after which the laminar fluid was "renewed" by fresh bulk fluid. This theory gave the correct proportionality of the transfer coefficient to diffusivity, i.e., $\beta \propto D^{1/2}$. Subsequently, Danckwerts[29] allowed for a random distribution of times between these renewal events, which is more typical of what occurs in a turbulent fluid, and arrived at an often cited result $\bar{\beta} = (D/\tau)^{1/2}$ where $\bar{\beta}$ is the average interfacial transfer coefficient, and $\tau$ is the average time between renewal events.

Predictions using this method proved to be correct for simple systems where the average time between renewals was easy to identify, e.g., a clean bubble rising smoothly through laminar fluid,[30] or calm fluid exposed to a diffusive scalar for some known length of time.[31] However, for turbulent systems the specification of $\tau$ is not as clear. Researchers studying this model in the 1960s and 70s utilized theories of turbulence scaling and turbulent cascades in order to determine $\tau$. The first of these to emerge was the "large-eddy" model of Fortescue and Pearson,[31] which postulated that the largest eddies present in the turbulence are the ones dominating the surface renewal process. At about the same time, Banerjee et al. developed a "small-eddy" model,[32] based on the contradictory idea that the smallest eddies, i.e., the dissipative timescales, were the most important. Assuming Kolmogorov scaling the dissipative timescale may be estimated as $(v/\varepsilon)^{1/2}$, where $\varepsilon$ is the dissipation rate and $v$ is the kinematic viscosity. The dissipation rate, in turn, may be estimated as $u^3/L$, where $u$ is the turbulence intensity, so long as a turbulent cascade is valid. Both models agreed with empirical measurements under their own sets of conditions, but gave very different results when applied under the same conditions. This discrepancy was resolved by showing that the two models represented the asymptotic behavior at small and large turbulent Reynolds numbers, respectively.[33]

While the large- and small-eddy models were derived for situations with continuous interfaces, they may also be applied to more complex situations. The small-eddy model in particular offers a method of estimating the mass-transfer coefficient, provided the system is highly turbulent. In a dispersed gas-liquid system, e.g., a bubbly flow, the dissipation scales on the liquid side can be estimated from CFD calculations, and the small-eddy model may be applied. There is, however, a problem with this type of approach. The energy cascade from large-scale turbulent structures to the fine-scale occurs in flows where the turbulence arises primarily from large-scale structures, e.g., wake flows or large bubbles. However, if the turbulence is largely generated by interfacial shear, then the interface will exhibit "bursting" and other kinematic phenomena.[34] While models of this nature, e.g., small-eddy or large eddy, might work, their physical basis is weak in such situations. In view of all this, attempts have been made to construct more realistic models for sheared interfaces, based on the intermittent nature of the turbulence generated. Such models are now considered.

If the interfacial shear is strong enough to cause turbulent motions in the liquid, but not strong enough to create breaking interfacial waves, then both direct numerical simulations[35] and experiments[34] indicate that turbulent "bursting" motions form on the liquid side of the interface in a manner quite similar to that in wall turbulence. These bursts carry fluid away from the interface in "ejections" and bring fluid to the interface in "sweeps", and facilitate interfacial scalar transfer. The time between bursts was found to be centered at $50v(u^*)^{-2}$, where $u^*$ is the interfacial friction velocity. If this time is taken as the average surface renewal rate $\tau$, then the surface renewal model makes predictions that agree with measurements.[35] Figure 2 shows this agreement.

This version of the surface renewal model compares well with experiments as long as the gas-liquid interface does not "break", i.e., fold onto itself. Such breaking begins to occur as interfacial shear rate is increased and small gravity waves of approximate length $\sim$10 cm appear. These small waves, commonly called "microbreaking" waves, introduce new turbulence generation

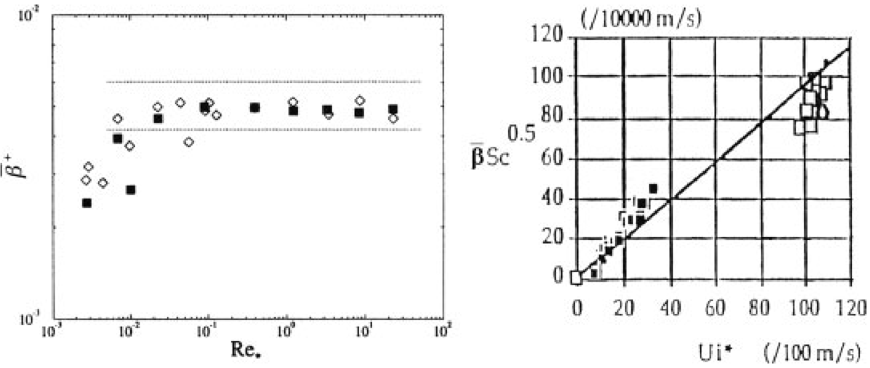


**Figure 2. Agreement between the small-eddy model and empirical data.**

On the left is data from a wind wave channel, Ocampo-Torres et al.[36] with symbols being experimental data and the dotted lines being the prediction of the surface renewal model.[26] On the right is data from a bubble column, Cockx et al.[37] with the line being the surface renewal model prediction. The renewal time was estimated by the bursting frequency.

mechanisms that affect scalar exchange rates. In fact, a dramatic change in the rate of interfacial transfer occurs in air-water systems when the wind speed, as measured 10 m above the interface, exceeds about 3 m/s. This change is now known to arise due to the onset of microbreaking waves.[21] The surface renewal type theories appear to be less applicable in such situations as multiple timescales are introduced, associated with wave and turbulence phenomena. The most successful approach appears to be the surface divergence theory, which is now discussed.

## Surface Divergence Models

As mentioned previously, the surface divergence model for interfacial scalar transfer has its origins in an article by Chan and Scriven[27]. The model is based on the realization that, for large *Sc*, the important fluid motions lie in a region of very small interface-normal dimensions, i.e., smaller than viscous length scales and eddy length scales. This allows the flow to be modeled as "stagnation" flow as mentioned previously. These flows are completely specified by a single parameter, the surface divergence strength $\gamma$. Negative $\gamma$ refers to fluid flowing toward the interface and interface-parallel liquid diverging. Positive $\gamma$ refers to fluid flowing away from the interface and interface-parallel motions converging. This manner of specifying the flow field led to the name "surface divergence" models. Chan and Scriven[27] had shown that, given a time series of $\gamma$ values, they could directly calculate the interfacial scalar transfer. McCready et al.[38] further developed this model to show that a synthetic time series of $\gamma$, with statistics similar to that of real turbulence, leads to $\bar{\beta} \sim D^{1/2}(\overline{\gamma^2})^{1/4}$ where $\overline{\gamma^2}$ is the mean-square surface divergence. Recent experimental measurements have confirmed this equation,[20,21] as seen in the left hand side of Figure 3.

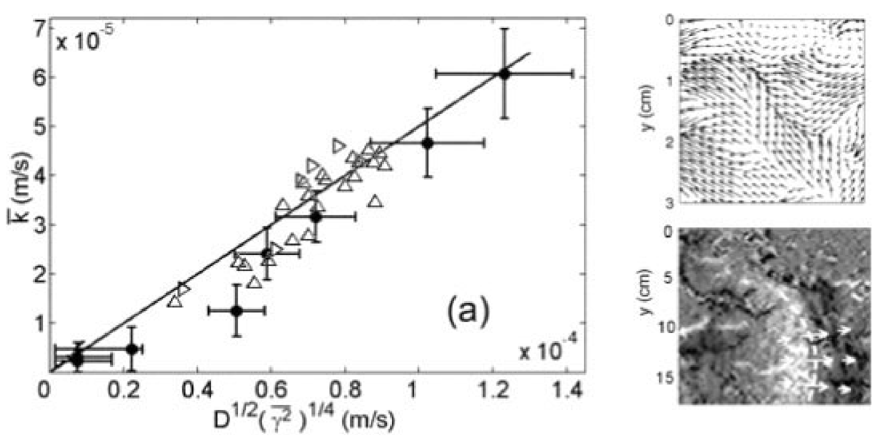


**Figure 3. Comparison of the surface divergence model with empirical data.**

The solid line is the surface divergence model prediction by McCready et al.[38] The solid dots and upward-pointing triangles are empirical data from sheared gas-liquid interfaces.[21,39] These sheared interfaces showed bursting phenomena at the lower end of the abscissa and microbreaking waves at the higher end. The right-pointing triangles are empirical data from stirred tanks.[20] On the righthand side are two plan-view images of interfacial flows, taken from experimental investigations. On the top are velocity vectors obtained using PIV with a grid stirred tank.[20] On the bottom is a grayscale graph of surface divergence calculated from PIV velocity vectors on a microbreaking interface.[21] In this grayscale graph, darker shade means greater convergence and lighter shade means greater divergence. A microbreaking wave is positioned in the lower center of the grayscale graph, its crest denoted by a dashed line and PIV vectors denoted by arrows.

Table 1. Parameterizations for the Gas-Liquid Scalar Exchange Coefficient ($\bar{\beta}$)

| Model | Parameterization | References |
|---|---|---|
| Large eddy | $\bar{\beta}Sc^{1/2} = C_1 u \; \mathrm{Re}_t^{-1/2}$ | 31 |
| Small eddy | $\bar{\beta}Sc^{1/2} = C_2' \; (\varepsilon v)^{1/4}$ where $\varepsilon$ is dissipation | 32 |
| | $\bar{\beta}Sc^{1/2} = C_2 u \; \mathrm{Re}_t^{-1/4}$ if $\varepsilon$ is estimated as $u^3/L$ | |
| Surface Divergence (no interfacial shear) | $\bar{\beta}Sc^{1/2} = C_3 u \; [0.3(2.83 \; \mathrm{Re}_t^{3/4} - 2.14 \; \mathrm{Re}_t^{2/3})]^{1/4} \; \mathrm{Re}_t^{-1/2}$ | 26 |
| Surface Renewal (with interfacial shear, no waves) | $\bar{\beta}Sc^{1/2} = 0.1u^*$ | 26,34 |
| Surface Divergence | $\bar{\beta}Sc^{1/2} = (v\bar{\gamma})^{1/2}$ | 26,38 |

The surface divergence model is emerging as a theory that is applicable to a wide range of flow conditions. Figure 3 illustrates this by plotting measurements of $\beta$ from stirred tanks, bursting shear flows, and microbreaking wavy flows on a single theoretical surface divergence line. Much current research is devoted to characterizing $\gamma$ under various flow conditions, and then relating scalar exchange rates to it. Laboratory examples of surface divergence patterns are seen on the right sides of Figure 1 and Figure 3.

Another intriguing finding, seen by numerous researchers, is a linear proportionality between interfacial rms slope, termed roughness, and interfacial transfer coefficients. Recent research[21] has also found a linear proportionality between rms slope and rms surface divergence values. Since roughness is routinely measured over the ocean by satellite remote sensing this has led to the idea of making rapid predictions of $\beta$ distributed across the entire oceanic region of the planet. Little work has been published on such a possibility, and it remains an idea for future research.

Further development of the surface divergence model came from Banerjee,[26] who considered the case of a homogeneous turbulent flow interacting with a flat interface. The kinematics of this situation were introduced a few sections earlier, with the Hunt and Graham[23] "method of images" discussion. Banerjee[26] used the Hunt and Graham theory, along with some developments by Brumley and Jirka,[24] to theoretically derive the rms surface divergence at the gas-liquid interface. Banerjee then used the surface divergence model to produce an equation that predicts the interfacial transfer coefficient. The only free parameter in the resulting equation is the turbulent Reynolds number, i.e., $\mathrm{Re}_t = u^* L/v$, of the far-field turbulence. Intriguingly, this equation agrees with the large-eddy surface renewal model at low $Re_t$, and also with the small-eddy surface renewal model at large $Re_t$. It also applies at intermediate values of $Re_t$, and provides a deeper understanding of the dynamics at play. Recent DNS has verified the prediction of rms surface divergence made by Banerjee.[40]

## Conclusions

This Perspective has covered the evolution of fluid-fluid interfacial turbulent scalar transport models, focusing on the surface renewal and surface divergence theories. The emphasis was on predicting the interfacial-transfer coefficient based on kinematic properties of the flow, e.g., rms velocities, length-scales, surface divergence patterns. Table1 above gives a survey of the models.

Most of the comparisons between theory and experiments were for separated gas-liquid flows, but there is some evidence that models of this nature also apply for dispersed flows, e.g., bubbly flows, where the dimensions of the dispersed phase are much larger than the turbulence scales. For low-shear conditions in which the interface does not break, parameterizations for scalar exchange rates in terms of the surface renewal model are found to be accurate. A more universal approach based on the interfacial surface divergence was found to give agreement both against DNS and experimental results under a broad range of conditions. While these developments continue to aid in the design and scaleup of chemical reactors and phase-change heat transfer equipment, much of the current interest arises from the need to better predict scalar exchange rates in environmental systems, most crucially $CO_2$ uptake by global water bodies. It is likely that the future will bring assessment of theories and laboratory experiments in large-scale environmental settings, utilizing satellite remote sensing techniques to obtain information indicative of surface divergence rates.

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