# Absorption of gas into a wavy falling film

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## Abstract

In this work, we present results of a study of gas absorption into a falling film on a vertical substrate. The film flow is accompanied by the formation of nonlinear waves which strongly influence the diffusion layer that develops from the film surface. As a result, significant enhancement of mass transfer has been observed in experiments. We use recent advances in modelling of the hydrodynamics and solve a two-dimensional convective–diffusion equation for the solute concentration. Numerical solutions for the finite-amplitude wave regimes and associated integral absorption rates are obtained for a range of flow conditions. Our results show clearly the influence of waves on the development of the diffusion layer and, in particular, the enhancement of absorption due to the waves; the existence of optimal conditions for maximizing the absorption rate is also demonstrated.
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## 1. Introduction

We consider gas absorption into a falling film of incompressible viscous liquid. This process is of central importance in chemical engineering applications and has received considerable attention in the literature (Kholpanov and Shkadov, 1990; Killion and Garimella, 2001). These applications may be divided into two categories. In the first category are applications where it is desired to achieve the maximum concentration of absorbed gas within the liquid. In the second category, the objective is to remove as much as possible of the solute gas from the gas phase; this has obvious applications in extraction processes. The flow distance required to achieve these objectives provides a measure of the efficiency of these mass transfer processes.

In order to provide a quantitative measure of the process efficiency, previous authors have characterised liquid saturation in different ways. Emmert and Pigford (1954) used the following mass transfer coefficient:

$$k_{\text{sat}} = \frac{\tilde{Q}}{\tilde{L}} \ln \frac{\tilde{C}_s - \tilde{C}_{\text{in}}}{\tilde{C}_s - \tilde{C}_{\text{out}}}, \tag{1}$$

where $\tilde{L}$ is the distance between the flow inlet and outlet, where the respective average concentrations of absorbed gas in the liquid are $\tilde{C}_{\text{in}}$ and $\tilde{C}_{\text{out}}$, $\tilde{C}_s$ is the concentration of absorbed gas in the liquid at the interface, and $\tilde{Q}$ denotes the flowrate. Experimental results for flows without waves, which were achieved through the use of surfactant, were presented as plots of $\tilde{Q}/k_{\text{sat}}\tilde{L}$ against $\tilde{H}^2/D\theta$ in which $\tilde{H}$ is the film thickness, $D$ is the molecular diffusivity, and $\theta$ is the time of contact of the gas–liquid interface. Emmert and Pigford presented data for the absorption of carbon dioxide and oxygen by water; the magnitude of the mass transfer coefficient, $k_{\text{sat}}$, was found to be less than that calculated by Pigford (1941) for the corresponding stationary diffusion problem (referred to below as the 'theoretical' value). In contrast, the $k_{\text{sat}}$ values associated with experiments featuring rippled film flows (with $Re = \tilde{Q}/v = 25$– $500$) were found to be 2.5 times larger than the theoretical value.

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Similar results were obtained by Oliver and Atherinos (1968) in experiments involving the absorption of carbon dioxide into a water film flowing down an incline at $Re = 20$– 300. These investigators measured the saturation parameter defined by

$$k_{sat,OA} = \frac{\tilde{Q}}{\tilde{L}} \ln \frac{\tilde{P}_s - \tilde{P}_{in}}{\tilde{P}_s - \tilde{P}_{out}},$$

where $\tilde{P}_s$ is the partial pressure of $CO_2$ in the gas, and $\tilde{P}_{in}$ and $\tilde{P}_{out}$ are the partial pressures of $CO_2$ in equilibrium with the liquid at the inlet and outlet. This formula is the same as (1) if a linear Henry's law relationship between $\tilde{C}_s$ and $\tilde{P}_s$ is accepted.

The local concentration profiles of carbon dioxide absorbed by a water film flowing down an inclined plate at relatively large Reynolds numbers $Re = 180$– 460 were directly measured by Jepsen et al. (1966). This study showed that in the vicinity of the inlet region, mass transfer takes place via molecular diffusion. Following the appearance of waves, the carbon dioxide penetrates rapidly to the bottom of the film, which leads to the subsequent increase of the concentration in that region of the film. Careful analysis of the concentration field showed that this increase greatly exceeded that predicted by theories based on molecular diffusion. The concentration profile at a given point was found to be the same below wave peaks or wave troughs; waves moving past the 'observation point' simply acted to 'compress' and 'expand' the concentration profile. Correspondingly, in the experiments carried out by Inazumi et al. (1986), a sudden increase in the rate of growth of the concentration with the streamwise coordinate was measured at the critical point where ripples in the film appeared.

Won and Mills (1982) presented the following empirical formula for the mass transfer coefficient

$$k_{sat} = (g\nu)^3 \times 6.97 \times 10^{-9} \frac{(4Re)^{n_1}\gamma^{3/2}}{Sc^{n_2}} \frac{\tilde{R} - \tilde{H}}{\tilde{R}},$$

$$\tilde{H} = 0.068 \left(\frac{\nu^2}{g}\right)^{1/3} (4Re)^{2/3}, \quad n_1 = \frac{3.49}{\gamma^{0.21}},$$

$$n_2 = 0.137\gamma^{0.17}. \tag{2}$$

This takes into account surface tension $\sigma$ through the Kapitza number (Kapitza, 1948) $\gamma = \sigma\rho^{-1}(g\nu^4)^{-1/3}$, in which $\nu$ is the liquid viscosity, $g$ is gravity and $\rho$ is the density. The formula was derived for $Re \in (250, 2500)$, $Sc \equiv \nu/D \in (80, 2700)$, $\gamma \in (420, 5800)$. In this work, the liquid film flowed on the inside wall of a vertical glass tube of radius $\tilde{R}$. The absorption of carbon dioxide and oxygen into methanol, ethanol and aqueous propanol solution was investigated. The authors did not discuss the effect of wave regimes on absorption; the hydrodynamics were characterised only in terms of the mean film thickness (2).

Yoshimura et al. (1996) carried out experiments to determine the dependence of the Sherwood number

$$Sh_{sat} = \frac{k_{sat}\tilde{H}}{D}, \quad \tilde{H} = \left(\frac{3\nu\tilde{Q}}{g}\right)^{1/3}, \tag{3}$$

on the wave regimes at moderate Reynolds numbers $Re \leqslant 70$. The effect of wave regimes on the absorption of oxygen into a vertically falling water film was investigated for both naturally developing waves and waves excited via oscillations imposed at the film inlet. It was shown that the wave frequency for which the largest value of the Sherwood number, $Sh_{sat}$, was recorded increased with the Reynolds number, $Re$.

Henstock and Hanratty (1979) conducted experiments on the absorption of oxygen into a falling film of water on the inner surface of a vertical pipe at $Re = 85$– 2700. The rate of absorption per unit area, $\tilde{q}_a$, was characterised through the following expression:

$$k_m = \frac{\tilde{q}_a}{\tilde{C}_s - \tilde{C}_\infty}, \tag{4}$$

where $\tilde{C}_\infty$ is the concentration in the liquid film away from the surface, and the following correlation for $k_m$ was provided

$$k_m = 0.067 \frac{(gD)^{1/3}}{\nu^{1/6}} (5.65\, Re^{2.5} + 0.000147 Re^{4.9})^{1/6},$$

which suggests that $\tilde{q}_a$ increases with $Re$. These authors further showed that a co-current gas flow leads to an increase in the value of the Sherwood number based on the absorption flux, $Sh_m = k_m\tilde{H}/D$. McCready and Hanratty (1985) subsequently found that the saturation Sherwood number, $Sh_{sat}$, (3) for oxygen absorption from turbulent air by a concurrently flowing liquid film depends on the wave regime.

The effect of wave regimes on gas absorption has been investigated for other types of flow. Muenz and Marchello (1966) showed that the rate of gas absorption into a horizontal water layer depends on the frequency of small amplitude waves generated at the interface. For the case of standing waves in a horizontal layer, Goren and Mani (1968) showed experimentally that the mass transfer rate increases with wave amplitude. Back and McCready (1988) investigated sheared thin films flowing under concurrent gas flows; they found that the saturation coefficient $k_{sat}$ (1) strongly depends on the wave regime for Reynolds numbers in the range $Re = 25$– 2500.

A large majority of the theoretical studies that have examined gas absorption into falling films have employed the so-called penetration theory, which corresponds to solutions of the diffusion equation in one dimension. This theory provides the following prediction for the mass transfer