Semiempirical Equation for Liquid-Phase Mass-Transfer Coefficient for Drops

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The absorption of gases into drops is of great importance to many fields, including scrubber and atmospheric systems. Some experimental works on this subject have been reported. Garner and Lane (1959), in their study on the absorption of CO_2 into water drops using two drop diameters, 0.422 and 0.585 cm, found that absorption rates were greater than those predicted by the fully circulation model of Kronig and Brink (1950). Kaji et al. (1985) measured the liquid-phase physical mass-transfer coefficient for SO_2 absorption by pure water drops of 0.22 cm diameter. They found that the coefficients decreased slowly from 0.0350 to 0.0275 cm/s when the falling drops accelerated through the absorption column. Their data agreed with those predicted by Handlos and Baron (1957) only at long contact times. Altwicker and Lindhjem (1988) performed the absorption of CO_2 by water drops of 0.06 and 0.12 cm diameter. They also found that absorption rates were greater than those predicted by the model of Kronig and Brink, and liquid-phase mass-transfer coefficients were independent of drop falling velocity. They compared their experimental mass-transfer coefficients and those obtained by Garner and Lane (1959) and Kaji et al. (1985) with three theoretical models: Handlos and Baron (1957), Angelo et al. (1966), and Ruckenstein (1967). They found that among the three models the surface stretch model of Angelo et al. (1966) seemed to work best over the entire drop size range. Recently, Huo (1991) studied the absorption of SO_2 into drops of 1.0 N HCl solution with drop diameters of 0.279 and 0.444 cm and gas-phase SO_2 concentrations of 3,000 and 9,000 ppm. He found that the liquid-phase physical mass-transfer coefficients were nearly independent of gas-phase SO_2 concentration, drop falling velocity and contact time, and varied between 0.024 and 0.030 cm/s and 0.028 and 0.029 cm/s for drops of 0.279 and 0.444 cm dia., respectively. His experimental results agreed better with the model of Angelo et al. (1966).

The surface stretch model for oscillating drops proposed by Angelo et al. (1966) postulated that during drop oscillation the fresh surface area is formed and returned to the bulk, which is assumed to be well mixed. In terms of k_L^o, the liquid-phase mass-transfer coefficient, their model can be expressed as:

$$k_L^o = \frac{2}{\sqrt{\pi}}\,[fD(1+\epsilon+3\epsilon^2/8)]^{1/2} \qquad (1)$$

where f is the oscillation frequency of drop, D is the diffusivity of the absorbed gas in the liquid, and ϵ is the distortion parameter which represents the maximum difference in surface areas between the actual drop and a perfect sphere having the same drop volume. Frequency f can be predicted from the equation given by Lamb (Clift et al., 1978). His equation for drops falling in gases reduces to:

$$f = \left[\frac{8\sigma}{3\pi m}\right]^{1/2} \qquad (2)$$

where σ is the surface tension of liquid and m is the mass of the drop. Combining Eqs. 1 and 2 yields:

$$k_L^o = \frac{4}{\pi}\left[\frac{\sigma}{\rho}\right]^{1/4}[D(1+\epsilon+3\epsilon^2/8)]^{1/2}d^{-3/4} \qquad (3)$$

It can be seen from Eq. 3 that k_L^o is proportional to drop diameter with an exponent of $-3/4$. This relation between k_L^o and drop diameter, however, has not been confirmed experimentally.

In this work, we used the data reported in the literature to study the relation between the liquid-phase mass-transfer coefficient and drop diameter, and proposed a semiempirical equation which is based on the surface stretch model to correlate the experimental data available.

Results and Discussion

According to Eq. 3, the experimental data can be plotted in terms of $k_L^o/[4/\pi(\sigma/\rho)^{1/4}D^{1/2}]$ vs. drop diameter in logarithmic scale to find the relation between k_L^o and drop diameter. The experimental data we used, their experimental conditions as well as the physical properties are summarized in Table 1. The

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Table 1. Experimental Conditions, k_L^o, and Physical Properties

Investigators	Gas-Drop	d(cm)	T(°C)	k_L^o(cm/s)	$D\times10^5$(cm^2/s)	σ(dyne/cm)	ρ(g/cm^3)
Altwicker &	CO_2-Water	0.060	23±1	0.101±0.025	1.90	72.28	0.997
Lindhjem (1988)	CO_2-Water	0.120	23±1	0.059±0.005	1.90	72.28	0.997
Kaji et al. (1985)	SO_2-Water	0.220	20	0.028~0.035	1.40	72.75	0.998
Huo (1991)	SO_2-HCl(lN)	0.279	29.5	0.024~0.030	1.87	71.27	0.996
Garner & Lane (1959)	CO_2-Water	0.422	20	0.018~0.030	1.85	72.75	0.998
Huo (1991)	SO_2-HCl(lN)	0.444	29.5	0.028~0.029	1.87	71.27	0.996
Garner & Lane (1959)	CO_2-Water	0.585	20	0.011~0.029	1.85	72.75	0.998

surface tension of 1.0 N HCl solution was taken as that of pure water (Dean, 1973). The diffusivity of SO_2 in water is 1.65×10^{-5} cm^2/s at 25°C (Roberts and Friedlander, 1980). Diffusivities at other temperatures were calculated using the Stokes-Einstein equation, $D\mu_L/T=$constant. The diffusivity of SO_2 in 1.0 N HCl solution was taken as that in pure water. The diffusivities of CO_2 in water were those reported in the referred articles (see Table 1). As shown in Figure 1, the solid line obtained from the least-squares fit of experimental data has a coefficient of correlation of 0.90 and a slope of -0.70 ± 0.10, which is close to the value of $-3/4$ predicted by Eq. 3. The results of Eq. 3 with $\epsilon=0$ and 0.3 are also plotted in Figure 1. It can be seen that both lines are almost parallel to the least-squares fit line and lie above that line. Thus, although Eq. 3 gives the correct relation between k_L^o and d, it overpredicts the k_L^o value. By comparing the difference between the results for the cases of $\epsilon=0$ and $\epsilon=0.3$ with the range of data scattering, one can see that k_L^o predicted by the surface stretch model is relatively insensitive to ϵ.

Based on the above discussions, Eq. 1 can be modified as:

$$k_L^o=\alpha\frac{2}{\sqrt{\pi}}[fD]^{1/2} \tag{4}$$

where α is a correction factor, and ϵ in Eq. 1 has been set to zero. Combining Eqs. 2 and 4 yields:

$$k_L^o=\alpha\frac{4}{\pi}\left[\frac{\sigma}{\rho}\right]^{1/4}D^{1/2}d^{-3/4} \tag{5}$$

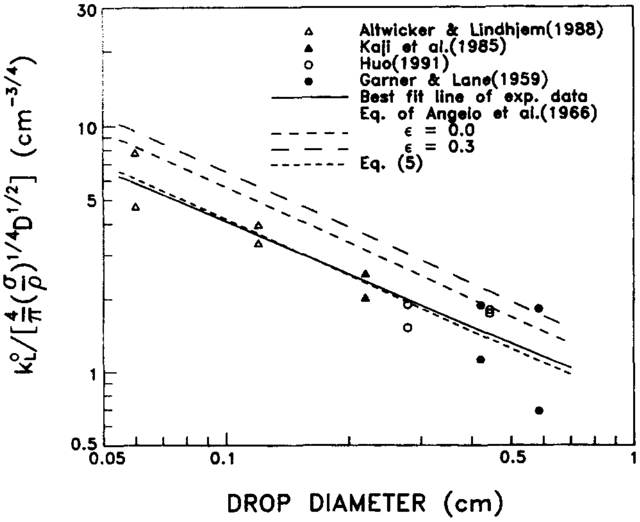

Figure 1. Variation of liquid-phase mass-transfer coefficient with drop diameter.

From the least-squares fit of data using Eq. 5, the correction factor, α, was found to be 0.78 ± 0.21. As shown in Figure 1, the line representing Eq. 5 is almost coincidental with the best fit line of experimental data.

Altwicker and Lindhjem (1988) have mentioned that the surface stretch model matched the data much better if the frequency of oscillation was approximately one-half that predicted by Eq. 2. Their finding implies a correction factor of 0.71, which is close to the value obtained in this work.

Thus, we may conclude that for the prediction of liquid-phase mass-transfer coefficients for drops in the size range 0.06 to 0.6 cm dia., a semiempirical equation which is based on the surface stretch model and best represents the data available is:

$$k_L^o=0.88[fD]^{1/2} \tag{6}$$

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Notation

D = diffusivity of CO_2 or SO_2 in liquid, cm^2/s
d = diameter of drop, cm
f = frequency of drop oscillation, 1/s
k_L^o = liquid-phase physical mass-transfer coefficient, cm/s
m = mass of droplet, $(\pi/6)d^3\rho$, g
S_{max} = maximum surface area during drop oscillation, cm^2
S_o = surface area of spherical drop, cm^2
T = absolute temperature, K
u = velocity, cm/s

Greek letters

α = correction factor
ϵ = distortion parameter, $(S_{max}/S_o)-1$
μ_L = viscosity of liquid, cp
ρ = density of liquid, g/cm^3
σ = surface tension of liquid, dyne/cm

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