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Gas Absorption by Non-Newtonian Fluids in Agitated Vessels

Carbon dioxide was absorbed by aqueous Carbopol solutions in a turbine-agitated vessel for the cases of absorption across an unbroken interface and absorption with the gas bubbling through the liquid. The rheological behavior of the solutions was described by the non-Newtonian power law model with flow behavior indices varying from 0.92 to 0.59. An effective viscosity technique which had previously been developed to correlate agitated vessel rates of viscous dissipation and heat transfer with power law pseudoplastic fluids was used to correlate the data.

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SCOPE

Gas absorption in mechanically agitated vessels is used in a variety of industrial applications and has been studied extensively for Newtonian liquids in various types of apparatus. In the work described here, gas absorption by non-Newtonian pseudoplastic liquids in agitated vessels is studied. The particular system chosen for the study is carbon dioxide absorption in various aqueous Carbopol (carboxy polymethylene) solutions. The rheological behavior of these solutions is well described by the Ostwald-de Waele power law model with the flow behavior index varying between 0.92 and 0.59. The agitated vessel geom-

etry corresponded to the Standard Tank Configuration (Holland and Chapman, 1966); the impeller used was a six flat blade turbine.

Two methods of gas-liquid contact were studied. In the first method, the gas was introduced into the absorption vessel above the gas-liquid interface. This technique provided an unbroken gas-liquid interface of known area. In the second case, mass transfer occurred in a gas-liquid dispersion caused by introducing the gas through a sparge tube located in the liquid directly below the impeller.

CONCLUSIONS AND SIGNIFICANCE

The gas absorption data were successfully correlated using an effective viscosity technique which previously had been developed to correlate rates of viscous dissipation and heat transfer for pseudoplastic liquids. The effective viscosity is defined as the ratio of the shear stress

corresponding to an average shear rate existing in the agitated vessel to this average shear rate. The average shear rate is empirically found to be proportional to the impeller speed. For the turbine impeller used in the experiments the effective viscosity is given by Equation (8).

The effective viscosity given by Equation (8) replaces the liquid viscosity in the correlations previously developed for Newtonian liquids.

For gas absorption across an unbroken interface the data are correlated with a standard deviation of 8.5% by

$$\frac{k_c d}{D} = 5.11 \times 10^{-3} \left(\frac{d^2 N \rho}{\mu_e} \right)^{0.926} \left(\frac{\mu_e}{\rho D} \right)^{0.5}$$

For absorption in a mechanically agitated gas-liquid dis-

persion in the absence of surface foam the data are correlated with a standard deviation of 5.0% by

$$\frac{k_c a d^2}{D} = 21.2 \left(\frac{d^2 N \rho}{\mu_e} \right)^{1.11} \left(\frac{\mu_e}{\rho D} \right)^{0.5} \left(\frac{dV_s}{\sigma} \right)^{0.447} \left(\frac{\mu_g}{\mu_e} \right)^{0.694}$$

The exponents found in these equations are in agreement with published work for Newtonian liquids.

Gas absorption is important in a number of industrial processes. One of the major types of equipment used to accomplish this unit operation is the mechanically agitated vessel. The purpose of the research reported here was to study gas absorption into power law non-Newtonian liquids in agitated vessels.

There are many industrially important fluids such as rubbers, solutions of various polyesters, aqueous clay suspensions, surface coating materials and solutions of cellulose and its derivatives whose rheological behavior is non-Newtonian and whose shear-stress shear-rate relationships can be fairly well represented by the Ostwald-de Waele power law relationship

$$\tau = K \dot{\gamma}^n \quad (1)$$

The liquids used in this study were various aqueous Carbopol-934 (carboxy polymethylene) solutions chosen because of their close adherence to Equation (1). The gas used for the absorption studies was carbon dioxide, chosen because of its relatively high solubility since high solubilities result in larger mass transfer rates. The vessel geometry was designed to conform to the Standard Tank Configuration as described by Holland and Chapman (1966); the agitation was provided by a six flat blade turbine impeller. Two different cases were studied. In the first, the gas was introduced into the absorption vessel above the gas-liquid interface (Unbroken Interface Case). In the second, mass transfer occurs in a gas-liquid dispersion caused by introducing the gas through a sparge tube located in the liquid directly below the impeller (Bubbling Case).

There have been several studies of gas absorption by agitated Newtonian liquids across an unbroken gas-liquid interface. Kozinski and King (1966) have summarized this work and have obtained data of their own. A dimensional analysis indicates that the mass transfer coefficient may be correlated by an equation of form

$$\frac{k_c d}{D} = C \left(\frac{d^2 N \rho}{\mu} \right)^b \left(\frac{\mu}{\rho D} \right)^c \quad (2)$$

Sideman et al. (1966) provide a review of mass transfer in mechanically agitated gas-liquid dispersions. For this case the gas flow rate and the physical properties of the gas will affect the mass transfer rate. These authors point out the form that the mass transfer correlations take for the bubbling case.

$$\frac{k_c a d^2}{D} = C \left(\frac{d^2 N \rho}{\mu} \right)^b \left(\frac{\mu}{\rho D} \right)^c \left(\frac{dV_s}{\sigma} \right)^e \left(\frac{\mu_g}{\mu} \right)^f \quad (3)$$

The term on the left side of Equation (3) is a modified Sherwood number which includes the effective interfacial area; the terms on the right side are the impeller Reynolds number, the Schmidt number, a dimensionless gas flow group, and the ratio of dispersed phase and continuous phase viscosities.

METHOD OF CORRELATION

Equations (2) and (3) cannot be used directly to correlate mass transfer data with non-Newtonian liquids since the liquid viscosity which appears in Equations (2) and (3) is not defined for non-Newtonian liquids. However in this work it is proposed to use an effective viscosity as used by Skelland and Dimmick (1969) and Sandall and Patel (1970) in correlating heat transfer rates to non-Newtonian liquids in agitated vessels.

In this correlation method, an effective viscosity based on an average shear rate in the agitated vessel as proposed by Metzner and Otto (1957) is defined as

$$\mu_e = \frac{\tau(\dot{\gamma}_a)}{\dot{\gamma}_a} \quad (4)$$

Metzner and Otto found that in applying Equation (4) to correlate rates of viscous dissipation in agitated vessels, the average shear rate $\dot{\gamma}_a$ could be simply related to the impeller speed by

$$\dot{\gamma}_a = A_s N \quad (5)$$

where A_s is a proportionality constant. Calderbank and Moo-Young (1961) extended the Metzner-Otto approach to allow the proportionality constant in Equation (5) to be a function of the flow behavior index n

$$\dot{\gamma}_a = B N \left(\frac{4n}{3n+1} \right)^{\left(\frac{n}{1-n} \right)} \quad (6)$$

For pseudoplastic fluids, Calderbank and Moo-Young determined the value of the constant B by matching power numbers for non-Newtonian agitation to the corresponding correlation for Newtonian agitation. The reported value for B for turbines and propellers is

$$B = 11 \pm 10\% \quad (7)$$

The effective viscosity for the liquids and impeller used in this work is obtained from Equations (1), (4), (6), and (7).

$$\mu_e = K(11N)^{n-1} \left(\frac{3n+1}{4n} \right)^n \quad (8)$$

The effective viscosity given by Equation (8) replaces the liquid viscosity in the mass transfer correlations, Equations (2) and (3).

EXPERIMENTAL APPARATUS AND PROCEDURE

A schematic diagram of the gas absorption apparatus is shown in Figure 1. The absorption vessel consisted of a 30.5-cm long section of 15.2-cm inside diameter glass pipe sealed on each end with an O-ring and flange. The vessel was equipped with four baffles and was filled with liquid to a depth of 15.2 cm in conformance with Standard Tank Configuration (Hol-

land and Chapman, 1966). Separate agitators for the gas and liquid phases were mounted on a single shaft which went through an O-ring seal in the top flange. The gas phase was agitated to ensure that there would not be a build-up of inert gas in the absorption vessel. The agitators were driven by a ¼ HP variable speed motor. The liquid agitator was a 5.1-cm diameter, six flat blade turbine impeller located 5.1 cm from the vessel bottom. Table 1 lists the important geometrical characteristics of the absorption apparatus. The gas was introduced into the vessel in two different ways. In the first case (Unbroken Interface Case), the gas entered the vessel above the liquid surface. In the second case (Bubbling Case), the gas was introduced through a 4-mm I.D. sparge tube located 3.2 cm directly below the turbine impeller. The liquids used in the experiments were degassed by recycling them through a 10.2-cm diameter desorption column packed to a height of 121.9 cm with 8-mm glass Raschig rings.

Fifty ml. liquid samples were withdrawn from the inlet and outlet streams approximately 30 min. after adjusting the impeller speed. As a check for steady state a second set of samples were usually taken 15 min. later. The solutions were analyzed for carbon dioxide by precipitation as barium carbonate from a solution of barium chloride and standard sodium hydroxide; the excess sodium hydroxide was titrated to a phenolphthalein end point with hydrochloric acid.

Liquid and gas flow rates were determined by rotameters. The impeller speed was measured using a stroboscope. Further details may be found in the original thesis (Perez, 1973).

PHYSICAL PROPERTIES

The densities of all solutions were measured at 24°C to within ±0.001 g/ml by weighing and were found to be identical within experimental accuracy to the density of water. Thus the densities of all solutions at temperatures other than 24°C were taken to be those of water. Vapor pressures of all aqueous solutions were assumed to be the same as for pure water and were taken from Perry et al. (1963). The surface tension of all solutions was measured against carbon dioxide gas at 23°C using a du Nouy ring tensiometer. The temperature correction for the surface tension of all solutions was assumed to be the same as that which applies to the surface tension of water against air as obtained from the Handbook of Chemistry and Physics (1970). The rheological properties of the Carbopol solutions were determined from capillary tube viscometer measurements. Carbon dioxide solubilities in water and aqueous Carbopol solutions were determined experimentally (Perez and Sandall, 1974). The molecular

TABLE 1. AGITATED VESSEL EQUIPMENT SPECIFICATIONS

(A) Glass pipe:	
Length, cm	30.48
Inside diameter, cm	15.24
(B) Copper baffles:	
Length, cm	20.32
Width, cm	1.52
Thickness, cm	0.1587
(C) Liquid impeller;	
6 flat blade disk turbine:	
Stirring shaft diameter, cm	1.27
Impeller diameter, cm	5.08
Disk diameter, cm	3.81
Height from tank bottom, cm	5.08
Blade width, cm	1.02
Blade length, cm	1.27
Length of blade on disc, cm	0.64
(D) Vessel tubes:	
Outside diameter, cm	0.64
Inside diameter, cm	0.41
(E) Gas sparging tube:	
Inside diameter, mm	4.0
Height below impeller, cm	3.18

TABLE 2. PHYSICAL PROPERTIES

Liquid	Surface tension against carbon dioxide @ 23°C, dynes/cm	Diffusion coef. @ 25°C, cm ² /s × 10 ⁵	Henry's constant (mmHg cc/gmole) × 10 ⁻⁷		
			24°C	30°C	35°C
Water	69.94	1.98	2.171	2.522	2.843
0.25% Carbopol	65.08	2.09	2.222	2.639	2.943
0.75% Carbopol	64.44	1.74	2.816	3.337	3.732
1.00% Carbopol	63.43	1.58	3.407	4.096	4.751

TABLE 3. RHEOLOGICAL PROPERTIES

Liquid	Temperature, °C	Flow behavior index, <i>n</i>	Consistency index, <i>K</i> (g/cm s ²⁻ⁿ)
0.25% Carbopol	24	0.916	0.0428
	30	0.916	0.0379
	35	0.916	0.0355
0.75% Carbopol	24	0.773	0.507
	30	0.773	0.483
	35	0.773	0.469
1.00% Carbopol	24	0.594	5.29
	30	0.594	4.83
	35	0.594	4.12

diffusion coefficients of carbon dioxide gas in water and in various Carbopol solutions were determined experimentally in a wetted wall column (Perez and Sandall, 1973). Table 2 lists the important physical properties; the power law parameters are given in Table 3.

RESULTS

Mass transfer coefficients were calculated for both cases based on the assumption of complete mixing in the liquid. For the Unbroken Interface Case the area for mass transfer is known and the mass transfer coefficient is calculated from

$$k_c = \frac{L}{A} \left[\frac{C_2 - C_1}{\frac{H}{p} - C_2} \right] \quad (9)$$

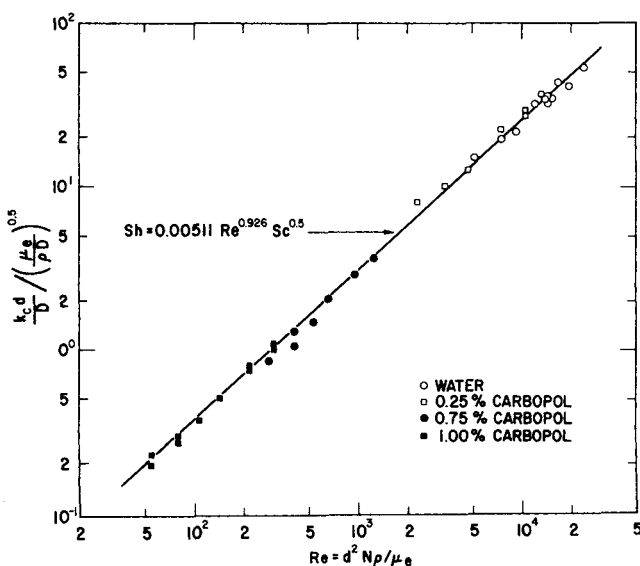


Fig. 1. Schematic drawing of absorption apparatus.

For the Bubbling Case the interfacial area is unknown and therefore cannot be separated from the mass transfer coefficient. The volumetric mass transfer coefficient in this case is determined from

$$k_c a = \frac{L}{V} \left[\frac{C_2 - C_1}{\frac{H}{p} - C_2} \right] \quad (10)$$

Figure 2 shows the mass transfer coefficients for the Unbroken Interface Case as a function of impeller speed. As can be seen, there exists two separate regions with respect to effect of the agitator. The first region is one where the agitator has no effect on the rate of mass transfer. For agitator speeds below approximately 100 rev./min. the mass transfer coefficient is relatively constant. Free convection is more important than forced convection at these low speeds. This critical value of the agitator speed agrees with the data of Hutchinson and Sherwood (1937) for absorption into water in a similar apparatus. The data shown in Figure 2 are for a constant liquid flow rate which was different for each solution used. It was found, however, that the mass transfer coefficient is not a function of liquid flow rate.

The volumetric mass transfer coefficients obtained for the Bubbling Case are shown as a function of agitator speed in Figure 3. The data shown in Figure 3 are for a superficial gas velocity of 0.162 cm/s through the sparge tube. Agitator speeds less than about 200 rev./min. have little effect on the absorption rate in this case. This is in approximate agreement with the data of Mehta and Sharma (1971) for chemical absorption into aqueous solutions. For these low agitation rates the interfacial area is unaffected by impeller speed.

The mass transfer rates for the 0.75 and 1.00 wt. % Carbopol solutions shown in Figure 3 for the Bubbling

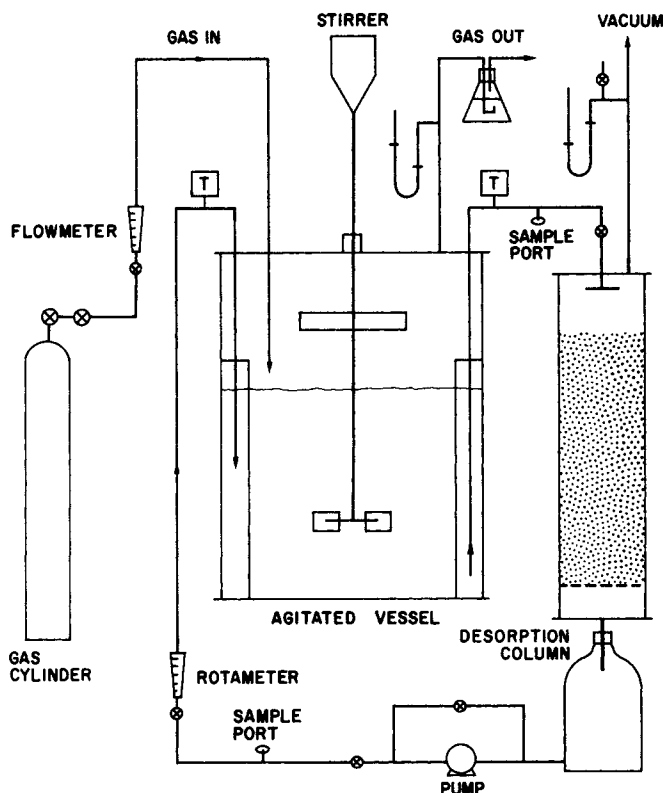


Fig. 2. Unbroken interface mass transfer coefficient as a function of impeller speed.

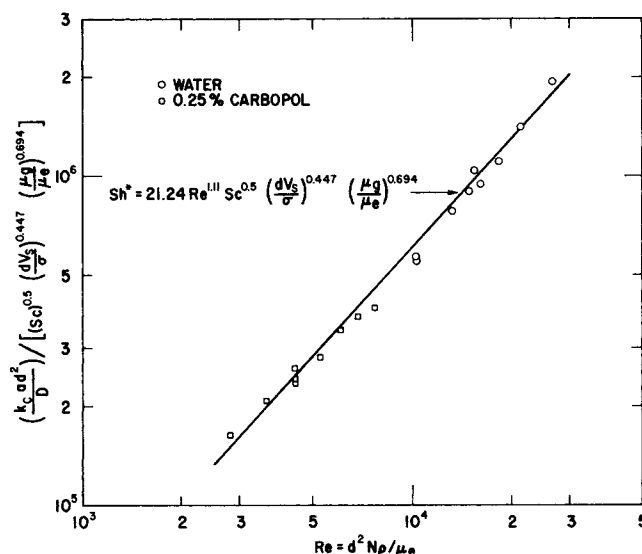


Fig. 3. Bubbling case volumetric mass transfer coefficient as a function of impeller speed.

TABLE 4. UNBROKEN INTERFACE CORRELATION RESULTS
 $Sh = CRe^b Sc^c$

C	b	c	Mean deviation, %	Standard deviation, %
7.40×10^{-3}	0.901	0.476	6.74	8.38
5.11×10^{-3}	0.926	0.500*	6.69	8.47

* Fixed.

TABLE 5. 95% CONFIDENCE LIMITS ON CORRELATION PARAMETERS FOR UNBROKEN INTERFACE CASE
 $Sh = CRe^b Sc^c$

Fitted parameter	Value	95% confidence limits
C	7.40×10^{-3}	$\pm 1.74 \times 10^{-4}$
b	0.901	± 0.090
c	0.476	± 0.086

Case are higher than would be expected when compared to the pure water and 0.25 wt. % solution results. This is due to the formation of foam for these solutions. Since the foam provides an extremely large area for mass transfer and since there was a constant foam collapse and renewal, abnormally high mass transfer rates were measured for the 0.75 and 1.00 wt. % solutions.

CORRELATION OF RESULTS

The mass transfer data for the Unbroken Interface Case obtained at impeller speeds greater than 100 rev./min. in the region where agitation has an effect were fitted to Equation (2) with the liquid viscosity replaced by the effective viscosity expression, Equation (8). The results of the least squares analysis are given in Tables 4 and 5. The mean and standard deviations shown in Table 4 are defined as

$$\text{mean deviation} = \frac{100}{N} \sum_{i=1}^N \left| \frac{Sh_{\text{calc}} - Sh_{\text{obs}}}{Sh_{\text{calc}}} \right|_i \quad (11)$$

standard deviation

$$= 100 \left[\frac{1}{N-1} \sum_{i=1}^N \left(\frac{Sh_{calc} - Sh_{obs}}{Sh_{calc}} \right)^2 \right]^{1/2} \quad (12)$$

The Higbie (1935) penetration theory or the Danckwerts (1951) surface renewal theory are commonly accepted models for liquid phase mass transfer across a gas-liquid interface, and both of these models predict an exponent of 0.5 on the Schmidt number in Equation (2). Since an exponent of 0.5 lies within the 95% confidence limits for the Schmidt number exponent as shown in Table 5, c was fixed at 0.5 and the data were again fitted to Equation (2). The resulting correlation is

$$Sh = 5.11 \times 10^{-3} Re^{0.926} Sc^{0.5} \quad (13)$$

The data fit Equation (13) with a standard deviation of 8.5%. The data are shown plotted according to Equation (13) in Figure 4.

The summary of previous investigations given by Kozinski and King (1966) for liquid phase mass transfer across a contiguous gas-liquid interface shows that the exponent on the Reynolds number varies from 0.75 to 1.2 for Newtonian liquids in baffled tanks. Thus, the exponent of 0.926 found in this work compares favorably with previous work.

A total of 53 data points in Region II of Figure 2 were used in obtaining Equation (13). The range in the variables covered in the experiments is given in Table 6.

Since the mass transfer coefficient data indicate a square root dependence on the diffusivity, the data can be interpreted in terms of the Danckwerts surface renewal theory. For surface renewal theory the mass transfer coefficient is related to the fractional rate of surface renewal s by the expression

$$k_c = \sqrt{Ds} \quad (14)$$

Comparing Equations (13) and (14) gives an expression for the rate of surface renewal.

$$s = 2.61 \times 10^{-5} \left(\frac{\mu_e}{\rho d^2} \right) Re^{1.852} \quad (15)$$

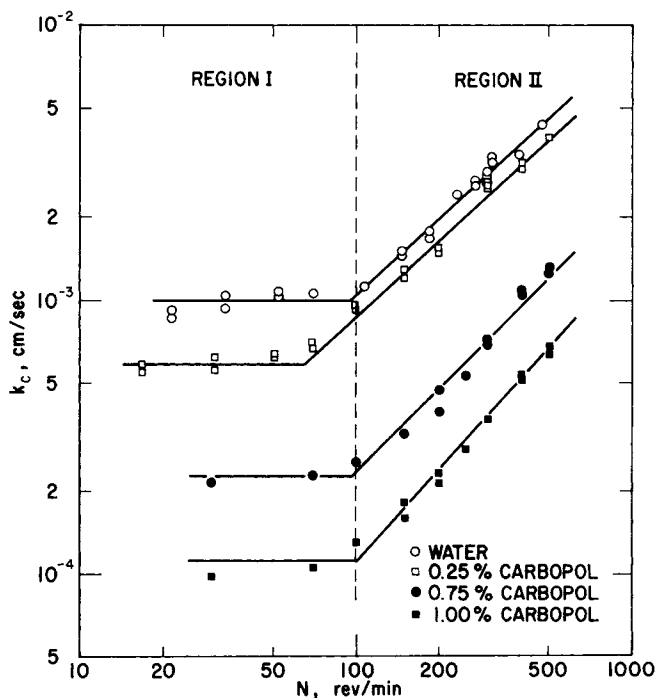


Fig. 4. Unbroken interface mass transfer correlation.

TABLE 6. RANGE OF VARIABLES FOR UNBROKEN INTERFACE CASE CORRELATION

Variable	Range
Speed, N (rev/min)	100-500
Flow behavior index, n	0.594-1.00
Consistency index, K (g/cm-s ²⁻ⁿ)	0.009-4.5
Sherwood number, Sh	56.2-1,160
Reynolds number, Re	54.5-23,800
Schmidt number, Sc	454-81,900

TABLE 7. BUBBLING CASE CORRELATION RESULTS

$$Sh^* = C Re^b Sc^c \left(\frac{dV_s}{\sigma} \right)^e \left(\frac{\mu_g}{\mu_e} \right)^f$$

C	b	c	e	f	Mean deviation, %	Standard deviation, %
19.66	1.11	0.538	0.447	0.730	2.86	4.97
21.24	1.11	0.500*	0.447	0.694	2.86	4.97

* Fixed.

TABLE 8. 95% CONFIDENCE LIMITS ON CORRELATION PARAMETERS FOR UNBROKEN INTERFACE CASE

$$Sh^* = C Re^b Sc^c \left(\frac{dV_s}{\sigma} \right)^e \left(\frac{\mu_g}{\mu_e} \right)^f$$

Fitted parameter	Value	95% confidence limits
C	19.66	± 0.68
b	1.11	± 0.247
c	0.538	± 0.283
e	0.447	± 0.218
f	0.730	± 0.259

The mass transfer data for the Bubbling Case obtained for $N > 200$ rev./min. in Region II of Figure 3 were fitted to Equation (3) with the liquid viscosity given by Equation (8) using a least squares procedure. The data for the 0.75 and 1.00 wt. % Carbopol solutions were excluded from this correlation because of the foam formation difficulties experienced with these solutions. The results of this data fitting are given in Tables 7 and 8. As was the case for the nonbubbling data correlation, an exponent on the Schmidt number of 0.5 is within the 95% confidence limits for this parameter. If the exponent on the Schmidt number is fixed at 0.5 the resulting correlation is

$$Sh^* = 21.2 Re^{1.11} Sc^{0.5} \left(\frac{dV_s}{\sigma} \right)^{0.447} \left(\frac{\mu_g}{\mu_e} \right)^{0.694} \quad (16)$$

The data fit Equation (16) with a standard deviation of 5.0%. The standard deviation from Equation (16) increases to 255% if the 0.75 and 100 wt. % Carbopol data are included. Figure 5 shows the data plotted according to Equation (16). A total of 24 data points for water and the 0.25 wt. % Carbopol solution in Region II of Figure 3 were used to obtain Equation (16). The range of variables covered in the experiments is given in Table 9.

For mass transfer in an agitated gas-liquid dispersion Calderbank (1959) has concluded that the mass transfer coefficient k_c is independent of gas flow rate and liquid agitation rate. Thus, the dependence of the modified Sherwood number on these variables as given by Equation (16) indicates their effect on the effective interfacial area.

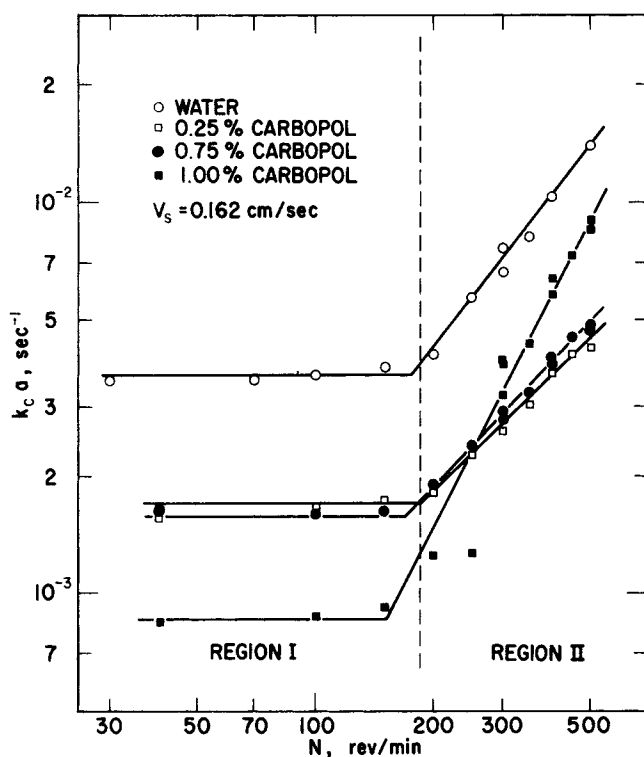


Fig. 5. Bubbling case mass transfer correlation.

TABLE 9. RANGE OF VARIABLES FOR BUBBLING CASE CORRELATION
(Water and 0.25 wt. % Carbopol)

Variable	Range
Speed, N (rev./min)	200-500
Flow behavior index, n	0.916-1.00
Consistency index, K	0.009-0.04
Superficial gas velocity, cm/s	0.162-0.466
Modified Sherwood number, Sh^*	2,340-20,900
Reynolds number, Re	2,810-26,700
Schmidt number, Sc	455-1,490
Gas flow group, dV_s/σ	1.89×10^{-5} - 2.10×10^{-4}
Viscosity ratio, μ_g/μ_e	0.005-0.019

NOTATION

a	= effective interfacial area per unit volume, cm^{-1}
A	= interfacial area for Unbroken Interface Case, cm^2
A_s	= proportionality constant in Equation (5)
b	= exponent on Reynolds number in Equations (2) and (3)
B	= proportionality constant defined by Equation (7)
c	= exponent on Schmidt number in Equations (2) and (3)
C	= proportionality constant in Equations (2) and (3)
C_1	= concentration of carbon dioxide in inlet liquid, g moles/ cm^3
C_2	= concentration of carbon dioxide in outlet liquid, g moles/ cm^3
d	= impeller diameter, cm
D	= diffusion coefficient, cm^2/s
e	= exponent on gas flow group in Equation (3)
f	= exponent on viscosity ratio in Equation (3)
H	= Henry's constant, mmHg cc/g mole
k_c	= mass transfer coefficient, cm/s
K	= consistency index, $\text{g}/\text{cm s}^{2-n}$

L	= liquid flow rate, cm^3/s
n	= flow behavior index
N	= impeller speed, rev/s
p	= partial pressure of carbon dioxide, mmHg
Re	= Reynolds number = $d^2 N \rho / \mu_e$
s	= fractional rate of surface renewal, s^{-1}
Sc	= Schmidt number = $\mu_e / \rho D$
Sh	= Sherwood number = $k_c d / D$
Sh^*	= Sherwood number for bubbling case = $k_c a d^2 / D$
V_s	= superficial gas velocity through sparge tube, cm/s

Greek Letters

γ	= shear rate, s^{-1}
γ_a	= average shear rate, s^{-1}
μ	= viscosity, $\text{g}/\text{cm s}$
μ_e	= effective viscosity defined by Equation (4), $\text{g}/\text{cm s}$
μ_g	= gas viscosity, $\text{g}/\text{cm s}$
ρ	= liquid density, g/cm^3
σ	= surface tension, dynes/cm
τ	= shear stress, dynes/ cm^2

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