FILM ABSORPTION IN LIQUIDS OF DIFFERENT VISCOSITIES

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Received February 26th, 1980

Experimental results on absorption of CO_2 in liquid film flowing downwards the string of spheres are presented for liquids with viscosities in the range from 1 to 10·1 mPa s. The results correspond qualitatively to the Levich's analysis of mass transfer into the flowing film with wavy surface. Equation relating dimensionless numbers is proposed for calculation of the volumetric mass transfer coefficient.

Kinetics of physical absorption in the liquid film, where the viscosity of liquid differs significantly from that of water at temperatures close to 20° C has not yet been studied thoroughly, even though the problem is industrially significant. Onda¹ and coworkers have published evaluation of data for absorption in the bed of packing in solvents whose viscosities were from 0.8 to 1.5 mPa s. A considerably wider range of viscosities of solvents has been used in experiments on laboratory types of absorbers. The absorption has been performed for viscosities of solvents on the dire column² in the range from 0.6 to 3.0 mPa s, on modification of the dire column³⁻⁵ from 0.4 to 25 mPa s, and on the string of spheres⁶ from 0.6 to 1.0 mPa s, on wetted pipe⁷ from 0.9 to 2.9 mPa s and on the series of inclined surfaces⁸ from 1.0 to 3.4 mPa s.

Comparison of results of various authors can be made by use of dimensionless relations for the mass transfer coefficient. This relation for absorption in the packed column has the form:

$$St_{sv} = C \left(\frac{a_s}{a_1}\right)^{\beta_1} \left(\frac{a_w}{a_1}\right)^{\beta_2} (a_i d_p)^{\beta_3} \operatorname{Re}^{\beta_4} \operatorname{Sc}^{\beta_5} \operatorname{Ga}^{\beta_6} \operatorname{We}^{\beta_7}$$
(1)

and the values of coefficients C and β_i resulting from the literature data are given in Table I. The kinetics of absorption in laboratory absorbers can be expressed by a similar equation:

$$St_{sm} = C' \left(\frac{u_s}{d_p}\right)^{\beta'_1} \operatorname{Re}^{\beta'_2} Sc^{\beta'_3} \operatorname{Ga}^{\beta'_4} We^{\beta'_5}$$
(2)

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and the published experimental results supply the values C' and β'_i which are given in Table II.

From the data given in the tables is obvious a considerable nonuniformity of the experimental results. This leads to relatively great differences in the estimate of the mass transfer coefficient in the case the viscosity of the solvent significantly differs from that of water. Viscosity changes affect both molecular diffusivity and convection. The opinion of various authors on the effect of surface tension differs and frequently is not taken into consideration.

It is advantageous to study the effects of individual factors on a simple geometry. In a recent study on absorption in the wetted wall column, Chung and Mills⁷ have assumed the validity of the relation

$$Sh = f \operatorname{Re}^{n} \operatorname{Sc}^{1/2} \operatorname{Ga}^{1/3},$$
 (3)

where, unlike other authors, they found that the exponent with the Reynolds number depends on viscosity. The factor f is not constant and has been expressed in the mentioned study graphically in dependence on the modified form of the Weber number. But the authors mention the small range of surface tensions of the studied liquids. The measurements were performed at large liquid flow rates mostly in the region of turbulent regime of the liquid film.

In this study are presented experimental results on absorption in the liquid film flowing down the string of spheres which is a system with a simple geometry and simulates conditions in the bed of packing better than the wetted wall column.

EXPERIMENTAL

Apparatus. The used laboratory model of the column, Fig. 1, formed by one or seven columns of spheres has the basic parameters identical with the model described in the earlier study⁶. Description of individual elements is given in the legend to the figures. The construction changes made enabled simpler dismantling and operation and were necessary also due to the combination of construction materials steel-glass-teflone²². Basic geometric parameters: diameter of spheres 15 mm, number of spheres in one column 12, diameter of the cylinder 15 is 600 mm, diameter of cylinder 15 is 600 mm, diameter of cylinder 1 is 80 mm, height of cylinder 1 is 1000 mm, diameter of the central wire 1 mm, total height of the column 180 mm, distance between the opening of the nozzle and the first sphere 10 mm, distance between the upper edge of the liquid collecting tube and the last sphere 10 mm, surface area of the column 848 $\cdot 10^{-3}$ m².

Procedure. The gas and liquid circuits inclusive the desorption part of the unit are in detail described in the earlier study²².

After the liquid and gas temperatures reached steady state values, flow rates of both phases were fixed and perfect watting of the string of spheres was ensured, if necessary, by use of the auxiliary nozzle directed into the space ϑ . The distance of liquid surface below the edge of the liquid collector has been kept about 5 mm. At small flow rates the setting was more accurate with the amplitude 2 to 3 mm. When steady state was reached, four samples were taken successively by the system of pipettes. The liquid flow rate has been determined during the experiment by

Summary of Standardized Correlati	ons for Packing								
Authors	U	β1	β2	β3	β4	βs	β6	βη	Re
Sherwood, Holloway ⁹ Sebulman, Ulrich ¹⁰ Fujita, Hayakawa ¹¹ Onda, Sada, Otubo ¹² Onda, Sata, Sutto ¹³	$C \cdot a_1^{m-2}$ $2 \cdot 51 \cdot 10^{1}$ $6 \cdot 30 \cdot 10^{-2}$ $4 \cdot 64 \cdot 10^{-3}$ $2 \cdot 0 \cdot 10^{-2}$	0-000	0	0 -0.55 -0.64	H 0.50	10-10-10-00	0.05	00000	3-700 3-700 1-500 0.5-500 0.5-500
Onda, Sada, Kido ¹⁵ Mada, Shiboara ¹⁶ Komori, Hirai ¹⁷ Monda, Takeuchi, Koyama ¹⁸ Onda, Takeuchi, Okumolo ¹⁹ Mika ⁶	$\begin{array}{c} 1.05 & 10^{-1} \\ 4.2 & 4.2 \\ 0.016 & 10^{-2} \\ 1.5 & 10^{-3} \\ 5.1 & 10^{-3} \\ 1.5 & 10^{-2} \\ 1.5 & 10^{-2} \end{array}$	0-00000	1 0 0.40 0.17		0.83 0.83 0.83	+11 +11 +11 +11 +11 +11 +11	0 0 0 0 0 0 0	0 0 0 0 0 0 0 0	$\begin{array}{l} 0.5-600\\ 0.3-700\\ 0.2-1250\\ 0.5-500\\ 0.5-600\\ 20-700\\ Re<30\\ \end{array}$
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TABLE I

Authors	ú	β'	β'2	β'3	B'4	β'5	Re	Sc
Yoshida, Koyanagi ²¹ Hikita and coworkers ² Norman, Sammak ³ Norman, Sammak ³ Vitowc ⁵ Vitowc ⁵ Vitowc ⁵ Vitowc ⁵ Vitowc ⁵ Vitowc ² Vyrowc ² Vyroal ²² Vyroal ²²	3.40.10-2 2.14.10-1 3.03.10-1 6.34.10-1 6.34.10-1 2.11.10-2 2.11.10-2 7.06.101 7.06.101 9.50.101 3.37.10-2 3.37.10-2 3.37.10-2	$\begin{array}{c} 0 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\$	0.50 3 0.61 0.47 0.47 0.70 0.70 0.71 0.63 0.63	0 0 0 0 0 0 0 0 0 0 0 0 0 0	0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	0 0 0 0 0-21 0.14	5-200 5-250 0:25-200 0:25-200 5-200 5-200 5-200 5-200 5-200 5-1150 6-1153 6-1153 3-145	250586 218-1 760 108-4 350 108-4 350 123586 123586 123586 123586 123586 123586 5523 200 5523 205

TABLE II Summary of Standardized Correlations of Absorber Models

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measuring the mass of collected water after ending the sampling in the selected time interval. The liquid flow rate of one string has been from $1 \cdot 10^{-3}$ to $9 \cdot 10^{-3}$ kg s⁻¹. All experiments were performed at the gas flow rate $8 \cdot 5 \cdot 10^{-5}$ m³ s⁻¹.

Type of absorbent	10^3 kg m^{-3}	η mPa s	$10^{-9} \text{ m}^2 \text{ s}^{-1}$	c* 10 ⁻² kmol m ⁻³	10^{-2} kg s ⁻²
Glycerol-water	1.068	2.34	0.914	2.89	7.15
Glycerol-water	1.125	5-85	0.429	2.14	6.97
Glycerol-water	1.150	10.1	0.274	1.89	6.87
Water	0-998	1.00	1.82	3.92	7.28
n-Propyl alcohol	0.806	2.22	2.62	11.2	2.38
n-Butyl alcohol	0.810	3.03	2.13	9.13	2.46
n-Amyl alcohol	0.814	4.92	1.88	8.08	2.43
n-Hexyl alcohol	0.819	5.39	1.53	7.69	2.45

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TABLE III Properties of Used Absorbents at 20°C

FIG. 1

Column with the String of Spheres

1 Glass jacket of the column, 2 liquid inlet, 3 liquid outlet, 4 gas inlet, 5 gas outlet, 6 connection to atmosphere, 7 exchangable nozzle, 8 device for fixing of nozzles, 9 inlet nozzle, 10 glass spheres, 11 supporting wire, 12 outlet tube, 13 liquid overflow, 14 thermometer jacket, 15 glass cylinder, 16 homogenisation vessel, 17 upper flange of the column, 18 central part of column, 19 bottom flange of column, 20 lower part of column.



Absorbents used. To cover the intended range of viscosities and surface tensions of absorbents the series of aliphatic alcohols and aqueous solutions of glycerol were considered. Aqueous solutions of glycerol with viscosities 1.00, 2.34, 5.85 and 10.1 mPa s and n-propyl alcohol, n-butylalcohol, n-amyl alcohol and n-hexyl alcohol were used. The needed physical properties of absorbents are given in Table III.

Analytical methods used. The purity of gaseous CO_2 was checked by azotometer. This procedure enabled determination of concentration of inert gases smaller than 0.1 vol.% with the relative error less than 5%. The determined CO_2 concentrations in pressure flasks were within the range from 99-7 to 99-8 vol.%.

For determination of CQ₂ in the liquid phase both the titration method described in the earlier studies^{5,6} for determination of CO₂ in water and its modifiation for the case of organic solvents were used. For low concentrations of CO₂ in the used solvents the basic analytical methods were modified so that the relative error of the method was smaller than 2%. The method is in detail described in the study²².

RESULTS AND DISCUSSION

At small values of the Reynolds number the flow of the liquid film is laminar and if its surface is smooth, the absorption rate is described by the Highbie model of mass transfer

$$k_c = 2(Dv/\pi h)^{1/2}, \qquad (4)$$

where the surface velocity is calculated from the Nusselt relation and the geometric film length corrected for end effects. After substitution and transformation to the dimensionless form from Eq. (4) results

$$K = \frac{k_{\rm c} {\rm Sc}^{1/2}}{(gv)^{1/3}} = 1.15 \ {\rm Re}^{1/3} \ {\rm Ga}^{-1/6} = 1.15 \left(\frac{\Gamma_{\rm v}^2}{g}\right)^{1/6} h^{-1/2}$$
(5)

i.e. the expression for the quantity K is independent of viscosity.

The measurements of the absorption rate of CO_2 into solutions of glycerol in water, performed in the wetted wall column designed according to literature²³ which satisfied the conditions of smooth laminar film have supplied values of K independent of viscosity. The values of the Schmidt number substituted into relation (5), were calculated from diffusivity coefficients obtained from absorption measurements in the laminar jet. This result can be also considered as verification of results of both measurements.

At flow of the liquid film on the string of spheres expansion and contraction of the film takes place, where at the points of contact of individual spheres the film could be mixed. The measurements of the authors of this laboratory apparatus¹⁴ have demonstrated, that the expansion and contraction of the film has no significant effect on the kinetics of absorption so that at small liquid flow rates it is possible to determine the diffusion coefficient by measurements of the absorption rate in the

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film on one sphere. But with the string of spheres, similarly as with a longer wetted wall column the surface of the film usually does not remain smooth and the formed waves significantly affect the kinetics of absorption. This is manifested by the change of the exponent with the Reynolds number in comparison with the value resulting from Eq. (5). If it is possible to consider the dimensionless relation as the product of powers of dimensionless numbers, the exponent with the Galiei number can also change. Instead of Eq. (5) it is then possible to expect the validity of equation

$$K = a \operatorname{Ga}^{\mathfrak{b}} \operatorname{Re}^{\mathfrak{a}}, \tag{6}$$

where a, b and c are empirical coefficients.

In Figs 2 and 3 is plotted the experimentally obtained dependence of K on $\Gamma_{\mathbf{v}}$ for the string of spheres. It is obvious that in logarithmic coordinates the relation $K(\Gamma_{\mathbf{v}})$, can be considered, in certain limits, linear while for small values of $\Gamma_{\mathbf{v}}$ and large viscosities it is possible to observe a fracture and change to the dependence $K(\Gamma_{\mathbf{v}}^{1/3})$ corresponding to the theoretical relation for the laminar film flow with smooth surface. Even though it was not possible for experimental reasons (the necessity of a perfectly wetted surface) to reach at all liquid viscosities the above





Dependence of K on Γ_{v} for Water and Solutions of Glycerol, Absorption on the String of Spheres

○. Water, ④ solution of glycerol: $\eta =$ = 2·34 mPa s, € $\eta = 5.85$ mPa s, ○ $\eta = 10.1$ mPa s.





Dependence of K on Γ_v for Alcohols, Absorption on the String of Spheres \circ n-Propyl alcohol $\eta = 2 \cdot 22$ mPas, \bullet nbutyl alcohol $\eta = 3 \cdot 03$ mPas, \odot n-amyl alcohol $\eta = 4 \cdot 92$ mPas, \odot n-hexyl alcohol $\eta = 5 \cdot 39$ mPas. given conditions, Fig. 3 demonstrates the possibility of independence of K on viscosity at these conditions. At higher liquid flow rates it is possible to find, similarly as in the study⁷, the change in the slope of the straight line $K(\Gamma_v)$ which means its rise with increasing viscosity.

The dependence $K(\Gamma_{v})$ for aqueous solution of glycerol with the viscosity 2.3 mPs s and for n-propyl alcohol with the viscosity 2.2 mPa s is plotted in Fig. 4. Both these liquids have approximately the same viscosities, while the surface tension for the solution of glycerol is $7.15 \cdot 10^{-2}$ kg s⁻² and for n-propylalcohol is $2.38 \cdot 10^{-2}$ kg s⁻². At small liquid flow rates no difference has been observed between both these liquids, at higher flow rates a systematic deviation toward small values of K has been observed for the liquid with lower surface tension. But it is necessary to state that this deviation is within the range of scatter of experimental data. It could not have been the result of different viscosities since to the lower viscosity correspond larger values of K. Such behaviour corresponds qualitatively to the analysis by Levich²⁴ on mass transfer into the film with wavy surface. At laminar film flow the independence of the absorption kinetics of surface tension results from his analysis and at fully developed turbulent flow of the film the absorption rate shall be indirectly proportional to the second root of surface tension. The found decrease is smaller than corresponds to turbulent flow which could be explained by the fact that even at larger liquid flow rates the turbulent flow has not developed in the film. This follows also from values of the Reynolds number, which has always been smaller than 250, which is the value considered to represent transition between the laminar and turbulent flow of the film.

The experimental data for the kinetics of absorption on the string of spheres were evaluated in the form of equation



$$Sh = 3.78 \cdot 10^{-3} \text{ Re}^{0.63} \text{ Sc}^{0.62} \text{ Ga}^{0.47} \text{ We}^{-0.14}$$
, (7)

while the estimate of standard deviation of linearized relations is s = 0.069 and of the exponents $s_1 = 0.016$, $s_2 = 0.085$, $s_3 = 0.24$, $s_4 = 0.16$, while $t_{0.05} = 1.96$. But with about the same accuracy are the experimental data represented by the regression relation not including the Weber number

$$Sh = 3.37 \cdot 10^{-2} Re^{0.63} Sc^{0.55} Ga^{0.26}$$
, (8)

where the corresponding estimates of standard deviations are s = 0.069, $s_1 = 0.016$, $s_2 = 0.018$, $s_3 = 0.016$. It is also possible to observe a significant decrease in the error of the exponent with the Schmidt number which is approaching in this case the expected value 0.5.

Possible application of the experimental results presented here for packings has been tested by comparison of the experimental volumetric mass transfer coefficient for CO_2 into several solvents with the values calculated according to Eq. (8). For this purpose Eq. (8) has been arranged, at the assumption that the effective surface area for absorption can be considered to be identical with the wetted surface area of the packing^{9,29}, into the form

$$k_{e}a = 3.37 \cdot 10^{-2}a_{t} \left(\frac{a_{w}}{a_{t}}\right)^{0.37} \left(\frac{\dot{m}}{Sa_{t}\eta}\right)^{0.63} \frac{g^{0.26}v^{0.48}}{d_{p}^{0.22}(v/D)^{0.45}}$$
(9)

TABLE IV Comparison of Experimental and Calculated (according to Eq. (9)) Values of $k_c a$

Packing	Solvent	$\frac{m}{Sa_1\eta}$	$(k_c a)_{exp}$ $10^{-3} s^{-1}$	$(k_ca)_{calc}$ $10^{-3} s^{-1}$	Author
Raschig	water	2	9.7	11.2	Onda and coworkers ¹
rings	ethanol	2	14.0	11-6	Onda and coworkers ¹
4 mm	decaline	2	9.0	11.8	Onda and coworkers ¹
	methanol	2	15.5	15.0	Onda and coworkers ¹
	benzene	2	15.0	17-1	Onda and coworkers ¹
	tetrachloromethane	2	14.5	17.1	Onda and coworkers ¹
Raschig	methanol	2	2-8	2.9	Yoshida,
rings	methanol	10	10.0	9.3	Koyanagi ²¹
25 mm	methanol	20	16.9	15-2	Yoshida,
Berl	methanol	2	3.0	3-9	Koyanagi ²¹
saddles	methanol	10	11-0	12.5	Yoshida,
25 mm	methanol	20	19.5	20.5	Koyanagi ²¹

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Ratio of the wetted and dry surface area of the packing is calculated from relation¹⁸

$$\frac{a_{\mathbf{w}}}{a_{t}} = 1 - \exp\left[-1.45(ga_{t})^{0.05} \left(\frac{\eta^{3}}{\varrho\sigma^{2}}\right)^{0.1} \left(\frac{\sigma_{c}}{\sigma}\right)^{0.75} \left(\frac{\dot{m}}{Sa_{t}\eta}\right)^{0.4}\right].$$
(10)

The values $(k_ca)_{cale}$ for conditions of the experiments from literature^{1,21} are given in Table IV. The range of liquid flow rates covered by literature was relatively narrow, therefore was the comparison performed for one value of the Reynolds number, in the second paper²¹ were considered three values covering the measured range of flow rates. The experimental data denoted in Table as $(k_ca)_{exp}$ were obtained from tabulated data from both quoted papers. From Table IV is obvious a good agreement of the calculated and experimental data, if the scatter of experimental data in compared systems and the error in the relation for calculation of the wetted surface area of the packing are taken into consideration.

On the contrary, if the relations from literature are used for the estimate of the by us found mass transfer coefficients for absorption of CO_2 into liquids with higher viscosity, the results are obtained which mutually differ by up to one order of magnitude²².

CONCLUSIONS

Studied was the kinetics of absorption into the film of liquids with differing viscosities on the laboratory model of an absorber. The results correspond qualitatively to the analysis by Levich²⁴ of mass transfer into the downward flowing film with wavy surface. The obtained dimensionless relation has supplied a satisfactory estimate of the volumetric mass transfer coefficient of CO_2 absorption in various liquids in the packed bed when the wetted surface area of the packing has been substituted for the effective surface area for absorption. So has been demonstrated applicability of this method⁶ to liquids other than water.

LIST OF SYMBOLS

- a_c specific effective surface area of the packing, L^{-1}
- a_1 specific surface area of dry packing, L^{-1}
- $a_{\rm w}$ specific wetted surface area of the packing, L^{-1}
- c^* equilibrium solubility of the gas in liquid, N L⁻³
- C constant in Eq. (1)
- C' constant in Eq. (2)
- $d_{\rm p}$ nominal diameter of the element of packing, L
- \dot{D}_{T} internal diameter of the column, L
- f proportionality factor in Eq. (3)
- g gravitational acceleration, L T⁻²
- h length of liquid film, L

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D diffusivity, $L^2 T^{-1}$

- k_c mass transfer coefficient for liquid phase at physical absorption, L T⁻¹
- K dimensionless quantity defined by Eq. (5)
- m mass flow rate of liquid, M T⁻¹
- n exponent in Eq. (3)
- s estimate of standard deviation of the regression function
- s_i estimate of standard deviations of exponents
- S cross sectional area, L^2
- us wetted perimeter, L
- $t_{0.05}$ critical value of the Student's distribution
- z height of packing in the column, L
- β_i exponents in the standardized dimensionless Eq. (1)
- β'_1 exponents in the standardized dimensionless Eq. (2)
- η dynamic viscosity of liquid, M L⁻¹ T⁻¹
- v kinematic viscosity of liquid, $L^2 T^{-1}$
- σ surface tension of liquid, M T⁻²
- $\sigma_{\rm c}$ surface energy of material, M T⁻²
- ϱ liquid density, M L⁻³
- Γ linear mass wetting intensity, M L⁻¹ T⁻¹
- $\Gamma_{\rm v}$ linear volumetric wetting intensity, L² T⁻¹

Dimensionless Numbers

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Translated by M. Rylek.