

Available online at www.sciencedirect.com



International Journal of HEAT and MASS TRANSFER

PERGAMON

International Journal of Heat and Mass Transfer 46 (2003) 1715–1724

www.elsevier.com/locate/ijhmt

Mass transfer into liquid falling film in straight and helically coiled tubes

Mohammed Salah Hameed *, Mehdi Saleh Muhammed

P.O. Box 29168, Abu Dhabi, United Arab Emirates Received 1 January 2001; received in revised form 25 November 2002

Abstract

Helically moving liquid films are principally used in many heat and mass transfer processes, which are usually practiced to minimize longitudinal dispersion, maximize secondary currents and provide compact spacing.

In the present work, practical experiments are carried out to determine mass transfer coefficients for thin films of liquid falling in case of straight and coiled tubes. The system used in the present investigation is the absorption of CO_2 into liquid films of distilled water, or ethyl alcohol (96.25%), or ethylene glycol of 12% or 5.2%. The obtained experimental results indicated higher mass transfer coefficient in the helically liquid film.

Empirical correlations are concluded by using dimensional analysis method. The correlations relate Sherwood number to the following dimensionless groups for laminar and turbulent regimes.

 $Sh = f(Re_f, Sc, Ga_f)$ for inclined straight tube $Sh = f(De_f, Sc, Ga_f)$ for helically coiled tube

where De_f , Ga_f and Re_f are Dean, Gallileo and Reynolds numbers for flowing films, respectively. While Sc and Sh are Schmidt and Sherwood numbers.

Crown Copyright © 2003 Published by Elsevier Science Ltd. All rights reserved.

Keywords: Mass transfer coefficient; Liquid falling film; Film Reynolds number; Helically coil tubes; Film Sherwood number

1. Introduction

In industrial situations, it is desirable to have high mass transfer coefficients in contacting stages. Flow regime needs to be investigated, where high mass transfer coefficient can be expected. In the present work an attempt is made to define hydrodynamic conditions in thin liquid films in straight inclined or helically coiled tubes. If the provided conditions are selected carefully, these films could be kept laminar or turbulent within operating variables range. The hydrodynamic understandings can be applied to study the gas absorption rates by using several systems of carbon dioxide as an absorbing gas and distilled water, or ethyl alcohol (96.25%), or ethylene

* Corresponding author.

glycol of (5.2%) or (12%) as liquids. The carbon dioxide is absorbed into a thin film of liquid or ripple free surface running down a straight inclined or helically coiled transparent plastic tube. The latter case simulates the case of mass transfer to liquid films system in contacting stages.

Considerable experimental data is available in literature on liquid phase controlled mass transfer in case of liquid film falling along inclined surfaces. In such cases the liquid shear is usually absence. However, careful experimental studies to determine mass transfer coefficient for thin sheared liquid films appears to be scarcely or non-existent.

The literature on mass transfer into liquid falling film has been concerned mainly with the dependence of the mass transfer coefficients on the molecular diffusivity. There are number of mass transfer models exist in literature to simulate the absorption phenomena of gas in liquid, such as the film model, renewal or penetration

E-mail address: mhameed@emirates.net.ae (M.S. Hameed).

^{0017-9310/03/\$ -} see front matter Crown Copyright © 2003 Published by Elsevier Science Ltd. All rights reserved. doi:10.1016/S0017-9310(02)00500-8

Nomencl	ature
---------	-------

$egin{array}{c} A \\ C \\ C_0 \end{array} \\ C_i \\ C^* \end{array}$	tube radius (cm)Rmass transfer area (cm²)Reconcentration (gmol/cm³)Scinlet concentration of CO2 in liquid (gmol/Shcm³)uoutlet concentration of CO2 in liquid (gmol/zcm³)equilibrium concentration of CO2 in liquid (gmol/Gr (cmol/cm^3) ϕ	R Re _f Sc Sh u z Greek s φ	Rcoil diameter (cm) $Re_{\rm f}$ film Reynolds number $(\Delta\Gamma/v)$ Sc Schmidt number (D/v) Sh Sherwood number $(k_{\rm L}\delta/D)$ u velocity (cm/s) z longitudinal direction (cm)Greek symbols ϕ central angle (rad)			
$egin{array}{c} D \ De_{ m f} \ g \ Ga_{ m f} \ k_{ m L} \ L \ N_{A} \end{array}$	diffusion coefficient (cm ² /s) film flow Dean number ($Re_f(a/R)^{1/2}$) gravitation constant (cm/s ²) film flow Gallileo number ($\delta^3 g \sin \psi/v^2$) film mass transfer coefficient (cm/s) width of liquid film (cm) mass flux (gmol/cm ² s)	δ Δ υ σ ψ	(cm ² /s) film thickness (cm) dimensionless concentration kinematic viscosity (cm ² /s) surface tension (dyne/cm) angle of inclination (degree)			

model, film penetration model, and eddy diffusivity oriented model. But there are limited attempt was made towards relating the absorption rates to hydrodynamic conditions. To have a good understanding of the mass transfer process in sheared film, it is necessary to develop correlation that relates the mass transfer process to fluid velocity field.

The object of the present work is to measure the absorption rates of gas into liquid film in systems of falling films down inclined straight and helically coiled tubes and then relate the measured results to the film hydrodynamic behavior.

2. Experimental measurements

Carbon dioxide gas is used with either distilled water, 96.25% ethyl alcohol, 5.2 or 12% ethylene glycol as liquids to study the mass transfer rate for both systems of straight and helically coiled tubes. The CO₂ gas used in this investigation is of high purity grade so that water vapor presents in gas does not effect the rate of mass transfer [1]. The liquids selected with scarcely CO₂ absorption so that diffusion and mass transfer of absorbed gas would not affect liquid film velocity. Moreover, the liquids used have only purely physical absorption of CO₂ with no chemical reaction. The liquids were chosen with broad Schmidt number ranges from 441 to 1090.

The gas was flowing counter currently with liquid at Reynolds number below 300 to provide free falling of liquid and give constant film thickness [2].

The used apparatus is shown schematically in Fig. 1. Liquid, from constant head tank (A), was fed to 140 cm long flexible transparent tube placed in section (C). The liquid flow rates to the tube were adjusted by means of needle valve and calibrated rotameter. The gas was supplied from gas cylinder (D), to the test section, at pre-set rate measured by adjusting the rotameter (B). A counter currently gas flow with liquid is used in testing section (C). The gas is maintained at constant temperature, 20 °C, by passing it through a glass coil immersed in constant temperature water bath before entering the test section. Two sampling points were positioned at the inlet and outlet of test section to measure the CO₂ concentration in the entering and leaving liquid to the test section. The outlet liquid was collected in tank (F) before degassing in desorption unit (G). The latter unit was constructed from glass column 80 cm height and 10 cm inside diameter with upper brass cover fixed with two brass outlet tubes of 1 cm inside diameters each. One of the tubes is used for liquid feedings, while the second tube is leading to a vacuum system through a liquid trap (H). The lower end of the column was connected through a valve to the main feeding tank that supplies the system with liquid through the centrifugal pump (I).

Liquid samples were collected to analyze CO_2 concentration by using back titration method for NaOH by means of hydrochloric acid and phenolphthalein indicator.

It is know from literature over a wide range of $Re_{\rm f}$, that liquid falling films are practically smooth for angles of inclination less than 5°. Limited ripples formed at higher range of $Re_{\rm f}$. For this purpose, the angle of inclinations are used between 2.5–5°. In straight inclined tube experiments, the flexible tube is mounted on a flat steel plate. The far end of the tube away from the entrance is supported on a horizontal stand. It can be moved vertically up or down to a suitable position that can be conveniently measured by a traveling microscope in order to set the required inclined angle at (2.5°, 3°, 5°).



Constant head tank	Α
Rotameter	В
Angle adjustment board	С
CO ₂ Supply bottle	D
Water bath	E
Collecting tank	F
CO2 Gas desorption unit	G
Liquid trap	Н
Recycling pump	I
Sample point	ΙΎ
On-Off valve	R
Three way valve	£
Needle valve	Ø

Fig. 1. Schematic diagram of absorption apparatus.

The length of the developing region was taken to be about 20-film thickness [3].

The film thickness is measured at 50 cm distance away from the entrance of the tube to insure that all measurements are made in the fully developed region.

A special micrometer used in measuring the film thickness through holes made in the upper part of the flexible tube. It consists of two micrometers arranged so that the vertical and horizontal distances of the cross sectional area of the flow can be measured. The whole measurement arrangement supported on a vertical stand, which allows the two micrometers to move freely to the required position near the system.

The helical coil was formed by wrapping a flexible tube around different PVC tubes in accordance with the required curvatures. Three PVC tubes of different diameters are selected. The flexible tube is wrapped around the PVC tubes at adjusted angles of inclinations, the same as that used for straight tube. Since curvature has a calming action, the developing region in curved tubes is expected to be shorter than that of straight tubes.

3. Mass transfer coefficient

Lamouralle and Sandal [4] found that the mass transfer coefficients predicted from penetration (Higbie model), renewal surface (Dankwert model), and film model theories do not agree with the measured falling film mass transfer coefficients. As each of these models depend on one variable only; it is the contact time for the first two models and the constant film thickness for film model. Each of these theories has a preferred range of application. The film theory is used for steady flow situation in which a binary fluid mixture flows horizontally along a plane surface where mass transfer occur. The boundary layer theory preferred for laminar flow along stationary plane boundaries and in pipe entrances. Whereas the penetration theory is usually preferred for liquids at gas–liquid interfaces. In other situations the choice must be based on judgment or when possible, experimental evidence.

These models found that increasing Reynolds number has only small effect on changing the mass transfer coefficient. The actual experimentally data for mass transfer coefficient did not agree with calculated ones when using the stated models as shown in Fig. 2. The figure was plotted by feeding the actually measured experimental data into these models and calculate the mass transfer coefficient form the relevant model and then plot the calculated results in Fig. 2.

To overcome the inconsistency in predicting the values of $k_{\rm L}$ as function of $Re_{\rm f}$ by using various models and system configurations, a mathematical correlation is



Fig. 2. Comparison of different methods for mass transfer coefficient calculation.

derived below based on mass and momentum transfer phenomenon and it can be employed for all kinds of surface configurations.

By taking a mass balance on an elementary shell of the falling film showing in Fig. 3, the following equation can be obtained:

$$N_A \,\mathrm{d}A = \mathrm{d}C(\Gamma)(\phi a) \tag{1}$$

Since

1718

$$N_A = k_{\rm L} (C^* - C) \tag{2}$$

By substituting, obtain N_A from Eq. (2) into Eq. (1) and using A = L dz, the following equation is concluded:

$$k_{\rm L}(C^* - C)(L\,\mathrm{d}z) = \mathrm{d}C(\Gamma)(\phi a) \tag{3}$$

Eq. (3) can be integrated between the limits:

$$z = 0 \quad C = C_i$$

$$z = z \quad C = C_0$$

$$k_L = \left(\frac{\phi a \Gamma}{A}\right) In(1/\Delta)$$
(4)

where \varDelta represents the dimensionless concentration.



Fig. 3. Elementary shell of falling film inside tube.

In an inclined tube, the length of falling film can be measured easily, and consequently the mass transfer area can be calculated. While in case of helical tube, the film length is affected by the influence of centrifugal force on the liquid falling film and hence an average value is usually used in calculating *A*. The surface waves usually introduce about 3% error in the calculation of mass transfer area (as cited in [5]), that can be neglected in engineering calculations.

4. Results and discussion

Several experiments were performed and in each of them the quantity of absorbed carbon dioxide gas in the falling films was calculated. Different film Reynolds numbers, ranging from 200 to 3500, were used at low gas flow rates. The absorption was taken place within the 140 cm long inclined straight or helically coiled tubes. The obtained experimental results for both configurations are discussed separately.

4.1. The inclined straight tubes

The variation in mass transfer coefficient's with liquid film Reynolds number is shown in Fig. 4. Several liquids were used that differ in their physical properties as listed in Table 1. In general, the increase rate of mass transfer coefficient k_L seems to be low with film Reynolds number (Re_f) in laminar flow region while it increases at higher rate in turbulent region. The higher rate of increase in k_L in the turbulent region is related to the higher turbulence and mixing that would be created



Fig. 4. Mass transfer coefficient versus film Reynolds number.

Table 1Physical properties of liquids used

Liquid	<i>T</i> (°C)	υ	<i>T</i> (°C)	$D imes 10^5$	<i>T</i> (°C)	$C^* imes 10^5$	Sc	
Ethyl alcohol	18	0.015	20	3.45	25	12.50	441	
Distilled water	25	0.008	20	1.77	20	3.29	452	
5.2% Ethylene glycol	20	0.0112	25	1.58	25	3.02	709	
12% Ethylene glycol	20	0.0133	25	1.22	25	2.79	1090	

within the film layer. It is clear from Fig. 4 that the molecular diffusion (causes slow transport) plays a main role in the mass transfer occurs within the laminar region, while convective transfer is appeared to be more prominent in the turbulent region.

Present experimental results also indicated that the tube inclination angles ranging from (3° to 5°) has no real effect on $k_{\rm L}$ for all the tested liquids used.

The experimental results have been developed further to show, as in Fig. 5, the variation of Sherwood number $(k_L \delta/D)$ with (Re_f) at different inclined angles. The increase angle of inclination seems to decrease the Sherwood number value at certain film Reynolds numbers (Re_f) . This trend is mainly due to hydrodynamic effect rather than mass transfer effect, as the (Re_f) and diffusion coefficient are both stay constant with change of



Fig. 5. Variation of Sherwood number with falling film Reynolds number.

angle of inclination while the film thickness changes with angle of inclination.

The critical film Reynolds number (Re_f) that is related to the flow change from laminar to turbulent can be predicted from mass transfer studies. It seems to have an approximately constant value equal to 1000 for all liquids tested. This is an important result that concludes the momentum transfer behavior from mass transfer studies.

Empirical correlations, to predict mass transfer coefficient, is derived by using dimensional analysis technique. The dimensionless groups affecting mass transfer into the falling film has been recognised by assuming that the mass transfer coefficient is a function of the following parameters:

$$k_{\rm L} = f(v, \sigma, \delta, u, D, g) \tag{5}$$

Now, by using the dimensional analysis technique, the Sherwood number was found to be a function of Re, Sc and Ga of the liquid film as given in the following equation:

$$Sh = f(Re_{\rm f}, Sc, Ga_{\rm f}) \tag{6}$$

By using the multi-variable least squares methods, the constant and powers of dimensionless groups are found as follows:

For laminar region

$$Sh = 4.64 \times 10^{-3} (Re_{\rm f})^{0.35} (Sc)^{0.61} (Ga_{\rm f})^{0.14}$$
(7)

For turbulent region

$$Sh = 2.136 \times 10^{-4} (Re_{\rm f})^{0.4} (Sc)^{0.65} (Ga_{\rm f})^{0.52}$$
(8)

The two above correlations show that Sherwood number is more dependent on film Reynolds and Gallileo numbers in turbulent region than in case of laminar regions; which indicates that mass transfer mechanism in turbulent region is more dependent on convection phenomena. While Schmidt number is almost constant for both regions within the investigated range of (Re_f). This conclusion is in full agreement with Koziol et al. [6] findings.

4.2. Helically coiled tubes

In general, experimental results [7] indicated higher absorption rate of CO_2 in liquids for the case of helically coiled tubes incomparison with the case of straight tubes. This fact is expected to occur as a result of the difference in the hydrodynamic behavior between the two cases.

The obtained experimental results, as shown in Fig. 6, indicated that the change of helical tube pitch of (3-5)

degrees has no real effect on mass transfer coefficient for all the liquids used. On the other hand, the tube curvature (a/R) has a noticeable effect on mass transfer coefficient for the liquids investigated and as illustrated in Fig. 7 for the case of distilled water.

Mass transfer coefficient in helical tube seems to decrease with decreasing tube curvature (increasing helical diameter) and eventually approaches that obtained for straight tube or minimum curvature, particularly in laminar region. Fig. 7 shows that (k_L) values for straight tube coincide with that of 41.31 cm helical tube diameter within laminar region while the two cases divert in values in turbulent region. The higher $k_{\rm L}$ for helical tube may be due to increase secondary flow in case of low curvature at high flow rate. This phenomenon again support the existing similarity between mass transfer and hydrodynamic studies, as the critical Reynolds number found from mass transfer studies agrees with the value obtained in hydrodynamic studies. The critical Reynolds number was found to be approximately equal to 1000 in both cases.

Fig. 8 shows the mass transfer coefficient variation with different liquid films used at constant pitch and curvature. As in the case of straight tube, k_L increases as the value of saturation concentration of CO₂ is increased at the liquid interface. It can be seen clearly in Fig. 8 how k_L is related to C^* in Table 1. Ethyl alcohol has the highest saturation concentration and subsequently expected to have the higher k_L . Hence k_L is expected to decrease as a result of a decrease in CO₂ concentration at saturation.



Fig. 6. Mass transfer coefficient versus Reynolds number for different tube pitch at constant inclination for CO_2 absorption in 5.2% ethylene glycol.



Fig. 7. Mass transfer coefficient versus Reynolds number for absorption of CO₂ in distilled water at different configuration.



Fig. 8. Mass transfer coefficient versus Reynolds number for absorption of CO_2 in different liquids used for helical tube of constant pitch.

By using the dimensional analysis technique, empirical equations may be concluded to calculate mass transfer coefficient. Assume that $k_{\rm L}$ is function of the parameters given in Eq. (9).

$$k_{\rm L} = f(v, \sigma, \delta, R, D, u, g). \tag{9}$$

By substituting (a/R) instead of (δ/R) so that to include Dean's number in the correlation, the following relation is obtained:

$$Sh = f(De_{\rm f}, Sc, Ga_{\rm f}) \tag{10}$$

The exact equations for laminar and turbulent regions are concluded by using the multivariable least squares method to evaluate the constant and the power of the dimensionless groups and conclude the following equations.

For laminar flow region

$$Sh = 1.4 \times 10^{-3} (De_{\rm f})^{0.13} (Sc)^{0.73} (Ga_{\rm f})^{0.5}$$
(11)



Fig. 9. Comparison of experimental and calculated, from Eq. (11), Sherwood number, for laminar film flow in helical tube.

For turbulent flow region

$$Sh = 1.0 \times 10^{-3} (De_{\rm f})^{0.5} (Sc)^{0.54} (Ga_{\rm f})^{0.45}$$
(12)

where $De_{\rm f}$ is the Dean number of the flowing liquid film and it is equal to $Re_{\rm f}(a/R)^{1/2}$.

The validity of using Eqs. (11) and (12) to predict the experimental mass transfer coefficient is shown in Figs. 9 and 10, respectively.



Fig. 10. Comparison of experimental and calculated, from Eq. (12), Sherwood number for turbulent film flow in helical tube.

By comparing the obtained correlations for the two liquid film systems flowing in straight and helical tubes, it can be seen from Eqs. (7) and (11) in laminar region that the powers of film Reynolds number (incorporated within Dean number in case of helical tube) is lower for helical tube. While Schmidt number is higher in case of helical tube as compared to the case of straight tube. This result confirms that the molecular diffusion is being



Fig. 11. The increase in mass transfer coefficient due to increase coil curvature at 3° inclination.

more effective in helical tube and the contact time between the gas and the liquid is more influential. While by comparing the two systems of straight and helical tubes for turbulent regions from Eqs. (8) and (12), it can be seen that the values of the powers of the dimensionless groups, in general, are closer to each other than in case of laminar flow. The higher power of film Reynolds number for helical tube is due to the predominant effect of secondary flow, which is the controlling factor in this region.

Fig. 11 concluded that higher mass transfer coefficient up to 30–40% would be obtained in helical tube of higher curvature in case of distilled water used as a liquid medium for example.

5. Conclusion

The following conclusions can be drawn from the present work.

 Variation of mass transfer coefficient, k_L exists with liquid film Reynolds number (*Re*_f).

- 2. The critical $Re_{\rm f}$ at the point of transfer from laminar to turbulent regions can be accurately predicted from mass transfer phenomena.
- 3. Higher mass transfer coefficient is obtained with liquid film in helical tubes than in straight falling tubes.
- 4. Mass transfer coefficient increases with increasing helical tubes curvature.
- 5. Higher mass transfer coefficient is obtained with higher saturation concentration of CO₂ in liquid.

References

- [1] L.E. Scriven, R.L. Pigford, AIChE, J 4 (1958) 439-444.
- [2] D.R. Oliver, T.E. Atherinos, Chem. Eng. Sci. 23 (1968) 525– 536.
- [3] B. Atkinson, R.L. McKee, Chem. Eng. Sci. 19 (1964).
- [4] A.P. Lamourelle, O. Sandal, Chem. Eng. Sci. 27 (1972) 1035–1043.
- [5] E. Blass, Int. Chem. Eng. 19 (2) (1979) 183-195.
- [6] K. Koziol, L. Broniarz, T. Nowika, Int. Chem. Eng. 20 (1980) 136–142.
- [7] M.S. Muhammed, Gas diffusion study into several liquids film layers falling inside helical tube, M.Sc. Thesis, submitted to the University of Technology, Baghdad, 1988.