FREE CONVECTION MASS TRANSFER: LAMINAR AND TURBULENT

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(Received 11 September 1974 and in revised form 26 February 1975)

Abstract—Natural convection mass transfer from vertical plates has been studied using a novel technique for a Rayleigh number range of $4 \times 10^9 - 2 \times 10^{13}$. The Rayleigh number marking the onset of laminar-turbulent transition was found to be 8×10^{10} . The regression equation for the overall Sherwood number for $2 \times 10^{11} < Ra < 2 \times 10^{13}$ was

$\overline{Sh} = 0.258 \, Ra^{0.289}$

The measured overall mass-transfer coefficients in the turbulent flow agreed fairly well with the limited experimental data available in the literature.

NOMENCLATURE

- D, diffusion coefficient $[cm^2/s]$;
- g, acceleration of gravity [cm/s²];
- k, local mass-transfer coefficient [cm/s];
- k, overall mass-transfer coefficient [cm/s];
- L, length of plate from the leading edge [cm];
- Ra, Rayleigh number;
- Sh, local Sherwood number;
- \overline{Sh} , overall Sherwood number;
- μ , viscosity [g/cm s];
- ρ , fluid density [g/cm³];
- ρ_{w} , fluid density at the polymer surface-liquid interface:
- ρ_{∞} , fluid density in the fluid bulk.

INTRODUCTION

FLUID flow in natural convection is due to a density differential within the fluid. In natural convection mass transfer, such a density differential is due to the material transferring from a surface, which has a different density than the working fluid. When the surface of the transferring material is in the form of a vertical plate, the flow within the boundary layer is laminar for short distances from the plate leading edge. At some distance down from the leading edge (the transferring material is assumed to be heavier than the working fluid), the flow within the boundary layer becomes turbulent. Usually, along a vertical plate, a transition region exists that separates the laminar and the turbulent regimes.

Mass transfer in natural convection from vertical surfaces is of considerable importance in the solution mining industries where leaching of salts occurs under natural convection mass transfer. Schmidt numbers for such salt-water systems are of the order of 1500 and consequently heat transfer in natural convection, where the Prandtl number is usually low, is not of direct application in mass-transfer operations.

Although heat and mass transfer in natural convection for vertical plates under laminar conditions and that for the heat transfer under turbulent conditions, have been extensively studied, work on turbulent natural convection mass transfer is scarce.

The only study on mass transfer in turbulent natural convection for vertical plates that deals with a Rayleigh number up to 10^{15} and a Schmidt number of about 2000 is that of Fouad and Ibl [1]. They measured overall mass-transfer coefficients and consequently they evaluated the corresponding overall Sherwood numbers. Such mass-transfer coefficients include contributions from the laminar, the transition and the turbulent flow regimes over the vertical plate. No measurements in the laminar regime alone were made by Fouad and Ibl.

Lloyd et al. [2] presented the most reliable experimental data to date for the local mass-transfer coefficients for vertical plates under laminar natural convection. Unfortunately their data in the turbulent regime covered a very small range in Rayleigh number and consequently no correlation can be deduced for the local Sherwood number in the turbulent regime.

Husband [3] measured the overall mass-transfer coefficient for vertical plates of up to 6 m long using dissolution of inverted sugar in water for a Rayleigh number range $4 \times 10^5 - 9 \times 10^{14}$. In the laminar regime his data gave a much higher value for the overall Sherwood number for a given Rayleigh number than those of the accepted correlation of Wilke *et al.* [4]. However, Husband's system had a rather high Schmidt number, namely Sc = 14400 and this high value of Sc could account for the high values of the overall mass-transfer coefficients.

Two methods are generally used in natural convection mass-transfer studies, namely, the electrochemical and the dissolution techniques. The electrochemical technique is the most widely used method in masstransfer measurements. This method can be employed most readily in measuring local transfer rates; Wilke *et al.* [4] and Lloyd *et al.* [2]. In the dissolution technique, the rate of the dissolution of a solid, e.g. benzoic and salicylic acids and sugars, is directly used in the measurements of overall mass transfer coefficients; Wilke *et al.* [4] and Husband [3].

In this paper overall mass-transfer coefficients, in terms of Sherwood numbers, will be evaluated in laminar and turbulent natural convection for a vertical plate using a new technique not hereto employed in natural convection studies.

EXPERIMENTAL PROCEDURE

The new technique employed in this study of natural convection mass transfer is that of Macleod and Todd [5]. The essence of their technique is to coat the solid surface of interest with a polymer which is inert to the working fluid. This polymer coating is then swollen in a reversible manner by a swelling agent. Transfer of the swelling agent from the surface of the polymer coating to the working fluid stream can then be recorded as a weight loss and consequently masstransfer rates can be measured. Macleod and Todd employed this technique in mass transfer studies involving forced convection. In order for this technique to be useful in mass transfer studies, it is essential that a "constant rate period" is maintained while measurements of the mass transfer are taken. Such a constant rate period would then indicate that the resistance to the mass transfer is within the working fluid and not in the polymer coating.

In natural convection studies, the swelling agent should satisfy the following criteria: (i) a large density differential; (ii) low vapour pressure; (iii) moderately swells the polymer coating; (iv) slightly soluble in the working fluid; (v) readily available and non reactive with either the polymer coating or the working fluid. Each of these criteria will be discussed:

As natural convection is caused by a density differential within the working fluid, a large density difference between the swelling agent and the working fluid would give a relatively large value for the Rayleigh number for short lengths of plates. Consequently relatively short plates will suffice in studying the turbulent natural convection mass transfer. As for the second criterion, a low vapour pressure of order less than 0.5 mm Hg is very desirable since losses of the swelling agent to air during plate handling should be kept at a minimum. A low vapour pressure would give a low swelling agent concentration at the surface of the polymer coating and consequently a small driving force for mass transfer in air.

Condition (iii) is necessary as a very high degree of swelling distorts the polymer coating and consequently the polymer film would not adhere to the plate (substrate) due to the movement of the coating during swelling. A swelling of up to 50 per cent (weight of swelling agent to weight of dry polymer film) is suitable. Condition (iv) is required so that a constant rate period could be maintained for a reasonable length of time during which mass transfer rate measurements are taken. A high solubility of a swelling agent in the working fluid could lead to a fast depletion of the polymer coating from its swelling agent, leading to a short constant rate period. The necessity of requirement (v) is obvious.

Glass plates of various lengths and 40 mm wide were coated on one side only with silicone rubber* and this polymer coating was cured according to the manufacturer's instructions. The silicone rubber coating thickness was about 1 mm. 2-Bromopyridine was found to be the best choice as a swelling agent. The working fluid was water.

Determination of the solubility of 2-bromopyridinc in water and the density of the saturated water solution were made in our laboratories. The relevant data are given in Table 1.

Table 1. Physical properties of the system at 20°C

Density of 2-bromopyridine, ρ	$1.657 \mathrm{g/cm^3}$
Density of pure water, ρ_{∞}	$0.9982 \mathrm{g/cm^3}$
Density of saturated 2-bromo-	
pyridine solution (in water), ρ_w	$1.0049 g/cm^3$
Solubility of 2-bromopyridine in water	$0.0164 \frac{g}{g \text{ of solution}}$
Vapour pressure of 2-bromo- pyridine	0.5 mm Hg (at 20°C) (estimated)
Diffusion coefficient of 2-bromo- pyridine in water, D	$4 \times 10^{-6} \mathrm{cm}^2/\mathrm{s}$

The data in Table 1 indicates that 2-bromopyridine gives a relatively large value for the density driving force although its saturation concentration in water is small. It is then clear that by a proper choice of a swelling agent, this technique becomes very useful in that the fluid properties are constant across the diffusion boundary layer and no uncertainties exist in the evaluation of the fluid properties, in contrast to the other methods employed in natural convection studies.

Pre-swollen silicone rubber films were swelled with 2-bromopyridine by immersing the glass plates on which the silicone polymer is coated, in the swelling agent bath for a period of 24h to insure that the equilibrium state between the polymer film and the swelling agent was reached. This equilibrium state is considered to have been reached when no further increase in the weight of the polymer coating occurs with immersion time.

Once the equilibrium swelling state is reached, the plate can then be used for mass-transfer studies. The plate was then taken from the swelling agent bath and dried with a lint free tissue paper. The weight of the plate was then recorded and it was suspended vertically in a large constant temperature water bath (300 mm dia \times 900 mm height). The height of the water above the upper edge of the plate was about 100 mm. The plate was then reimmersed in the swelling agent bath for a period of at least 3 h before it was re-used.

Initial tests were made to ensure that the drying technique is adequate. It was found that by wetting with water a non-swollen plate and drying it with the lint free tissue paper, the plate weight varied by 0.0002 g when the procedure was repeated several times.

^{*}RTV-615 silicone rubber manufactured by General Electric, Waterford, New York 12188.

DISCUSSION OF RESULTS

The Sherwood number is used as a dimensionless representation of the mass-transfer coefficient. The local Sherwood number is given by

$$Sh = \frac{kL}{D} \tag{1}$$

where k is based on the concentration difference of the swelling agent at the coating surface and in the fluid bulk. Here, the concentration is given as the mass of the swelling agent in water per unit volume of solution.

The average or the overall Sherwood number is defined as

ſL

$$\overline{Sh} = \frac{kL}{D} \tag{2}$$

where

$$\frac{\int_{0}^{k} k \,\mathrm{d}L}{L} = k. \tag{3}$$

Previous studies indicated that mass transfer in natural convection can be related to the product of the Grashof and the Schmidt numbers, namely, the Rayleigh number. The Rayleigh number is given by

$$Ra = \frac{\rho \boldsymbol{g}(\rho_{w} - \rho_{\infty})L^{3}}{\mu^{2}} \cdot \left(\frac{\mu}{\rho D}\right). \tag{4}$$

It is customary to evaluate ρ , μ and D at the average concentration of the diffusion boundary layer. The value of the solution density at the polymer surface-liquid interface, ρ_w , was taken at saturation.

The weight loss due to the transfer of the swelling agent from the polymer film (silicone rubber coating) to the water for the various plate lengths with time is shown in Fig. 1. For all the different plates the variation



FIG. 1. Weight loss of polymer coating with time.

of the weight loss with time is linear. Such a constant rate period can only be maintained so long as the swelling agent concentration within the polymer at the surface is not greatly different from the original bulk concentration. Alternatively, such a constant rate period occurs so long as the polymer offers little resistance to mass transfer where by the surface concentration is always maintained at the original bulk concentration of the swelling agent in the polymer. This constant rate period can be interpreted as having the controlling resistance to mass transfer in the water stream, not in the coating, Macleod and Todd.

It is interesting to note that this constant rate period is maintained while a considerable percentage of the swelling agent has left the polymer film. For example, for the 160 mm plate, the dry weight of the silicone rubber coating was 4·1817g and the amount of the swelling agent present before immersion in the water tank was 0·4792g. After an eight minute immersion in water, 0·112g of swelling agent diffused out from the coating. This amount constitutes 23·3 per cent of the original swelling agent present in the coating.

The straight lines of Fig. 1 do not pass through the origin as would be expected from a constant rate period criterion. This could be due to losses during handling of the plates and to the initial unsteady state natural and forced convection mass transfer. Visual observations indicated that the water disturbances due to the plate immersion die out after about 1 min.

The diffusion coefficient of 2-bromopyridine is not available in the literature. It was decided that as the laminar natural convection is well correlated both experimentally and theoretically, our experimental data in this regime would be used to evaluate the diffusion coefficient of the swelling agent. The data in the laminar regime are then made to coincide with Wilke's equation

$$\overline{Sh} = 0.666 Ra^{\frac{1}{4}} \tag{5}$$

by choosing an appropriate value for D. This value of D gave the best fit with the experimental data and it is given in Table 1. It should be emphasized that this procedure is effectively a method of finding a value for D so that the constant in equation (5) is satisfied. This procedure does not change the position of any data points relative to each other.

Figure 2 shows that the present experimental data is excellent in the laminar regime in so far as giving a slope of $\frac{1}{4}$ as indicated by equation (5).

A change in the slope of the linear relationship of log (Sh) against log (Ra) appears to occur at a Rayleigh number of about 8×10^{10} indicating the onset of instability. Lloyd *et al.* gave the onset of the transition at a Rayleigh number of 2×10^{11} using local measurements of Sherwood numbers. It is interesting to note that the onset of instability for the thermal natural convection is usually taken at 8×10^8 , where in such studies the Prandtl number is low; Lloyd and Sparrow [6]. Experimental work of Husband [3] for a Schmidt number of about 14400 indicated that the onset of instability is at a Rayleigh number of 6×10^{13} . As suggested by Fouad and Ibl, it scems quite possible



FIG. 2. Variation of overall Sherwood number with Rayleigh number.

that the onset of the instability might not be a unique function of Rayleigh number but a function of both Schmidt and Rayleigh numbers.

Although the experimental data of Fouad and Ibl for the overall Sherwood number exhibit a large degree of scatter, they do indicate a general agreement with the present work as shown in Fig. 2.

The regression equation for the overall Sherwood number in a Rayleigh number range $2 \times 10^{11} - 2 \times 10^{13}$ is given as

$$\overline{Sh} = 0.258 \, Ra^{0.289}. \tag{6}$$

The above correlation for the overall Sherwood number includes contributions from the laminar, transition and turbulent regimes. The maximum deviation of any data points is 6 per cent from equation (6). However, by excluding one data point, the maximum deviation becomes only 3 per cent. The above regression equation for the overall Sherwood number is the best available correlation to date for natural convection mass transfer for a Rayleigh number up to 2×10^{13} .

CONCLUSION

The method suggested here, whereby a swollen polymer coating is used in natural convection mass transfer, offers a new scope in natural convection studies. Different swelling agents and working fluids can conveniently be used to arrive at different values of Schmidt numbers. Also, this method provides a relatively large density differential with little change in the physical properties of the system, leading to a correct estimate of the Schmidt and Rayleigh numbers.

A new correlation for the overall Sherwood number for a Rayleigh number up to 2×10^{13} is given and it represents the experimental data to within 6 per cent.

Acknowledgements—The authors wish to acknowledge the financial assistance from the Saskatchewan Research Council and from the National Research Council of Canada.

REFERENCES

- 1. M. G. Fouad and N. Ibl, Natural convection mass transfer at vertical electrodes under turbulent flow conditions, *Electrochim. Acta* 3, 233 (1960).
- J. R. Lloyd, E. M. Sparrow and E. R. G. Eckert, Laminar transition and turbulent natural convection adjacent to inclined and vertical surfaces, *Int. J. Heat Mass Transfer* 15, 457 (1972).
- W. H. W. Husband, Mass transfer from high vertical surfaces to liquids under free convection conditions, Ph.D. Thesis, Department of Chemistry and Chemical Engineering, University of Saskatchewan, Saskatoon, Sask., Canada (1969).
- C. R. Wilke, C. W. Tobias and M. Eisenberg, Free convection mass transfer at vertical surfaces, *Chem. Engng Prog.* 49, 663 (1953).
- N. Macleod and R. B. Todd, The experimental determination of wall-fluid mass transfer coefficients using plasticized polymer surface coatings, *Int. J. Heat Mass Transfer* 16, 485 (1973).
- 6. J. R. Lloyd and E. M. Sparrow, On the stability of natural convection flow on inclined plates, J. Fluid Mech. 42, 465 (1970).

TRANSFERT DE MASSE EN CONVECTION NATURELLE LAMINAIRE ET TURBULENTE

Résumé—Le transfert de masse en convection naturelle sur des plaques verticales a été étudié à l'aide d'une nouvelle technique dans un domaine de nombres de Rayleigh allant de $4\cdot10^9$ à $2\cdot10^{13}$. Le nombre de Rayleigh marquant le déclenchement de la transition laminaire-turbulent a été trouvé égal à $8\cdot10^{10}$. L'équation de corrélation pour le nombre de Sherwood global dans le domaine $2\cdot10^{11} < Ra < 2\cdot10^{13}$ est la suivante:

$$Sh = 0.258 Ra^{0.289}$$
.

Les coefficients globaux de transfert massique mesurés dans l'écoulement turbulent sont en bon accord avec les données expérimentales disponibles.

STOFFÜBERTRAGUNG BEI FREIER KONVEKTION: LAMINAR UND TURBULENT

Zusammenfassung – Mit Hilfe eines neuen Verfahrens wurde die Stoffübertragung durch freie Konvektion an vertikalen Platten für Rayleigh-Zahlen von 4.10⁹ bis 2.10¹³ untersucht. Dabei wurde festgestellt, daß der Übergang laminar-turbulent durch die Rayleigh-Zahl $Ra = 8.10^{10}$ gekennzeichnet ist. Die Regressionsgleichung für sämtliche Sherwood-Zahlen für 2.10¹¹ < $Ra < 2.10^{13}$ ist $\overline{Sh} = 0.258$. $Ra^{0.289}$. Alle gemessenen Stoffübertragungskoeffizienten für die turbulente Strömung stimmten ziemlich gut mit den wenigen vorhandenen Versuchswerten aus der Literatur überein.

СВОБОДНОКОНВЕКТИВНЫЙ МАССООБМЕН: ЛАМИНАРНЫЙ И ТУРБУЛЕНТНЫЙ

Аннотация — С помощью новой методики исследовался свободноконвективный перенос массы от вертикальных пластин в диапазоне значений числа Релея от $4 \times 10^{\circ}$ до 2×10^{13} . Найдено, что значение числа Релея, при котором наступает передох от ламинарного течения к турбулентному, равно 8×10^{10} . При $2 \times 10^{11} < Ra < 2 \times 10^{13}$ уравнение регрессии для суммарного числа Шервуда составляет $Sh = 0,258 Ra^{0,289}$. Измеренные суммарные коэффициенты массообмена в турбулентном потоке довольно хорошо согласуются с имеющимся в литературе ограниченным числом экспериментальных данных.