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Free convective mass transfer at up-pointing pyramidal electrodes

J. KRYSA
Department of Inorganic Technology, Institute of Chemical Technology, Technicka 5, 166 28 Prague 6, The Czech Republic

and

A. A. WRAGG†
School of Engineering, University of Exeter, North Park Road, Exeter EX4 4QF, U.K.

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Abstract—A limiting current technique was used for the measurement of the free convective mass transfer rate at entire upward pointing pyramids. The mass transfer rate at a single inclined pyramidal surface was correlated by the equation:

$$Sh_L = 0.828(Ra_{L,\theta})^{0.25}$$

for the $Ra_{L,\theta}$ range 1×10^7 to 2×10^{11}. It was found that the mass transfer rate for the combination of all inclined faces was lower than that for the single inclined surface. The down-pointing horizontal base surface has a strong influence on the total mass transfer rate for entire upward pointing pyramids. The experimental total mass transfer data were correlated using a method which includes an interference factor taking account of the fact that the three inclined triangular faces are exposed to fluid which has already been depleted in copper ions as it flows upward from the base. The dependence of the interference factor on the aspect ratio of the pyramid (L/b) was not so strong as for vertical cylinders or down-pointing pyramids.
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1. INTRODUCTION

This paper describes an experimental investigation of mass transfer in free convection at up-pointing pyramidal electrodes using the well-known limiting diffusion current technique (LDCT) of mass transfer measurement. The up-pointing pyramidal electrode is a case of a three dimensional object with one down facing horizontal triangular face and three up-facing inclined triangular faces. The objectives of the present study were:

(a) to measure the mass transfer rates at the triangular inclined surfaces and the horizontal base of an up-pointing pyramid;

(b) to measure the mass transfer rate at entire up-pointing pyramids of varying aspect ratio;

(c) to correlate data for such a geometry taking account of interaction between the individual surfaces.

Such mass transfer data for electrochemical systems in natural convection are pertinent to the kinetics and mechanisms of processes such as electrodeposition, electroforming and electropolishing. Mass transfer measurements are also known to offer extremely good simulation of heat transfer by free convection [1] so that the work is also relevant to estimation of heat dissipation from, for example, electronic devices.

2. PREVIOUS INVESTIGATION

Lloyd *et al.* [2] reported an extensive study for up facing rectangular surfaces inclined at between 0 and +45° from the vertical. In the laminar regime the mass transfer data agreed well with the equation

$$Sh_L = 0.499(Ra_L \cos\theta)^{0.25}. \qquad (1)$$

Results for turbulent flow were correlated with a one third exponent on the Rayleigh number but the coefficient exhibited a marked dependence on the inclination angle.

Patrick *et al.* [3] studied free convective mass transfer to inclined up facing rectangular surfaces. Schlieren photography indicated a number of different possible flow structures. Attached flow conditions were obtained for up facing inclinations where θ was small. Separation of the flow occurred at higher values of θ (>60°). The results for attached boundary layer conditions were well correlated by the equation

† Author to whom correspondence should be addressed.

NOMENCLATURE

A_t	total surface area of pyramid
A_i	area of single inclined surface of pyramid
A_{it}	area of all inclined pyramid surfaces
A_h	area of horizontal base of pyramid
b	base of pyramid [Fig. 1(b)]
c_b	bulk concentration of copper ions
c_{CuSO_4}	bulk concentration of copper sulphate
$c_{H_2SO_4}$	bulk concentration of sulphuric acid
D	diffusion coefficient of copper ions
f	interference factor
F	Faraday constant
g	gravitational acceleration
H_p	height of pyramid
H_h	vertical height of pyramid triangular base [Fig. 1(b)]
I_L	limiting diffusion current
k	mass transfer coefficient
L	length of inclined surface of pyramid
L_w	characteristic length of Weber [equation (5)]
n	charge number of copper ion
Ra	Rayleigh number
$Ra_{L,\theta}$	Rayleigh number based on inclined surface length and on $g\cos\theta$
Sh	Sherwood number
Sh_L	Sherwood number based on inclined surface length
T	electrolyte temperature.

Greek symbols

$\Delta\rho$	density difference between bulk solution and interface
ρ	density
μ	dynamic viscosity
θ	inclination angle from the vertical [see Fig. 1(b)]
ψ	sphericity.

$$Sh_L = 0.68(Ra_L \cos\theta)^{0.25} \tag{2}$$

in the range $2\times10^6 \leqslant Ra \leqslant 3\times10^{10}$.

Data for down facing surfaces, coupled with upward flow as in the present case, is scarce but Loomba [4] correlated free convective mass transfer at down facing horizontal discs by the equation

$$Sh_d = 0.64 Ra_d^{0.22}. \tag{3}$$

Weber *et al.* [5] measured and correlated natural convection mass transfer from a variety of spherical and non-spherical three dimensional objects, including pyramids in several orientations. The characteristic dimension for such objects, which is sensitive to orientation, was defined as

$$L_w = \frac{\text{surface area}}{\text{perimeter projected onto horizontal plane}}. \tag{4}$$

For objects with sphericity $\psi > 0.6$ for $Sc \approx 2000$ Weber *et al.* [5] obtained the universal correlation:

$$Sh = Sh_0 + 0.53 Ra^{0.256} \tag{5}$$

where Sh_0 is the stagnant medium Sherwood number.

Previous papers from this group [6–8] have described investigations of free convective mass transfer at horizontally oriented cuboids [6], vertical cylinders with active upper and lower ends [7] and down-pointing pyramids [8]; it was found that the overall measured rate of mass transfer was lower than the value predicted by summing the rates of mass transfer at the different individual faces. The experimental total mass transfer data were correlated using a method which includes an interference factor.

3. EXPERIMENTAL

The experiments were performed in a cylindrical 3 dm^3 glass container of internal diameter 15 cm and height 25 cm. Copper sulphate concentration varied from 0.020 to 0.262 M. Each solution contained 1.5 M sulphuric acid as a supporting electrolyte. The electrolyte temperature was carefully measured and always lay within the range 18–21°C being constant to ± 0.1°C during each individual experiment. The pyramidal electrodes were machined from solid copper. Each electrode was supported using 1 mm diameter steel wire glued into a hole in the center of the horizontal base. This wire also served as a current carrier. The wire was lacquered to insulate it from the electrolyte. The pyramidal cathodes were placed in the center of the container. The counter electrode (anode) was a cylindrical copper mesh of diameter 14 cm and height 24 cm. The arrangement of the apparatus is similar to that described previously [7, 8] and is shown in Fig. 1(a). The actual Cu^{2+} concentration was periodically determined by spectrophotometric analysis to an accuracy of approximately $\pm 2\%$.

The usual electrical circuit for limiting current measurement was employed, consisting of a dc power supply with a voltage regulator, a high impedance voltmeter and a multi-range ammeter. Limiting currents were obtained by the well known procedure reported previously [9]. The anode acted as a reference

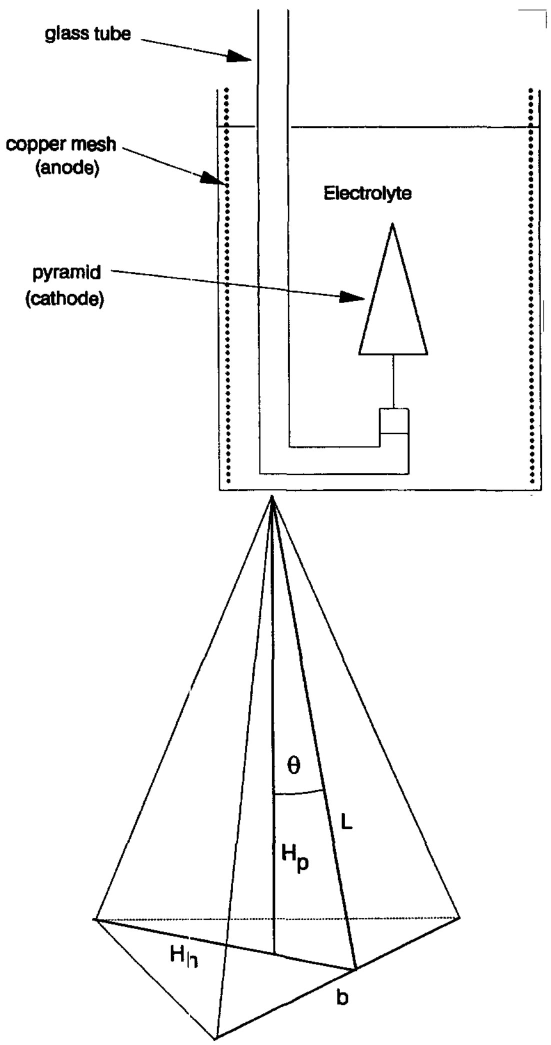

Fig. 1. (a) Apparatus. (b) Geometric parameters of up-pointing pyramids.

electrode in view of its high area compared to that of the cathode. Under such conditions polarization is negligible at the anode and the cell current-voltage relationship depends only on the conditions prevailing at the cathode. The onset of the limiting current was sharp and reproducible and its value was determined to an accuracy of $\pm 3\%$.

Table 1 lists all the geometric characteristics of the pyramids used. This work considers sets of pyramids with constant inclination angle θ, constant length of inclined surface L and varying aspect ratio (length to side of base). In this investigation L ranged from 0.57 up to 7.86 cm and inclination angle from 4.5 up to 50.8°. Values of L are estimated to be accurate within 1–2%. Inclination angles are counted positive to indicate the up-facing orientation of the inclined triangular pyramid faces and for consistency with the work of Patrick *et al.* [3].

The mass transfer controlled limiting currents at the inclined surface and at the down facing horizontal surface were measured separately and in combination e.g. at the pyramid with three active inclined surfaces and at the entire pyramid with the down facing bottom surface also simultaneously active. Surfaces not required to be active were stopped off with lacquer (Lacomit).

The geometric parameters of the up-pointing pyramids are illustrated in Fig. 1(b).

4. RESULTS AND DISCUSSION

4.1. *Mass transfer data calculation*

For each experiment the mass transfer coefficient was calculated from the measured limiting current using the equation:

$$k = \frac{I_L}{AnFc_b}. \qquad (6)$$

The area in this equation was the total available for mass transfer for the particular experiment. The correction for the attachment of the supporting wire was small, never being more than 1.5% of the exposed area and usually being much less.

For the inclined triangular surfaces the data were expressed in the form of a slant height Sherwood number and a slant height Rayleigh number

$$Sh_L = \frac{kL}{D} \qquad (7)$$

$$Ra_{L,\theta} = \frac{g\Delta\rho L^3}{\mu D}\cos\theta. \qquad (8)$$

This approach to data treatment was previously used for the correlation of free convective heat and mass transfer at inclined plates [2, 3, 10] and at cones with an insulated base [11].

The diffusivity of the Cu^{2+} ions was calculated using the equation

$$\frac{\mu D}{T} = (2.495 + 0.0173c_{CuSO_4} + 0.0692c_{H_2SO_4}) \times 10^{-15}\frac{N}{K} \qquad (9)$$

due to Fenech and Tobias [12]. Density and viscosity were calculated using data of Eisenberg *et al.* [13]. The $\Delta\rho$ terms were taken from Wilke *et al.* [14]. The effect of migration on the copper deposition rate was negligible; the migration contribution for the highest concentration of copper sulphate (0.26 M) was $\approx 1.5\%$ [15]. Values of Sh_0 were calculated by the method outlined by Clift *et al.* [16].

4.2. *Single active surface*

Mass transfer coefficients for a single inclined triangular surface are dependent both on the length and inclination angle [3, 8]. Due to this fact, the effect of pyramid length and inclination angle were investigated separately. The mass transfer coefficients to a single inclined surface were measured at pyramids 1–5 (constant inclination angle, varying length) at pyra-

Table 1. Electrode geometries

Pyramid	b	L	θ	H_p	H_h	A_i	A_{it}	A_{pt}	A_{it}/A_h	L/b	L_w	ψ
1	1.05	0.57	31.5	0.49	0.83	0.30	0.90	1.33	2.06	0.54	0.42	0.62
2	2.15	1.19	31.5	1.02	1.86	1.28	3.87	5.87	1.93	0.55	0.91	0.64
3	3.60	1.97	31.5	1.68	3.12	3.55	10.64	16.26	1.89	0.55	1.5	0.64
4	5.34	2.96	31.5	2.52	4.62	7.90	23.71	36.04	1.92	0.55	2.25	0.64
5	7.14	3.95	31.5	3.37	6.18	14.1	42.31	64.37	1.92	0.55	3.0	0.64
6	1.16	1.96	10	1.93	1.00	1.14	3.41	4.02	5.81	1.69	1.15	0.62
7	2.31	1.97	20	1.85	2.00	2.28	6.83	9.14	2.95	0.85	1.32	0.67
8	3.39	1.96	30	1.70	2.94	3.22	10.00	14.98	2.00	0.59	1.47	0.64
9	4.34	1.98	40	1.52	3.76	4.30	12.90	21.05	1.58	0.46	1.62	0.59
10	5.22	1.99	50	1.28	4.52	5.20	15.58	27.38	1.32	0.38	1.75	0.51
11	2.17	0.805	50.8	0.51	1.88	0.87	2.62	4.66	1.28	0.37	0.71	0.51
12	2.22	1.20	32.2	1.06	1.92	1.33	3.99	6.12	1.87	0.54	0.92	0.64
13	2.20	2.06	17.9	2.00	1.91	2.27	6.81	8.91	3.24	0.94	1.35	0.68
14	2.17	3.97	9.0	3.93	1.88	4.31	12.93	15.03	6.16	1.83	2.31	0.62
15	2.13	7.86	4.5	7.83	1.84	8.37	25.11	27.07	12.8	3.70	4.24	0.53

mids 6–10 (constant length, varying inclination angle) and at pyramids 11–15 (varying length and inclination angle).

The effect of the pyramidal length on the mass transfer rate for a single up-facing inclined pyramidal surface of constant (31.5°) and varying (50.8–4.5°) inclination angle is shown in Fig. 2(a). In both cases the mass transfer rate decreases with surface length. The decrease for constant inclination angle is more gradual. It is clear that the mass transfer coefficient decreases not only with increasing length but also with decreasing inclination angle.

The effect of the inclination angle on the mass transfer rate for a single up-facing inclined pyramidal surface of constant (1.98 cm) and varying (7.86–0.805 cm) length is shown in Fig. 2(b). There is a pronounced increase in k as the pyramid length becomes smaller: values are higher for shorter lengths and lower for longer lengths as compared with the constant length data. From Fig. 2(a) and (b) it follows that the mass transfer coefficient for a single up-facing inclined pyramidal surface depends predominantly on the surface length.

A plot of Sh_L against $Ra_{L,\theta}$ for single inclined up-facing triangular surfaces is shown as Fig. 3. A least squares fit of the data yields the relation

$$Sh_L = 1.135(Ra_{L,\theta})^{0.235}. \qquad (10)$$

Forcing a slope of 1/4 as has been reported in the literature for laminar flow on inclined planes [2, 3, 10, 11] gives

$$Sh_L = 0.828(Ra_{L,\theta})^{0.25} \qquad (11)$$

for data in the $Ra_{L,\theta}$ range 1×10^7 to 2×10^{11}. This equation is plotted as the line in Fig. 3 and can be seen to represent the data well. The exponent of 0.25 in equation (11) (as in equation (2) of Patrick *et al.* [3]) indicates the presence of attached flow. The coefficient in equation (11) (0.828) for a single inclined triangular face of an up-facing pyramid is slightly higher than that in the equation of Krysa and Wragg [8] (0.78) for a single inclined triangular face of a down-pointing pyramid and again higher than that in equation (2) of Patrick *et al.* [3] (0.68) for an inclined rectangular surface.

Results for mass transfer to the horizontal triangular down-facing surface are also shown in Fig. 3. with the height of the triangular horizontal base (H_h) as characteristic dimension in the Sherwood and Rayleigh numbers. Least square analysis gives the correlation

$$Sh_{H_h} = 0.45(Ra_{H_h})^{0.243} \qquad (12)$$

and forcing a slope of 1/4 gives

$$Sh_{H_h} = 0.39(Ra_{H_h})^{0.25} \qquad (13)$$

for Ra_{H_h} in the range 1×10^8 to 1×10^{11}. This equation is plotted as the broken line in Fig. 3.

4.3. *Mass transfer at combinations of surfaces*

Mass transfer coefficients for all the inclined triangular surfaces for different pyramid lengths of pyramids 11–15 are shown in Fig. 4 for four different $CuSO_4$ concentrations. The mass transfer correlation for all the inclined surfaces simultaneously active is shown in Fig. 5. Forcing a slope of 1/4 gives the relationship

$$Sh_L = 0.757(Ra_{L,\theta})^{0.25} \qquad (14)$$

for the same $R_{L,\theta}$ range as equation (11). The coefficient in equation (14) is lower than that in (11) since the mass transfer rates at the single active surfaces are not additive because of interactions at the pyramid edges. However, mass transfer is still higher then that for inclined rectangular faces [2, 3, 10, 11].

Mass transfer coefficients for the separate surfaces and for the surfaces in combination are plotted in Fig. 6

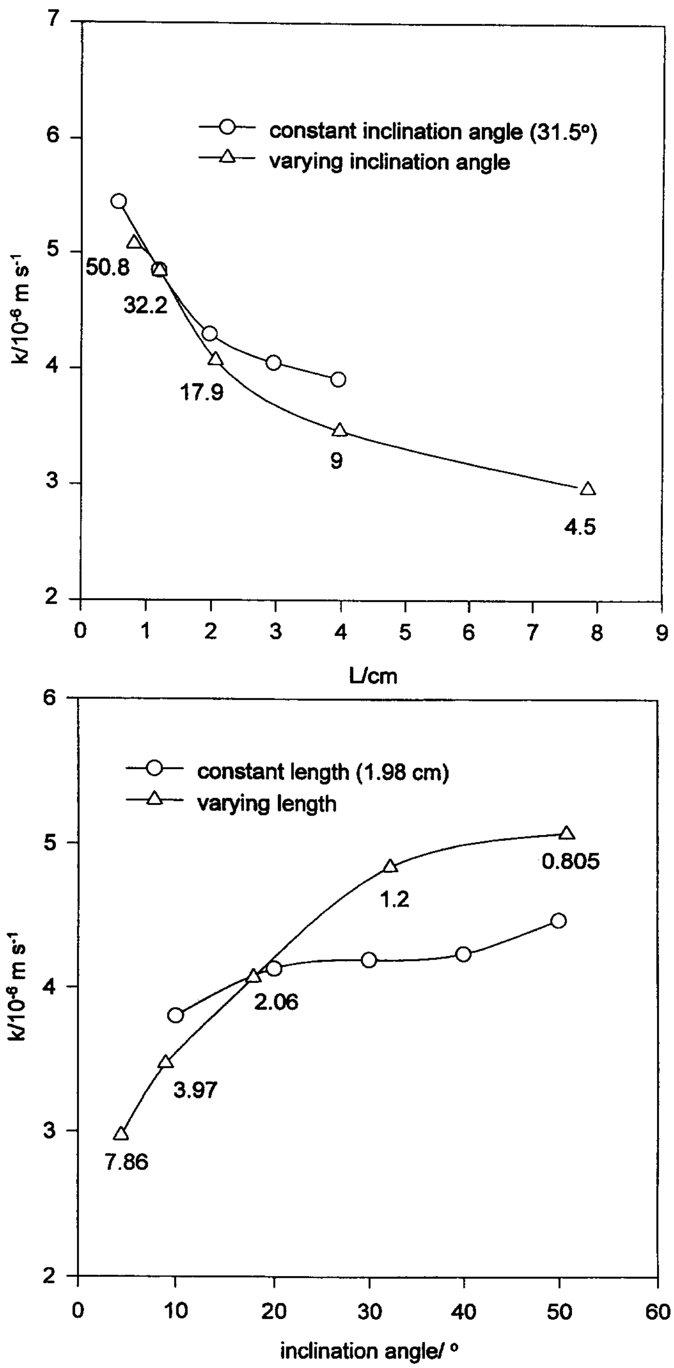

Fig. 2. The effect of (a) length on the mass transfer coefficient for a single up-facing inclined surface of a pyramid of constant (31.5°) and varying inclination angle. (b) Inclination angle on the mass transfer coefficient for a single up-facing inclined surface of a pyramid of constant (1.98 cm) and varying length.

for a single concentration of copper sulphate (0.16 M). It can be seen that a single triangular inclined surface gives higher mass transfer than the combined inclined surface. The down-facing surface gives low mass transfer and strongly influences the total mass transfer rate for the entire up-pointing pyramid.

The effect of pyramid length, L, on the mass transfer coefficient for the entire pyramids 11–15 in up- and down-pointing orientation is shown in Fig. 7. It is clear that the up-pointing case gives a lower mass transfer coefficient than down-pointing. This situation is caused by the dominating behaviour of the horizontal surface; the down-facing horizontal surface (associated with the up-pointing pyramid) not

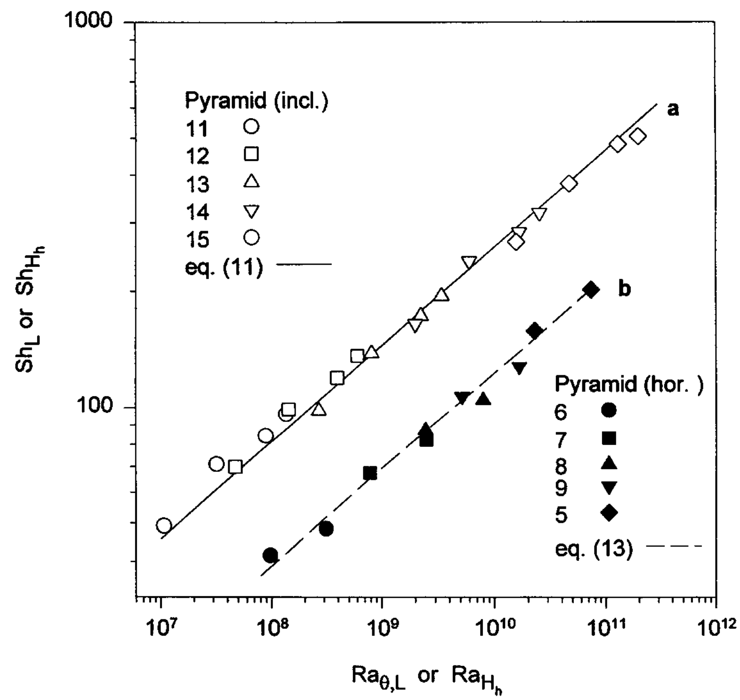

Fig. 3. Mass transfer correlation for (a) single inclined up-facing triangular surfaces; (b) down-facing horizontal triangular surfaces.

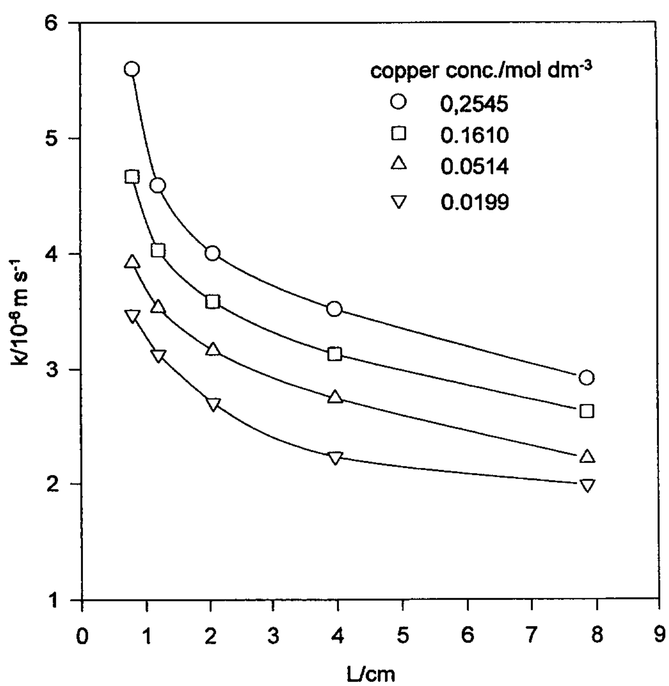

Fig. 4. The effect of length on mass transfer coefficient for all inclined surfaces simultaneously active (constant pyramid base).

only has an intrinsically lower mass transfer performance than an up-facing surface, but also feeds depleted solution to the 'leading edges' of the inclined up-facing surfaces, thus decreasing their contribution to the total behaviour.

4.4. *Overall data correlation*

4.4.1. *Approach using Weber characteristic dimension.* The total mass transfer results from pyramids 11–15 are plotted using the characteristic length defined by Weber *et al.* [5] in Fig. 8. The Weber cor-

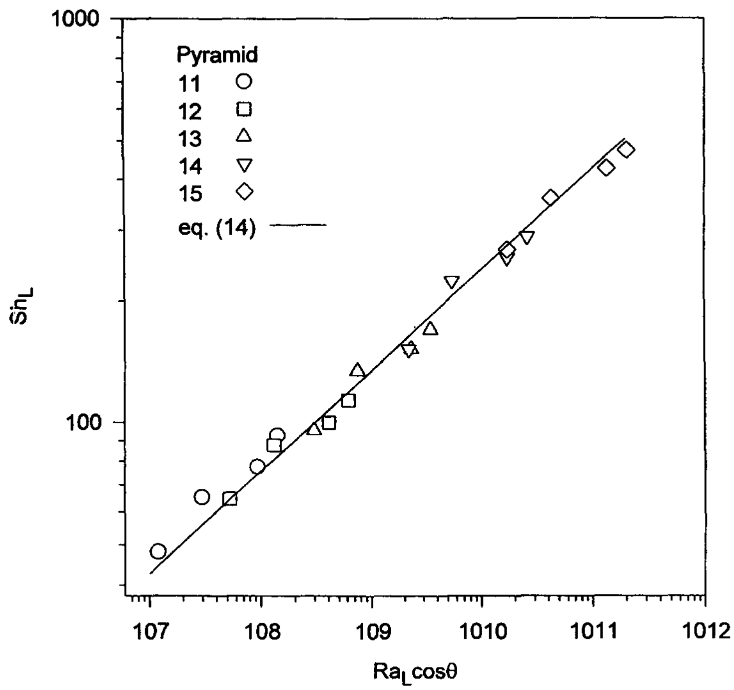

Fig. 5. Mass transfer correlation for all inclined faces simultaneously active.

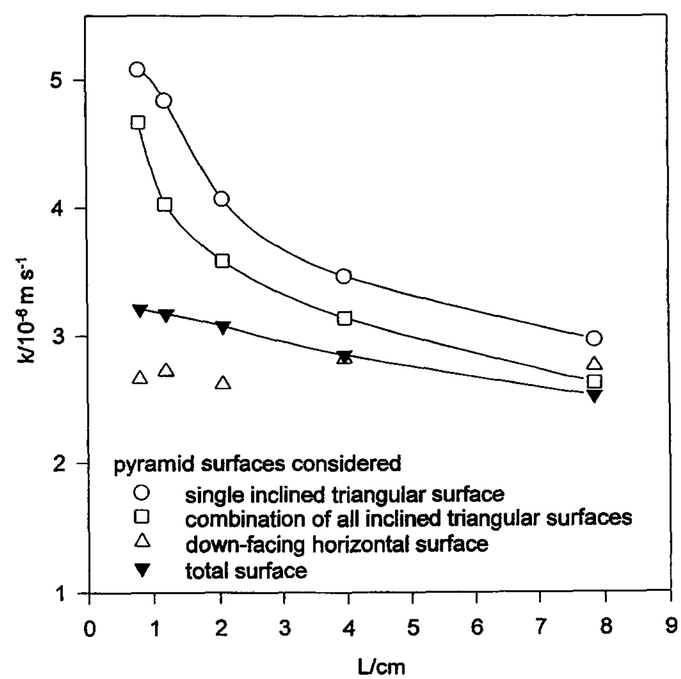

Fig. 6. The effect of length on mass transfer coefficient for different surfaces for a single concentration of copper sulphate (constant pyramid base).

relation equation (5) is also shown. It is seen that the data from all the present pyramids lie under equation (5) and this equation is not suited to the correlation of free convective mass transfer at up-pointing pyramids. This fact may be attributed to the behaviour of the down-facing horizontal surface which strongly effects total mass transfer as described above.

Worthington [6] and the present authors [7, 8] also observed discrepancy with the Weber approach when correlating natural convective mass transfer to

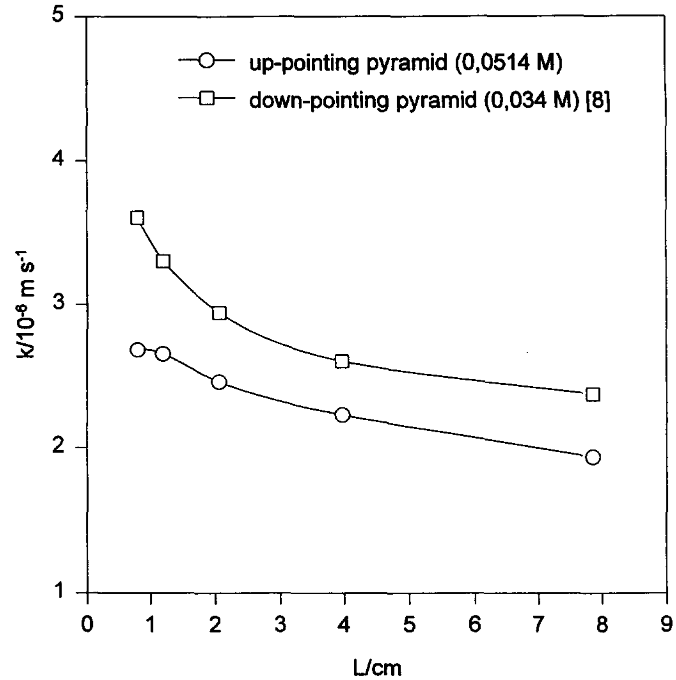

Fig. 7. Comparison of total mass transfer rate for pyramids with constant base in up- and down-pointing orientation.

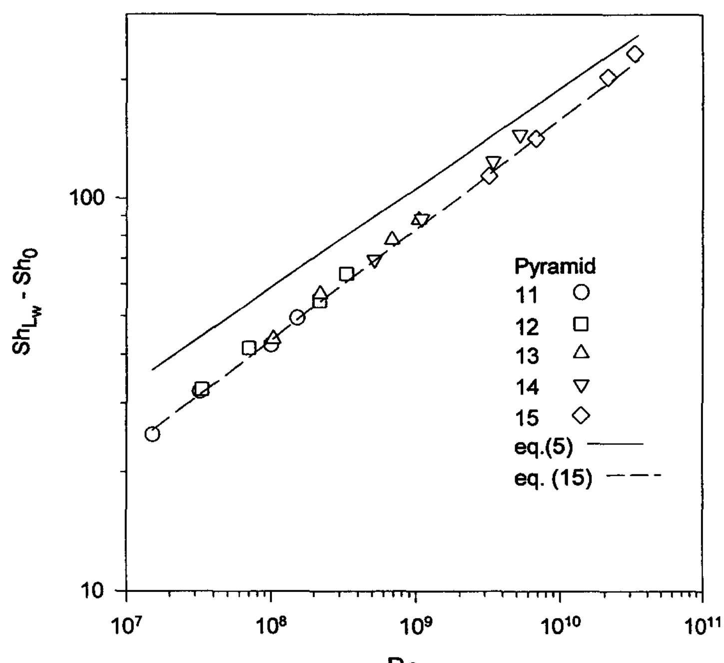

Fig. 8. Correlation of total pyramidal mass transfer data using the Weber *et al.* [5] characteristic dimension.

cuboids [6], vertical cylinders with active ends [7] and to down-pointing pyramids [8]. Nevertheless this probably presents the best possible correlation based on a single characteristic length and a least squares fit through data yields the relation

$$Sh_{L_w} - Sh_0 = 0.222(Ra_{L_w})^{0.287} \qquad (15)$$

for Ra_{L_w} in the range 1×10^7 to 3×10^{10}. This equation is plotted as the broken line in Fig. 8.

4.4.2. *Summation approach to correlation for entire up-pointing pyramids.* By summing the mass transfer rates from correlations for the separate sides a total mass transfer performance can be predicted. The advantage of this method is discussed in [6, 7]. In the present approach the correlation for all the inclined surfaces was taken as equation (14). For the down facing horizontal surfaces the correlation was taken as equation (13). The mass transfer rates for separate surfaces were correlated using length of pyramid (L) and height of horizontal base of pyramid (H_h) as characteristic lengths. The resultant predicted natural convection mass transfer rate for up-pointing pyramids becomes:

$$Sh_L - Sh_0 = \frac{[0.67b + 4.542L(\cos\theta)^{0.25}]}{6L + b\sqrt{3}} Ra_L^{0.25} \tag{16}$$

in the region $9\times10^6 \leqslant Ra_L \leqslant 2\times10^{11}$.

Introducing the relationship

$$\cos\theta = \frac{\sqrt{L^2 - \frac{1}{12}b^2}}{L} \tag{17}$$

to equation (16) yields

$$Sh_L - Sh_0 = \frac{[0.67b + 4.542L^{3/4}(L^2 - \frac{1}{12}b^2)^{1/8}]}{6L + b\sqrt{3}} Ra_L^{0.25} \tag{18}$$

where L and b are the inclined surface pyramid length and the base of the horizontal surface of the pyramid respectively and Ra_L is a Rayleigh number based on the length of the inclined surface of the pyramid.

4.4.3. *Summation approach with interference.* The behaviour predicted by equation (18) was compared with the mass transfer data for pyramids 11–15. It was found in every case that the predicted rate was higher than the actual data but that the shape and gradient of the prediction were well matched. The flow of the fluid around the pyramid base means that the up-facing inclined surfaces are exposed to solution which has already been depleted of copper ions. Thus the overall mass transfer for a pyramid is lower than that for the equivalent summed separate surfaces. A multiplying factor, f (an interference factor), may be introduced to equation (18) to represent the lower rate of mass transfer at up-facing inclined triangular surfaces. This equation now becomes:

Table 2. Dependence of interference factor, f, on geometry of up-pointing pyramid

Pyramid	11	12	13	14	15
f	0.72	0.743	0.761	0.809	0.826

$$Sh_L - Sh_0 = \frac{[0.67b + f4.542L^{3/4}(L^2 - \frac{1}{12}b^2)^{1/8}]}{6L + b\sqrt{3}} Ra_L^{0.25}. \tag{19}$$

Values of f were obtained from equation (19) and calculated for each pyramid. These are tabulated in Table 2. f varies between 0.72 for the smallest pyramid and 0.826 for the tallest pyramid. $Sh_L - Sh_0$ has been plotted to show the data from pyramids 11–15 against $\Phi(L, b).Ra_L$ in Fig. 9. $\Phi(L, b)$ is given by

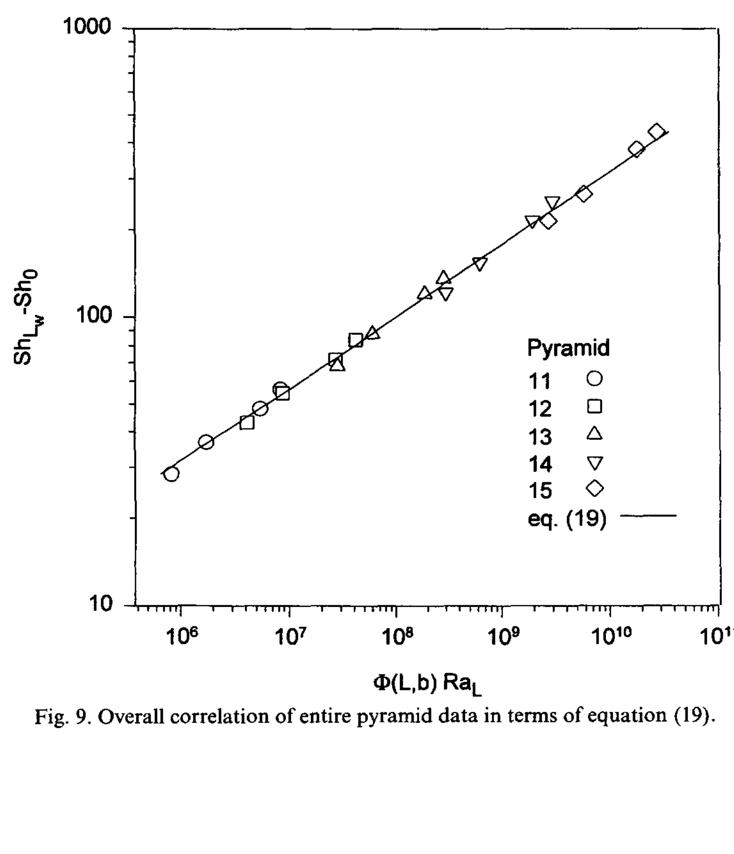

Fig. 9. Overall correlation of entire pyramid data in terms of equation (19).

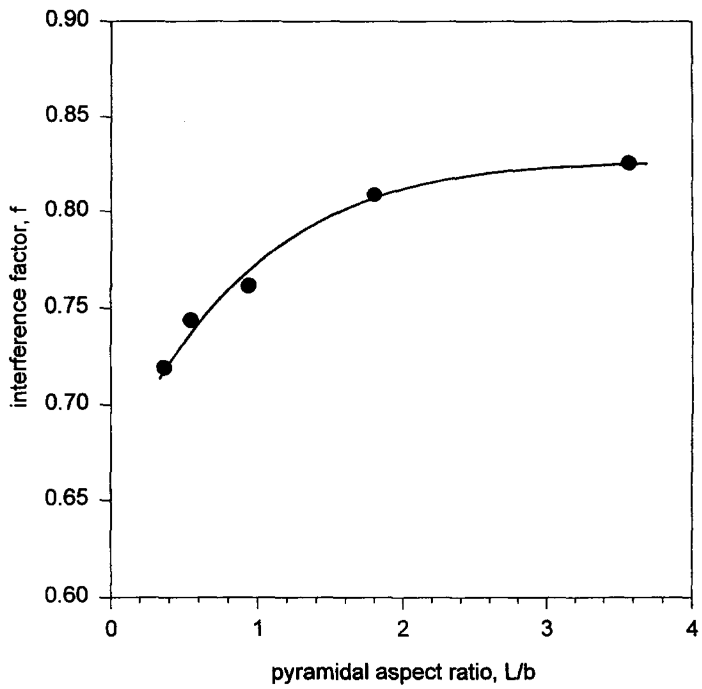

Fig. 10. Plot showing variation of interference factor, f, with pyramidal aspect ratio L/b.

$$\phi(L,b) = \left[\frac{0.67b + f4.542L^{3/4}(L^2 - \frac{1}{12}b^2)^{1/8}}{6L + b\sqrt{3}}\right]^4. \quad (20)$$

It can be seen that all the data fit the line representing equation (19) well. Figure 10 shows the dependence of f on the pyramidal aspect ratio (L/b). The dependence of the interference factor on the aspect ratio of the pyramid (L/b) is not so strong as for vertical cylinders or down-pointing pyramids. Similarly to the case for vertical cylinders [7], the interference factor f increases with aspect ratio. This is caused by the fact that, for the set of vertical cylinders and up-pointing pyramids, the length of the surface exposed to fluid which has already been depleted in copper ions increases and the diameter d (cylinder) or base b (pyramid) of the down-facing horizontal base are the same.

For down-pointing pyramids [8], the interference factor f decreases with pyramidal aspect ratio. This can be explained by the fact that the up-facing horizontal base exposed to fluid which has already been depleted in copper ions is the same. It would be of interest to further investigate total mass transfer rate to a set of up-pointing pyramids where L is constant and change in the ratio L/b is caused by a change in the pyramid base b.

As with work reported in [6–8] the separate sides summation approach incorporating an interference factor has proved highly successful in correlating mass (heat) transfer data for natural convection at a three dimensional solid, and may be recommended for design calculation purposes.

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