FREE CONVECTIVE MASS TRANSFER AT ISOSCELES TRIANGULAR SURFACES OF VARYING INCLINATION

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The limiting diffusion current technique was used for investigation of free convective mass transfer at down-pointing up-facing isosceles triangular surfaces of varying length and inclination. As the mass transfer process, copper deposition from acidified copper(II) sulfate solution was used. It was found that the mass transfer rate increases with inclination from the vertical to the horizontal position and decreases with length of inclined surface. Correlation equations for 7 angles from 0 to 90° were found. The exponent in the Sh_L–Ra_L correlation ranged from 0.247 for the vertical case, indicating laminar flow, to 0.32 for inclinations of 60 to 90°, indicating mixed or turbulent flow. The general correlation $Sh_L = 0.358(Ra_L \sin \theta)^{0.30}$ for the $Ra_L \sin \theta$ range from 7×10^6 to 2×10^{11} and inclination range from 15 to 90° was obtained.

Keywords: Free convection; Triangular surface inclined; Copper deposition; Laminar flow; Turbulent flow; Limiting diffusion current technique; Electrochemistry.

This paper describes an experimental investigation of free convective mass transfer at down-pointing up-facing isosceles triangular surfaces with varying inclination from vertical to horizontal (0 to 90°) using the limiting diffusion current technique (LDCT). The possible mass transfer configurations for inclined triangular surfaces are shown in Fig. 1. The LDCT is an accurate method for determination of mass transfer coefficients and also constitutes a valuable method of simulation for heat transfer[1].

A physical insight into free convection problem is schematically shown in Fig. 2. As an electrochemical system the cathodic deposition of copper on a vertical surface is chosen. The concentration gradient of cupric ions in the diffusion layer results in a density difference between the surface and electrolyte bulk. Therefore the diffusion layer is lighter than ambient fluid

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and it causes a buoyancy force on the fluid close to the cathode surface. As a result a velocity profile in the vicinity of the surface occurs.

Several papers deal with the problem of free convective mass and heat transfer on inclined up-facing surfaces. Patrick *et al.*[2] studied free convective mass transfer on inclined up-facing rectangular surfaces. By using schlieren visualization a number of different possible flow structures was observed. For inclinations 0–30° (from the vertical direction), there was an attached flow along the inclined surface; for higher inclinations, the flow was separated from the surface.

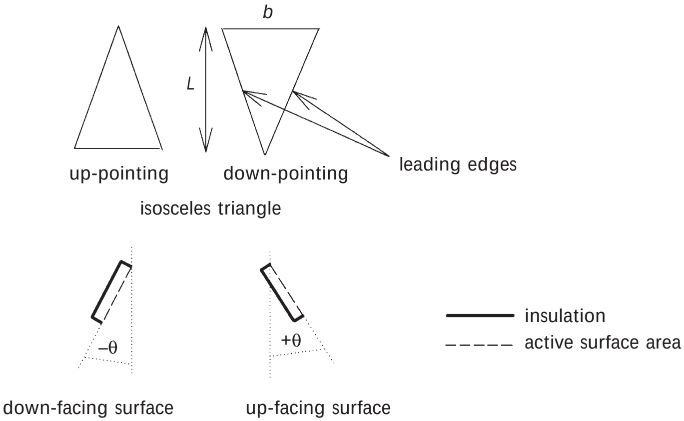

Fig. 1
Possible experimental configurations of triangular surfaces

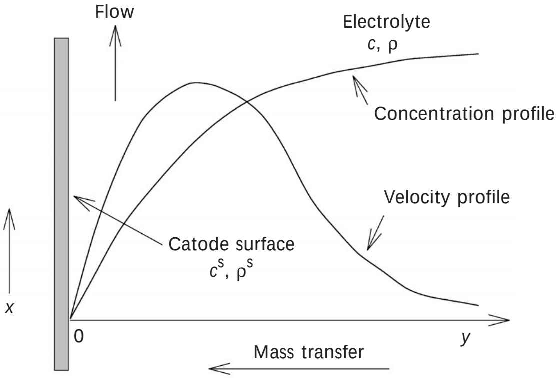

Fig. 2
Velocity and concentration distribution in free convection flow over a vertical surface

Krýsa and Wragg[3] measured mass transfer rates using the LDCT on up-pointing vertical pyramids with triangular bases. The angle of all the inclined triangular surfaces varied from 4.5 to 50.8°. Mass transfer for the single active triangular inclined surface alone was correlated using the equation

$$Sh_\mathrm{L} = 0.828(Ra_\mathrm{L} \cos \theta)^{0.25} \tag{1}$$

for the $Ra_\mathrm{L} \cos \theta$ range from 1×10^7 to 2×10^{11}. For all three triangular surfaces simultaneously active, Krýsa and Wragg[3] obtained the equation

$$Sh_\mathrm{L} = 0.757(Ra_\mathrm{L} \cos \theta)^{0.25} \tag{2}$$

for the same range of $Ra_\mathrm{L} \cos \theta$ as Eq. (*1*). The coefficient in Eq. (*2*) is lower than that in Eq. (*1*) since the mass transfer rates at the single active surfaces are not additive because of interactions occuring at the pyramid edges.

Krýsa *et al.*[4] investigated free convective mass transfer on circular disks with varying inclination. The disks were active on both sides. From schlieren photography it was observed that the flow on the down-facing surface always stays attached to the electrode, whereas the flow on the up-facing surface becomes turbulent and separates at a certain distance from the leading edge depending on inclination and disk diameter. A general correlation $Sh = ARa^B$ was proposed[4] where the coefficients A and B are dependent on the inclination angle.

The present work is a continuation of the previous investigation of Krýsa *et al.*[5] dealing with down-facing down-pointing triangles which proposed the correlation

$$Sh_\mathrm{L} = 0.774(Ra_\mathrm{L} \cos \theta)^{0.25} \tag{3}$$

for $2 \times 10^6 < Ra_\mathrm{L} \cos \theta < 2 \times 10^{11}$.

The problem in free convection is to obtain a single correlation for up-facing inclined surfaces (rectangles, triangles, discs). The situation is complex due to the flow separation from the surface at a certain critical distance resulting thus possibly in a mixture of flow regimes. The aim of this work was to obtain a general mass transfer correlation that would provide the prediction of mass (heat) transfer coefficients for triangular surfaces of various length and up-facing inclination.

EXPERIMENTAL

For the mass transfer measurement, copper deposition from acidified copper(II) sulfate solution was used. The Cu^{2+} concentration was varied from 50 to 250 mol m^{-3}. As a supporting electrolyte, 1500 mol m^{-3} sulfuric acid was used. The Cu^{2+} concentration was determined for each experiment spectrophotometrically at a wavelength of 510 nm with an accuracy ±2%.

The temperature of the electrolyte ranged between 18.2 and 23.9 °C being constant during each experiment to ±0.05 °C. The experiments were carried out in a Perspex tank with dimensions 28 × 15 × 30 cm. Two copper plates situated at the sides of the tank and equidistant from the test cathode were used as anodes (Fig. 3).

The geometric parameters of the triangles are listed in Table I. The length of the base was 2 cm for all triangles. The electrodes were fabricated from 5 mm thick brass sheet. Only one triangular surface was active, the other surfaces being insulated from the electrolyte by coating with lacquer (Lacomit). Before each experiment the active surface was polished with very fine emery paper (grit size 4/0) and then thoroughly washed with distilled water. The triangular electrodes were held in their position by an insulated 2 mm brass wire which also provided electrical connection to the cathode. The objects were held in a chosen position using a Perspex support mechanism (see Fig. 3). Similarly to the previously described work, the inclination angle for the up-facing surface was taken as positive[2,3].

The electric circuit for limiting current measurement consisted of a DC power supply with voltage regulator, a high-impedance voltmeter and a digital ammeter connected to a computer. The anodes acted as reference electrodes due to their high active area (about 500 cm^2) in comparison with the working electrode (maximum 8 cm^2). Under such conditions, polarization is negligible at the anode and the cell current–voltage relationship depends only on the conditions at the cathode.

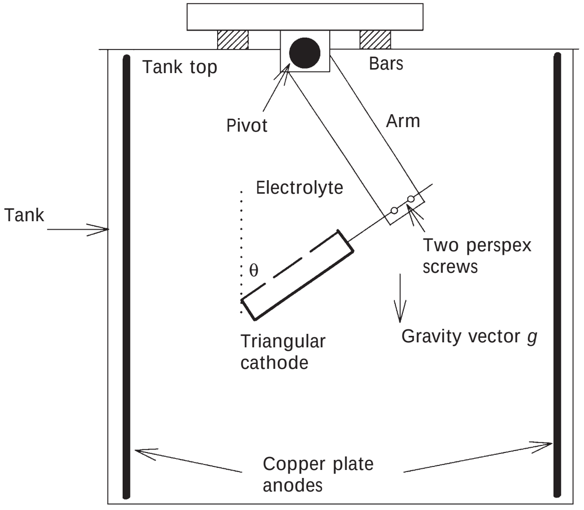

FIG. 3
Support mechanism for down-pointing objects

A polarization curve for each triangle/solution combination was constructed so that the potential at which the limiting current occurred could be determined. When the homogeneous steady state was achieved, limiting currents were obtained by a chronoamperometric technique by applying the cathodic limiting current potential (450–650 mV). The value of limiting current was determined with an accuracy ±1.5%.

RESULTS AND DISCUSSION

Mass Transfer Data Calculations

For each experiment, the mass transfer coefficient, k, was calculated from the measured limiting current, I_{lim}, and the bulk concentration of Cu^{2+} ions, c, using the equation

$$k = \frac{I_{\text{lim}}}{nSFc}\,, \tag{4}$$

where S is the electrode area available for mass transfer and n is the number of electrons for Cu^{2+} ion reduction ($n = 2$). The data were expressed in the form of dimensionless Sherwood and Rayleigh numbers defined as

$$Sh_{\text{L}} = \frac{kL}{D} \tag{5}$$

$$Ra_{\text{L}} = Gr_{\text{L}} Sc = \frac{\Delta\rho g L^3 \rho}{\mu^2}\,\frac{\mu}{\rho D} = \frac{\Delta\rho g L^3}{\mu D}\,, \tag{6}$$

where g is the gravitational acceleration, $\Delta\rho$, ρ, μ and D are physical properties of the medium (see Symbols) and L is the length of the triangular sur-

TABLE I
Geometric parameters of investigated triangular surfaces

No.	L, cm	b, cm	L/b	S, m^2
1	0.52	1.94	0.27	0.000050
2	1.01	1.98	0.51	0.000100
3	2.02	1.98	1.02	0.000200
4	4.08	1.99	2.05	0.000406
5	8.03	2.00	4.02	0.000803

face (Fig. 1). The physical properties of copper(II) sulfate solution acidified with 1500 mol m^{-3} sulfuric acid were evaluated from literature data. The $\Delta\rho$ dependence on the concentration of Cu^{2+} ions was taken from Wilke *et al.*[6] Density and viscosity were calculated using the data of Eisenberg *et al.*[7] The Cu^{2+} ion diffusion coefficient was calculated from the equation

$$\frac{\mu D}{T} = (2495 + 0.0692 c_{\mathrm{CuSO_4}} + 0.0173 c_{\mathrm{H_2SO_4}}) \times 10^{-18} \tag{7}$$

due to Fenech and Tobias[8], where the concentrations, c, are expressed in mol m^{-3}, viscosity, μ, in kg m^{-1} s^{-1}, diffusion coefficient, D, in m^2 s^{-1} and thermodynamic temperature, T, in K. The effect of migration on the copper deposition rate was negligible: the highest contribution (about 3%) was for the highest concentration of Cu^{2+} ions (250 mol m^{-3}) and the lowest contribution (0.3%) was for the lowest concentration of Cu^{2+} ions (50 mol m^{-3}). For more information see Ibl and Dossenbach[9] or Krýsa *et al.*[10]

Mass Transfer Measurement

The effect of inclination angle on the mass transfer coefficient for down-pointing up-facing triangles of various lengths for a single concentration of Cu^{2+} ions (172 mol m^{-3}) is shown in Fig. 4. The shape of the plots was similar for all concentrations (50, 113 and 250 mol m^{-3}).

In considering free convection at horizontal surfaces the orientation of the horizontal surface must be taken into account. In systems with metal

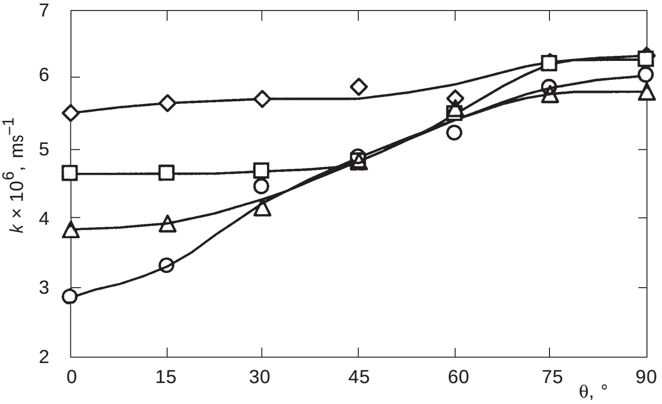

FIG. 4

Effect of inclination angle, θ, on mass transfer coefficient for down-pointing up-facing triangles of various lengths (cm): ◇ 0.5, □ 1, △ 2, ○ 8; Cu^{2+} concentration 172 mol m^{-3}

deposition (mass transfer) or a solid body of higher temperature than the ambient fluid (heat transfer) the down-facing orientation always exhibits a much lower mass (heat) transfer coefficient than the up-facing one[11,12]. This is due to the fact that a down-facing horizontal surface hinders flow resulting from the density difference. On the other hand flow at up-facing horizontal surface leads to flow instability. Depending on the concentration, temperature and the size of surface various turbulent regimes appear, which can flow freely upward from the surface[2].

The highest difference between horizontal and vertical positions was observed for the longest triangular surface (L = 8 cm). In general, the mass transfer coefficient increases with increasing inclination (from vertical to horizontal) and decreases with rising inclined surface length. In the inclination region from ≈75 to 90°, the mass transfer coefficients for all lengths are similar and the influence of length is small.

For the shortest electrode (L = 0.5 cm), inclination has a very small influence on the mass transfer coefficients; there is only a slight increase from the vertical (0°) to the horizontal (90°) position. For the longer electrodes (L = 1–4 cm), there are three regions on the plot. In the first from 0 to ≈30° (L = 1 cm) and from 0 to 15° (L = 2–4 cm), the values of mass transfer coefficients are almost constant. In the second region from 45° (L = 1 cm) and from 30° (L = 2–4 cm) to 75°, the mass transfer coefficients increase with increasing inclination. In the last region from 75 to 90° the mass transfer coefficients are again almost constant for all three triangles (L = 1–4 cm). For the longest electrode (L = 8 cm), there are only 2 regions. From 0 to 75° k increases and from 75 to 90° it is almost constant.

The effect of the length of inclined triangular surface with various inclinations on mass transfer coefficient for concentration of Cu^{2+} ions of 50 mol m^{-3} is shown in Fig. 5. The influence of length is the most significant in vertical position (0°) where the mass transfer coefficient decreases over the entire length, while for higher inclinations (45 and 90°) in a region L = 2–8 cm the mass transfer coefficient is almost length independent.

Mass Transfer Data Correlation

Mass transfer data for the inclined triangular surfaces were correlated using a general equation of the form

$$Sh_{\mathrm{L}} = A(Ra_{\mathrm{L}})^{B} \,. \qquad (8)$$

An example of this correlation is shown in Fig. 6 for an inclination of 30°. A least-squares fit gives the equation

$$Sh_L = 0.331(Ra_L)^{0.293} \tag{9}$$

which is shown as the solid line in Fig. 6.

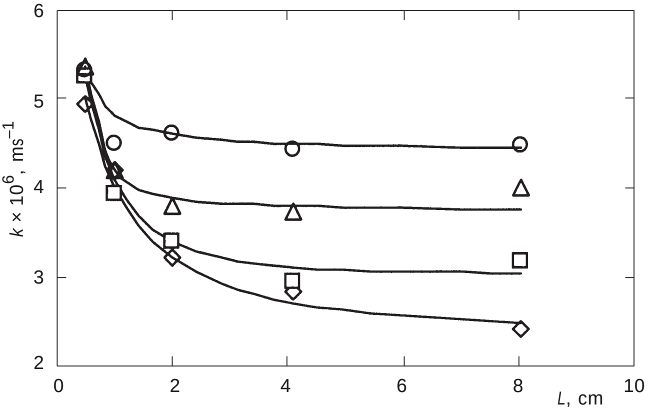

FIG. 5
Effect of the length of inclined surface for several inclination angles (°): ◇ 0, □ 30, △ 45, ○ 90; Cu^{2+} concentration 50 mol m^{-3}

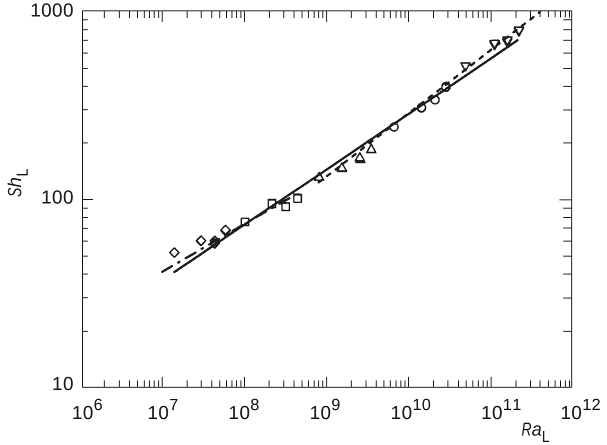

FIG. 6
Correlation of mass transfer data from down-pointing triangular surfaces inclined at +30°; L (cm): ◇ 0.5, □ 1, Δ 2, ○ 4.1, ∇ 8; —— Eq. (*9*), – – – Eq. (*10*), – · – · Eq. (*11*)

It is evident that for low and high *Ra* numbers the data are higher than the correlating line. It has previously been observed for up-facing inclined rectangular surfaces[2] that there is a transition from laminar attached flow to separated flow which depends on length and inclination. As the inclination increases from vertical to horizontal, the critical length at which the flow starts to separate from the surface decreases. Therefore, for constant inclination at short electrodes, the flow is predominantly laminar while at longer electrodes the flow is predominantly turbulent. This approach was applied to the correlation for inclination 30°.

Mass transfer data at shorter lengths (0.5, 1.0 cm) were correlated by forcing an exponent to 0.25.

$$Sh_\mathrm{L} = 0.733(Ra_\mathrm{L})^{0.25} \qquad (10)$$

On the other hand, mass transfer at longer electrodes (2–8 cm) was correlated by applying an exponent of 0.33.

$$Sh_\mathrm{L} = 0.132(Ra_\mathrm{L})^{0.33} \qquad (11)$$

These particular equations are shown in Fig. 6 as dash-dotted and dotted lines and it can be seen that the fit is better than in the case of Eq. (*9*).

Sh–*Ra* correlations were performed for all other inclination angles. Coefficients *A* and *B* were obtained using a least-squares analysis (Table II). Both coefficients *A* and *B* depend on inclination. The coefficient *B* in the correlation for inclinations of 0° (vertical) was very close to 0.25 and the coefficients in the correlations for inclinations of 60, 75 and 90° were close to 0.33. Therefore, these coefficients were forced to these correlations and appropriate coefficients are shown in brackets (Table II). The assumption of turbulent flow for inclinations of 60–90° is based, not only on the exponent in the *Sh*–*Ra* correlations, but also on the visualization of flow at similar objects such as rectangles[2] and discs[4].

The general correlation for triangular surfaces of various inclinations can be written as

$$Sh_\mathrm{L} = f_\mathrm{A}(Ra_\mathrm{L})^{f_\mathrm{B}}\,, \qquad (12)$$

where f_A and f_B are functions of inclination angle, where f_A was approximated by a Boltzmann function (Eq. (*13*)) and f_B by a polynomial function (Eq. (*14*)).

$$f_A = \frac{0.5873}{1 + \exp\left\{\dfrac{\theta - 24.6085}{7.0202}\right\}} + 0.1741 \qquad (13)$$

$$f_B = 0.2476 + 6.9534 \times 10^{-4}\,\theta + 2.8037 \times 10^{-5}\theta^2 - 2.8231 \times 10^{-7}\theta^3 \qquad (14)$$

Coefficients *A* and *B* obtained using least-squares analysis for single inclination correlations and shown in Table II are plotted in Fig. 7 as a function of inclination. For inclinations 0, 60, 75 and 90°, coefficients in brackets (Table II) were used.

The functions (*13*) and (*14*) are also plotted in Fig. 7 and compared with the coefficients obtained using least-squares analysis (Table II). There is a good agreement between values of coefficients and functions f_A and f_B. Therefore, using Eqs (*12*)–(*14*), a mass transfer coefficient for any inclination angle between 0 and 90° can be predicted.

There is another possible approach to the correlation of mass transfer data for all inclinations (except for 0°). The term sin θ was introduced into the correlation to take into account the effect of inclination and the overall correlation produced is shown in Fig. 8.

TABLE II
Coefficients *A* and *B* for various inclinations

θ, °	*A*	*B*	R^2
0	0.774 (0.734)	0.247 (0.25)	0.9965 (0.9980)
15	0.663	0.256	0.9929
30	0.331	0.293	0.9854
45	0.265	0.307	0.9907
60	0.210 (0.155)	0.321 (0.33)	0.9960 (0.9945)
75	0.245 (0.162)	0.317 (0.33)	0.9980 (0.9964)
90	0.266 (0.168)	0.315 (0.33)	0.9965 (0.9977)

A least squares fit through the data (80 points) gives the correlation

$$Sh_L = 0.358(Ra_L \sin \theta)^{0.30} . \qquad (15)$$

The exponent 0.30 suggests that on most triangles the flow is transitional between laminar and turbulent.

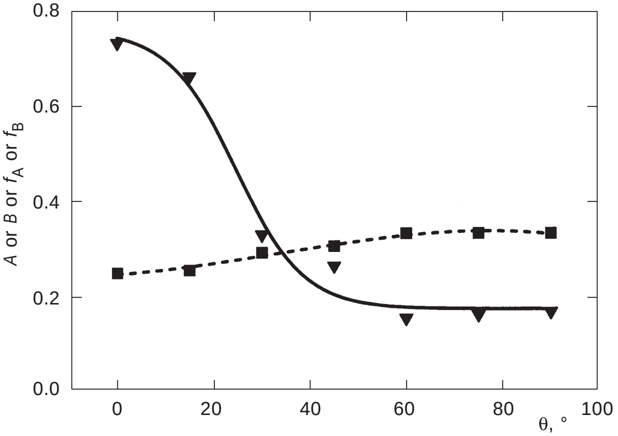

FIG. 7
The effect of inclination on the coefficients *A* and *B* in correlating equation (*12*); ▼ *A*, ■ *B*; —— Eq. (*13*), – – – Eq. (*14*)

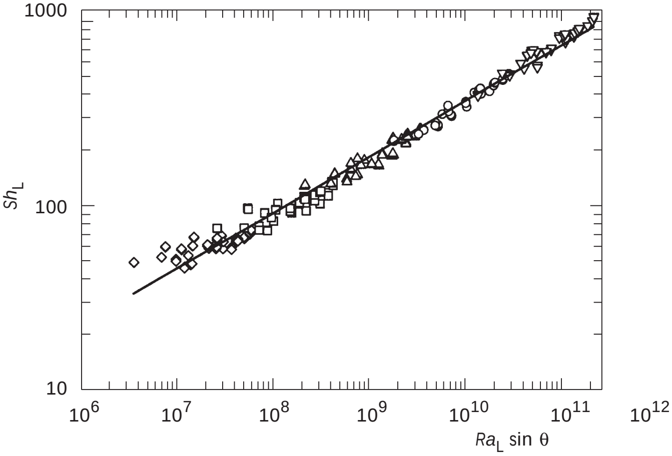

FIG. 8
Overall correlation of mass transfer data from up-facing down-pointing triangular surfaces inclined in the range from 15 to 90°; *L* (cm): ◇ 0.5, □ 1, △ 2, ○ 4.1, ∇ 8; —— Eq. (*15*)

CONCLUSIONS

The effect of inclination on mass transfer coefficient is very small for short surfaces ($L/b \leq 0.25$).

The coefficients A and B in the correlation $Sh_L = A(Ra_L)^B$ were found to be dependent on inclination. Exponent B shows that for the vertical orientation the flow is laminar, for inclinations 15 to 45° it is transitional and for inclinations 60 to 90° it is turbulent.

A general correlation for up-facing down-pointing inclined triangular surfaces was obtained in the form $Sh_L = f_A(Ra_L)^{f_B}$ where f_A and f_B are functions of inclination angle in the range from 0 to 90°.

A simplified overall correlation was also obtained by introducing the term sin θ

$$Sh_L = 0.358(Ra_L \sin\theta)^{0.30}$$

for the Ra_L sin θ range from 7×10^6 to 2×10^{11} and inclination angles betwen 15 and 90°.

SYMBOLS

b	length of base of triangular surface, m
c	bulk concentration of Cu^{2+} ions, mol m^{-3}
c^s	surface concentration of Cu^{2+} ions, mol m^{-3}
D	diffusion coefficient of Cu^{2+} ions, m^2 s^{-1}
F	Faraday constant, 96 487 C mol^{-1}
g	gravitational acceleration, 9.81 m s^{-2}
Gr_L	Grashof number based on the length of inclined surface L, Eq. (*6*)
I_{lim}	limiting diffusion current, A
k	mass transfer coefficient, m s^{-1}, Eq. (*4*)
L	length of inclined surface, m
n	charge number of Cu^{2+} ion, 2
Ra_L	Rayleigh number based on the length of inclined surface L, Eq. (*6*)
S	active electrode area, m^2
Sc	Schmidt number, Eq. (*6*)
Sh_L	Sherwood number based on the length of inclined surface L, Eq. (*5*)
T	electrolyte temperature, K
μ	dynamic viscosity, kg m^{-1} s^{-1}
θ	inclination angle from vertical direction, °
$\Delta\rho$	density difference between bulk solution and surface, kg m^{-3}
ρ	density of bulk solution, kg m^{-3}
ρ^s	surface density of solution, kg m^{-3}

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