# Natural convection mass transfer at spheres

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Received 18 March 1983

Mass transfer by natural convection at spheres has been studied by an electrochemical technique involving limiting current measurement for the anodic dissolution of copper spheres in phosphoric acid. Acid concentration and sphere diameter were changed to provide values of *Sc. Gr* ranging from  $2.85 \times 10^{10}$  to  $2.15 \times 10^{11}$ ; under these conditions the mass transfer data was correlated by the equation:

 $Sh = 0.15 (Sc. Gr)^{0.33}$ 

#### Nomenclature

- I Limiting current density
- K mass transfer coefficient
- F Faraday's constant
- *C* Saturation solubility of copper phosphate in phosphoric acid
- Z number of electrons involved in the reaction
- Sh Sherwood number = Kd/D
- Sc Schmidt number =  $v_{ave}/D$
- Gr Grashof number =  $gd^3(\rho_i \rho_b)/\nu_{ave}^2\rho_i$
- d Sphere diameter
- D diffusivity of copper ions
- $u_{\mathbf{b}}$  viscosity in the bulk liquid
- $u_i$  viscosity at the interface
- $\rho_{\mathbf{b}}$  density in the bulk liquid
- $\rho_i$  density at the interface
- g acceleration due to gravity
- $v_{ave}$  average Kinematic viscosity
- Nu Nusselt number
- Pr Prandtl number

#### 1. Introduction

Although some work has been reported on forced convection mass and heat transfer at spheres, little has been done on free convection heat and mass transfer at such surfaces despite the practical importance of the subject. Schutz [1], using an electrochemical technique involving the cathodic deposition of copper from acidified copper sulphate, studied natural convection mass transfer at spheres in the range  $1.22 \times 10^7 < Sc. Gr < 1.51 \times 10^{10}$ . He correlated his data with the equation:

$$Sh = 2 + 0.59 (Gr. Sc)^{0.25}$$
 (1)

In the area of heat transfer, Merk and Prins [2] made a theoretical analysis of natural convection at spheres in the laminar flow region and deduced the equation:

$$Nu = 2 + 0.55 (Gr. Pr)^{0.25}.$$
 (2)

Yuge [3] obtained the following equation for free convection heat transfer from spheres to air:

$$Nu = 2 + 0.43 \, (Gr. \, Pr)^{0.25}. \tag{3}$$

Mikheyev [4] correlated heat transfer data at spheres in the range  $2 \times 10^7 < Gr$ .  $Pr < 1 \times 10^{13}$  by the equation:

$$Nu = 0.135 (Gr. Pr)^{0.33}.$$
 (4)

The object of the present work is to study free convection mass transfer at spheres under conditions providing values of *Gr. Sc* which extend beyond the range studied by Schutz [1]. To this end the limiting current of the anodic dissolution of copper spheres in phosphoric acid was measured and used to calculate the mass transfer coefficient. The diffusion controlled dissolution of copper in phosphoric acid was successfully used in previous natural convection studies [5, 6] and was found to produce results in agreement with those obtained by the cathodic deposition of copper from acidified copper sulphate. The advantage of the technique over copper deposition is that the electrode surface remains smooth during the experiment without interference from rough deposits as in the case of copper deposition from high CuSO<sub>4</sub> concentrations.

#### 2. Experimental technique

Figure 1 shows the cell and electrical circuit used in the present work. The cell consisted of a cylindrical glass container of diameter 11 cm and a height of 15 cm divided into two compartments by a cylindrical porous PVC diaphragm of diameter 9 cm. This diaphragm served to prevent stirring by the hydrogen bubbles evolved at the cathode, interfering with the natural convection process at the anode. The cathode consisted of a cylindrical copper sheet of diameter 10 cm and height 14 cm placed in the outer compartment of the cell and it acted also as a reference electrode by virtue of its high surface area compared to that of the anode. The anode was made of a copper sphere ranging in diameter from 2.1 to 5.75 cm. The electrical circuit consisted of a 6 volt d.c. power supply, a variable resistance and a multirange ammeter connected in series with the cell. A high impedance voltmeter was connected in parallel with the cell to measure its potential. Three concentrations of phosphoric acid were used: 8, 10 and 12 mol  $dm^{-3}$ ; all were prepared from AR grade phosphoric acid.

Polarization curves, from which the limiting current was determined, were plotted by increasing the applied current step-wise and measuring the corresponding steady state potential. Before each run, the anode was polished with fine emery paper, degreased with trichloroethylene, washed with alcohol and finally rinsed in distilled water. The anode was positioned in the centre of the cell. An insulated copper wire (2 mm diameter) brazed to the sphere held the anode in position and acted as the electrical contact. Temperature was kept constant at 22°C. Each run was carried out using a fresh solution and a new electrode. The sphere diameter was measured before and after each experiment to make sure that no significant dimensional change had taken place during the experiment. Each experiment was repeated two or three times.

#### 3. Results and discussion

Figure 2 shows typical polarization curves with a well defined limiting current plateau; from these curves the limiting current density, *I*, was determined and used to calculate the mass transfer coefficient according to the equation:

$$\frac{I}{ZF} = KC.$$
 (5)

The saturation solubility of copper phosphate in different phosphoric acid concentrations (C)was obtained from the data of Sedahmed *et al.* [5]. The variation of the mass transfer coefficient (K) with sphere diameter is shown in Fig. 3; within the range of experimental error the mass



Fig. 1. Cell and electrical circuit.





transfer coefficient is almost independent of the sphere diameter thus denoting a turbulent flow mechanism. An overall mass transfer correlation was envisaged in terms of the dimensionless groups Sh, Sc and Gr. Values of solution densities, viscosities and diffusivities required to calculate these groups were determined in a previous study [5]. Table 1 summarizes the values of the dimensionless groups used to obtain the overall mass transfer correlation. Figure 4 shows that the present mass transfer data can be correlated within the range

 $2.85 \times 10^{10} < Sc. Gr < 2.15 \times 10^{11}$  by the equation:

$$Sh = 0.15 (Sc. Gr)^{0.33}$$
. (6)

The average deviation was  $\pm 7.76\%$ . Again the exponent 0.33 in Equation 6 reveals a turbulent flow mechanism, this is consistent with the finding of Schutz that his range of study  $(1.22 \times 10^7 < Sc. Gr < 1.51 \times 10^{10})$  was in the transition region. On comparing Equation 6 with the corresponding heat transfer equation (Equation 4), it would be found that Equation 4 predicts higher transfer



Fig. 3. Effect of sphere diameter on the mass transfer coefficient. Symbols as Fig. 2.

$CH_{3}PO_{4}$ (mol dm <sup>-3</sup> )	Sphere diameter (cm)	$10^{5} \text{ K}$ (cm s <sup>-1</sup> )	Sh	Sc	Gr
8	2.10	14.44	300	8.128 × 10 <sup>4</sup>	125000
8	2.97	16.85	494.7	$8.128 \times 10^{4}$	352200
8	3.74	13.92	315.2	$8.128 \times 10^4$	705500
8	5.02	15.36	763.6	$8.128 \times 10^4$	1710000
8	5.75	15.88	846.3	$8.128 \times 10^{4}$	2559400
10	2.10	11.04	351.2	$1.68 \times 10^{5}$	60283
10	2.97	9.93	446.2	$1.68 \times 10^{5}$	169850
10	3.74	8.28	468.9	$1.68 \times 10^{5}$	340200
10	5.02	9.35	711.3	$1.68 \times 10^{5}$	824500
10	5.75	8.28	720.6	$1.68 \times 10^{5}$	1234200
12	2.10	5.94	311.7	$3.59 \times 10^{5}$	29350
12	2.97	5.31	393.5	3.59 × 10⁵	82705
12	3.74	5.31	496.2	$3.59 \times 10^{5}$	165660
12	5.02	6.6	828.5	$3.59 \times 10^{5}$	401500
12	5.75	5.31	762.5	$3.59  imes 10^{s}$	600980

Table 1. Dimensionless groups used in data correlation. Temperature =  $22^{\circ}$  C

rates under the same conditions. This discrepancy may be attributed to the difference in Pr and Sc, since Equation 4 was obtained for fluids of Pr = 0.7. A similar discrepancy was noted in the case of horizontal plates. Wragg and Loomba [7] represented turbulent flow free convection mass transfer at horizontal plates by the equation:

$$Sh = 0.18 (Sc. Gr)^{0.33}$$
. (7)

The corresponding heat transfer equation was found by Fishenden and Saunders [8] to be:

$$Nu = 0.15 (Pr. Gr)^{0.33}.$$
 (8)

Mikheyev [4] found that Equation 4 not only

applies for spheres but also for other geometries such as vertical plates and cylinders, and horizontal plates and cylinders. A comparison between Equation 6 and Equation 7 shows that such generalization could not be extended to mass transfer where the geometry of the transfer surface affects the rate of mass transfer; again this difference in behaviour between heat and mass transfer may be attributed to the difference in the value of Sc and Pr.

### References

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Fig. 4. Overall mass transfer correlation.

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