Note

Effect of System Geometry on Gas Phase Mass Transfer in Electromagnetic Levitation Assembly

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1. Introduction

The levitation technique is frequently employed in the study of reaction kinetics of high temperature gas-liquid systems. This technique offers a number of advantages over the conventional crucible method since the melt is free from contamination by container materials and the reaction interface is well defined. Further, it is also useful to simulate the environment of intensive reactors in which liquid drops encounter reactive gases.

The quantitative description of process kinetics under levitation conditions requires accurate determination of mass transfer coefficients in the gas phase. This may pose some difficulty in a levitation system since the mass transfer coefficient is dependent on the properties of the fluid, system geometry, temperature *etc.* The work of previous investigators has highlighted the difficulty in describing mass transfer in levitation conditions where large temperature and concentration gradients exist. There are a number of correlation equations developed to predict mass transfer coefficients for spherical geometries.^{1–11)} However, using such equations in conditions which are significantly different to those in which they were developed must be undertaken with caution. Any such correlation equation must be validated in the conditions where it is employed. This work shows the importance of confirming the validity of a correlation equation when applied to the levitation system. Naphthalene evaporation is used to determine the mass transfer coefficient.

2. Experimental

The gas phase mass transfer coefficients of a droplet in the levitation geometry were determined by the evaporation of naphthalene spheres at the ambient

temperature. **Figure 1** shows a schematic diagram of the levitation apparatus used for kinetic studies of gas-metal reactions. Experiments were performed with naphthalene spheres with this apparatus in order to quantify the forced convection characteristics of the system. Beads of naphthalene were cast of similar shape to that of the levitated metal drop (6.0×10^{-3} m in diameter). The accurate dimensions of the drop were obtained by dipping it in liquid naphthalene to increase its diameter or abrading the surface of the bead to reduce it. These were mounted on a 1 mm brass wire. Each bead was then placed within the levitation apparatus to occupy the position of the levitated drop. Nitrogen gas was then passed over the bead at varying gas flowrates for varying durations. In order to determine the effect of the container walls and the levitation coil on gas phase mass transport within the present geometry, naphthalene evaporation experiments were also undertaken in the absence of the coil. The rate of mass loss of naphthalene was obtained by recording the mass loss on a single pan digital balance.

3. Results and Discussion

The mass loss on evaporation of naphthalene in the levitation vessel for various gas flowrates was measured. The change in bead dimensions was negligible for the measured mass loss and hence the mass transfer coefficient is given by Eq. (1):

$$k_g = \frac{\dot{W}RT}{128.17AP_s} \quad (\text{m s}^{-1}) \quad \dots\dots\dots(1)$$

where k_g is the gas mass transfer coefficient (m s^{-1}), $\dot{W}$ is the rate of mass loss of naphthalene, R is the universal gas constant ($\text{m}^3 \text{atm mol}^{-1} \text{K}^{-1}$), T is the ambient temperature (K), 128.17 is the molecular weight of naphthalene, A is the surface area of the sample (m^2), and P_s is the vapour pressure of naphthalene (atm).

Then the mass transfer coefficients obtained from

Fig. 1. Schematic diagram of experimental apparatus.

Fig. 2. Experimental results of naphthalene evaporation at different gas flow rates. Prediction by the Steinberger-Treybal correlation equation is superimposed.

above may be used to compare with predictions made by a correlation equation for the similar geometry to the present system. The Steinberger-Treybal correlation equation⁵⁾ was developed to describe mass transfer to or from a sphere in an infinite fluid medium and in the absence of any significant temperature gradients. It is given by Eq. (2):

$$Sh = 2 + 0.569(Gr'Sc)^{0.25} + 0.347(Re^{0.62}Sc^{0.31}) \quad \dots(2)$$

where Sh is the Sherwood number, Gr' is the mean Grashof number, Sc is the Schmidt number, and Re is the Reynolds number. Details of these numbers are given in the list of symbols. This equation assumes that the contributions to mass transfer from diffusion, natural convection and forced convection are linearly additive. In ambient room temperature conditions, the natural convection term (the second term on the right hand side of Eq. (2)) is negligible and hence Eq. (2) is reduced to a linear equation of the form given by Eq. (3):

$$Sh = 2 + n(Re^{0.62}Sc^{0.31}) \quad \dots(3)$$

where n is the geometry dependent coefficient of the forced convection term. This coefficient may be obtained by a cold model study for the system geometry of interest. The modified Steinberger-Treybal equations can then be used to calculate the gas phase mass transfer coefficient for the system. Several authors^{12,13)} have applied this strategy to their levitation experiments. However, the complexity of these systems warrants that the mass transfer coefficients be validated by comparing them with values obtained independently in levitation conditions.

Figure 2 shows the experimental results with predictions of the Steinberger-Treybal correlation. It clearly shows that, in the flow regimer where the Reynolds number is smaller than about 300 ($Re^{0.62} \cong 35$), the Steinberger-Treybal correlation significantly underestimates the mass transfer coefficient. When the Reynolds number is greater than 600 ($Re^{0.62} = 53$), then the mea-

sured mass transfer rates are in good agreement with the prediction of the Steinberger-Treybal correlation. Figure 2 also shows a transition regime between the Reynolds numbers of 300 and 600 where the effect of enhanced mass transfer due to the system geometry is rapidly diminished. The observations given above imply that the presence of the levitation coil enhances the gas phase mass transport at lower Reynolds numbers. This behaviour may be explained by the fact of the flow separation around the sphere. The work of several investigators¹¹⁾ have shown that there is significant variation of the local mass transfer rates over the surface of a sphere. The mass transport rates in the front of the sphere can be significantly greater than that at the rear for increasing gas velocity. Therefore, the contribution from enhanced mass transfer behind the sphere diminishes since the total mass transport rate is dominated by transfer from the front of the sphere. Hence the presence of the levitation coil has little effect on gas phase mass transfer for large Reynolds numbers. These results clearly identify that the geometry of the reaction system is an important factor which influences mass transport in the gas phase. For the geometry employed in the present study, therefore the Steinberger-Treybal correlation equation need to be modified to fit the measured data in the low flow rate regime ($Re < 300$) shown in Fig. 2. The linear regression resulted in Eq. (4).

$$Sh = 2 + 0.569(Gr'Sc)^{0.25} + 0.56(Re^{0.62}Sc^{0.31}) \quad \text{for } Re < 300 \quad \dots(4)$$

The enhancement of mass transport at low Reynolds number was observed in the results of previous investigators^{14,15)} of decarburisation of Fe-C melts using similar geometries. They also obtained higher values (>0.5) of the forced convection coefficient ' n ' when compared with that (0.347) in Eq. (2),

Although Eq. (4) is limited in its applicability to the system geometry of the present study, the present work clearly shows that using a correlation equation in conditions which are significantly different to those in which it was developed must be undertaken with caution, and hence the equation must be validated in the conditions where it is employed. The fact that the gas phase mass transfer is very sensitive to the system geometry may be beneficially used for identifying the influence of the gas phase transport to the overall reaction kinetics of a gas-metal reaction. For example, if the mass transfer in the gas phase is involved in the rate-controlling step, the overall rate will follow the trend similar to what is shown in Fig. 2.

In the present analysis, however, the mutual interaction of free and forced convections that may exist in the presence of simultaneous transfers of large heat and mass is not considered.

4. Conclusions

The present study has shown that the mass transfer in the gas phase, by the forced convection in particular, is very sensitive to the system geometry. For the geometry of the present electromagnetic levitation system

having the induction coil around the spherical sample, the correlation equation of Steinberger and Treybal was found to be adequate to describe the gas phase mass transfer only if the Reynolds number is large ($Re > 600$), but significantly underestimate the gas phase mass transfer coefficient for low Reynolds numbers ($Re < 600$). The results of this work emphasise the need to validate the application of a correlation equation when applied to a particular geometric system. The fact that the gas phase mass transfer is sensitive to the system geometry may be beneficially used for identifying the influence of the gas phase transport to the overall reaction kinetics of a gas-metal reaction.

Nomenclature

- A : surface area of sample (m^2)
 W : rate of mass loss ($kg\ s^{-1}$)
 P_s : vapour pressure of naphthalene in atmosphere
 R : universal gas constant ($m^3\ atm\ mol^{-1}\ K^{-1}$)
 T : temperature (K)
 T_f : film temperature $\left(= \frac{T_a + T_s}{2} \right)$ (K)
 T_b : bulk gas temperature (K)
 T_i : melt surface temperature (K)
 d : diameter of sample (m)
 k_g : gas mass transfer coefficient ($m\ s^{-1}$)
 Sh : Sherwood number $\left(= \frac{k_g d}{D} \right)$
 Gr' : Mean Grashof number $\left(= Gr_m + \left(\frac{Sc}{Pr} \right)^{0.5} Gr_h \right)$
 Sc : Schmidt number $\left(= \frac{\mu_f}{\rho_f D} \right)$
 Re : Reynolds number $\left(= \frac{\rho_f d v}{\mu_f} \right)$
 Gr_m : Grashof number for mass transfer
 $\left(= \frac{\rho_f g d^3 (\rho_i - \rho_b)}{\mu_f^2} \right)$

Gr_h : Grashof number for heat transfer

$$\left(= \frac{\rho_f^2 g d^3 (\rho_i - \rho_b)}{\mu_f^2 T_f} \right)$$

Pr : Prandtl number $\left(= \frac{\mu_f C_p}{\kappa} \right)$

D : interdiffusivity in the gas mixture ($m^2\ s^{-1}$)

μ : viscosity of gas ($kg\ m^{-1}\ s^{-1}$)

ρ : density of gas ($kg\ m^{-3}$)

v : velocity of gas ($m\ s^{-1}$)

C_p : heat capacity of gas ($J\ kg^{-1}\ K^{-1}$)

κ : thermal conductivity of gas ($J\ m^{-1}\ s^{-1}\ K^{-1}$)

g : acceleration due to gravity ($m\ s^{-2}$)

Subscript

f: property at the film temperature

b: property at the bulk gas temperature

i: property at the melt surface temperature

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