

# Interphase Mass Transfer with Bulk Flow Normal to the Phase Boundary

Gilsoo Han\* and Hong Yong Sohn\*\*\*,†

\*Departments of Metallurgical Engineering and of \*\*Chemical and Fuels Engineering,  
University of Utah, Salt Lake City, Utah 84112, U. S. A.

(Received 21 June 2003 • accepted 11 November 2003)

**Abstract**—The effect of bulk flow normal to the interface on the interphase mass transfer rate is analyzed by using a convective mass transfer coefficient, and a method of deriving the correct choice of the reference mole fraction in the bulk flow term is developed. The effect of the bulk flow and the definition of the proper reference mole fraction have been derived based on diffusion in a binary gas mixture across the boundary layer and expanded to the system of an interfacial gas-solid reaction. The results show that the bulk flow effect together with the proper choice of the reference mole fraction in the bulk flow term is important in obtaining an accurate expression of interphase mass transfer rate. In an extreme situation, the bulk flow effect can cause mass transfer to occur in the direction of increasing concentration. The theoretical development is applied to the rate analysis of the hydrogen reduction of silica.

Key words: Boundary Layer, Bulk Flow Effect, Diffusion, Interphase, Mass Transfer, Volume Change

## INTRODUCTION

Certain mass transfer processes taking place at phase boundaries between fluids and condensed phases accompany a bulk flow perpendicular to the interface. Although important in many mass transfer operations such as evaporation, absorption, membrane filtration, drying, and heterogeneous chemical reactions, the effect of such a bulk flow component is usually not considered in the analysis of the mass transfer rate. The only textbook that treats this effect at any length is *Transport Phenomena* by Bird et al. [1960, 2002a]. It is the purpose of this paper to examine this effect by using a more fundamental approach and develop an unambiguous method of rate analysis for such a system.

An example of the processes to which the mass transfer analysis is applied in this paper is gas-solid reactions, which are important in metal extraction and material processing operations. These reactions occur at phase boundaries, and thus the gaseous reactant must be transferred from the bulk gas phase to the reaction interface and the gaseous products away from the interface to the bulk. Therefore, in addition to chemical reaction, mass transport usually plays an important role in determining the overall reaction rate [Szekely et al., 1976]. The effect of bulk flow normal to the interface due to the volume change of gas should be considered when the total number of moles of gaseous products is different from that of the gaseous reactants. Despite the significant effect of such a bulk flow, most previous work has neglected this effect. An investigation has been performed to determine the effect of bulk flow normal to the interface on the rate of a gas-solid reaction that is controlled by the convective mass transfer process together with the rigorous expression of the effect of bulk flow when using convective mass transfer coefficient.

## CONVECTIVE MASS TRANSFER WITH BULK FLOW NORMAL TO THE INTERFACE

Diffusion in a fluid mixture is described by the following Maxwell-Stefan equation [Bird et al., 2002a]:

$$\nabla x_i = \sum_{j=1}^n \frac{1}{C_T D_{ij}} (x_i N_j - x_j N_i) \quad (1)$$

where  $D_{ij}$  is the diffusivity of the pair i-j in a binary mixture which is nearly independent of composition, and  $C_T$  is total molar concentration of the solution. For certain systems it is convenient to use an equation analogous to Fick's law for a binary mixture, as follows [Bird et al., 1960]:

$$N_i = -C_T D_{im} \nabla x_i + x_i \sum_{j=1}^n N_j \quad (2)$$

where  $D_{im}$  is an effective diffusivity of species i in the mixture. The first term on the right side represents the molar flux of i due to the molecular flux or diffusion and the second term is the molar flux of i caused by the convective flux or bulk flow normal to the interface.

Interphase mass transfer across a laminar boundary layer is usually described by defining a mass transfer coefficient,  $h_D$ , according to the following equation, with the positive direction of  $N_i$  defined to be from the interface toward the bulk stream [Bird et al., 2002b; Sohn and Sohn, 1980]:

$$N_i = h_D C_T (x_{is} - x_{ib}) + \tilde{x}_i \sum_{j=1}^n N_j \quad (3)$$

where  $\tilde{x}_i$  is a reference mole fraction (yet to be determined). This is the mass transfer equivalent to the expression of convective heat transfer between a solid surface and a bulk fluid. Such a mass convection expression is used when the solution using Eq. (1) combined with a proper species conservation equation for a given situation is highly complex either because the system geometry and flow configuration are complex or the flow is turbulent. The last

\*To whom correspondence should be addressed.

Email: hysohn@mines.utah.edu

†This paper is dedicated to Professor Hyun-Ku Rhee on the occasion of his retirement from Seoul National University.

terms in these equations account for the effect of bulk flow in the direction of mass transfer, i.e., perpendicular to the interface.

In Fick's law, given in Eq. (2),  $x_i$  is a function of position that is determined when this equation for species flux is incorporated into the species balance equation for a specific system. In the interphase mass transfer expression of Eq. (3), the flux is expressed in terms of the interfacial and bulk concentrations and thus  $\bar{x}_i$  must also have a fixed value, which must lie somewhere between  $x_{is}$  and  $x_{ib}$ . The proper choice of  $\bar{x}_i$  is not clearly defined in the literature. Typically,  $x_{is}$  or  $x_{ib}$  is used for this term [Bird et al., 2002b; Sohn and Sohn, 1980]. This is not appropriate because the effect of the bulk flow term is affected by the mole fraction of the diffusing species, which in general varies considerably between the interface and the bulk. Furthermore, confusion can also arise when the same system is described in terms of one or the other species. In the following, we will develop a procedure for determining the proper value of  $\bar{x}_i$ .

Let us consider interphase mass transfer in a binary mixture of gases A and C, as illustrated in Fig. 1. In the following, the boundary layer thickness will be assumed to be unaffected by the bulk flow normal to the interface, which usually is much smaller than the external flow parallel to the interface. The application of Eq. (2) to the gas A yields

$$N_A = -C_T D_{AC} \frac{dx_A}{dz} + x_A (N_A + N_C) \quad (4)$$

Let us define the ratio of molar fluxes as follows:

$$-\frac{N_C}{N_A} = v \quad (5)$$

where the sign of  $v$  shows the relative direction of the fluxes: A positive value means the directions are opposite to each other, and vice versa. Substituting Eq. (5) in Eq. (4) and rearranging, we get

$$N_A = \frac{-C_T D_{AC}}{1+x_A(v-1)} \frac{dx_A}{dz} \quad (6)$$

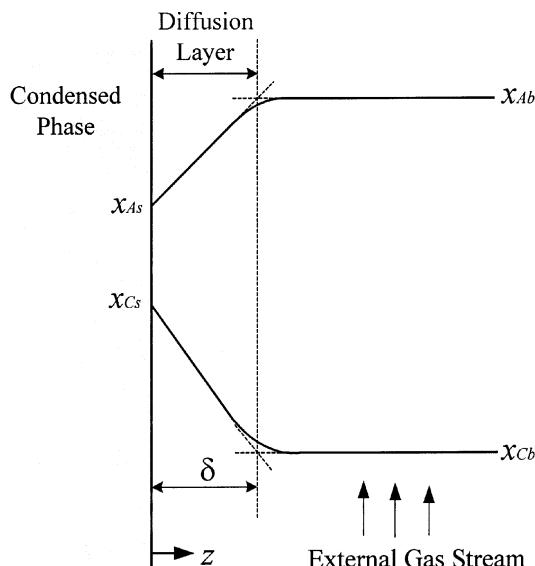


Fig. 1. Concentration profiles for a binary gas mixture near a gas-condensed phase interface.

The mass balance within the diffusion boundary layer over a condensed phase is given by the following, assuming pseudo-steady state:

$$\frac{dN_A}{dz} = 0 \quad (7)$$

with the boundary conditions:

$$\text{at } z=0, x_A = x_{As} \quad (8)$$

$$\text{at } z=\delta, x_A = x_{Ab} \quad (9)$$

Substituting Eq. (6) into Eq. (7) and integrating twice with respect to  $z$ , and applying the boundary conditions, we get

$$N_A = \frac{C_T D_{AC}}{\delta} \frac{1}{v-1} \ln \frac{1+(v-1)x_{As}}{1+(v-1)x_{Ab}} \quad (10)$$

Eq. (10), as well as similar equations appearing in the subsequent parts of this paper, contains the special case of  $v=1$ , which can be evaluated by the application of L'Hospital's rule. In the case of Eq. (10), this yields the following familiar result:

$$N_A = \frac{C_T D_{AC}}{\delta} (x_{As} - x_{Ab}) \quad (10a)$$

The molar flux of A can also be described by Eq. (3) for interphase mass transfer:

$$N_A = h_D C_T (x_{As} - x_{Ab}) + \bar{x}_A (N_A + N_C) \quad (11)$$

Applying Eqs. (10) and (11) to the case of equimolar counter diffusion, the mass transfer coefficient is determined to be equivalent to

$$h_D = \frac{D_{AC}}{\delta} \quad (12)$$

Substituting Eq. (12) into Eq. (11) and comparing Eqs. (10) and (11), we obtain after some rearrangement

$$1+(v-1)\bar{x}_A = \frac{[1+(v-1)x_{As}] - [1+(v-1)x_{Ab}]}{\ln \frac{1+(v-1)x_{As}}{1+(v-1)x_{Ab}}} \quad (13)$$

and

$$N_A = \frac{h_D C_T}{v-1} \ln \frac{1+(v-1)x_{As}}{1+(v-1)x_{Ab}} \quad (14)$$

The reference mole fraction and the molar flux can be obtained in terms of gas C through the same procedure:

$$1 - \left( \frac{v-1}{v} \right) \bar{x}_C = \frac{\left[ 1 - \frac{v-1}{v} x_{Cs} \right] - \left[ 1 - \frac{v-1}{v} x_{Cb} \right]}{\ln \frac{1 - \frac{v-1}{v} x_{Cs}}{1 - \frac{v-1}{v} x_{Cb}}} \quad (15)$$

and

$$N_C = -\frac{h_D C_T v}{v-1} \ln \frac{v - (v-1)x_{Cs}}{v - (v-1)x_{Cb}} \quad (16)$$

An expression equivalent to Eq. (14) was derived in Bird et al. [2002b] for the case of a high net mass transfer rate across the phase

boundary. The results derived here indicate, however, that these expressions are applicable regardless of the net rate of mass transfer. As can be seen in Eqs. (13) and (15),  $\tilde{x}_i$  and thus expression of  $N_i$  depends on the ratio of mole fluxes. If  $x_{is}$  and  $x_{ib}$  are given, the molar fluxes can be obtained from Eqs. (14) and (16), and the definition of  $\tilde{x}_i$  from Eqs. (13) and (15). However, in most cases including gas-solid reactions,  $v$  and bulk concentrations are fixed rather than all the values of surface concentrations.

### APPLICATION TO GAS-SOLID REACTIONS

For a simple gas-solid reaction of the following form



the above results for a binary gas mixture are directly applicable. For this system,  $v$  is given by the reaction stoichiometry  $c$  and  $x_{Ab}$  is fixed for a given condition.  $x_{As}$  is then obtained from the equilibrium condition, if the interfacial reaction is fast, or in combination with reaction kinetics.

Fig. 2 shows the effect of bulk flow normal to the interface due to a gas volume change upon reaction on the flux in a binary gas mixture for the case of  $x_{Ab}=0.9$ , as an example. The effect of bulk flow depends on the  $x_{As}$  and  $x_{Ab}$ , and the value of  $v$ . When the value of  $v$  is unity, i.e., when there is no change in the gas volume, there is no bulk flow effect. However, when the value of  $v$  is different from unity, the change in the gas volume upon reaction causes a bulk flow effect, with the deviation from the equimolar case increasing as the ratio of  $x_{As}$  to  $x_{Ab}$  increases. In addition to affecting the magnitude of flux, the bulk flow effect can even change the direction of mass transfer in the case of  $v<0$ , that is, the overall flux of a gaseous component can be in the direction of increasing mole fraction due to the bulk flow in that direction. This rather unusual and interesting situation may not be encountered too often in a real system, but it is a possibility when the binary system involves just two

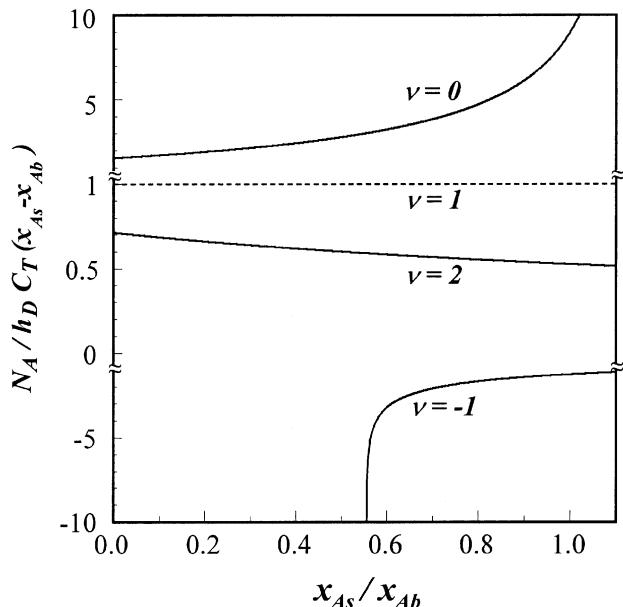


Fig. 2. Effect of bulk flow normal to the interface on the molar flux of A in a binary gas mixture with  $x_{Ab}=0.9$ .

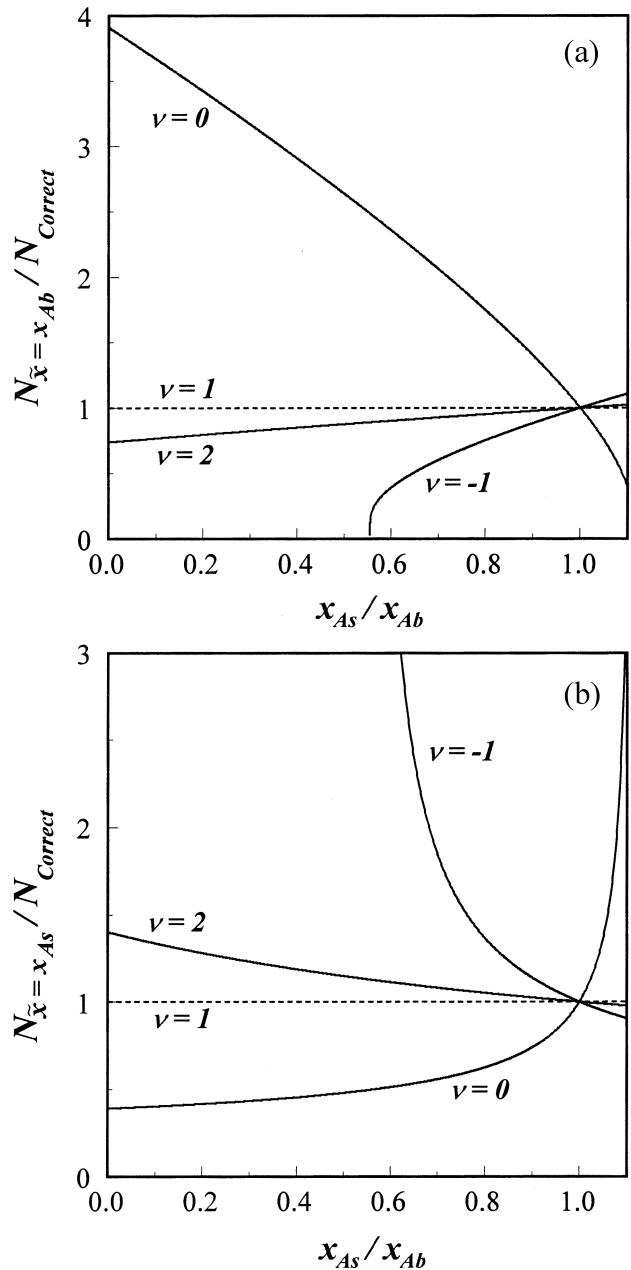


Fig. 3. Effect of the choice of reference mole fraction in Eq. (3) on the molar flux of A in a binary gas mixture with  $x_{Ab}=0.9$ : (a)  $N_{x=x_{Ab}} / N_{Correct}$ ; (b)  $N_{x=x_{As}} / N_{Correct}$ .

gaseous reactants or just two gaseous products, or when a certain gaseous component is selectively absorbed into a condensed phase from a binary gas mixture.

Fig. 3 illustrates an example of the effect of the choice of  $\tilde{x}_i$  on the flux in a binary gas mixture with  $x_{Ab}=0.9$ . As mentioned earlier,  $x_{ib}$  or  $x_{is}$  is usually used for  $\tilde{x}_i$ . However, as can be seen in Eqs. (13) and (15),  $\tilde{x}_i$  is in general different from  $x_{ib}$  or  $x_{is}$ . Hence, the correct flux of gas A is compared with the fluxes calculated for  $\tilde{x}_A=x_{Ab}$  or  $\tilde{x}_A=x_{As}$  in this figure. The effect of the choice of  $\tilde{x}_i$  varies with the ratio between  $x_{As}$  and  $x_{Ab}$  and the value of  $v$ . As the difference between  $x_{As}$  and  $x_{Ab}$  increases, the importance of this effect increases, as expected.

## REDUCTION OF FUSED SILICA IN HYDROGEN

The reduction of silica in a hydrogen environment, forming silicon monoxide gas and water vapor, is important in many industries because the volatilization of silica can cause serious problems such as heat-exchanger fouling and product contamination as well as deterioration of the properties of high-temperature ceramic materials [Gardner, 1974; Lewis, 1979; Schwerdtfeger, 1966; Szekely et al., 1976; Tso and Park, 1982]. The reduction reaction between fused silica and hydrogen gas is represented by



This reaction forms no coherent solid product layer, and generates a net increase in gas volume, necessitating the consideration of the bulk flow effect. This reaction can be expressed by the following general equation:



Incorporating the assumption of pseudo-steady state and unchanging boundary layer thickness, the following is derived for the cases in which  $D_{Cm} = D_{Qm}$  and the diffusivities are independent of concentration (e.g. when  $x_C, x_Q \ll x_A$  and  $D_{CA} = D_{QA}$ ). The same procedure as in the binary gas mixture yields the reference mole fraction and the molar flux for reactant gas A, as follows:

$$1 + (c + q - 1)\bar{x}_A = \frac{[1 + (c + q - 1)x_{As}] - [1 + (c + q - 1)x_{Ab}]}{\ln \frac{1 + (c + q - 1)x_{As}}{1 + (c + q - 1)x_{Ab}}} \quad (20)$$

and

$$N_A = \frac{h_D C_T}{c + q - 1} \ln \frac{1 + (c + q - 1)x_{As}}{1 + (c + q - 1)x_{Ab}} \quad (21)$$

The molar flux and the reference mole fraction of products C and Q can be obtained through the same procedure as for A:

$$1 - \frac{c + q - 1}{c} \bar{x}_C = \frac{\left[1 - \frac{c + q - 1}{c} x_{Cs}\right] - \left[1 - \frac{c + q - 1}{c} x_{Cb}\right]}{\ln \frac{1 - \frac{c + q - 1}{c} x_{Cs}}{1 - \frac{c + q - 1}{c} x_{Cb}}} \quad (22)$$

$$N_C = \frac{h_D C_T}{-(c + q - 1)/c} \ln \frac{1 + \frac{(c + q - 1)}{c} x_{Cs}}{1 + \frac{(c + q - 1)}{c} x_{Cb}} \quad (23)$$

$$1 - \frac{c + q - 1}{q} \bar{x}_Q = \frac{\left[1 - \frac{c + q - 1}{q} x_{Qs}\right] - \left[1 - \frac{c + q - 1}{q} x_{Qb}\right]}{\ln \frac{1 - \frac{c + q - 1}{q} x_{Qs}}{1 - \frac{c + q - 1}{q} x_{Qb}}} \quad (24)$$

and

$$N_D = \frac{h_D C_T}{-(c + q - 1)/q} \ln \frac{1 + \frac{(c + q - 1)}{q} x_{Qs}}{1 + \frac{(c + q - 1)}{q} x_{Qb}} \quad (25)$$

The above expressions can be generalized for any multicomponent systems in which all diffusivities are equal, by defining

$$Y_i = 1 - \frac{\sum \nu_j}{\nu_i} x_i \quad (26)$$

and

$$\bar{Y}_i = L(Y_{is}, Y_{ib}) \quad (27)$$

Here,  $L(Y_{is}, Y_{ib})$  represents the logarithmic mean of  $Y_{is}$  and  $Y_{ib}$ , and  $\nu_i$  is the stoichiometry coefficient of gaseous species i (negative for gaseous reactants and positive for gaseous products). Then,

$$\bar{x}_i = \frac{\nu_i}{\sum \nu_j} + \frac{x_{is} - x_{ib}}{\ln \frac{\nu_i - (\sum \nu_j)x_{is}}{\nu_i - (\sum \nu_j)x_{ib}}} \quad (28)$$

and

$$N_i = \frac{h_D C_T (x_{is} - x_{ib})}{\bar{Y}_i} = - \frac{h_D C_T \nu_i}{\sum \nu_j} \ln \frac{\nu_i - (\sum \nu_j)x_{is}}{\nu_i - (\sum \nu_j)x_{ib}} \quad (29)$$

Eq. (28) represents the proper definition of  $\bar{x}_i$  to be used with Eq. (3) for the correct description of the interphase mass transfer rate in a multicomponent system in which the assumptions described earlier are applicable.

The experimental data obtained by Lewis [1979] at the University of Utah will be analyzed based on the theoretical derivation described above. The experiment was conducted by measuring the weight change of silica using a thermogravimetric apparatus at three temperatures, 1,285 °C, 1,386 °C, and 1,450 °C. It consisted of a microbalance, a kanthal-wound furnace, an alumina reactor tube and a gas train for purification. The hydrogen gas was passed through Dri-Rite ( $\text{CaSO}_4$ ) and Anhydrene [ $\text{Mg}(\text{ClO}_4)_2$ ] before entry into the furnace. The bottom of the tube was filled with alumina chips to preheat the incoming gas. The sample was suspended from the balance by a molybdenum wire in the alumina tube. Pure, natural fused quartz (99.99%  $\text{SiO}_2$ ) was obtained from the General Electric Company in the form of tubing (6 mm i.d.  $\times$  8 mm o.d.). The samples were sectioned to be 5 mm in length. Prior to experiment, the samples were cleaned with soap and water, and rinsed with a 7% HF solution. The values of the parameters used to calculate and analyze the reaction rate are summarized in Table 1.

**Table 1. Parameters used in mass transfer calculation for the hydrogen reduction of silica**

Temperature [K]	1558	1659	1723
$D = D_{H_2O-H_2}$ [cm <sup>2</sup> /s]	18.434	20.576	21.958
$\mu/\rho = \mu_{H_2}/\rho_{H_2}$ [cm <sup>2</sup> /s]	19.387	21.485	22.864
$K_{eq}^*$	$8.056 \times 10^{-9}$	$1.004 \times 10^{-7}$	$4.239 \times 10^{-7}$
$C_T$ [mol/cm <sup>3</sup> ]	$6.7267 \times 10^{-6}$	$6.3171 \times 10^{-6}$	$6.0825 \times 10^{-6}$
d [cm]	0.5		
$P_T$ [atm]**	0.85		
$x_{H_2O, b}, x_{SiO, b}$	0		
$x_{H_2, b}$	1		

\*Data from HSC Chemistry [1999].

\*\*Atmospheric pressure in Salt Lake City, Utah, U.S.A.

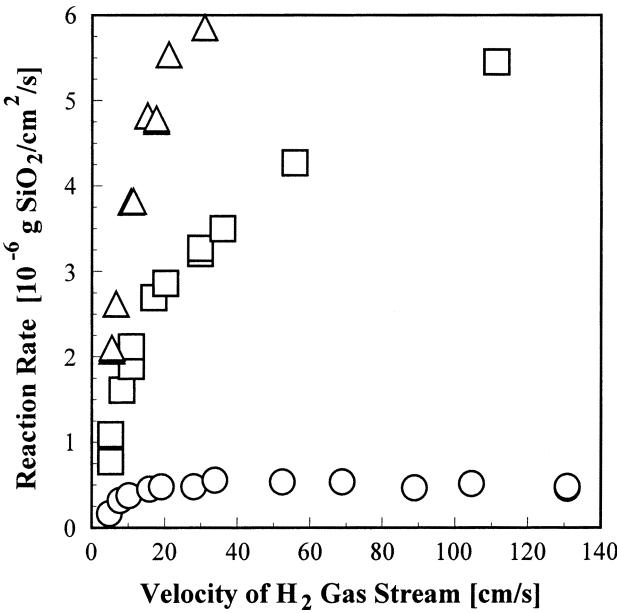


Fig. 4. Reaction rate vs. velocity of hydrogen gas stream at different temperatures:  $\circ$  1,285 °C;  $\square$  1,386 °C;  $\triangle$  1,450 °C.

Fig. 4 shows the reaction rate of  $\text{SiO}_2$  as a function of the flow rate of hydrogen gas at three different temperatures. As can be seen, the mass flux increased with the velocity of the hydrogen gas stream, which indicates that the rate of this reaction is affected by the mass transport in the boundary layer. However, at the lowest temperature investigated, 1,285 °C, the rate of reaction per unit surface area is not affected by the velocity of gas stream except at very low flow rates. This flow-rate-independent condition is expected when the overall rate is controlled by chemical reaction at the surface.

As shown in Eq. (18), it is necessary to consider the bulk flow effect in the mass flux in a boundary layer. In this case, the theoretical value of the interdiffusivity of the pair  $\text{H}_2\text{-SiO}$  is almost the same as that of  $\text{H}_2\text{-H}_2\text{O}$ , and the mole fractions of  $\text{H}_2\text{O}$  and  $\text{SiO}$  are much smaller than that of  $\text{H}_2$  due to the small equilibrium constant for the reaction. Thus, if the rate of reaction is controlled by mass transport in the boundary layer, Eqs. (20) to (25) can be used to describe the reaction rate. Thus, Eq. (21) yields

$$N_{H_2} = h_D C_T \ln \frac{1+x_{H_2,s}}{1+x_{H_2,b}} \quad (30)$$

which for  $x_{H_2,b}=1$  reduces to

$$N_{H_2} = C_T h_D \ln \frac{1+x_{H_2,s}}{2} \quad (31)$$

Since the mole fraction of hydrogen at the reaction surface is nearly equal to that in the bulk because of the extremely small equilibrium constant in the temperature range tested, expansion of the logarithmic term in Eq. (31) or rearrangement of Eq. (11) yields

$$N_{H_2} \approx \frac{1}{2} h_D C_T (x_{H_2,s} - 1) \quad (32)$$

in which the bulk flow effect appears in the factor 1/2, that is, the correct molar flux of the reactant is half that without considering the bulk flow effect.

The average mass-transfer coefficient over the reacting surface is obtained from the following equation [Benitez, 2002] that is valid for a flat plate with laminar flow, because the Reynolds number for the conditions in this experiment is lower than 4:

$$Sh = 0.664 Re^{1/2} Sc^{1/3} \quad \text{for } Re < 3 \times 10^5 \quad (33)$$

Substituting the definitions of the Sherwood, the Reynolds, and the Schmidt numbers, which are given in Nomenclature, we obtain

$$h_D = 0.664 \cdot D^{2/3} (\mu/\rho)^{-1/6} (V/d)^{1/2} \quad (34)$$

Substituting Eq. (34) in Eq. (32), we get

$$N_{H_2} = 0.332 \cdot D^{2/3} (\mu/\rho)^{-1/6} (V/d)^{1/2} C_T (x_{H_2,s} - 1) \quad (35)$$

Here,  $D$  is the hydrogen diffusivity in a mixture with  $\text{H}_2\text{O}$  gas. Since  $D_{H_2\text{-}H_2\text{O}}$  and  $D_{H_2\text{-}SiO}$  are almost equal, the calculated value of  $D_{H_2\text{-}H_2\text{O}}$  was used in this work.

The reaction rate can also be expressed from the viewpoint of the product  $\text{H}_2\text{O(g)}$  through the same procedure as for the reactant  $\text{H}_2\text{(g)}$ , which gives

$$N_{H_2\text{O}} = -h_D C_T \ln \frac{1-x_{H_2\text{O},s}}{1-x_{H_2\text{O},b}} \quad (36)$$

which for  $x_{H_2\text{O},b}=0$  reduces to

$$N_{H_2\text{O}} = C_T h_D \ln \frac{1}{1-x_{H_2\text{O},s}} \quad (37)$$

Since the mole fraction of  $\text{H}_2\text{O(g)}$  at the reaction surface approaches the small value in the bulk (zero in this case) due to the extremely small equilibrium constant,

$$N_{H_2\text{O}} \approx C_T h_D x_{H_2\text{O},s} \quad (38)$$

in which the bulk flow effect does not appear, that is, the bulk flow effect can be ignored when mass transfer is described based on the flux of the species that exists at low concentrations.

Substituting Eq. (34) into Eq. (38), we get

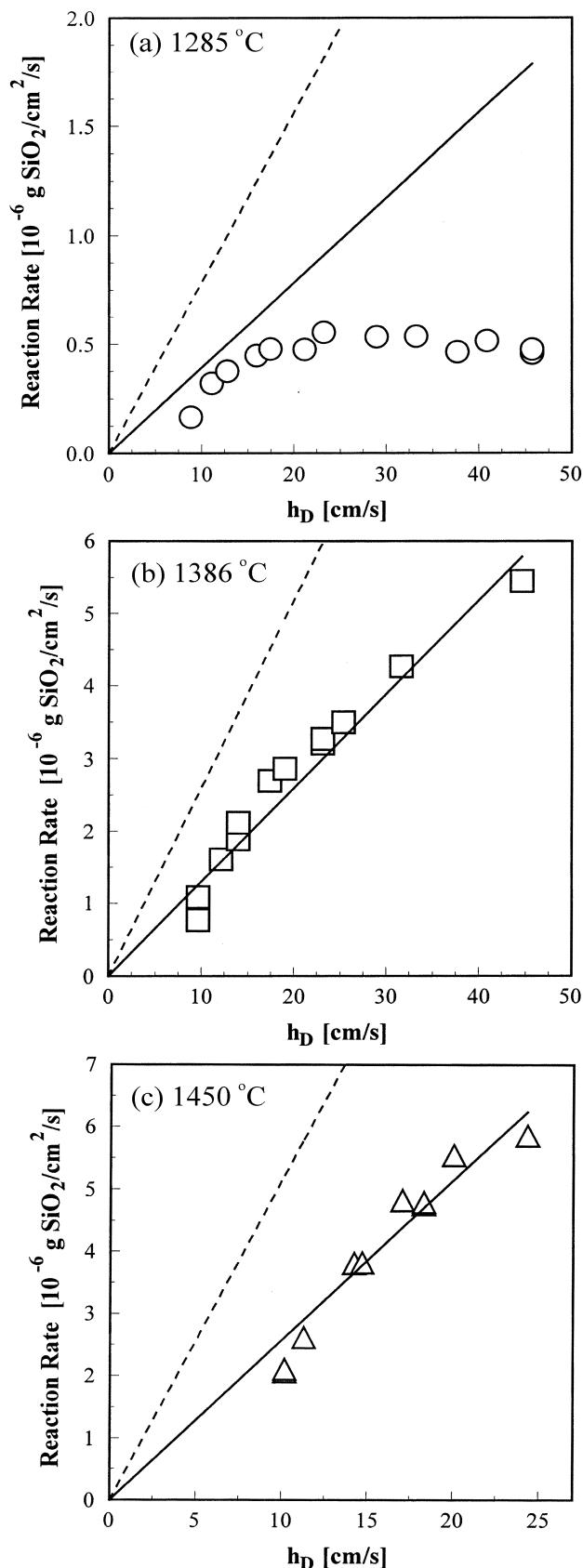
$$N_{H_2\text{O}} = 0.664 \cdot D^{2/3} (\mu/\rho)^{-1/6} (V/d)^{1/2} C_T x_{H_2\text{O},s} \quad (39)$$

The value of mole fraction of  $\text{H}_2\text{O(g)}$  on the reaction surface can be obtained by using the equilibrium constant:

$$K_{eq} = \left[ \frac{P_{SiO} \cdot P_{H_2\text{O}}}{a_{SiO} \cdot p_{H_2}} \right]_{eq} = \left[ P_T \frac{x_{H_2\text{O}}^2}{1-2 \cdot x_{H_2\text{O}}} \right]_{eq} \quad (40)$$

Here, we assumed ideal gas mixture, and  $x_{SiO}=x_{H_2\text{O}}$  based on the reaction stoichiometry and the same diffusivities of  $\text{SiO}$  gas and water vapor. Further in this equation, the partial pressures must be expressed in atm unit so that they become numerically equal to the activities (dimensionless). It is noted that the atmospheric pressure in Salt Lake City is 0.85 atm. The consumption rate of  $\text{SiO}_2$  can be obtained from  $N_{H_2}$  or  $N_{H_2\text{O}}$  by the stoichiometric relationship.

In Fig. 5, the experimental reaction rate is compared with the calculated reaction rate obtained from Eq. (35) or (39). At the low temperature of 1,285 °C, the calculated rate does not agree well with the experimental rate, which indicates that mass transfer does not control the overall rate, as mentioned earlier. At the high temperatures, the calculated results agree well with the experimental data, which indicates that the overall reaction rate is controlled by the convective mass transfer in the diffusion boundary layer. The cal-



**Fig. 5. Comparison of calculated and experimental rate of silica consumption: ○, □, and △ experimental data; — calculated results incorporating bulk flow effect; - - - calculated results without considering bulk flow effect.**

culated rates based on hydrogen transfer without considering the bulk flow effect are also shown in Fig. 5. The latter is seen to give a poor agreement with the experimental data. Thus, inclusion of the bulk flow effect is essential to obtain accurate results.

## CONCLUDING REMARKS

The effect of bulk flow normal to the interface on the rate of interphase mass transfer expressed in terms of a convective mass transfer coefficient has been examined. It has been shown that the effect of bulk flow and the choice of the reference mole fraction to be used to represent the bulk flow term are important for correctly describing interphase mass transfer rates.

The effects of bulk flow and choice of reference mole fraction depend on the difference between the mole fraction at the interface and that in the bulk gas stream as well as the ratio of molar fluxes. In an extreme situation, the bulk flow effect can cause mass transfer to occur in the direction of increasing concentration. As the ratio of the mole fraction at the surface to that in the bulk stream becomes far from unity, the choice of reference mole fraction becomes increasingly important.

The theoretical derivations have been applied to the reduction of silica in hydrogen. The calculated reduction rate that properly incorporates the bulk flow effect agrees well with the experimental data, while the rate calculated without considering the bulk flow effect shows a poor agreement with the experimental data. Thus, the incorporation of the bulk flow effect and the proper choice of the reference mole fraction are important for correctly describing mass transfer rates.

## ACKNOWLEDGMENTS

Hong Yong Sohn wishes to make a special acknowledgment and pay the highest tribute to his long time friend and college classmate, Professor Hyun-Ku Rhee, on his retirement from our Alma Mater Seoul National University. It has been a source of great pleasure and pride to follow his outstanding contributions as a teacher, scholar and professional leader throughout his long career. Although our careers have kept us apart across the Pacific, our friendship has remained strong and pure. Heart-felt congratulations on Professor Rhee's lifetime accomplishments and best wishes for a long and happy retirement!

Gilsoo Han wishes to express his gratitude for the receipt of a Cooper-Hansen Fellowship from the University of Utah during the course of this work.

## NOMENCLATURE

$a_i$	: activity of i
A	: fluid reactant
b	: stoichiometry coefficient of B
B	: solid reactant
c	: stoichiometry coefficient of C
C	: fluid product
$C_T$	: total molar concentration of gas
d	: length of plate
D	: interdiffusivity of gases

$D_{ij}$	: diffusivity of the pair i-j in a binary mixture
$D_{im}$	: effective diffusivity of species i in a multicomponent mixture
$h_D$	: mass-transfer coefficient
$K_{eq}$	: equilibrium constant
$L(Y_{is}, Y_{ib})$	: logarithmic mean of $Y_{is}$ and $Y_{ib}$
$N_i$	: molar flux of species i relative to stationary coordinate
$p_i$	: partial pressure of i
$P_T$	: total pressure of gas
$q$	: stoichiometry coefficient of Q
$Q$	: fluid product
$R$	: gas constant
$Re$	: Reynolds number = $(V \cdot d) / (\mu / \rho)$
$Sc$	: Schmidt number = $(\mu / \rho) / D$
$Sh$	: Sherwood number = $h_D \cdot d / D$
$T$	: absolute temperature
$V$	: bulk velocity of gas stream
$x_i$	: mole fraction of species i
$\bar{x}_i$	: reference mole fraction of species i defined in Eq. (3)

#### Greek Letters

$\mu$	: viscosity
$\nu$	: ratio of molar fluxes in a binary gas mixture defined by Eq. (5) (positive when the fluxes are in the opposite directions)
$\nu_i$	: stoichiometry coefficient of gaseous species i (positive for product gases and negative for reactant gases)
$\rho$	: fluid mass density

#### Subscripts

A, C, Q : fluid species A, C, and Q, respectively

i	: species i
b	: value in bulk stream
s	: value at the interface

#### REFERENCES

Benitez, J., "Principles and Modern Applications of Mass Transfer Operations," John Wiley & Sons, Inc., New York, 85 (2002).

Bird, R. B., Stewart, W. E. and Lightfoot, E. N., "Transport Phenomena," 1<sup>st</sup> ed., John Wiley & Sons, New York, 563 (1960).

Bird, R. B., Stewart, W. E. and Lightfoot, E. N., "Transport Phenomena," 2<sup>nd</sup> ed., John Wiley & Sons, Inc., New York, 536 (2002a).

Bird, R. B., Stewart, W. E. and Lightfoot, E. N., "Transport Phenomena," 2<sup>nd</sup> ed., John Wiley & Sons, Inc., New York, 671 (2002b).

Gardner, R. A., "The Kinetics of Silica Reduction in Hydrogen," *J. Solid State Chem.*, **9**, 336 (1974).

HSC Chemistry, Version 4.0, Outokumpu Research Oy, Finland (1999).

Lewis, S. D., "Reduction of Fused Silica in Hydrogen," Thesis, University of Utah (1979).

Schwerdtfeger, K., "The Rate of Silica Reduction in Reducing Gases at 1,500 °C," *Trans. Met. Soc. AIME*, **236**, 1152 (1966).

Sohn, H. Y. and Sohn, H.-J., "The Effect of Bulk Flow Due to Volume Change in the Gas Phase on Gas-Solid Reaction: Initially Nonporous Solids," *Ind. Eng. Chem. Process Des. Dev.*, **19**, 237 (1980).

Szekely, J., Evans, J. W. and Sohn, H. Y., "Gas-Solid Reactions," Academic Press, New York, 66 (1976).

Tso, S. T. and Park, J. A., "Reaction of Fused Silica with Hydrogen Gas," *J. Am. Ceram. Soc.*, **65**, 457 (1982).