Particle–gas mass transfer under plasma conditions

SHRIKANT V. JOSHI,* JIN Y. PARK,*† P. R. TAYLOR[‡] and L. S. RICHARDSON[§]

 Department of Chemical Engineering, [†] Department of Metallurgical and Mining Engineering, University of Idaho, Moscow, ID 83843, U.S.A.
 § Materials Science Division, EG&G Idaho, Inc., Idaho Falls, ID 83415, U.S.A.

(Received 25 November 1985 and in final form 19 May 1986)

Abstract—A simple analytical method is proposed for particle–gas mass transfer calculations under plasma conditions. This method, called the Z-potential method, fully accounts for the temperature variation of the gas transport properties while permitting use of the convenient isothermal expression Sh = 2.0. The Z-potential approach is also found to be extremely useful in ascertaining the Knudsen discontinuum effect on mass transfer between the plasma gas and a small particle. A predictive model for the Knudsen effect, developed by combining the Z-potential approach with the jump theory, is illustrated in detail by means of a case study on a nitrogen plasma system.

1. INTRODUCTION

IN RECENT years, the unique advantages offered by thermal plasmas have led to a growing interest in their use for particle processing. Various plasma-particle operations involving physical transformations have been listed by Fauchais and Baronnet [1]. The feasibility of several heterogeneous gas-solid reactions involving the injection of solid particles into a reactive plasma has also been established [2-6]. However, a thorough understanding of the transport processes under plasma conditions is still lacking. Plasmaparticle mass transfer has been a particularly neglected area of research in the past. Few predictive models are presently available to account for the steep temperature gradients typically encountered in plasma-particle systems. The noncontinuum effects, which could be significant when the particle in question is very small in size, have also yet to be investigated. These two important problems are addressed in this paper in order to facilitate more accurate plasma-particle mass transfer calculations.

In plasma-particle systems, the difference between the surface temperature of the particle and the bulk plasma temperature can be as high as 10,000 K. Due to the resulting steep temperature gradients, the plasma transport properties can vary considerably in the vicinity of the particle. This can have a significant effect on the heat and mass transfer rates. The choice of a single temperature for evaluating the transport properties of the plasma gas is difficult in such cases. For moderate temperature differences, the arithmetic mean temperature is often used [7,8]; however, this method is unsatisfactory under plasma conditions where dissociation and/or ionization could lead to peaks in the plasma property-temperature relationships [9]. Therefore, a reliable method of determining the representative value of a plasma property over the temperature range in question is necessary.

The plasma-particle heat transfer process has been quite extensively studied in the past. Bourdin *et al.* [9] have shown that the integral mean thermal conductivity of the gas, $\overline{\lambda}$, should be used to evaluate the heat transfer coefficient, *h*, when steep temperature gradients exist. When the convective effects are negligible, as in the case of a small particle entrained in a plasma stream, the use of $\overline{\lambda}$ leads to the well-known Ranz-Marshall expression for moderate temperature conditions [10], $Nu = hd/\overline{\lambda} = 2.0$. The $\overline{\lambda}$, defined specifically to account for the temperature variation of the thermal conductivity, is given by

$$\bar{\lambda} = \frac{I(T_{\infty}) - I(T_{\rm s})}{T_{\infty} - T_{\rm s}} \,. \tag{1}$$

In the above equation T_{∞} and T_s are the temperatures of the bulk plasma and the particle surface, respectively, and I(T) is the heat conduction potential defined as

$$I(T) = \int_{T_{\rm r}}^{T} \lambda(T) \,\mathrm{d}T \tag{2}$$

where λ is the thermal conductivity of the gas and T_r is a reference temperature. For a given gas, the heat conduction potential is a function of temperature only and may be tabulated as any other thermophysical property of the gas [9, 11]. Apart from accounting for the nonisothermal effects, the heat conduction potential method has also been found to be extremely useful in predicting the influence of the noncontinuum effects on plasma-particle heat transfer [12, 13].

While the above research efforts have significantly

[†]To whom correspondence should be addressed.

NOMENCLATURE			
С	concentration [kmol m^{-3}]	\bar{T}	arithmetic mean temperature [K]
d	particle diameter [m]	v	mean molecular speed $[m s^{-1}]$
D	molecular diffusivity $[m^2 s^{-1}]$	Y	mole fraction
D	integral mean diffusivity $[m^2 s^{-1}]$	Ζ	Z-potential $[J \text{ kmol}^{-1}]$.
h H I k Kn Kn I M	heat transfer coefficient $[W m^{-2} K^{-1}]$ specific enthalpy $[J kg^{-1}]$ heat conduction potential $[W m^{-1}]$ mass transfer coefficient $[m s^{-1}]$ Knudsen number effective Knudsen number mean free path of gas molecules $[m]$ molecular weight of the gas	Greek symbols γ specific heat ratio, C_p/C_v λ thermal conductivity [W m ⁻¹ K ⁻¹] $\overline{\lambda}$ integral mean thermal conductivity [W m ⁻¹ K ⁻¹] μ gas viscosity [kg m ⁻¹ s ⁻¹] ρ gas density [kg m ⁻³] θ thermal accommodation coefficient	
N Du	mass flux [kmol m ⁻² s ⁻¹] Brondtl number	$\theta_{\rm m}$	surface condensation coefficient.
r Q	heat flux $[W m^{-2}]$	Subscript	scripts
r	radial position [m]	0	in the gase phase in the immediate
R	gas constant $[J \text{ kmol}^{-1} \text{ K}^{-1}]$		particle vicinity
Sc	Schmidt number	с	using the continuum approach
Sc*	modified Schmidt number	s	at the particle surface
Т	temperature [K]	∞	in the bulk of the plasma gas.

reduced the difficulty of plasma-particle heat transfer calculations, little attention has been devoted so far to mass transfer in plasma-particle systems. Here we present a simple method, called the Z-potential method, recently developed by the authors [14] to account for the steep temperature gradients. The Zpotential approach then is combined with the jump theory to predict the extent of the Knudsen effect on plasma-particle mass transfer when the injected particle is small enough to cause discontinuities in velocity, temperature and composition at the particle surface. For illustrative purposes, results of calculations on particles of various sizes immersed in a nitrogen plasma are also presented. The analysis focuses exclusively on self-diffusion but the method is equally applicable to certain cases of equimolar counterdiffusion and for small concentrations of the diffusing species. The extension of this method to more complicated systems is currently under investigation [15].

The present study was primarily conducted to investigate the effect of steep temperature gradients typically encountered in thermal plasma processing of fine particles and to investigate the influence of the noncontinuum effects on plasma-particle mass transfer. The results of this study will enable a more realistic interpretation of the conversion data obtained from heterogeneous gas-solid reactions in a reactive thermal plasma. In addition, the calculation methods presented in this paper will also find application in various other high temperature, nonplasma processes, such as pulverized coal combustion [8], which involve small solid particles in contact with a gaseous phase at a significantly different temperature.

2. EFFECT OF STEP TEMPERATURE GRADIENTS

Consider a single spherical particle of diameter dinjected into and entrained by a plasma gas in local thermodynamic equilibrium. Due to the small size of the particle and the low relative velocity between the particle and the gas typical of plasma processing operations, let us assume the convective effects to be negligible. However, let the particle be large enough so that the discontinuum effect on the transport processes is not significant. Further, let the temperature T, the total gas concentration C and the mole fraction of the diffusing species Y at the particle surface and in the bulk of the gas phase be denoted by subscripts s and ∞ , respectively.

2.1. The Z-potential

Neglecting radiation effects, the governing equation for the steady state heat transfer between the plasma and the particle may be written as

$$\frac{1}{r^2} \frac{\mathrm{d}}{\mathrm{d}r} \left[r^2 \lambda(T) \frac{\mathrm{d}T}{\mathrm{d}r} \right] = 0.$$
(3)

Similarly, for the case of self-diffusion under consideration here, as also for certain cases of equimolar counterdiffusion and for $Y \ll 1$, the governing equation for mass transfer is

$$\frac{1}{r^2}\frac{\mathrm{d}}{\mathrm{d}r}\left[r^2C(T)D(T)\frac{\mathrm{d}Y}{\mathrm{d}r}\right] = 0 \tag{4}$$

and the mass flux at the particle surface, N, is given by

$$N = C(T)D(T)\frac{\mathrm{d}Y}{\mathrm{d}r}\bigg|_{r=d/2}.$$
 (5)

When the variations in plasma properties due to changes in the gas composition are insignificant as in the cases under consideration, it has been shown elsewhere [14] by the present authors that the expression for N may be written as

$$N = \frac{2(Y_{\infty} - Y_{\rm s})}{d} \frac{[I(T_{\infty}) - I(T_{\rm s})]}{[Z(T_{\infty}) - Z(T_{\rm s})]}$$
(6)

where Z(T), which will henceforth be referred to as the Z-potential, is defined as

$$Z(T) = \int_{T_r}^T \frac{\lambda(T)}{C(T)D(T)} \,\mathrm{d}T \tag{7}$$

 $T_{\rm r}$ being a reference temperature.

The use of the Z-potential is no more difficult than that of the heat conduction potential in heat transfer calculations; Z(T), too, may be tabulated as any other thermophysical gas property. Figure 1 shows the Zpotentials of argon and nitrogen plasmas, calculated using the data reported in literature [16, 17], as functions of temperature. It should, however, be pointed out that the diffusivity calculations of Amdur and Mason [17] neglect the effects of excitation, dissociation and ionization. Since these effects could be very significant, especially at the high temperatures typical of plasma processing operations, calculations based on the Z-potential data of Fig. 1 can only serve the purpose of illustration and further work is essential to generate more accurate diffusivity data at high temperatures.

2.2. The integral mean diffusivity

The mass flux can also be expressed in terms of the mass transfer coefficient, k, as



FIG. 1. The Z-potentials of argon and nitrogen as a function of temperature (reference temperature = 300 K).

While the selection of the bulk gas concentration, C_{∞} , as the reference concentration in the above expression may appear somewhat arbitrary, it is consistent with the fact that C_{∞} can be measured most accurately.

As shown by the present authors in a previous paper [14], the integral mean diffusivity of the gas, \overline{D} , defined as

$$\bar{D} = \frac{1}{C_{\infty}} \frac{I_{\infty} - I_{s}}{Z_{\infty} - Z_{s}}$$
(9)

where I_{∞} refers to $I(T_{\infty})$, Z_{∞} to $Z(T_{\infty})$ and so on, should be used to evaluate the mass transfer coefficient when steep temperature gradients exist. The above definition of \overline{D} is such that the well-known Ranz-Marshall correlation, $Sh = kd/\overline{D} = 2.0$, is valid even under extremely nonisothermal conditions typically encountered in plasma-particle systems. In terms of \overline{D} , the plasma-particle mass transfer rate may be written as

$$N = \frac{2\bar{D}C_{\infty}}{d} \left(Y_{\infty} - Y_{\rm s}\right). \tag{10}$$

As mentioned earlier, the film temperature (an arithmetic mean of the bulk gas and particle temperatures) has frequently been used in the past to evaluate the transport properties in nonisothermal problems. The expression for mass flux using the film temperature approximation, $N_{\rm fr}$ is given by

$$N_{\rm f} = \frac{2C(\bar{T})D(\bar{T})}{d} \left(Y_{\infty} - Y_{\rm s}\right) \tag{11}$$

where $\overline{T} = (T_s + T_{\infty})/2$.

The ratio of the mass flux calculated using the above expression to the exact mass flux calculated using equation (10) is plotted as a function of the particle surface temperature, T_s , in Fig. 2 for different nitrogen plasma temperatures. As the figure indicates, the film temperature may overpredict or underpredict the mass flux by as much as 20% in some cases depending upon the prevailing system conditions. It is possible that the errors may be even more significant if more accurate diffusivity data, which takes into account excitation, dissociation and ionization, is used in the calculations. Whether the film temperature overpredicts or underpredicts the mass flux will be determined by the temperature dependencies of I(T)and Z(T) as well as by the temperature at which dissociation/ionization of the plasma gas begins. For example, the cases considered in Fig. 2 correspond to film temperatures ranging from 3150 K (i.e. a 300 K particle in a 6000 K plasma) to 7500 K (i.e. a 4000 K particle in a 11,000 K plasma). In this range of \overline{T} values, the product $C(\bar{T})D(\bar{T})$ increases between 3150 K and 6000 K but then drops due to the onset of dissociation. This explains the observed reversal of trends which is clearly illustrated by the curve for a plasma temperature of 9000 K.

As previously pointed out, the I(T) and Z(T) values can be tabulated for any given plasma gas and this makes the use of the integral mean diffusivity



FIG. 2. Ratio of the mass flux calculated using the arithmetic mean temperature [equation (11)] to the exact mass flux [equation (10)] as a function of the particle surface temperature, T_s , for different plasma temperatures.

extremely convenient. Besides, the inadequacy of the use of the film temperature, as illustrated by Fig. 2, provides additional incentive for using \overline{D} to determine the mass transfer coefficient in plasma-particle mass transfer calculations.

3. THE NONCONTINUUM EFFECTS

The importance of noncontinuum effects in plasmaparticle systems has long been recognized [18]. The extent of this effect, which is generally referred to as the Knudsen effect, is determined by a dimensionless parameter called the Knudsen number defined as [19]

$$Kn = \frac{l}{d} \tag{12}$$

where d is the particle diameter and l the mean free path of the gas molecules. Typically, the particles employed in plasma processing are $1-100 \,\mu\text{m}$ in diameter while the molecular mean free path is of the order of $1-10 \,\mu\text{m}$ and the conditions correspond to Knudsen numbers in the range 0.01 < Kn < 10. In such cases, the Knudsen effect could be significant and the continuum transport mechanisms such as Newton's, Fourier's and Fick's laws fail to completely describe the transport processes. Availability of analytical expressions for the heat and mass fluxes in the presence of the Knudsen effect is, therefore, a prime requirement for realistic interpretation of solid conversion data obtained from a plasma-solid reaction system.

The noncontinuum effects in the Knudsen number range 0.001–0.1 (called the jump regime) can be well estimated using the jump theory which was first proposed by Maxwell [20]. According to this theory, discontinuities in velocity, temperature and composition develop at the gas-particle interface when the mean free path is comparable to the particle size. Chen and Pfender have recently revised this theory for plasma-particle momentum and heat transfer calculations [12, 13] and these authors have also shown that the predictions of the jump theory may be extended to Knudsen numbers of 0.8 with little error.

However, the extent of the Knudsen effect on mass transfer under plasma conditions remains yet to be investigated. The Z-potential approach provides a convenient tool to deal with this problem. A predictive model for the Knudsen effect is developed in this section by combining the Z-potential approach with the jump theory. Since the method also necessitates the use of the heat transfer results, relevant expressions from Chen and Pfender's derivation for the heat transfer case have been incorporated into the following analysis—the reader is referred to the paper by these authors [12] for mathematical details of the derivation.

As in the previous section, consider a single spherical particle injected into and entrained by the plasma gas and let all the assumptions made therein also hold. However, in addition, suppose that the particle size is small enough such that the molecular mean free path is comparable with it and the Knudsen number lies in the range corresponding to the jump regime. Then, the Knudsen effect cannot be ignored since discontinuities in velocity, temperature and composition may develop at the plasma-particle interface. Apart from the conditions at the particle surface and in the bulk plasma specified in Section 2, let T_0 and Y_0 be the temperature and the mole fraction of the diffusing species in the gas phase in the immediate vicinity of the particle (henceforth referred to as the immediate gas phase)-these, it must be noted, are not a priori known quantities and need to be determined using the corresponding jump and flux expressions as explained later.

3.1. Determination of T_0

Chen and Pfender [12] have shown that the ratio of the heat flux with the Knudsen effect, Q_0 , to that calculated using the continuum approach, Q_{0c} , is given by

$$\frac{Q_0}{Q_{0c}} = \frac{I_{\infty} - I_0}{I_{\infty} - I_s}$$
$$= 1 / \left[1 + 4 \left(\frac{2 - \theta_h}{2\theta_h} \right) \left(\frac{2\gamma_s}{\gamma_s + 1} \right) \frac{Kn^*}{Pr_s} \right]$$
(13)

where θ_h is the thermal accommodation coefficient, γ_s the specific heat ratio and Pr_s the Prandtl number. Kn^* is the effective Knudsen number defined as

$$Kn^* = \frac{2Pr_s}{\rho_s v_s d} \frac{I_0 - I_s}{H_0 - H_s}$$
(14)

H being the specific enthalpy of the gas, ρ the gas density and v the mean molecular speed which can be calculated from the kinetic theory using the relation

$$v = 2 \left(\frac{2RT}{M\pi}\right)^{1/2}.$$
 (15)

In the above expression, R is the gas constant and M the molecular weight of the gas.

The value of T_0 , which is necessary to perform the mass transfer calculations discussed later, can be determined from equation (13). However, the equation is only implicit in I_0 (or, equivalently, T_0) and, therefore, an iterative procedure needs to be used to find T_0 .

3.2. Composition jump at the particle surface

Following a procedure similar to that described by Kennard [20] for the temperature jump, an expression for the composition jump at the particle surface may be written as

$$Y_0 - Y_s = \frac{2 - \theta_m}{2\theta_m} \frac{4M}{\rho v} CD\left(\frac{dY}{dr}\right)_0$$
(16)

where θ_m may be thought of as a surface 'condensation' coefficient. It may be mentioned here that the concept of a condensation coefficient and of a mole fraction discontinuity due to noncontinuum effects has been previously used by Harvey and Meyer [21] in their theoretical analysis of the vaporization of liquid metal droplets in arc-heated gas streams. However, these authors have used film temperatures for property evaluation since they were concerned only with nonionized gases and such is not the case in the present study.

Using equation (16), the mass flux at the immediate gas phase can also be written as

$$N_0 = CD\left(\frac{\mathrm{d}Y}{\mathrm{d}r}\right)_0 = \frac{2\theta_{\mathrm{m}}}{2-\theta_{\mathrm{m}}}\frac{(\rho/M)v}{4}(Y_0 - Y_{\mathrm{s}}). \quad (17)$$

Replacing I_s , Z_s and Y_s in equation (6) by I_0 , Z_0 and Y_0 , respectively, to account for the jumps, an expression for the mass flux at the immediate gas phase, N_0 , may also be written as

$$N_0 = \frac{2}{d} \frac{I_{\infty} - I_0}{Z_{\infty} - Z_0} (Y_{\infty} - Y_0).$$
 (18)

For a near-isothermal case, the gas properties may be assumed to be approximately constant and equal to those corresponding to the particle surface temperature T_s . Then, equation (18) reduces to

$$N_0 = \frac{2}{d} C_s D_s (Y_\infty - Y_0).$$
 (19)

Using equations (17) and (19) to solve for Y_0 and introducing the expression for gas viscosity at room temperature ($\mu_s \cong 0.5\rho_s v_s l_s$), we obtain

$$\frac{Y_{\infty} - Y_{\rm s}}{Y_{\infty} - Y_{\rm 0}} = 1 + 4 \frac{2 - \theta_{\rm m}}{2\theta_{\rm m}} \frac{K n_{\rm s}}{S c_{\rm s}}$$
(20)

where Sc_s is the Schmidt number defined as $Sc_s = \mu_s / \rho_s D_s$.

Similarly, for large temperature differences such as those typically encountered in plasma-particle operations, equations (17) and (18) need to be used to solve for Y_0 . After approximating $\rho v \cong \rho_s v_s$, as done by Chen and Pfender [12] in the case of heat transfer, the solution can be rearranged to

$$\frac{Y_{\infty} - Y_{s}}{Y_{\infty} - Y_{0}} = 1 + 4 \frac{2 - \theta_{m}}{2\theta_{m}} \frac{Kn^{*}}{Sc^{*}}$$
(21)

where Kn^* is the effective Knudsen number given by equation (14) while Sc^* is the modified Schmidt number defined as

$$Sc^{*} = \frac{I_{0} - I_{s}}{I_{\infty} - I_{0}} \frac{Z_{\infty} - Z_{0}}{H_{0} - H_{s}} \frac{Pr_{s}}{M}.$$
 (22)

Equation (21) is explicit with respect to Y_0 . Therefore, no trial and error procedure is necessary to perform the composition jump calculation once the heat transfer calculation has been completed and the value of T_0 known.

3.3. The Knudsen effect on the plasma-particle mass transfer

As in the case of heat transfer [12], the plasmaparticle mass transfer rate may be affected significantly as a result of the Knudsen effect. In order to ascertain the extent of this effect, it is most informative to compare the mass transfer rate in the presence of the temperature and composition jumps to that calculated using the continuum approach. When the continuum transport mechanisms can be extended all the way to the particle surface, there are no temperature or composition jumps and the mass flux using the continuum approach, N_{0c} , is given by equation (6). Then, from equations (6) and (18), we have

$$\frac{N_{0}}{N_{0c}} = \frac{I_{\infty} - I_{0}}{I_{\infty} - I_{s}} \frac{Z_{\infty} - Z_{s}}{Z_{\infty} - Z_{0}} \frac{Y_{\infty} - Y_{0}}{Y_{\infty} - Y_{s}}.$$
 (23)

From the above expression, the calculation of the ratio of the mass fluxes with and without the Knudsen effect is extremely straightforward. Once the value of T_0 is determined from equation (13) using a trial and error procedure, the first two terms on the RHS of the above expression can be evaluated using the tabulated

I and Z values while the third term can be calculated using equation (21).

4. RESULTS AND DISCUSSIONS

4.1. Example calculation

Some calculations were performed on an actual thermal plasma-particle system to illustrate the extent of the Knudsen effect on mass transfer. Computations were carried out for $5-80 \,\mu\text{m}$ diameter particles at surface temperatures in the range $1000-4000 \,\text{K}$ immersed in a nitrogen plasma at temperatures up to $15,000 \,\text{K}$ —all conditions typical of those encountered in actual plasma-processing operations. For nitrogen properties such as density, thermal conductivity and diffusivity, the data provided in refs. [16, 17] was used. Figures 3–7 illustrate the results of this case study and each of these figures is individually discussed below.

It is seen from Fig. 3 that the Knudsen effect on plasma-particle mass transfer could be extremely significant, the effect becoming more pronounced with decreasing particle size. For example, the reduction of mass flux due to the Knudsen effect in a nitrogen plasma at 10,000 K and for $\theta_{\rm h} = \theta_{\rm m} = 0.8$, is under 3 % for a 80 μ m particle, approximately 15% for a 20 μ m particle and as high as 43% for a 5 μ m particle.

An interesting aspect of the Knudsen effect on mass transfer in plasma-particle systems is illustrated in Fig. 4. From the figure, it is clear that a mass transfer enhancement because of the Knudsen effect is possible under certain conditions. A plausible explanation for such a behavior would be that the temperature jump at the particle surface effectively enhances the diffusion coefficient and the thermal conductivity. If, and when, the conditions are such that the term $(Z_{\infty} - Z_{s})/(Z_{\infty} - Z_{0})$ in equation (23) is sufficiently large to offset the effect of the other two terms, which tend to decrease the mass flux, enhancement will occur. The determining factor, therefore, is the nature of variation of the heat conduction potential and the Z-potential of the plasma gas with temperature. It should be pointed out here that the low value of the thermal accommodation coefficient (0.2) is more realistic for a relatively uncontaminated particle surface or for an evaporating particle [22]. It must also be mentioned that the case of mass flux



PLASMA TEMPERATURE, K

FIG. 3. The mass flux ratio, N_0/N_{0c} , as a function of the plasma temperature, T_{∞} , for particles of different size in a nitrogen plasma ($T_s = 1000 \text{ K}$, $\theta_h = 0.8$, $\theta_m = 0.8$).



PLASMA TEMPERATURE, K

FIG. 4. Mass flux enhancement due to the Knudsen effect in a nitrogen plasma ($T_s = 1000$ K, $\theta_h = 0.2$, $d = 20 \,\mu$ m).



PLASMA TEMPERATURE, K

FIG. 5. Effect of the condensation coefficient, θ_m , on the mass flux ratio, N_0/N_{0c} , in a nitrogen plasma $(T_s = 1000 \text{ K}, \theta_h = 0.8, d = 5 \,\mu\text{m}).$



PLASMA TEMPERATURE, K

FIG. 6. Effect of the thermal accommodation coefficient, θ_h , on the mass flux ratio, N_0/N_{0c} , in a nitrogen plasma ($T_s = 1000 \text{ K}$, $\theta_m = 0.8$).



PLASMA TEMPERATURE, K

FIG. 7. Effect of the particle surface temperature, $T_{\rm s}$, on the mass flux ratio, N_0/N_{0c} , in a nitrogen plasma $(\theta_{\rm h} = 0.8, \theta_{\rm m} = 0.8, d = 5 \,\mu{\rm m}).$

enhancement appears to be an exception to the previous generalization that the Knudsen effect becomes more pronounced with decreasing particle size.

Figure 5 illustrates the strong influence of the condensation coefficient, θ_m , on the Knudsen effect. The figure, which exhibits the results for a $5 \mu m$ particle immersed in a nitrogen plasma for the case $\theta_h = 0.8$, shows that the mass flux ratio is greater than 0.60 when $\theta_m = 1$ but can drop to as low as 0.2 for smaller values of θ_m .

For a given particle size, the extent of the Knudsen effect on mass transfer does not seem to depend as strongly on the thermal accommodation coefficient, θ_h , as on θ_m . Figure 6 shows the results of calculations with two different θ_h values (0.8 and 0.2) for a single particle size and it is clear that a big change in θ_h affects the mass flux ratio only slightly. Another observation, which contrasts the one made by Chen and Pfender [12] for heat transfer, stands out: the Knudsen effect on heat transfer invariably becomes more pronounced as θ_h is decreased but this is not always true in the case of mass transfer. A comparison of the predictive expressions for the heat and mass flux ratios explains the differing trends.

A decrease in θ_h is accompanied by an increase in T_0 . From the expression for the heat flux ratio developed by Chen and Pfender [equation (13)], it is clear that the ratio Q_0/Q_{0c} always decreases with an increase in T_0 . However, the term $(Z_{\infty} - Z_s)/(Z_{\infty} - Z_0)$ which also appears in the mass flux expression of equation (23), always increases with T_0 . Therefore, the effect of θ_h on N_0/N_{0c} can only be determined on an individual basis from the temperature dependencies of I and Z for the plasma gas in question and from the prevailing system conditions.

Finally, Fig. 7 illustrates the effect of the particle surface temperature on the mass flux ratio. As in the case of θ_h , the particle surface temperature is seen to have a small effect on plasma-particle mass transfer. A generalization as to whether the mass flux ratio increases/decreases with surface temperature again does not appear to be possible; however, this does not present a problem since the prediction method developed in this paper is so convenient that every plasma-particle system can be dealt with individually.

It must be mentioned that the prediction method for the Knudsen effect on mass transfer presented in this paper is developed based on the jump theory which is considered valid for Knudsen numbers less than 0.1. Chen and Pfender [12] have shown by comparing their theoretical predictions with experimental data that, at least in the case of small temperature differences between the gas and the particle surface, the predictions of the jump theory for heat transfer can be extended up to Kn values of 0.8 with little error. While the same may also be true in the case of mass transfer, verification is not possible at this stage because of the unavailability of experimental mass transfer data and, therefore, the method should be used with some caution for higher Kn values. Thermal diffusion, which could play a significant role in plasma-particle mass transfer [14], was also neglected in the present study; the extension of the method to include thermal diffusion, and to be applicable to more complicated systems, is being currently investigated [15].

5. CONCLUSIONS

An analysis leading to the Z-potential and the integral mean diffusivity, both of which have served to eliminate the problems posed by steep temperature gradients in plasma-particle mass transfer calculations, is presented. Combining the Z-potential approach and the jump theory, a predictive model is developed for the calculation of the Knudsen effect on mass transport. The use of the proposed model is found to be extremely convenient, requiring no substantial computation once the corresponding heat transfer problem is solved.

The results of some example calculations indicate that the Knudsen effect on plasma-particle mass transfer could be significant, with the effect becoming more appreciable as the particle size decreases. The plasma temperature and the condensation coefficient are also found to have a strong influence on the ratio of the mass flux with and without the Knudsen effect.

The particle surface temperature and the thermal accommodation coefficient have only a slight effect on the mass flux ratio. The nature and extent of their effect is determined by the temperature dependencies of the heat conduction potential and the Z-potential of the plasma gas.

Under certain conditions, it is observed that the Knudsen effect can even cause a mass flux enhancement. Whether or not any enhancement is possible is determined by the I vs T and the Z vs T data of the plasma gas in question. Due to the varied nature of the I(T) and Z(T) values of plasma gases, and the fact that the calculation results are quite sensitive to these values, it is clear that any plasmaparticle system should be studied on an individual basis to determine the extent of the Knudsen effect under the prevailing system conditions.

Acknowledgements—This research was performed as part of the Idaho National Engineering Laboratory's Strategic and Critical Materials research program sponsored by the Interior Department's Bureau of Mines and conducted under Department of Energy contract No. DE-AC07-76ID01570.

REFERENCES

- P. Fauchais and J. M. Baronnet, State of the art plasma chemical synthesis of homogeneous and heterogeneous products, *Pure appl. Chem.* 52, 1669–1705 (1980).
- R. K. Rains and R. H. Kadlec, The reduction of Al₂O₃ to aluminum in a plasma, Met. Trans. 1, 1501–1506 (1970).
- 3. G. Thursfield and G. J. Davies, Sulphide processing in induction plasmas, Trans. Instn Min. Metall. 83,

C191-C192 (1974).

- H. L. Gilles and C. W. Clump, Reduction of iron ore with hydrogen in a direct current plasma jet, *Ind. Engng Chem. Proc. Des. Dev.* 9, 194-207 (1970).
- 5. C. S. Stokes, In *Reactions Under Plasma Conditions* (Edited by M. Venugopalan), Vol. 2, Chap. 15. Wiley-Interscience, New York (1971).
- J. D. Chase and J. F. Skriven, Plasma-jet process for beneficiation of titaniferous ores, AIChE Symposium on Plasma Chemical Processes, Atlantic City (1976).
- 7. J. Szekely and N. J. Themelis, *Rate Phenomena in Process Metallurgy*, p. 459. Wiley-Interscience, New York (1971).
- M. A. Field, D. W. Gill, B. B. Morgan and P. G. W. Hawksley, Combustion of pulverized coal, The British Coal Utilization Research Association (1967).
- E. Bourdin, P. Fauchais and M. I. Boulos, Transient heat conduction under plasma conditions, *Int. J. Heat* Mass Transfer 26, 567-582 (1983).
- W. E. Ranz and W. R. Marshall, Evaporation from drops: part II, Chem. Engng Prog. 48, 173-180 (1952).
- 11. Xi Chen and E. Pfender, Heat transfer to a single particle exposed to a thermal plasma, *Plasma Chem. Plasma Process.* 2, 185-212 (1982).
- Xi Chen and E. Pfender, Effect of the Knudsen number on heat transfer to a particle immersed into a thermal plasma, *Plasma Chem. Plasma Process.* 3, 97-113 (1983).
- 13. Xi Chen and E. Pfender, Behavior of small particles in a

thermal plasma flow, Plasma Chem. Plasma Process. 3, 351-366 (1983).

- S. V. Joshi, J. Y. Park, P. R. Taylor and L. S. Richardson, The integral mean diffusivity for particlegas mass transfer calculations under thermal plasma conditions, *Plasma Chem. Plasma Process.* 5, 143-161 (1985).
- 15. S. V. Joshi, Ph.D. thesis in progress, University of Idaho.
- M. I. Boulos, Internal Report, Université de Sherbrook (May 1984).
- I. Amdur and E. A. Mason, Properties of gases at very high temperatures, *Physics Fluids* 1, 370–383 (1958).
- B. Waldie, Review of recent work on the processing of powders in high temperature plasmas. Part II—Particle dynamics, heat transfer and mass transfer, *Chem. Engng*, *Lond.* 259, 188–193 (1972).
- S. A. Schaaf and P. L. Chambre, In Fundamentals of Gas Dynamics (Edited by H. W. Emmons), Vol. 3. Princeton University Press, Princeton, NJ (1958).
- 20. E. H. Kennard, *Kinetic Theory of Gases*, Chap. 8. McGraw-Hill, New York (1938).
- F. J. Harvey and T. N. Meyer, A model of liquid metal droplet vaporization in arc heated gas streams, *Metall. Trans.* 9, B615-B621 (1978).
- Xi Chen, Y. C. Lee and E. Pfender, The importance of Knudsen and evaporation effects on modeling in thermal plasma processing, *Proc. 6th Int. Symposium on Plasma Chemistry*, Montreal, Canada, pp. 51–58 (1983).

TRANSFERT MASSIQUE PARTICULE-GAZ DANS UN PLASMA

Résumé—On propose une méthode analytique simple pour calculer le transfert massique particule-gaz dans un plasma. Cette méthode, appelée méthode du potentiel Z, tient compte des variations des propriétés de transport du gaz en fonction de la température en permettant l'utilisation de l'expression Sh = 2. L'approche par le potentiel Z est trouvé pratique en tenant compte de l'effet discontinu de Knudsen sur le transfert massique entre le plasma et une petite particule. Un modèle de l'effet de Knudsen, développé en combinant l'approche par Z avec la théorie du saut, est illustré en détail par l'étude du cas relatif au plasma d'azote.

STOFFÜBERGANG ZWISCHEN PARTIKELN UND GAS UNTER PLASMABEDINGUNGEN

Zusammenfassung—Es wird eine einfache analytische Methode für die Berechnung des Stoffübergangs zwischen Partikeln und Gas unter Plasmabedingungen vorgeschlagen. Diese Methode, genannt die Z-Potential-Methode, berücksichtigt die Temperaturabhängigkeit der Transporteigenschaften des Gases vollständig. Sie untersagt jedoch die Anwendung des bequemen isothermen Ausdrucks Sh = 2,0. Die Z-Potential-Näherung ist auch sehr nützlich zur Ermittlung des Knudsen-Diskontinuum-Effekts beim Stoffübergang zwischen Plasmagas und kleinen Teilchen. Ein Vorhersagemodell für den Knudseneffekt wird im Detail anhand eines Stickstoffplasmasystems vorgestellt. Das Modell wurde durch Kombination der Z-Potential-Methode mit der Sprungtheorie entwickelt.

МАССООБМЕН МЕЖДУ УАСТИЦЕЙ И ГАЗОМ В УСЛОВИЯХ ОБРАЗОВАНИЯ ПЛАЗМЫ

Аннотация — Предложен простой аналитический метод расчета массообмена между частицей и газом в условиях образования плазмы. Этот метод, называемый методом Z-потенциала, полностью учитывает зависимость переносных свойств газа от температуры, позволяя при этом использовать удобное изотермическое выражение Sh = 2,0. Показано, что метод Z-потенциала очень удобен для установления влияния кнудсеновского слоя на перенос массы между газом плазмы и малой частицей. Модель расчета эффекта Кнудсена, разработанная путем сочетания метода Z-потенциала с теорией скачка, детально проиллюстрирована на примере исследования плазмы азота.