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Particle Dynamics and Particle Heat and Mass Transfer in Thermal Plasmas. Part II. Particle Heat and Mass Transfer in Thermal Plasmas

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This paper is concerned with a review of heat and mass transfer between thermal plasmas and particulate matter. In this situation various effects which are not present in ordinary heat and mass transfer have to be considered, including unsteady conditions, modified convective heat transfer due to strongly varying plasma properties, radiation, internal conduction, particle shape, vaporization and evaporation, noncontinuum conditions, and particle charging. The results indicate that (i) convective heat transfer coefficients have to be modified due to strongly varying plasma properties; (ii) vaporization, defined as a mass transfer process corresponding to particle surface temperatures below the boiling point, describes a different particle heating history than that of the evaporation process which, however, is not a critical control mechanism for interphase mass transfer of particles injected into thermal plasmas; (iii) particle heat transfer under noncontinuum conditions is governed by individual contributions from the species in the plasma (electrons, ions, neutral species) and by particle charging effects.

KEY WORDS: Thermal plasmas; heat and mass transfer; review.

1. INTRODUCTION

In thermal plasma processing, heat and mass transfer between the plasma and the injected particles plays a crucial role. In plasma synthesis, for example, complete evaporation of the injected particles may be required for achieving the desired results. On the other hand, in plasma spraying, completely molten particles without evaporation are desired for making high-quality coatings. These examples indicate that proper particle heat and mass transfer is essential. In continuation of a previous survey on particle

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Table I. Effects Involved in Particle Heat and Mass Transfer in a Thermal Plasma

1. Unsteady condition
2. Modified transfer coefficients due to strongly varying plasma properties
3. Vaporization and evaporation
4. Noncontinuum effect
5. Radiation
6. Internal conduction
7. Particle shape
8. Particle charging
9. Combination of above effects

dynamics in thermal plasma flows,(1) this survey will be devoted to the heat and mass transfer situation of particulate matter injected into thermal plasmas.

Compared to the relatively few studies associated with particle dynamics in thermal plasmas, a large number of papers have been published about particle heat and mass transfer in thermal plasmas. As mentioned in the previous survey,(1) particles injected into a thermal plasma will experience a number of effects which are not present in ordinary gases. Waldie(2) described in a comprehensive review the most important effects which have to be considered, including heat transfer associated with strongly varying plasma properties, vaporization, dissociation, recombination, radiation, and noncontinuum situations. Over the past years, many papers have been published concerned with one or several of these effects.(3-20) The results of these studies will be summarized and discussed in later sections of this review.

Although most of these effects have been known for many years, there is still no satisfactory understanding of their relative importance and of their synergistic interplay in high-temperature environments. It is felt that a survey on this subject is timely, considering the increasing attention which thermal plasma processing has attracted over the past years.

The various effects which are known today are listed in Table I. Emphasis will be on phenomena associated with vaporization and evaporation of particles and on particle heat transfer under noncontinuum conditions, including particle charging effects.

2. GENERAL CONSIDERATIONS

For a spherical particle with symmetric boundary conditions, heat transfer within the particle is described by the conduction equation, i.e.,

$$\rho_p C_p \frac{\partial T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(K_p r^2 \frac{\partial T}{\partial r} \right) \tag{1}$$

where r is the radial distance from the center of the particle, and ρ_p, C_p, and K_p are the density, specific heat, and thermal conductivity of the particle, respectively.

The following effects imposed by the plasma environment on the particle have to be considered.

2.1. Heat Transfer

2.1.1. Convective Heat Transfer

Convective heat transfer is described by the simple equation

$$q_{\text{conv}} = h(T_\infty - T_w) \tag{2}$$

where h is the convective heat transfer coefficient, T_∞ is the surrounding plasma temperature, and T_w is the surface temperature of the particle. The convective heat transfer coefficients may be derived from semiempirical correlations. For a medium with constant properties, one finds

$$\text{Nu} = \frac{hD_p}{K_p} = 2.0 + 0.6\text{Re}^{1/2}\text{Pr}^{1/3} \tag{3}$$

where D_p is the particle diameter, Nu the Nusselt number, Re the Reynolds number, and Pr the Prandtl number. A number of corrections are needed to make this correlation compatible with the plasma environment. These corrections will be discussed in Section 3.2.

2.1.2. Radiative Heat Transfer

This heat transfer mechanism refers to radiation emitted by a particle, i.e.,

$$q_{\text{rad}} = \varepsilon \sigma T_w^4 \tag{4}$$

where σ is the Stefan–Boltzmann constant, and ε the emissivity.

2.2. Mass Transfer

2.2.1. Vaporization of a Particle

Vaporization is defined as a mass loss process at temperatures below the boiling point. As the temperature of the particle increases, its vapor pressure increases also, leading to mass losses by vaporization, i.e.,

$$\dot{m} = \rho h_m M \ln\left(\frac{p}{p - p_v}\right) \tag{5}$$

where M is the molecular weight of the particle material, h_m the mass transfer coefficient, p the partial vapor pressure with respect to saturation, and p_v the partial vapor pressure with respect to the surface temperature of the particle. The mass transfer coefficient can be derived from a correlation similar to Eq. (3) based on an analogy between mass and heat transfer, i.e.,

$$\mathrm{Sh} = \frac{h_m D_p}{\mathcal{D}} = 2.0 + 0.6\mathrm{Re}^{1/2}\mathrm{Sc}^{1/3} \tag{5a}$$

where Sh is the Sherwood number, Sc the Schmidt number, and $\mathcal{D}$ the interdiffusivity.

2.2.2. Evaporation

If the surface of a particle reaches the boiling point, evaporation will take place which may be described by

$$\dot{m} = q_{\mathrm{net}}/L_e \tag{6}$$

where L_e is the latent heat of evaporation and q_{net} the net heat transfer to the evaporating particle.

In the following, boundary conditions will be specified for the previously mentioned heat and mass transfer relationships.

2.3. Boundary Conditions

2.3.1. Convection and Radiation Boundary Conditions

From an energy balance follows

$$K_p \left.\frac{\partial T}{\partial r}\right|_{r=r_0} = h(T_\infty - T_w) - \varepsilon\sigma T_w^4 \tag{7}$$

where r_0 is the radius of the particle.

2.3.2. Interface between Two Phases

The interface between the solid and the liquid phase may be described by

$$K_p \left.\frac{\partial T}{\partial r}\right|_{r=r_i^-} = K_p \left.\frac{\partial T}{\partial r}\right|_{r=r_i^+} + \rho L_f \frac{dr_i}{dt} \tag{8}$$

and

$$T_{r_i} = T_{\mathrm{melting}} \tag{9}$$

where r_i is the radius of the interface, and L_f the latent heat of fusion.

2.3.3. *Diffusion Boundary of Vaporizing Particle* ($T < T_{\text{boiling}}$)

This boundary condition may be described by

$$K_p \left.\frac{\partial T}{\partial r}\right|_{r=r_0} = h(T_\infty - T_w) - \varepsilon\sigma T_w^4 - \rho h_m M L_e \ln\left(\frac{p}{p - p_v}\right) \tag{10}$$

2.3.4. *Intense Evaporation Boundary Condition for a Particle*

In this case, the balance equation may be written as

$$K_p \left.\frac{\partial T}{\partial r}\right|_{r=r_0} = h(T_\infty - T_w) - \varepsilon\sigma T_w^4 + \rho L_e \frac{dr_0}{dt} \tag{11}$$

and

$$T_w = T_{\text{boiling}} \tag{12}$$

In addition, one more symmetry boundary condition is needed, i.e.,

$$\left.\frac{\partial T}{\partial r}\right|_{r=0} = 0 \tag{13}$$

and the initial condition is

$$T_{t=0} = T_{\text{carrier gas}} \tag{14}$$

The governing equations with the associated boundary conditions can be solved by numerical methods. In addition, for particulate matter with high thermal conductivities, such as metals, the governing equations can be further simplified. By assuming that the thermal conductivity is infinite inside the particle, a uniform temperature field results for the interior of the particle.(7)

The effects listed in Table I will affect the governing equations as well as the boundary conditions. Their relative importance for heat and mass transfer will be reviewed in the following.

3. ESTABLISHMENT OF THE GOVERNING EQUATIONS

3.1. Pseudo-Steady-State Situation

When a particle is injected into a thermal plasma jet reactor, it will experience large variations of the surrounding plasma temperature and velocity fields. This dramatically changing environment requires modifications of the conventional basic equations.

The correlation for convective heat transfer coefficients [e.g., Eq. (3)] has been derived for steady-state, ideal conditions and for a uniform plasma flow past a spherical particle. If the temperature and velocity fields in the boundary layers surrounding a particle do not relax fast enough when the particle is exposed to drastically varying temperatures and velocities, then the so-called pseudo-steady-state conditions are not reached, i.e., steady-state expressions are not valid.

Bourdin *et al.*(15) studied this problem by considering unsteady heat conduction to a particle under suddenly changing plasma conditions. They found that the relaxation time is approximately 1 μs, which is far less than typical particle residence times (≈1 ms). Based on this finding, they concluded that the steady-state expressions for describing plasma-to-particle heat transfer are still valid. Konopliv and Sparrow(21) suggested a dimensionless time scale for convection analysis for unsteady heat transfer assuming Stokesian flow around a sphere, i.e.,

$$\tau \equiv 3\left(\frac{u_\infty^2 K_p}{\rho_p C_p D_p^4}\right)^{1/3} t \tag{15}$$

The dimensionless relaxation time corresponds roughly to $\tau = 1.0$. Based on this number, the relaxation time can be estimated. For a film temperature of 5000 K, a plasma velocity of 100 m/s, and a particle diameter of 100 μm, the relaxation time t becomes approximately 0.5 μs (≪1 ms). Hence a pseudo-steady state will exist for convective heat transfer even for rapidly changing temperature fields.

In addition to changing temperature fields, there is another effect due to changing velocity fields. The characteristic relaxation time for this situation is(22)

$$t \approx D_p / U_\infty \tag{16}$$

For velocities around 100 m/s, and particle diameters of 100 μm, $t \approx$ 1 μs, i.e., the relaxation time is also by far less than the typical particle residence time. Therefore, it can be concluded that locally changing velocity fields do not affect pseudo-steady-state assumptions.

There is, however, one more possible effect, namely a history effect (analogous to the Basset history term for particle dynamics). Unfortunately, no information on this effect is available. Since the influence from locally changing fields is negligible and the time scale for relaxation is much less than the typical residence time (≈1 μs compared to 1 ms), it is expected that the accumulated history effect will not be substantial.

Similar considerations apply to the calculation of mass transfer coefficients. In general, it can be concluded that pseudo-steady-state relations are valid at each location in the plasma, i.e., the expressions derived from steady-state conditions can be used for calculating heat and mass transfer coefficients.

3.2. Variable Property Effects

Equations (3) and (5a) for calculating convective heat and mass transfer coefficients have been derived for constant properties, i.e., without large temperature variation across the boundary layer. In reality, however, there is a large temperature drop across the boundary layer so that correction factors for these expressions are required. Instead of using film-temperature approximations, other correction procedures will be discussed in the following.

For heat transfer, the formula for the Nusselt number can be modified as follows:

(i) Lewis and Gauvin(3) proposed

$$\mathrm{Nu_f} = (2 + 0.515\mathrm{Re_f^{1/2}})(\nu_f/\nu_\infty)^{0.15} \tag{17}$$

where ν is the kinematic viscosity. The subscript f refers to properties corresponding to the film temperature, and the subscript ∞ refers to properties corresponding to the free-stream temperature.

(ii) Fiszdon(9) suggested

$$\mathrm{Nu_f} = (2 + 0.6\mathrm{Re_f^{1/2}Pr_f^{1/3}})\left(\frac{\rho_\infty\mu_\infty}{\rho_w\mu_w}\right)^{0.6} \tag{18}$$

where μ is the dynamic viscosity. The subscript w refers to properties corresponding to the wall temperature.

(iii) Sayegh and Gauvin(10) proposed another expression, i.e.,

$$\mathrm{Nu_w} = 2f_0 + 0.473\mathrm{Pr}^m\mathrm{Re}_{0.19}^{0.552} \tag{19}$$

where $\mathrm{Re}_{0.19}$ is the Reynolds number corresponding to $T_{0.19}$, and $\mathrm{Pr} = 0.672$ (assumed constant for an argon plasma).

$$T_{0.19} \equiv T_w + 0.19(T_\infty - T_w)$$

$$m = 0.78\mathrm{Re}_{0.19}^{-0.145}$$

$$2f_0 = 2\left[1 - \left(\frac{T_w}{T_\infty}\right)^{1+x}\right] \Big/ \left[(1+x)\left(1 - \frac{T_w}{T_\infty}\right)\left(\frac{T_w}{T_\infty}\right)^x\right]$$

with $\mu \sim T^x$, and $x = 0.8$ for argon.

(iv) Lee *et al.*(12) proposed the expression

$$\mathrm{Nu_f} = (2 + 0.6\mathrm{Re_f^{1/2}Pr_f^{1/3}})\left(\frac{\rho_\infty\mu_\infty}{\rho_w\mu_w}\right)^{0.6}\left(\frac{C_{p_\infty}}{C_{p_w}}\right)^{0.38} \tag{20}$$

(v) Vardelle *et al.*(20) suggested the relation

$$\mathrm{Nu}_{\mathrm{avg}} = (2 + 0.515\mathrm{Re}_{\mathrm{avg}}^{1/2}) \tag{21}$$

where the dimensionless numbers are defined by average properties, which are defined as

$$\bar{\phi} \equiv \int_{T_w}^{T_\infty} \phi \, dT/(T_\infty - T_w)$$

$$\mathrm{Re}_{\mathrm{avg}} \equiv \bar{\rho} U_\infty D_p / \bar{\mu}$$

$$\mathrm{Nu}_{\mathrm{avg}} \equiv h D_p / K$$

Since heat transfer plays such an important role for plasma processing of particulate matter, the previously mentioned heat transfer coefficients will be further examined and compared.

Figures 1-3 show the results calculated with the previously mentioned correlations. For facilitating comparisons, the results are plotted for the same parameters (free-stream and surface temperatures). Three cases are considered, namely $T_\infty = 4000$ K and $T_w = 1000$ K, $T_\infty = 10{,}000$ K and $T_w = 2500$ K, $T_\infty = 12{,}000$ K and $T_w = 3000$ K. In addition to the five curves shown in Figs. 1-3, the results of numerical computation are also included in these figures. Lee's simulations(13) are shown for every case, whereas Sayegh and Gauvin's simulations(10) are only shown for the first case.

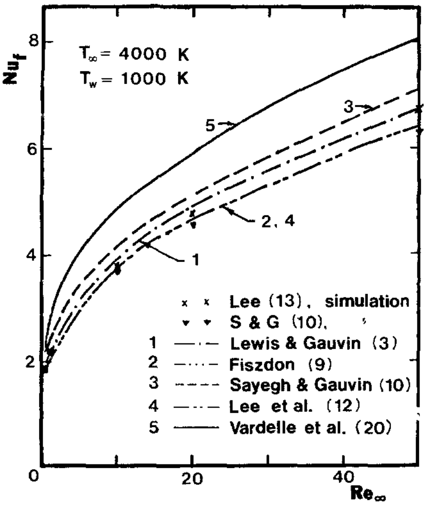

Fig. 1. Nusselt numbers derived by different authors and by computer simulation.

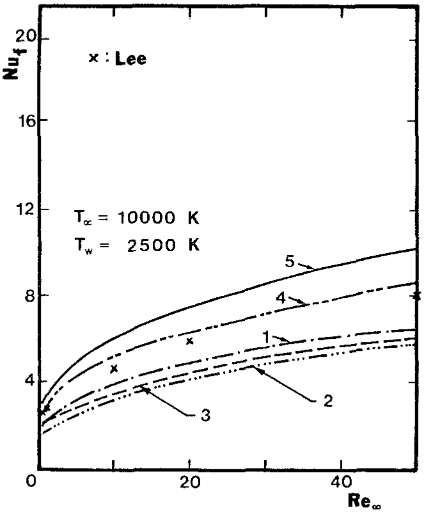

Fig. 2. Nusselt numbers derived by different authors and by computer simulation. (Number and symbol identification are the same as in Fig. 1.)

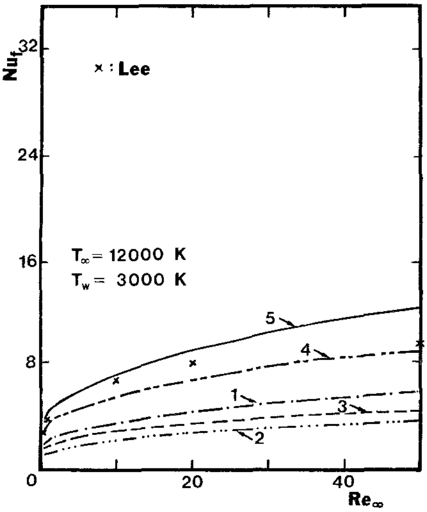

Fig. 3. Nusselt numbers derived by different authors and by computer simulation. (Number and symbol identification are the same as in Fig. 1.)

Figure 1 indicates that all methods except that of Ref. 20 produce similar results for relatively low plasma temperatures. Figures 2 and 3 show that for $T_\infty \geq 10^4$ K where ionization becomes important, the results of Ref. 12 depart from the other results due to changes of the specific heat at higher temperatures.

In every case, the results of Ref. 20 show the highest values. Even though their method is exactly fitted to the case of conduction only,(15) the reason for the large departure from all the other results is not obvious at this time.

Although the results of Ref. 12 have been fitted to the data derived from computer simulations, there is no claim that this method is superior to the others, because the computer simulation has been restricted to an argon plasma.

The analysis for the mass transfer situation is quite similar to the heat transfer case and, therefore, it will not be repeated here.

From the previous discussion it is obvious that large discrepancies exist among various approaches for calculating heat transfer coefficients. This finding already indicates the need for further studies, especially for particle heat and mass transfer under various plasma conditions, in order to develop reliable relationships. At present, there are almost no experimental data available. Thus, computer simulation of the plasma flow over a sphere remains an important tool for determining heat transfer coefficients.

3.3. Radiation Transport

Radiative heat transfer has been discussed extensively for particles immersed into a thermal plasma.(2,4,5,15,17) For low particle loading rates, radiation from the plasma to the particles may be neglected, as well as radiative exchange among particles. Radiative heat losses from particles, however, are frequently taken into account in modeling work.(8,9,12)

Radiative heat transfer becomes important for the following three cases(17): (a) large particles, (b) high surface temperatures and high emissivities of the particles, and (c) low enthalpy differences between the surfaces of particles and the plasma. Radiation losses from particle are negligible except for particles with surface temperatures exceeding 2000 K immersed into plasmas (for example, argon or nitrogen) at temperatures below 4000 K.(15) Johnston(5) also pointed out that radiation is an important factor in the case of augmented flames owing to the lower operating temperature ($T \leq 4500$ K) compared to a plasma jet.

For high particle loading rates, radiation exchange among particles becomes important. In this case, the radiation field produced by emitting/absorbing particles in the plasma is no longer optically thin and radiation absorbed by a particle may be substantial. Unfortunately, little is known for this particular situation.

3.4. Internal Conduction

Internal conduction within a particle in a thermal plasma may lead to large differences between the surface and the center temperature of a particle. The Biot number, defined as the ratio of convective to conductive heat transfer, serves as a criterion for determining the relative importance of heat conduction within a particle.

$$\mathrm{Bi} = \frac{hD_\mathrm{p}}{K_\mathrm{p}} \tag{22}$$

where h is the convective heat transfer coefficient, K_p is the thermal conductivity of a particle, and D_p is the particle diameter.

If $\mathrm{Bi} \ll 0.1$, internal conduction is relatively high, i.e., temperature variations within the particle are negligible. For particles immersed into thermal plasmas, this criterion depends heavily on the material of the particle and on the thermal conductivity of the plasma. Bourdin *et al.*(15) proposed a method for calculating the Biot number assuming that conduction is the governing heat transfer mechanism (small Reynolds numbers) for particle heating in the plasma, i.e.,

$$\mathrm{Bi} = \frac{\bar{K}}{K_\mathrm{p}} \tag{23}$$

where $\bar{K}$ is the average thermal conductivity of the plasma across the boundary layer.

They found that the difference between the surface and center temperature of a particle becomes less than 5% of the difference between the plasma and the surface temperature of a particle if $\mathrm{Bi} < 0.02$.

Since the plasma temperature field is highly nonuniform and since particle heat transfer may not be entirely by conduction, it is safer to take the upper limit of the critical Biot number of the particle during its flight as

$$\mathrm{Bi}_\mathrm{crit} = 2 \times \frac{\bar{K}(T_\mathrm{avg})}{K_\mathrm{p}(T_\mathrm{w,avg})} \tag{24}$$

where the factor of 2 incorporates the convective influence, and thermal conductivities of the particle and the plasma are chosen with respect to estimated average temperatures.

Based on this criterion the internal conduction resistance of the particle is negligible if $\mathrm{Bi}_\mathrm{crit} \ll 0.1$. For this case, a simplified approach can be used to calculate heat and mass transfer.(7)

This simplified approach reduces computing time, and also includes irregular particle shapes. For an actual process, hundreds of simulations are needed for determining the overall performance. Therefore, complicated calculation methods are not practical even if temperature variations within the particle are not entirely negligible. The simplified approach represents probably the best choice.

3.5. Particle Shape Effects

Both particle shape and orientation may have large effects on the rate of heat and mass transfer. There are, however, no suitable correlations available to take these effects into account, in particular for plasma heat transfer.

Pasternack and Gauvin(23) proposed a characteristic length for substituting the diameter term in Eqs. (3) and (5a), i.e.,

$$L' = \frac{\text{Total surface area}}{\text{Maximum perimeter perpendicular to the flow}} \tag{25}$$

This definition is quite different from Wadell's sphericity definition(1) because the orientation effect of the particle is also included. Unfortunately, their studies(23) were limited to a Reynolds number range of $500 < \mathrm{Re} < 5000$. The validity of this approach for small particles injected into thermal plasmas (typically $\mathrm{Re} < 50$) is questionable.

If it is sufficient to consider conduction only, the total heat transfer is, in a first approximation, governed by the total surface area of a particle. Because the total energy transfer should be the same, the following relation for sphericity may be established

$$\begin{aligned} \Psi &= \frac{q_{1,\text{particle}}}{q_{2,\text{sphere with same volume}}} \\ &= \frac{\text{Surface area of the sphere}}{\text{Surface area of the particle}} \end{aligned} \tag{26}$$

where q is the local heat flux.

The sphericity seems in this case to be a reasonable scaling parameter. Because of the small Reynolds numbers experienced in thermal plasma processing, this consideration may be close to reality.

The nonspherical shape of particles also invalidates a simple one-dimensional analysis for heat transfer from a plasma to a particle. Two- or three-dimensional conduction analysis is needed if internal conduction is not negligible. The increase of computing time and complexity for multidimensional analysis is quite substantial.

3.6. Vaporization and Evaporation

Vaporization and evaporation are physical processes concerned with mass transfer across a liquid–vapor interface. Vaporization is defined as a mass transfer process driven by vapor concentration gradients existing between the free stream and the particle surface. In contrast, evaporation accounts for large amounts of mass transfer as the surface temperature reaches the boiling point.

At the interface between liquid and vapor phases, a heat balance is maintained and at the same time a bulk flow of material crosses the interface. For high mass transfer rates, the transfer coefficients become functions of the mass transfer rate, thus causing nonlinearities in the transport equations. For example, Chen and Pfender(16) have shown that the heat flux through a liquid–vapor interface is reduced due to the absorption of heat by the vapor. This, however, does not invalidate the definitions of the transfer coefficients.

It is convenient to use transport relations analogous to the heat transfer relations, but add correction factors to the transfer coefficients, since most of the available information on transfer coefficients is limited to small mass transfer rates. In the following, the transport relations for high mass transfer rates will be discussed.

The "stagnant film theory" represents a simplified unidirectional transport model for predicting the variation of the momentum, heat, and mass transfer coefficients with mass transfer rate. Similar to the boundary layer hypothesis, the sole resistance to momentum, heat, and mass transfer is supposed to reside in a film of stagnant fluid surrounding the body. The film thicknesses are assumed to be constant along the surfaces and independent of the mass transfer rate. Detailed derivations can be found in the literature.(24,25) This model provides satisfactory results for systems where the rate of mass transfer is moderate. It has been widely used for interphase heat and mass transfer problems, and it also has been adopted for calculating vaporization rates at liquid and vapor interfaces.(6,8,9) However, the assumption that the film thicknesses remain unaffected by varying mass transfer rates is questionable and can only be tested by pertinent experiments or by more realistic calculations.

In general, a flux may be expressed as the product of a transfer coefficient with a gradient. The latter may be considered as a driving force. For mass transfer, this driving force is called the transfer number. Thus the normal velocity component v of the flux associated with mass transfer through the interface can be determined from the driving force and the transfer coefficient. There are as many different forms of transfer numbers possible as differential equations can be formed from the fundamental balance equations. Since there is a unique value of v, each of the equations based on different transfer numbers valid for a given system must yield the same value in their normalized form.(26)

According to the stagnant film theory, the mass transfer rate increases with increasing driving force (vapor pressure or temperature). As the temperature approaches the boiling point, the driving force approaches infinity, resulting in an infinite mass transfer rate which, of course, is not realistic. Hence, it may be concluded that Eq. (5) is no longer valid as the temperature approaches the boiling point.

In order to circumvent this problem, a simple approach is proposed. If q_{net} is the net surface heat flux to a particle, a heat balance at the surface yields

$$q_{\text{net}} = \dot{m}L_{\text{e}} + q_{\text{int}} \tag{27}$$

where $\dot{m}$ is the mass flow rate of vaporized material, L_{e} is the latent heat of evaporation, and q_{int} the heat flux which raises the internal energy of the particle (sensible heat). The mass flow rate reaches a maximum for $q_{\text{int}} = 0$. Thus, an accommodation factor $f(T)$, which may be a function of temperature, is introduced so that

$$\dot{m}_{\max} = f(T) \cdot (q_{\text{net}}/L_{\text{e}}) \tag{27a}$$

with $0 \leq f(T) \leq 1$. By introducing $f(T)$, it is easy to investigate the relative significance of the surface temperature upon the vaporization rate driven by the vapor pressure. In other words, the control mechanism for this physical process can be determined.

Numerical simulation of a particle residing in a high-temperature surrounding has been performed based on the model described in Ref. 13. Also, the noncontinuum effect, which will be discussed in the next section, will be adopted. With $f(T)$, two limiting cases for vaporization can be established. For $f(T) = 0$, no mass vaporization (or mass diffusion) is taken into account.(7) In the case of $f(T) = 1$ the vaporization rate driven by the vapor pressure is determined by Eq. (5).(6,8)

The results of corresponding calculations are presented in Figs. 4–9, for two different materials (tungsten and iron) and for two different free-stream temperatures (10^4 and 15,000 K). In these figures normalized temperatures are plotted as a function of time after the particle has been exposed to a thermal plasma. Particular attention is focused on the region where vaporization or evaporation occurs. The figures indicate that the surface

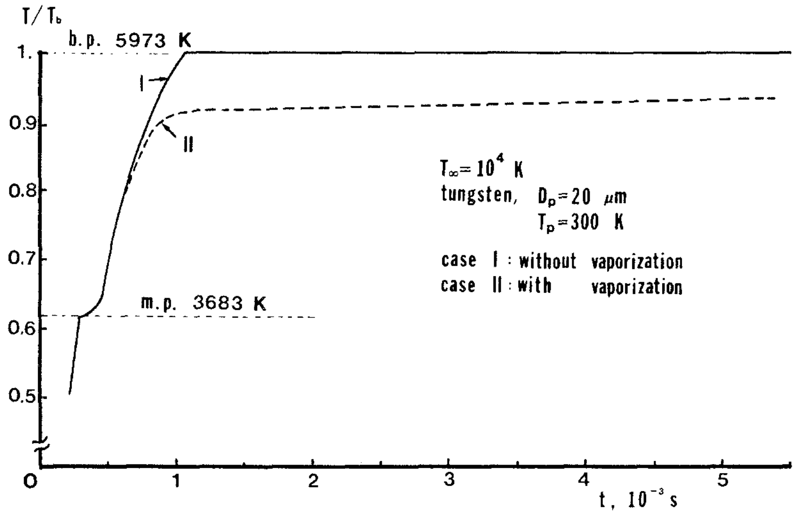

Fig. 4. Normalized temperature history of vaporization and evaporation (evaporation starts as the particle surface reaches the boiling point).

temperatures reach a "plateau" (cases II) at some temperatures below the boiling point and continue to rise very slowly for the case of $f(T) = 1$. For cases I, there is no vaporization before the surface temperature reaches the boiling point. The results of Figs. 5, 7, and 9 show that the total mass transfer through the interface is almost the same for both cases, which is an important finding. Furthermore, the results demonstrate that the

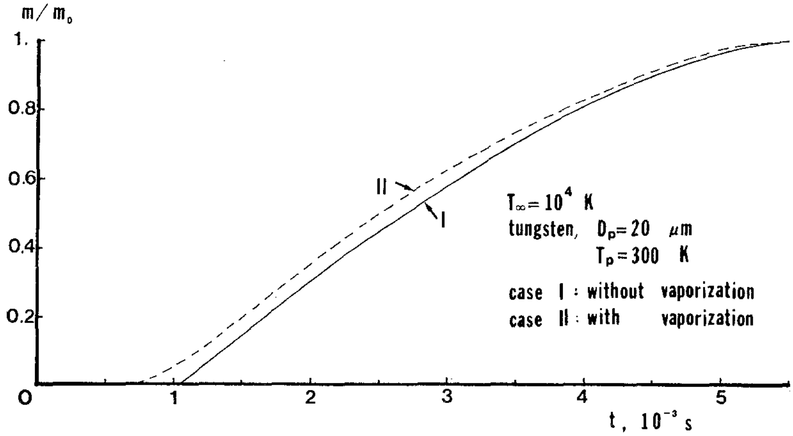

Fig. 5. Normalized total mass transfer from a particle (m_0 = initial mass of the particle).

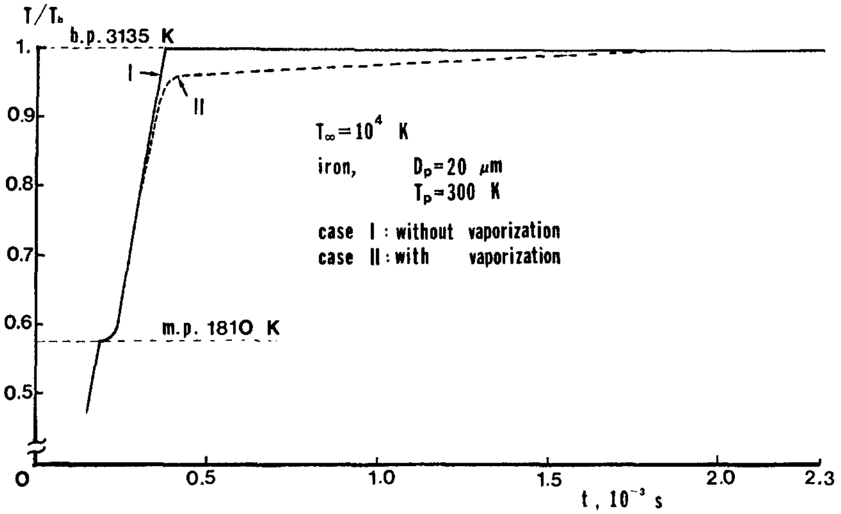

Fig. 6. Normalized temperature history of vaporization and evaporation (evaporation starts as the particle surface reaches the boiling point).

difference between the two limiting cases becomes less significant for higher free-stream temperatures and/or lower-boiling-point materials.

These results can be explained in terms of the relatively high free-stream temperature compared to the surface temperature of a particle. The heat input from the surroundings to a particle is governed by the temperature difference between the surroundings and the particle surface. This tem-

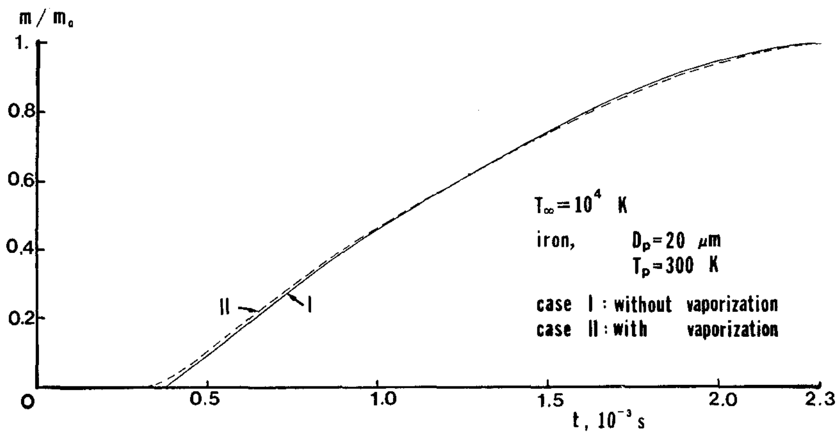

Fig. 7. Normalized total mass transfer from a particle (m_0 = initial mass of the particle).

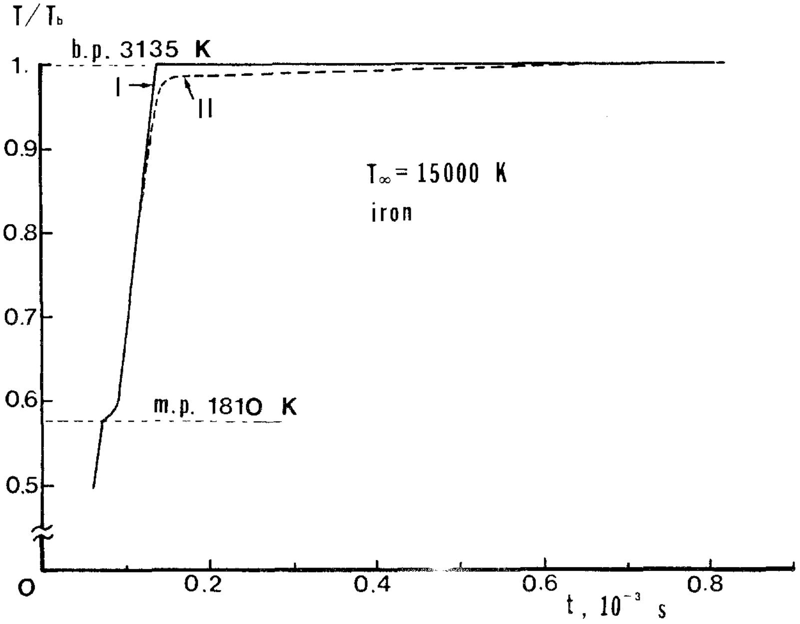

Fig. 8. Normalized temperature history of vaporization and evaporation (evaporation starts as the particle surface reaches the boiling point).

perature difference is little affected by possible variations of the particle surface temperature. As a consequence, the total heat input is not sensitive to the particle surface temperature. Hence, the total mass transfer curves show little sensitivity to the different mechanisms or different surface temperatures. This result reveals that the choice of the equation for calculating vaporization rates is not critical for this interphase mass transfer process.

It is felt that the results shown in Figs. 4–9 should provide an interpretation of the long-standing puzzle about heat and mass transfer through a liquid–vapor interface. A final settlement of this controversy must await experimental verification of these predictions.

3.7. Noncontinuum and Particle Charging Effects

As mentioned previously, the particle sizes used for plasma processing may be of the same order of magnitude as the molecular mean free path lengths in the plasma. This "rarefaction effect" may exert a strong influence on heat transfer.

The noncontinuum effect on heat transfer has been studied by Chen and Pfender(18) in the temperature jump regime, resulting in a proposed

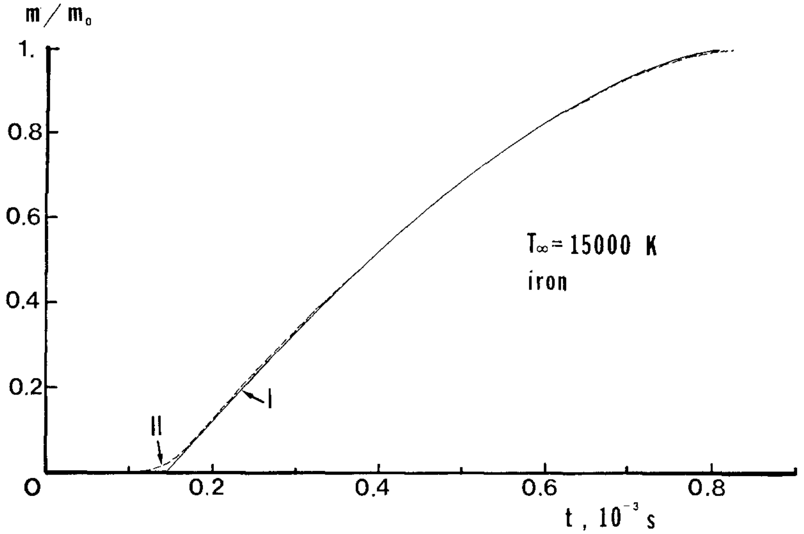

Fig. 9. Normalized total mass transfer from a particle (m_0 = initial mass of the particle).

correction:

$$\frac{q_{\text{noncont}}}{q_{\text{cont}}}=\frac{1}{1+(Z^*/r_p)} \tag{28}$$

where

$$Z^*=\left(\frac{2-a}{a}\right)\left(\frac{\gamma}{1+\gamma}\right)\left(\frac{4\bar{K}}{\rho_w\bar{V}_w\bar{C}_p}\right)$$

is the jump distance. In these equations, a is the thermal accommodation coefficient, γ the specific heat ratio, $\bar{K}$ the average thermal conductivity, $\bar{C}_p$ the average specific heat, $\bar{V}_w$ the mean molecular velocity corresponding to the wall temperature, ρ_w the mass density corresponding to the wall temperature, and r_p the particle radius.

The noncontinuum effect becomes substantial for small particles. Therefore, it is crucial for modeling associated with thermal plasma processing(27) when small particles (<20 μm) are involved. The approach used in Ref. 27 is based on the so-called temperature jump which is valid for Knudsen numbers in the range $0.001 < \mathrm{Kn} < 1$.

Rykalin *et al.*(11) and Godard and Chang(28) studied the extreme case of free molecular flow ($\mathrm{Kn} \geq 10$). Although the free molecular regime is of little concern for modeling of typical thermal plasma processes, the considerations of individual contributions (electron, ion, and neutral particles) and of the particle charging effect used in their work provide useful guidelines for modifying the relations described in Ref. 18.

The temperature jump approach used in Ref. 18 assumes a Knudsen layer around the particle. Free molecular transport is assumed within this layer, while continuum transport prevails outside the layer. Continuity of temperature and of the heat flux corresponding to these two regimes is required for this approach.

In the case of free molecular transport, plasmas, however, are no longer in a local thermodynamic equilibrium (LTE) state, i.e., every species has to be treated separately. Therefore, the conventional temperature jump approach cannot be used directly under plasma conditions. It is more reasonable to use an approach describing individual contributions to heat transfer by neutral species, ions, electrons, and recombination of ions and electrons.(11) Under such circumstances, the particle charging effect may become influential.

A particle injected into a thermal plasma is always negatively charged due to the different thermal velocities and mobilities of electrons and ions. By taking the flux of charged species from Refs. 29 and 30, one finds

for electrons

$$I_e = n_e \left(\frac{k_B T_e}{2\pi m_e}\right)^{1/2} \exp(X_p) \tag{29}$$

for ions

$$I_i = n_i \left(\frac{k_B T_i}{2\pi m_i}\right)^{1/2} \exp(-X_p) \tag{30}$$

where n_e is the number density of electrons, n_i the number density of ions, k_B the Boltzmann constant, T_e the electron temperature, T_i the ion temperature, m_e the mass of the electron, m_i the mass of an ion, e the elementary charge, ϕ_p the surface potential (<0), T_w the surface temperature, and X_p the dimensionless surface potential (<0), i.e.,

$$X_p = \frac{e\phi_p}{k_B T_w}$$

The charging process is very quick, i.e., it happens immediately upon particle injection. The characteristic time for reaching steady-state conditions is typically around 1 μs. A steady-state surface potential is obtained as soon as the fluxes balance, i.e.,

$$I_e = I_i \tag{31}$$

By assuming that only singly ionized species exist in the plasma, i.e.,

$$n_e = n_i \tag{32}$$

the potential becomes

$$X_\mathrm{p} = \tfrac{1}{4} \ln \left[\left(\frac{T_i}{T_e} \right) \left(\frac{m_e}{m_i} \right) \right] \tag{33}$$

A negative surface potential will retard electron bombardment and increase the ion flux. Godard and Chang(28) studied the case of free molecular flow in an ionized gas, considering heat transfer to a sphere. They demonstrated that the particle surface potential plays an important role.

A modified approach based on Ref. 18 has been pursued according to these findings. A limiting sphere concept, assuming a spherical shell surrounding the particle, proposed by Fuchs(31) will be used here. It is assumed that the distance between the shell and the particle is the smallest value among the mean free path lengths of atoms, ions, and electrons, including the Debye length. Free molecular transport prevails within the shell while continuum transport is assumed outside the shell. The number density of each species on the surface of the shell is assumed to be the same as that under LTE conditions corresponding to the jump temperature. Heat transfer of these species must be the same as the continuum transfer outside the shell.

According to Ref. 31, the number density of charged species arriving on the sphere will not be the same as that on the shell due to the required adjustment for the electric potential, i.e.,

$$n_e = n_{e,0} / \{\exp(-X_\mathrm{p}) + 3r_\mathrm{p}[\exp(X_\mathrm{p}) - 1]/4\lambda_e X_\mathrm{p}\} \tag{34}$$

$$n_i = n_{i,0} / \{\exp(X_\mathrm{p}) + 3r_\mathrm{p}[\exp(X_\mathrm{p}) - 1]/4\lambda_i X_\mathrm{p}\} \tag{35}$$

where $n_{e,0}$ is the number density of electrons and $n_{i,0}$ the number density of ions on the shell, X_p the dimensionless negative surface potential, r_p the particle radius, and λ_e, λ_i the mean free path lengths of electrons and ions, respectively.

The energy transport equations taken from Refs. 11 and 27 assume the following form:

for atoms

$$q_\mathrm{atom} = n_a \frac{2k_\mathrm{B}^{3/2} T_\mathrm{jump}^{1/2}}{\sqrt{2\pi m_a}} (T_\mathrm{jump} - T_\mathrm{w}) \tag{36}$$

for ions

$$\begin{aligned} q_\mathrm{ion} = {} & n_i \frac{(k_\mathrm{B} T_\mathrm{jump})^{1/2}}{\sqrt{2\pi m_i}} (1 - X_\mathrm{p} + 0.5X_\mathrm{p}^2)(2k_\mathrm{B} T_\mathrm{jump}) \\ & - 2n_i \frac{(k_\mathrm{B} T_\mathrm{w})^{3/2}}{\sqrt{2\pi m_a}} \left(\frac{T_\mathrm{jump}}{T_\mathrm{w}} \right)^{1/2} \end{aligned} \tag{37}$$

for electrons

$$q_e = n_e \frac{(k_B T_{\text{jump}})^{1/2}}{\sqrt{2\pi m_e}} (2k_B T_{\text{jump}}) \exp(X_p) \tag{38}$$

Recombination between ions and electrons yields

$$q_{\text{recom}} = n_i \frac{(k_B T_{\text{jump}})^{1/2}}{\sqrt{2\pi m_i}} E_i \exp(-X_p) \tag{39}$$

where the q's represent the heat fluxes, T_{jump} the jump temperature, T_w the particle surface temperature, the m's the molecular masses of the species, and E_i the recombination energy.

The number densities of the species are determined by solving the Saha equation for the jump temperature. The jump temperature is determined by the continuity requirement of the heat flux on the shell.

If the electron temperature in the free molecular region is assumed to be equal to the ion temperature, then the dimensionless surface potential X_p assumes a value of −2.8 [see Eq. (33)].

For the case of pure conduction, modified results are shown in Fig. 10 which indicates substantial deviations from previous results.(18)

Figure 11 shows the individual contributions of atoms and of recombination for different particle sizes. If conditions approach the continuum situation, the kinetic energy of the atoms represents the major mechanism for heat transfer. As the particle size decreases, the recombination mechanism becomes dominant. The contributions due to the kinetic energies of

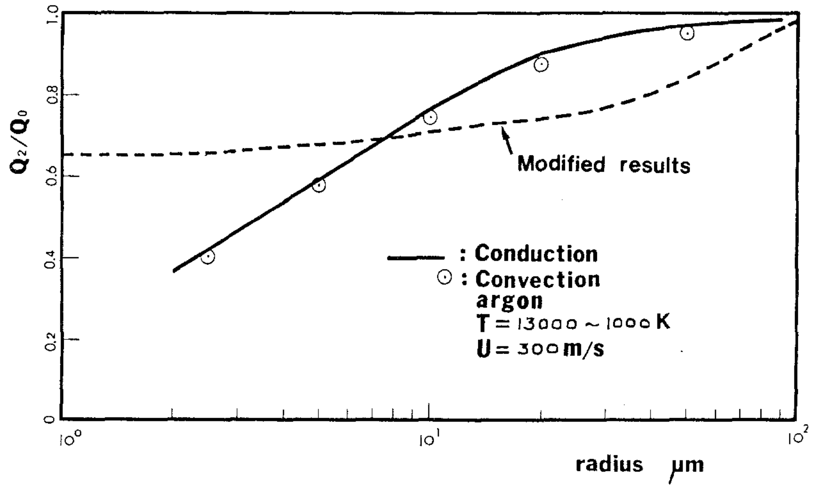

Fig. 10. Heat transfer under noncontinuum conditions (Q_2: with Knudsen effect; Q_0: without Knudsen effect).

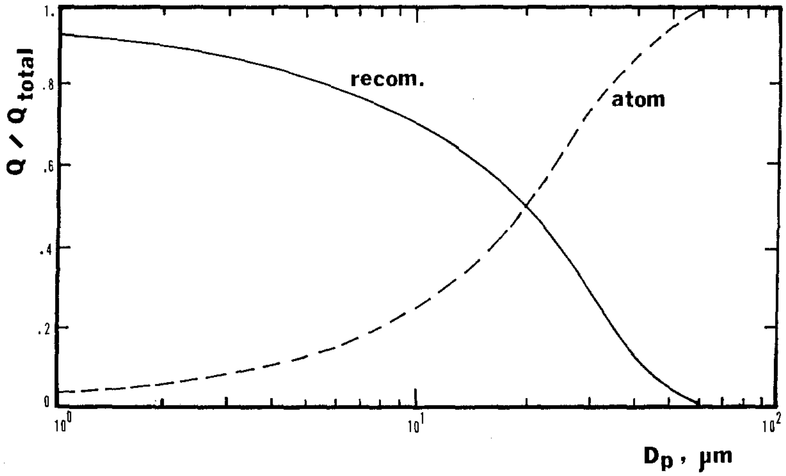

Fig. 11. Individual contributions within the free molecular regime.

ions and electrons are negligible, because of the small ion energies and reduced electron bombardment rate caused by the negative surface potential. Since the contribution of recombination is determined by the ion flux which increases substantially due to particle charging, it seems that the particle charging effect becomes influential under noncontinuum or nonequilibrium (frozen) conditions. For LTE situations in the boundary layer, this effect becomes negligible.

4. SUMMARY

The particle heat and mass transfer processes in thermal plasmas have been reviewed with emphasis on heat transfer coefficients and particle vaporization. In spite of substantial progress in this field, more fundamental work is required, especially in terms of experimental verification of analytical predictions.

The results of this study may be summarized as follows:

1. Convective heat transfer coefficients require substantial modifications to account for strongly varying plasma properties. There are substantial discrepancies among the predictions of various investigators.
2. Vaporization of particles, defined as a mass transfer process corresponding to particle surface temperatures below the boiling point, describes a different particle heating history than that of the evaporation process. But regardless of the postulated mass transfer process,

the mass loss of a particle as a function of time remains almost the same.

3. Considering the situation in the boundary layer surrounding a particle, it has been shown that the contribution of the recombination process does play an important role if frozen conditions prevail in the boundary layer or if one approaches the molecular flow regime. Particle heating due to recombination of electrons and ions on the particle surface is enhanced by the net negative charge carried by the particles.

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