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A unified approach to plasma–particle heat transfer under non-continuum and non-equilibrium conditions

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Abstract—A model for the heat transfer from a stationary, infinite plasma to a particle is developed so as to include rarefaction effects over the entire range of Knudsen numbers, and thermal and chemical nonequilibrium in the vicinity of the particle surface. The overall heat transfer rate is obtained by separately determining the contributions due to heavy species conduction and to energy transport by ion flow to the particle surface. Knudsen effects are accounted for by means of Sherman's interpolation formula for the conduction problem and its equivalent in electrostatic probe theory for the ion transport to the surface. Results are presented for argon plasmas in the pressure range 0.1–1 atm, and for particle sizes in the range 10–100 μm.

1. INTRODUCTION

THE CORRECT prediction of the behavior of particles immersed in a plasma stream is of central importance to the successful application of plasma jets to spraying and coating technologies. The fundamental problem lies in determining realistically the coupling between the particles and the plasma stream, in terms of momentum transfer (which determines the particle trajectories and residence times within the plasma jet), and energy transfer (which affects directly the particle temperature, state, size, shape and therefore, indirectly, the momentum transfer). Much work has been done in the area of numerical modeling of this quite complex physical situation, and the treatment of the coupled problem seems to be well established, provided suitable models are available for the separate problems of momentum and heat transfer to a single particle. Unfortunately, due to the nature of the plasma, and due to constraints on particle size and operating pressure, one finds that the description of these phenomena, which is a prerequisite to the overall description of the coupled problem, is complicated by at least two effects which preclude the application of otherwise well-established models. One is non-equilibrium, thermal and chemical, which may change the relative importance of certain energy transfer mechanisms as well as introduce new ones that cannot be described by equilibrium theory. The second is that the problem neither satisfies the requirements of a continuum approach, nor those of free-molecular theory. Deviations from either limit are not necessarily small, and no established theory is available for the transitional region. The aim of this work is to propose one possible way of including both effects in a model of the heat transfer to a particle, without resorting to a priori assumptions regarding the extent of non-equilibrium, or the magnitude of the deviation from continuum. Analogies with established results in electric probe theory and in heat transfer in rarefied gases (under non-plasma conditions) will be instead introduced, that, in spite of not having a strong theoretical foundation, seem appealing for their ability to correctly recover known limits, and their agreement with experiments. Simplifying assumptions will unavoidably be made, but it is felt that their relaxation should not introduce qualitative differences from the method described here.

2. BACKGROUND

That heat transfer rates to particles may be substantially affected by deviations from continuum was first shown by Chen and Pfender, who carried out the analysis of plasma–particle heat transfer at atmospheric pressure [1], for pressures ranging from 0.01 to 2 atm [2], and for situations where the flow around the particle may contribute a convective component [3]. The deviations from continuum (known as Knudsen effect) associated with the non-negligible Knudsen number $Kn \equiv \lambda/R_p$, where λ is a characteristic mean free path length in the plasma and R_p the particle radius, were treated by means of a conduction potential discontinuity, in analogy to the classical temperature jump approach [4]. Joshi *et al.* [5] kept the mass transfer and heat transfer problems separate, and suggested the use of a generalized diffusivity, implying the validity of some sort of heat and mass transfer analogy. The non-continuum effects were also accounted for by a diffusion potential jump approach. It must be stressed that in both Chen and Pfender's and Joshi *et al.*'s lines of work a plasma in local thermodynamic equilibrium (LTE) was postulated. This assumption implies that all diffusional driving

NOMENCLATURE

A	particle surface area [m^2]
C	electron (ion) molar fraction in the undisturbed plasma
D	diffusion coefficient [m^2 s^{-1}]
$\mathcal{D}$	Damköhler number
e	electron charge [C]
$\mathbf{E}$, E	electric field [V m^{-1}]
f, g, h	Shorthand notations for functions of n, τ, C
h_c	heat transfer coefficient [W m^{-2} K^{-1}]
I	electric current [A]
j_c	dimensionless current density at the particle surface
$\mathbf{J}$, J	current density [A m^{-2}]
k	Boltzmann's constant [J K^{-1}]
K	heavy species translational thermal conductivity [W m^{-1} K^{-1}]
Kn	Knudsen number
Kn_j	effective Knudsen number for ion diffusion
Kn_q	effective Knudsen number for conduction
L	characteristic length [m]
L_R	recombination length [m]
m	mass
Ma	Mach number
n	dimensionless number density
N	number density [m^{-3}]
$\dot{N}$	ionization rate [m^{-3} s^{-1}]
Nu_r	Nusselt number based on R_p and $K_{h\infty}$
P_{tot}	total pressure [Pa]
q_c	dimensionless heat flux at the particle surface
Q	heat transfer rate [W]
Q_{cond}	Heat transfer rate by heavy species conduction [W]
Q_{rec}	energy transport rate due to ion diffusion currents [W]
Q''	heat flux [W m^{-2}]
R	radius, radial coordinate [m]
T	temperature [K]
v	thermal velocity [m s^{-1}]
Z	number of collisions.

Greek symbols

α_{LF}	collisionless ion collection coefficient
α_{TA}	thermal accommodation coefficient
α'	recombination coefficient [m^6 s^{-1}]
λ	mean free path [m]
ρ	plasma density [kg m^{-3}]
Σ	collision cross section [m^2]
τ	dimensionless temperature
Φ_{ioniz}	ionization potential [V]
$\Delta\Phi_{sheath}$	sheath potential drop [V]
$\Omega^{(1,1)}$	first approximation to the ion–neutral momentum transfer collision integral [m^2].

Subscripts

C	continuum limit
e	electron
FM	free-molecular limit
h	heavy (ion and/or neutral)
i	ion
n	neutral
p	particle
T	total (ions + electrons + neutrals)
∞	undisturbed plasma.

forces can essentially be expressed in terms of the temperature gradient and, therefore, could be accounted for in a generalized heat conduction equation by the inclusion of a reactional component in the thermal conductivity of the plasma. The slip and jump methods were applied by Chang [6] for the calculation of drag and convective heat transfer to particles under plasma spraying conditions varying from atmospheric pressure to soft vacuum. Chang allowed for non-equilibrium effects treating number densities and temperatures separately and obtaining density and temperature jumps at the particle surface. His energy transfer rates clearly distinguish between the various contributions, due to heavy species translational energy transport, recombination energy release due to the neutralization of ions on the particle surface and electron enthalpy transport.

Although its use as a 'corrective' boundary condition to the continuum form of the governing equation lacks theoretical justification, the jump approach is generally accepted for Knudsen numbers ranging from 0.001 to 0.1. This implies that the method is expected to describe perturbation of the continuum situation of small magnitude. The authors of ref. [1], based on a comparison with Takao's results [7] for rarefied cold flow heat transfer data for spheres in the transition regime, find agreement up to $Kn = 0.8$. Given the lack of theoretical support to the use of the jump approach, which assumes a zero-order continuum behavior to dominate near the surface, matched to kinetic expressions that strictly apply only to plane geometry, there seems to be little guarantee that the results will be physically realistic and not forced by the nature of the approach itself. It is the authors' opinion that the range of applicability of the jump approach method should be judged according to the magnitude of the deviations from the continuum limit, and therefore its use should be limited to small jump distances relative to the particle characteristic length.

An alternative approach to the problem was introduced by Lee [8] and refined by Chyou [9], and was based on the concept of a limiting Knudsen sphere. The thickness of the shell surrounding the particle is of the order of the shortest among the relevant mean free paths and the Debye length, and transport is free molecular inside the shell and continuum outside. Continuity is required at the continuum–free molecular interface, and is used to provide boundary conditions to the continuum problem. Their results, as discussed in a later section, do not however recover the free-molecular limit for small particle size.

Another major stumbling block of most previous approaches is the assumption of prevailing LTE conditions in the plasma surrounding the particle. This assumption strongly affects the evaluation of properties and, in particular, of the diffusive fluxes of charged particles to the surface. Since equilibrium in relatively high density plasmas is achieved primarily by collisional mechanisms, it is obvious that any study of the rarefaction effects on the heat transfer to small particles must account for possible nonequilibrium, chemical and thermal. This last aspect needs to be carefully considered in view of the potentially important contribution to heat transfer rates due to the release of recombination energy to the surface, and of the effect on the results of the transport property calculations.

3. PROPOSED APPROACH

3.1. *General description*

The approach described here aims to quantify the importance of ion diffusion and recombination energy release, and of thermal conduction in the energy transfer to the surface of particles in the 10–100 μm diameter range, immersed into argon plasmas at pressures ranging from 0.1 to 1 atm. This choice of parameters essentially covers the continuum end of the transition regime (for Kn calculated on the basis of undisturbed plasma properties and particle size, as described later in more detail). No assumptions about thermal or chemical equilibrium will be made, and, for consistency, possible simple expressions for non-equilibrium property calculation will be suggested. Flow effects will be neglected. In dealing with deviations from the continuum situation an alternative approach to the jump model will be adopted: this approach consists of obtaining, for Knudsen numbers in the transition range, the value of the quantity of interest (be it the ion current or the conduction heat transfer rate to the particle) by interpolation between the values calculated in the continuum and free-molecular limits. The interpolation formula may indeed be quite arbitrary, and the only two constraints that one may impose is that it should asymptotically yield the correct continuum and free-molecular limits for $Kn \to 0$ and $Kn \to \infty$, respectively. The arbitrariness of the interpolation formula can only be removed by comparison with experimental data.

This idea was first introduced by Sherman [10] for the correlation of heat transfer data in rarefied gases in the transition regime. The interpolation formula for spheres yielded excellent agreement in spite of its simplicity

$$\frac{Q}{Q_{\mathrm{FM}}} = \frac{1}{1+Kn_q^{-1}}; \quad Kn_q \equiv \frac{Q_{\mathrm{C}}}{Q_{\mathrm{FM}}}$$

where Q_{FM} and Q_{C} are values calculated by approaching the problem in the free-molecular and continuum limits, respectively, and Q represents the actual heat transfer rate. This treatment of the transition regime is known as Sherman's interpolation method, and has been proven to be of a fairly general nature. Cercignani [11] discusses its merits as compared to Millikan's empirical formula and variational methods for the calculation of the drag force on a sphere in the transitional flow regime.

As far as the evaluation of the ion current collected by the particle is concerned, the particle can be treated as a floating spherical probe, and electric probe theory results may be applied. For a physical probe in the transition regime, Talbot and Chou [12] proposed an interpolation formula for some key integral quantities for the prediction of the current collection obtained in an earlier fully kinetic analysis of the problem [13]. Thornton [14] later proposed and substantiated with experimental results, a simpler correlation, the spirit of which is essentially the same as that of Sherman's

$$\frac{I}{I_{\mathrm{FM}}} = \frac{1}{1+Kn_j^{-1}}; \quad Kn_j \equiv \frac{I_{\mathrm{C}}}{I_{\mathrm{FM}}}$$

where I_{C} and I_{FM} are again the values obtained from the continuum and free-molecular limiting solutions, respectively, and I is the actual ion current reaching the particle.

That $Q_{\mathrm{C}}/Q_{\mathrm{FM}}$ and $I_{\mathrm{C}}/I_{\mathrm{FM}}$ may be interpreted as effective Knudsen numbers (related to a single Knudsen number based on undisturbed plasma conditions and on particle size), may not be obvious, and will be shown in the following.

Ion current. In the continuum limit, the ion current is essentially driven by diffusion induced by the density gradient that the appropriate boundary condition (zero ion density at the particle, for a catalytic surface) causes. One can qualitatively expect that the diffusion ion current will scale as $I_{\mathrm{C}} \propto AD_{\mathrm{in}\infty}N_{\mathrm{i}\infty}/L$, where, for frozen or near-frozen chemistry, L is a physical dimension of the particle, typically R_{p} for a spherical particle. In the free-molecular limit $I_{\mathrm{FM}} \propto AN_{\mathrm{i}\infty}v_{\mathrm{i}\infty}$. Apart from coefficients which can be expected to be of order unity

$$\frac{I_{\mathrm{C}}}{I_{\mathrm{FM}}} \propto \frac{D_{\mathrm{in}\infty}}{R_{\mathrm{p}}v_{\mathrm{i}\infty}} \propto \frac{\lambda_{\mathrm{in}\infty}}{R_{\mathrm{p}}} \equiv Kn_{\infty}.$$

The relevant mean free path for the Knudsen number is therefore the ion–neutral mean free path in the undisturbed plasma.

Conduction heat transfer rate. In the continuum limit one expects $Q_{\mathrm{C}} \propto AK_{\mathrm{h}\infty}(T_{\infty} - T_{\mathrm{p}})/R_{\mathrm{p}}$, whereas in the free-molecular limit, $Q_{FM} \propto A N_{h\infty} v_{h\infty}(T_\infty - T_p)$, where $v_{h\infty} = v_{i\infty}$ is the thermal velocity for the heavy species (ions and atoms are assumed in thermal equilibrium). By taking the ratio of the two, one finds $Q_C/Q_{FM} \propto K_{h\infty}/N_{h\infty}v_{i\infty}R_p$. Jumping to some results of the property evaluation discussed later, one finds that, to an acceptable degree of accuracy, $K_{h\infty} \propto N_{h\infty}D_{in\infty}$, with some dependence on the degree of ionization. Therefore

$$\frac{Q_C}{Q_{FM}} \propto \frac{D_{in\infty}}{R_p v_{i\infty}} \propto \frac{\lambda_{in\infty}}{R_p} \equiv Kn_\infty .$$

It is important to note that the two expressions for I_C/I_{FM} and Q_C/Q_{FM}, due to a difference in the numerical factors, will yield different values in spite of their analogous dependence on Kn_∞. This suggests that the ion collection and the energy transfer to the particle respond differently, in terms of transitional behavior, to the same undisturbed plasma conditions. This seems one additional reason for analyzing these two processes separately rather than in a single generalized heat conduction approach.

3.2. *Property calculation*

Ion–neutral diffusion coefficient. The first approximation to the ion–neutral binary diffusion coefficient in the Chapman–Enskog approach is given by [15]

$$D_{in} = \frac{3(2\pi k T_h/m_{in})^{1/2}}{16 N_T \Omega^{(1,1)}} .$$

Based on the ion–neutral resonant charge exchange cross section adopted by Devoto [15], $\Omega^{(1,1)}$ can be shown to vary with the heavy species temperature as $\Omega^{(1,1)} \propto T_h^{-0.19}$, and therefore the separate dependence of D_{in} on T_h and N_T is

$$D_{in} \propto \frac{T_h^{0.69}}{N_T} .$$

It should be noted that for all conditions of interest D_{in} is an excellent approximation to the more complex ordinary diffusion coefficient in a three-species system. Comparison with equilibrium properties from Hsu's work [16] also results in excellent agreement (Fig. 1).

Heavy species translational thermal conductivity. From simple kinetic considerations, one expects for the translational heavy species thermal conductivity $K_h \propto v_h(\lambda_i N_i + \lambda_n N_n)$ where

$$\lambda_i N_i \propto \left[\Sigma_{ii} + \frac{N_n}{N_i}\Sigma_{in}\right]^{-1}, \quad \lambda_n N_n \propto \left[\Sigma_{nn} + \frac{N_i}{N_n}\Sigma_{in}\right]^{-1} .$$

It should be noted that Σ_{in} and Σ_{nn} are approximately of the same order of magnitude, whereas, due to the long range nature of the Coulomb potential, Σ_{ii} is substantially larger than Σ_{in}. Considering various regimes of ionization, some conclusions may be drawn about the relative importance of $\lambda_i N_i$ and $\lambda_n N_n$ in the expression for K_h.

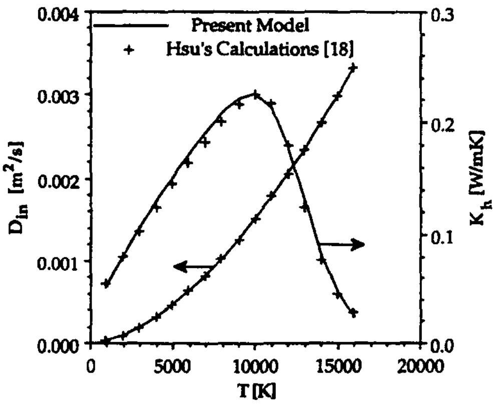

FIG. 1. Ion–neutral diffusion coefficient and heavy species translational thermal conductivity. Comparison between the adopted expressions and rigorous transport property calculations for 1 atm LTE argon plasma.

For $N_i/N_n \ll 1$ (weakly ionized plasma):

$$\Sigma_{ii} \ll N_n\Sigma_{in}/N_i, \quad \Sigma_{nn} \gg N_i\Sigma_{in}/N_n,$$
$$N_n\Sigma_{in}/N_i \gg \Sigma_{nn} \quad \text{and} \quad \lambda_i N_i \ll \lambda_n N_n .$$

For N_i/N_n not small (moderately to strongly ionized plasma):

$$\Sigma_{ii} \gg N_n\Sigma_{in}/N_i \quad \text{and} \quad \lambda_i N_i \ll \lambda_n N_n .$$

So, except for practically fully ionized plasmas ($N_i/N_n \approx \Sigma_{ii}/\Sigma_{in} \gtrsim 10$, $\xi \gtrsim 0.9$), the heavy species translational thermal conductivity is dominated by ion–neutral and neutral–neutral collisions. Adopting the approximation $\Sigma_{in} \approx 5\Sigma_{nn}$, one obtains $\lambda_n N_n \propto [\Omega^{(1,1)}(N_i/N_n + 1/5)]^{-1}$, and K_h can be related to D_{in} via $\Omega^{(1,1)}$. A slight modification of the numerical factors to improve the comparison with available data yields

$$K_h = \frac{4kN_T}{\frac{1}{4} + \frac{N_i}{N_n}} D_{in}$$

a simple expression for the translational thermal conductivity of heavy species, which is believed to be suitable for non-equilibrium conditions since the dependence on temperature and density is kept separate. This expression, when tested vs the equilibrium calculations of Hsu [16] resulted in agreement within 5% of the more accurate values (Fig. 1). The extension to include the fully ionized limit is straightforward, but will not be applied here.

3.3. *Continuum model*

Ion diffusion. Following Hirschfelder *et al.* [17], the appropriate form for the ion diffusion flux $\mathbf{J}_i$ in a monatomic singly ionized plasma at constant total pressure is

$$\mathbf{J}_i = \frac{m_i D_{in}}{\rho}\left[-N_n \nabla N_i + \frac{N_T^2 N_i}{P_{tot}} e\mathbf{E} + N_i \nabla N_n\right]$$

where the electric field E, in the case of a floating particle, is ambipolar in nature. For a floating particle, since $\mathbf{J}_i = \mathbf{J}_e$ and $D_{in} \ll D_e$ one can conclude, by order of magnitude analysis of the electron diffusion equation

$$e\mathbf{E} \approx -kT_e \frac{\nabla N_i}{N_i}$$

to a very good degree of approximation. Therefore

$$\mathbf{J}_i = -\frac{m_i D_{in}}{\rho}\left[\left(N_n + \frac{N_T^2 kT_e}{P_{tot}}\right)\nabla N_i - N_i \nabla N_n\right].$$

The term ∇N_n must be evaluated consistently with the assumption of constant total pressure P_{tot} by means of Dalton's law

$$P_{tot} = N_n kT_h + N_i k(T_h + T_e).$$

It will be assumed here that the electron temperature T_e can be taken as essentially constant, and treated as a parameter of the problem. This assumption is supported by the fact that there seems to be strong evidence, both experimental [18] and computational [6, 19], that substantial deviations from thermal equilibrium should be expected in the vicinity of cold boundaries, even for atmospheric pressure plasmas, with the electron temperature remaining quite high. This should certainly hold for the plasma surrounding relatively small particles. A suitable criterion for electron cooling can be based on the following argument: an electron will remain in the vicinity of the particle (within a typical distance R_p) for a number of collisions of order $Z \approx (R_p/\lambda_e)^2$ (based on a random walk model), and will lose at each collision with a cold ion or neutral a fraction $2m_e/m_i (\approx O(10^{-5}))$ of its energy. Substantial electron cooling will therefore only take place if $Z(2m_e/m_i) \approx O(1)$, that is only in the presence of very large particles.

In the following, the ion density N_i will be scaled with respect to the undisturbed plasma ion density $N_{i\infty}$ as $n \equiv N_i/N_{i\infty}$, the heavy species temperature will be scaled with $T_{e\infty}$ as $\tau \equiv T_h/T_{e\infty}$. The undisturbed plasma will for simplicity be considered in LTE with $T_{h\infty} = T_{e\infty} = T_\infty$. This last assumption may be readily removed, if necessary, without requiring but minor modifications in the equations presented here. Another parameter will be introduced as $C \equiv N_{i\infty}/N_{T\infty}$, related to the undisturbed plasma degree of ionization ξ as $\xi = C/(1-C)$, where $\xi \equiv N_{i\infty}/(N_{i\infty}+N_{n\infty})$ (a fully ionized plasma is then characterized by $C = 0.5$). D_{in} may be expressed in terms of $D_{in\infty}$ as

$$D_{in} = D_{in\infty}\frac{\tau^{0.69}}{N_T/N_{T\infty}}.$$

Scaling the gradient operator on $(R_p)^{-1}$, and manipulating the expression for $\mathbf{J}_i$, the natural scaling for the ion diffusion flux becomes evident as

$$\mathbf{j}_i \equiv \frac{\mathbf{J}_i}{N_{i\infty}D_{in\infty}/R_p} = -\frac{\tau^{0.69}}{fg}[(\tau+g^2)\nabla n + nf\nabla\tau]$$

where f and g are shorthand notation for functions of n and τ defined as

$$f = f(n; C) \equiv 1 - nC;\ g = g(n, \tau; C) \equiv 1 - nC(1-\tau).$$

The conservation equation for the diffusion flux is of the form

$$\nabla\cdot\mathbf{J}_i = \dot{N}_i$$

where the right-hand term represents the net ionization rate per unit volume, which for dominating three-body recombination (ion–electron–electron process described by the coefficient α') can be written as

$$\dot{N}_i = \alpha'(T_e)N_e\left[\left(\frac{N_e^2}{N_n}\right)_{eq} N_n - N_e^2\right].$$

Introducing the scaled variables in the continuity equation one obtains

$$\nabla\cdot\mathbf{j}_i = \mathscr{D}n(s-n^2)$$

where

$$s = s(n, \tau; C) \equiv \frac{1 - Cn(1+\tau)}{\tau(1-2C)}$$

and $\mathscr{D}$ is the Damköhler number defined as

$$\mathscr{D} \equiv \frac{R_p^2}{D_{in\infty}/\alpha' N_{i\infty}} \equiv \left(\frac{R_p}{L_R}\right)^2.$$

Typically, the recombination length calculated on the basis of the Hoffert–Lien [20] recombination coefficient is of the order of a few hundred micrometers ($L_R \sim 10^{-3}$–10^{-4} m) for an atmospheric pressure plasma. When dealing with micrometer-size particles ($R_p \sim 10^{-5}$–10^{-6} m) the Damköhler number is of the order $\mathscr{D} \sim 10^{-2}$–10^{-4}, indicating that frozen conditions tend to prevail in the vicinity of the particle. In Table 1 the recombination length and the Damköhler number for a 100 μm diameter particle in an atmospheric pressure plasma are compiled. It is important to note that, due to the structure of the recombination term, even for $\mathscr{D} \sim 0.1$ near-frozen conditions should be expected. Since approximately $\mathscr{D} \propto P_{tot}$, frozen conditions become even more dominant at subatmospheric pressures.

Assuming therefore that frozen conditions prevail in the plasma around the particle, the continuity equation becomes

$$\nabla\cdot\mathbf{j}_i = 0.$$

Table 1. Characteristic recombination lengths and Damköhler numbers for a 100 μm diameter particle in an atmospheric pressure argon plasma

T_∞ [K]	L_R [m]	$\mathscr{D} = (R_p/L_R)^2$
8000	2.32×10^{-3}	4.6×10^{-4}
10000	5.47×10^{-3}	8.4×10^{-3}
12000	2.35×10^{-3}	4.5×10^{-2}
14000	1.65×10^{-3}	1.1×10^{-1}

In spherical coordinates, scaling the radial coordinate R as $r = R/R_p$, one obtains the ion diffusion equation in integral form as

$$\frac{\tau^{0.69}}{fg}\left[(\tau+g^2)\frac{dn}{dr}+nf\frac{d\tau}{dr}\right]=\frac{j_c}{r^2}$$

where j_c is a scaled current density such that $I_C = j_c(4\pi e R_p N_{i\infty} D_{in\infty})$ (taken positive when collected by the particle). j_c must be determined so as to satisfy the appropriate boundary conditions ($n \to 1$ for $r \to \infty$, and $n = 0$ at $r = 1$).

Heavy species heat conduction. In the heavy species energy equation, the dominant energy transfer mechanism is assumed to be conduction. Collisional coupling with electrons is assumed to be weak (due to the low electron–heavy mass ratio, and to the fact that where N_e is large $(T_e - T_h)$ is small, and vice versa), and will be neglected. The appropriate form of the energy equation under such assumptions reduces to a heat conduction equation, involving the translational thermal conductivity of the heavy species. After scaling the equation, consistently with the ion diffusion equation, and observing that

$$K_h = K_{h\infty}\frac{1+2C}{1-2C}\frac{1-nC(1-3\tau)}{1-nC(1+\tau)}\tau^{0.69}$$

the resulting integral conduction equation becomes

$$\frac{d\tau}{dr}=\frac{h}{\tau^{0.69}}\frac{q_c}{r^2}$$

where

$$h = h(n,\tau;C) \equiv \frac{1-2C}{1+2C}\frac{1-nC(1+\tau)}{1-nC(1-3\tau)}$$

and q_c is a scaled heat flux such that $Q_C = q_c(4\pi R_p K_{h\infty} T_\infty)$. The boundary conditions for τ are $\tau \to 1$ for $r \to \infty$, and $\tau = \tau_p = T_p/T_\infty$ at $r = 1$.

To sum up, the governing equations and boundary conditions for the coupled problem of ion diffusion and heavy species heat conduction are

$$\frac{\tau^{0.69}}{fg}\left[(\tau+g^2)\frac{dn}{dr}+nf\frac{d\tau}{dr}\right]=\frac{j_c}{r^2}$$

$$f=f(n;C)\equiv 1-nC;\quad g=g(n,\tau;C)\equiv 1-nC(1-\tau)$$

$$\frac{d\tau}{dr}=\frac{h}{\tau^{0.69}}\frac{q_c}{r^2}$$

$$h = h(n,\tau;C) \equiv \frac{1-2C}{1+2C}\frac{1-nC(1+\tau)}{1-nC(1-3\tau)}$$

$$n\to 1 \quad \text{for} \quad r\to\infty, \quad \text{and} \quad n=0 \quad \text{at} \quad r=1$$

$$\tau\to 1 \quad \text{for} \quad r\to\infty, \quad \text{and} \quad \tau=\tau_p=T_p/T_\infty \quad \text{at} \quad r=1.$$

The solution to this non-linear set of coupled ordinary differential equations can be readily obtained by a Runge–Kutta algorithm, together with a shooting method for two-point boundary value problems. The solution of the continuum problem in terms of the scaled variables will yield j_c and q_c as the appropriate constants of integration which ensure satisfaction of the boundary conditions. j_c and q_c are functions only of the parameters C (related to the degree of ionization in the undisturbed plasma) and τ_p (the ratio of the particle surface temperature to that of the plasma). In particular, q_c can be analytically approximated as a function of C and τ_p to within 3% of the numerical solution by the expression

$$q_c = (1-1.73C)\frac{1+2C}{1-2C}\frac{1-\tau_p^{1.69}}{1.69}.$$

The group $q_c/(1-\tau_p)$ may also be interpreted as a conventional Nusselt number based on the particle radius ($Nu_r \equiv h_c R_p/K_{h\infty}$) for heavy species translational conduction. In the limit $C \to 0$, and $\tau_p \to 1$ (small temperature differences), $q_c \to 1$ and $Nu_r \to 1$ as expected. j_c is not amenable to a simple analytical description, although an attempt to collapse the results onto a single curve is presented in Fig. 2. The specific plasma conditions and particle size (T_∞, P_{tot}, R_p) enter the solution only in the redimensionalization of j_c and q_c to J_C and Q_C. It should however be noted that neglecting the term $\lambda_i N_i$ with respect to $\lambda_n N_n$ in the approximation for K_h leads to the non-physical situation of $K_h \to 0$ in the fully ionized limit. The model is therefore not expected to yield accurate results for $\xi \gtrsim 0.9$, or $C \gtrsim 0.47$.

3.4. *Knudsen number expressions*

Plasma–particle Knudsen number. The Knudsen number Kn_∞ based on the undisturbed plasma properties and on the particle radius will be defined as

$$Kn_\infty \equiv \frac{D_{in\infty}}{\left(\frac{kT_\infty}{m_i}\right)^{1/2} R_p}.$$

Assuming an undisturbed plasma in LTE, the explicit dependence of Kn_∞ on T_∞ and P_{tot} is $Kn_\infty \propto T^{1.19}/P_{tot}$.

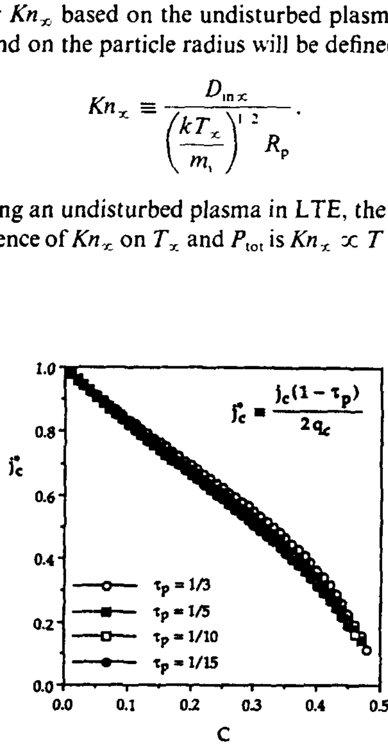

FIG. 2. Rescaled dimensionless ion flux at the particle surface in the continuum limit.

Numerically

$$Kn_\infty \cong \frac{1.045}{R_p} \frac{(T_\infty/10^4)^{1.19}}{P_{tot}}$$

where T_∞ is in K, R_p in μm, and P_{tot} in atm.

Ion diffusion Knudsen number. The effective Knudsen number for ion current collection was defined as $Kn_j = I_C/I_{FM}$. From the solution of the continuum problem, I_C (the current collected in the continuum limit) can be calculated as

$$I_C = j_c(eN_{i\infty}D_{in\infty}/R_p)A.$$

In the free-molecular limit Laframboise's theory for electrostatic probes in the collisionless regime [21] yields, in the analytical representation due to Kiel [22]

$$I_{FM} = \alpha_{LF}(N_{i\infty}v_{i\infty}/4)A$$

where α_{LF} is a coefficient that depends on the ratio of ion and electron temperature in the undisturbed plasma. For $(T_h/T_e)_\infty = 1$, and for a thin sheath, $\alpha_{LF} = 0.72$ [22]. By taking the ratio of I_C to I_{FM}, one obtains

$$Kn_j = 2.50\frac{j_c}{\alpha_{LF}}Kn_\infty.$$

Conduction heat transfer Knudsen number. The Knudsen number for the heat transfer rate by conduction was defined as $Kn_q = Q_C/Q_{FM}$. From the solution of the continuum problem, Q_C can be calculated as

$$Q_C = q_c(K_{h\infty}T_\infty/R_p)A.$$

In the free-molecular limit, Q_{FM} can be obtained from kinetic theory as

$$Q_{FM} = \alpha_{TA}[(N_{n\infty}+\alpha_{LF}N_{i\infty})v_{h\infty}/4][2k(T_\infty - T_p)]A$$

where α_{TA} is the thermal accommodation coefficient associated with the particle surface. The importance of α_{TA} is a matter that should not be passed over lightly: its effects may be substantial, and little more than guessing 'reasonable' values can at this stage be done. From here on α_{TA} will be assumed to be 1: this is of course a limiting case, and one could expect it to be realistic only for particles of very irregular shape, or high degree of porosity. However, here, the choice is made to avoid introducing further effects that would increase the effective Kn_q.

Introducing the expression for $K_{h\infty}$

$$K_{h\infty} = \frac{16kN_{T\infty}}{\frac{1+2C}{1-2C}}D_{in\infty}$$

and observing that $(N_{n\infty}+\alpha_{LF}N_{i\infty}) = [1-(2-\alpha_{LF})C]N_{T\infty}$, one obtains for the effective Knudsen number Kn_q

$$Kn_q = 20.0\frac{(1-2C)/(1+2C)}{1-(2-\alpha_{LF})C}\frac{q_c}{(1-\tau_p)}Kn_\infty.$$

For $\alpha_{TA} < 1$, the effective Kn_q would simply be increased by a factor of $1/\alpha_{TA}$. It is important to note here that although Kn_j and Kn_q are different, they are clearly related to a single parameter of the undisturbed plasma, that is Kn_∞. Their exact values depend on the solution of the continuum problem, but since j_c and $(1-2C)q_c/(1+2C)$ (the latter being a scaled temperature gradient at the particle surface) should both be of the order unity, Kn_∞ and the above formulae give a simple and rather accurate estimate of the effects of the deviation from continuum on ion diffusion and heavy species heat conduction.

3.5. *Energy transfer rates*

The total energy transferred to the particle from the surrounding plasma can be computed on the basis of the calculated I (ion current collected by the particle) and Q (translational energy transport by the heavy species). The contribution due to Q is straightforward. The diffusion current also contributes to the overall energy transfer: ions carry ionization energy that is released upon neutralization on the particle surface (assumed catalytic), and are accelerated across the electron retarding sheath immediately adjacent to the particle surface. Moreover, due to the fact that the particle obviously collects no net electrical current, there will also be an electron current, equal in magnitude but carrying opposite charge, reaching the surface and contributing an amount of energy proportional to I and to the electron temperature. For a floating particle the surface work function is irrelevant to the calculation of the overall heat transfer rate, since ions and electrons contribute terms of equal magnitude and opposite sign. The expression for the overall heat flux to the particle should then read

$$\underset{\text{Total}}{Q_{tot}} = \underbrace{\underset{\substack{\text{Heavy species}\\ \text{translational}\\ \text{energy transport}}}{Q}}_{\equiv Q_{cond}} \underbrace{+\underset{\substack{\text{Ionization}\\ \text{energy}\\ \text{transport}}}{I\cdot\Phi_{ioniz}} + \underset{\substack{\text{Sheath potential}\\ \text{contribution}}}{I\cdot(-\Delta\Phi_{sheath})}}_{\equiv Q_{rec}} + \underbrace{\underset{\substack{\text{Electron}\\ \text{translational}\\ \text{energy transport}}}{I\cdot 2kT_\infty/e}}_{\equiv Q_e}.$$

For argon, $\Phi_{ioniz} \approx 15.76$ V. The contribution due to the sheath potential drop (essentially a particle charging effect due to the high electron mobility) is not so clearly defined. However, in the fully collisionless case, for a thin sheath around a floating particle, according to Laframboise's analysis

$$\Delta\Phi_{sheath} \approx -5.9(kT_\infty/e).$$

In the continuum limit, Bohm's criterion [23] can be expected to hold in some form, with a 'presheath' existing in front of the collisionless thin sheath. In this case, the sheath plus presheath potential drop can be estimated as

$$\Delta\Phi_{\rm sheath} \approx -5.2(kT_{\infty}/e).$$

In view of the dominance of the ionization energy transport term, the difference between the two values for the sheath potential drop will be neglected, and the first of the two will be employed throughout.

4. RESULTS AND DISCUSSION

The calculation of the plasma–particle energy transfer rates hinges on the determination of the nondimensional parameters j_c and q_c from the continuum solution. From these parameters, the overall energy transfer in the continuum limit, and the Knudsen numbers for ion diffusion (Kn_i) and heat conduction (Kn_q), which describes the deviations from that limit, may be obtained. In the adopted model, j_c and q_c depend only on the degree of ionization of the undisturbed plasma and on the particle-to-plasma temperature ratio $\tau_p \equiv T_p/T_{\infty}$. The dependence of the continuum limit solution on τ_p is generally weak, and can be analytically reduced (collapsing the curves completely in the case of q_c and at least partially in the case of j_c). This result seems particularly interesting in view of the drastic simplification it would allow in the overall plasma–particle coupling problem. In the following discussion, no further mention of the effects of τ_p on the heat transfer rates will be made other than

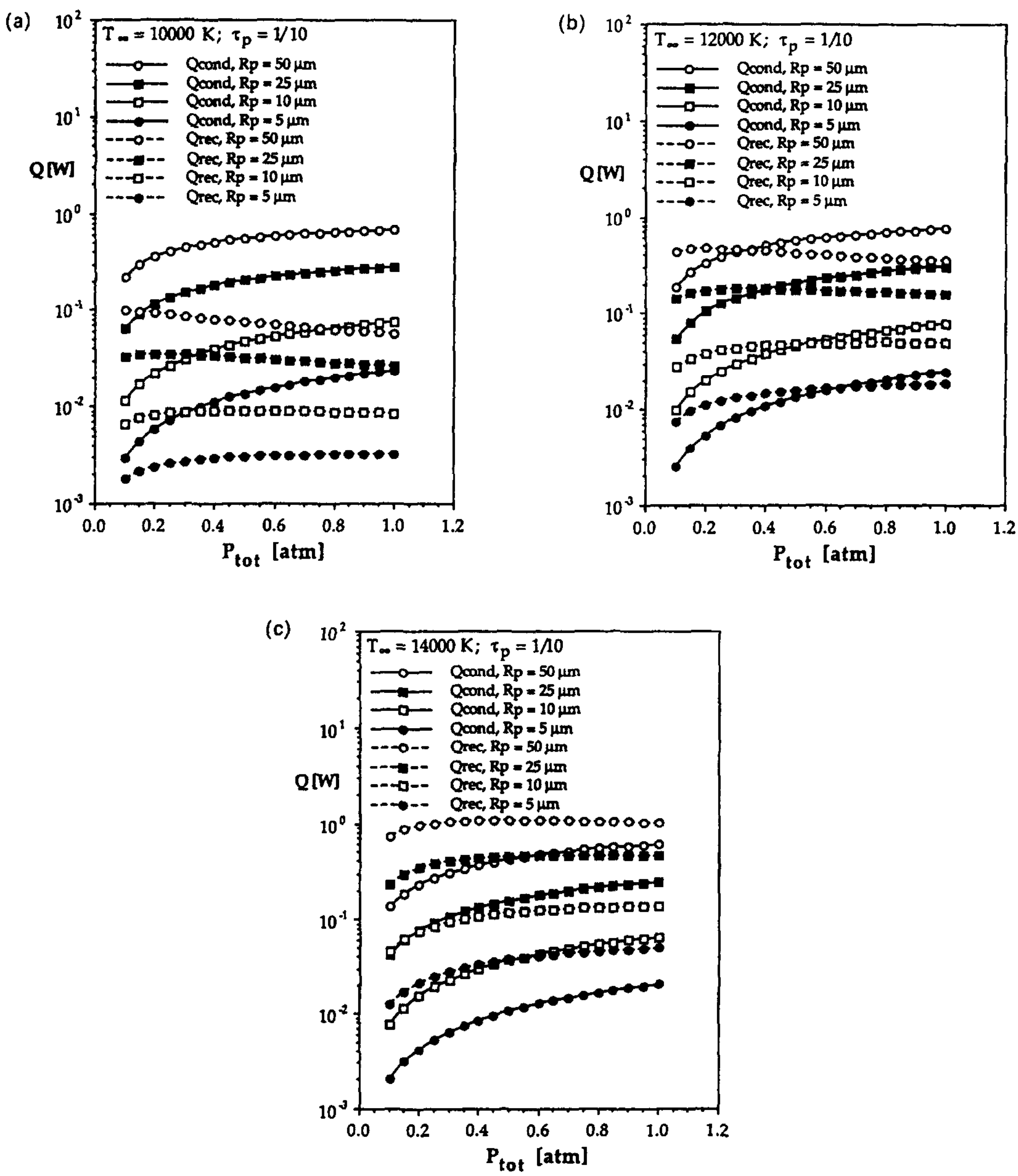

FIG. 3. Heavy species conduction ($Q_{\rm cond}$) and ion current energy transport ($Q_{\rm rec}$) to particles of radius R_p vs total pressure for undisturbed plasma at (a) 10 000 K, (b) 12 000 K, (c) 14 000 K.

reminding the reader that in the free-molecular limit the translational energy exchange is proportional to $T_\infty(1-\tau_p)$.

The results will be presented in the form of separate contributions to the energy transfer rates from Q_{rec} and from Q_{cond}. The calculations were carried out for pressures P_{tot} in the range 0.1–1 atm, for plasma temperatures T_∞ up to 15 000 K, and for spherical particles of radius R_p of 50, 25, 10 and 5 μm. This choice of parameters, in addition to its relevance to applications, is consistent with the primary objective of this work, that is the description of the heat transfer in the transitional regime between the continuum and free-molecular limits. The deviations from either limit will be presented in the form of the specific Knudsen numbers Kn_j and Kn_q, from which the quantities of interest may be obtained as

$$\frac{Q}{Q_{FM}} = \frac{1}{1+Kn_q^{-1}}; \quad \frac{Q}{Q_C} = \frac{1}{1+Kn_q}$$

$$\frac{I}{I_{FM}} = \frac{1}{1+Kn_j^{-1}}; \quad \frac{I}{I_C} = \frac{1}{1+Kn_j}.$$

First, the dependence of Q_{cond} and Q_{rec} on pressure, with the particle radius as a parameter, and for three plasma temperatures will be presented. At 10 000 K (Fig. 3(a)), Q_{cond} dominates over Q_{rec}, whereas at 12 000 K (Fig. 3(b)) a cross-over between the two effects occurs depending on pressure and particle radius. At 14 000 K (Fig. 3(c)) Q_{rec} dominates the heat transfer, independently of particle radius and pressure. This is due to the substantial increase in charged species density over this temperature range. The dependence of Q_{rec} on pressure shows an inversion in trend with different particle radius. In particular, for near-continuum situations (low T_∞, high P_{tot}, large R_p, and resulting small $Kn_\infty \propto T_\infty^{1.19}/(P_{tot}R_p)$), Q_{rec} increases with decreasing pressure. This behavior is attributable to the fact that the ion current reaches the particle surface by diffusion, and the larger diffusion coefficient and higher degree of ionization associated with lower pressures initially overcompensate the 'thinning' of the plasma, which, eventually, for further decreases in pressures, dominates, limiting the contribution of Q_{rec}. In terms of the dependence on the particle radius, both Q_{rec} and Q_{cond} are larger for larger particle size. However, the increase is not proportional to the increase in the surface area available for heat transfer: indeed, the energy transfer rates per unit area (Fig. 4) show that the fluxes are higher for smaller particles. This effect is consistently observed for all conditions, and is more pronounced as continuum is approached (in fact, in the free-molecular limit, the fluxes become independent of the particle size). The physical reason for this to happen is that, for given plasma temperature and pressure conditions, smaller particles are characterized by larger Knudsen numbers, and therefore their conditions lie closer to the free-molecular limit, which represents the upper limit in terms of possible plasma–particle energy exchange. This effect is expected to be important for the optimization of particle size vs expected residence times towards the most efficient particle heating. When the energy flux plots vs temperature are considered (for three different pressures, 0.1, 0.5, and 1 atm, Figs. 5(a)–(c), respectively), the striking feature is the existence of a temperature range, approximately 2000–3000 K wide, where a change in the dominant energy transfer mechanism occurs, namely from ordinary conduction by the heavy species to recombination and sheath energy release due to the ion flow to the particle, for increasing temperatures. Lowering the pressure causes the crossover to take place at a lower temperature, as a result of the increasing degree of ionization.

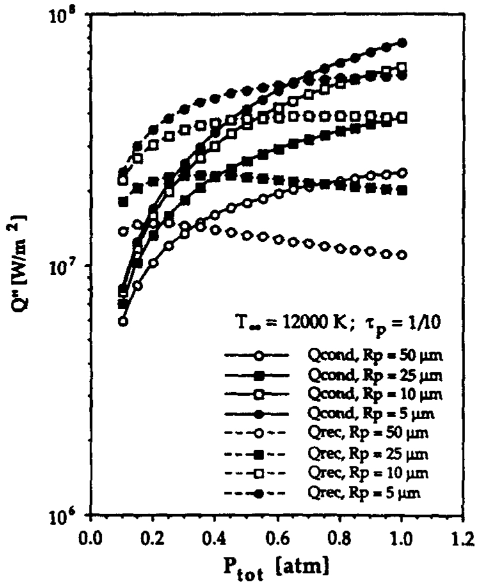

FIG. 4. Heavy species conduction (Q_{cond}) and ion current energy transport (Q_{rec}) per unit to particles of radius R_p vs total pressure for undisturbed plasma at 12 000 K.

As far as the dependence on pressure of the specific Knudsen numbers Kn_j and Kn_q is concerned, Fig. 6 shows the wide spectrum of conditions swept by the present choice of parameters, with rarefaction generally more heavily affecting conduction than ion diffusion. It is important to note how most of the conditions are characterized by Knudsen numbers well above 0.1, indicating that the deviations from the continuum limit are generally larger than 10%, and indeed it appears that at low pressures, or for small particle sizes, one should rather worry about deviations from the free-molecular limit than from continuum. This is somewhat in disagreement with previous analyses, as can be seen from Fig. 7 where the results from the present model are compared with those from refs. [1, 8, 9], for the specific case of an atmospheric pressure plasma at 15 000 K. It must be pointed out that a direct comparison of the absolute values for the energy transfer rates is not possible, since none of the mentioned references reports the

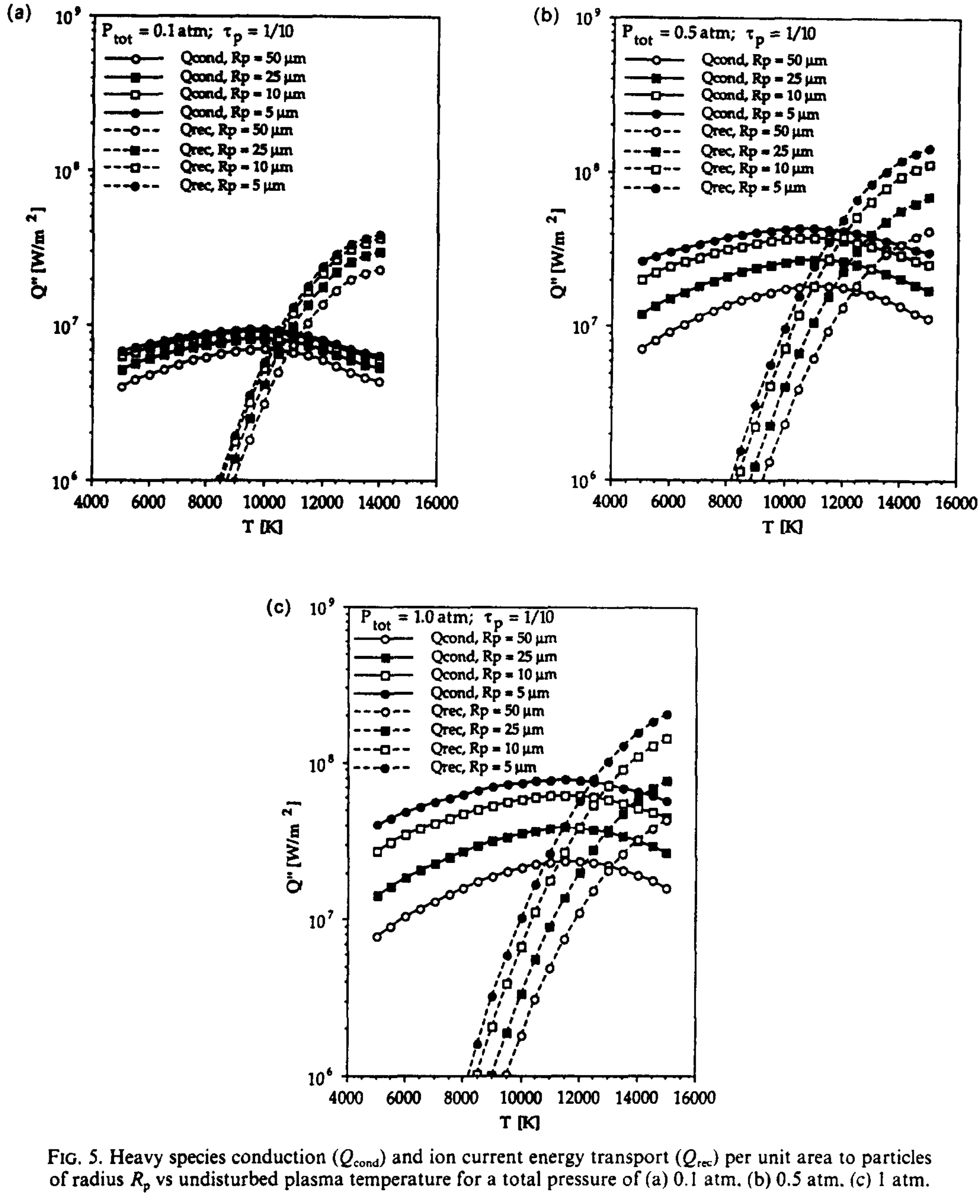

FIG. 5. Heavy species conduction (Q_{cond}) and ion current energy transport (Q_{rec}) per unit area to particles of radius R_p vs undisturbed plasma temperature for a total pressure of (a) 0.1 atm, (b) 0.5 atm, (c) 1 atm.

calculated values for the continuum limit, but only the deviation from such limits. Discrepancies in the values for Q_c'' may therefore be partly responsible for such differences. For the purpose of a meaningful comparison with Chen and Pfender's results [1], obtained by a jump method, the results of the equilibrium version of the present model are also presented. Two sets of property values were used (from ref. [16], indicated as LTE1, and from ref. [9], indicated as LTE2), that should bracket the values used in ref. [1]. It is clear that, with respect to the present model, the jump approach substantially underestimates the Knudsen effect, and, indeed, for particle radii less than about 20 μm, predicts energy fluxes that exceed the free-molecular limit values (represented by the straight lines labeled as Q_{FM}''/Q_C''). It is interesting to note that, however, the jump approach appears to have the ability to attain an asymptotic free-molecular behavior for small particle sizes. The problem with it seems to be the systematic underprediction of the relevant Knudsen number, due to the strong bias which the initial assumption of small deviation from continuum puts on the property evaluation. As far as the present model is concerned, it is important to note that the particular conditions considered in Fig. 7 correspond to a situation where the energy transfer rates are dominated by the recombination term, and therefore the overall deviation

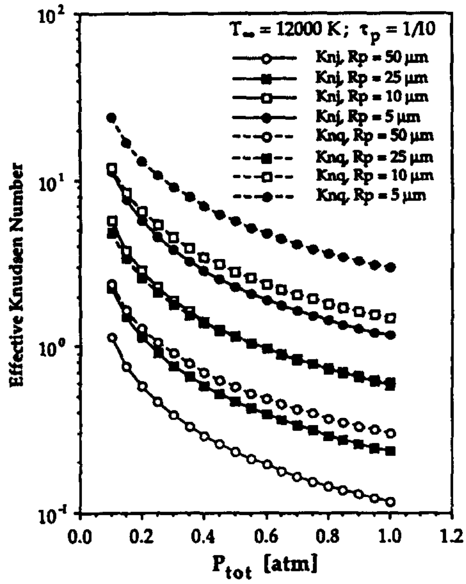

FIG. 6. Effective Knudsen numbers for ion diffusion (Kn_j) and for conduction (Kn_q) for particles of radius R_p vs total pressure for undisturbed plasma at 12 000 K.

from continuum is strongly dependent on the specific Knudsen number Kn_j for ion diffusion and only weakly on Kn_q. The situation is different at somewhat lower temperatures, where the two contributions are comparable, with a resulting 'hybrid' interpolation curve, affected simultaneously by Kn_j and Kn_q. As for Lee's and Chyou's models [8, 9], where the jump approach is abandoned in favor of the limiting sphere concept, problems arise mainly in the recovery of a free-molecular behavior. This is probably due to the fact that the continuum–free molecular interface is positioned at the sheath edge, a few Debye lengths away from the particle surface. Since the Debye length is substantially shorter than the ion–neutral mean free path, this results in a simple increase in the effective particle size seen by the continuum solution, without a real description of rarefaction effects. The model evidently computes the Debye length on the basis of the continuum solution, approaching the particle from the undisturbed plasma, therefore linking the Debye length to the particle size. The result is that Q''/Q''_C for Lee and for Chyou actually represents a ratio between two continuum energy transfer rates for particles of different radii that, however, vary in the same functional way. This, for small particle sizes, gives rise to a constant Q''/Q''_C ratio. The model may account for some changes in the temperature boundary condition for the continuum problem as the particle size is reduced, but, as the present model clearly indicates, these should only weakly affect the solution.

As a final remark, it is interesting to note that non-equilibrium reduces the particle size induced rarefaction effects on the plasma–particle heat transfer rates, at least for an atmospheric plasma at 15 000 K. This is probably attributable to the fact that the recombination energy transfer, which dominates the heat transfer under these conditions, is somewhat less sensitive to the Knudsen effect than conduction, as can be seen from Fig. 6.

5. CONCLUSIONS

Independently of the model adopted to describe the phenomenon, it is evident that, for the set of conditions considered, the energy fluxes based on the continuum approach are sufficiently close in magnitude to their counterpart in the free-molecular limit that methods based on the assumption of small perturbations from either limit appear inadequate. In such translational regions of the Knudsen number range, Sherman's interpolation formula for conduction heat transfer and its analogous formula (due to Thornton) in electrostatic probe theory seem to

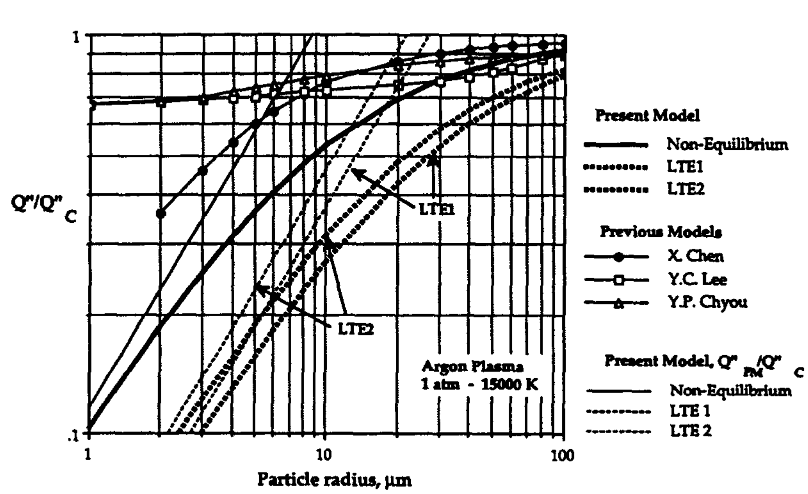

FIG. 7. Effect of particle size on the deviation from continuum total energy transfer fluxes. The data for previous models are taken from ref. [9].

offer a physically sound and computationally advantageous approach to plasma–particle heat transfer problems. Non-equilibrium effects may be included in a simple way, as long as the plasma properties are consistently evaluated.

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UNE APPROCHE UNIFIEE DU TRANSFERT THERMIQUE PLASMA–PARTICULE DANS DES CONDITIONS DE NON-CONTINUITE ET DE NON-EQUILIBRE

Résumé—Un modèle pour le transfert thermique d'un plasma infini à une particule est développé de façon à inclure les effets de raréfaction sur le domaine complet des nombres de Knudsen, du non-équilibre thermique et chimique au voisinage de la surface de la particule. Le flux global de chaleur est obtenu en déterminant séparément les contributions de la conduction des espèces lourdes et du transport d'énergie par l'écoulement ionique à la surface de la particule. Les effets de Knudsen sont pris en compte au moyen de la formule d'interpolation de Sherman pour le problème de conduction et son équivalent dans la théorie électrostatique pour le transport ionique à la surface. Les résultats sont présentés pour des plasmas d'argon dans le domaine de pression 0,1–1 atm et pour des tailles de particules dans le domaine 10–100 μm.

EINE EINHEITLICHE NÄHERUNG FÜR DEN WÄRMEÜBERGANG ZWISCHEN PLASMA UND PARTIKELN UNTER NICHTKONTINUUMS- UND NICHTGLEICHGEWICHTSBEDINGUNGEN

Zusammenfassung—Es wird ein Modell für den Wärmeübergang von einem ruhenden unendlich ausgedehnten Plasma an ein Partikel entwickelt. Dabei werden die Einflüsse einer starken Verdünnung über den gesamten Bereich der Knudsen-Zahlen berücksichtigt, ebenso die thermischen und chemischen Nichtgleichgewichtsbedingungen in der Nähe der Partikeloberfläche. Der Gesamtwärmetransport wird durch getrennte Bestimmung der Einzelbeiträge ermittelt: Wärmeleitung und zusätzlicher Energietransport durch Ionenströmung zur Partikeloberfläche. Knudsen-Effekte werden mit Hilfe der Interpolationsgleichung nach Sherman für das Wärmeleitungsproblem berücksichtigt, für den Ionentransport wird das entsprechende elektrostatische Analogon herangezogen. Es werden Ergebnisse für ein Argonplasma im Druckbereich 0,1–1 atm und für Partikelgrößen im Bereich 10 bis 100 μm vorgestellt.

ЕДИНЫЙ ПОДХОД К ТЕПЛОПЕРЕНОСУ МЕЖДУ ПЛАЗМОЙ И ЧАСТИЦЕЙ ПРИ ОТСУТСТВИИ СПЛОШНОСТИ И РАВНОВЕСИЯ

Аннотация—Разработана модель теплопереноса от неподвижной бесконечной плазмы к частице для учета эффектов разрежения во всем диапазоне значений числа Кнудсена, а также теплового и химического неравновесия вблизи поверхности частицы. Скорость суммарного теплопереноса получена методом раздельного определения вкладов за счет теплопроводности тяжелых частиц и переноса энергии ионным потоком к поверхности частицы. Кнудсеновский эффект учитывается интерполяционной формулой Шермана для задачи теплопроводности и ее эквивалента в электростатике применительно к переносу ионов к поверхности. Представлены результаты для аргонной плазмы при давлении 0,1–1 атм и размерах частиц 10–100 мкм.