Modeling of clusters deposition under the effect of thermophoresis during thermal plasma flash evaporation process

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Received 15 December 2000; accepted 11 January 2001

Abstract

In the thermal plasma flash evaporation process, due to the high quenching rate of the impinging plasma jet and the steep temperature gradient in front of the substrate, thermophoretic force is expected to have a significant effect on the cluster transportation process. This paper aims at investigating the cluster transportation and deposition processes by the method of numerical simulation and presents the qualitative description of the effects of thermophoresis on the deposition efficiency. Eulerian approach is employed in this research and the corresponding convection–diffusion governing equation is established for the cluster concentration field. The environment pressure for the plasma jet is set to be 50 Torr and the working gas is argon plasma. The velocity and temperature fields are simulated firstly and then the cluster transportation process is modeled within the established flow field but the computational domain is only limited to a narrow region in front of the substrate. It is assumed that clusters of uniform size are generated in a certain region within the boundary layer and it is treated as the source term in the cluster transportation equation. The results of cluster concentration field and the radial distribution of deposition flux are achieved for the clusters in the size range 1–10 nm, respectively. Results are also given for the comparative cases without considering thermophoresis effects. It is found that the concentration boundary layer is significantly suppressed by the thermophoretical force, and the effect of thermophoresis plays a dominant role than that of diffusion thus almost uniform deposition efficiency is achieved for clusters of different sizes. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Clusters; Thermophoresis effect; Modeling; Plasma; Deposition

1. Introduction

For small aerosol particles suspended in a gas where a temperature gradient exists, the particles will experience a force in the direction opposite to that of the temperature gradient. In literature, this phenomenon is called thermophoresis effect and the corresponding force is called thermophoretic force. In engineering practices and scientific research, thermophoresis effect can be found in many processes and applications, thus has gained great interest from researchers in recent decades. Some typical cases have been investigated experimentally or numerically, such as particle contaminations on the wafer during the semiconductor process [1–3], fabrication of optical fibers by modified chemical vapor deposition [4–7] and other CVD processes [8], particle deposition onto the blade surface of gas turbine [9,10], fouling of heat exchanger pipes [11], gas cleaning and filtration process, soot collecting and thermal precipitation [12–18], and thermal spraying process [19]. Among these occasions, thermophoresis effects often affect particle deposition and transportation processes jointly with other mechanisms including diffusion, convection, sedimentation and electrophoresis, and in some cases it might play a dominant role.

On the other side, as it is known, the gas temperature of thermal plasma is always in the range of \(10^4\) K. As the result, the temperature gradient caused by such a high temperature gas in a typical thermal plasma reactor will be several orders higher than that of above-mentioned cases, where the temperature difference is always in the order of \(10^1–10^3\) K. In the process of the thermal plasma flash evaporation [20], due to the high quenching rate of the plasma jet when impinging upon the substrate, the vapor of the feedstock materials will be in the state of supersaturation thus clusters or ultrafine particles will be generated within the boundary layer. Because the steep temperature gradient in front of the substrate (\(10^9–10^{10} \text{ K m}^{-1}\)), thermophoretic force is expected to have a significant effect on the cluster transportation process and thus will influence the deposition efficiency prominently. And also due to the ionization state
of thermal plasma gases, it is believed that thermophoresis effect will involve some unique and complex mechanisms in this situation. In this aspect, Refs. [21–24] have derived the theoretical formulas for the thermophoretic force exerted on a single spherical particle in a rarefied plasma flow. But concerning the overall effect of thermophoresis on the clusters behavior during solid films deposition, there are few relevant researches available in literature up till now.

In the present work, the method of numerical simulation is used to investigate the effect of thermophoresis on clusters deposition during thermal plasma flash evaporation process. It is anticipated that not only the physical trends can be revealed but also the semi-quantitative description can be provided to some extent. It is helpful for developing more understandings and insights into the complex phenomena of clusters deposition under the thermal plasma conditions.

2. Particle transport model and the simulation method

2.1. Theoretical background

For the thermophoresis phenomenon in ordinary un-ionized gases, there are a few theoretical papers in literature, which provided analytical formulas for the thermophoretic force acting on small spherical particles. Among them, Ref. [25] is widely accepted and the same expression is employed in the present research

\[ F_T = -3 \pi \mu d_p \nu K_T \frac{\nabla T}{T} \]  

(1)

where \( d_p \) is the diameter of the particle, \( \mu \) the gas viscosity, \( \nu = \mu / \rho, \rho \) and \( T \) are the density and temperature of gas. The thermophoretic coefficient \( K_T \) is expressed as:

\[ K_T = \frac{2 C(Kn) C_t (k_g/k_p) + C_t K_n}{(1 + 3 C_m K_n ) [1 + 2 (k_g/k_p) + 2 C_t K_n]} \]  

(2)

where \( C_t, C_t \) and \( C_m \) are the thermal creep, temperature jump and velocity jump coefficients, respectively. The currently accepted values of these coefficients are \( C_t = 1.147, C_t = 2.20 \) and \( C_t = 1.146 \) [10]. \( K_n \) is Knudsen number and is defined as \( K_n = \lambda / d_p \) and \( \lambda \) is the mean free path of the gas molecule. \( C(Kn) \) is the slip correction factor and is given as

\[ C(Kn) = 1 + Kn \left[ 1.257 + 0.4 \exp \left(-\frac{1.1}{Kn} \right) \right] \]  

(3)

In most situations, the thermophoretic coefficient \( K_T \) is usually varying in the range from 0.2 to 1.2. From Ref. [25], it is believed that the fitting formula of Eq. (2) is applicable in the \( K_n \) range spanning the free-molecular to the continuum regime. For thermal plasma flash evaporation process, if assuming the temperature of 3000 K for the gases within the boundary layer and 200 Torr as the chamber pressure, then for argon plasma the typical mean free path can be estimated as follows:

\[ \lambda = \frac{2 \mu}{\nu} (\text{nm}) = \frac{2 \mu}{[8 k_B T/(m^2 \text{nm})]^{1/2} (\text{nm})} = 4.8 \mu \text{m} \]  

(4)

From the corresponding experimental measurement, the clusters generated by thermal plasma flash evaporation processes was estimated to be in the size range of 1–9 nm [26]. So it can be confirmed that the cluster will be in the free-molecular regime. In this case, it is found that \( K_T \) will be independent of the particle size and composition, and Eq. (2) will give the constant value of 0.55, just as the conclusions derived by Ref. [1]. So the cluster transportation behavior is believed to be irrelevant with the cluster materials in the thermal plasma flash evaporation process.

In many applications, the theoretical expression for the thermophoretic force is always transformed into another more useful and convenient physical quantity of thermophoretic velocity, \( \tilde{V}_T \), which is gotten under the assumptions of the balance between the drag force and thermophoretic force when the inertia force can be neglected. It is expressed as

\[ \tilde{V}_T = -K_T \frac{\mu}{\rho} \frac{\nabla T}{T} \]  

(5)

2.2. Simulation method

Concerning the simulations of particle transportation phenomena under the effect of thermophoresis, there are basically two different methods. The first is the particle tracking method using Lagrangian approach, which treats trajectories of individual particles. In the present simulation, however, Eulerian approach is employed. It views the aerosol particle system as a continuum phase thus treats them as a species concentration field. In this case, the particle number density conservation equation can be expressed as

\[ \nabla \cdot (n \tilde{V}) = \nabla \cdot (D \nabla n) - \nabla \cdot (n \tilde{V}_T) + S_{\text{cluster}} \]  

(6)

where \( n \) is the cluster/particle number density, \( \tilde{V}(u, \nu) \) the convection velocity, and \( D \) is called Stokes–Einstein diffusion coefficient and expressed by the following formula:

\[ D = \frac{k_B T C(Kn)}{3 \pi \mu d_p} \]  

(7)

where \( k_B \) is Boltzmann constant. The four terms appearing in Eq. (6) are terms due to convection transport, diffusion transport, thermophoretic transport and the cluster/particle generation source, respectively.

For simplicity, in such simulations it is always assumed that the particle concentration is dilute so that the effect of cluster/particle distribution on the gas flow, fluid thermodynamic and transportation properties can be neglected. The decoupling between fluid flow and particle transportation allows fluid velocity and temperature fields to be calculated independently at the first step. Then the aerosol particle transportation and deposition process will be simulated.
Table 1
Parameters for the impinging thermal plasma jet

| Parameter                      | Value          |
|-------------------------------|----------------|
| Plasma gas: argon             |                |
| Chamber pressure: $p_{chamber}$| 50 Torr        |
| Torch exit radius: $R_0$      | 0.02 m         |
| Standoff distance: $L_o$      | 0.27 m         |
| Substrate temperature: $T_{sub}$ | 800 K         |

within the already established temperature and velocity field. It should be noted that in front of the substrate, the plasma temperature is only in the range of 1000–3000 K, so that the ionization degree is negligible and the non-equilibrium phenomena will not affect the electrically neutral condition inside the boundary layer significantly. So in the first approximation, the local thermodynamic equilibrium (LTE) model will be used in the calculation and clusters are assumed to be electrically neutral.

In Table 1, the assumed parameters are listed for the thermal plasma jet ejected from a hybrid thermal plasma torch. The velocity and temperature profiles at the torch exit cross-section are assumed as (Swirling velocity is omitted in the first approximation):

$$u(r) = u_{max}[1 - (r/R_0)^3]$$  

$$T(r) = (T_{max} - T_w)[1 - (r/R_0)^3] + T_w$$  

where it is postulated that $u_{max} = 50$ m s$^{-1}$, $T_{max} = 6000$ K and $T_w = 300$ K. Thus the total mass flow rate of argon gas and the total net power in the jet can be calculated

$$m = 2.9 \times 10^{-4} \text{ kg s}^{-1} (-10.7 \text{ STP l min}^{-1})$$  

$$P_{jet} = 700 \text{ W}$$

The same set of conservation equations as Ref. [27] is employed for the modeling of thermal plasma jet in laminar flow regime. SIMPLE program [28] is used as the algorithm to solve them. The 0.27 m(z) × 0.1 m(r) computational domain is divided by the 122(z) × 82(r) grid nodes. The achieved temperature field for the plasma jet is shown in Figs. 1 and 2 gives the variations of the temperature and temperature gradient along axis just before the substrate surface.

After the velocity and temperature field are obtained for

![Fig. 1. Contours of the temperature distribution of the plasma jet (unit: 10³ K).](image1)

![Fig. 2. The variation of temperature and temperature gradient along the axis in front of the substrate.](image2)
the whole computational domain, the cluster number density conservation equation is only solved in a narrow region of 0.005 m(z) x 0.1 m(r) just before the substrate. The corresponding computational mesh is 52(z) x 82(r) in this region. This equation is solved using SIMPLE algorithm too, but a transformation is adopted to change it to the standard convection–diffusion form required by SIMPLE algorithm

\[(\vec{V} + \vec{V}_T) \nabla n = \nabla (D \nabla n) - n \nabla (\vec{V} + \vec{V}_T) + S_{\text{cluster}}\] (12)

In z–r coordinates, this equation can be rewritten as

\[
(u + u_T) \frac{\partial n}{\partial z} + (v + v_T) \frac{\partial n}{\partial r} = \frac{\partial}{\partial z} \left( \frac{D}{r} \frac{\partial n}{\partial z} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( \frac{D}{r^2} \frac{\partial n}{\partial r} \right)
\]

\[
-n \left[ \frac{\partial (u + u_T)}{\partial z} + \frac{\partial (v + v_T)}{\partial r} + \frac{v + v_T}{r} \right] + S_{\text{cluster}}\] (13)

2.3. Assumptions of the source term and boundary conditions

Another unsolved problem is the cluster generation source term \(S_{\text{cluster}}\). In literature, there are no available simulation results of the cluster nucleation and growth in such drastic quenching process by thermal plasma impinging jet. Modeling research for this phenomenon is another much more challenging task which is not intended to be involved in the present work. For simplicity, it is assumed that in the range of 0.003 m < l < 0.004 m (l is the distance to the substrate surface), the cluster generating rate per unit volume is assumed to follow this function for clusters of uniform size

\[S_{\text{cluster}} = \dot{n}_{\text{cluster}} = 10^{14} \exp(-r/0.01) \quad (s^{-1} m^{-3})\] (14)

By this simplified method, it is believed that the effect of the thermophoretic force on the clusters deposition during the thermal plasma evaporation process could be revealed qualitatively.

Then the total amount of nucleated clusters within the boundary layer can be calculated as

\[S_{\text{total}} = \int_V \dot{n}_{\text{cluster}} \, dV\] (15)

At the upstream boundary of the computational domain for cluster number density conservation equation, the inlet
cluster number density $n$ is set to be 0. At the exit boundary of $r = R$ and at the axis of the jet, the derivatives of $n$ are set to be 0. It is assumed that all the clusters reached the substrate will adhere to the substrate surface, that is $n_{\text{sub}} = 0$. So the local cluster deposition flux is

$$j_{\text{sub}} = \left. \left( D \frac{\partial n}{\partial r} \right) \right|_{r=0}$$

And the total deposition rate at the substrate surface can be calculated as

$$J = 2\pi \int_0^R j_{\text{sub}} r \, dr$$

Then the cluster total thermodeposition efficiency, which is more meaningful than the absolute value, can be defined as

$$\eta(\%) = \frac{J}{S_{\text{total}}}$$

For most problems involving particle thermodeposition, the characteristic value of particle Schmidt number ($Sc = \nu/D$) is rather high so that it will cause very rapid changes of number density inside the concentration boundary layer near the wall for cool surfaces. To obtain accurate and convincing solutions, it is necessary to distribute the computational grids much more densely near the substrate wall. In the present modeling work, the variable grid spacing is adopted and it is found that the minimum spacing should be set as $2 \times 10^{-7}$ m along axial direction just in front of the substrate for the case of $d_p = 10$ nm. The check of total cluster number balance is proved to be a useful indicator for the effectiveness of grid distribution. The following requirement is examined during iterations and is kept to be satisfied for all the specific simulation cases in this research:

$$\int_{S_{\text{total}}} (J_{\text{diffusion}} + J_{\text{convection}} + J_{\text{deposition}}) > 99\%$$

It is found if the spacing of grid distribution is not sufficiently fine, this requirement cannot be sustained.

3. Simulation results and discussions

Since the main concerns of the present modeling is the transportation behavior of clusters generated in the process of thermal plasma flash evaporation film deposition, the clusters ranging from 1 to 10 nm in diameter with an interval of 2 nm will be modeled respectively, so the physical trend with the variations of cluster size could be revealed.

Fig. 3 shows the cluster number density distribution for the case of $d_p = 1$ nm. To reveal the relative effect of thermophoresis, another comparable case is modeled with the assumption of $K_T = 0$, where only diffusion transportation will be in effect. The result is given in Fig. 4. It can be found that the distributions of these two cases are basically similar to each other but the concentration boundary layer is thinner to some extent for the case of considering thermophoresis effect. So the local cluster deposition flux is increased greatly as shown in Fig. 5.

More obvious differences are shown for the computational case of $d_p = 10$ nm in Figs. 6–8. When assuming $K_T = 0$, the obtained contours (Fig. 7) shows the concentration boundary layer with the thickness around $3 \times 10^{-4}$ m. Whereas for the case considering the thermophoresis effect, the thickness of the concentration boundary layer is significantly suppressed that the resolution of Fig. 6 is not enough to discern it. However, from the results data, it is found that the thickness is reduced to around $4 \times 10^{-5}$ m for the case in Fig. 6. For the distribution of local cluster deposition flux along the substrate surface, as shown in Fig. 8, the effect of

![Fig. 5. The cluster local deposition flux along the substrate surface for $d_p = 1$ nm (Solid: with thermophoresis effect. Dashed: without thermophoresis effect).](image-url)
Fig. 6. Contours of cluster number density for $d_p = 10$ nm under the effect of thermophoresis (unit: $10^9$ m$^{-3}$).

Fig. 7. Contours of cluster number density for $d_p = 10$ nm without the effect of thermophoresis (unit: $10^9$ m$^{-3}$).

Fig. 8. The cluster local deposition flux along the substrate surface for $d_p = 10$ nm (Solid: with thermophoresis effect. Dashed: without thermophoresis effect).

Thermophoresis will improve the deposition flux strikingly and the absolute value of $j_{ab}$ at $r = 0$ almost matches the case for $d_p = 1$ nm. The most important reason for this tendency is the relationship of cluster diffusion coefficient with the diameter $D \sim 1/r^2$. For larger size particles, the contributions of diffusion transportation for cluster deposition will be reduced significantly and is even of little importance compared with the contribution of thermophoretic deposition. So in some modeling researches [9], the diffusion term in the particle conservation equation (Eq. (6)) is even omitted for particles in micron-meter range.

To gain more insight into the effect of quenching under
Fig. 9. The variation of total cluster deposition efficiency with the change of cluster diameters under different effects of temperature gradient.

thermal plasma conditions, Fig. 9 shows the variations of total cluster deposition efficiency $\eta$ with the change of cluster diameters for different degrees of temperature gradient. Here the actual temperature gradient field for the impinging plasma jet is imaginarily reduced to 10 and 1% of the original value, respectively, which represent cases of ordinary temperature differences encountered in many other applications. It is shown that for the cases of relatively lower temperature gradient, the effect of diffusion and thermodeposition will act jointly. The improvement of total deposition efficiency for larger size particles will be more significant than for the smaller size particles, but the absolute values of the former one are always lower due to the lower contribution from diffusion. This type of results are the common results given by the simulation under the temperature difference of $10^1$–$10^3$ K in many ordinary applications of thermophoresis phenomena. However, for the impinging thermal plasma jet, it is clearly shown by Fig. 9 that the effect of thermophoretic force is overwhelming due to the steep temperature gradient before the substrate surface, irrespective of the diameter of cluster.

This conclusion is instructive for the film deposition techniques by thermal plasma flash evaporation process, because the deposition of clusters with larger size will contribute much more portions to the film growth rate. Instead of the common tendency of high deposition efficiency for smaller size clusters under relatively lower temperature gradient, fortunately the deposition efficiency is almost the same within the cluster size concerned here under thermal plasma conditions. Although in fact, the sticking coefficient for clusters with different size is another important parameter needed to investigate, it is a crucial precondition that the large size clusters reach the substrate surface by convection, diffusion or thermodeposition effect.

4. Conclusions

Numerical analysis is conducted to investigate the clusters deposition phenomena under the effect of thermophoresis during film deposition by thermal plasma flash evaporation process. Eulerian approach is adopted and the cluster transportation equation is solved after the velocity and temperature fields are achieved for the impinging thermal plasma jet firstly. The number density distributions for clusters with the uniform size of 1–10 nm are obtained respectively for the case of considering the thermophoresis effect or not considering it. It is found that in the conditions of thermal plasma impinging jet, the thickness of cluster concentration boundary layer is suppressed by the thermophoretic force acted on the clusters, and this tendency is much more significant for the larger sized clusters. The transportation and deposition by thermophoresis mechanism is dominant than the contribution of diffusion, and almost uniform cluster total deposition efficiency can be gotten for clusters of all sizes concerned here. Future research will be focused on the modeling of cluster generation and growth phenomena inside the quenching region of the thermal plasma flash evaporation process.

Acknowledgements

This research was financially supported by the Japan Society for the Promotion of Science (JSPS) under the program “Research for the Future: 97R15301”. The authors thank Prof. X. Chen at Tsinghua University for his valuable suggestions and helpful discussions regarding this work.

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