Investigation of the Propagation of the Heavy Monoatom Beam*

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In this paper, the numerical analyses on the behavior of a heavy monoatom beam are presented. Some numerical methods for the analysis of rarefied gas flow have been proposed thus far, which have been successfully applied to the flow of $Kn < 1$, such as in the semiconductor manufacturing processes and other vacuum technologies. However, since the atomic number density of the beam is extremely low, the flow here is categorized as molecular ($Kn > 1$), in which the atomic free path is rather large compared with the representative scale of the beam. Hence, the probability density function of the atomic free path in the nonequilibrium field was taken into account in the numerical simulation of the beam propagation. Consequently, the calculated results of the beam diffusion due to mutual atomic scattering agreed well with the measured data obtained using gadolinium vapor.

Key Words: Computational Fluid Dynamics, Rarefied Gas, Molecular Flow, Evaporation, Space Engineering

1. Introduction

Recently, fundamental research on rarefied gas dynamics in a vacuum has become indispensable in highly advanced technologies. Gas flow is classified according to the continuity of the fluid as viscous, intermediary or molecular as shown in Table 1⁽¹⁾. Since collision frequency of fluid molecules is considerably large, viscous flow has been conventionally considered to be continuously randomized flow and macroscopic equations such as the Navier-Stokes equation are taken to be valid in describing fluid motion of viscous flow. However, discontinuity of fluid

media appears in the so-called rarefied flow represented by intermediary flow, or viscous flow very similar to intermediary flow, and collisional scattering processes of fluid molecules should be taken into account. The Boltzmann equation describes transport phenomena of fluid molecules, in which the velocity distribution function is a variable defined in seven-dimensional phase space built by time, molecular coordinates and velocity, whereas macroscopic quantities like pressure, temperature and flow velocity are given as statistically averaged quantities. As for rarefied gas flow, some methods of numerical simulation such as the Bird method or the Nanbu method have been applied to various aspects of semiconductor processing or aerospace technologies^{(1)~(4)}.

Here, numerical and experimental studies of the propagation of a heavy monoatom beam as molecular flow are presented, and this study examines the contribution to flow analysis in chemical vapor deposition (CVD), molecular beam epitaxy (MBE) or metal isotope separation by laser beam irradiation⁽⁵⁾. In molecular flow, the collision frequency of fluid molecules decreases to such a low level that the molecular mean free path becomes large compared with the

Table 1 Classification of flow

Viscous flow	$Kn < 0.1$
Intermediary flow	$0.1 < Kn < 10$
Molecular flow	$Kn > 10$

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representative scale of the flow. Therefore, in the numerical analysis of beam propagation, precise evaluation of the probability function of free paths in the nonequilibrium field involving nonuniform density and high flow velocity is necessary. Here, collisional diffusion in atomic beam propagation was analyzed numerically and was experimentally verified using gadolinium vapor.

List of Symbols

c_p : specific heat at constant pressure
 c_v : specific heat at constant volume
 d : beam width
 d_s : collimator width
 E : energy
 e : energy density
 F : force
 f : velocity distribution function
 G : universal gravitation constant
 g : density of collision probability
 I : moment of inertia of system
 J : molecular flux
 K : kinetic energy of system
 Kn : Knudsen number
 k : Boltzmann constant
 l : free path
 M : mass of system
 m : molecular mass
 N : molecular number of system
 n : molecular number density
 P : noncollision probability
 p : pressure
 q : thermal input
 R : random number = [0, 1]
 r : intermolecular distance
 r_m : closest distance between colliding molecules
 S : entropy
 T : temperature
 t : time
 U : internal energy of system
 u : flow velocity
 v : molecular velocity
 $\bar{v}$: thermally averaged velocity
 x : travelling distance of molecule
 γ : ratio of specific heat
 γ^* : polytropic constant
 δ : dimension of analytical element
 ϵ : depth of intermolecular potential
 η : ratio of radii
 θ : tilted angle of flux sensor
 λ : mean free path
 μ : reduced mass
 ν : collision frequency
 ξ : beam dissipation

ρ : impact parameter
 Σ : scattering cross section
 σ : collision diameter
 τ : collision time
 ϕ : intermolecular potential
 χ : scattering angle
 χ_m : cut-off angle
 ϕ : azimuthal angle
 Ω : potential energy of system

(Superscripts)

* : collision partner
 ' : quantity after collision

(Subscripts)

k : subregion

2. Numerical Analysis of Beam Propagation

2.1 Governing equations

The Boltzmann equation, the governing equation of rarefied flow describing fluid molecular transport phenomena, is shown in Eq. (1). The first and second terms of the right-hand side of Eq. (1) are the energy transfer term from outside and elastic collisional term, respectively, and the molecular beam generation energy transfer to molecular vapor from the electron beam is given in Eq. (2). Generally, high-energy electrons, in the case of electric discharge or electron beam heating processes, are the medium transferring electron energy to fluid molecules, and energy transfer must be calculated in terms of mutual interaction between the molecular vapor and the electron beam⁽⁶⁾.

$$\frac{\partial f}{\partial t} + \vec{v} \cdot \nabla f + \frac{\vec{F}}{m} \frac{\partial f}{\partial \vec{v}} = \left(\frac{\partial f}{\partial t} \right)_e + \left(\frac{\partial f}{\partial t} \right)_c \quad (1)$$

$$q = \frac{\partial e}{\partial t} = \frac{\partial}{\partial t} \int \frac{m}{2} \vec{v}^2 f d^3 \vec{v} = \int \frac{m}{2} \vec{v}^2 \left(\frac{\partial f}{\partial t} \right)_e d^3 \vec{v} \quad (2)$$

Because of the collision term shown on the right-hand side of Eq. (1), the theoretical solution of the Boltzmann equation cannot be obtained except in a few cases. Equation (3) shows the two-body collisional term, in which superscripts ' and * denote the quantity after collision and that of the collision partner, respectively. Here, the molecular number density is so low and the criterion shown in Eq. (4), which guarantees approximation of two-body collisions, is satisfied, that multibody collisions with more than three bodies can be neglected. Therefore, Eq. (3) is applicable and the Monte Carlo method is employed here to allow Eq. (3) into the calculation of the Boltzmann equation.

$$\left(\frac{\partial f}{\partial t} \right)_c = \int d^3 \vec{v}^* \int d\chi \frac{d\Sigma}{d\chi} |\vec{v} - \vec{v}^*| (f' \cdot f'^* - f \cdot f^*) \quad (3)$$

$$n^{-1/3} \sigma^{-1} \gg 7 \quad (4)$$

This study focuses on the behavior of the molecular flow where Knudsen number is $Kn > 1$, as illus-

trated by propagation characteristics of the heavy monoatom beam. Since the mean free path of the beam atom in the flow of $Kn > 1$ is a few times larger than the beam diameter, the representative scale of the flow, anisotropy of the mean free path due to density, and flow velocity distributions should be taken into consideration.

2.2 Analytical method of beam propagation

2.2.1 Scattering characteristics Scattering characteristics of monoatoms can be calculated employing the Lennard-Jones model of intermolecular potential in which anisotropy of molecular orientation is not considered, as shown in Fig. 1, since the monoatom has no axis of a chemical bond⁽⁷⁾. Potential parameters of the gadolinium atom are $\sigma = 4.0 \text{ \AA}$ and $\epsilon/k = 230 \text{ K}$, where k denotes the Boltzmann constant⁽⁸⁾.

$$\phi(r) = 4\epsilon \left\{ \left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right\} \quad (5)$$

Fig. 1 Lennard-Jones(12, 6)-type potential

Fig. 2 Modelling of two-body collision processes

Equation (6) is the kinetic equation of the two-body problem, and the scattering angle calculated by integrating Eq. (6) is shown in Eq. (7). Here, the scattering angle χ , defined in the center-of-mass coordinate system depicted in Fig. 2, is obtained by translating the two-body problem into a single-body problem with the introduction of reduced mass.

$$\mu \frac{d^2 \vec{r}}{dt^2} = -\nabla \phi(r). \quad (6)$$

$$\chi = \pi - 2 \int_{r_m}^{\infty} \frac{\frac{\rho}{r^2} dr}{\sqrt{1 - (\rho/r)^2 - \phi(r)/\mu v_{\infty}^2}}. \quad (7)$$

In this paper, Eq. (6) is integrated by the Crank-Nicholson method to obtain the scattering orbit of the atom with impact parameter ρ and relative velocity v_{∞} between two colliding atoms under initial conditions. The lower boundary r_m of integration of Eq. (7) indicates the closest distance between two colliding atoms, and the calculated scattering angle χ of the gadolinium monoatom is shown in Fig. 3. The differential cross section, shown as Eq. (8), offers the probability distribution of the appearance of the scattering angle χ . Since Eq. (8) has a singular point at $\chi = 0^\circ$, cut-off angle χ_m is introduced in order to eliminate minute scattering angles around $\chi = 0^\circ$ having no physical meaning. Therefore, Eq. (8) is integrated to give the total cross section, Eq. (9), in which the lower boundary of the integral is set as χ_m , and hence, the minute angle $\chi < \chi_m$ is neglected. In actual fact, the total cross section, Eq. (9), depends on the value of the cut-off angle χ_m which can be referred to as the parameter of numerical simulation, and it is ideal that the cut-off angle χ_m is set at a considerably small value. However, an exceedingly small cut-off angle leads to excess central processing unit (CPU) time. Accordingly, χ_m is set at 5° .

The Monte Carlo simulation, in general, requires random numbers under a certain probability function. Here, the rejection method is employed in order to calculate the scattering angle χ in the center-of-mass coordinate system shown in Fig. 2(a). The azimuthal

Fig. 3 Scattering characteristics of gadolinium atom

angle ϕ is also calculated statistically as a homogeneous random number $= [0, 2\pi]$. Then, the scattering angle χ and azimuthal angle ϕ are translated into those in the experimental room coordinate system as shown in Fig. 2(b).

$$\frac{d\Sigma}{d\chi} = 2\pi\rho \left| \frac{d\rho}{d\chi} \right| \quad (8)$$

$$\Sigma = \int_{x_m}^{\pi} \frac{d\Sigma}{d\chi} \cdot d\chi = 2\pi \int_{x_m}^{\pi} \rho \cdot \left| \frac{d\rho}{d\chi} \right| \cdot d\chi \quad (9)$$

2.2.2 Atomic free path in nonequilibrium field

The method of estimation of the atomic free path is described below. The collision frequency of multibody systems is defined in the flow velocity coordinate system as shown in Eq.(10), which moves with local flow velocity.

$$\begin{aligned} \nu &= \frac{\iint f \cdot f^* |\vec{v} - \vec{v}^*| \Sigma d^3 \vec{v} d^3 \vec{v}^*}{\int f^* d^3 \vec{v}^*} \\ &\simeq \frac{\Sigma \int \sqrt{2} f v d^3 \vec{v} \cdot \int f^* d^3 \vec{v}^*}{\int f^* d^3 \vec{v}^*} \\ &\simeq \sqrt{2} n \Sigma \bar{v}. \end{aligned} \quad (10)$$

Mean free path λ is given as follows:

$$\lambda = \int f \tau(v) v dv \simeq \frac{1}{\sqrt{2} n \Sigma} \quad (11)$$

where

$$\tau(\bar{v}) = \int_0^{\infty} \nu t \exp(-\nu t) dt = \frac{1}{\nu} \simeq \frac{1}{\sqrt{2} n \Sigma \bar{v}} \quad (12)$$

$$\bar{v} = \int f v dv. \quad (13)$$

In terms of atomic travelling distance x defined in the flow velocity coordinate system, the noncollision probability function $P(x)$ satisfies Eq.(14), giving the probability of travelling to point x with no collision from $x=0$, the point of the most recent collision.

$$P(x+dx) = \left(1 - \frac{x}{\lambda}\right) P(x). \quad (14)$$

$P(0)=1$ is obvious from initial conditions at $x=0$, and Eq.(15) is obtained from Eq.(14). However, the local mean free path in the experimental room coordinate system varies according to the location, and Eq.(15) depends on both the atomic number density and flow velocity. Accordingly, it is, in principle, impos-

sible to forecast the next point of collision at the time the atom is located at $x=0$.

$$P(x) = \exp\left(-\int \frac{dx}{\lambda}\right). \quad (15)$$

Here, as shown in Fig. 4, Eq.(15) is translated into the discrete form of Eq.(16) by dividing the travelling distance x into the subregions, in which the subscript k denotes the subregion k , and A_k represents the factor determined from boundary conditions.

$$P_k(x) = A_k \exp\left(-\frac{x}{\lambda_k}\right) (x_{k-1} < x < x_k). \quad (16)$$

$$\lambda_k = \frac{1}{\sqrt{2} n_k \Sigma}. \quad (17)$$

The condition of continuity of $P(x)$ at every boundary $x=x_k$ leads to Eq.(18). Furthermore, since $P(0)=1$ obviously leads to $A_1=1$, Eq.(19) is obtained by solving Eq.(18).

$$A_k \exp\left(-\frac{x_k}{\lambda_k}\right) = A_{k+1} \exp\left(-\frac{x_k}{\lambda_{k+1}}\right). \quad (18)$$

$$P_k(x) = \exp\left(-\sum_{m=1}^{k-1} \frac{\Delta x_m}{\lambda_m} - \frac{x - x_{k-1}}{\lambda_k}\right). \quad (19)$$

Consequently, the probability distribution function $g_k(x)$ of the atom undergoing a second collision at $x=x$ after the first collision at $x=0$ is given as Eq.(20). However, Eq.(21), the normalization condition of $g_k(x)$, is valid only in the case where there is no evacuated region ($\lambda_k=\infty$, $n_k=0$) and each region is filled with gas if number density is so low.

$$g_k(x) = P_k(x) \frac{1}{\lambda_k} = \frac{1}{\lambda_k} \exp\left(-\sum_{m=1}^{k-1} \frac{\Delta x_m}{\lambda_m} - \frac{x - x_{k-1}}{\lambda_k}\right). \quad (20)$$

$$\begin{aligned} \int_0^{\infty} g(x) dx &= \sum_{k=1}^{\infty} \int_{x_{k-1}}^{x_k} \frac{1}{\lambda_k} \exp\left(-\sum_{m=1}^{k-1} \frac{\Delta x_m}{\lambda_m} - \frac{x - x_{k-1}}{\lambda_k}\right) \\ &\quad \cdot \frac{x - x_{k-1}}{\lambda_k} = \lim_{k \rightarrow \infty} \left\{ 1 - \exp\left(-\sum_{m=1}^k \frac{\Delta x_m}{\lambda_m}\right) \right\} = 1. \end{aligned} \quad (21)$$

As described above, random numbers generated under the probability distribution function $g_k(x)$ correspond to the atomic free path l . In Eq.(22), the cumulative function of $g_k(x)$ is expressed as equivalent to the homogeneous random number $R=[0, 1]$. In terms of the inverse function method, with the solution to Eq.(22), the atomic free path as a probability variable can be obtained in Eq.(23).

$$R = \int_0^l g(x) dx = 1 - \exp\left(-\sum_{m=1}^k \frac{\Delta x_m}{\lambda_m} - \frac{x - x_k}{\lambda_{k+1}}\right). \quad (22)$$

$$l = x_k - \lambda_{k+1} \left\{ \ln(1-R) + \sum_{m=1}^k \frac{\Delta x_m}{\lambda_m} \right\} (x_k < x < x_{k+1}). \quad (23)$$

It should be noted that since the atomic free path obtained in Eq.(23) is the thermal travelling distance defined in the flow velocity coordinate system, the travelling distance due to flow velocity must be added to the atomic free path in the experimental room

Fig. 4 Atomic free path in nonequilibrium field

coordinate system. Therefore, the occurrence of collision in the range of $x_k < x < x_{k+1}$ is identified in terms of Eq.(24), as free path l successively revised with travelling.

$$\sum_{m=1}^k |\vec{v} - \vec{u}_m| \Delta t_m < l < \sum_{m=1}^{k+1} |\vec{v} - \vec{u}_m| \Delta t_m. \quad (24)$$

2.2.3 Boundary conditions The heavy monoatom beam is reformed through a collimator located above the atomic swarm evaporated from the surface of hot molten metal. The velocity distribution function of evaporated atoms under the evaporation boundary condition is assumed as shown in Eq.(25), such that evaporation flux J_0 and thermally averaged velocity $\bar{v}$ should coincide with those of the saturation condition with evaporation temperature T_0 . As for the wall boundary condition, the wall surface here is maintained at such a low temperature that incident atoms colliding with the wall should be trapped regardless of their energy or incident angle.

$$f_i(v) = \pi n_0 \left(\frac{m}{2\pi k T_0} \right)^{3/2} \cdot v_i^2 \exp\left(-\frac{mv_i^2}{2kT_0}\right). \quad (25)$$

$$J_0 = \int_0^\infty v_i \cdot f_i(v) dv = \frac{1}{4} n_0 \bar{v}. \quad (26)$$

$$\bar{v} = \int_0^\infty v_i \cdot f_i(v) dv / \int_0^\infty f_i(v) dv = \left(\frac{8kT_0}{\pi m} \right)^{1/2}. \quad (27)$$

2.2.4 Treatments of scattering media In the Monte Carlo simulation of the behavior of the monoatom beam as rarefied flow, the partner atoms must be prepared corresponding to each traced atom as the scattering medium. In this study, the scattering angle χ of traced atoms is calculated statistically using this partner atom in the center-of-mass coordinate system and is then translated into that in the experimental room coordinate system as depicted in Fig. 2.

Since no energy relaxation is considered here, the traced atom changes only its velocity orientation in the center-of-mass coordinate system. The atomic free path is successively revised and the occurrence of collisions is identified based on the travelling history, as described in 2.2.2. In a numerical simulation such as that of the Bird method, since two atoms are selected from a certain cell as collision partners, the number of atoms residing in a certain cell should not be lower than 20. Furthermore, it is also required that the mean free path λ in a cell is much smaller than cell dimension δ and that collisions occur so frequently in a cell that flow in and around the cell can be assumed to be random. Hence, Eq.(28) should be satisfied in order that the conventional method is valid.

$$Kn \approx \frac{\lambda}{d} < \frac{\lambda}{\delta} < 1. \quad (28)$$

However, since the mean free path λ in a monoatom beam is larger than beam width d and Eq.(28) is not satisfied, the Knudsen number becomes Kn

$= \lambda/d > 1$. Here, partner atoms are generated statistically according to Eq.(29), where Eq.(30) represents the atomic number density and flow velocity calculated by a sampling of behavior of traced atoms. Consequently, traced and partner atoms are destined to suffer mutual collision as expressed by Eq.(29).

$$f \approx n \left(\frac{m}{2\pi k T} \right)^{3/2} \cdot \exp\left\{ -\frac{m}{2kT} (\vec{v} - \vec{u})^2 \right\}. \quad (29)$$

$$n = \int f \cdot d^3 \vec{v}, \quad \vec{u} = \frac{1}{n} \int f \cdot \vec{v} d^3 \vec{v}. \quad (30)$$

In molecular flow, fluid temperature loses its physical meaning because flow is less randomized, and flow velocity appears to depend on energy transfer by the electron beam. The theoretical model of free expansion of a rarefied gas is investigated in the following chapter.

3. Rarefaction Model of Multibody System

3.1 Macroscopic equations

Diffusion processes of a heavy monoatom beam are described by the rarefaction model of multibody systems in Fig. 5, showing mainly the fluid behavior in the randomized region ($Kn < 1$) around the beam source O. Macroscopic equations governing rarefaction processes are shown in Eqs.(31) ~ (34). Equation (31) ~ (33) represent conservation laws of mass, momentum and energy, respectively, and Eq.(34) is the equation of state.

$$\frac{\partial n}{\partial t} + \nabla \cdot n \vec{u} = 0 \quad (31)$$

$$n \frac{\partial \vec{u}}{\partial t} + n \vec{u} \cdot \nabla \vec{u} + \frac{1}{m} \nabla p = 0 \quad (32)$$

$$n \frac{\partial}{\partial t} \left(E + \frac{\vec{u}^2}{2} \right) + n \vec{u} \cdot \nabla \left(E + \frac{\vec{u}^2}{2} \right) + \frac{1}{m} \nabla \cdot p \vec{u} = \frac{q}{m} \quad (33)$$

$$p = nkT \quad (34)$$

Assuming that the problem here is stationary and spherically symmetric, Eqs.(31) ~ (33) can be transformed into Eqs.(35) ~ (37), where r denotes the distance from point O.

Fig. 5 Rarefaction model of multibody system

$$\frac{\partial}{\partial r}(r^2 nu) = 0 \quad (35)$$

$$nm u \frac{\partial u}{\partial r} + \frac{1}{r^2} \frac{\partial}{\partial r}(r^2 p) = 0 \quad (36)$$

$$nm u \frac{\partial}{\partial r} \left(E + \frac{u^2}{2} \right) + \frac{1}{r^2} \frac{\partial}{\partial r}(r^2 p u) = q \quad (37)$$

Quantity q represents the quantity of energy transfer in the process of monoatom beam generation by electron beam heating. Equation (38), in addition to Eqs. (34) and (35), leads to Eq. (39).

$$q = nm T \frac{DS}{Dt} = nm T \left(\frac{\partial S}{\partial t} + u \frac{\partial S}{\partial r} \right) = nm T u \frac{\partial S}{\partial r} \quad (38)$$

$$c_v dT - \frac{p}{n^* m} dn = T dS \quad (39)$$

Since the polytropic constant γ^* is described as Eq. (40), Eq. (39) is integrated to give Eq. (41).

$$\gamma^* = \frac{c_p - T dS/dT}{c_v - T dS/dT} \quad (40)$$

$$\frac{T}{T_0} = \left(\frac{n}{n_0} \right)^{\gamma^*-1}, \quad \frac{p}{p_0} = \left(\frac{n}{n_0} \right)^{\gamma^*} \quad (41)$$

As the entropy increase dS is given in Eq. (42), Eq. (43) is necessary such that $dn < 0$ and $ds > 0$ are simultaneously valid.

$$dS = c_v (\gamma^* - \gamma) \frac{dn}{n} \quad (42)$$

$$\gamma^* < \gamma \quad (43)$$

As for flow velocity u , Eqs. (44) and (45) are obtained by integrating Eq. (36). Therefore, according to Eqs. (41) and (45), temperature and flow velocity are estimated in terms of the polytropic constant γ^* .

$$\frac{1}{2} m u^2 + \int \gamma^* p_0 n_0^{-\gamma^*} n^{\gamma^*-2} dn \quad (44)$$

$$u = \sqrt{\frac{2\gamma^*}{\gamma^*-1} \frac{p_0}{n_0 m} \left\{ 1 - \left(\frac{n}{n_0} \right)^{\gamma^*-1} \right\}} \quad (45)$$

3.2 Polytropic constant

From the point of view of the experimental coordinate system, the multiparticle system generated from beam source O expands upward. It may also be true that from the point of view of the center-of-mass coordinate system, which moves with local flow velocity, it expands spherically around point O', the mass center of the system. Equation (46) shows equilibrium conditions of gravity in the center-of-mass coordinate system, in which the pressure gradient at $r' = r'_i$ balances the synthetic gravity force toward point O'. Here, $M(r'_i)$ represents the total mass of the system contained in the region of $r' < r'_i$, and superscript ' is omitted since no confusion is expected below.

$$\left(\frac{\partial p}{\partial r} \right)_i = - \frac{GM(r'_i)}{r_i^2} n_i m \quad (46)$$

Equation (46) holds even at $r = r_j$, and with introduction of parameter, the ratio of radii $\eta = r_i/r_j$, for which the conditions of homogeneous rarefaction and total mass conservation are shown in Eqs. (47) and

(48), respectively. Hence, the pressure relationship between p_i and p_j is presented in Eq. (49).

$$n_i = \eta^{-3} n_j \quad (47)$$

$$M(r_i) = M(r_j) = \text{const.} \quad (48)$$

$$dp_i = - \frac{GM(r_i)}{r_j^2} \eta^{-4} n_j m r_j = \eta^{-4} dp_j \quad (49)$$

Equation (50), the differential representation for the equation of state, is transformed into Eq. (51) by Eqs. (47) and (49).

$$dp_i = k T_i dn_i + k n_i dT_i \quad (50)$$

$$dp_j = k T_j \eta dn_j + k n_j \eta dT_j \quad (51)$$

According to Eq. (52), obtained by Eqs. (41) and (47), Eq. (51) can finally be transformed into Eq. (53).

$$\frac{T_i}{T_j} = \left(\frac{n_i}{n_j} \right)^{\gamma^*-1} = \eta^{-3(\gamma^*-1)} \quad (52)$$

$$dp_j = k T_j \eta^{-3\gamma^*+4} dn_j + k n_j \eta^{-3\gamma^*+4} dT_j \quad (53)$$

On the other hand, Eq. (54) is also introduced at $r = r_j$ as Eq. (50) holds at $r = r_i$.

$$dp_j = k T_j dn_j + k n_j dT_j \quad (54)$$

In the comparison of Eq. (54) with Eq. (53), the polytropic constant γ^* is given in Eq. (55).

$$\gamma^* = \frac{4}{3} \quad (55)$$

4. Results

4.1 Experimental setup and conditions

Figure 6 shows the experimental setup for verification of the calculated results. Here, the atomic beam source is the surface of a high-temperature heavy metal heated by electron beam (maximum power 10 kW, accelerated energy 8 keV), which supplies atomic vapor in the evaporation chamber. The vapor from the beam source is reformed into a rectangular shape by the collimator, and introduced into the beam channel. Atomic beam flux in the evaporation chamber and beam channel is measured using a quartz oscillator. The atomic number density n is actually one of the main parameters of the beam, as well as collimator width d_s . It is determined by the beam source condition stemming from the atomic number

Fig. 6 Experimental setup

density n_0 of saturated vapor of the beam source. Equation (56) shows the saturated vapor characteristics at temperature T_0 of gadolinium vapor.

$$\log n_0 = -\frac{17\,295}{T_0} + 12.63 - 0.488 \log T_0 - \log kT_0. \quad (56)$$

4.2 Atomic number density distributions

Figure 7 shows the relationship between the atomic number density n_0 of saturated gadolinium vapor and the electron beam power input W , which is estimated experimentally from the weight reduction of the beam source through the experiment. Figure 8 shows calculated atomic number density distributions of gadolinium vapor, in which the shape of the beam source is a 5 mm-diameter circle, and temperature is $T_0 = 1\,780$ K. Two cases of collimator widths, 10 mm and 20 mm, are calculated here. As for atomic number densities in the evaporation chamber, Fig. 9 indicates the calculated and experimental results for gadolinium and copper vapor.

Fig. 7 Atomic number density n of saturated vapor (gadolinium)

(a) Collimator width = 20 mm (b) Collimator width = 10 mm

Fig. 8 Calculated results of atomic number density ($W = 0.4$ kW)

4.3 Collisional diffusion of the atomic beam

Figure 10 shows the effect of collimator width d_s on gadolinium atom flux J_b in the beam channel. Since the atomic beam near the collimator already assumes molecular flow of $Kn > 1$, beam flux J_b shows little dependence on collimator width d_s . Beam velocity u_b is obtained by substituting $n/n_0 \ll 1$ and $\gamma^* = 4/3$ (see Eq.(55)) into Eq.(45). Beam dissipation ξ due to collisional diffusion of beam atoms was defined in Eq. (57) as the ratio of flux measurements J_a and J_b by quartz oscillator A and B, respectively.

$$\xi = \frac{J_a}{J_b}. \quad (57)$$

Figures 11~13 show the dependence of beam dissipation ξ on collimator width d_s , electron beam power input W and tilted angle θ of quartz oscillator A, where the solid line and $\circ$ indicate the calculated and experimental results, respectively.

Fig. 9 Vapor atomic number density (calculated/experimental)

Fig. 10 Beam flux (calculated/experimental)

Fig. 11 Beam dissipation (dependence on collimator width)

Fig. 12 Beam dissipation (dependence on electron beam power)

Fig. 13 Beam dissipation (dependence on tilted angle of sensor)

5. Discussion

5.1 Atomic free path

On the assumption of equilibrium conditions, the atomic free path is assumed to be constant, independent of the atomic coordinate. Equation (59) is obtained by substituting Eq.(58) into Eq.(23).

$$\lambda_k \approx \lambda_0 \approx \text{const.} \quad (58)$$

$$l = -\lambda_0 \log_e(1 - R). \quad (59)$$

Accordingly, at a certain point of collision, the atomic free path to the next point of collision can be calculated statistically in advance, regardless of travelling history. In particular, since, in the flow of $Kn < 1$, the approximation of Eq.(58) is applicable, the numerical calculation of free path is quite easy. However, in molecular flow of $Kn > 1$, it is necessary to revise the free path successively, according to the procedure employed here, and Eq.(23) may be taken as a general representation of Eq.(59). Above all, precise calculation of the free path is essential in the quantitative estimation of beam dissipation ξ .

5.2 Beam velocity

According to molecular motion theory, kinetic energy K and internal energy U of multibody systems are as follows, in the case where the molecule is a monoatom.

$$dK = \frac{3}{2} N k dT = \frac{3}{2} N c_v (\gamma - 1) dT. \quad (60)$$

$$dU = N c_v dT - N T dS = N c_v \frac{\gamma - 1}{\gamma^* - 1} dT. \quad (61)$$

Hence, Eq.(62) is obtained below.

$$K = \frac{3}{2} (\gamma^* - 1) U. \quad (62)$$

Since $K > 0$ is obvious, the polytropic constant γ^* must satisfy Eq.(63) as well as Eq.(43). In the actual beam generating process, thermal input (entropy increase) reduces γ^* to unity, and, in addition, raises beam velocity u_b .

$$\gamma > \gamma^* > 1. \quad (63)$$

6. Conclusions

Beam propagation of the gadolinium atom, as molecular flow, was numerically analyzed by means of the Monte Carlo method, and the following conclusions were obtained.

(1) In the analysis of molecular flow where $Kn > 1$, it is necessary to introduce a nonequilibrium probability distribution function of the molecular free path, although in the case of viscous or intermediary flow, where $Kn < 1$, the mean free path is smaller than the dimensions of the analytical cell, and the equilibrium condition is valid. In particular, in flow fields of high velocity and inhomogeneous density, it is advisable to introduce an algorithm to successively revise the free path, according to the history of molecular travel.

(2) In the numerical simulation of molecular scattering, it is advisable to introduce Lennard-Jones type potential as intermolecular potential which can describe the interaction between actual molecules, rather than using a simple model such as an isotropic scattering model or rigid sphere model. Although in

flow where $Kn < 1$, the difference due to inter-molecular potential is expected to be randomized until it disappears, such a difference appears in the flow where $Kn > 1$.

(3) The polytropic constant $\gamma^* = 4/3$ was obtained in the homogeneous rarefaction model of gravity balanced multibody systems. However, in the electron beam heating process of heavy metals, there exists molecular excitation due to nonelastic collisions between high-energy electrons and molecules⁽⁶⁾. Accordingly, further investigation must be executed, since energy transfer may reduce the polytropic constant γ^* , and therefore raise the beam velocity u_b .

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