CHARACTERISTICS OF THE FINE PARTICLES
GENERATED IN PLASMA PROCESS

Aguru YAMAMOTO, Kenji MATSUBAYASHI, Masaaki SUZUKI

Graduate School of Science and Engineering, Tokyo Institute of Technology,
O-okayama, Meguro-ku, Tokyo, 152-8552, Japan

Abstract

Modeling of fine particles generation in plasma process under an atmospheric pressure is investigated. Our model consists of two physical phenomena, evaporation of metal and particle growth, were studied with computer simulation. Evaporation phenomenon is simulated by lattice Boltzmann method and evaporation rate of iron is estimated. Particle growth is simulated by using general dynamic equation. Agglomerate consisting of primary particles which is taken size distribution into account is represented by fractal dimension connecting number density of primary particles. Particle size and particle size distribution at various temperatures are obtained and discussed.

1. Introduction

When thermal plasma is used in nuclear energy industrial process, the generation of fine particles is a problem from a viewpoint of safety. Plasma has high temperature and high enthalpy and is suitable for treatments of metal having high melting point. But metal particles generate through evaporation. These fine particles are not desirable, so that we have studied their characteristics. It is objective in this study to make the characteristics of the fine particles generated in plasma process under an atmospheric pressure clear and to investigate how to control those generations.

The first step of particle generation is evaporation of metal. Evaporation phenomenon has been studied in losses of light bulb filaments by many researchers. Effects of surrounding gas pressure on evaporation rate of filament metal are reported [1]. Experimental results that are reported have wide range. However, theoretical work is scarce except Langmuir’s ideal evaporation rate. The second step of particle generation is particle growth. Evaporated gas of
metal grows up to be particles through collisions and coalescences. Particle growth has been studied very well. Numerical simulation of the particle growth is governed by general dynamic equations (GDE). When particle size distribution is not assumed, discrete-sectional model is popular in modeling of particle size [2]. Agglomerate consisting of spherical primary particles that are easy to recognize is represented by using a fractal dimension [3].

In the present study, the evaporation phenomenon is simulated with lattice Boltzmann method. Lattice Boltzmann method (LBM) is based on microscopic models and mesoscopic kinetic equations. The LBM uses description for the evaluation of the velocity distribution function of a fluid [4]. Applying multi-speed to discrete velocity model to represent the velocity distribution function makes it possible to use energy equations, and therefore to simulate thermohydrodynamics [5]. Two-component and three-speed LBM is used in the present study to simulate the evaporation phenomenon. Evaporation rates of iron in argon under an atmospheric pressure are estimated at various temperatures. Particle growth is studied by using preparatory experiments and simulations in the present study. The detailed model of particle growth that suits the experimental results is made. The GDEs about zeroth and first moment with discrete-sectional model are used. An additional quantity, the number density of primary particles, makes it possible to treat agglomerate consisting of primary particles with size distribution. Particle growth is simulated with evaporation rate estimated with LBM. Particle size and particle size distribution at various temperatures are obtained.

2. Evaporation phenomenon

**Lattice Boltzmann method**

Boltzmann equation with BGK type single relaxation model in the collision term is [6],

\[
\frac{\partial f}{\partial t} + c \cdot \frac{\partial f}{\partial x} = \frac{f - f^{eq}}{\tau}, \tag{1}
\]

where \( f \) is the distribution function, \( f^{eq} \) is the equilibrium distribution function and \( \tau \) is the relaxation time. The application of BGK type single relaxation time simplifies Boltzmann equation. Particles collide and particle size distribution relaxes toward equilibrium. Eq. (1) is the governing equation of LBM, generally. The distribution function is represented with discrete velocity. Maxwell-Boltzmann distribution of the equilibrium distribution function is expanded though Taylor expansion. The coefficients appeared in the expansion are obtained by using definitions of macroscopic fluid variables and comparing coefficient of Navier-Stokes equation,

(mass) \( \rho = \int f \, dc \), \( \tag{2} \)

(momentum) \( \rho u = \int ef \, dc \), \( \tag{3} \)

(energy) \( e = \frac{1}{\rho} \int \frac{1}{2} (c - u)^2 f \, dc \), \( \tag{4} \)

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(pressure tensor) \[ P = \iint (c - u)^2 \, f \, dc = Dp, \quad (5) \]

(comparison with N-S) \[ u \left( \rho \frac{\partial u}{\partial t} + \frac{1}{2} \rho u^2 + p \right) = \frac{1}{2} \iint c^2 \, f \, dc, \quad (6) \]

where \( D \) is the number of dimensions in space.

The relaxation time \( \tau \) is generally obtained by connecting Boltzmann equation with N-S equation. In the present study the relaxation time is, however, derived microscopically [7]. Relaxation is originated by collisions with like particles and unlike particles. Momentum is also considered because of two components having different masses. The relaxation time of the first component is the following form,

\[ \tau_1 = \frac{m_1}{N_{11} \frac{m_1}{m_1 + m_l} + N_{12} \frac{m_2}{m_1 + m_2}}, \quad (7) \]

where \( m_i \) is the molecular mass of component \( i \) and the number of collisions between component \( i \) and \( j \), \( N_{ij} \) is,

\[ N_{ij} = \frac{1}{2} n_i n_j (d_i + d_j) \left( \frac{1}{m_i} + \frac{1}{m_j} \right) \sqrt{\frac{2 \pi k_B T}{m_i m_j}}, \quad (8) \]

where \( d_i \) is the molecular diameter of component \( i \) and it is assumed that colliding particles were rigid sphere. The relaxation time of the second component is derived, similarly.

Two operators of eq. (1), translation term and relaxation term, are split, then one time step have two steps. Evolution of the local distribution function, \( f(x,t) \), is analyzed. The translation term is discretized with up-wind scheme and the term of time evolution is treated explicitly.

**Evaporation phenomenon**

Evaporation phenomenon is evaluated by using evaporation rate of macroscopic values. Ideal evaporation rate in a vacuum is theoretically derived by Langmuir and is the following form,

\[ J = P_v \sqrt{\frac{1}{2 \pi M R T}}, \quad (9) \]

where \( P_v \) is the vapor pressure, \( M \) the molecular weight, \( T \) the temperature, and it is assumed that evaporating vapor is saturated at a surface of evaporation.

**Simulation**

Evaporation phenomenon needs two-component system, evaporating gas and surrounding gas, to simulate. Each component has own governing equation took the form of eq. (1), where BGK type single relaxation time is applied to the collision term. One-dimension and seven-velocity model (1D7V model), including a stationary particle, is used for discrete velocity. The 1D7V model is three-speed and two-direction and a stationary particle, \{+3c, -3c, +2c, 0, -2c, -3c, +3c\},
\(-2c, +c, -c, 0\), where \(c\) is the standard velocity. Equilibrium distribution functions of each discrete velocity are following.

\[
f(0) = \rho \left[ 1 - \frac{c}{c^2} \left( \frac{10c^2}{3e^4} - \frac{14c}{3e^2} + \frac{49}{18} \right) \right],
\]

\[
f(+1) = \rho \left[ \frac{c}{c^2} \left( \frac{5c^2}{2e^4} - \frac{13c}{4e^2} + \frac{3}{2} \right) \right] \left[ 1 + \frac{cu}{2e^2} + \frac{c^2u^2}{4e^4} - \frac{u^2}{4e^2} + \frac{c^3u^3}{48e^6} - \frac{cu^3}{8e^2} \right],
\]

\[
f(-1) = \rho \left[ \frac{c}{c^2} \left( \frac{5c^2}{2e^4} - \frac{13c}{4e^2} + \frac{3}{2} \right) \right] \left[ 1 - \frac{cu}{2e^2} + \frac{c^2u^2}{4e^4} - \frac{u^2}{4e^2} + \frac{c^3u^3}{48e^6} + \frac{cu^3}{8e^2} \right],
\]

\[
f(+2) = \rho \left[ \frac{c}{c^2} \left( \frac{c^2}{4} - \frac{c}{e^2} + \frac{3}{20} \right) \right] \left[ 1 + \frac{cu}{2e^2} + \frac{c^2u^2}{4e^4} + \frac{u^2}{4e^2} + \frac{c^3u^3}{48e^6} - \frac{cu^3}{4e^2} \right],
\]

\[
f(-2) = \rho \left[ \frac{c}{c^2} \left( \frac{c^2}{4} - \frac{c}{e^2} + \frac{3}{20} \right) \right] \left[ 1 - \frac{cu}{2e^2} + \frac{c^2u^2}{4e^4} - \frac{u^2}{4e^2} + \frac{c^3u^3}{48e^6} + \frac{cu^3}{4e^2} \right],
\]

\[
f(+3) = \rho \left[ \frac{c}{c^2} \left( \frac{c^2}{6e^4} - \frac{c}{12e^2} + \frac{1}{90} \right) \right] \left[ 1 + \frac{3cu}{2e^2} + \frac{9c^2u^2}{8e^4} - \frac{u^2}{4e^2} + \frac{9c^3u^3}{16e^6} - \frac{3cu^3}{8e^2} \right],
\]

\[
f(-3) = \rho \left[ \frac{c}{c^2} \left( \frac{c^2}{6e^4} - \frac{c}{12e^2} + \frac{1}{90} \right) \right] \left[ 1 - \frac{3cu}{2e^2} + \frac{9c^2u^2}{8e^4} - \frac{u^2}{4e^2} + \frac{9c^3u^3}{16e^6} + \frac{3cu^3}{8e^2} \right].
\]

It is possible to avoid the connection with N-S equations, eq. (6), when a stationary particle is not used like 1D6V model, \(\{+3c, -3c, +2c, -2c, +c, -c\}\) without a stationary particle. Since two-component and thermodynamic fluid, however, tend to be unstable, a stationary particle have to be used to make simulation stable.

Diameter of evaporating molecule is given by [8].

\[d = 1.221 V^{1/3}, \quad (10)\]

where \(V\) is the molecular volume of solid metal at its melting temperature.

3. Particle growth

Experiments

In the experiments with DC-arc plasma, generated particles were collected with a membrane filter. Spherical primary particles, which were easy to recognize and to measure size, were observed by using a scanning electron microscope and were about 20[nm] in diameter. Mean diameter hardly changed at various conditions. Primary particles were coagulated each other and particles were agglomerates.
Simulation

It is assumed that particle size distribution is lognormal distribution and self-preserving. Detailed informations are, however, required in application to safety assessment in the nuclear field. We pay attention to size distribution of particles with discrete-sectional model. Particles grow through collisions and subsequent coalescences. Our numerical simulation of particle growth is governed by GDE, rate equation of losses and products of particles at each size. The GDEs about zeroth and first moment, number density and mass density, respectively, are used to simulate. In the free-molecule regime, the collision rate between $v$ and $\bar{v}$ in volume is,

$$
\beta(v, \bar{v}) = \frac{1}{2} (d + \bar{d})^2 \sqrt{2\pi k_B T \left( \frac{1}{m} + \frac{1}{\bar{m}} \right)},
$$

which is derived from the kinetic theory of gases [7]. And number density of primary particles is an additional conserved quantity and is used to consider size distribution of primary particles. The additional number density makes it possible to simulate particle growth of agglomerates consisting of primary particles with any kind of size distributions. The fractal dimension $D_f$ is useful to represent agglomerate consisting of spherical primary particles. The fractal dimension linked to agglomerate consisting of primary particles with size distribution is defined by the following equation,

$$
\nu = \xi^{3 - \nu} \nu_m,
$$

where $\xi$ is the number of primary particles of an agglomerate, $i$ the number of molecules of a primary particle and $\nu_m$ the volume of a molecule.

Particle growth divided into two parts, condensation and coagulation. In condensation part, particles collide and coalesce, then spherical particles generate. In coagulation part, particles collide and stick together, then agglomerates generate.

4. Results and Discussion

Evaporation phenomenon

Simulations for evaporation of iron in rare gas were carried out on LBM and the evaporation rate was obtained. It is assumed that there were saturated zone near evaporating surface, which is the same as what Langmuir assumes to derive eq. (9). It is also assumed that evaporated particles were in gas phase and not affected by external forces. The evaporation rate is normalized with ideal evaporation rate, defined by eq. (9).

Simulated results of Ar-Fe system, evaporation of iron in Ar, are shown in Fig. 1, normalized evaporation rate as a function of $Pt \times L/T$, where $Pt$ is the total pressure, $L$ the size of simulated region and $T$ the temperature. In Fig. 1, simulated results of Kr-Fe system are also included. Every simulations of same system result in one curve. Reduction of the normalized evaporation rate of Kr-Fe system is larger than that of Ar-Fe system, because krypton particle is heavier than argon particle. Fig. 2 shows the normalized evaporation rate as a function of evaporating temperature. The normalized evaporation rate hardly depends on evaporating temperature. On the other hand, total amount of evaporation heavily depend on
evaporating temperature because of the dependence of vapor pressure on temperature (Fig. 3). The results shown in Fig. 3 are estimated by \(L=0.25\text{[mm]}\) at \(T=2200\text{[K]}\) and \(P_T=760\text{[Torr]}\) [9]. The relation that \(L\) is proportional to \(T^{3/4}P_T^{-1/2}\), is also used and it is assumed that \(L\) is the thickness of stagnant film of surrounding gas. Evaporation rate of iron in argon under an atmospheric pressure is about one hundredth of the ideal rate at each temperature.

Fig. 1 Normalized evaporation rate of Ar-Fe and Kr-Fe system as a function of \(P_T/LT\)

Fig. 2 Normalized evaporation rate of Ar-Fe system as a function of temperature under an atmospheric pressure

Fig. 3 Evaporation rate of Ar-Fe system as a function of temperature

**Particle growth**

Time evolution of size distribution of agglomerates is estimated with the computer simulation. It is assumed that critical size of particle is smaller than a molecule, and every sized particle is stable. Time evolutions of the geometric mean diameter of agglomerates at various temperatures are shown in Fig. 4, where it is assumed that the geometric mean
The diameter of primary particles was 20[nm] and the fractal dimension was 2.0. Initial amount of molecules are the results of the simulation of LBM. If the period of particle growth does not depend on temperature, an example of distribution of generated particles is obtained. Particle size distribution of iron surrounded by argon under an atmospheric pressure at various temperatures are shown in Fig. 5.

![Graph showing particle growth](image1)

![Graph showing particle size distribution](image2)

**Fig. 4** Simulated particle growth of Ar-Fe (Geometric mean diameter of primary particle is 20nm)

**Fig. 5** Particle size distribution at various evaporating temperature

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### 5. Conclusion

Simulations for evaporation phenomenon and particle growth were carried out. Evaporation rate of iron surrounded by argon under an atmospheric pressure was one hundredth of ideal evaporation rate derived by Langmuir. Krypton was more effective to reduce evaporation rate than argon. Temperature has an effect on evaporation rate, that is influences particle growth. Though temperature was not determined and still important unknown parameter, it was shown that generating particles through evaporation could be estimated. It was confirmed that the outline of characteristics of the fine particles generated in plasma can be presumed appropriately.

### References


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