MODELING OF ULTRAFINE PARTICLE FORMATION IN A REACTIVE THERMAL PLASMA

Atsushi KANZAWA, Jun MITSUHASHI, Takuya HONDA
and Takayuki WATANABE
Department of Chemical Engineering
Tokyo Institute of Technology
O-okayama, Meguro-ku, Tokyo 152, JAPAN

ABSTRACT

An Al_2O_3 ultrafine particle is formed in an RF plasma using an aluminum powder and an oxygen gas. This process was simulated in this paper. The evaporation of the aluminum powder was estimated using the model of liquid droplet combustion, and the Al_2O_3 ultrafine particle formation was predicted from the nucleation of the produced Al_2O_3 vapor. The calculated particle size distribution was compared with the measured one.

1. INTRODUCTION

So far various kinds of ultrafine particles have been formed by using a thermal plasma, but the studies on the mechanism of the formation are few and still in progress /1,2,3/. Especially, for the case where the chemical reaction occurs, the mechanism is not clear yet.

In the present paper, the formation of an Al_2O_3 ultrafine particle in an RF thermal Ar-O_2 plasma was simulated. The modeling of the ultrafine particle formation consists of three phases: heating of the injected aluminum powder; evaporation and reaction between the evaporated aluminum vapor and oxygen; homogeneous nucleation and particle growth due to the condensation of the produced Al_2O_3 vapor.

Experimental investigations on the Al_2O_3 ultrafine particle formation were also conducted. This experimental conditions were used for the above simulation.

2. EXPERIMENTAL

2.1 Apparatus and Procedure

1) Experimental apparatus

Figure 1 shows the experimental setup used for the Al_2O_3 ultrafine particle formation. The RF power source used has the frequency of 4 MHz and the maximum power of 35 kW. The plasma torch is made of a quartz tube of 40 mm i.d. and 160 mm long. This tube is cooled by water and outside of it an induction coil is winded. The reactor of 40 mm i.d. and 480 mm long is attached to the torch exit to promote the ultrafine particle formation and to exclude the influence of air mixing.

The aluminum powder is fed with the argon carrier gas through the water-cooled tube installed at the center inside of the torch.

The plasma gases, argon and oxygen, are supplied from the top of the torch.

Two plates are set up: one is below the reactor exit to receive the unevaporated aluminum powder, and the other is outside of the reactor exit to trap the ultrafine particle.

2) Experimental procedure

The argon and oxygen gases are supplied into the torch at the fixed flow rate and then a plasma is generated by an RF power source. After becoming steady, the aluminum powder is fed at a constant rate. The unevaporated aluminum powder and the ultrafine particle are collected on each plate.

3) Experimental conditions

The conditions used in this experiments are shown in Table 1.

2.2 Results

1) Amount of unevaporated aluminum

The unevaporated powder which is confirmed to be only aluminum was collected and weighed. This results are shown by the open circles in Fig.6, and it is noticed that the amount of the unevaporated aluminum powder decreases with the content of oxygen.

2) Characterization of ultrafine particle

The obtained ultrafine particle was observed by X-ray diffraction, and it was confirmed to be γ-Al_2O_3.

3) Size of ultrafine particle

The size distribution of the ultrafine Al_2O_3 particle was obtained from TEM photograph as shown in Fig.2. The size of the ultrafine particle is found to be 10 - 350 nm (peak is at 40 nm).

3. CALCULATION OF PLASMA FIELD

3.1 Model and Basic Equations

1) Model

To simulate the ultrafine particle formation, it is necessary to know the plasma fields, i.e.,temperature, velocity and concentration. These fields were obtained from the calculation under the same conditions as the experimental ones. The assumptions used in this calculation are as follows; steady, laminar, LTE except for dissociation reaction of oxygen, axial symmetry, optically thin, constant pressure, neglecting viscous dissipation and neglecting thermal diffusion.

2) Basic equations

Hydrodynamic conservation equations (mass, momentum, energy and species) and Maxwell's equation are used. These equation were solved numerically by using SIMPLER method.

3) Properties

The effect of the dissociation and recombination reactions of oxygen is taken into consideration for thermal conductivity and specific heat at constant pressure. For the other properties, the values of LTE condition are used.

The recombination reaction rate of oxygen atom is given by the next relation.

$$k = 1.9x10exp(7500/RT) \quad \text{for} \quad O + O + M = O_2 + M$$

The dissociation reaction rate can be obtained from the recombination rate constant and equilibrium constant.

3.2 Results and Discussions

Figure 3 shows the result calculated at the oxygen flow rate of 2 l/min. In Fig.3, (a) is streamlines, (b) is isotherm contours and (c) is dissociation degree contours.

The temperature of the $Ar-O_2$ plasma is lower near the induction coil and higher in the reactor compared with the Ar plasma. This is caused by the dissociation(heat absorption) and recombination (heat release) of oxygen. The supplied oxygen is almost dissociated except for the region near the wall.

4. CALCULATION OF ALUMINUM POWDER EVAPORATION

4.1 Model and equations

The feed rate of the aluminum powder is so small that the interaction among the particles and the plasma temperature decreases due to the evaporation may be neglected, so it is sufficient to consider only the behavior of a single particle. The single aluminum particle is assumed to be spherical and to move only towards one direction at the center of the torch. The equation of solid particle motion was used to obtain the trajectory of the aluminum particle.

The heat transfer rate to the single aluminum particle is obtained by using the modified Nusselt number in which the effect of the property change is taken into consideration/4/.

The aluminum particle is heated and fused to the boiling point, and then evaporated. The amount of the evaporated powder was calculated from the model of liquid droplet combustion/5/. This model is shown in Fig.4 and the heat released by the reaction between aluminum vapor and oxygen promotes the vaporization. Two reactions were considered in this calculation and they are

1. $Al + O = AlO + 492$ kJ/mol
2. $Al + (3/2)O = (1/2)Al_2O_3 + 1529$ kJ/mol

4.2 Results

The history of the aluminum particle temperature is shown in Fig.5. At the melting point and the boiling point, the particle temperature is kept constant.

The calculated amounts of unevaporated aluminum powder is shown in Fig.6 and compared with the experimental results. From this figure, it is noticed that the calculated results considering the reaction are in the similar tendency as the experimental results and the effect of the reaction on the evaporation is recognized.

5. CALCULATION OF ULTRAFINE PARTICLE FORMATION

5.1 Model

The product of the reaction between the aluminum vapor and oxygen atom is assumed to be Al_2O_3 vapor according to the reaction 2, and the ultrafine Al_2O_3 particle is formed from this produced vapor. The nucleation begins when this Al_2O_3 vapor is saturated. The homogeneous nucleation model is used here.

The produced nuclei grows by the heterogeneous condensation of Al_2O_3 vapor, and the particle size distribution is obtained from the volume population balance.

5.2 Results

The Al_2O_3 vapor pressure is varied with the position in the reactor as shown in Fig.7, in which the saturated vapor pressure is also illustrated. The difference between these two lines promotes the nucleation. Figure 8 shows the nucleation rate. The nucleation and the growth are noted to begin at the position where the Al_2O_3 vapor pressure exceeds the saturated one.

The particle size distribution at the exit of the reactor is shown in Fig.9. The calculated and experimental results are almost the same, but the calculated ultrafine particle size is a little smaller than the experimental one. This may be caused by the agglomeration among the ultrafine particles.

6. CONCLUSION

(1) The obtained ultrafine particle was verified to be Al_2O_3.
(2) The heat released by the exothermic reaction(aluminum oxidation) promotes the evaporation and this was explained by a combustion model.
(3) The ultrafine particle formation was simulated and the particle size was almost the same as the experimental one.

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Table 1 Plasma condition

plasma source power	8 kW
plasma gas flow rate	Ar : 20 l/min
	O_2 : 0 - 2 l/min
carrier gas flow rate	Ar : 2 l/min
powder feed rate	Al : 0.5 g/min
powder feed position	100 mm from top of torch (no effect of eddy)
powder size distribution	44-105 μm

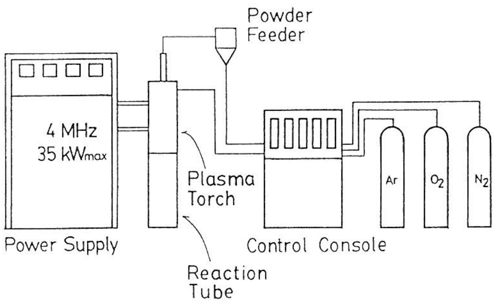

Fig.1 Experimental setup

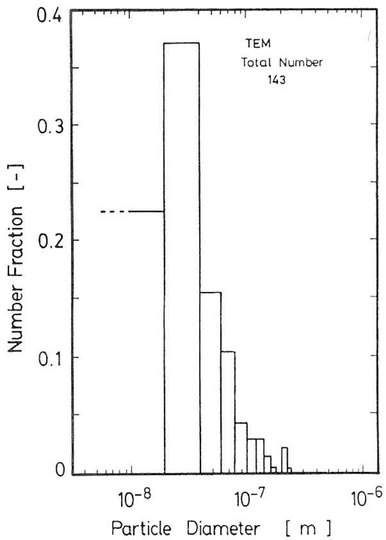

Fig.2 Measured particle size distribution

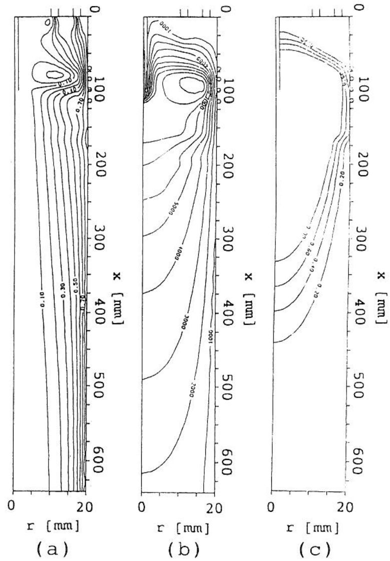

Fig.3 Calculated plasma fields

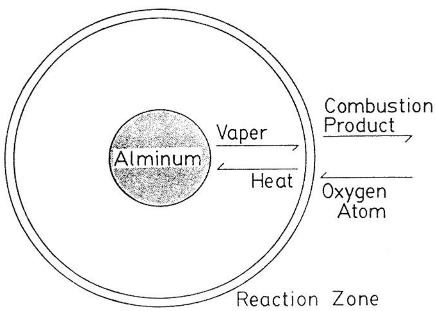

Fig.4 Liquid droplet combustion model

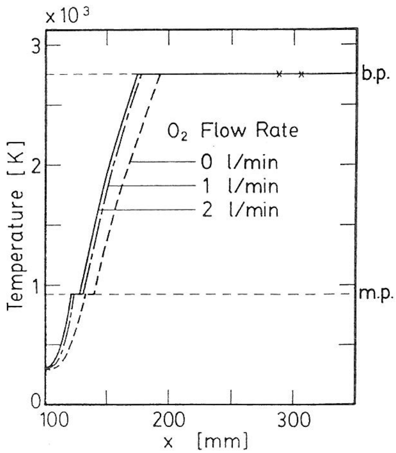

Fig.5 History of particle temperature

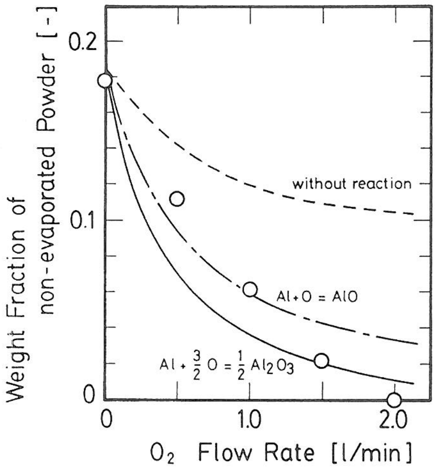

Fig.6 Unevaporated Al powder

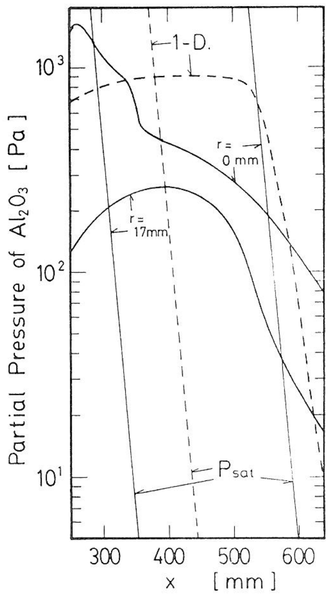

Fig.7 Calculated Al_2O_3 vapor

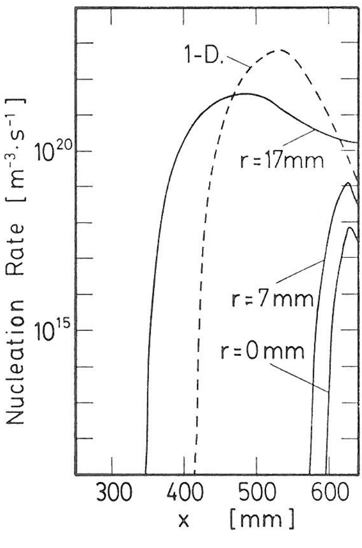

Fig.8 Nucleation rate

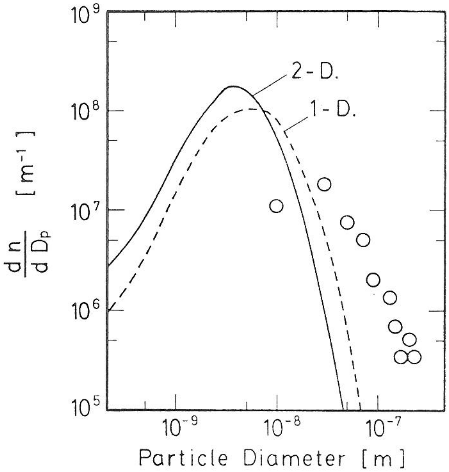

Fig.9 Calculated particle size distribution