Thermal Plasma Modification of Titanium Carbide Powder -Numerical Analysis of Powder Behavior in Plasmas-

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ABSTRACT Modeling of behavior of TiC powders in an Ar-H_2 RF plasma has been performed with the numerical analysis of the plasma fields of velocity, temperature and concentration. The transferred energy to the powders decreases with an increase in the powder feed rate, and with a decrease in the pressure. These parameters have important effects on the powder modification.

1. INTRODUCTION

Radio-frequency thermal plasmas have a number of applications: synthesis of ultrafine powders, deposition of thin films, plasma spraying and powder treatments. Powders injected into plasmas are subjected to modifications of their chemical composition, morphology and crystal structure. The in-flight treatment of TiC powders in an Ar-H_2 or Ar-N_2 plasma induces the formation of carbon-site vacancies followed by the substitution by nitrogen [1,2]. Characteristics of the treated powders are dependent on the plasma composition and pressure and the powder feed rate.

The understanding of the behavior of the powders injected into plasmas is indispensable to investigation of the mechanism. The interactions between the powders and the plasmas can be estimated by the numerical analysis of the heat transfer from the plasma to the powders. The modeling of the powder behavior involves the prediction of the flow, temperature and concentration fields in the plasma with chemical reactions as well as the prediction of the powder trajectory and the temperature history. The interactions can be also evaluated from the powder characteristics such as particle size distribution and compositional modification, because the modification reflects the history of the powders during the processing.

Some modeling of RF thermal plasmas including chemical reactions have been proposed. Zhao *et al.* [3] presented modeling with reactions between $SiCl_4$ and H_2. McKelliget and El-Kaddah [4,5] also calculated plasma fields including the dissociation of $SiCl_4$. Girshick *et al.* [6] reported the simulations of an Ar-H_2 plasma. The authors [7-9] presented the modeling of argon plasmas with molecular gas in consideration of its dissociation and recombination kinetics.

The next task is the estimation of heat and momentum transfer between the powders and plasmas. The estimation has been studied by many authors assuming negligible effect of the powders on the plasma fields. The powders with high injection rate have strong effects on the heat extracted by the powders from the plasma. The interactions of the powders with an RF plasma was modeled by Proulx *et al.* [10] using Particle-Source-In-Cell (PSI-Cell) method.

The purpose of the present work is to investigate the effect of the process parameters such as plasma pressure and the powder feed rate on the characteristics of TiC powders. Numerical simulations of TiC powder behavior injected into an Ar-H_2 RF thermal plasma have been performed, including the effect of the heat extracted by the powders on the plasma fields.

2. NUMERICAL FORMULATION

The modeling of powder behavior in plasmas involves the prediction of the plasma fields of the flow, temperature and concentration. The next task is the estimation of heat and momentum transfer between the powders and the plasma.

2.1 A Model for Plasma Velocity, Temperature and Concentration

A model of the RF plasma torch and reaction chamber is illustrated in Fig. 1, and the operating conditions are summarized in Table 1. The model is concerned with a steel injection tube along the torch axis. Argon is injected from the water-cooled steel tube and from the inner slots. The sheath gas injected with swirl from the outer slots is Ar with H_2. Also Ar and/or H_2 from the circumference slots located below the plasma torch is injected. The dissociation and recombination rates of H_2 are considered.

The fields of flow, temperature and concentration in the plasmas have been calculated by solving the two-dimensional continuity, momentum (x,r,θ), energy and species conservation equations with Maxwell's equations. The electromagnetic fields have been analyzed by a two-dimensional modeling approach with the electric field intensity as the fundamental variable [11]. The governing conservation equations have been solved using SIMPLEC algorithm [12], which is a revision of SIMPLER algorithm [13]. Grid points 54 by 43 were used for both x and r directions.

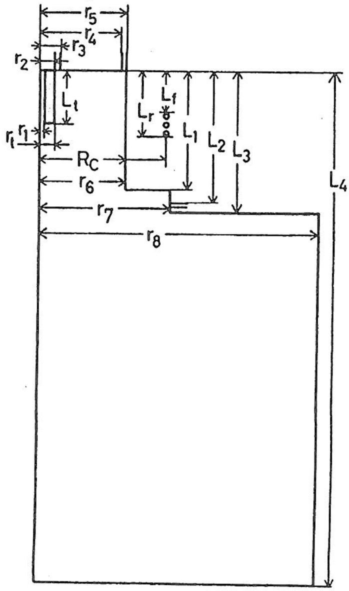

Fig. 1 A schematic of the torch and reaction chamber.

Table 1 Parameters of the RF plasma torch.

Torch power	25 kW
Working frequency	2 MHz
Reactor pressure	53.3 - 101.3 kPa
Coil radius (R_c)	34 mm
Coil turn number	3
Distance to frontal end of coil (L_f)	60 mm
Distance to rear end of coil (L_r)	90 mm
Wall thickness of quartz tube (w)	2 mm
Insertion length of steel tube (L_t)	80 mm
Torch length (L_1)	210 mm
Distance to circumference slot (L_2)	230 mm
Distance to reation chamber (L_3)	240 mm
Total length of reactor (L_4)	740 mm
Flow rate of carrier gas (Ar)	4 liters/min
Flow rate of plasma gas (Ar)	6 liters/min
Flow rate of sheath gas (Ar)	30 liters/min
Flow rate of sheath gas (H_2)	3.5 liters/min
Flow rate of tail injection gas (H_2)	5 liters/min
Inner radius of injection tube (r_1)	1.25 mm
Outer radius of injection tune (r_t)	3.75 mm
Inner radius of inner slot (r_2)	3.75 mm
Outer radius of inner slot (r_3)	5 mm
Inner radius of outer slot (r_4)	22.5 mm
Outer radius of outer slot (r_5)	23 mm
Inner radius of quartz tube (r_6)	23 mm
Inner radius of circumference slot (r_7)	35 mm
Inner radius of reaction chamber (r_8)	75 mm

2.2 A Model for Behavior of a TiC Particle

The powders are injected from the steel tube into the plasma ($x = 80$ mm, $r = 0 \sim 1.25$ mm). The heat transfer from the plasma have been estimated with the model of a single particle in a rarefied gas [14]. The heat transfer equation for a particle in a continuum fluid can not be used because the Knudsen number is larger than 1. The diameter of the powders used is smaller than the mean free path length of the plasma. The continuum concepts are no longer applicable at high Knudsen numbers. The heat transfer equation in a rarefied gas can be expressed as:

$$q = q_n + q_i + q_e \qquad (1)$$

where q is the total heat transfer from the plasma to a particle, q_n, q_i and q_e are the heat transfer due to the neutral species, ions and electrons, respectively. The heat transfer due to neutral species consists of the terms due to argon: q_{Ar}, hydrogen atom: q_H, and hydrogen molecule: q_{H2}.

$$q_n = q_{Ar} + q_H + q_{H2} \qquad (2)$$

The trajectory of a particle can not be also estimated from the continuum concepts. The another approach for a free molecular flow has to be employed. Chen and Pfender [15] pointed out the importance of non-continuum effects on the particle motion and proposed a correction term for the drag coefficient in the Knudsen number regime $10^{-2} < Kn < 1$. Their formula is no longer applicable in our case, therefore the particle velocity is assumed to be the same as the plasma velocity as the first approximation. The particle trajectory has been also estimated using the model by Chen and Pfender. The numerical simulations showed that the particle velocity is the same as the plasma velocity except the injection point.

2.3 The Plasma-Particle Interaction

The initial powders are classified into 12 different diameters. The particle size distribution before the injection and the total feed rate have been adjusted to the experimental conditions. The total heat transfer to all powders has been calculated considering the particle size distribution, after the estimation of the trajectory and temperature of a single particle of every classification.

The transferred energy to the powders is expected to affect the temperature and velocity distributions in the plasma because of the large transferred energy. The PSI-Cell method is one of the best models for the plasma-particle interaction, however this method is not available in our case. Our model includes the dissociation and recombination of hydrogen and the complicated geometry of the calculation domain with an injection gas at the reaction chamber. Therefore obtaining the stable solution for only the plasma fields is extremely difficult with long CPU time. Another simple method has to be employed for the estimation of the plasma-particle interaction. The plasma temperature and velocity distributions have been recalculated with the assumption that the energy is extracted uniformly from the plasma in the region of the powder passage.

3. RESULTS AND DISCUSSION

The calculated isotherms and concentration contours of H are shown in Fig. 2 (a) and (b), respectively. Calculations were performed for Ar-H_2 at 66.7 kPa with the tailflame injection gas (Ar: 10 liters/min; H_2: 5 liters/min). The injection gas from the steel tube and from the circumference slots are heated up rapidly. The concentration profiles show that H concentration near the torch wall below the coil region is the highest because the sheath gas is entrapped by the recirculation and is thermally decomposed.

The history of heat flow of a TiC particle in an Ar-H_2 plasma is shown in Fig. 3. The transferred energy has been estimated for a particle with 2 μm at the pressure of 66.7 kPa. The heat flow due to argon atom is comparable to that due to hydrogen atom. The contribution due to hydrogen molecule can be negligible owing to the high

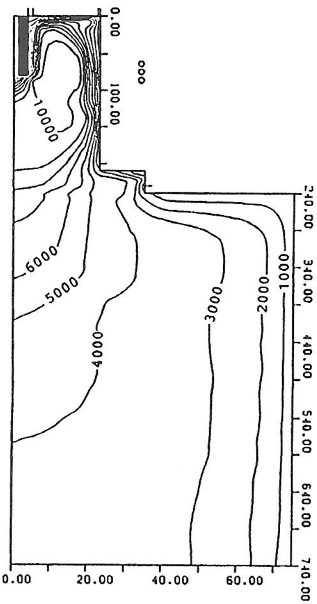

Fig. 2(a) Isotherms.

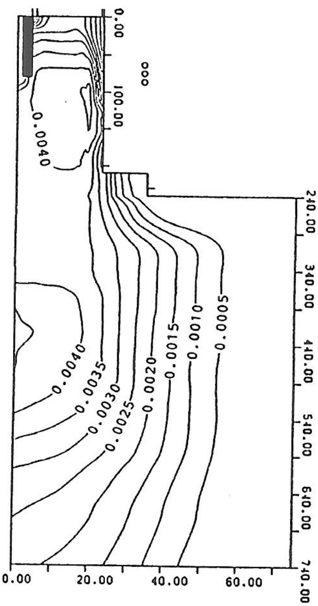

Fig. 2(b) Concentration contours of H.

dissociation. The contribution due to ions or electrons can be also negligible owing to the low ionization in the plasma.

Effects of the powder feed rate on the transferred energy, and on the evaporation and melting fraction are shown in Figs. 4 and 5, respectively. The transferred energy decreases with an increase in the powder feed rate, because the injection powders with larger feed rate leads to larger decrease in the plasma temperature. All of the powders melts with the powder feed rate under 2 g/min, and the almost powders remains in the solid state over 5 g/min. While the evaporated fraction increases under the feed rate of 3 g/min. The powder feed rate has strong effects on the powder characteristics, and the feed rate between 2 and 3 g/min is the optimum in this condition.

Effects of the pressure on the transferred energy, and on the evaporation and melting fraction are shown in Figs. 6 and 7, respectively. The transferred energy increases with the pressure, because of larger

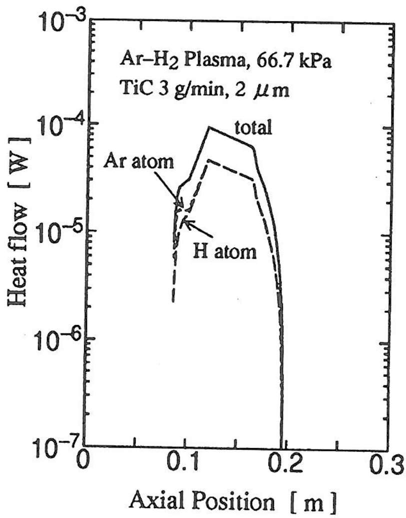

Fig. 3 Heat flow to a TiC particle.

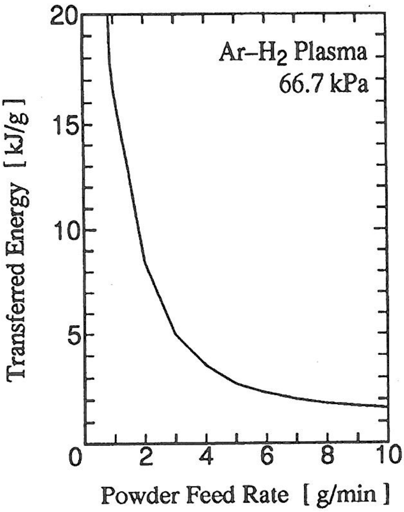

Fig. 4 Effects of the powder feed rate on transferred energy to the powders.

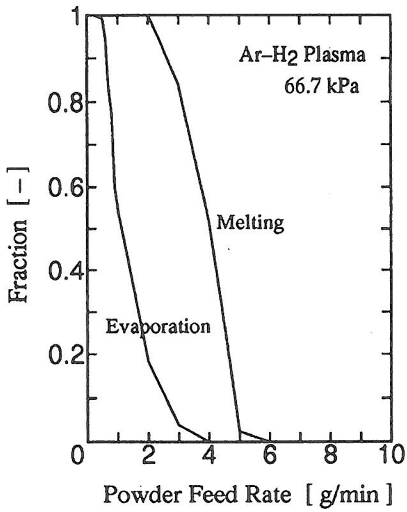

Fig. 5 Effects of the powder feed rate on melting and evaporation fraction of the powders.

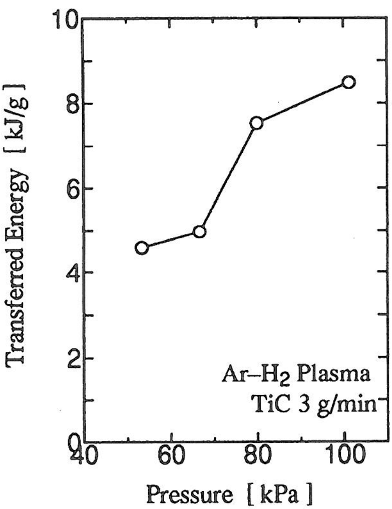

Fig. 6 Effects of the pressure on transferred energy to the powders.

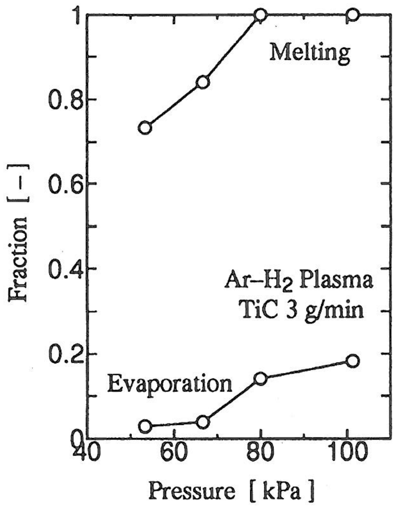

Fig. 7 Effects of the pressure on melting and evaporation fraction of the powders.

number density of each species, and of lower particle velocity resulting longer residence time. The pressure is also the important parameter for the in-flight powder treatment.

Comparisons of the calculated and measured powder size distribution are shown in Fig. 8. The particle size distribution of the powders, which were dispersed in a 0.2 wt % of sodium hexametaphosphate aqueous solution, was measured by Brookheaven Instrument, BI-XDC. The transferred energy can be evaluated indirectly from the size distribution of the treated powders, because the characteristics reflect the history of the powders in the plasma. Both of the calculated and measured results present that the powder size decreases with an increase in the pressure. These results are due to larger total heat transfer at higher pressure, and these results are consistent with the results of the calculated transferred energy.

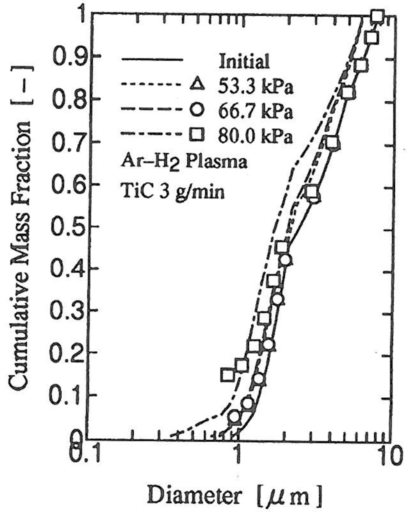

Fig. 8 Calculated and measured particle size distribution in an Ar-H_2 plasma; keys: calculations; lines: experiments.

4. CONCULUSIONS

Modeling of behavior of TiC powders in an Ar-H_2 RF thermal plasmas has been performed as well as the numerical analysis of the plasma fields. The transferred energy to the powders affects the temperature and velocity distributions in the plasma because of the large transferred energy. These distributions have been recalculated with the assumption that the energy is extracted uniformly from the plasma in the region of the powder passage. The transferred energy to the powders decreases with an increase in the powder feed rate, and with a decrease in the plasma pressure. The measured powder size also indicates that the heat transfer to the powders is larger at larger pressure .

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