

## Effect of Frequency on Characteristics of Radio Frequency Argon-Hydrogen Plasmas

Takayuki Watanabe, Atsushi Kanzawa, Mina Sakano\*, Naohiko Shimura\* and Yoshimichi Ohnishi\*

Department of Chemical Engineering, Tokyo Institute of Technology  
2-12-1 O-okayama, Meguro-ku, Tokyo 152, Japan

\*Heavy Apparatus Engineering Laboratory, Toshiba Corporation  
2-1 Ukishima-cho, Kawasaki-ku, Kawasaki 210, Japan

### Abstract

Modeling of radio frequency thermal plasmas has been performed to investigate the effect of induction frequency on the plasma characteristics. Computations are carried out for Ar-H<sub>2</sub> plasmas over the frequency range 0.5-4 MHz. The induction frequency is strongly related to the skin depth resulting in the off-axis peak distribution of the temperature. The peak position of the temperature distribution shifts toward the center with a decrease in the frequency.

### 1. INTRODUCTION

Radio-frequency (RF) induction thermal plasmas have been used for production of high-quality and high-performance materials, such as synthesis of ultrafine powders, deposition of thin films, plasma spraying and treatments of powders. The plasma is formed by applying a RF current across a coil around a quartz tube. The applied frequency is strongly related to the size of the torch and also to the distributions of the temperature, velocity and species concentration. Numerical modeling has been a powerful tool in investigation of the effect of frequency on the plasma characteristics.

A theoretical investigation of the frequency effect on local thermodynamic equilibrium (LTE) conditions was performed by Mostagimi and Boulos [1]. Their computations were carried out for argon plasmas over the frequency ranging from 3 to 40 MHz. They showed that higher frequency results in lower temperature, and the difference between the electron and the atom/ion temperatures is increased. The effect of frequency on the dynamic behavior of RF plasmas was calculated by Sakuta *et al.* [2]. They showed that a frequency of a few hundred hertz is the lower limit applicable. The authors [3-5] presented the modeling of argon plasmas mixed with molecular gas in consideration of finite-rate of the dissociation and recombination.

The purpose of the present study is to investigate the effect of induction frequency in Ar-H<sub>2</sub> thermal plasmas on the distributions of the temperature, velocity and species concentration. The induction frequency is important in determining the torch size, since lower frequency results in an increase in the torch size from the theory of induction heating. The modeling would give the estimation of the optimum torch diameter according to the induction frequency.

## 2. NUMERICAL FORMULATION

A model of the RF plasma torch and reaction chamber is shown in Fig. 1, and the operating conditions are summarized in Table 1. The plasma torch is surrounded by a water-cooled induction coil. The coil consists of two turns and applies the induction frequency from 500 kHz to 4 MHz to the plasma. The plate power is 120 kW and the actual power level at the torch has been assumed to be 60 kW. The model is concerned with a water-cooled copper injection tube along the torch axis. The sheath gas injected with swirl from outer slots protecting the inner surface of the quartz tube is the mixture of argon and hydrogen.

The calculations are based on the following assumptions: (a) steady-state laminar flow [6]; (b) axial symmetry; (c) optically thin; (d) negligible viscous dissipation; (e) negligible displacement current and flow-induced electric field; (f) local thermodynamic equilibrium for the ionization [7], while the dissociation and recombination rates of hydrogen have been considered.

The fields of flow, temperature and concentration of the RF thermal plasma can be calculated by solving the two dimensional continuity, momentum, energy and species conservation equations in consideration of swirl flow of the sheath gas.

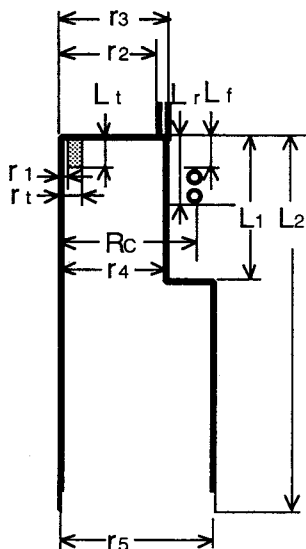


Fig. 1 Geometry of the calculation domain of the RF plasma torch and reaction chamber.

Table 1 Parameters of the RF plasma torch.

Torch power	60.0 kW
Working frequency	0.5 - 4 MHz
Reactor pressure	101.3 kPa
Coil radius (Rc)	56.5 mm
Coil turn number	2
Distance to frontal end of coil (Lf)	23 mm
Distance to rear end of coil (Lr)	62 mm
Wall thickness of quartz tube (w)	1.5 mm
Insertion length of steel tube (Lt)	45 mm
Torch length (L1)	240 mm
Total length of torch and reactor (L2)	740 mm
Flow rate of carrier gas (Ar)	2 liters/min
Flow rate of sheath gas (Ar)	40 liters/min
Flow rate of sheath gas (H2)	2.65 liters/min
Inner radius of injection tube (r1)	1.0 mm
Outer radius of injection tube (rt)	4.5 mm
Inner radius of outer slot (r2)	39 mm
Outer radius of outer slot (r3)	41 mm
Inner radius of quartz tube (r4)	41 mm
Inner radius of reaction chamber (r5)	55 mm

Continuity:  $\nabla \cdot (\rho \mathbf{u}) = 0$  (1)

where  $\rho$  is the density and  $\mathbf{u}$  is the velocity.

Momentum:  $\rho \mathbf{u} \cdot \nabla \mathbf{u} = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \mathbf{J} \times \mathbf{B}$  (2)

where  $p$  is the pressure and  $\boldsymbol{\tau}$  is the viscous stress tensor. The last term in the right-hand side is due to the Lorentz force.

Energy:

$$\rho \mathbf{u} \cdot \nabla h = \nabla \cdot \left( \frac{k}{C_p} \nabla h \right) + \nabla \cdot \left\{ \frac{\mu}{Pr} \sum_i (Le_i - 1) h_i \nabla c_i \right\} - Q_r + \mathbf{J} \cdot \mathbf{E}$$
 (3)

where  $h$  is the enthalpy,  $k$  is the thermal conductivity,  $C_p$  is the specific heat at constant pressure,  $Q_r$  is the radiation loss per unit volume, and  $c$  is the mass fraction. The second term in the right-hand side is due to the energy transfer caused by the diffusion, and the last term is due to the Joule heating.

Species:  $\rho \mathbf{u} \cdot \nabla c = \nabla \cdot (\rho D \nabla c) + S$  (4)

where  $D$  is the diffusion coefficient and  $S$  is the source term owing to the dissociation and recombination of hydrogen. In these equations, the conduction current  $\mathbf{J}$ , the magnetic flux density  $\mathbf{B}$ , and the electric field intensity  $\mathbf{E}$  have been obtained from Maxwell's equation.

The boundary conditions along the centerline were set to insure axial symmetry. At the wall of the plasma torch and the reactor, no slip conditions are maintained for the velocity, and the concentrations have zero gradient. The temperature at the inside wall of the plasma torch was calculated assuming the outside wall was maintained at 300 K by water cooling. The temperature at the reactor wall was assumed to be at 300 K. The injection tube was assumed to be at 500 K. At the inlet, carrier gas from the central injection tube and sheath gas along the wall have been considered. The sheath gas has swirl velocity component. Each gas stream has constant axial velocity with zero radial velocity having temperature at 300 K. The outflow boundary conditions at the reactor were assumed that gradient of the variables are zero.

The electromagnetic (EM) fields in this study have been analyzed on the basis of the two-dimensional modeling approach with the electric field intensity as the fundamental EM field variable [8]. Maxwell's equations are expressed in terms of the electric field intensity as follows:

$$\nabla^2 \mathbf{E} - \xi \sigma \frac{\partial \mathbf{E}}{\partial t} = 0$$
 (5)

where  $\xi$  is the magnetic permeability and  $\sigma$  is the electrical conductivity. A sub-domain inside the plasma torch has been used for the calculation of the EM fields. The associated boundary conditions for the EM fields are identical to the boundary conditions used in Ref. 8.

The recombination rate  $k_r$  of hydrogen [9] considering the three-body reactions with neutral species  $M$  can be evaluated from Eq. (6):

$$\begin{aligned} \text{H} + \text{H} + \text{M} &= \text{H}_2 + \text{M} \\ k_r &= 1.0 \times 10^6 \text{ T}^{-1.0} \end{aligned}$$
 (6)

The dissociation rate has been calculated using the equilibrium constant.

The governing conservation equations have been solved using SIMPLEC algorithm [10], which is a revision of SIMPLER (Semi-Implicit Method for Pressure Linked Equation Revised) algorithm [11]. The governing equations and the electric field intensity equation and the associated boundary conditions were discretized into finite difference form using the control-volume technique.

### 3. RESULTS AND DISCUSSION

#### 3-1 Plasma fields

The calculated isotherms and concentration contours of hydrogen atom are shown in Figs. 2 and 3, respectively. Computations were performed for Ar-H<sub>2</sub> plasmas at the induction frequency of 1 MHz. The temperature field shows off-axis peak distribution, because the time-varying magnetic field can not penetrate into the inner part of the plasma. This phenomenon is quantified in terms of the skin depth  $\delta$  defined as

$$\delta = (\pi \xi \sigma f)^{-1/2} \quad (7)$$

where  $f$  represents the induction frequency. The skin depth indicates the penetration depth of the time-varying magnetic field into the plasma. The skin depth increases with a decrease in the frequency. Therefore the temperature distribution is strongly related to the induction frequency.

The corresponding concentration profiles show that the concentration of hydrogen atom around the torch exit is the highest, because the hydrogen injected as the sheath gas is entrapped by the recirculation and is thermally decomposed into the atom. This indicates that the sufficient mixing of the sheath gas is achieved at the torch exit.

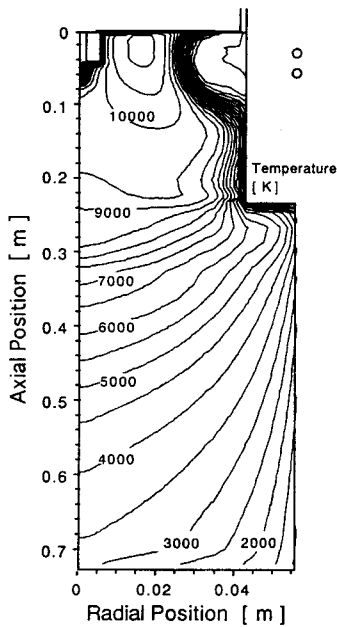


Fig. 2 Isotherms in Ar-H<sub>2</sub> plasmas at 1 MHz.

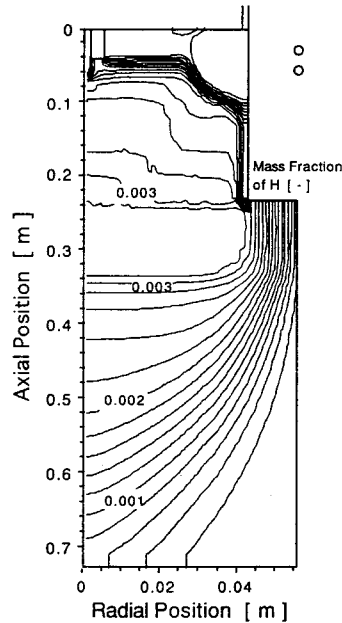


Fig. 3 Concentration contours of hydrogen atom in Ar-H<sub>2</sub> plasmas at 1 MHz.

### 3-2 Effect of induction frequency

The effect of induction frequency in Ar-H<sub>2</sub> thermal plasmas on the distributions of the temperature and the velocity is investigated. The radial distributions of the plasma temperature at the coil region ( $x = 52.5$  mm) with changing the frequency from 0.5 to 4 MHz are presented in Fig. 4. These distributions show the off-axis peak resulting from the skin effect which is quantified by Eq. (7). The peak position of the temperature distribution shifts toward the center with a decrease in the frequency. A decrease in the frequency leads to an increase in the skin depth, resulting in the deep penetration of the high temperature region. These results are in qualitative agreement with the distributions of the Joule heating rate as shown in Fig. 5. The peak position of the distribution also shifts toward the center with a decrease in the frequency.

The temperature distributions in Fig. 4 represent the low temperature region near the wall. The low temperature region is attributed to the inflow of the cold sheath gas caused by the Lorentz force toward the center. The area of the low temperature region increases with a decrease in the frequency. This effect may lead to a decrease in the convection heat loss from the plasma to the torch wall.

The radial velocity distributions are shown in Fig. 6. The radial velocity toward the center is caused by the radial

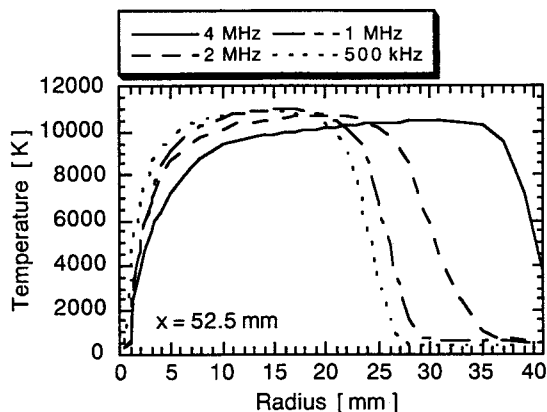


Fig. 4 Effect of frequency on the radial profiles of the temperature at the coil region.

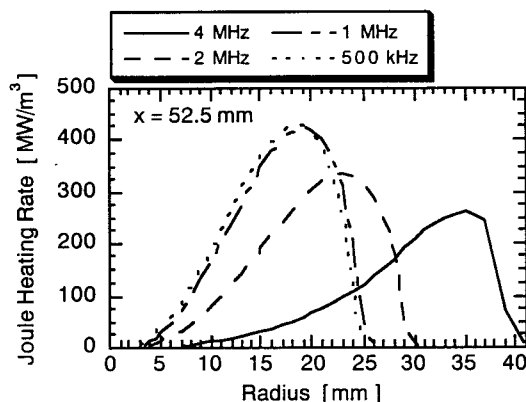


Fig. 5 Effect of frequency on the radial profiles of the Joule heating rate at the coil region.

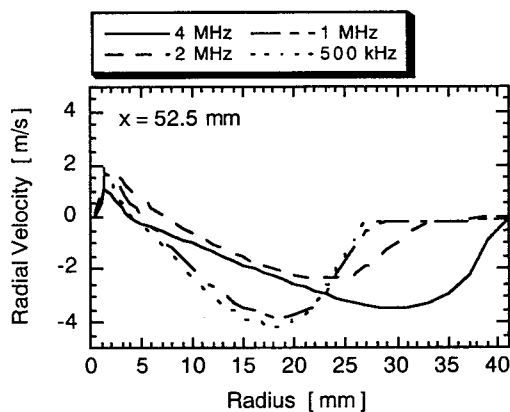


Fig. 6 Effect of frequency on the radial profiles of the radial velocity at the coil region.

Lorentz force as shown in Fig. 7. Stronger radial velocity and stronger radial Lorentz force are generated at lower frequency. The peak position of these distributions shifts toward the center with a decrease in the frequency. This is due to an increase in the skin depth with a decrease in the frequency.

The effect of frequency on the distribution of hydrogen atom, which is not shown here, is not important at the coil region. Hydrogen is dissociated completely except near the torch wall owing to its relatively low dissociation energy.

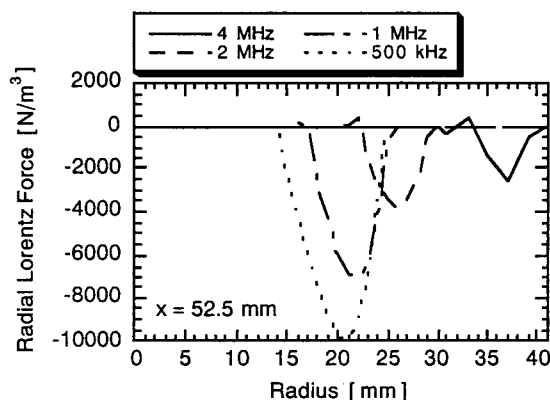


Fig.7 Effect of frequency on the radial profiles of the radial Lorentz force at the coil region.

#### 4. CONCLUSIONS

Modeling of Ar-H<sub>2</sub> RF thermal plasmas with changing the induction frequency from 500 kHz to 4 MHz has been performed. The induction frequency has strong effect on the skin depth resulting in the off-axis peak distribution of the temperature. A decrease in the frequency leads to an increase in the skin depth, therefore the peak position of the temperature distribution shifts toward the center with a decrease in the frequency. The choice of induction frequency is important in determining the optimum torch diameter.

#### References

1. J. Mostaghimi and M. I. Boulos, *J. Appl. Phys.*, **68**, 2643 (1990).
2. T. Sakuta, *et al.*, *Plasma Sources Sci. Technol.*, **2**, 67 (1993).
3. T. Watanabe, *et al.*, *J. Chem. Eng. Jpn.*, **24**, 25 (1991).
4. T. Watanabe, *et al.*, *Proc. Symp. Plasma Sci. Mater.*, **6**, 211 (1994).
5. T. Watanabe, *et al.*, *J. Mater. Res.*, to be published.
6. K. Chen and M. I. Boulos, *J. Phys. D*, **27**, 946 (1994).
7. J. Mostaghimi, P. Proulx and M. I. Boulos, *J. Appl. Phys.*, **61**, 1753 (1987).
8. X. Chen and E. Pfender, *Plasma Chem. Plasma Processing*, **11** (1991) 103.
9. K. Kuratani, in *Formation Mechanism and Controls of Pollutants in Combustion System* (The Japan Society of Mechanical Engineers, Tokyo, 1980), p.5.
10. J. P. Van Doormaal and G. D. Raithby, *Num. Heat Transfer*, **7**, 147 (1984).
11. S. V. Patanker, *Numerical Heat Transfer and Fluid Flow* (McGraw-Hill, New York, 1980).