NUMERICAL MODELLING OF A NONEQUILIBRIUM RF INDUCTION REACTIVE PLASMA USING MULTICOMPONENT MODEL

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Abstract

A numerical modelling of RF induction nonequilibrium argon-nitrogen plasma with chemical reactions is carried out at atmospheric pressure using multicomponent model. Especially, dissociation and ionization, thermal nonequilibrium effect and further variable transport properties in the RF induction argon-nitrogen plasma are taken into account in this modelling. It is investigated how the thermofluid and diffusion characteristics of plasma species and further plasma parameters in the RF induction plasma are influenced by the injection locations of premixed argon-nitrogen gas and by the inlet flow rate differences.

1. Introduction

Plasma is regarded as a multifunctional fluid having high energy density, chemical reactivity, variable transport properties and also electromagnetic controllability [1]. In various kinds of plasma, an inductively coupled plasma has the advantages of large volume with low velocity, clean high-energy and chemical reactivity since it can be produced by an induction RF electric field using reactive gas selectively without electrodes. Especially, it is suitable for chemical processes such as synthesis of ultrafine powders, spherodization, reactive plasma spraying, diamond film deposition, decomposition and recovery of harmful waste materials [2]. To enhance the multifunctions of an inductively coupled plasma, it is supposed to be effective to utilize the chemical reactions and the variations of transport properties by gas mixtures [3]. Furthermore, it is very important to clarify the thermofluid characteristics, plasma parameters under the complex mixing processes and the various kinds of chemical reactions and also to control a plasma flow precisely for high quality plasma processings. In this case nonequilibrium effect and simultaneous dissociation and ionization should be taken into account for plasma modelling. As far as the authors know, there are some numerical [4] and experimental [5] studies on RF induction mixed plasma with chemical reactions. However, there is very few paper on the numerical modelling of a nonequilibrium RF induction mixed plasma accompanied with simultaneous dissociation and ionization.

Then the main objective of a present study is to formulate a numerical model that describes the thermofluid and concentration fields of chemical species and also plasma parameters of a nonequilibrium RF induction argon-nitrogen plasma at atmospheric pressure. This formulation, which takes into account the dissociation and ionization, thermal nonequilibrium effect and further temperature dependent variable transport properties of gas mixtures in the induction electromagnetic field, is presented here using multicomponent model. A numerical simulation is carried out how the thermofluid and diffusion characteristics of plasma species and further plasma parameters in the nonequilibrium RF induction argon-nitrogen plasma are influenced by the injection locations and flow rates of premixed argon-nitrogen gas.

2. Numerical model

2.1 Governing equations

A schematic illustration of the RF induction argon-nitrogen plasma torch and the coordinate system are shown in figure 1. In order to derive the governing equations, the following assumptions are introduced here for a nonequilibrium RF induction plasma with the axial injection of argon-nitrogen mixtures from nozzles 1, 2 and 3 respectively at atmospheric pressure.

(i) The plasma is regarded as an ideal gas and a continuous fluid in thermodynamic nonequilibrium.

(ii) The plasma is an optically thin. The ionization and also dissociation of argon-nitrogen mixture are considered.

(iii) The flow field is steady and laminar. The velocity, temperature and concentration fields are two dimensional and axisymmetric.

(iv) The induction electromagentic field with sinusoidal high frequency is two-dimensional and axisymmetric.

(v) The displacement current, gravity, viscous dissipation and further hall parameter are not considered.

(vi) The effect of anisotropy of the thermodynamic and transport properties in the induction electromagnetic field is not considered.

(vii) The ion diffusion is controlled by ambipolar diffusion.

Under these given assumptions, the governing equations are presented as follows.
Conservation of mass for all species:

$$\nabla \cdot (\rho \mathrm{u}) = 0 \tag{1}$$

Momentum equations:

$$\nabla \cdot (\rho \mathrm{u}\mathrm{u}) = -\nabla p + \nabla \cdot \bar{\bar{\tau}} + \mathrm{F} \tag{2}$$

Where the axial and radial components of the Lorentz force are respectively given by

$$F_z = -\frac{1}{2}\mu_0\sigma Real(E_\theta \overline{H}_r), \qquad F_r = \frac{1}{2}\mu_0\sigma Real(E_\theta \overline{H}_z) \tag{3}$$

Energy equations for heavy particles and electron:

$$\nabla \cdot (\rho_h h_h \mathrm{u}) = \nabla \cdot \left(\frac{\lambda_h}{C_{p_h}} \nabla h_h\right) - \nabla \cdot (p_h \mathrm{u}) + E_{eh} \tag{4}$$

$$\nabla \cdot (\rho_e h_e \mathrm{u}) = \nabla \cdot \left(\frac{\lambda_e}{C_{p_e}} \nabla h_e\right) - \nabla \cdot (p_e \mathrm{u}) + Q_j - E_{eh} - Q_r \tag{5}$$

Here the Joule heating is given by

$$Q_j = \frac{1}{2}\sigma E_\theta \bar{E}_\theta \tag{6}$$

Species conservation:

$$\nabla \cdot (n_s \mathbf{u}) = \nabla \cdot (D_s \nabla n_s) + S_s \tag{7}$$

where plasma species s : N_2, N, N^+, N_2^+, Ar, Ar^+, e, n_s : number density of s species, $D_s = Lewis \times \lambda_s / m_s n_s C_{p_s}$: diffusion coefficient, S_s : source term from ionization, dissociation and recombination.

$$S_s = \begin{cases} \dot{n}_{Ar^+} & s : Ar^+ \\ -\dot{n}_N - \dot{n}_1 & s : N_2 \\ \dot{n}_N + \dot{n}_1 - \dot{n}_{N^+} - \dot{n}_2 & s : N \\ \dot{n}_{N^+} - \dot{n}_1 & s : N^+ \\ \dot{n}_1 + \dot{n}_2 & s : N_2^+ \end{cases} \tag{8}$$

Reactive processes:

$$\left.\begin{array}{rl} N_2 + M \rightleftarrows N + N + M, & \dot{n}_N = k_f n_{N_2} n_M - k_b n_N^2 n_M \\ N + e \rightleftarrows N^+ + e + e, & \dot{n}_{N^+} = k_{ion} n_N n_e - k_{re} n_{N^+} n_e^2 \\ N_2 + N^+ \rightleftarrows N + N_2^+, & \dot{n}_1 = k_{f1} n_{N_2} n_{N^+} - k_{b1} n_N n_{N_2^+} \\ N + N \rightleftarrows N_2^+ + e, & \dot{n}_2 = k_{f2} n_N^2 - k_{b2} n_{N_2^+} n_e \\ Ar + e \rightleftarrows Ar^+ + e + e, & \dot{n}_{Ar^+} = k_{ion} n_{Ar} n_e - k_{re} n_{Ar^+} n_e^2 \end{array}\right\} \tag{9}$$

and also from electrical neutral

$$n_e = n_{Ar^+} + n_{N^+} + n_{N_2^+} \tag{10}$$

equation of state

$$p = \sum \frac{m_s n_s}{M_s} RT_s \tag{11}$$

where M_s : molecular weight of s species.
Vector potential equation:
Using phase vector potential from Maxwell's equations,

$$\nabla^2 \mathbf{A}_c - i\mu_0 \omega \sigma \mathbf{A}_c = 0 \tag{12}$$

$$\mathbf{E}_c = -i\omega \mathbf{A}_c \tag{13}$$

$$\mathbf{B}_c = \nabla \times \mathbf{A}_c \tag{14}$$

2.2 Thermodynamic and transport properties

Each of the thermodynamic and transport properties such as viscocity, specific heat at constant pressure, thermal conductivity, ionization and dissociation rate coefficients [6], electrical conductivity are given by numerical data as a function of temperature. The thermodynamic and transport properties of argon and nitrogen gas mixture are evaluated by mixed gas rule [7].

2.3 Boundary conditions

At the inner surface of the tube the no-slip condition, the thermal condition and the electrical insulation are considered. The thermal equilibrium condition is assumed and nitrogen mole fraction is given at the inlet. The outer surface of the tube is kept 300 K.

2.4 Numerical procedure

A summary of the torch geometry and the operating conditions in the present study is given in the table 1. The distributions of the velocity, temperature and concentration at the nozzle exits are assumed to the top hat in the thermal equilibrium.

Case 1 and case 2 correspond to the flow rate differences from each nozzle especially focusing carrier gas flow rate difference from nozzle 1. A small amount of nitrogen is premixed with argon as a sheath gas or as a carrier gas in the present study.

3. Numerical results and discussion

Figures 2 show the velocity field, heavy particles and electron temperatures, chemical species number densities for different injection locations of argon-nitrogen premixed gas in the case 2 compared with the case of argon only. These figures show the effect of carrier gas flow or sheath gas flow including nitrogen on the thermofluid characteristics and the chemical species distribution with the recirculating flow region. When the nitrogen gas is injected in a sheath gas, the recirculation center shifts toward the inlet coil region and heavy particles and electron temperatures decreases compared with those in the case of only argon, showing larger thermal nonequilibrium condition. This is due to the small production of Joule heating and large dissociation of nitrogen. On the other hand, there is little effect of injection of premixed nitrogen molecule as a carrier gas on the thermofluid field due to the small dissociation and diffusion of nitrogen. The electron and argon ion number densities are basically similar in all cases since there is large ionization of argon in the present study. The maximum degree of ionization is about 0.06 in the recirculating region. Corresponding to the nitrogen injection locations, nitrogen molecule are dense along the inlet wall or in the inlet center region respectively. In the case of nitrogen carrier gas injection, it moves outward in the inlet region due to the blocking of recirculating flow. In the case of nitrogen sheath gas injection, high number densities of nitrogen atom and nitrogen atom ion exist in the recirculating flow region, which does not coincides exactly the high temperature region. This could result from the complex production mechanism of nitrogen species in the convective and diffusive region and also the different rates of ionization and dissociation as a function of temperature. The maximum dissociation degree of nitrogen is about 0.4 in the inlet coil region due to the relatively high dissociation energy of nitrogen. Even small amount of nitrogen molecular ion exists in the recirculation region in the case of sheath gas injection.

4. Conclusions

The present study describes the numerical model for a nonequilibrium RF induction argon-nitrogen plasma with chemical reactions at atmospheric pressure using multicomponent model. The summary of the results obtained here by numerical simulation are as follows.

1. There exists the recirculating flow in the coil region due to the small carrier flow rate and large radial Lorentz force. In this case, high temperature region becomes smaller than the case without recirculating flow region, showing large thermal nonequilibrium condition but trapping some charged plasma species.
2. When a nitrogen gas is injected as a sheath gas, nitrogen gas is easily diffused and then nitrogen atom and nitrogen atom ions are produced due to the active dissociation and ionization in the high temperature recirculating region.
3. The electrons are mainly produced by argon ionization in the present study. As for the nitrogen gas there is mainly dissociation with very little ionization due to the different chemical reaction rates and complex diffusion in the high temperature region.

References

(1) JSME, Functional Fluids and Intelligent Fluids, Corona Pub. Co. Ltd., (2000) (in Japanese).
(2) M. I. Boulos, IEEE Trans. on Plasma Sci., **19** (1999), 1078-1089.
(3) H. Nishiyama, Y. Muro and S. Kamiyama, J. Phys. D: Appl. Phys., **29** (1996), 2634-2643.
(4) T. Watanabe, N. Tonoike, T. Honda and A. Kanzawa, J. Chem. Eng. Japan, **24** (1991), 25-32.
(5) G. Y. Zhao, J. Mostaghimi and M. I. Boulos, Plasma Chem. Plasma Process., **10** (1990), 151-157.
(6) R. N. Gupta, J. M. Yos, R. A. Thompson and P. Lee, NASA Reference Publication, RP1232 (1990).
(7) C. R. Reid, J. M. Prausntiz and T. K. Sherwood, The Properties of Gases and Liquids, McGraw-Hill, (1977).

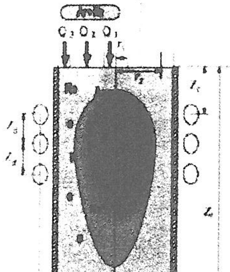

Figure 1 A schematic illustration of the RF induction argon-nitrogen plasma torch and the coordinate system

Table 1 The torch geometry and operating conditions

torch radius	r_w	25 (mm)
nozzle 1 radius	r_1	5 (mm)
nozzle 2 radius	r_2	20 (mm)
torch length	z_l	75 (mm)
wall thickness	δ_w	2 (mm)
coil location	Z_c	15 (mm)
coil space	Z_d	10 (mm)
coil radius	r_c	33 (mm)
frequency	f	3 (MHz)
input power	P	8 (kW)
operating pressure	p	1.01325×10^5 (Pa)
inlet temperature	T_i	350 (K)
wall temperature	T_w	300 (K)
case 1		
carrier gas	Q_1	5 (ℓ/min)
plasma gas	Q_2	4 (ℓ/min)
sheath gas	Q_3	7 (ℓ/min)
case 2		
carrier gas	Q_1	0.01 (ℓ/min)
plasma gas	Q_2	1 (ℓ/min)
sheath gas	Q_3	10 (ℓ/min)
volume fraction of nitrogen gas (carrier)	Q_1	1.8 (%)
volume fraction of nitrogen gas (sheath)	Q_3	1 (%)

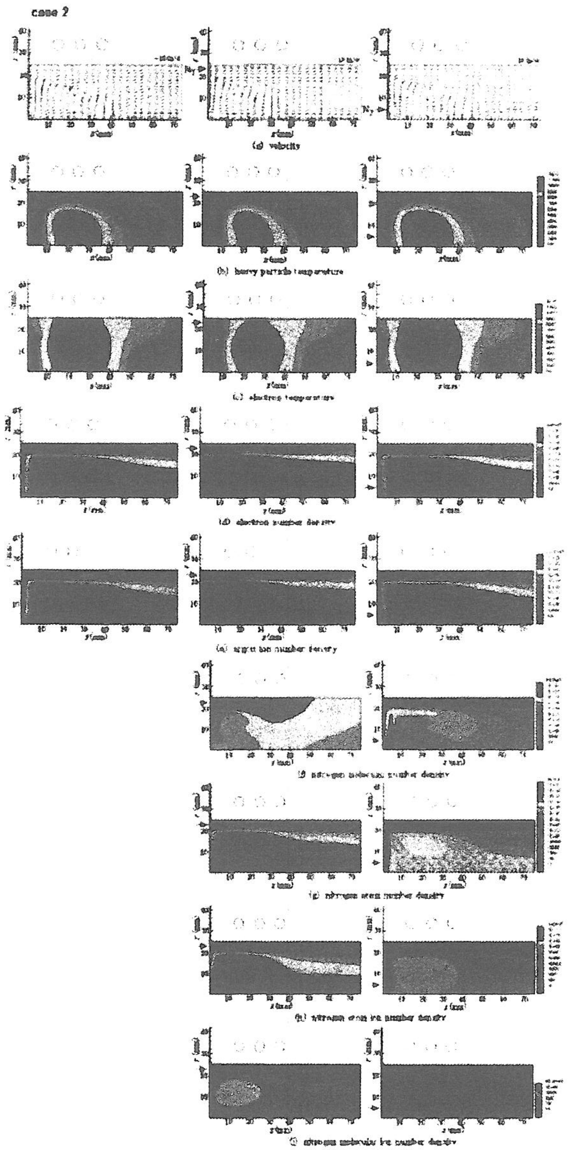

Figure 2 Plasma characteristics for the nitrogen injection locations