Numerical study of heating and evaporation processes of quartz particles in RF inductively coupled plasma

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Abstract. Numerical simulations of heat and evaporation processes of quartz particles in Ar radio frequency inductively coupled plasma (ICP) are investigated. The quartz particles are supplied by the carrier gas into the ICP within gas-cooling. It is shown that with the increase of amplitude of discharge current above critical value there is a toroidal vortex in the ICP torch at the first coil. The conditions for the formation of vortex and the parameters of the vortex tube have been evaluated and determined. The influence of vortex, discharge current, coil numbers and feed rate of carrier gas on the evaporation efficiency of quartz particles have been demonstrated. It was found that the optimal discharge current is close to the critical value when the quartz particles with initial sizes up to 130 µm can be fully vaporized in the ICP torch with thermal power of 10kW. The heat and evaporation processes of quartz particles in the ICP torch have significant importance for the study of one-step plasma chemical reaction method directly producing silicon from silicide (SiO₂) in the argon-hydrogen plasma.

1. Introduction
Currently, in connection with the development of various new techniques [1-4], the interest in the inductively coupled plasma (ICP) steadily increases [5, 6]. The method [1] of one-step plasma chemical reaction directly producing silicon from silicide (SiO₂) in the argon-hydrogen ICP plasma torch is proposed and investigated. In our study the solid particles of SiO₂ are introduced by hydrogen into argon plasma, which is created by the ICP torch. There are three different physic-chemical processes occurring consistently in the heterogeneous flows: 1) evaporation and decomposition of SiO₂ solid particles with the formation of silicon and oxygen atoms; 2) non-equilibrium gas-phase chemical reaction with the binding of oxygen and hydrogen atoms, but without formation of silicon monoxide SiO; 3) condensation of silicon vapor with the formation of polycrystalline silicon particles or films.

Analysis of the results [1] showed that the high purity of the polycrystalline silicon can be acquired by using the electrodeless torches of induction frequency plasma with the fulfillment of optimal heat and evaporation conditions for the solid particles of SiO₂. At present, a wide range of theoretical and experimental studies is needed for the design and optimization of structural and operating parameters of the ICP torches. In this paper, the results of numerical simulation of the heat and evaporation processes of quartz particles in the ICP plasma torch with axial gas-cooling are presented.
2. Physical and mathematical model

It is assumed that heating and evaporation of quartz particles happens in sparse heterogeneous plasma flows. The fundamental equations included Maxwell’s equations, plasma dynamics equations with the account of influence of electromagnetic force and Joule heat, as well as the equations of movement, heat and mass transfer of particles.

The scheme and geometric parameters of ICP plasma torch with axial gases feeds are shown in Figure 1. In our work, the elementary sizes of ICP plasma torch are set as: $Z_2 = 50 \text{ mm}$, $Z_{inf} = 63 \text{ mm}$, $Z_{at} = 123 \text{ mm}$, $Z_4 = 400 \text{ mm}$, $R_1 = 1.7 \text{ mm}$, $R_2 = 18.8 \text{ mm}$, $R_3 = 25 \text{ mm}$, $R_4 = 33 \text{ mm}$, $d_{coil} = 6 \text{ mm}$, $\delta_1 = 3 \text{ mm}$, $\delta_2 = 2.2 \text{ mm}$, $\delta_3 = 3.5 \text{ mm}$. The feed rates of central and sheath gases are $G_z = 1.02 \times 10^{-4} \text{ kg/s}$ and $G_3 = 1.41 \times 10^{-3} \text{ kg/s}$. However, the feed rate of carrier gas ranges from $G_{t_{min}} = 0.87 \times 10^{-6} \text{ kg/s}$ to $G_{t_{max}} = 0.42 \times 10^{-4} \text{ kg/s}$ and the length of the injection probe $Z_i$ ranges from 3 to 50 mm.

![Figure 1. Scheme and geometric parameters of the ICP plasma torch: 1) injection probe of carrier gas and quartz particles, 2) channel of central gas, 3) channel of sheath gas.](image)

The coils ($N = 2, 3$ or $5$) are located at a fixed length $l_{coil} = Z_{at}$, $Z_{inf} = 60 \text{ mm}$ along the ICP quartz pipe. The amplitude of the discharge current $J_{coil}$, varying sinusoidally with frequency $\omega = 3 \text{ MHz}$ was chosen in the range 20–170 A.

In this model, it is assumed that the spiral inductor can be regarded as a system of cylindrically symmetric parallel coils. The states of gas flows at the inlets to the ICP channels are azimuthally symmetrical and stationary. With the fulfillment of these assumptions, the electromagnetic and gas dynamics equations can be solved in a two-dimensional cylindrical coordinate system $(r, z)$ (2D-model).

In the present study, the calculation of gas-dynamic parameters of the laminar ($Re < 10^3$) subsonic axisymmetric stationary plasma flows is carried out on the basis of plasma dynamics equations [7], in which the electromagnetic forces and Joule heat (energy loss by radiation is negligible) have been taken into account. Argon is considered as the carrier, central and sheath gas. The thermal parameters of argon plasma are determined as approximation of local thermodynamic equilibrium (LTE) from known data [8].

At the inlets ($Z = 0$) the temperature ($T = 300\text{K}$) and the axial component of the velocities of central ($U_{z2} = 0.054 \text{ m/s}$) and sheath gas ($U_{z3} = 2.1 \text{ m/s}$) are set constant. The axial component of the carrier gas $U_{z1}$ ranges from 0.054 m/s to 2.1 m/s. At the outlet $Z = Z_4$, constant pressure $P = P_{atm} = 10^5 \text{ Pa}$ is specified.
The calculations of electromagnetic field parameters, intensity of the Joule heat and electromagnetic forces are made on the basis of Maxwell's equations, which is written by the magnetic vector potential [9]. The boundary conditions for the magnetic vector potential are given in [10].

The calculations of the movement, heat and mass transfer of quartz particles in plasma flows with steady distribution of velocity and temperature are carried out by the assumption that: the quartz particles have spherical shape with initial diameter \( d_0 = 50 - 200 \mu m \); the interactions between quartz particles are neglected and the temperature in the particles is equally distributed. The equation system includes:

- momentum equation of quartz particles in plasma flows with a due consideration of aerodynamic drag force, gravity and virtual mass force;
- energy equation of quartz particles, taking into account the convective heat transfer from plasma and the latent heat resulting from the evaporation of particles.

The evaporation of particles is simulated within the framework of diffusion model, in which the coefficient of heat and mass transfer is calculated by Ranz-Marshall formula [11]. The velocity and entry angle \( \beta \) at the outlet of injection probe (relatively to the axis) are determined by the solution of momentum equation of quartz particles. The thermal parameters of quartz particles are determined from the known data [8].

Numerical solution of the equations system was made in ANSYS using finite volume method. The grid model was built in ANSYS ICEM package using HEXA_8 block of hexagonal structure. Detailed descriptions of the application of physical and mathematical model and numerical solution are presented in [12-16].

3. Results and Discussion

It is found that the maximal temperature in the ICP torch ranges from 8 to 10 kK when the total feed rates of carrier, central and sheath gases \( G_1+G_2+G_3 \approx 1.55 \times 10^{-3} \text{ kg/s} \) and the amplitude of discharge current \( J_{col} \) ranges from 20 to 170 A. In order to ensure the normal thermal state of quartz pipe (wall temperature less than 800-900 K), the discharge current \( J_{col} \) in ICP inductor with three and five coils should not exceed 160-170 A and 120-130 A so that the thermal power in the ICP torch does not exceed 10 kW.

It was established that there can be two different kinds of plasma flow patterns in the ICP torch, which appear alternately with the change of discharge current \( J_{col} \). When the \( J_{col} \) is above the critical value \( J_{cr} \), there is a toroidal vortex (Figure 2) in ICP torch at the first coil. The formation of vortex is connected with the appearance of high pressure zone around the ICP axis because of the influence of axial component of electromagnetic force. With the increase of the discharge current, the intensity of the vortex becomes higher. However, when \( J_{col} \) is below the critical value \( J_{cr} \), the electromagnetic force decreases, and the high pressure zone disappears. As a result, the toroidal vortex is not formed. Under this kind of plasma flow pattern, the central and carrier gases flow almost along the ICP axis into high temperature zone.

When the geometry parameters of the ICP-torch and gases feed rates are defined, the critical value \( J_{cr} \) are determined by the specific number of coils \( n = N/f_{col} \). For two (\( n = 0.33 \text{ cm}^1 \)), three (\( n = 0.5 \text{ cm}^1 \)) and five (\( n = 0.83 \text{ cm}^1 \)) coils, \( J_{cr} \) is equal to 110-120 A, 80-90 A and 40-50 A, respectively.

The intensity of vortex depends both on the discharge current, and on the feed rate of carrier gas. The reduce of feed rate of carrier gas from \( 0.42 \times 10^{-4} \text{ kg/s} \) (Figure 2, upper half-plane) to \( 0.87 \times 10^{-6} \text{ kg/s} \) (Figure 2, lower half-plane) can lead to a substantial increase of vortex area and intensity. However, it was found that the length of injection probe \( Z_i \) have low influence on the vortex locations and intensity.
Figure 2. The streamlines in the channels of ICP torch \((N = 3, Z_1 = 3 \text{ mm})\) under \(J_{\text{coil}} = 170 \text{ A}\) and different feed rates of carrier gas: upper half-plane - \(G_{\text{max}} = 0.42 \times 10^{-4} \text{ kg/s} (U_{Z1} = 2.1 \text{ m/s}, U_{Z2} = 0.054 \text{ m/s})\); lower half-plane - \(G_{\text{max}} = 0.87 \times 10^{-6} \text{ kg/s} (U_{Z1} = U_{Z2} = 0.054 \text{ m/s})\).

The evaporation efficiencies of particles \(\varepsilon = 1 - (d_k/d_0)\) \((d_k\) is the diameter of particles at the ICP torch outlet \(Z = Z_t, d_0\) is the diameter of particles at the injection probe inlet\) depends on the discharge current \(J_{\text{coil}}\), plasma flow patterns, initial particle diameter \(d_0\) and entry angle of particles \(\beta\) at the outlet of injection probes. It has been studied that when the particles are transported into the injection probe under the cone angle of \(45^\circ\) at the inlet of injection probe, the range of entry angles \(\beta\) at the outlet of injection probe depends on the initial diameter of particles: \(\beta = 0–10^\circ\) for particles with \(d_0 \leq 100 \text{ \mu m}\), \(\beta = 0–25^\circ\) for particles with \(d_0 \leq 200 \text{ \mu m}\).

As discussed above, the fine particles \((d_0 \leq 100 \text{ \mu m})\) are transported into the ICP torch under entry angle \(\beta \leq 10^\circ\). They fly through the cavity of the vortex or by the streamlines into the high temperature zone. The trajectories of such particles have a spindle shape. Investigations show that particles with diameter \(d_0 \leq 100 \text{ \mu m}\) can be fully vaporized before they fly out of the ICP within coils with the number \(N \geq 3\) and discharge currents \(J_{\text{coil}}\) about 40–50 A.

It was found that large particles \((150 \text{ \mu m} < d_0 \leq 200 \text{ \mu m})\) preserve their velocity at the outlet of injection probe when they fly in the ICP torch despite the different kinds of plasma flow patterns because of their large inertia. The evaporation efficiency \(\varepsilon\) of these particles weakly depends on the discharge current \(J_{\text{coil}}\) and does not exceed 0.2 for the ICP with \(N \leq 3\) and \(J_{\text{coil}} \leq 170 \text{ A}\).

The results show that the evaporation efficiency of particles with \(d_0\) from 100 < \(\leq 150 \text{ \mu m}\) and entry angle of particles \(\beta \leq 10^\circ\) have maximal values when the discharge current \(J_{\text{coil}}\) is close to the critical value \(J_{\sigma}\) (Figure 3, solid lines). There can be several reasons for this.

First, when the discharge current is below the critical value \(J_{\sigma}\), there is no vortex in the ICP torch. The particles fly directly through the high-temperature zones. With the increase of discharge current, the evaporation efficiency \(\varepsilon\) becomes higher due to the rise of plasma temperature and the intensification of heat and mass transfer processes in the ICP torch.

Second, when the discharge current is above the critical value \(J_{\sigma}\), the increase of discharge current can significantly promote the plasma velocity along the axis (by 2 times, then \(J_{\text{coil}} < J_{\sigma}\) because of the formation of vortex in the ICP torch, which respectively leads to a remarkable reduction of particle resident times in the high temperature zone. As a result, the evaporation efficiency \(\varepsilon\) drops. When \(\beta \geq 10^\circ\) the evaporation efficiency of particles with 100 \text{ \mu m} < \(d_0 \leq 150 \text{ \mu m}\) monotonically increases with the growth of discharge current (Figure 3, dashed lines).
Figure 3. The dependence of evaporation efficiency of particles on discharge current under different initial diameters of particles, which are supplied into the ICP torch ($N = 3, U_{Z1} = 2.1 \text{ m/s}, U_{Z2} = 0.054 \text{ m/s}$) under axial direction of injection (solid lines) and cone injection angle $45^\circ$ (at the inlet of injection probe) (dashed lines): 1) $d_0 = 120 \mu\text{m}$; 2) $d_0 = 130 \mu\text{m}$; 3) $d_0 = 150 \mu\text{m}$.

4. Conclusion

There are two different kinds of plasma flow patterns in the ICP torch depending on the discharge current $J_{col}$. When the discharge current $J_{col}$ is above the critical value $J_c$, there is a toroidal flow of vortex in the ICP torch at the first coil. However, if the discharge current $J_{col}$ is below $J_c$, the carrier and central gases flow almost axially into the high temperature zone (without a vortex).

The evaporation efficiency of particles depends on the discharge current $J_{col}$, plasma flow patterns and initial diameter of particles $d_0$. First, the fine particles ($d_0 \leq 100 \mu\text{m}$) can be completely vaporized in the ICP torch with the number of coils $N \geq 3$ and discharge current $J_{col}$ about 40-50 A. Second, for the particles with $100 \mu\text{m} < d_0 \leq 150 \mu\text{m}$, the evaporation efficiency $\varepsilon$ is maximal when the discharge current $J_{col}$ is near the critical value $J_c$ (for $\beta < 10^\circ$). In the case they are injected at $\beta > 10^\circ$, the evaporation efficiency of particles monotonically increases with the growth of discharge current. Third, for the large particles ($150 \mu\text{m} < d_0 \leq 200 \mu\text{m}$), the evaporation efficiency does not exceed 0.2 under the allowable working states (thermal power in ICP $\leq 10 \text{ kW}$).

The ICP torches with gas-cooling and energy power up to 10 kW can be effectively used for heating and evaporating silica particles with maximum size no greater than 150 $\mu\text{m}$. The processes are significant for the plasma-chemical method for producing silicon from quartz particles in argon-hydrogen plasma [17].

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