NUMERICAL SIMULATION OF THE ACTIVATION PROCESS OF SUPERSONIC GAS FLOWS BY A MICROWAVE DISCHARGE

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Diamond films are widely used due to their unique physical and chemical properties, such as a large Young's modulus, the highest among all the materials known in nature, thermal conductivity, high breakdown voltage, transparency in the wavelength range from ultraviolet to infrared and chemical resistance. In the past few decades, there has been an intensive development of the technology of chemical vapor deposition (CVD) of diamond films. Activation of the gas mixture is usually carried out by the methods of hot filament, microwave plasma, arc plasma torches and glow discharge. The presence of electrodes or filament near the substrate affects the quality of the diamond. When using such activation methods, an additional impurity gets into the deposited diamond layer. In this regard, the method of activation of the gas mixture using microwave plasma is one of the most suitable from the point of view of the purity of the diamonds obtained. At the same time, this method allows to achieve high growth rates of diamond films, which makes it even more attractive.

The use of microwave radiation to activate gas mixtures containing hydrogen and carbon in the diamond synthesis is widespread. In the process of diamond film deposition, generation of a plasma cloud above the substrate is most frequently used. From that cloud active molecular fragments diffuse to the surface of the substrate [1]. However, a new deposition method has recently emerged, in which active fragments formed in a plasma cloud flow from a resonant chamber into a vacuum deposition chamber in form of a supersonic jet [2]. This method allows the use of plasma at very high pressures.

The study and optimization of microwave plasma generation process is a difficult task for modern experimental physics, due to the presence of extreme temperatures, high rate reactions and complex geometry of the setup. In this regard, numerical simulation may be very helpful as it allows testing the effect of many different parameters on the efficiency of the system in a short time. A popular tool for simulating the microwave plasma dynamics is COMSOL Multiphysics. Previously, this program was used for modeling of diamond deposition with microwave plasma activation [3]. Also, this software package was used to simulate the flow in a space thrusters [4].

It should be noted that the presence of atomic hydrogen in the active zone ensures efficient growth of diamond films. Atomic hydrogen quickly converts graphite, formed in the process of the reactions, again into gaseous hydrocarbon, practically without affecting the diamond. Atomic hydrogen is created mainly by thermal dissociation and electron impact reactions with H_2 molecules. Each of the processes prevails in its temperature range. When using microwave radiation of high power (about 3000 W), the resulting plasma has a temperature of about 3000-6000 degrees. Under such conditions, the process of thermal dissociation of hydrogen is dominant.

It is well known that the microwave radiation field has inhomogeneous distribution in the volume of resonant chamber. Because of this, with the fixed net radiation power, the magnitude of the electric field may vary significantly in different parts of the chamber. Thus,

in the optimization of microwave heating device, the right choice of the resonant chamber geometry plays an important role.

In this paper, we present the results of optimization of the geometrical parameters of the resonant chamber achieved by means of numerical simulation. A baseline chamber design was chosen to mimic an experimental setup from [2].

The baseline case was a cylindrical chamber with an outlet nozzle in the bottom wall in a shape of a 1mm hole. To optimize the microwave heating process, additional calculations were performed with an outlet nozzle in the shape of a truncated cone and a hemisphere playing the role of a microwave field (and heating) concentrator in a resonant chamber.
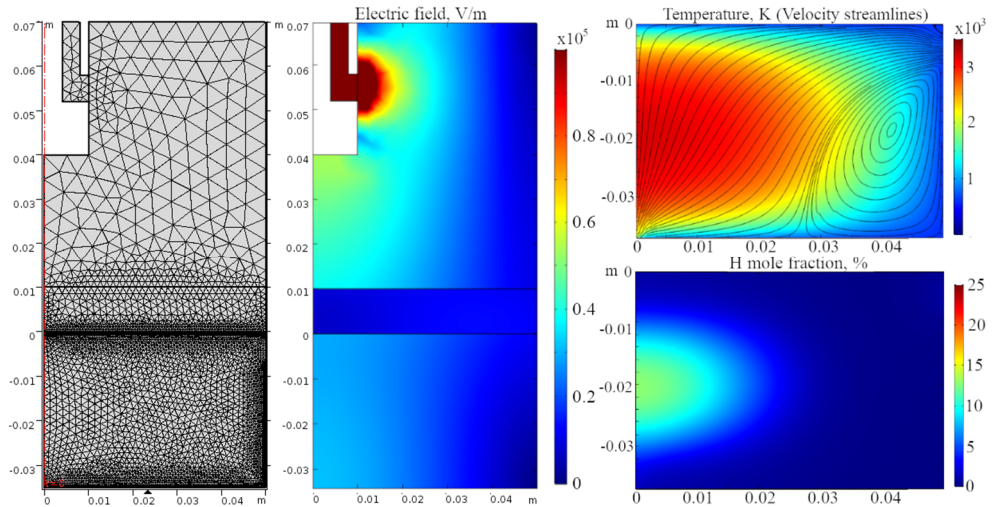

Fig. 1. Simulation grid (left), electric field (middle), temperature and dissociation degree (right) distributions for the case without the nozzle modifications.

The simulations were conducted with the finite element software COMSOL Multiphysics. The simulations were carried out in axisymmetric formulation. A cylindrical resonant chamber was chosen as computation domain (Fig. 1). The diameter and height of the chamber were 100 mm and 145 mm, respectively. Microwave radiation was supplied through a coaxial port located in the center of the top base of the resonant chamber. The radiation frequency was 2.45 GHz, and the deposited power was ~1.5 kW. Walls of the chamber were set perfectly conducting. Microwave radiation was used to heat the working gas. Molecular hydrogen was chosen as the working gas. The gas was flowing through the bottom part of the resonant chamber, separated by a quartz wall. The gas enters the chamber through a hole in the side wall, with a constant flow rate of 10 l/m. The diameter of the exit hole was 1 mm. A fixed pressure of 2 Torr was set at the exit hole, while inside the chamber the pressure was sustained at the level of about 300 torr. At the metal boundaries a fixed temperature of 500K was set (according to preliminary experimental measurements). The computational grid consisted of about 16,000 triangular nodes (Fig. 1 left). The grid was refined toward the domain boundaries. The turbulent fluid dynamics was simulated using the standard k-ε model. Due to the fact that the flow behind the nozzle was not resolved, and the gas velocity in the chamber didn't exceed 300 m/s, it was possible to use the sub-sonic compressible fluid approximation (Ma <0.3). The rates of hydrogen dissociation reactions were taken from [5] presented in Table 1. Dissociation energy, denoted in the table as D,

was 435 kJ/mol. Forward sticking coefficient for surface reaction $H + H = H_2$ was 0.03. This value of a forward sticking coefficient is typical for some metals. At the initial moment of time, the whole chamber was filled with molecular hydrogen, all atomic hydrogen was obtained as a result of reactions.

Table 1. Rate coefficients for hydrogen dissociation and recombination reactions.

Reaction	Rate coefficient, $cm^3 mole^{-1} \sec^{-1}$
$H_2 + H_2 = H_2 + 2H$	$k = 1.8 * 10^{17} T^{-1.5} \exp(-D/RT)$
$H_2 + H = 3H$	$k = 1.2 * 10^{20} T^{-0.5} \exp(-D/RT)$
$H + H = H_2$	$k = 1.66 * 10^{-3} T^2$

The simulations were carried out in a segregated manner. At the first stage, the stationary velocity distribution in the chamber was found for the given temperature field distribution inside the chamber. At the second step, the equations describing the microwave heating of the gas, the equations of thermal dissociation and recombination of hydrogen, and the equations of the heat transfer for the mixture were jointly solved, taking into account the velocity distribution obtained at the first stage. Then these two steps were repeated until the equilibrium solution was found.

To verify the model, we simulated the equilibrium thermal dissociation of hydrogen for various temperatures in the interval from 2000 to 5000K. The simulation results showed a good convergence with reference data [6]. Comparison of the equilibrium values of the degree of thermal dissociation for different temperatures with the data from the literature are presented in Table 2.

Table 2. Equilibrium thermal dissociation.

Temperature, K	H2 dissociation degree (reference)	H2 dissociation degree(simulated)
2000	0.088	0.13
2500	1.31	1.33
3000	8.34	8.38
3500	29.6	29.5
4000	63.9	63.8
4500	82.3	81.6
5000	95.8	93.1

At the first stage, simulations were carried out for the surrounding space of the nozzle in the form of a hole in the center of the lower base of the computational domain. Under such conditions, the electric field is not focused near the exit nozzle, but is almost uniformly distributed over the entire chamber with a magnitude of 15kV/m. In this case, the temperature distribution is determined by the distance from the walls (due to the wall heat losses), and therefore the maximum temperature and degree of dissociation, equal to 3190 K and 12% respectively, are located in the center of the chamber, which significantly reduces the amount of atomic hydrogen at the nozzle exit. Near the exit nozzle in this case, the temperature was only 2350K and the degree of dissociation was 2.1%.

To achieve greater focusing of the electric field near the exit nozzle, it was decided to change the shape of the exit nozzle by adding a hemisphere or a truncated cone, that both also had a 1 mm exit hole (same as the initial nozzle). Figure 2 presents the simulation results for cases with nozzles in the form of a hemisphere and a truncated cone with different values of the radius of the upper base. It was found that the field focuses on the edge of a truncated cone, and as its upper base radius increases, the field maximum moves away from the exit hole, which indicates a decrease in the efficiency of microwave heating. It was found that optimal focusing of the electric field near the exit hole for a cone type nozzle takes place when the upper base radius is 2 mm. The smaller upper base radius would be harder to achieve from an engineering point of view because the hole should be made in a thin tungsten foil wrapped around the cone and it would not give much of additional electric field strength. The field strength near the nozzle for this case was about 65kV/m, the maximum temperature was 3420K and the corresponding value of the dissociation degree near the nozzle exit was 23.2%.
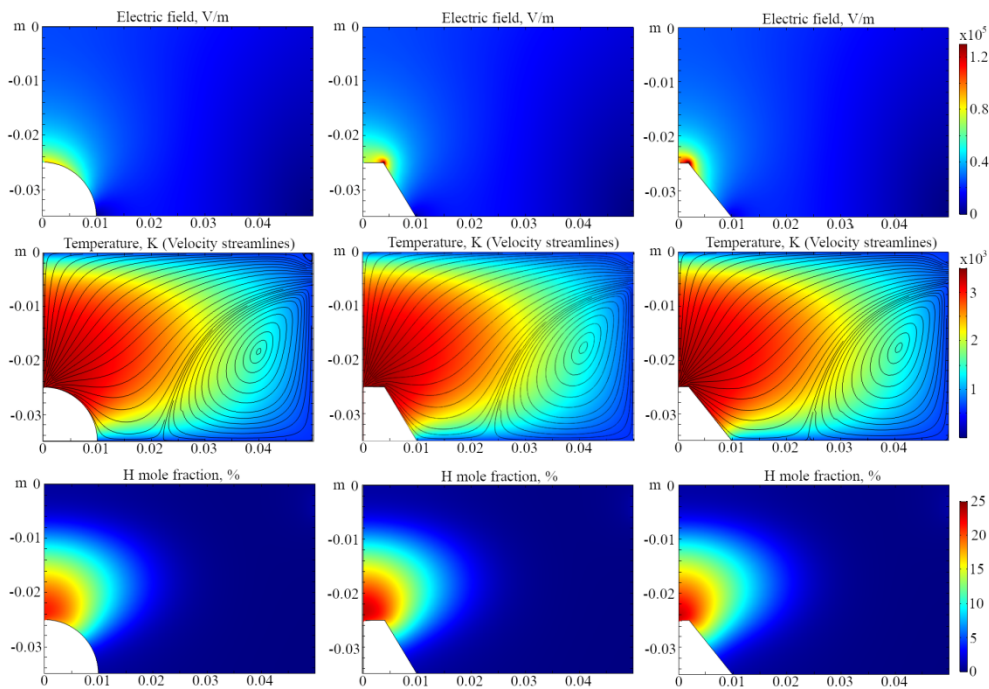

Fig. 2. Electric field, velocity, temperature and dissociation degree distributions for cases with hemisphere and a truncated cone (with 2mm, 4mm top base radius) nozzles.

A hemisphere nozzle was considered attractive as it reduces the recombination losses on the inner walls of the nozzle compared to a cone and at the same time maintains high field strength. It is observed that the maximum field strength in this case reaches 60 kV/m. The maximum temperature in this case was 3400K and the corresponding value of the degree of dissociation was 22.8% at the nozzle exit which is only slightly less than for a cone. Figure 2 shows that the change in flow of gas does not significantly affect the temperature distribution and concentration of atomic hydrogen for the used flow rates. This is associated with a significant difference in the characteristic time scales for fluid dynamics and for the processes of dissociation and recombination.
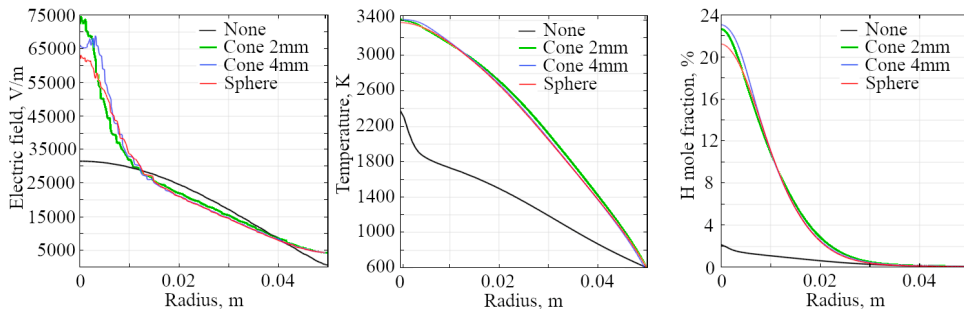
Fig. 3. Electric field, temperature, H mole fraction profiles through a section located 2 mm from the exit hole.

The profiles in Figure 3 show the efficiency of using additional nozzles. Using a truncated cone or hemisphere as a nozzle gives a gain in the electric field and temperature by a factor of two relative to the base case. It is important to note that the dissociation degree depends on temperature non-linearly, and when using an additional nozzle, the degree of dissociation increases by almost 10 times.

Also, it is necessary to make a nozzle (or at least a part of it near the exit hole) from a thin and very heat-resistant material (e.g. tungsten). In this case, higher temperatures could be achieved and the recombination rate on such hot wall will be much lower.

The preliminary experimental tests with setup from [2] showed that such modification (either cone or hemisphere) increased a working temperature inside the chamber significantly and made the discharge more stable even for higher pressures (up to 300 torr. and more).

First experimental results on the diamond deposition with using analyzed external nozzle configurations have been positive and encouraging. These tests with setup from [2] showed that such modification (either cone or hemisphere) increased a working temperature inside the chamber significantly and made the discharge more stable even for higher pressures (up to 300 torr. and more).

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