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Supersonic plasma outflow in a plasmochemical method of amorphous silicon thin films formation

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Abstract. As a result of the numerical modeling of gasdynamic functions of a nozzle of Laval there obtained its parameters which form supersonic plasma jet outflow in a process of amorphous silicon thin films deposition. According to the nozzle design parameters, there obtained amorphous silicon thin films and studied uniformity of the thickness of the synthesized coatings. It was also performed that due to a low translational temperature at the nozzle exit the relaxation losses reduce significantly, "freezing" the vibrational degrees of freedom and the degrees of freedom of the transverse motion of the particles, and increasing the energy efficiency of the film formation process. All this is caused by the fact that on the surface of a growing film only the products of primary interaction of electrons with molecules of a silicon-containing gas in the plasmatron do interact.

1. Introduction

Among other methods of hydrogenous amorphous silicon thin films (a-Si:H) synthesis the most perspective ones are supposed to be jet methods nowadays. They differ according to the way of gaseous reagents activation [1, 2]. Traditional methods of a-Si:H and mc-Si thin films deposition based on the use of plasma flows of various electrical discharges limit the development of electronics of large areas and have a relatively low growth rate of the films. Any attempts to increase growth rates lead to a decrease in the quality of the produced films. It is suggested to overcome the disadvantages of traditional methods using the method of producing amorphous silicon films based on silane decomposition in plasma of a radio-frequency capacitive discharge (RFC-discharge) outside the disposition chamber with the following formation of supersonic jets from the decomposition products that outflow into the vacuum deposition chamber through a system of supersonic nozzles.

The aim of the study is to increase the effectiveness of technological deposition processes of amorphous silicon thin-film coatings basing on the jet plasmochemical method with a preliminary activation of a gas in the area remoted from the growth chamber [3].

2. Problem Definition

To create supersonic outflow in order to deliver the decomposition products of the silanecontaining gas mixture from the plasmatron to the substrate and to freeze recombination processes during the delivery there were made calculations of parameters of a nozzle of Laval (figure 1) and the ones of gasdynamic functions of the pressure, temperature and speed of gas in the jet (figures 2–5).

Calculations of pressure and temperature distributions along the nozzle length are necessary for determination of the initial conditions at which the outflow of the silane decomposition products in the plasmatron to the film deposition substrate is carried out by a supersonic jet, which provides the «freezing» of the vibrational and transverse degrees of freedom according to the direction of motion. The products of primary interaction of electrons with molecules of a silicon-containing gas in the jet do not interact with each other.

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A nozzle of Laval, which forms supersonic outflow of silane dissociation products to deliver a

A nozzle of Laval, which forms supersonic outflow of silane dissociation products to deliver a «frozen» composition of the dissociation products in the plasmatron to the film deposition substrate, is profiled by means of certain gasdynamic calculations [4].

To analyze the gas flow in a nozzle of Laval the following assumptions were made:

1. The gas is considered to be ideal.

2. The gas flow is isentropic and adiabatic.

3. The gas flow is stationary and one-dimensional, all flow parameters at any point of the nozzle vary only along the axis of the nozzle.

- 4. Mass flow rate of the gas in all cross sections of the flow is the same.
- 5. The influence of all external fields (including gravitational) and forces is neglected.

6. The nozzle symmetry axis is a spatial coordinate.

The following initial parameters were used for calculations:

 $-P_1=26.66$ Pa - pressure in the plasmatron chamber;

- $-a_1 = 40^\circ \text{convergence angle};$
- $-a_2 = 15^{\circ} angle of expansion;$
- $-T_1$ =500 K stagnation temperature;
- -R=207.85 J/(kg·K) universal gas constant;
- $-G=3.10^{-7}$ kg/s mass flow of the gas;
- $-P_2=10^{-2}$ Pa pressure in the environment;
- -k = 1.67 adiabatic index.

3. Results and discussion

Stagnation parameters are such gas parameters which correspond to the zero-flow velocity. The relationship between the stagnation parameters and the flow ones can be determined by means of gasdynamic functions: P – pressure, T – temperature, V – velocity.

One of the gasdynamic function arguments will be the Mach's number which is also supposed to be local and depends on the coordinate: x: M=V/a, where V is a local flow velocity, a is a speed of a sound in the gas. Another argument is a velocity coefficient which is also known as reduced velocity λ (the ratio of the speed of gas in the given section of the nozzle to the speed of a sound in the critical section):

 $\lambda = V/a_{cr}$,

where a_{cr} is the speed of a sound in the critical section.

The mass flow rate G is related to the critical section area S_{cr} , the pressure P_1 and the gas temperature T_1 as follows (1) [4]:

$$G = \frac{P_1}{\sqrt{T_1}} S_{cr} \left(\frac{2}{k+1}\right)^{\frac{k+1}{2(k-1)}} \sqrt{\frac{k}{R}} .$$
 (1)

The critical section S_{cr} and the length of the subsonic part of the nozzle l_u are expressed from the flow rate equation.

Parameters in any section of the nozzle can be found if the parameters in the plasmatron chamber are known. They can be calculated from the following relationships:

pressure:

$$\pi = \frac{P}{P_*} = \left(1 - \frac{k-1}{k+1}\lambda^2\right)^{\frac{k}{k-1}}.$$
(2)

temperature:

$$\frac{T_*}{T} = 1 + \frac{k-1}{2}M^2.$$
 (3)

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One more gasdynamic function used in the calculations is specific time flow $q(\lambda)$ which is expressed by means of the ratio of the areas occupied by a non-viscous core in the critical and analyzed sections,

$$q(\lambda) = \left[\frac{d_{cr} - 2\delta_{cr}^*}{d - 2\delta^*}\right]^2,\tag{4}$$

where d_{cr} , and d are the diameters of the critical and analyzed sections (in mm), δ_{cr}^* and δ^* are the boundary layer displacement thicknesses for the critical and analyzed sections (in mm) respectively.

As a first approximation the values of the boundary layer displacement thickness δ_{cr}^* and δ in the calculations of gasdynamic functions are assumed to be zero. In the following approximations their values are obtained due to boundary layer calculation.

The gasdynamic function takes its maximum value $q(\lambda)=1$ in the critical section. In this section the velocity coefficient has the value of $\lambda=1$. The relation between the velocity coefficient λ and the function $q(\lambda)$ is expressed as follows:

$$q(\lambda) = \lambda \left[\left(1 - \frac{k-1}{k+1} \lambda^2 \right) \left(\frac{k+1}{2} \right) \right]^{\frac{1}{k-1}},\tag{5}$$

For the diverging section the expressions for the supersonic section S_d and the length of the sound part of the nozzle l_d are valid:

$$d_d = 2\sqrt{\frac{S_d}{\pi}},\tag{6}$$

$$l_{d} = \frac{(d_{u} - d_{d})}{\left(2 \ tn \frac{\alpha_{2}}{2}\right)},\tag{7}$$

Other parameters can be calculated according to the section length. The flow in the converging section of a nozzle of Laval moves with a subsonic velocity where the velocity coefficient has the value of $\lambda < 1$. The convergence of the result is provided by the scheme:

$$\lambda = \frac{q(\lambda)}{\left(\frac{k+1}{2}\right)^{\frac{1}{k-1}} \left(1 - \frac{k-1}{k+1}\lambda^2\right)^{\frac{1}{k-1}}}$$
(8)

the Mach's number:

$$M = \sqrt{\frac{\frac{2}{k+1}\lambda^2}{1 - \frac{k-1}{k+1}\lambda^2}}$$
(9)

The critical velocity is:

$$a_{cr} = \sqrt{\frac{k}{k+1}RT^*} \tag{10}$$

 T^* is the temperature of the stagnated flow.

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Figure 1. Profile of a nozzle of Laval.

As a result of the numerical modeling of the supersonic nozzle the following values of its parameter are obtained (figure 1):

The length of the subsonic part of the nozzle constituted $l_u=1.6$ mm, the general length was l=50 mm, the areas of the subsonic part, the critical and the diverging sections of the nozzle had the values of $S_u=4.1$, $S_{cr}=1.5$, $S_d=140$ mm² respectively.

Figures 2-5 presents the charts of the dependence of the speed of gas and the speed of a sound, the Mach's number, the pressure and the temperature on nozzle length (where the initial point is 0):



Figure 2. Dependence of speed of gas and speed of a sound on nozzle length.

The speed of gas increases in the area of the critical section (vertical dashed line) (see figure 2). It reaches the value of 900 m/s along the length of the nozzle. The Mach's number (figure 3) reaches the value M=6 which is not something unusual for such kinds of pressures [5].

Figure 4 shows that the condition of the supersonic outflow is being fulfilled:

$$P_{cr} = P_* \left(\frac{2}{k+1}\right)^{\frac{k}{k-1}} > P_e, \tag{11}$$

where P_* is the pressure in the chamber (stagnation pressure); P_{cr} is the pressure in the critical section of the nozzle; P_e is the pressure in the environment; k is the heat capacity ratio.

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Figure 3. Dependence of Mach's number and speed of a sound on nozzle length.



Figure 4. Gasdynamic function of pressure on nozzle length.

The analysis of gasdynamic temperature function on nozzle length (figure 5) shows that due to a low value of translational temperature at the nozzle exit the relaxation losses reduce significantly, «freezing» the vibrational degrees of freedom and the degrees of freedom of the transverse motion of the particles, and increasing the energy efficiency of the process.



Figure 5. Chart of gasdynamic function of temperature on nozzle length.

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To measure the dynamic pressure in the free stream of the flowing plasma of the argon a Prandtl tube and thermistor pressure sensors were used [6]. The results of the measurements are presented in figure 6. The measurements were made without a sample (substrate). The chart shows that the pressure decreases while it spreads from the nozzle which is demonstrated by the modeling calculations as well.



Figure 6. Dependence of the dynamic pressure in the free stream of the flowing plasma of the argon from the distance from the nozzle.

One should remember that thermistor pressure sensors have a relatively low accuracy and their measurement error constitutes \pm (10%–60%). Thus, the obtained results are rather of a qualitative nature.

The deposition of amorphous silicon thin films was carried out in a vacuum unit NNV6.1. The glow-discharge plasma was excited in a plasmatron with a diameter of 70 mm, and a length of 30 mm. Parameters of nozzle: $S_u=4.1$, $S_{cr}=1.5$, $S_d=140 \text{ mm}^2$. The pressure in the chamber varied within 26.66 Pa and was controlled by the capacitive vacuum sensor SETRA 760. Radio frequency glow discharge was excited by the generator with the oscillating power 600 W (field intensity constituted E = 50 V/cm, frequency f =13.56 MHz). Two sliding vane rotary pumps and a diffusion pump evacuated the vacuum chamber till the ultimate vacuum pressure of $6 \cdot 10^{-4}$ Pa. Mass flow controller allowed to control the flow of the working gas within the range of 4-60 sccm. After the pressure in the plasmatron chamber was increased to the performance range, the glow discharge was excited.

Figure 7 demonstrates the results of thickness measurements of the amorphous silicon films from distance from the center. The deposition was carried out for different values of the distance from the nozzle to the substrate (30 and 50 mm), the deposition time was 20 min. It is clearly seen that for the case of 30 mm the films have a highly uneven thickness due to the uneven plasma flow density. There is also a large pressure drop in the center and along the edges of the jet. If we put the substrate in the distance of 50 mm from the nozzle edge, the drop in pressure will constitute approximately 30 % and on the substrate there will be an area with a radius of 60 mm. However, in this case the deposition velocity decreases sharply which can be explained by a decrease of the local gas density near the substrate. To get uniform thickness coatings with a non-uniform coating thickness of less than 5 %, it is suggested to install more than one nozzle and even a nozzle array arranged so that gas jets intersect 60% of the distance from the nozzle to the substrate [7].

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Figure 7. Film thickness from distance from the center.

Conclusions

Calculations of pressure and temperature distributions along the length of the nozzle prove that the delivery of silane dissociation products in the plasmatron to the substrate is accomplished by a supersonic jet. It was also performed that due to a low translational temperature at the nozzle exit the relaxation losses reduce significantly, "freezing" the vibrational degrees of freedom and the degrees of freedom of the transverse motion of the particles, and increasing the energy efficiency of the film formation process. All this is caused by the fact that on the surface of a growing film only the products of primary interaction of electrons with molecules of a silicon-containing gas in the plasmatron do interact. When using the nozzle array, the supersonic outflow is maintained by the increase of the mass flow rate of the gas.

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