"Supersonic Gas Jets Activated by an Electron Beam"

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Supersonic gas jets activated by an electron beam are used in plasma chemistry for thin films deposition of SiO_2, a-Si:H, etc [1-2].

This method has several advantages in operating, controlling and investigating the deposition process, and embodies two modern tendencies in plasma chemistry: a) the use of supersonic jets for direct and quick gas transportation to the surface on which plasma-chemical process takes place; b) the use of electron beams for the activation of gas phase or surface reactions instead of the conventional DC or RF discharges.

A very interesting feature of the method is that the activating electron beam (E_b=10keV, J_b=10mA) has the power P_b=100W but deposits into a gas jet less than 1W (two or three orders of magnitude less than in conventional CVD methods). However, relatively high growth rates (~2nm/s) have been achieved [2].

Another important feature of the method is very small residence time of activated particles on their way from a source nozzle to a substrate: $\tau=10^{-4}$sec. In low density free jets, it leads to an important conclusion: in the first approximation, film growth is determined by surface processes and the flow rates of molecules activated by an electron beam and secondary electrons in a free jet.

In this paper, a model for the method of electron-beam activation of gas jet is presented. It takes into account electron energy distribution function generated by an electron beam (in Ar-SiH_4 nonuniform supersonic jet). The model reveals ways to increase the method efficiency by optimizing E-beam energy, gas jet parameters, and SiH_4/Ar ratio, and outlines the advantages and limits of the method.

1. Integral Boltzmann Equation for secondary electrons.

For the determination of EEDF spatial dependence in steady state conditions, it is necessary to solve integral Boltzmann equation for electron distribution function with

a source term describing secondary electrons generated in gas by an electron beam:

$$f(\vec{r},e) = \alpha\int\frac{d^3\vec{r}'}{4\pi}\,\frac{\exp[-\alpha\sigma_t(e)\tau(\vec{r},\vec{r}')]}{|\vec{r}-\vec{r}'|^2}\,n_g(\vec{r}')\cdot \qquad (1)$$

$$\Big\{\chi(\vec{r}')q(E_b,e) + \sigma_m(e)f(\vec{r}',e) + \sum_j^{E_m}\sigma_j(e+\varepsilon_j)f(\vec{r}',e+\varepsilon_j) +$$

$$+\int_{e+I}^{2e+I} d\varepsilon\sigma_i(\varepsilon)q(\varepsilon,\varepsilon-e-I)f(\vec{r}',\varepsilon) + \int_{2e+I} d\varepsilon\sigma_i(\varepsilon)q(\varepsilon,e)f(r',\varepsilon)\Big\}$$

where $q(E,e)$ is the spectrum for the generation of secondary electrons with the energy e during molecule ionization by the electrons with the energy E normalized by the condition

$$\int_0^{(E-I)/2} q(E,e)de = 1$$

$\chi(\vec{r})=n_b(\vec{r})/n_b$ is the dimensionless function of electron beam profile; n_b is a characteristic number density of primary electrons on the beam axis; $n_g(\vec{r})$ is gas density normalized by n_0. In Eq.(1), all quantities are normalized in the following way: cross sections- by the value of $\sigma_0=10^{-16}cm^2$; energies - by 1 eV; distances - by characteristic radius of electron beam, r_b; and dimensionless parameter $\alpha=n_0r_b\sigma_0$ is introduced.

The quantity $\tau(\vec{r},\vec{r}')$ is proportional to the "optical length" and equal to

$$\tau(\vec{r},\vec{r}') = |\vec{r}-\vec{r}'|$$

in a constant density gas.

EEDF, $f(\vec{r},e)$, is normalized in the following way

$$n_s(\vec{r},e)/n_b = \sigma_i(E_b)\int_e^{E_m} de(E_b/e)^{0.5}f(\vec{r},e),$$

where $n_s(\vec{r},e)$ is number density of secondary electrons having energy greater than e at point $\vec{r}$. Then, the excitation rate of j-th molecule state will be equal to

$$F_j(\vec{r}) = \Phi_{bi}\int_{\varepsilon_j}^{E_m} de\sigma_j(e)f(\vec{r},e), \quad \Phi_{bi}=n_gn_bV_b\sigma_i(E_b),$$

where Φ_{bi} is the rate of ionization by the primary electrons; $E_m=(E_b-I)/2$. For simplicity, we will suppose that all cross sections are isotropic ones. For the elastic scattering (j=m), we will take momentum cross section $\sigma_m(e)$ with the average energy loss $\varepsilon_m=(2m/M)$ in collision

with molecule having mass M. $\sigma_i(e)$ is ionization cross section, I is ionization threshold, $\sigma_t(e)=\sigma_m(e) + \Sigma_j\sigma_j(e) + \sigma_i(e)$ is the total cross section of electron-molecule collision. Such choice corresponds to the conventional approximation of distribution function expansion into spherical harmonics.

It is convenient to solve Eq.(1) for constant gas density using Fourier-Bessel transformation. Secondary electron distribution function then can be expressed in the form (in a uniform gas)

$$f(\vec{r},e) = \int \frac{d^3\vec{r}'}{4\pi} \frac{\chi(\vec{r}')\tau(\vec{r},\vec{r}')^2}{|\vec{r}-\vec{r}'|^2} G(\vec{r},\vec{r}',e), \qquad (2)$$

where $G(\vec{r},\vec{r}',e)$ is the Green's function of the initial Boltzmann equation which is equal to

$$G(\vec{r},\vec{r}',e) = \frac{2}{\pi} \int_0^\infty d\nu\nu \frac{\sin[\nu\tau(\vec{r},\vec{r}')]}{\tau(\vec{r},\vec{r}')} \rho(\nu,e). \qquad (3)$$

The function $\rho(\nu,e)$ satisfies the equation

$$\sigma_t(e)\rho(\nu,e) = A(\nu,e)\Big\{q(E_b,e) + \sum_j \sigma_j(e+\varepsilon_j)\rho(\nu,e+\varepsilon_j) \qquad (4)$$

$$+ \int_{e+I}^{2e+I} d\varepsilon\sigma_i(\varepsilon)q(\varepsilon,\varepsilon-e-I)\rho(\nu,\varepsilon) + \int_{2e+I}^{E_m} d\varepsilon\sigma_i(\varepsilon)q(\varepsilon,e)\rho(\nu,\varepsilon) \Big\},$$

$$A(\nu,e) = \frac{\alpha\sigma_t(e)}{\nu} \operatorname{arctg}\left[\frac{\nu}{\alpha\sigma_t(e)}\right], \quad A(\nu=0,e) = 1$$

Eq.(2) with the Green's function (3) represents an "exact" solution of the Boltzmann integral equation (1) for an arbitrary spatial secondary electrons source of generation. The function $\rho(\nu,e)$ is the Fourier-Bessel image or a "spectral function" of the distribution function $f(\vec{r},e)$ and must be determined from the equations (4).

2. Diffusional approximation.

As it is seen from the solution of Eq.4, the dependence of spectral function $\rho(\nu,e)$ on ν/α at given energy e has a characteristic form: for small ν/α it is equal to the almost constant value $\rho(0,e)$ and then it transfers to the asymptotic dependence corresponding to the equation (4) for large values ν/α. The analysis of the numerical solution shows that spectral function $\rho(\nu,e)$ can be approximated by simple Lorentz's dependence with the accuracy higher than 10% for the whole range of values ν/α:

$$\rho(\nu,e) \approx \rho_1(\nu,e) + \rho_d(e)/[1+(\nu/\alpha\sigma_d(e))^2] \qquad (5)$$

where $\rho_1(\nu,e) = A(\nu,e)q(E_b,e)/\sigma_t(e)$; $\sigma_d(e)$ and the function $\rho_d(e)$ can be obtained from Eq.(4):

$$(\sigma_d(e) \sim \sigma_t(e)/3);\ \rho_d(e) \approx \rho(\nu=0,e)$$

In the range of "frequencies" $\nu \leqslant \alpha\sigma_t(e)$ which makes an essential contribution to the value of integral (3), the inequality $\rho(\nu,e) \gg \rho_1(0,e)$ is fulfilled usually. Thus, $\rho_d(e) \approx \rho(0,e)$. The distribution function $f(\vec{r},e) \approx f_d(\vec{r},e)$ after integration (3) can be presented as follows

$$\frac{f_d(\vec{r},e)}{\rho_d(e)(\alpha\sigma_d(e))^2} = \qquad (6)$$

$$= \int \frac{d^3\vec{r}'}{4\pi} \frac{\chi(\vec{r}')n_g(\vec{r}')\tau(\vec{r},\vec{r}')}{|\vec{r}-\vec{r}'|^2} \exp[-\alpha\sigma_d(e)\tau(\vec{r},\vec{r}')]$$

Thus $f_d(\vec{r},e)$ is diffusionlike distribution function of secondary electrons produced by an electron beam. Expression (6) is modified to take into account a spatial nonuniformity of a gas.

3. Gas jet activation.

Further, we will consider the case of silane - argon mixture expansion from the sonic nozzle, d_* is its diameter. An electron beam crosses the gas jet at z_b (Fig.1). Silane and argon concentrations are equal to c_S and c_A ($c_S \leqslant 0.1 \ll c_A \sim 0.9 - 1.$).

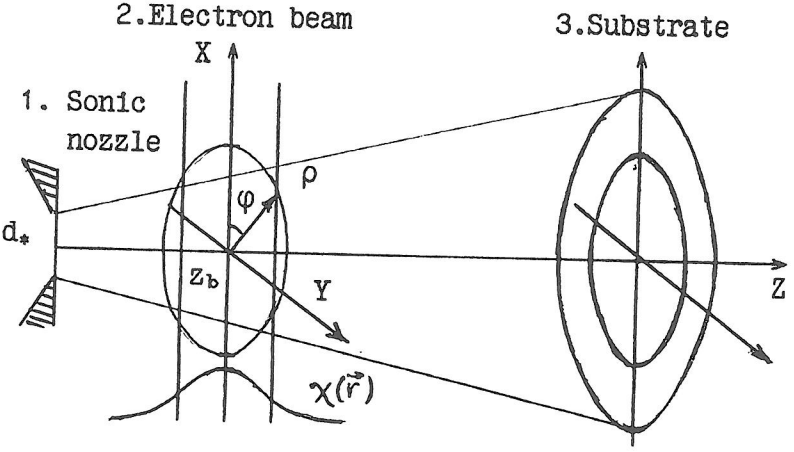

Fig. 1. Gas jet activated by an electron beam.

One atomic gas density distribution along the free jet axis, z, and in the radial direction, ρ, for $z \gg d_*$ can be approximated by the expression

$$n_g(z,\rho) = (n_0/2\pi)\ (d_*/z_b)^2/(z^2+\beta^2\rho^2), \qquad (7)$$

where $\beta \approx 2$, n_0 is stagnation density of argon-silane mixture. Variables z and ρ given below are normalized by z_b.

The "optical length" for secondary electrons in supersonic gas jet is approximately equal to

$$\tau(\vec{r},\vec{r}') \approx \frac{|\vec{r} - \vec{r}'|}{\tilde{r}\ \tilde{r}'}\ , \qquad (8)$$

$$\tilde{r} = \sqrt{z^2+\beta^2\rho^2}, \qquad \tilde{r}' = \sqrt{z'^2+\beta^2\rho'^2}$$

Equations (6) can integrated for density distribution (7). Then excitation rate for j-level of x-kind molecule can be expressed as

$$\frac{F_j^x(\vec{r})}{n_b v_b \sigma_i(E_b)} \approx c_x \Phi_j^x \int \frac{d^3\vec{r}'\chi(\vec{r}')}{4\pi\ \tilde{r}\ \tilde{r}'} B^2\ \frac{\exp(-B|\vec{r} - \vec{r}'|)}{|\vec{r} - \vec{r}'|} \qquad (9)$$

$$B = \frac{n_0\sigma_0}{2\pi}\ \frac{d_*^2}{z_b^2}\ \frac{\sigma_d(X,j)}{\tilde{r}\ \tilde{r}'}\ ,$$

where Φ_j^x is the excitation rate of corresponding molecule level in a uniform gas by a uniform electron beam, when secondary electrons energy distribution function is obtained from Eq.(4) and equals to $\rho_d(e)$ (Fig.2). All Φ_j^x are normalized by total primary electrons ionization rate. Parameter $\sigma_d(X,j)$ depends slightly on the kind of energy level j.

Excitation rate in the whole jet can be obtained from Eq.(9) and expressed in the form:

$$\Phi_j^x \approx \frac{J_b}{e}\ \sigma_i(E)\alpha\ c_x\Phi_j^x\ \Psi(\alpha, z_b/d_*, r_b/z_b), \quad \alpha = \frac{n_0 d_*^2 \sigma_0}{4z_b} \qquad (10)$$

In Eq.(10), the function $\Psi(\alpha, z_b/d_*, r_b/z_b)$ takes into account the overlapping of the gas jet profile, $n_g(r)$, and the electron beam profile, $\chi_b(r)$. It describes the escape of secondary electrons from the gas jet for relatively small gas densities at the point where an electron beam crosses the axis of the gas jet, and the contraction of the EEDF to the electron beam profile for high gas

densities.

For $\alpha \gg 1$ ($p_0 > 10$Torr, $d_* \sim z_b \sim 1$cm) $\Psi \to 1$ but the gas will be excited only in the "volume" of electron beam and some part of gas in the jet will not be activated. For $\alpha \ll 1$ the majority of secondary electrons will be gone away from the jet. Thus, the optimal condition for gas jet activation is $\alpha \approx 1$.

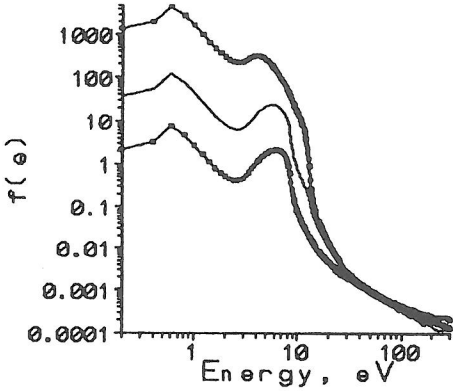

Fig. 2. EEDF of secondary electrons in Ar-SiH_4 mixture. Silane concentration:
1. - 0.1%,
2. - 5%,
3. - 99%.

The presented model provides the ground for estimating gas activation in supersonic jets by an electron beam and choosing the optimal conditions. Some examples of EB-activation of argon-silane jets are given in accompanying paper [3].

In conclusion, we point out that the efficiency of the gas jet activation can be considerably increased by reducing electron beam energy from E=10keV to the values of 1keV or lower, where primary electrons energy loss function is larger.

Another way to increase the efficiency is to combine the electron beam excitation with the use of electric fields witch shift a large low energy electrons group into an intermediate energy region. Under such an arrangement, the electron distribution function will be formed both by the energy loss of high energy secondary electrons produced by an electron beam and by energy gain from the electric field.

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2. R.G.Sharafutdinov et al. Zh.Techn.Fis., 1995, v.65, 181-185.
3. G.I.Sukhinin. Excitation of Ar-SiH_4 and Ar-CH_4 Mixtures by an Electron Beam". To be presented at ISPC-95.