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Modelling Mass and Heat Transport during Gas Phase Epitaxy in a Reactor with Rotating Substrate. On Possibility to Improve Of Properties of Films

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Abstract: In this paper we analysed convective diffusion processes in vertical and horizontal reactors for epitaxy from gas phase with disk substrate holder at atmosphere and low (~0.1 atmosphere) pressure. We formulate conditions to improve properties of grown structure. We introduce an analytical approach to solve equations of convective diffusion. We apply the obtained results for MOCVD of gallium arsenide. Epitaxy MOCVD could be used to growth of nanoscale semiconductor structures (quantum pits; arrays of quantum pits; structures with two-dimensional electronic gas). The same procedure could be also used to growth structures with higher dimensions.

Keywords: gas phase epitaxy; semiconductor layer; convection diffusion model.

1 Introduction

Epitaxial growth of monocrystalline layers from gas phase along with molecular beam epitaxy (MBE) is a main technological operation of modern micro- and nanoelectronics [1-11]. Methods of epitaxy are continuously improved to ensure the specified electrophysical parameters of semiconductor structures. A perspective method of manufacturing of semiconductor structures $A_{III}B_V$ and $A_{II}B_{VI}$ and a solid solutes based on them is epitaxy from gas phase with using appropriate organometallic compounds and hydrides as starting materials (MOCVD) [3,4,8-11].

Method MOCVD gives a possibility to manufacture semiconductor nanodimension structures, which includes into itself quantum pits; arrays of quantum dots; structures with two-dimensional electronic gas. One of aspects of the further development and improvement of methods of gas phase epitaxy is analytical modeling of series physical processes occurring in the gas phase and on the surface of the growing epitaxial layer. In particular, at the moment there is no enough full solution and fully complete analysis of problems convective diffusion, which determines the mass transfer at the gas-phase epitaxy in a flow system. In this situation estimations of correlations between technological parameters of kinetics and mechanism of the growth of epitaxial layers may have a significant error.

Our aim framework this paper is analytical modeling of processes of convective diffusion. The modeling gives a possibility to analyze flow of flux of mixture of gas in zone of growth of epitaxial layers. We consider the epitaxial

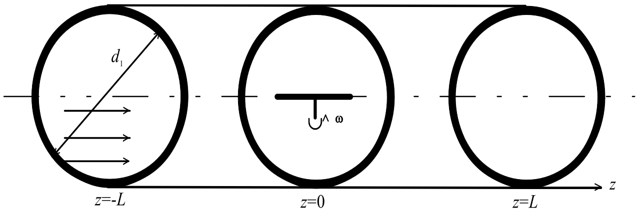

Fig. 1*a*. Structure of a horizontal reactor for epitaxy from gas phase

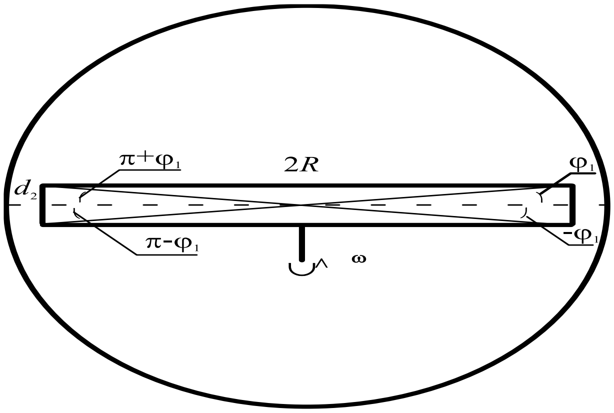

Fig. 1*b*. Structure of a disk substrate holder.

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growth at atmospheric pressure and at lower pressure (~ 0.1 atmosphere) in vertical or horizontal reaction chamber with high-frequency heating of rotating disk substrate holder.

The considered technological systems are main systems for epitaxial processes MOCVD. A flow of mixture of gases come in growth area of reaction chamber. The mixture includes into itself gaseous reagents for growth and doping epitaxial layers and hydrogen, which has been used as a gas-carrier.

2 Method of analysis

To model processes of convective diffusion in a vertical reaction chamber (see Fig. 1*a*) we assume, that vector of linear speed of flow in inlet part of area of growth is perpendicular to disk substrate holder. The substrate holder is rotating with angular velocity ω. We also assume, that distribution of concentration of growth component has been calculated near surface of disk with radius R. The distance from the disk is approximately equal to thickness of diffusion layer [12]. To consider our aims it is necessary to solve jointly the equation of Navier-Stokes and the second Fick's law. The Navier-Stokes equation for our case could be written as

$$\frac{\partial C(r,\varphi,z,t)}{\partial t}=div\left\{D\cdot grad\left[C(r,\varphi,z,t)\right]-\right.$$
$$\left.-\vec{\upsilon}(r,\varphi,z,t)\cdot C(r,\varphi,z,t)\right\}, \quad (1)$$

where r, φ, z and t are the cylindrical coordinates, which correspond to geometry of reaction chamber, and time; $C(r,\varphi,z,t)$ is the spatio-temporal distribution of concentration of growth component; D is the diffusion coefficient of growth component; $\vec{\upsilon}$ is the speed of flow of gases. The speed of flow of gases could be determine by solving the Navier-Stokes equation

$$\frac{\partial\vec{\upsilon}}{\partial t}+\left(\vec{\upsilon}\cdot\nabla\right)\vec{\upsilon}=-\nabla\left(\frac{P}{\rho}\right)+\nu\,\Delta\,\vec{\upsilon}, \quad (2)$$

where P is the pressure of gases in the reaction chamber; ν is the kinematical viscosity.

We consider regime of limiting diffusion of growth component. In this situation all coming near to the considered disk particles of growth components precipitating and regularizing on the substrate. The model corresponds to known experimental data for MOCVD [3,11]. With account laminarity of flow of gases in growth zone of vertical chamber boundary and initial conditions for differential conditions (1) and (2) could be written as

$$C|_{z=-L}=C_0,\ C|_{z=0}=0,\ C|_{\varphi=0}=C|_{\varphi=2\pi},\ C|_{r=0}\neq\infty,\ \upsilon_r|_{z=0}=0,$$
$$\left.\frac{\partial C}{\partial\varphi}\right|_{\varphi=0}=\left.\frac{\partial C}{\partial\varphi}\right|_{\varphi=2\pi},\ \left.\frac{\partial C}{\partial r}\right|_{r=R}=0,\ \left.\frac{\partial\upsilon_r}{\partial r}\right|_{r=0}=0,$$
$$\left.\frac{\partial\upsilon_\varphi}{\partial\varphi}\right|_{\varphi=0}=\left.\frac{\partial\upsilon_\varphi}{\partial\varphi}\right|_{\varphi=2\pi},\ \left.\frac{\partial\upsilon_\varphi}{\partial\varphi}\right|_{\varphi=0}=\left.\frac{\partial\upsilon_\varphi}{\partial\varphi}\right|_{\varphi=2\pi},\ \left.\frac{\partial\upsilon_r}{\partial r}\right|_{r=R}=0,$$
$$\upsilon_r|_{z=-L}=0,\ \upsilon_r|_{z=0}=0,\ C|_{t=0}=C_0\delta(z+L),\ \upsilon_r|_{z=L}=0,\ \upsilon_r|_{\varphi=0}=\upsilon_r|_{\varphi=2\pi},$$
$$\upsilon_r|_{r=0}\neq\infty,\ \upsilon_\varphi|_{z=0}=\omega r,\ \upsilon_\varphi|_{z=-L}=0,\ \upsilon_\varphi|_{z=L}=0,\ \upsilon_\varphi|_{\varphi=0}=\upsilon_\varphi|_{\varphi=2\pi}, \quad (3)$$
$$\upsilon_\varphi|_{r=0}\neq\infty,\ \upsilon_z|_{z=-L}=\upsilon_0,\ \upsilon_z|_{z=0}=\upsilon_0,\ \upsilon_z|_{z=L}=0,\ \upsilon_z|_{\varphi=0}=\upsilon_z|_{\varphi=2\pi},$$
$$\upsilon_z|_{r=0}\neq\infty,\ \upsilon_r|_{t=0}=0,\ \upsilon_\varphi|_{t=0}=0,\ \upsilon_z|_{t=0,\,z=-L}=\upsilon_0,$$

where υ_0 is the linear velocity of gas flow in inlet part of area of growth; ω is the frequency of rotation of disk substrate holder. Boundary condition with δ-function means, that concentration of growth component is equal to C_0 in inlet part of the growth area.

Further we consider convective diffusion to the rotating disk substrate holder for horizontal reaction chamber. To save generalized (cylindrical) system of coordinate we consider native hindrance for gas flow (disk substrate holder) by two sloping lines. These sloping lines passing through the center of the substrate holder in its diametric cross-section (see Fig. 1*b*). In this case due to usual relation between dimensions of the substrate holder $d_2<<2R$ angle of the sloping between these lines φ_1 is small ($\varphi_1<<1$ rad). In this situation one can assume, that vector of linear velocity in inlet part into growth zone is parallel to the surface of the disk (see Fig. 1*b*), boundary and initial conditions could be written as

$$C|_{z=-L}=C_0,\ C|_{t=0}=C_0\delta(z+L),\ C|_{r=0}\neq\infty,$$
$$C|_{\varphi=-\varphi_1}=C|_{\varphi=\varphi_1}=C|_{\varphi=\pi-\varphi_1}=C|_{\varphi=\pi+\varphi_1},\ \left.\frac{\partial C}{\partial r}\right|_{r=R}=0,$$
$$\left.\frac{\partial\upsilon_r}{\partial r}\right|_{r=0}=0,\ \upsilon_r|_{\varphi=-\varphi_1}=\upsilon_r|_{\varphi=\varphi_1}=\upsilon_r|_{\varphi=\pi-\varphi_1}=\upsilon_r|_{\varphi=\pi+\varphi_1},$$
$$\upsilon_r|_{z=-L}=0,\upsilon_r|_{z=L}=0,\upsilon_r|_{t=0}=0,\ \upsilon_r|_{r=0}\neq\infty,\ \upsilon_z|_{z=0}=0,$$
$$\upsilon_\varphi|_{\varphi=-\varphi_1}=\upsilon_\varphi|_{\varphi=\varphi_1}=\upsilon_\varphi|_{\varphi=\pi-\varphi_1}=\upsilon_\varphi|_{\varphi=\pi+\varphi_1},\ \left.\frac{\partial\upsilon_r}{\partial r}\right|_{r=R}=0,$$
$$\upsilon_\varphi|_{\varphi=-\varphi_1}=\upsilon_\varphi|_{\varphi=\varphi_1}=\upsilon_\varphi|_{\varphi=\pi-\varphi_1}=\upsilon_\varphi|_{\varphi=\pi+\varphi_1},\ \upsilon_z|_{r=0}\neq\infty,$$
$$v_z|_{r\pm R,t=0}tg(\varphi)=\omega z,\ \upsilon_z|_{r\pm R,t=0}tg(\varphi)=\omega z, \quad (4)$$
$$\upsilon_z|_{\varphi=-\varphi_1}=\upsilon_z|_{\varphi=\varphi_1}=\upsilon_z|_{\varphi=\pi-\varphi_1}=\upsilon_z|_{\varphi=\pi+\varphi_1},\ \upsilon_z|_{z=-L}=\upsilon_0,$$

$\upsilon_z|_{z=L}=\upsilon_0$, $\upsilon_\varphi|_{z=-L}=0$, $\upsilon_\varphi|_{z=L}=0$, $\upsilon_z|_{r=0}\neq\infty$, $\upsilon_\varphi|_{r=0}\neq\infty$,

$\upsilon_r|_{r=0}\neq\infty$, $\upsilon_z|_{z=0}=0$, $\upsilon_\varphi|_{t=0}=0$.

In this situation equations for components of velocity with account cylindrical system of coordinate could be written as

$$\frac{\partial\,\upsilon_r}{\partial\,t}=\nu\left\{\frac{1}{r}\frac{\partial}{\partial\,r}\left[r\frac{\partial\,\upsilon_r(r,\varphi,z,t)}{\partial\,r}\right]+\frac{1}{r^2}\frac{\partial^2\upsilon_r(r,\varphi,z,t)}{\partial\varphi^2}+\right.$$

$$\left.+\frac{\partial^2\upsilon_r(r,\varphi,z,t)}{\partial\,z^2}\right\}-\upsilon_r\frac{\partial\,\upsilon_r}{\partial\,r}-\frac{\upsilon_\varphi}{r}\frac{\partial\,\upsilon_\varphi}{\partial\,\varphi}-\upsilon_z\frac{\partial\,\upsilon_z}{\partial\,z}-$$

$$-\frac{\partial}{\partial\,r}\left(\frac{P}{\rho}\right)\tag{5a}$$

$$\frac{\partial\,\upsilon_\varphi}{\partial\,t}=\nu\left\{\frac{1}{r}\frac{\partial}{\partial\,r}\left[r\frac{\partial\,\upsilon_\varphi(r,\varphi,z,t)}{\partial\,r}\right]+\frac{1}{r^2}\frac{\partial^2\upsilon_\varphi(r,\varphi,z,t)}{\partial\varphi^2}+\right.$$

$$\left.+\frac{\partial^2\upsilon_\varphi(r,\varphi,z,t)}{\partial\,z^2}\right\}-\upsilon_r\frac{\partial\,\upsilon_r}{\partial\,r}-\frac{\upsilon_\varphi}{r}\frac{\partial\,\upsilon_\varphi}{\partial\,\varphi}-$$

$$-\upsilon_z\frac{\partial\,\upsilon_z}{\partial\,z}-\frac{1}{r}\frac{\partial}{\partial\,\varphi}\left(\frac{P}{\rho}\right)\tag{5b}$$

$$\frac{\partial\,\upsilon_z}{\partial\,t}=\nu\left\{\frac{1}{r}\frac{\partial}{\partial\,r}\left[r\frac{\partial\,\upsilon_z(r,\varphi,z,t)}{\partial\,r}\right]+\frac{1}{r^2}\frac{\partial^2\upsilon_z(r,\varphi,z,t)}{\partial\varphi^2}+\right.$$

$$+\frac{\partial^2\upsilon_z(r,\varphi,z,t)}{\partial\,z^2}-\upsilon_r\frac{\partial\,\upsilon_r}{\partial\,r}-\frac{\upsilon_\varphi}{r}\frac{\partial\,\upsilon_\varphi}{\partial\,\varphi}-$$

$$-\upsilon_z\frac{\partial\,\upsilon_z}{\partial\,z}-\frac{\partial}{\partial\,z}\left(\frac{P}{\rho}\right).\tag{5c}$$

Now we determine solution of the above system of equations by using method of averaging of function correction [13-15]. Framework the approach to determine the first-order approximation of components of flow of mixture of gases we replace the components on their not yet known average values $\upsilon_r\to\alpha_{1r}$, $\upsilon_\varphi\to\alpha_{1\varphi}$, $\upsilon_z\to\alpha_{1z}$ in right sides of Eqs.(5). The considered substitution gives us possibility to obtain equations for the first-order approximation of the considered components in the following form

$$\frac{\partial\,\upsilon_{1r}}{\partial\,t}=-\frac{\partial}{\partial\,r}\left(\frac{P}{\rho}\right),\ \frac{\partial\,\upsilon_{1\varphi}}{\partial\,t}=-\frac{1}{r}\frac{\partial}{\partial\,\varphi}\left(\frac{P}{\rho}\right),$$

$$\frac{\partial\,\upsilon_{1z}}{\partial\,t}=-\frac{\partial}{\partial\,z}\left(\frac{P}{\rho}\right).\tag{6}$$

Solutions of the above equations could be written as

$$\upsilon_{1r}=-\frac{\partial}{\partial\,r}\int\limits_0^t\frac{P}{\rho}d\,\tau\,,\ \upsilon_{1\varphi}=-\frac{1}{r}\frac{\partial}{\partial\,\varphi}\int\limits_0^t\frac{P}{\rho}d\,\tau\,,$$

$$\upsilon_{1z}=-\frac{\partial}{\partial\,z}\int\limits_0^t\frac{P}{\rho}d\,\tau\,.\tag{7}$$

The second-order approximations of components of flow of mixture of gases could be calculated by replacement of the required components in the right sides of the Eqs. (5) on the following sums: $\upsilon_r\to\alpha_{2r}+\upsilon_{1r}$, $\upsilon_\varphi\to\alpha_{2\varphi}+\upsilon_{1\varphi}$, $\upsilon_z\to\alpha_{2z}+\upsilon_{1z}$. Equations of these approximations could be written as

$$\frac{\partial\,\upsilon_{2r}}{\partial\,t}=\nu\left\{\frac{1}{r}\frac{\partial}{\partial\,r}\left[r\frac{\partial\,\upsilon_{1r}(r,\varphi,z,t)}{\partial\,r}\right]+\right.$$

$$\left.+\frac{1}{r^2}\frac{\partial^2\upsilon_{1r}(r,\varphi,z,t)}{\partial\varphi^2}+\frac{\partial^2\upsilon_{1r}(r,\varphi,z,t)}{\partial\,z^2}\right\}-$$

$$-(\alpha_{2r}+\upsilon_{1r})\frac{\partial\,\upsilon_{1r}}{\partial\,r}-\frac{\alpha_{2\varphi}+\upsilon_{1\varphi}}{r}\frac{\partial\,\upsilon_{1\varphi}}{\partial\,\varphi}-$$

$$-(\alpha_{2z}+\upsilon_{1z})\frac{\partial\,\upsilon_{1z}}{\partial\,z}-\frac{\partial}{\partial\,r}\left(\frac{P}{\rho}\right)\tag{8a}$$

$$\frac{\partial\,\upsilon_{2\varphi}}{\partial\,t}=\nu\left(\frac{1}{r}\frac{\partial^2\upsilon_{1r}}{\partial\,r\,\partial\,\varphi}+\frac{2}{r^2}\frac{\partial^2\upsilon_{1\varphi}}{\partial\,\varphi^2}-\frac{1}{r^2}\frac{\partial^2\upsilon_{1r}}{\partial\,\varphi\,\partial\,z}+\right.$$

$$\left.+\frac{\partial^2\upsilon_{1\varphi}}{\partial\,z^2}\right)-\frac{1}{r}\frac{\partial}{\partial\,\varphi}\left(\frac{P}{\rho}\right)-(\alpha_{2r}+\upsilon_{1r})\frac{\partial\,\upsilon_{1\varphi}}{\partial\,r}-$$

$$-\frac{(\alpha_{2\varphi}+\upsilon_{1\varphi})}{r}\frac{\partial\,\upsilon_{1\varphi}}{\partial\,\varphi}-(\alpha_{2z}+\upsilon_{1z})\frac{\partial\,\upsilon_{1\varphi}}{\partial\,z},\tag{8b}$$

$$\frac{\partial\,\upsilon_{2z}}{\partial\,t}=\nu\left(\frac{\partial^2\upsilon_{1r}}{\partial\,z^2}+\frac{\partial^2\upsilon_{1z}}{\partial\,r^2}+\frac{1}{r^2}\frac{\partial^2\upsilon_{1z}}{\partial\,\varphi^2}\right)-$$

$$-\frac{\partial}{\partial\,z}\left(\frac{P}{\rho}\right)-(\alpha_{2r}+\upsilon_{1r})\frac{\partial\,\upsilon_{1z}}{\partial\,r}-\frac{(\alpha_{2\varphi}+\upsilon_{1\varphi})}{r}\frac{\partial\,\upsilon_{1z}}{\partial\,\varphi}-$$

$$-(\alpha_{2z}+\upsilon_{1z})\frac{\partial\,\upsilon_{1z}}{\partial\,z}.\tag{8c}$$

Integration of the above equations leads to the following results

$$\upsilon_{2r} = \nu \int_0^t \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \upsilon_{1r}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \upsilon_{1r}}{\partial \varphi^2} + \frac{\partial^2 \upsilon_{1r}}{\partial z^2} \right] d\tau -$$

$$- \frac{\partial}{\partial r} \left(\int_0^t \frac{P}{\rho} d\tau \right) - \int_0^t \left(\alpha_{2r} + \upsilon_{1r} \right) \frac{\partial \upsilon_{1r}}{\partial r} d\tau -$$

$$- \int_0^t \frac{\left(\alpha_{2\varphi} + \upsilon_{1\varphi} \right)}{r} \frac{\partial \upsilon_{1r}}{\partial \varphi} d\tau -$$

$$- \int_0^t \left(\alpha_{2z} + \upsilon_{1z} \right) \frac{\partial \upsilon_{1r}}{\partial z} d\tau \, , \quad (8г)$$

$$\upsilon_{2\varphi} = \nu \int_0^t \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \upsilon_{1\varphi}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \upsilon_{1\varphi}}{\partial \varphi^2} + \frac{\partial^2 \upsilon_{1\varphi}}{\partial z^2} \right] d\tau -$$

$$- \frac{1}{r} \frac{\partial}{\partial \varphi} \left(\int_0^t \frac{P}{\rho} d\tau \right) - \int_0^t \left(\alpha_{2r} + \upsilon_{1r} \right) \frac{\partial \upsilon_{1\varphi}}{\partial r} d\tau - \quad (8\partial)$$

$$- \int_0^t \frac{\left(\alpha_{2\varphi} + \upsilon_{1\varphi} \right)}{r} \frac{\partial \upsilon_{1\varphi}}{\partial \varphi} d\tau - \int_0^t \left(\alpha_{2z} + \upsilon_{1z} \right) \frac{\partial \upsilon_{1\varphi}}{\partial z} d\tau \, ,$$

$$\upsilon_{2z} = \upsilon_0 + \nu \int_0^t \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \upsilon_{1z}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \upsilon_{1z}}{\partial \varphi^2} + \frac{\partial^2 \upsilon_{1z}}{\partial z^2} \right] d\tau -$$

$$- \int_0^t \left(\alpha_{2r} + \upsilon_{1r} \right) \frac{\partial \upsilon_{1z}}{\partial r} d\tau - \int_0^t \frac{\left(\alpha_{2\varphi} + \upsilon_{1\varphi} \right)}{r} \frac{\partial \upsilon_{1z}}{\partial \varphi} d\tau -$$

$$- \frac{\partial}{\partial z} \left(\int_0^t \frac{P}{\rho} d\tau \right) - \int_0^t \left(\alpha_{2z} + \upsilon_{1z} \right) \frac{\partial \upsilon_{1z}}{\partial z} d\tau \, . \qquad (8e)$$

We determine average values α_{2r}, $\alpha_{2\varphi}$, α_{2z} by using the following standard relations

$$\alpha_{2r} = \frac{1}{\pi \Theta R^2 L} \int_0^\Theta \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \left(\upsilon_{2r} - \upsilon_{1r} \right) dz\, d\varphi\, dr\, dt \, ,$$

$$\alpha_{2\varphi} = \frac{1}{\pi \Theta R^2 L} \int_0^\Theta \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \left(\upsilon_{2\varphi} - \upsilon_{1\varphi} \right) dz\, d\varphi\, dr\, dt \, , \qquad (9)$$

$$\alpha_{2z} = \frac{1}{\pi \Theta R^2 L} \int_0^\Theta \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \left(\upsilon_{2z} - \upsilon_{1z} \right) dz\, d\varphi\, dr\, dt \, .$$

Here Θ is the growth time. Substitution of the first- and the second-order approximations of components of speed of growth into the relations Eqs (9) gives a possibility to obtain system of equations for the required averaged values

$$\begin{cases} A_1 \alpha_{2r} + B_1 \alpha_{2\varphi} + C_1 \alpha_{2z} = D_1 \\ A_2 \alpha_{2r} + B_2 \alpha_{2\varphi} + C_2 \alpha_{2z} = D_2 \\ A_3 \alpha_{2r} + B_3 \alpha_{2\varphi} + C_3 \alpha_{2z} = D_3 \end{cases} \qquad (10)$$

where $A_1 = 1 + \int_0^\Theta (\Theta - t) \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \frac{\partial v_{1r}}{\partial r} dz\, d\varphi\, dr\, dt$,

$$B_1 = \int_0^\Theta (\Theta - t) \int_0^R \int_0^{2\pi} \int_{-L}^{L} \frac{\partial v_{1r}}{\partial \varphi} dz\, d\varphi\, dr\, dt \, , \; C_1 =$$

$$= C_2 = \frac{\pi}{2} \Theta^2 R^2 V_0 \, , \; D_1 = \nu \int_0^\Theta (\Theta - t) \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \left[\frac{\partial^2 \upsilon_{1r}}{\partial z^2} + \right.$$

$$\left. + \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \upsilon_{1r}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \upsilon_{1r}}{\partial \varphi^2} \right] dz\, d\varphi\, dr\, dt - \Theta^2 \times$$

$$\times \frac{\pi}{8} R^2 \upsilon_0^2 - \int_0^\Theta (\Theta - t) \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \upsilon_{1r} \frac{\partial \upsilon_{1r}}{\partial r} dz\, d\varphi\, dr\, dt -$$

$$- \int_0^\Theta (\Theta - t) \int_0^R \int_0^{2\pi} \int_{-L}^{L} \upsilon_{1\varphi} \frac{\partial \upsilon_{1r}}{\partial \varphi} dz\, d\varphi\, dr\, dt \, , \; A_2 = \int_0^\Theta (\Theta - t) \times$$

$$\times \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \frac{\partial \upsilon_{1r}}{\partial r} dz\, d\varphi\, dr\, dt \, , \; B_2 = 1 + \int_0^\Theta (\Theta - t) \times$$

$$\times \int_0^R \int_0^{2\pi} \int_{-L}^{L} \frac{\partial v_{1r}}{\partial \varphi} dz\, d\varphi\, dr\, dt \, , \; D_2 = \nu \int_0^\Theta (\Theta - t) \int_0^R r \times$$

$$\times \int_0^{2\pi} \int_{-L}^{L} \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \upsilon_{1\varphi}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \upsilon_{1\varphi}}{\partial \varphi^2} + \frac{\partial^2 \upsilon_{1\varphi}}{\partial z^2} \right] dz\, d\varphi\, dr\, dt -$$

$$- \int_0^\Theta (\Theta - t) \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \upsilon_{1r} \frac{\partial \upsilon_{1r}}{\partial r} dz\, d\varphi\, dr\, dt - \int_0^\Theta (\Theta - t) \times$$

$$\times \int_0^R \int_0^{2\pi} \int_{-L}^{L} \upsilon_{1\varphi} \frac{\partial \upsilon_{1r}}{\partial \varphi} dz\, d\varphi\, dr\, dt - \frac{\pi}{8} \Theta^2 R^2 \upsilon_0^2 \, , \; A_3 =$$

$$= \int_0^\Theta (\Theta - t) \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \frac{\partial \upsilon_{1z}}{\partial r} dz\, d\varphi\, dr\, dt \, , \; B_3 = \int_0^\Theta (\Theta - t) \times$$

$$, \times \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \frac{\partial \upsilon_{1z}}{\partial \varphi} dz\, d\varphi\, dr\, dt` , \; C_3 = 1 + \frac{\pi}{2} \Theta^2 R^2 \upsilon_0 \, ,$$

$$D_3 = \nu \int_0^\Theta (\Theta - t) \int_0^R r \int_0^{2\pi} \int_{-L}^{L} \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \upsilon_{1z}}{\partial r} \right) + \frac{\partial^2 \upsilon_{1z}}{\partial z^2} + \right.$$

$$+\frac{1}{r^2}\frac{\partial^2 \upsilon_{1z}}{\partial \varphi^2}\Bigg]\,d\,z\,d\,\varphi\,d\,r\,d\,t-\int\limits_0^\Theta(\Theta-t)\int\limits_0^R r\int\limits_0^{2\pi}\int\limits_{-L}^{L}\frac{\partial\,\upsilon_{1z}}{\partial\,r}\times$$

$$\times\,\upsilon_{1r}d\,z\,d\,\varphi\,d\,r\,d\,t-\int\limits_0^\Theta\int\limits_0^R r\int\limits_0^{2\pi}\int\limits_{-L}^{L}\frac{\partial\,\upsilon_{1z}}{\partial\,\varphi}\upsilon_{1\varphi}d\,z\,d\,\varphi\,d\,r\times$$

$$\times(\Theta-t)\,d\,t-\frac{\pi}{8}\Theta^2R^2\upsilon_0^2\cdot$$

The above system could be solved by standard approaches [16] and could be written as:

$$\alpha_{2r}=\Delta_r/\Delta,\ \alpha_{2\varphi}=\Delta_\varphi/\Delta,\ \alpha_{2z}=\Delta_z/\Delta, \quad (11)$$

where $\Delta=A_1(B_2C_3-B_3C_2)-B_1(A_2C_3-A_3C_2)+(A_2B_3-A_3B_2)\times$ $\times\,C_1,\ \Delta_r=D_1(B_2C_3-B_3C_2)-B_1(D_2C_3-D_3C_2)+$ $+C_1(D_2B_3-D_3B_2),\ \Delta_\varphi=D_1(B_2C_3-B_3C_2)-B_1\times$ $\times\,(D_2C_3-D_3C_2)+C_1(D_2B_3-D_3B_2),\ \Delta_z=A_1\times$ $\times\,(B_2D_3-B_3D_2)-B_1(A_2D_3-A_3D_2)+D_1(A_2B_3-A_3B_2)$.

In this section we calculate components of speed of flow of gas mixture framework the second-order approximation by using method of averaging of function correction. The second-order approximation is usially enough good approximation for qualitative analysis of the considered processes and to make several quantitative estimations.

Let us consider the Eq.(1) in the cylindrical system of coordinate

$$\frac{\partial C(r,\varphi,z,t)}{\partial t}=\frac{1}{r}\frac{\partial}{\partial r}\left[r\,D\frac{\partial C(r,\varphi,z,t)}{\partial r}\right]+\frac{1}{r^2}\frac{\partial}{\partial\varphi}\left[D\frac{\partial C(r,\varphi,z,t)}{\partial\varphi}\right]+$$

$$+\frac{\partial}{\partial z}\left[D\frac{\partial C(r,\varphi,z,t)}{\partial z}\right]-\frac{1}{r}\frac{\partial}{\partial r}\left[r\,C(r,\varphi,z,t)\,\upsilon_r(r,\varphi,z,t)\right]-$$

$$-\frac{1}{r}\frac{\partial}{\partial\varphi}\left[C(r,\varphi,z,t)\,\upsilon_\varphi(r,\varphi,z,t)\right]-$$

$$-\frac{\partial}{\partial z}\left[C(r,\varphi,z,t)\,\upsilon_z(r,\varphi,z,t)\right]. \quad (12)$$

Let us consider the method of averaging of function correction to determine spatio- temporal distribution of concentration of growth component in flow of gas mixture. To determine the first-order approximation of the required concentration we replace the considered function on not yet known average value α_{1C} in the right side of the considered equation. Using the recently considered algorithm gives a possibility to obtain the first-order approximation of concentration of growth component in the following form

$$C_1(r,\varphi,z,t)=-\frac{\alpha_{1C}}{r}\int\limits_0^t\frac{\partial\left[r\,\upsilon_r(r,\varphi,z,\tau)\right]}{\partial\,r}d\,\tau-$$

$$-\frac{\alpha_{1C}}{r}\int\limits_0^t\frac{\partial\,\upsilon_\varphi(r,\varphi,z,\tau)}{\partial\,\varphi}d\,\tau-$$

$$-\,\alpha_{1C}\int\limits_0^t\frac{\partial\,\upsilon_z(r,\varphi,z,\tau)}{\partial\,z}d\,\tau+C_0\,. \quad (13)$$

We determine not yet known average value α_{1C} of the first-order approximation of the considered concentration by using the following relation

$$\alpha_{1C}=\frac{1}{\pi\Theta R^2L}\int\limits_0^\Theta\int\limits_0^R r\int\limits_0^{2\pi}\int\limits_{-L}^{L}C_1(r,\varphi,z,\tau)\,d\,z\,d\,\varphi\,d\,r\,d\,t\,. \quad (14)$$

Substitution of the first-order approximation of the considered concentration into the relation (14) leads to the following result [17]

$$\alpha_{1C}=C_0\Big/L\cdot\left[1+\frac{1}{\pi\Theta RL}\int\limits_0^\Theta(\Theta-t)\int\limits_0^{2\pi}\int\limits_{-L}^{L}\upsilon_r(R,\varphi,z,t)\,d\,z\,d\,\varphi\,d\,t+\frac{\Theta\,\upsilon_0}{RL}\right]\cdot \quad (15)$$

The second-order approximation of concentration of mixture of gases has been calculated framework standard procedure of method of averaged of function correction [11-16]. Framework the approach we replace the considered concentration in the Eq. (1) on the following sum: $C\to\alpha_{2C}+C_1$. In this situation the second-order approximation of the concentration in the following form

$$C_2(r,\varphi,z,t)=\frac{1}{r}\frac{\partial}{\partial\,r}\int\limits_0^t r\,D\frac{\partial\,C_1(r,\varphi,z,\tau)}{\partial\,r}d\,\tau+$$

$$+\frac{1}{r^2}\frac{\partial}{\partial\,\varphi}\int\limits_0^t D\frac{\partial\,C_1(r,\varphi,z,\tau)}{\partial\,\varphi}d\,\tau+C_0\delta(z+L)+$$

$$+\frac{\partial}{\partial\,z}\int\limits_0^t D\frac{\partial\,C_1(r,\varphi,z,\tau)}{\partial\,z}d\,\tau-$$

$$-\frac{1}{r}\frac{\partial}{\partial\,r}\left\{r\int\limits_0^t\left[\alpha_{2C}+C_1(r,\varphi,z,\tau)\right]\cdot\upsilon_r(r,\varphi,z,\tau)\,d\,\tau\right\}-$$

$$-\frac{1}{r}\frac{\partial}{\partial\,\varphi}\int\limits_0^t\left[\alpha_{2C}+C_1(r,\varphi,z,\tau)\right]\cdot\upsilon_\varphi(r,\varphi,z,\tau)\,d\,\tau-$$

$$-\frac{\partial}{\partial\,z}\int\limits_0^t\left[\alpha_{2C}+C_1(r,\varphi,z,\tau)\right]\cdot\upsilon_z(r,\varphi,z,\tau)\,d\,\tau\,. \quad (16)$$

Average value of the second-order approximation of concentration of mixture of gases α_{2C} could be determine by using the standard relation

$$\alpha_{2C} = \frac{1}{\pi \Theta R^2 L} \int_0^{\Theta} \int_0^{R} r \int_0^{2\pi} \int_{-L}^{L} \left(C_2 - C_1 \right) d\,z\,d\,\varphi\,d\,r\,d\,t\,. \quad (17)$$

Substitution of the first-order and the second-order approximations into the relation (17) gives a possibility to obtain relation for the average value α_{2C} in the following form

$$\alpha_{2C} = \frac{1}{\pi \Theta R^2 L} \int_0^{\Theta} \left(\Theta - t \right) \int_0^{R} r \int_0^{2\pi} D \left[\left. \frac{\partial C_1 \left(r, \varphi, z, \tau \right)}{\partial z} \right|_{z=L} - \right.$$

$$\left. - \left. \frac{\partial C_1 \left(r, \varphi, z, \tau \right)}{\partial z} \right|_{z=-L} \right] d\,\varphi\,d\,r\,d\,t - \frac{1}{\pi \Theta R^2 L} \int_0^{\Theta} \left(\Theta - t \right) \times$$

$$\times \int_0^{2\pi} \int_{-L}^{L} \left\{ \upsilon_r \left(R, \varphi, z, \tau \right) \cdot \left[\alpha_{2C} - \alpha_{1C} + C_1 \left(R, \varphi, z, \tau \right) \right] \right\} \times$$

$$\times r\,d\,z\,d\,\varphi\,d\,t - \frac{\upsilon_0}{\pi \Theta R^2 L} \int_0^{\Theta} \left(\Theta - t \right) \int_0^{R} r \int_0^{2\pi} \left(\alpha_{2C} - \alpha_{1C} + \right.$$

$$\left. + C_0 \right) d\,z\,d\,\varphi\,d\,r\,d\,t\,. \quad (18)$$

In the present paper we used the same approach to solve the Navier-Stokes equation (1) and the second Fick's law for both types reaction chambers. Necessary partial iterative solutions of the considered equations and formulas for calculations have been obtain for appropriate boundary and initial conditions.

3 Discussion

In this section we analyzed dependences of growth components framework flow of mixture of gases on frequency of rotation of disk substrate holder ω, diffusion coefficient D and linear speed of mixture of gases υ_0 (see Figs. 2, 3, 4) by calculated in previous section relations 8-11, 15,16,18. The obtained results have been obtained for epitaxial process of manufacturing of mono-crystalline layers of gallium arsenide by MOCVD method with using trimethylgallium and arsine as initial materials. We consider hydrogen as gas-carrier, volumetric speed of flow is 10 Lpm, specific length of reaction chamber is L =250 mm.

The Fig. 2 shows monotonous increasing of the considered concentration C at fixed value of concentration C_0 in the inlet part into the growth area. In this case one can find increasing of speed of epitaxial growth at increasing ω.

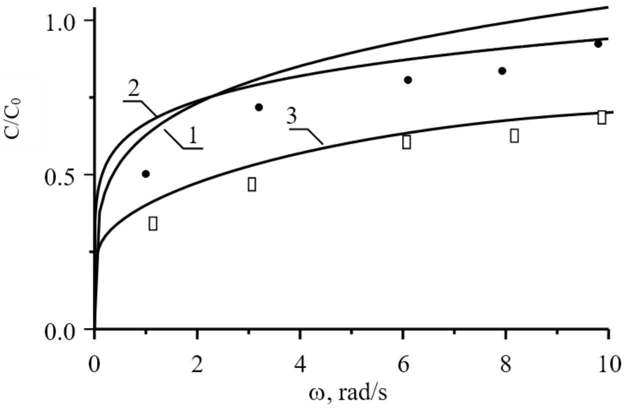

Fig. 2. Dependences of concentration of growth components on frequency of rotation ω of disk substrate holder. Curves 1 and 2 correspond to vertical reaction chamber for atmospheric pressure and low pressure (~10^4 Pa), respectively, (for d = 200 *mm*, R = 80 *mm*). Curve 3 corresponds to atmospheric pressure in horizontal reaction chamber (d = 150 *mm*, R = 50 *mm*). Experimental data correspond to vertical (●) and horizontal (□) reaction chambers (atmospheric pressure).

Fig. 3 shows dependence of concentration of growth component on value of diffusion coefficient D.

In this case increasing of value of diffusion coefficient leads to increasing of speed of particles of growth components. The increasing of speed leads to decreasing of concentration of these particles in growth zone. Curves 2 and 3 on Fig. 3 correspond to atmospheric pressure in reaction chamber. Curve 3 on Fig. 3 corresponds to decreased in 10 times pressure. The considered dependences (Figs. 2 and 3) show faster changing of concentration with faster achievement of saturation at low pressure in comparison with growth of epitaxial layers at atmospheric pressure. These figures also show comparison of theoretical results (solid lines) with experimental one (points) for MOCVD epitaxy of gallium arsenide. By using experimentally measured values (speed of growth of epitaxial layer and frequency of rotation of substrate holder) and taking into account linear dependence of speed of growth on concentration of main growth component C (trimethylgallium) [10,11] we obtain experimental dependences C/C_0 on ω (see Fig. 2). Reason of difference between experimental and theoretical data is probably existing adventitious reactions in gas phase [11]. In this situation only a part of gallium arsenide transforms to the solid phase in growth area. Similar experimental dependence has been also obtained in [8].

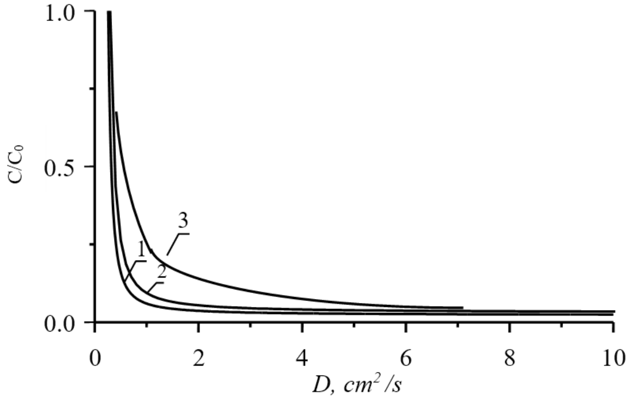

Fig. 3. Dependences of concentration of growth component on diffusion coefficient D (ω = 6 rad/c). Curves 1 and 2 correspond to vertical reaction chamber for low pressure (~10^4 Pa) and atmospheric pressure, respectively. Curve 3 corresponds to atmospheric pressure in horizontal reaction chamber.

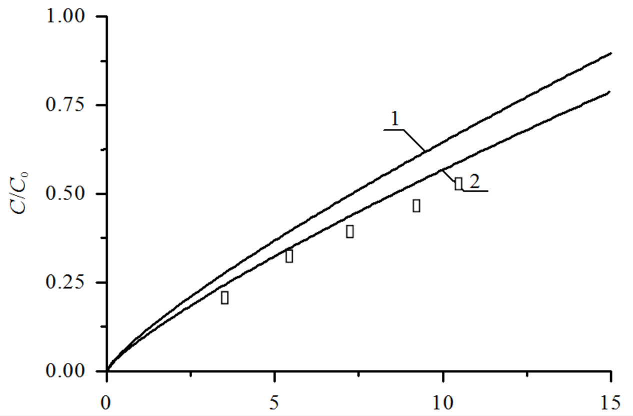

Fig. 4. Dependences of concentration of growth component on linear speed of gas flow υ_0, cm/s (ω = 6 rad/c, atmospheric pressure). Curve 1 corresponds to vertical reaction chamber. Curve 2 corresponds to horizontal reaction chamber. Symbols □ correspond to experimental data.

Analysis of calculated dependences and experimental data for horizontal reaction chamber (curve 2 of Fig. 3) shows more weak dependence of concentration C in horizontal reaction chamber and consequently speed of epitaxial growth of gallium arsenide on ω in comparison with vertical reaction chamber. Reason of this difference is difference in type of wrapping of flow of mixture of gases of rotating of disk substrate holder. Dependences of concentration of growth component in reaction zone on linear speed of gas flow are presented on Fig. 4. The dependences could be explained by increasing of induced convection during diffusion mass transport.

4 Conclusion

In this paper we introduce an analytical approach to model convective diffusion process, which take a place during epitaxy from gas phase with account rotating disk substrate holder. We estimate spatio-temporal distributions of concentration of growth component in flow of mixture of gases. We analyzed influence of main technological parameters on the concentration. The presented theoretical results agree well with the experimental data for MOCVD epitaxial gallium arsenide processes.

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