HEAT AND MASS TRANSFER IN AN EPITAXIAL REACTOR OF THE VERTICAL TYPE.

1. SYMMETRICAL HEATING OF THE REACTOR

V. P. Popov

UDC 546.281:536.244

Results are presented from a numerical study of the distribution of the dimensionless rate of deposition of an epitaxial layer along a reactor made in the form of a planar vertical convergent channel which is heated symmetrically.

This article examines heat and mass transfer in an epitaxial reactor in the form of a vertical quartz tube of rectangular cross section. Two graphite plate-type substrate holders with substrates are located on opposing walls of the reactor [1].

If the gap between the plates h = 2r (see Fig. 1) is much less than their width, the reactor can be represented as a plane channel formed by these plates. Heat and mass transfer in the reactor is examined using the example of the deposition of a layer of silicon from a mixture of silane and hydrogen. The gaseous mixture is introduced at the top of the reactor, as shown in Fig. 1. Silicon is deposited on the surface of thin silicon plates – substrates – and on the free surface of the substrate holders as a result of the rection

$$\mathrm{SiH_4} \rightleftarrows \mathrm{Si_{so}} + 2\mathrm{H_2}, \tag{1}$$

activated by heating of the gas by the holders and substrates. The temperature of the deposition surface (holders and substrates) is kept constant within the range T_s = 1323-1423 K.

In the case of laminar flow of the gas mixture, heat and mass transfer in the channel in the boundary-layer approximation is described by the system of conservation equations:

$$\frac{\partial}{\partial \tilde{x}'}(\tilde{\rho}\tilde{u}) + \frac{\partial}{\partial \tilde{y}}(\tilde{\rho}\tilde{v}) = 0, \tag{2}$$

$$\tilde{\rho}\tilde{u}\frac{\partial \tilde{u}}{\partial \tilde{x}'} + \tilde{\rho}\tilde{v}\frac{\partial \tilde{u}}{\partial \tilde{y}} = -\frac{\partial \tilde{P}}{\partial \tilde{x}'} \pm \frac{\mathrm{Ga}'}{\mathrm{Re}'}(\tilde{\rho} - 1) + \frac{\partial}{\partial \tilde{y}}\left(\tilde{\mu}\frac{\partial \tilde{u}}{\partial \tilde{y}}\right), \tag{3}$$

$$\tilde{\rho}\tilde{u}\frac{\partial \tilde{H}}{\partial \tilde{x}'} + \tilde{\rho}\tilde{v}\frac{\partial \tilde{H}}{\partial \tilde{y}} = \frac{1}{\mathrm{Pr}}\frac{\partial}{\partial \tilde{y}}\left(\frac{\tilde{\lambda}}{\tilde{c}_p}\frac{\partial \tilde{H}}{\partial \tilde{y}}\right), \tag{4}$$

$$\tilde{\rho}\tilde{u}\frac{\partial c_1}{\partial \tilde{x}'} + \tilde{\rho}\tilde{v}\frac{\partial c_1}{\partial \tilde{y}} = \frac{1}{\mathrm{Pr}_D}\frac{\partial}{\partial \tilde{y}}\left[\tilde{\rho}\tilde{D}\left(\frac{\partial c_1}{\partial \tilde{y}} + \frac{\alpha_T c_1(1-c_1)}{\tilde{T}}\frac{\partial \tilde{T}}{\partial \tilde{y}}\right)\right] \tag{5}$$

with the boundary conditions

$$\tilde{x}' = 0,\quad 0 < \tilde{y} < 1,\quad \tilde{u} = 1,\quad \tilde{T} = 1,\ c_1 = c_{01},$$

$$\begin{gathered} \tilde{x}' \geqslant 0,\quad \tilde{y} = \tilde{y}_{\mathbf{s}},\ \tilde{u} = 0,\\ \tilde{v} = \tilde{v}_{\mathbf{s}},\ \tilde{T} = \tilde{T}_{\mathbf{s}},\ c_1 = 0,\\ \frac{\partial c_1}{\partial \tilde{y}} + \frac{\alpha_T c_1(1-c_1)}{\tilde{T}}\frac{\partial \tilde{T}}{\partial \tilde{y}} = 0, \end{gathered} \tag{6}$$

A. V. Lykov Institute of Heat and Mass Transfer, Academy of Sciences of the Belorussian SSR, Minsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 56, No. 6, pp. 942-948, June, 1989. Original article submitted December 21, 1987.

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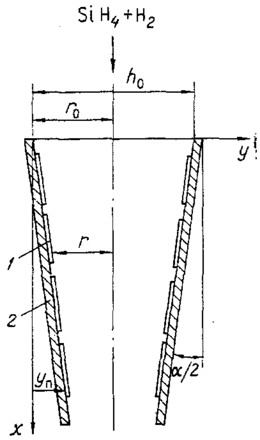

Fig. 1. Sketch of reactor: 1) substrate; 2) substrate holder.

$$\tilde{x}' \geqslant 0,\quad \tilde{y} = 1,\quad \frac{\partial \tilde{u}}{\partial \tilde{y}} = \frac{\partial \tilde{v}}{\partial \tilde{y}} = \frac{\partial \tilde{T}}{\partial \tilde{y}}\ \frac{\partial c_1}{\partial \tilde{y}} = 0. \qquad (6)$$

The physical properties of the gas mixture were assumed to be dependent on its temperature, pressure, and local composition. They were calculated as they were in [2].

It is known that Eq. (1) is the stoichiometric equation of the complete rection. The actual mechanism of the thermal decomposition of silane includes a large number of intermediate reactions [3, 4]. At a temperature $T_s \geq 1123$ K, the greatest contribution to the deposition of silicon is made by silicane [4] formed as a result of the homogeneous reaction

$$SiH_4 \rightleftarrows SiH_2 + H_2. \qquad (7)$$

Diffusing to the deposition surface, silicane is adsorbed on it. Silicon is formed on this surface as a result of several reactions that can be represented by the overall equation

$$SiH_2 \rightleftarrows Si_{(so)} + H_2. \qquad (8)$$

Reaction (8) takes place without a volume change. Thus, the velocity of the Stefan flow $\tilde{v}_s$ in boundary conditions (6) can be taken equal to 0. At a temperature $T_s \geq 1323$ K, almost no silane molecules reach the substrate, undergoing conversion to silicane in the interior of the reactor.

Since the main product of the homogeneous reactions is silicane and since there is little difference between silicane- and silicane-hydrogen mixtures from the viewpoint of heat and mass transfer, the results of numerical modeling with allowance for twenty homogeneous reactions at $T_s = 1323$ K and atmospheric pressure coincide with the results of the use of a simplified model considering only reaction (1) (which is assumed to be heterogeneous, occurring in the diffusion regime) and the results of experiments conducted under these conditions [4]. For atmospheric pressure and $T_s \geq 1323$ K, this allows us to adjudge the simplified model adequate, i.e., to ignore the homogeneous reactions and assume that the silicon deposition rate is limited by the rate of transport of silane molecules to the deposition surface, i.e., $c_{s1} = 0$.

In connection with the fact that hydrogen constitutes 99% (by vol.) of the silane-hydrogen mixtures used in processes involving deposition from the gas phase, the mixture can be considered diathermal. As a result, in energy equation (4) we omit terms accounting for heat transfer by diffusion and diffusional heat conduction. Estimates show that the heat fluxes caused by these effects are 3-4 orders of magnitude lower than the heat flux due to normal conduction.

Results of the solution of Eqs. (2-6) are shown in Figs. 2-4. The deposition rate is represented as being proportional to its dimensionless magnitude Nu_{0D}, which is determined from the concentration of silane at the inlet of the reactor.

If we ignore free convection and thermodiffusion, then Nu_{0D} decreases monotonically with an increase in the dimensionless distance $\tilde{x}_D$ from the front edge of the channel due to

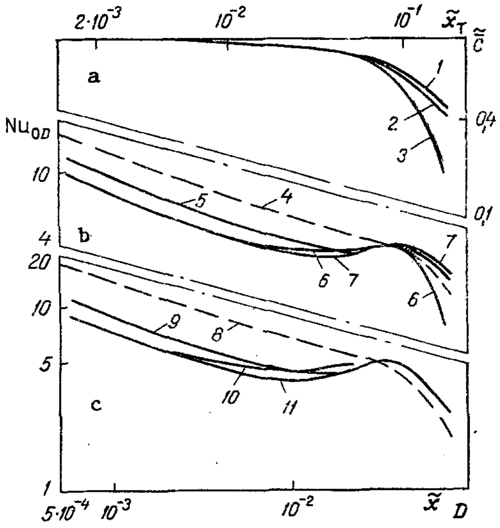

Fig. 2. Dependence of $\tilde{\tilde{c}}$ and Nu_{0D} on $\tilde{x}_{D,T}$ with allowance for the effect of free convection (P = 101 kPa, X_{01} = 0.0002, T_s = 1423 K, h_0 = 0.06 m): T_0 = 293 (8-11) and 473 K (1-7); α = 0° (1, 2, 4, 5, 7-9, 11) and 4 (3, 6, 10); Re tan α = 5.88 (3, 6) and 20.4 (10); Ga/Re = 0 (2, 4, 5, 8, 9), 465 (1, 3, 6, 7), 625.8 (11) and 688.4 (10); without thermodiffusion (3, 4, 8).

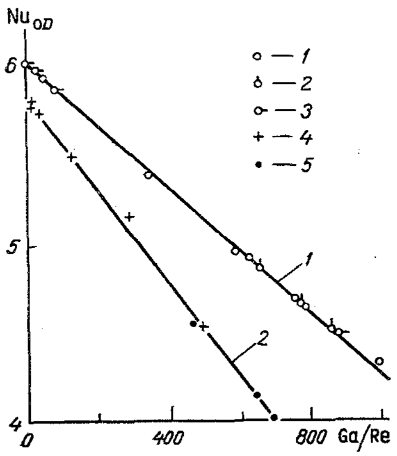

Fig. 3

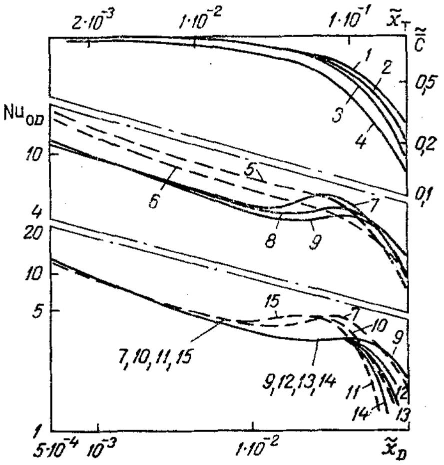

Fig. 4

Fig. 3. Dependence of Nu_{0D} on Ga/Re for different values of h and the temperature factor ψ (P = 101 kPa, X_{01} = 0.002, T_s = 1423 K, α = 0°, $\tilde{x}_T$ = 0.005); T_0 = 293 (1-3) and 473 K (4, 5), h_0 = 0.02 (3), 0.04 (2) and 0.06 m (1, 4, 5).

Fig. 4. Dependence of $\tilde{\tilde{c}}$ and Nu_{0D} on $\tilde{x}_T$ and $\tilde{x}_D$ in the absence of the effect of free convection (P = 101 kPa; X_{01} = 0.002, T_s = 1423 K, h_0 = 0.06 m): T_0 = 293 (3-5, 7, 10, 11, 15), 373 (2, 8) and 473 K (1, 6, 9, 12-14); α = 0° (1-9), 2° (10-13) and 4° (14, 15); Re tan α = 0 (1-9); 2.94 (12), 4.09 (10, 4.41 (13), 5.88 (14), 6.13 (11) and 20.4 (15); without thermodiffusion (5, 6).

depletion of silane from the gas phase (see curves 4 and 8 in Fig. 2). Allowing for thermodiffusion and free convection leads to the formation of a maximum on this relation. The presence of the maximum can be attribute to the effect of these two factors on deposition rate as the gas is heated (Fig. 2).

When the gas mixture is fed into the top of the reactor and flows downward, as shown in Fig. 1, free convection lowers the flow rate near the substrates and thereby reduces silicon deposition rate as well. The effect of free convection is manifest in the region of small

$\tilde{x}_T$. It changes little in the range $x_T \lesssim 0.015$ and nearly ceases upon attainment of the values $\tilde{x}_T' \approx 0.5{=}0.55$, which corresponds to the end of the thermal initial section of the symmetrically heated plane channel [5]. An increase in the temperature factor $\psi = T_s/T_0$ is accompanied by a decrease in the value of $\tilde{x}_T$ at which the effect of free convection on Nu_{OD} can be ignored (Fig. 2). This decrease corresponds to shortening of the thermal initial section with an increase in ψ. A similar dependence ot this length on ψ in the case of circular tubes was reported in [6].

It follows from Eq. (3) that the contribution of free convection to heat and mass transfer is characterized by the ratio Ga/Re. This criterion was also adopted in [5, 6].

A single straight line (see Fig. 3) in the coordinates Nu_{OD}–Ga/Re satisfactorily generalizes the value of Nu_{OD} obtained at constant ψ but different h, i.e., different Ga and Re. The dependence of Nu_{OD}(Ga/Re) weakens with an increase in ψ. At Ga/Re = 700 and $T_s = 1423$ K, the number Nu_{OD} decreases by 30% when $T_0 = 473$ K and by 20% when $T_0 = 293$ K compared to the case of the absence of free convection.

Reverse flow develops with an increase in Ga/Re to a certain limit. The distance from the channel inlet over which recirculation begins increases with a decrease in Ga/Re. This distance cannot exceed the length of the thermal initial section. Thus, at values of Ga/Re which are less than a certain critical value $(Ga/Re)_{cr}$, recirculation does not develop at any distance from the inlet.

Since Eqs. (2-5) are invalid in the region $Ga/Re \geq (Ga/Re)_{cr}$, the resulting values of $(Ga/Re)_{cr}$ are understated and very approximate. As the critical Ga/Re, we took the minimum value of this complex at which the iteration diverges in the determination of the longitudinal pressure gradient in the channel, i.e., when $\partial\tilde{p}/\partial\tilde{x}$ changes sign during the iteration.

The value of $(Ga/Re)_{cr}$ increases with an increase in the temperature factor. The value of $(Ga/Re)_{cr} = 760$ at $\psi = 3$, while $(Ga/Re)_{cr}$ increases to 1000 at $\psi = 4.86$.

The convergence of the channel increases $(Ga/Re)_{cr}$. This effect is reinforced with an increase in the product Re tan α. It is known that the velocity profile becomes fuller with an increase in Re tan α, which prevents the occurrence of recirculation. At low Re tan α, the curve $Nu_{OD}(\tilde{x}_D)$ has the same character as at $\alpha = 0$. The effect of free convection on Nu_{OD} decreases with an increase in α (see curves 5 and 6 in Fig. 2).

Due to the very low concentration of silane in the gas mixture, the effect of concentrational convection on deposition rate is negligible. A change in the molar fraction of silane at the reactor inlet from $1\cdot10^{-3}$ to $3\cdot10^{-3}$ at h = 0.06 m, $T_0 = 293$ K, $T_s = 1423$ K, and $u_0 = 0.7$ m/sec increases Ga/Re from 496 to 497.8. With fixed values of $\tilde{x}_T$, the values of Nu_{OD} differ by no more than 1.5%.

The reduction in Nu_{OD} due to free convection does not exceed 5% at Ga/Re less than 115 and 165 when $\psi = 3$ and 4.86, respectively (see Fig. 3). Thus, at $Ga/Re \leq 100$, the effect of free convection can be ignored.

The condition $Ga/Re \leq 100$ is not very restrictive. It may be frequently encountered in practice. For example, at atmospheric pressure, $h_9 = 0.03$, and a molar fraction of silane equal to 0.002, the value $Ga/Re \leq 100$ is reached when $u_0 \geq 0.86$ m/sec if $T_0 = 293$ K and when $u_0 \geq 0.236$ m/sec if $T_0 = 473$ K. The value of u_0 was calculated for 293 K in both cases.

Since the value of Ga/Re is proportional to the square of the pressure, at reduced pressures in the reactor it is possible to ignore free convection in nearly any case encountered in practice.

Thermodiffusion significantly reduces Nu_{OD} on the thermal initial section of the channel, as is evident from a comparison of curves 5 and 7, as well as 6 and 9, in Fig. 4.

The ratio of the mass flows of silane calculated with and without allowance for thermodiffusion (j_1 and j_1', respectively) is equal to

$$\frac{j_1}{j_1'} = \left[\frac{\partial X_1}{\partial \tilde{y}} + \frac{\alpha_T X_1 (1 - X_1)}{\tilde{T}} \frac{\partial \tilde{T}}{\partial \tilde{y}}\right] \Big/ \frac{\partial X_1}{\partial \tilde{y}} = 1 + K_{TD}, \qquad (9)$$

where

$$K_{TD} = \frac{\alpha_T X_1(1-X_1)}{\tilde{T}} \frac{\partial \tilde{T}}{\partial \tilde{y}} \Big/ \frac{\partial X_1}{\partial \tilde{y}}. \tag{10}$$

To obtain an approximate value of K_{TD}, we taken $X_1 << 1$, $X_{S1} = 0$,

$$X_1 = \frac{X_{01} + X_{S1}}{2} = \frac{X_{01}}{2}, \quad \frac{\partial X_1}{\partial y} = \frac{X_{01}}{\delta_D}, \quad \frac{\partial T}{\partial y} = \\ = \frac{T_0 - T_S}{\delta_T}, \quad T = \frac{T_0 + T_S}{2}. \tag{11}$$

It is known that the ratio of the thickness of the thermal and concentration boundary layers is equal to

$$\frac{\delta_T}{\delta_D} = \frac{\mathrm{Pr}_D}{\mathrm{Pr}}. \tag{12}$$

Inserting (11) and (12) into (10), we obtain

$$K_{TD} = \frac{\alpha_T (1 - \tilde{T}_S)\,\mathrm{Pr}}{(1 + \tilde{T}_S)\,\mathrm{Pr}_D}. \tag{13}$$

This estimate of K_{TD} turns out to be fairly exact. At $\tilde{x}_T \leq 0.012$, the ratio of the mass flows j_1/j_1' calculated from Eqs. (9), (13) differ by 3-5% from the ratio obtained by numerically solving Eqs. (2-5).

The value of Nu_{0D} is nearly independent of the temperature of the gas phase at the reactor inlet if $\tilde{x}_D \leq 0.02$-0.025. In the range $\tilde{x}_D = 0.02$-0.2, an increase in T_0 is accompanied by a decrease in Nu_{0D} (Fig. 4).

The temperature of the deposition surface T_S also has little effect on Nu_{0D}. This effect increases somewhat with an increase in $\tilde{x}_D$. The ratio of Nu_{0D} at $T_S = 1423$ K to this number at 1273 K changes from 1.018 to 1.088 with an increase in $\tilde{x}_D$ from 0.004 to 0.0215.

If the reaction has the form of a convergent channel which converges with the angle α (see Fig. 1), then for the governing linear dimension in the determination of Nu_{0D} it is convenient to take not the inlet value of h but the running value between the substrate holders, equal to $h = h_0 - x \tan \alpha$.

With such a definition of the Nusselt number, in the absence of free convection the relation $Nu_{0D}(\tilde{x}_D)$ is a single curve for all α in the investigated range Re tan α = 0-6 on the channel section $\tilde{x}_D \leq 0.03$-0.04. This section contracts with an increase in ψ and Re tan α.

Figure 2a and Fig. 4 show the change in the mass concentration of silane averaged over the cross section of the reactor. In most actual cases of deposition from the gas phase, $\tilde{x}_D$ does not exceed 0.02-0.03. Here, no more than 20-30% of the silane introduced into the reactor is used.

In the absence of thermodiffusion and free convection, the curves $Nu_{0D}(\tilde{x}_D)$ have a simple form (see Figs. 2 and 4) and can be approximated by expressions convenient for rough calculations. These expressions can be represented in the form

$$\mathrm{Nu}_{0D} = A\tilde{x}_D^{-n} \tag{14}$$

at $\tilde{x}_D \leq 0.03$ and

$$\mathrm{Nu}_{0D} = B \exp(-m\tilde{x}_D) \tag{15}$$

at $\tilde{x}_D > 0.03$.

The values of A, n, B, and m depend on $\tilde{x}_D$ and the temperature factor $\psi = T_S/T_0$. The following values of these quantities wre obtained from a numerical solution at $T_S = 1423$ K and $T_0 = 293$ and 473 K (ψ = 3 and 4.86):

$$0{,}0005 \leqslant \tilde{x}_D \leqslant 0{,}003,\ A = 0{,}28\psi + 0{,}11,\ n = 0{,}459 - 0{,}0263\psi;$$
$$0{,}003 \leqslant \tilde{x}_D \leqslant 0{,}03,\ A = 0{,}344\psi + 0{,}42,\ n = 0{,}372 - 0{,}0215\psi; \tag{16}$$
$$0{,}03 \leqslant \tilde{x}_D \leqslant 0{,}14,\ B = 1{,}93\psi + 0{,}67,\ m = 3{,}76\psi + 1{,}67.$$

The deviation of the results of approximate calculations by Eqs. (14-16) from the numerical solution without allowance for thermodiffusion and free convection is no greater than 3%. The effect of thermodiffusion and free convection can be evaluated from Eqs. (9-13) and Figs. 2-4.

NOTATION

c, mass concentration; c_p, isobaric heat capacity; D, coefficient of diffusion of silane in hydrogen; j_1, mass flux of silane; H, enthalpy; P, pressure,; T, temperature; α_T, thermodiffusion ratio; ρ, density; μ, absolute viscosity; λ, thermal conductivity; x, y, h, r, y_s, α, space coordinates and geometric dimensions of the reactor (see Fig. 1); u, v, velocity components in the x and y directions, respectively; $\psi = T_s/T_0$, temperature factor; $\tilde{c}_p = c_p/c_{p0}$; $\tilde{D} = D/D_0$, $\tilde{H} = H/H_0$, $\tilde{P} = (P_0 - g\rho_0)/(\rho_0 u_0^2)$, $\tilde{T} = T/T_0$, $\tilde{\rho} = \rho/\rho_0$, $\tilde{\lambda} = \lambda/\lambda_0$, $\tilde{u} = u/u_0$, $\tilde{v} = v\,\mathrm{Re}/u_0$, $\tilde{y} = y/r_0$, $\tilde{x} = x/(\mathrm{Re}\,h_0)$, $\tilde{x}' = x/(\mathrm{Re}' r_0')$ $\tilde{y}_\text{п} = x\,\mathrm{tg}\,(\alpha/2)/r_0$, $\mathrm{Re} = u_0 h_0 \rho_0/\mu_0$, $\mathrm{Re}' = u_0 r_0 \rho_0/\mu_0$, $\mathrm{Pr} = \mu_0 c_{p0}/\lambda_0$, $\mathrm{Pr}_D = \mu_0/(\rho_0 D_0)$, $\mathrm{Ga}' = g r_0^3 \rho_0^2/\mu_0^2$, $\mathrm{Ga} = g h^3 \rho_0^2/\mu_0^2 = 8\,\mathrm{Ga}'$, $\mathrm{Nu}_{0D} := 2 j_1 r/(c_{01}\rho_0 D_0)$, $\tilde{x}_T = \tilde{x}/\mathrm{Pr}$, $\tilde{x}_D = \tilde{x}/\mathrm{Pr}_D$. Indices: 0, at reactor inlet; s, on deposition surface; 1, silane; 2, hydrogen.

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