

ASYMPTOTIC SOLUTION FOR NONLINEAR CHEMICAL VAPOR DEPOSITION PROBLEMS

BY

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This paper is in the memory of one of the authors, Oleg Tikhomirov, who recently passed away. The coauthors wish to express their deep feeling of loss of a dear friend. We greatly miss him.

Abstract. The problem of chemical vapor deposition involving reaction kinetics of any order n at a heated substrate is considered. The deposition process is then described by a convective diffusion equation, coupled to nonlinear boundary conditions, describing the chemical reaction taking place at the heated substrate, where the nonlinearity is given in terms of the order n of the reaction kinetics. We derive boundary layer equations and use a combination of perturbation and similarity methods to find the deposition rate along the susceptor.

1. Introduction. The process of growing a thin film on a heated substrate by chemical vapor deposition epitaxial technique has evoked great interest. An understanding of the underlying mechanism of this process is needed.

The problem was investigated by Levich [1], Leveque [2], and Malmuth [3] for the case of first-order reaction kinetics, which resulted in a linear problem. The authors presented a solution for reaction kinetics of order n , see [4]. Recently, Wilder [5] solved the case of second-order reaction kinetics. The deposition rate along the susceptor was worked out and demonstrated for the example of GaAs deposition. This analytical work experienced great resonances in the field, and further analytical work is desired.

We are concerned with the process of modeling a reactor that employs two-dimensional multicomponent channel flow with the chemical reaction taking place at the susceptor (Fig. 1, p. 586). The concentration of the reacting species is assumed to be small compared to the concentration of the carrier gas. As a result, the variation of the physical properties of the overall gas due to the chemical reaction can be neglected.

The governing equations modeling this process are derived from the conservation

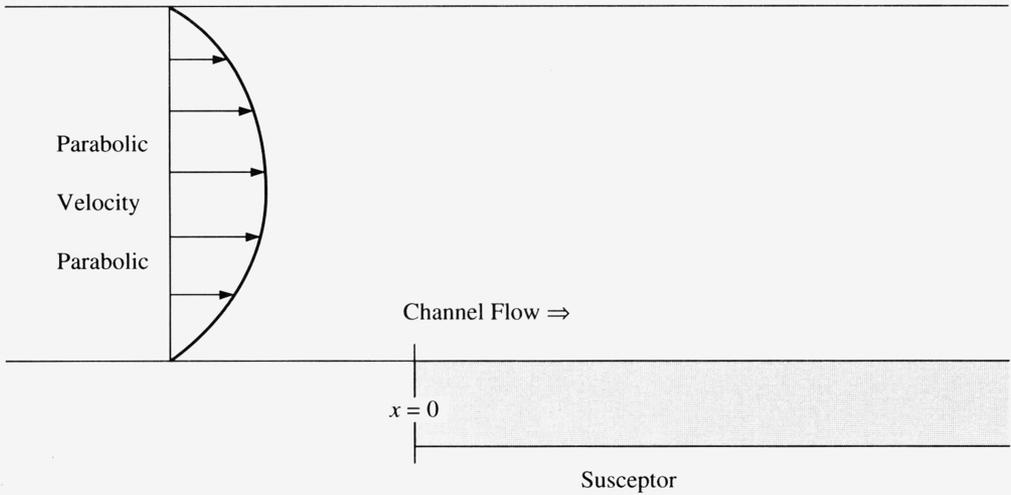


FIG. 1. Chemical vapor deposition apparatus

laws for laminar, incompressible, steady fluid flow and from empirical laws for diffusion of species, since at steady state the incoming flux at the susceptor must be balanced by the species reaction rate. One obtains a convective diffusion equation coupled to nonlinear boundary conditions describing the reaction at the heated substrate.

We solve the boundary value problem for arbitrary order n of reaction kinetics. Under the assumption that the concentration of the reacting species is low, and the diffusion coefficient in gases is considered independent of the composition, the governing equations that model this chemical vapor deposition process are

$$\text{Diffusion Equation } (\mathbf{v} \cdot \nabla)c = D\nabla^2 c \quad (1.1)$$

and

$$\text{Continuity Equation } \nabla \cdot \mathbf{v} = 0, \quad (1.2)$$

where \mathbf{v} is the barycentric gas velocity, c the concentration of the reacting species, and D the diffusion coefficient of the species.

The symmetry of the problem allows us to consider the concentration c and the velocity vector \mathbf{v} to be independent of the z component. Equations (1.1) and (1.2) are then the equations for two-dimensional flow in a channel and can, therefore, be written as

$$\text{Diffusion Equation } v_x \frac{\partial c}{\partial x} + v_y \frac{\partial c}{\partial y} = D \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} \right) \quad (1.3)$$

and

$$\text{Continuity Equation } \frac{\partial v_x}{\partial x} + \frac{\partial v_y}{\partial y} = 0, \quad (1.4)$$

where v_x and v_y are the x and y components of \mathbf{v} . We further neglect turbulent effects so that

$$v_y = 0, \quad (1.5a)$$

and together with the continuity equation (1.4) we conclude that

$$v_x(x, y) = u(y). \quad (1.5b)$$

We therefore obtain for the diffusion equation

$$u(y) \frac{\partial c}{\partial x} = D \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} \right). \quad (1.6)$$

For the boundary conditions we observe that the apparatus is arranged so that in the (x, y) -plane where $-\infty \leq x \leq \infty$ and $0 \leq y \leq \infty$, the susceptor is located at $y = 0$ and $x > 0$. For $x \leq 0$ we assume the velocity profile to be parabolic, see Fig. 1. The boundary conditions are specified at the reaction surface and in the bulk of the flow. Since we are considering heterogeneous reactions, the change in concentration occurs in a thin boundary layer on the reaction surface. We therefore have, for the condition in the bulk of the flow,

$$c(0, y) = c_\infty \quad (1.7a)$$

and

$$c(x, \infty) = c_\infty, \quad (1.7b)$$

where c_∞ is the bulk concentration of the reacting species. For the boundary condition on the reaction surface, we first recall that, according to Fick's law, there is a linear relation between the flux of the reacting species and its concentration gradient, and we obtain for the mass flux to the surface

$$j = D \left(\frac{\partial c}{\partial y} \right)_{(x, y)=(x, 0)}. \quad (1.8)$$

When the reaction mechanism is not explicitly given, one usually employs an empirical reaction law in the form of a power law for the reaction rate of the species. Then steady state implies that the incoming flux at the surface is balanced by the reaction rate of the species. We, therefore, have for the boundary condition at the susceptor

$$D \left(\frac{\partial c}{\partial y} \right)_{(x, y)=(x, 0)} = k(c(x, 0))^n, \quad (1.9)$$

where k is the rate constant of the reaction, which is independent of the concentration of the reactant and obeys the Arrhenius law $k = k_0 e^{-(E/RT)}$, with E being the activation energy and T the absolute temperature. The exponent n denotes the order of the reaction; it may be an integer or a fraction.

Throughout our investigations we are only interested in the solution to the boundary layer problem. In Sec. 2 we nondimensionalize the boundary value problem (1.6), (1.7), (1.9) and derive an asymptotic representation for the boundary layer equations. In Sec. 3 we present similarity solutions to the problem in terms of Kummer's functions, and from these solutions we derive, in Sec. 4, the expression for the deposition rate.

2. Boundary layer equations. We first reduce our boundary value problem to a dimensionless form. We introduce a characteristic length l along which the change in concentration takes place and a characteristic flow velocity U_0 .

We then define the dimensionless coordinates

$$x^* = \frac{x}{l} \quad (2.1a)$$

and

$$y^* = \frac{y}{l}. \quad (2.1b)$$

For the dimensionless velocity we let

$$U(y^*) = \frac{u(y)}{U_0}, \quad (2.2)$$

and for the dimensionless concentration

$$C(x^*, y^*) = \frac{c(x, y)}{c_\infty}. \quad (2.3)$$

Application of Eqs. (2.1)–(2.3) to the boundary value problem (1.6), (1.7), (1.9) yields the following nondimensional form:

$$U(y^*) \frac{\partial C}{\partial x^*} = \varepsilon \left(\frac{\partial^2 C}{\partial x^{2*}} + \frac{\partial^2 C}{\partial y^{2*}} \right), \quad (2.4)$$

$$C(0, y^*) = 1, \quad (2.5a)$$

$$C(x^*, \infty) = 1, \quad (2.5b)$$

$$\alpha \left(\frac{\partial C}{\partial y^*} \right)_{(x^*, y^*)=(x^*, 0)} = (C(x^*, 0))^n. \quad (2.5c)$$

We obtain two dimensionless parameters ε and α , which are defined as

$$\varepsilon = \frac{D}{\nu} \cdot \frac{\nu}{U_0 l} = \frac{1}{Sc} \cdot \frac{1}{Re} = \frac{1}{Pe_D} \quad (2.6a)$$

and

$$\alpha = \frac{D}{kc_\infty^{n-1} l} = \frac{1}{Da}, \quad (2.6b)$$

where Sc denotes the Schmidt number, Re the Reynolds number, Pe_D the Peclet number, Da denotes the Damköhler number, and ν is the kinematic viscosity. For gases $D/\nu = O(1)$ so that when the Reynolds number $Re \gg 1$ we have $\varepsilon \ll 1$. The dimensionless similarity parameter Da measures the reaction velocity compared to the rate of transfer of particles to the reaction surface. For rapid reactions we can also assume that $Da \gg 1$, i.e., $\alpha \ll 1$.

We now derive the boundary layer equations and, upon expanding $C(x^*, y^*)$ in an asymptotic series in ε , we will find a distinguished limit, relating both parameters.

Let δ be the thickness of the boundary layer on the reaction surface, and define the dimensionless quantity

$$\gamma = \frac{\delta}{l}. \quad (2.7a)$$

Denote the inner variable by

$$\tilde{y} = \frac{y^*}{\gamma}. \tag{2.7b}$$

Within the boundary layer at the reaction surface y^* is small, and we expand $U(y^*)$ as

$$U(y^*) = a_0 + a_1 y^* + a_2 y^{*2} + \dots. \tag{2.8}$$

We assume the reaction surface to be impermeable so that a no-slip condition requires that $a_0 = 0$. Hence, we obtain

$$U(y^*) = a_1 \gamma \tilde{y} + a_2 \gamma^2 \tilde{y}^2 + \dots. \tag{2.9}$$

We write $C(x^*, y^*; \epsilon)$ as an asymptotic expansion in terms of functions λ , which depend on ϵ .

$$\begin{aligned} C(x^*, y^*; \epsilon) &= C(x^*, \gamma \tilde{y}; \epsilon) = \tilde{C}(x^*, \tilde{y}; \epsilon) \\ &= C_0(x^*, \tilde{y}) + \lambda(\epsilon) C_1(x^*, \tilde{y}) + \dots. \end{aligned} \tag{2.10}$$

If we now substitute expansions (2.9) and (2.10) into the governing equation (2.4), we obtain

$$a_1 \gamma \tilde{y} \frac{\partial C_0}{\partial x^*} + \dots = \epsilon \left(\frac{\partial^2 C_0}{\partial x^{*2}} + \lambda(\epsilon) \frac{\partial^2 C_1}{\partial x^{*2}} + \frac{1}{\gamma^2} \frac{\partial^2 C_0}{\partial \tilde{y}^2} + \frac{\lambda(\epsilon)}{\gamma^2} \frac{\partial^2 C_1}{\partial \tilde{y}^2} + \dots \right). \tag{2.11}$$

If we balance the leading-order term, we obtain for the distinguished limit

$$\gamma = \epsilon^{1/3}$$

and the problem

$$a_1 \tilde{y} \frac{\partial C_0}{\partial x^*} = \frac{\partial^2 C_0}{\partial \tilde{y}^2}, \tag{2.12}$$

with boundary conditions

$$C_0(0, \tilde{y}) = 1, \tag{2.13a}$$

$$C_0(x^*, \infty) = 1, \tag{2.13b}$$

$$\beta \left(\frac{\partial C_0}{\partial \tilde{y}} \right)_{(x^*, \tilde{y})=(x^*, 0)} = (C_0(x^*, 0))^n. \tag{2.13c}$$

If we further introduce

$$\tilde{x} = \frac{x^*}{a_1} \tag{2.14}$$

and

$$C_0(x^*, \tilde{y}) = C_0(a_1 \tilde{x}, \tilde{y}) = \tilde{C}_0(\tilde{x}, \tilde{y}), \tag{2.15}$$

we can eliminate the constant a_1 from the problem and obtain

$$\tilde{y} \frac{\partial \tilde{C}_0}{\partial \tilde{x}} = \frac{\partial^2 \tilde{C}_0}{\partial \tilde{y}^2}, \tag{2.16}$$

$$\tilde{C}_0(0, \tilde{y}) = 1, \tag{2.17a}$$

$$\tilde{C}_0(\tilde{x}, \infty) = 1, \tag{2.17b}$$

$$\beta \left(\frac{\partial \tilde{C}_0}{\partial \tilde{y}} \right)_{(\tilde{x}, \tilde{y})=(\tilde{x}, 0)} = (\tilde{C}_0(\tilde{x}, 0))^n, \tag{2.17c}$$

where

$$\beta = \frac{\alpha}{\varepsilon^{1/3}}. \tag{2.18}$$

Observing the above, under the assumption of rapidly reacting surfaces, i.e., for $\alpha \ll \varepsilon^{1/3}$ (or $\beta \ll 1$), the concentration of the reactant is the bulk concentration c_∞ plus a small perturbation, i.e., a perturbation in terms of β .

Therefore, we expand \tilde{C}_0 in β as follows:

$$\tilde{C}_0(\tilde{x}, \tilde{y}; \beta) = C_0^0(\tilde{x}, \tilde{y}) + \omega(\beta)C_0^1(\tilde{x}, \tilde{y}) + \dots \tag{2.19}$$

If we apply this expansion to problem (2.16), (2.17) and balance the higher-order terms, we obtain

$$\omega^n = \beta \tag{2.20}$$

and the problem

$$\tilde{y} \frac{\partial C_0^0}{\partial \tilde{x}} = \frac{\partial^2 C_0^0}{\partial \tilde{y}^2}, \tag{2.21}$$

$$C_0^0(0, \tilde{y}) = 1, \tag{2.22a}$$

$$C_0^0(\tilde{x}, \infty) = 1, \tag{2.22b}$$

$$C_0^0(\tilde{x}, 0) = 0, \tag{2.22c}$$

plus the correction

$$\tilde{y} \frac{\partial C_0^1}{\partial \tilde{x}} = \frac{\partial^2 C_0^1}{\partial \tilde{y}^2}, \tag{2.23}$$

$$C_0^1(0, \tilde{y}) = 0, \tag{2.24a}$$

$$C_0^1(\tilde{x}, \infty) = 0, \tag{2.24b}$$

$$\left(\frac{\partial C_0^1}{\partial \tilde{y}} \right)_{(\tilde{x}, \tilde{y})=(\tilde{x}, 0)} = (C_0^1(\tilde{x}, 0))^n. \tag{2.24c}$$

Unlike the parameter-free form of problem (2.16)–(2.17c) (see Conclusions), those two coupled problems do indeed have similarity solutions, which we examine in the next section.

3. Similarity solution. An analysis of Eq. (2.21) shows the similarity variable to be of the form

$$\eta = \mu \frac{\tilde{y}}{\tilde{x}^{1/3}}, \tag{3.1}$$

with the constant $\mu = (\frac{1}{9})^{1/3}$.

Let us define the following functions:

$$C_0^0(\tilde{x}, \tilde{y}) = F_0(\eta), \tag{3.2}$$

$$C_0^1(\tilde{x}, \tilde{y}) = \frac{1}{\tilde{x}^{1/3n}} F_1(\eta), \tag{3.3}$$

where $F_0(\eta)$ and $F_1(\eta)$ denote the similarity solutions of the leading-order problem and the correction problem, respectively. Then, together with Eqs. (2.19) and (2.22), the solution can be given as

$$\begin{aligned} \tilde{C}_0(\tilde{x}, \tilde{y}) &= F_0(\eta) + \left(\frac{\beta^3}{\tilde{x}}\right)^{1/(3n)} F_1(\eta) + \dots \\ &= F_0(\eta) + \frac{1}{\tilde{x}^{1/(3n)}} F_1(\eta) + \dots \end{aligned} \tag{3.4}$$

From Eq. (3.4) we see that the solution, with $\beta \ll 1$, will be valid for $\tilde{x} \gg 1$ and $\eta = O(1)$.

Let us now find the similarity solutions $F_0(\eta)$ and $F_1(\eta)$. We apply Eq. (3.2) to the leading-order problem and obtain

$$F_0''(\eta) + 3\eta^2 F_0'(\eta) = 0, \tag{3.5}$$

$$F_0(\infty) = 1, \tag{3.6a}$$

$$F_0(0) = 0. \tag{3.6b}$$

We easily see this problem has the solution

$$F_0(\eta) = \frac{\int_0^\eta e^{-s^3} ds}{\int_0^\infty e^{-s^3} ds} = \frac{3}{\Gamma(\frac{1}{3})} \int_0^\eta e^{-s^3} ds. \tag{3.7}$$

If we apply Eq. (3.3) to the problem of the next order, Eqs. (2.23) and (2.24), we arrive, by implementing the result (3.7), at the following problem:

$$F_1''(\eta) + 3\eta^2 F_1'(\eta) + \left(\frac{3}{n}\right) \eta F_1(\eta) = 0, \tag{3.8}$$

$$\lim_{\eta \rightarrow \infty} \eta^{1/n} F_1(\eta) = 0, \tag{3.9a}$$

$$F_1(\infty) = 0, \tag{3.9b}$$

$$F_1(0) = \left(\mu \frac{3}{\Gamma(\frac{1}{3})}\right)^{1/n}. \tag{3.9c}$$

If we make a change of variable $t = \eta^3$, and let

$$F_1(\eta) = \eta e^{-\eta^3} \phi(\eta^3), \tag{3.10}$$

Eq. (3.8) transforms to Kummer's equation

$$t\phi''(t) + \left(\frac{4}{3} - t\right) \phi'(t) - \left(\frac{3n-1}{3n}\right) \phi(t) = 0, \tag{3.11}$$

which has the solution

$$\phi(t) = K_1 M\left(\frac{3n-1}{3n}, \frac{4}{3}, t\right) + K_2 U\left(\frac{3n-1}{3n}, \frac{4}{3}, t\right), \tag{3.12}$$

where K_1 and K_2 are constants to be determined by the boundary conditions. The functions $M(a, b, t)$ and $U(a, b, t)$ are Kummer's functions.

Equations (3.10) and (3.12), in conjunction with boundary condition (3.9a), yield

$$K_1 = 0 \quad (3.13)$$

and, in conjunction with boundary condition (3.9c), yield

$$K_2 = \frac{\Gamma(\frac{3n-1}{3n})}{\Gamma(\frac{1}{3})} \left(\mu \frac{3}{\Gamma(\frac{1}{3})} \right)^{1/n}; \quad (3.14)$$

hence, we have for F_1

$$F_1(\eta) = \frac{\Gamma(\frac{3n-1}{3n})}{\Gamma(\frac{1}{3})} \left(\mu \frac{3}{\Gamma(\frac{1}{3})} \right)^{1/n} \eta e^{-\eta^3} U\left(\frac{3n-1}{3n}, \frac{4}{3}, \eta^3\right). \quad (3.15)$$

Combining Eq. (3.4) with the similarity solutions (3.7) and (3.15) gives the solution for the concentration from which we can derive the deposition rate on the reaction surface; but before that, we will discuss the region of validity of our solution.

Consider the solution for $\tilde{C}_0(\tilde{x}, \tilde{y})$, which has the form

$$\tilde{C}_0(\tilde{x}, \tilde{y}) = \Gamma\left(\frac{1}{3}, \eta\right) + \left(\frac{\beta^3}{\tilde{x}}\right)^{1/(3n)} \frac{\Gamma(\frac{3n-1}{3n})}{\Gamma(\frac{1}{3})} \left(\frac{3^{1/3}}{\Gamma(\frac{1}{3})}\right)^{1/n} \eta e^{-\eta^3} U\left(\frac{3n-1}{3n}, \frac{4}{3}, \eta^3\right), \quad (3.16)$$

where $\Gamma(\frac{1}{3}, \eta)$ is the incomplete Gamma function, i.e., F_0 .

An analysis of this result shows that this solution is valid as long as for $\beta \ll 1$, we have $(\beta^3/\tilde{x})^{1/(3n)} \ll 1$ for $\eta = O(1)$. In other words, this means that the solution is valid as long as

$$\frac{a_1 U_0 D^2}{k^3 c_\infty^{3n-3} l} \ll x. \quad (3.17)$$

There are numerous ways of achieving this, e.g., by varying the length l of the plate or by varying the effective rate constant kc_∞^{n-1} , for example, by varying the temperature T .

The behaviour of our solution when $\eta \rightarrow 0$ has also been analysed. Figures 2 and 3 (see p. 593) show, for the case of $n = 1$, how the solution blows up near $\eta = 0$ when we increase the order of $(\beta^3/\tilde{x})^{1/(3n)}$ and that for increasing order of n this effect becomes much less severe.

The importance of our result is indicated in Fig. 4 (see p. 594), which compares solutions for the concentration for different n . We see, while the qualitative properties of the solution remain the same, that there is a considerable quantitative increase in concentration, which decreases along the similarity curves $\eta = \text{const}$. In the next section we will see the effect on the deposition rate.

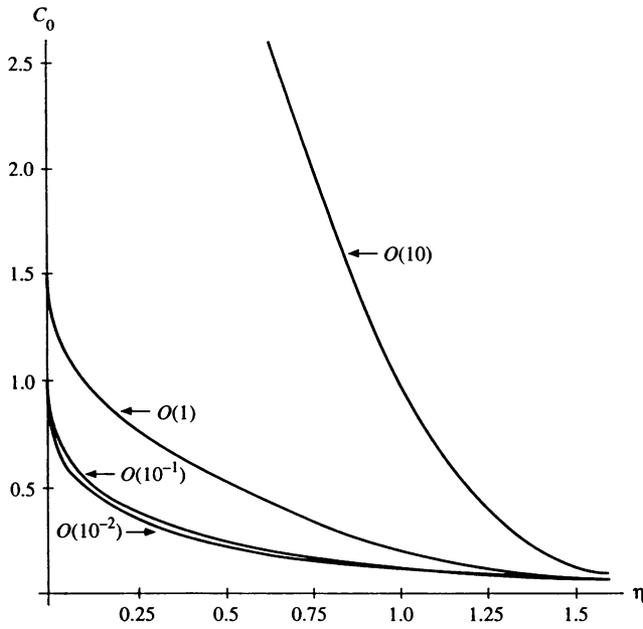


FIG. 2. Concentration \tilde{C}_0 with $n = 1$ and $\beta^3/\bar{x} = O(10^{-2})$ to $O(10^1)$

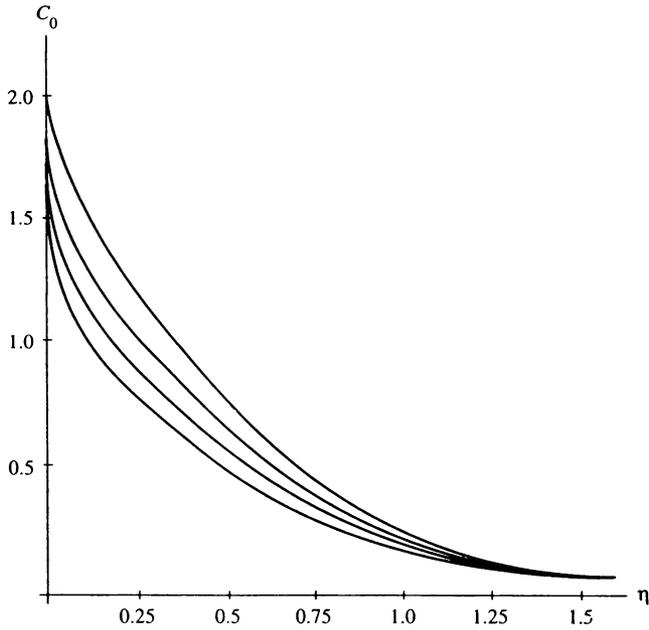


FIG. 3. Concentration \tilde{C}_0 with $n = 10$ and $\beta^3/\bar{x} = O(10^{-2})$ to $O(10^1)$

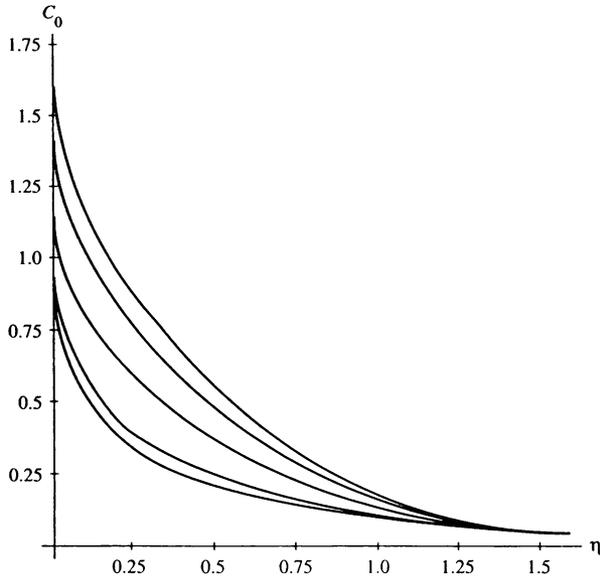


FIG. 4. Concentration \tilde{C}_0 with $n = 1$ to $n = 10$ and $\eta^3/\tilde{x} = O(10^{-2})$

4. Deposition rate. Since the main interest of the application of this analysis lies in the problem of prediction of deposition for chemical agents such as GaAs or HgCdTe, we will here solve for the deposition rate. We will derive a representation involving reaction rates of any order n .

The deposition rate is given by

$$G = D \left(\frac{\partial c}{\partial y} \right)_{(x,y)=(x,0)} \tag{4.1}$$

To find G , we return to dimensional coordinates. For convenience we also let

$$\tau = \frac{U_0}{9Da_1l^3}$$

and

$$\kappa^{1/3} = \frac{c_\infty}{\Gamma(\frac{1}{3})} \left(\frac{3D^2a_1U_0}{l} \right)^{1/3}$$

Then, by Eq. (3.16), we obtain

$$D \frac{\partial c}{\partial y} = e^{-\tau xy^2} \left[\left(\frac{\kappa}{x} \right)^{1/3} + \left(\frac{\kappa}{x} \right)^{(n+1)/(3n)} \left(\frac{1}{k} \right)^{1/n} \frac{\Gamma(\frac{3n-1}{n})}{9^{1/3}\Gamma(\frac{1}{3})} \cdot \left(U \left(\frac{3n-1}{3n}, \frac{4}{3}, \tau xy^2 \right) [1 - 3\tau xy^2] - \frac{3n-1}{3n} \tau xy^2 U \left(\frac{6n-1}{3n}, \frac{7}{3}, \tau xy^2 \right) \right) \right] \tag{4.2}$$

In order to find the deposition rate at the plate, in the above equation, we let $y \rightarrow 0$. As a result we have

$$G = \left(\frac{\kappa}{x}\right)^{1/3} - \left(\frac{\kappa}{x}\right)^{(n+1)/(3n)} \left(\frac{1}{k}\right)^{1/n} \frac{\Gamma\left(\frac{3n-1}{3n}\right)}{3^{5/3}\Gamma\left(\frac{1}{3}\right)^2\Gamma\left(\frac{2n-1}{3n}\right)} \frac{2\pi}{\sqrt{3}}. \quad (4.3)$$

Figure 5 shows the deposition rate for $n = 1$, and Fig. 6 (see p. 596) compares the deposition rates for $n = 1$, $n = 10$, and $n = 20$. We see that the $n = 1$ curve is the highest curve, so that, if one would not account for nonlinear effects, the deposition rate would be overestimated. For the answer to how great this difference is in a real experiment, one must first investigate an initial parabolic velocity profile. We calculated the deposition rates (Figs. 5, 6) using the parameters derived in [5].

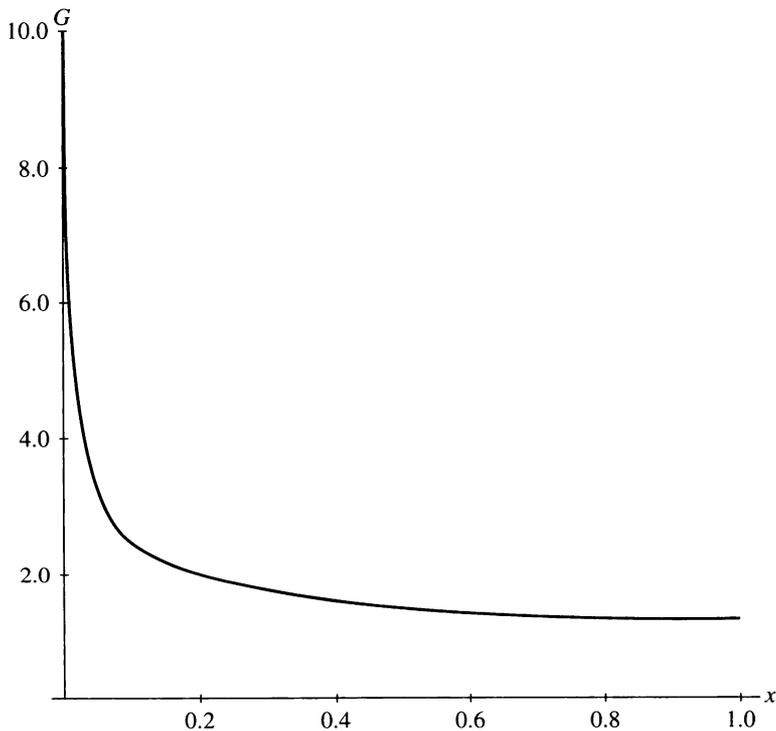


FIG. 5. Deposition rate for $n = 1$

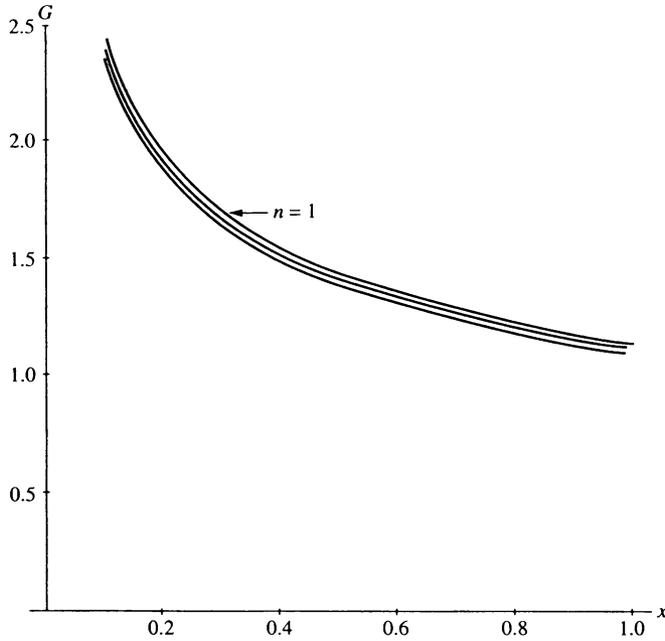


FIG. 6. Comparison of deposition rates for $n = 1$, $n = 10$, and $n = 20$.

Conclusions. We have solved the problem of chemical vapor deposition, involving reaction kinetics of any order n . Our investigations show that a neglect of the nonlinear effects results in an overestimate of the deposition rate.

The solutions of the convective diffusion equation, coupled to nonlinear boundary conditions, were found via perturbation and similarity methods. We should also observe at this point that via the transformation

$$\begin{aligned}\tilde{x} &= \beta^3 \bar{x}, \\ \tilde{y} &= \beta \bar{y},\end{aligned}$$

and

$$\tilde{C}_0(\tilde{x}, \tilde{y}) = \bar{C}_0(\bar{x}, \bar{y}),$$

the problem (2.16), (2.17) reduces to the parameter-free form

$$\bar{y} \frac{\partial \bar{C}_0}{\partial \bar{x}} = \frac{\partial^2 \bar{C}_0}{\partial \bar{y}^2},$$

$$\bar{C}_0(0, \bar{y}) = 1,$$

$$\bar{C}_0(\bar{x}, \infty) = 1,$$

$$\left(\frac{\partial \bar{C}_0}{\partial \bar{y}} \right)_{(\bar{x}, \bar{y})=(\bar{x}, 0)} = (\bar{C}_0(\bar{x}, 0))^n.$$

The full solution of problem (2.16), (2.17) requires the numerical solution of the parameter-free form, since this problem can be shown to have no similarity solutions. However, as we have seen for rapid reactions, our analysis gives an analytic solution, which can be presented, for the deposition rates, as an asymptotic expansion in $1/x$.

Finally, note that, if required, the accuracy may also be improved by including further terms in the asymptotic expansion (2.11).

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