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MASS TRANSPORT AND SURFACE REACTIONS IN LÉVÊQUE'S APPROXIMATION*

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Abstract—This paper addresses the general problem of multicomponent transport and heterogeneous reaction in Lévêque's approximation. The problem always admits an analytic solution when the surface reactions are first order in the concentrations. Here, explicit expressions for the concentration distributions are found for the case of a single reversible surface step, but the procedure equally applies to arbitrarily complex sequences of such steps. The behavior of the solution depends mainly on the value of a dimensionless axial parameter. Aside from axial distance and mean velocity, it involves all diffusion coefficients, reaction rate constants and stoichiometric coefficients. For small values of this parameter the solution behaves as if the surface were a source of constant flux; for large values, as if the surface were at constant concentration. Thus the solution exhibits a continuous change from a Neumann to a Dirichlet problem.

NOMENCLATURE

A,	species label;
$Ai(z)$,	Airy function ([18], p. 446);
B,	height of the channel;
C,	molar concentration;
C_0,	reactant input concentration;
D,	diffusion coefficient;
$f(x)$,	$\equiv f_3(x)$, function (2.12, B.2) describing surface quantities;
$g(x, y)$,	function (2.15) describing concentration profiles;
$\mathbf{j}$,	diffusion flux;
K,	$= k_r/k_p$, equilibrium constant;
k,	reaction rate constant;
L,	Laplace transform operator;
M,	molar mass;
N,	number of species;
$\mathbf{n}$,	unit normal, mass flux;
p,	Laplace transform variable;
R,	number of reactions;
r,	reaction rate;
u_r,	$= 1 - C_r/C_0$, normalized reactant concentration;
u_p,	$= C_p/C_0$, normalized product concentration;
$\mathbf{v}, \bar{v}$,	velocity field, average value;
X,	axial distance, dimensional;
x,	$= Bk_r^3X/6\bar{v}D_r^2$, axial distance, dimensionless;
Y,	transverse distance, dimensional;
y,	$= k_rY/D_r$, transverse distance, dimensionless.

Greek symbols

α,	a constant;
β,	a constant (2.9c);
$\Gamma(n, z)$,	incomplete gamma function ([18], p. 260);
$\Gamma(n)$,	$\equiv \Gamma(n, 0)$, ordinary gamma function ([18], p. 255);
Δ,	$= D_r/D_p$, diffusion ratio;
η,	$= y/(9x)^{1/3}$, transverse coordinate (2.16);
ν,	stoichiometric coefficient;
ξ,	$= \beta x^{1/3}$, axial coordinate (2.13);
ρ,	mass density.

Subscripts

a,	species index ($= 1 \ldots N$);
i,	reaction index ($= 1 \ldots R$);
r, p,	reactant, product.

1. INTRODUCTION

LÉVÊQUE's approximation [1] originally served to calculate temperature distributions in the thermal entrance region of a duct, i.e. that region where Graetz's solution [2, 3] converges slowly. There, heat or mass have not yet penetrated into the body of the fluid; they sample only the linear part of the axial velocity distribution. The appropriately normalized temperature or concentration field $u(x, y)$ then obeys the parabolic equation

$$y\frac{\partial u}{\partial x} = \frac{\partial^2 u}{\partial y^2} \tag{1.1}$$

and the boundary conditions

$$u(0, y) = u(x, \infty) = 0, \tag{1.2}$$

where x and y are normalized axial and transverse coordinates. Equations (1.1) and (1.2) admit similarity solutions under *constant* surface conditions ([4], pp. 307–309). Specifically, if $u(x, 0) = 1$, then

$$u = \Gamma(\tfrac{1}{3}, \eta^3)/\Gamma(\tfrac{1}{3}), \tag{1.3}$$

and if $\partial u/\partial x(x, 0) = -1$, then

$$u = [(9x)^{1/3}/\Gamma(2/3)] \times [\exp(-\eta^3) - \eta\Gamma(2/3, \eta^3)], \tag{1.4}$$

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in which $\eta = y/(9x)^{1/3}$ is the similarity variable. Problems of this type have been extended by Herbeck [5] to include axial diffusion, and by Soliman and Chambré [6] to account for time-dependence. Lévêque's approximation is closely related to that proposed by Fage and Falkner [7]. This last led Lighthill [8] and Chambré and Acrivos [9–11] to reduce the temperature and concentration boundary layer equations to a class of singular Volterra integral equations. These authors were thus able to deal numerically with a wide variety of boundary conditions and to give everywhere convergent series representations when the boundary conditions are linear. Riley [12] has given a recent review of concentration boundary layers.

Lévêque's problem (1.1) and (1.2) does *not* admit a similarity solution when surface conditions vary, e.g. when heterogeneous chemical reactions occur at finite rate. However, one expects closed form solutions for linear boundary conditions. This paper addresses Lévêque's approximation applied to multicomponent transport and heterogeneous reaction, the reaction rate being linear in the concentrations. The solution to this problem bears on catalytic conversion, on dissolution and etching processes, and on crystal growth by chemical vapor deposition. Furthermore, it can provide asymptotic estimates in the entrance region for analytic calculations that consider reactions in ducts [13–16], and, possibly, "starting" procedures for finite difference schemes. The next section presents the desired solution for a single heterogeneous reversible reaction step, and the third investigates its asymptotic behavior.

2. TRANSPORT EQUATIONS AND THEIR SOLUTION

Consider the isothermal, fully developed flow of an incompressible fluid mixture in a channel of height B. At station $X = 0$ a concentration C_0 of reactant A_r is injected into the "carrier". By carrier one means a solvant (liquid or gaseous) that can, however, participate in the chemical reactions. Convection and diffusion carry the reactant downstream and to a reaction surface $(X > 0, Y = 0)$ where it yields a product A_p according to

$$\text{carrier} + \nu_r A_r \rightleftarrows \nu_p A_p. \tag{2.1}$$

Amongst other things, equation (2.1) can represent a deposition or etching reaction; ν_r and ν_p are the absolute values of the stoichiometric coefficients. In Lévêque's approximation, the transport equations for reaction and product concentrations read

$$(6\bar{v}Y/B)\frac{\partial C_a}{\partial X} = D_a \frac{\partial C_a}{\partial Y^2}, \quad (a = r, p), \tag{2.2}$$

with boundary conditions

$$C_r(0, Y) = C_r(X, \infty) = C_0, \tag{2.3a}$$

$$C_p(0, Y) = C_p(X, \infty) = 0. \tag{2.3b}$$

The following boundary conditions hold at $Y = 0$:

$$(D_r/\nu_r)\frac{\partial C_r}{\partial Y} = -(D_p/\nu_p)\frac{\partial C_p}{\partial Y} = r, \tag{2.4a}$$

$$r = k_r C_r - k_p C_p. \tag{2.4b}$$

Equations (2.2) and (2.4) are justified in Appendix A; equation (2.4b) applies *if* the reaction rate of (2.1) is (pseudo-) first order in the concentrations. In dimensionless form (cf. nomenclature) one gets

$$\left.\begin{aligned} y\frac{\partial u_r}{\partial x} &= \frac{\partial^2 u_r}{\partial y^2}, \qquad &(2.5a)\\ \Delta y\frac{\partial u_p}{\partial x} &= \frac{\partial^2 u_p}{\partial y^2}, \qquad &(2.5b)\end{aligned}\right\} \text{ in } (x > 0, y > 0).$$

$$-\frac{1}{\nu_r}\frac{\partial u_r}{\partial y} = -\frac{1}{\Delta \nu_p}\frac{\partial u_p}{\partial y} = 1 - u_r - K^{-1}u_p. \tag{2.6}$$

on $(x > 0, y = 0)$, and both functions satisfy equation (1.2). Here $\Delta = D_r/D_p$, and $K = k_r/k_p$ is the equilibrium constant.

The Laplace transform [17] $L\{u(x, y)\} \equiv U(p, y)$ applied to the system (2.5) and (1.2) yields the general solution

$$U_r = \alpha_r Ai(p^{1/3}y) \tag{2.7a}$$

$$U_p = \alpha_p Ai(\Delta^{1/3}p^{1/3}y). \tag{2.7b}$$

Equations (2.6) then serve to determine the coefficients

$$\begin{aligned}\alpha_r &= \Delta^{-2/3}(\nu_r/\nu_p)\alpha_p\\ &= \frac{\nu_r}{p[(\nu_r + \nu_p\Delta^{2/3}/K)Ai(0) - p^{1/3}Ai'(0)]}.\end{aligned} \tag{2.8}$$

Using the known values ([18], p. 446) $Ai(0) = 1/3^{2/3}\Gamma(2/3)$, $Ai'(0) = -1/3^{1/3}\Gamma(1/3)$, and the reflection formula for the gamma function ([18], p. 256) in the form $\Gamma(1/3)/\Gamma(2/3) = (3^{1/2}/2\pi)[\Gamma(1/3)]^2$, one finally gets

$$U_r(p, y) = \frac{3^{1/3}\Gamma(1/3)\nu_r Ai(p^{1/3}y)}{p(p^{1/3} + \beta)}, \tag{2.9a}$$

$$U_p(p, y) = \frac{3^{1/3}\Gamma(1/3)\nu_p\Delta^{2/3}Ai(\Delta^{1/3}p^{1/3}y)}{p(p^{1/3} + \beta)}, \tag{2.9b}$$

with

$$\beta = (3^{1/6}/2\pi)[\Gamma(1/3)]^2(\nu_r + \nu_p\Delta^{2/3}/K). \quad (2.9c)^*$$

Both transforms (2.9a,b) are of the type $Ai(\alpha p^{1/3})/p(p^{1/3} + \beta)$ where $\alpha > 0$. The inverse transform of the denominator is proportional to the surface values, and its convolution with†

$$\begin{aligned} L^{-1}&[Ai(\alpha p^{1/3})]\\ &= (\alpha/2\pi 3^{5/6})x^{-4/3}\exp(-\alpha^3/9x),\end{aligned} \tag{2.10}$$

gives the full concentration distributions. Accordingly, first examine the surface values

$$\begin{aligned} u_r(x, 0) &= \Delta^{-2/3}(\nu_r/\nu_p)u_p(x, 0)\\ &= \frac{\nu_r}{1 + \Delta^{2/3}/K} f(\xi).\end{aligned} \tag{2.11}$$

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*The value of the numerical coefficient is approximately 1.37172_1.

†The inverse transform follows from the relation between the Airy function and a modified Bessel function, whose inverse is tabulated ([19], p. 283).

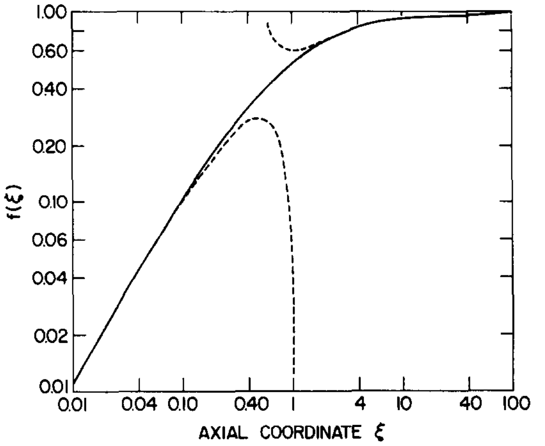

FIG. 1. The function f describes surface values of the concentrations and the reaction rate as a function of the normalized axial coordinate ξ. The dashed lines correspond to the asymptotic expressions [B.4b] and [B.5b] for $n = 3$, and in which only two and three terms of the series are summed, respectively.

where

$$f(x) \equiv f_3(x) = 1 - \frac{3^{3/2}}{2\pi} \int_0^\infty \mathrm{d}t \frac{\exp(-\beta^3 x t^3)}{t^2+t+1}, \tag{2.12}$$

as evaluated in Appendix B equation (B.2), is a universal function of $\beta x^{1/3} \equiv \xi$. It is useful to exhibit this variable in the following forms.

$$\begin{aligned}\xi &= (3^{1/6}/2\pi)[\Gamma(1/3)]^2(D_r/6B\bar{v})^{1/3} \\ &\quad \times (Bk_r/D_r)(v_r + v_p\Delta^{2/3}/K)(X/B)^{1/3} \\ &= (3^{1/6}/2\pi)[\Gamma(1/3)]^2(\bar{D}/6B\bar{v})^{1/3}(X/B)^{1/3}.\end{aligned} \tag{2.13}$$

The factor $(\bar{D}/6B\bar{v})$ is inversely proportional to the Péclet number referred to the generalized mean ([18], p. 10) diffusivity $\bar{D}^{1/3} = D_r^{1/3} v_r Da_r + D_p^{1/3} v_p Da_p$, in which the individual diffusivities are weighted by the stoichiometric coefficients and Damköhler numbers $Da_a = Bk_a/D_a$, $(a = r, p)$. The function $f(\xi)$ is displayed in Fig. 1.

Next, using equations (2.9–10) and (2.12) one calculates the full concentration distributions as the convolutions

$$\begin{aligned}u_r(x, y) &= \frac{v_r 3^{1/3}\Gamma(1/3)y}{2\pi 3^{5/6}\beta} \\ &\quad \times \int_0^x \mathrm{d}x' x'^{-4/3} \exp(-y^3/9x') f(x-x') \\ &= v_r(v_r + v_p\Delta^{2/3}/K)^{-1} g(x, y),\end{aligned} \tag{2.14a}$$

$$u_p(x, y) = v_p\Delta^{2/3}(v_r + v_p\Delta^{2/3}/K)^{-1} g(x, \Delta^{1/3}y), \tag{2.14b}$$

where

$$\begin{aligned}g(x, y) &= \frac{1}{\Gamma(4/3)} \int_{y/(9x)^{1/3}}^\infty \mathrm{d}s \exp(-s^3) \\ &\quad \times \left\{1 - \frac{3^{3/2}}{2\pi} \int_0^\infty \mathrm{d}t \frac{\exp[-\beta^3 x t^3(1 - y^3/9xs^3)]}{t^2+t+1}\right\}.\end{aligned} \tag{2.15}$$

It is easy to verify that $g(x, 0) = f(x)$, as should be the case. Again one sees that $g(x, y)$ is a universal function of the two variables ξ (cf. 2.13) and $\eta = \alpha/(9x)^{1/3}$, where $\alpha = y$ for the reactant (cf. 2.14a) and $\alpha = \Delta^{1/3}y$ for the product (cf. 2.14b). With dimensional quantities these "similarity" variables read

$$\eta_a = (2\bar{v}B/3D_a)^{1/3}(Y/B)(X/B)^{-1/3}, \qquad (a = r, p) \tag{2.16}$$

and coincide with the expressions used for the simple cases cited in the Introduction. The function $g(\xi, \eta)$ is displayed in Figs. 2 and 3.

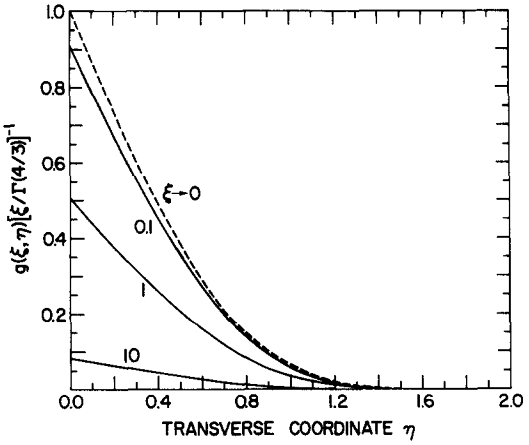

FIG. 2. The function g describes the full concentration distributions as a function of the normalized transverse coordinate η. For small values of ξ the distributions tend towards those [dashed, cf. equation (3.8)] characteristic of constant surface flux.

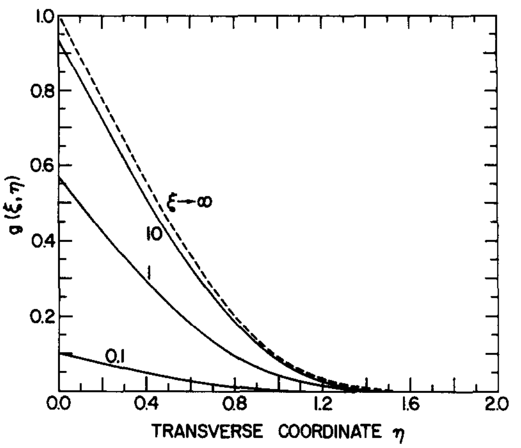

FIG. 3. Same as Fig. 2. For large values of ξ the distributions tend towards those [dashed, cf. equation (3.9)] characteristic of constant surface concentration.

3. DISCUSSION

The dimensional form of the concentration distributions is

$$C_r(X, Y) = C_0[1 - v_r(v_r + v_p\Delta^{2/3}/K)^{-1} g(\xi, \eta_r)], \tag{3.1a}$$

$$C_p(X, Y) = C_0 v_p\Delta^{2/3}(v_r + v_p\Delta^{2/3}/K)^{-1} g(\xi, \eta_p), \tag{3.1b}$$

in which [cf. equations (2.15) and (2.12)]

$$g(\xi,\eta) = [\Gamma(4/3)]^{-1} \times \int_{\eta}^{\infty} \mathrm{d}s \exp(-s^3) f[\xi(1-\eta^3/s^3)^{1/3}]. \quad (3.2)$$

Here f is considered a function of ξ. The surface concentrations are given formally by equations (3.1) in which f replaces g. Two related quantities of interest are the reaction rate (2.4b)

$$\begin{aligned} r(\xi) &= k_r[C_r(X,0) - K^{-1}C_p(X,0)] \\ &= k_r C_0[1-f(\xi)], \end{aligned} \quad (3.3)$$

and the product to reactant ratio

$$\frac{C_p(X,0)}{C_r(X,0)} = Kf(\xi)\left\{1 + \frac{\nu_r K}{\nu_p \Delta^{2/3}}[1-f(\xi)]\right\}^{-1}. \quad (3.4)$$

Equation (3.3) displays the local reaction rate in a remarkably simple form, and equation (3.4) allows a discussion of "closeness to equilibrium". To this end first observe that for small x, equation (B.4b) of Appendix B reads in this case

$$f(\xi) = \xi/\Gamma(4/3) + O(\xi^2), \quad (3.5)$$

and formula (B.5b) yields

$$f(\xi) \sim 1 - \xi^{-1}/\Gamma(2/3) + O(\xi^{-2}) \quad (3.6)$$

for large x. "Large" and "small" refer to the magnitude of the axial coordinate $\xi = \beta x^{1/3}$ given by equation (2.13) in dimensional form. For example, all other parameters being equal, ξ is smallest when $K \gg 1$, i.e. when the reaction (2.1) is driven to the right, or when $\Delta \ll 1$, i.e. the product diffuses faster than the reactant. Thus for $\xi \ll 1$, the reaction rate (3.3) approximates its maximum admissible value $k_r C_0$. Likewise the composition ratio (3.4) is of order $O(\xi)$ since the product has barely begun to form. On the other hand, for $\xi \gg 1$ the reaction rate approaches zero as $k_r C_0/\xi\Gamma(2/3)$, and the composition ratio tends towards $K[1+O(\xi^{-1})]$. Note that, in this case, C_p/C_r can be close to its equilibrium value K, yet the reaction rate can be quite large due to the possibly large multiplicative factor $k_r C_0$.

It is instructive to examine the full concentration distributions in these asymptotic limits. The retarded argument of f in equation (3.2) never exceeds ξ, so that expansion (3.5) applies when $\xi \ll 1$, and

$$g(\xi,\eta) \simeq [\Gamma(4/3)]^{-2} \times \int_{\eta}^{\infty} \mathrm{d}s \exp(-s^3)\xi(1-\eta^3/s^3)^{1/3}. \quad (3.7)$$

The substitution $t^{1/3} = \eta/s$ takes the integral into the form (C.1) of Appendix C. It is evaluated there and one gets

$$g(\xi,\eta) \simeq [\xi/\Gamma(4/3)][\exp(-\eta^3) - \eta\Gamma(2/3,\eta^3)]. \quad (3.8)$$

One recognizes expression (1.4) for constant surface flux since it is easy to see that $\xi/\Gamma(4/3) = (\nu_r + \nu_p\Delta^{2/3}/K)(9x)^{1/3}/\Gamma(2/3)$. On the other hand, for $\xi \gg 1$ the argument of f in equation (3.2) is large for most values of s so that expansion (3.6) holds. One then gets

$$\begin{aligned} g(\xi,\eta) &\sim [\Gamma(4/3)]^{-1} \int_{\eta}^{\infty} \mathrm{d}s \exp(-s^3) \\ &= \Gamma(1/3,\eta^3)/\Gamma(1/3), \end{aligned} \quad (3.9)$$

an expression identical to equation (1.3), i.e. the case of constant surface concentration. The dashed lines in Figs. 2 and 3 represent the asymptotic cases (3.8) and (3.9), respectively.

Finally, one recognizes that the intermediate region $\xi \simeq 1$ does not admit a similarity solution because surface conditions are variable [8–9]. Thus the solution for an irreversible reaction ($K \gg 1$) proposed by Bowen *et al.* {[20], equation (2.44)} is wrong. Of course, Lévêque's approximation breaks down in ducts when $Y \gtrsim \frac{1}{2}B$. Since the concentration distributions have a boundary-layer character, this puts an upper limit to the possible values of X, in accordance with equation (2.16). More complicated reaction schemes, sequential or parallel, can be analyzed in the same manner. These results will be published shortly.

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APPENDIX A

The notation of Bird *et al.* ([4] chapter 16) is used throughout. Consider the steady state flow of N species $A_a(a = 1 \ldots N)$ that do not react homogeneously. The following conservation laws hold in a region D

$$\nabla \cdot \mathbf{n}_a = 0, \tag{A.1}$$

and $\nabla \cdot \rho \mathbf{v} = 0$ by summation over species. Assume that these species react heterogeneously on a part Σ of ∂D according to the R reactions $(i = 1 \ldots R)$

$$\sum_a \nu_{ai} A_a \rightleftarrows 0. \tag{A.2a}$$

Here the stoichiometric coefficients are positive for products and negative for reactants ([21] p. 10). Each reaction must satisfy mass conservation

$$\sum_a \nu_{ai} M_a = 0, \tag{A.2b}$$

where M_a is the molar mass. If the reactive surface Σ is approximately immobile, and if the species A_a neither accumulate on it nor diffuse into the solid, then the following balance law must hold on Σ ([21] p. 19).

$$-\mathbf{n} \cdot \mathbf{n}_a + \sum_i \nu_{ai} M_a r_i = 0. \tag{A.3}$$

Here $\mathbf{n}$ is the unit normal to Σ pointing into the fluid, and r_i is the (molar) reaction rate of the ith reaction. Summing (A.3) over species and using (A.2b) gives $\mathbf{n} \cdot \rho \mathbf{v} = 0$. Since the tangential component of $\mathbf{v}$ must vanish, it follows that the boundary conditions on $\mathbf{v}$ for a mixture obeying (A.3) are the same as for a pure fluid, i.e., $\mathbf{v} \equiv 0$ on Σ. Inserting the diffusion fluxes $\mathbf{j}_a = \mathbf{n}_a - \rho_a \mathbf{v}$ into (A.1) and (A.3) gives then

$$\nabla \cdot \rho_a \mathbf{v} + \nabla \cdot \mathbf{j}_a = 0, \text{ in } D, \tag{A.4a}$$

$$-\mathbf{n} \cdot \mathbf{j}_a + \sum_i \nu_{ai} M_a r_i = 0, \text{ on } \Sigma. \tag{A.4b}$$

Now *if* the fluid is incompressible and *if* the diffusion currents can be expressed in the form of Fick's law $\mathbf{j}_a = -D_a \nabla \rho_a$, then equations (A.4) reduce to

$$\mathbf{v} \cdot \nabla \rho_a = D_a \nabla^2 \rho_a, \text{ in } D, \tag{A.5a}$$

$$D_a \partial \rho_a / \partial n + \sum_i \nu_{ai} M_a r_i = 0, \text{ on } \Sigma. \tag{A.5b}$$

Reaction rates are usually expressed as polynomials in the molar concentrations $C_a = \rho_a / M_a$; thus equations (A.5) assume the convenient final form

$$\mathbf{v} \cdot \nabla C_a = D_a \nabla^2 C_a, \text{ in } D, \tag{A.6a}$$

$$D_a \partial C_a / \partial n + \sum_i \nu_{ai} r_i = 0, \text{ on } \Sigma. \tag{A.6b}$$

Here the barycentric velocity $\mathbf{v}$ enters into the convection term of (A.6a) rather than the mole average velocity; the latter is not immediately obtainable from the Navier–Stokes equations.

APPENDIX B

Consider the inverse Laplace transform

$$f_n(x) = \frac{1}{2i\pi} \int_{c-i\infty}^{c+i\infty} \frac{\beta \exp(px)}{p(p^{1/n} + \beta)} \, \mathrm{d}p, \tag{B.1}$$

where $\beta > 0$ and n is real> 1. The integrand has a branch point at the origin and poles of phase $n\pi(2k - 1)$, where k is an integer. These latter lie *outside* the principal determination $[-\pi, \pi]$ for $n > 1$, and therefore do not contribute to the inverse transform. It is then easy to evaluate the integral over Bromwich's contour to give

$$f_n(x) = 1 - \frac{n \sin \pi/n}{\pi} \int_0^\infty \frac{\exp(-\beta^n x t^n)}{t^2 + 2t \cos(\pi/n) + 1} \, \mathrm{d}t. \tag{B.2}$$

Several remarks are in order. First, the variable x only occurs in the combination $\beta^n x$. Second, the integrand has no real poles, so the integral converges and can always be evaluated numerically. Third, the formula reduces to $f_2 = 1 - \exp(\beta^2 x) \operatorname{erfc}(\beta x^{1/2})$ as it should for $n = 2$ ([19] p. 233). Fourth, the limits $f_n(0) = 0$ and $f_n(\infty) = 1$ result from (B.2) and correspond to the appropriate limits of the Laplace Transform

$$F_n(p) = L[f_n(x)] = \frac{\beta}{p(p^{1/n} + \beta)}. \tag{B.3}$$

The asymptotic behavior is easily obtained from (B.3). For small values of x, it suffices to expand around $p \to \infty$ ([17] chapter 33):

$$F_n(p) = \sum_{k=0}^{\infty} (-1)^k \beta^{k+1} p^{-1-(k+1)/n}. \tag{B.4a}$$

Term by term inversion yields

$$f_n(x) = \sum_{k=0}^{\infty} (-1)^k (\beta x^{1/n})^{k+1} / \Gamma[1 + (k+1)/n], \tag{B.4b}$$

a series which converges for all values of $\beta x^{1/n}$. On the other hand, the behavior for large x arises from an expansion around $p = 0$ ([17] chapter 37)

$$F_n(p) = \sum_{k=0}^{\infty} (-1)^k \beta^{-k} p^{-1+k/n}, \tag{B.5a}$$

hence the asymptotic series

$$f_n(x) \sim \sum_{k=0}^{\infty} (-1)^k (\beta x^{1/n})^{-k} / \Gamma(1 - k/n). \tag{B.5b}$$

If n is an integer, then all terms of (B.5b) vanish for which k is a multiple of n.

APPENDIX C

Evaluate the integral

$$J(z) = \int_0^1 t^{-4/3} (1-t)^{1/3} \exp(-z/t) \, \mathrm{d}t. \tag{C.1}$$

The change of variables $s = (1-t)^{1/3} t^{-1/3}$ reduces it to

$$J(z) = 3 \int_0^\infty \frac{s^3 \exp[-z(1+s^3)]}{1+s^3} \, \mathrm{d}s, \tag{C.2}$$

and its derivative with respect to z is then

$$J'(z) = -\Gamma(4/3) z^{-4/3} \exp(-z). \tag{C.3}$$

Since $J(\infty) = 0$ one gets immediately

$$J(z) = \Gamma(4/3) \Gamma(-1/3, z). \tag{C.4}$$

A recursion formula

$$\Gamma(n+1, z) = n\Gamma(n, z) + z^n \mathrm{e}^{-z} \tag{C.5}$$

for the incomplete gamma function is obtained by parts just as in the case of the ordinary gamma function. Thus (C.4) can be written

$$J(z) = 3\Gamma(4/3)[z^{-1/3} \mathrm{e}^{-z} - \Gamma(2/3, z)]. \tag{C.6}$$

TRANSPORT MASSIQUE ET REACTION EN SURFACE DANS L'APPROXIMATION DE LEVEQUE

Résumé—Cet article concerne le problème général du transport de plusieurs composants et de la réaction hétérogène dans l'approximation de Lévèque. Le problème admet toujours une solution analytique quand les réactions pariétales sont du premier ordre en concentration. On trouve ici des expressions explicites pour des distributions de concentrations dans le cas d'un saut unique reversible à la surface, mais la procédure s'applique également à des séquences arbitrairement complexes de tels sauts. Le comportement de la solution dépend principalement de la valeur d'un paramètre axial adimensionnel. En plus de la distance à l'axe et de la vitesse moyenne, il gouverne les coefficients de diffusion, les constantes de vitesse de réaction et les coefficients stoichiométriques. Pour des petites valeurs de ce paramètre, la solution est comme si la surface était une source à flux constant ; pour des grandes valeurs, comme si la surface était à concentration constante. Ainsi la solution montre un changement continu du problème de Neumann à celui de Dirichlet.

STOFFÜBERGANG UND OBERFLÄCHEN-REAKTIONEN IN LÉVÊQUE'S NÄHERUNG

Zusammenfassung—Diese Arbeit behandelt das allgemeine Problem des Stoffübergangs mit heterogener chemischer Reaktion von mehreren Komponenten mit Lévêque's Näherung. Das Problem läßt immer eine analytische Lösung zu, wenn die Oberflächenreaktionen von erster Ordnung in den Konzentrationen sind. Hier wurden explizite Ausdrücke für die Konzentrationsverteilungen gefunden für den Fall eines einzigen reversiblen Oberflächenschnittes. Der analytische Lösungsablauf gilt jedoch genauso für willkürlich komplexe Folgen solcher Schritte. Die Eigenschaft der Lösung hängt hauptsächlich vom Wert eines dimensionslosen Axialparameters ab. Abgesehen von dem axialen Abstand und der mittleren Geschwindigkeit, enthält die Lösung alle Diffusionskoeffizienten, Reaktionskonstanten und stöchiometrischen Koeffizienten. Für kleine Werte des Axialparameters verhält sich die Lösung, als wäre die Oberfläche eine Quelle konstanten Stromes für große Werte, als bestände in der Oberfläche konstante Konzentration. Die Lösung weist also einen kontinuierlichen Wechsel von einem Neumann-Problem zu einem Dirichlet-Problem auf.

ПЕРЕНОС МАССЫ И ГЕТЕРОГЕННЫЕ РЕАКЦИИ В ПРИБЛИЖЕНИИ ЛЕВЕКА

Аннотация — Статья посвящена общей проблеме переноса в многокомпонентной системе с гетерогенной реакцией в приближении Левека. Данная задача всегда допускает аналитическое решение, если реакции на поверхности являются реакциями первого порядка по концентрациям. В явном виде получены выражения для распределения концентрации для одной обратимой стадии на поверхности, но методика равноприменима и к произвольной сложной последовательности таких стадий. Поведение решения зависит от величины безразмерного осевого параметра. Помимо аксиального расстояния и средней скорости он включает все коэффициенты диффузии, константы скоростей реакций и стехиометрические коэффициенты. При малых значениях этого параметра решение ведет себя таким образом, как если бы поверхность являлась источником постоянного массового потока, а при больших значениях — как если на поверхности имела место постоянная концентрация. Таким образом, решение демонстрирует непрерывный переход от задачи Неймана к задаче Дирихле.