01 May 1994

Forced Convective Diffusion and Interphase Heat and Mass Transfer: Computations of Radial Functions, Temperature and Concentration Fields, and Presentation of Local and Average Nusselt and Sherwood Numbers

Stephen Myers

X B. Reed Jr.

N. Y. Shah

A. E. Penn

FORCED CONVECTIVE DIFFUSION AND INTERPHASE HEAT AND MASS TRANSFER: COMPUTATIONS OF RADIAL FUNCTIONS, TEMPERATURE AND CONCENTRATION FIELDS, AND PRESENTATION OF LOCAL AND AVERAGE NUSSELT AND SHERWOOD NUMBERS

Opportunities for Undergraduate Research Experience Report

X B Reed, Jr.[1], N. Y. Shah[2], A. E. Penn[3], S. Myers[3,4]
Chemical Engineering Department
University of Missouri-Rolla
Rolla, MO 65401

[1]Professor, to whom correspondence should be addressed
[2]Doctoral student
[3]Senior student
[4]OURE student

Abstract

Theoretical calculations have been carried out for forced convective transport for uniform streaming and uniaxial and biaxial extensional axisymmetric flows past single spheres. Homogeneous and heterogeneous chemical reactions, both of first and of second order have also been or are presently being treated. Orthogonality and other properties of Legendre functions have been used, together with introduction of an eigenfunction expansion, to reduce the mathematical description from a partial differential equation with variable coefficients, which is nonlinear for homogeneous second order chemical reactions, to a system of coupled ordinary differential equations for the radial modes. The numerical solutions of the latter have been obtained using the robust, adaptive grid algorithm of Pereyra and Lentini. Plots of the radial functions for given Peclet and Damköhler numbers give insight into the role and interaction of L and of r_∞ (the number of terms necessary for convergence of the expansion and the finite radius at which the boundary conditions at infinity are imposed). From the radial modes, local and average Nusselt and Sherwood numbers, as well as the temperature and concentration fields, can be obtained. Plots of radial function families provide new insights that complement physicochemical understanding gained from isocontour plots of the temperature and concentration fields. Plots of local interphase transfer coefficients reflect the behavior of the flux field over the sphere surface and show how the average coefficients arise.

1. INTRODUCTION

This preliminary report covers a limited set of computations on several novel, potentially significant physicochemical processes while pointing out some computational and presentational limitations.

All but one of the basic elements of the physicochemical processes are hardly novel. We consider steady forced convective diffusion of temperature or of a reactant undergoing a homogeneous or a heterogeneous chemical reaction. The reaction may be first or second order. The geometry is spherical. The domain is the semi-infinite one external to a solid sphere. The convecting velocity field, moreover, is axisymmetric, solenoidal, and satisfies the steady, linearized Navier-Stokes equation and the no-slip condition at the surface of the sphere.

What is novel about this forced convective diffusion-reaction problem is the remaining element, the boundary condition imposed on the velocity field far from the sphere. It is not always the uniform streaming flow of Stokes (1842; see also Lamb, 1932; Happel and Brenner, 1965; Leal, 1992) but includes the low Reynolds number, axisymmetric extensional motion important in the rheology and flow of dispersions and elsewhere (see, e.g., Leal, 1992). This lends an importance to the problem beyond that of being an $Re \ll 1$ convective diffusion-reaction class of problems that are axisymmetric yet not the simplest problem of this class. The incorporation of axisymmetric extensional flows into the class of forced convection problems nonetheless enlarges

the class considerably, taking it in fundamentally new directions having significantly different ranges of applications.

The biaxial extensional flow approaches the sphere from the poles at $z = \pm \infty$ and departs in the equatorial latitudes, doing so radially symmetrically in the equatorial plane. The uniaxial extensional flow has the same streamlines as the biaxial one; the direction of the flow is, however, reversed along the same streamlines, approaching the sphere at the equatorial latitudes and departing in the opposite directions, from the poles of the sphere.

For a more complicated yet axisymmetric flow, what are some of the issues which might arise? For a given Peclet number, will the average Sherwood numbers have about the same values for the biaxial and uniaxial flows? Or will the magnitudes of the locally enhanced contributions in the stagnation region(s) of one flow, which correspond roughly to locally reduced contributions in the separation region(s) of the other flow, be significantly different? Or, again phrased oversimplistically, will they be comparable? Or will the area of enhanced (reduced) equatorial interphase mass transfer be greater or less than the area of reduced (enhanced) transfer at the poles for the uniaxial vs. the biaxial flow, whether or not the magnitudes of the fluxes are comparable in those regions.

The study of extensional flows for the forced convective diffusion problem without reaction was initiated by Shah and Reed (1994). For the convective diffusion problem, the passive

additive could be either temperature or concentration (Shah and Reed, 1994). The passive additive field $\theta(r,\theta)$ was obtained as a function of the Peclect number Pe, as were the average and local Nusselt numbers for heat or mass transfer, Nu and $Nu(\theta)$; in chemical engineering, the latter are more commonly called the average and local Sherwood numbers, Sh and $Sh(\theta)$, respectively, if the passive additive is the concentration field $c(r,\theta)$. In the present study, samples of $c(r,\theta)$, Sh, and $Sh(\theta)$ are reported as functions of Pe and of Da_{II} (or Da_{I}), the second (or first) Damköhler group, which is the chemical rate constant made dimensionless with the diffusive (or convective) time scale. Answers to such questions as were posed above will be answered in the course of this research program.

The effects are already subtle for the convective diffusion problem, and inferences about magnitudes of local fluxes and resultant averages over the sphere surface for convective diffusion-reaction conceivably could follow intuitively from the results for convective diffusion in the absence of homogeneous reactions. Conversely, the qualitative effects of the reaction may not be so trivially inferred from convective diffusion alone. Overall mass transfer coefficients for a given flow, whether biaxial or uniaxial, would be enhanced by reaction and increasingly so by a faster reaction. Yet local effects for biaxial flows could conceivably be affected differently by reaction than those for uniaxial flows.

In Section 2, we sketch the theoretical analysis leading to

the system of ordinary differential equations for the radial modes for axisymmetric extensional flows convecting a diffusing chemical species that may undergo a first order (or no) reaction; from the radial modes, the full concentration field can be obtained, as can the local and average Sherwood numbers. In Appendix II, the analogous analysis and results are outlined for a uniform streaming flow and for a second order reaction.

In Appendix I, a variety of results are presented. The computational limitations that can arise and the manner in which different limitations manifest themselves are discussed, both there and in Section 3, in which the perspective is broader than the litany of specific cases discussed in Appendix I. Finally, in Section 4 we stress the importance of the research program that is under weigh.

2. PHYSICOCHEMICAL BACKGROUND AND THEORETICAL FORMULATION

If by any physicochemical process the concentration of a reactant is maintained constant at the surface of a solid sphere, and if the reactant is yielded up to a surrounding fluid where it undergoes an irreversible first order homogeneous chemical reaction, as well as forced convective diffusion, then the reactant concentration satisfies the dimensionless partial differential equation

$$Pe\ v\cdot\nabla c = \nabla^2 c - Da_{II} c, \quad \text{for } 1<r<\infty, \tag{1}$$

subject to the boundary conditions

$$c = 1, \quad \text{at} \quad r = 1, \tag{2}$$

$$c \to 0, \quad \text{as} \quad r \to \infty. \tag{3}$$

The axisymmetric extensional velocity field convecting the reactant satisfies the adherence condition at r =1. The boundary condition at infinity is, in circular cylindrical coordinates,

$$(v_\rho, v_z) \to \pm (\rho, -2z), \tag{4}$$

with the plus sign indicating the biaxial flow, the negative sign the uniaxial flow. If further the velocity field is solenoidal and satisfies the linearized Navier-Stokes equation, then it may be expressed in terms of the spherical geometry as

$$v_r = \pm F(r) \ (1-3\cos^2\theta), \tag{5}$$

$$v_\theta = \pm G(r) \ (3\sin\theta\cos\theta), \tag{6}$$

$$v_\phi = 0, \tag{7}$$

in which

$$F(r) = r - \left(\frac{5}{2}\right) \frac{1}{r^2} + \left(\frac{3}{2}\right) \frac{1}{r^4},$$

$$G(r) = 1 - \frac{1}{r^5},$$

and in which +/- refers throughout to biaxial/uniaxial extensional flow.

The radius a of the sphere has been used as the characteristic length scale, and the strain rate $|E|$ of the flow far from the sphere has been employed to form the characteristic

velocity $|E|a$. The Peclet and the first and second Damköhler numbers are defined in the usual manner for a first order reaction, except that in an unusual though not singular manner the characteristic length is taken to be the radius a throughout (i.e, in the Sherwood numbers Sh and Sh(θ), as well):

$$Pe = |E|a^2/D,$$

$$Da_I = k/|E|,$$

$$Da_{II} = k\, a^2/D = Da_I Pe.$$

The local value of the Sherwood number is the local dimensionless flux at the surface,

$$Sh(\theta) = - \partial c(r,\theta)/\partial r \qquad \text{at} \quad r = 1. \tag{8}$$

The average or overall mass flux Sh is then given by

$$Sh = \tfrac{1}{2} \int_{\theta=0}^{\pi} Sh(\theta) \sin\theta \, d\theta \tag{9}$$

The dependent variables $c(r,\theta)$, $Sh(\theta)$, and Sh, are to be determined in terms of the parameters Pe and Da_{II} (or Da_I). The second Damköhler group has the advantage that it may be expressed solely in terms of physicochemical properties (the chemical rate constant and the molecular diffusivity) and the radius. All convective effects then reside in the Peclet number.

The eigenfunction expansion

$$c(r,\theta) = \sum_{\ell=0}^{\infty} c_\ell(r) P_\ell(\cos\theta), \tag{10}$$

in which the $P_\ell(\cos\theta)$ are Legendre functions and the radial functions $c_\ell(r)$ are the unknowns to be determined, is introduced

into Equation (1) and boundary conditions (2) and (3), along with (5), (6), and (7). Several properties of the Legendre functions are then used (Shah and Reed, 1994), the last of which is their orthogonality (e.g., Abramowitz and Stegun, 1964). The result of the calculations is an infinite system of coupled ordinary differential equations for the radial functions,

$$\pm \mathrm{Pe} \left\{ F(r) \frac{dc_k}{dr} - 3\left[\frac{(k+1)^2}{(2k+3)} + \frac{k^2}{(2k-1)}\right]\left[F(r)\frac{dc_k}{dr} - kG(r)c_k\right] \right.$$

$$- 3\left[\frac{k(k-1)}{(2k-1)(2k-3)}\right]\left[F(r)\frac{dc_{k-2}}{dr} - (k-2)G(r)c_{k-2}\right]$$

$$\left. - 3\left[\frac{(k+1)(k+2)}{(2k+3)(2k+5)}\right]\left[F(r)\frac{dc_{k+2}}{dr} + (k+3)G(r)c_{k+2}\right]\right\}$$

$$= \mathcal{L}_k c_k - \mathrm{Da}_{II} c_k,$$

in which,

$$\mathcal{L}_k = \frac{1}{r^2}\frac{d}{dr}\left(r^2\frac{d}{dr}\right) - \frac{k(k+1)}{r^2},$$

subject to the boundary conditions

$$c_o\ (r = 1) = 1, \qquad (12a)$$

$$c_\ell\ (r = 1) = 0,\ \ell \geq 1, \qquad (12b)$$

$$c_\ell\ (r \to \infty) \to 0,\ \ell \geq 0. \qquad (13)$$

In these equations, for $\ell<0$, $c_\ell \equiv 0$.

The velocity field plays the role of a pair of variable coefficients (Equations (5), (6)) in the forced convective

diffusion partial differential equation for $c(r,\theta)$, Equation (1). If there were no angular velocity component and if the radial component depended solely upon the radius, then the radial modes $c_\ell(r)$ would not be coupled to one another. That is, if $\underline{v} = v_r(r)\underline{i}_r$, then the system, Equations (11), would not be coupled. Neither is true, so that $v_\theta = v_\theta(r,\theta) \neq 0$ and $v_r = v_r(r,\theta)$ for different reasons necessitate no small amount of theoretical calculations in order to arrive at Equations (11), which become the corresponding equations of Shah and Reed (1994) upon setting $Da_{II} = 0$. The treatment of second order reactions for uniform streaming flow and the reduction to the corresponding system of ordinary differential equations is presented in Appendix II.

Numerical methods would be required in the solution of Equations (11) because of the variable coefficients in r, even if they were not coupled. Equations (11), (12), (13) constitute the boundary value problem which we solve using the adaptive grid method of Pereyra and Lentini (Pereyra, 1967, 1968, 1975, 1978; and Lentini and Pereyra, 1974, 1978).

3. COMPUTATIONAL FEATURES

To implement the algorithm, the series must be terminated at a finite value L (i.e., $c_\ell(r) = 0$, $\ell > L$), and the infinite domain must be made finite by selecting a finite radius r_∞ (denoted by R on the figures) at which the boundary conditions (13) are imposed. L must be large enough that the series (10) has

converged to the desired accuracy. Unfortunately, L cannot be known *a priori* because it must be specified before the $c_\ell(r)$ can be computed. The optimal selection of L depends upon Pe, Da_{II}, r_∞, and the computer available for the computations. Similarly, the optimal selection of r_∞ depends upon Pe and Da_{II} for a given computer.

Generally speaking, for a given computer, as Pe and Da_{II} increase, L must be increased and r_∞ decreased. The tolerance and the initial and the maximum number of mesh points must also be specified, and they play crucial roles which are both subtle and interrelated, not only with one another, but with the parameters of the problem Pe and Da_{II} and with the other major players of the algorithm, L and r_∞.

In earlier research on the forced convective diffusion of a passive additive by a uniform flow (Spears and Reed, 1991; Reed, Spears, and Shah 1994), and for forced convective diffusion by the same flow of a reactant undergoing a first order reaction (Reed, 1994) or a second order reaction (Shah, Kleinman, and Reed, 1994), moderate to high values of Pe and of $Da_{I \text{ or } II}$ could be reached on the local IBM 4381, the exception being a relatively mild one for Pe and a much stronger one for Da_I for the second order homogeneous reactions.

In the research described above, for which the convecting velocity is an extensional one, we are limited to much smaller values of Pe and $Da_{I \text{ or } II}$ by the IBM 4381.

In this preliminary report we do not attempt to present —

indeed, we do not yet have — extensive results. Instead, we focus on some of the different kinds of difficulties that can arise for the various problems, as well as indicating some typical results (Appendix I).

On the IBM 4381, the maximum number of radial modes L+1 for which computations can be carried out is about 33. More recently, computations have been carried out on the IBM RS 6000 and HP 9000 series, for which significantly higher values of L could be computed. This has enabled us to compute more accurately with more realistic values of r_∞. In particular, for r_∞ too small, the radial functions — and thence most derivative results except occasionally the average Nusselt and Sherwood numbers — can be distorted by the relatively close proximity of r_∞ to r=1.

In addition, the radial plots for the first vintage of results are all shown as one plot to a page (with typically 6 radial functions to a plot). This made it difficult to compare different cases, and it made it difficult to get an overview of all of the radial functions for a single case. Our first improvement was to reduce the plot size while putting more plots (6) to a page; this enabled us to present c_0 - C_L, for L up to 35, on a single page. With the acquisition by UMR of the NIC (numerically intensive computers, in local terminology) IBM RS 6000 and the HP 9000 workstations, we can reach significantly higher values of L than 35. Because smaller plots cannot be read and more curves to a plot make for "too busy" a plot, rather than

attempting to display all radial plots on a single page, we have now developed the capability of sequential pages, each with 6 plots to a page.

In Appendix III, radial plots for computations on the HP 9000 for L = 70 are shown for Pe = 10 and r_∞ = 100. An r_∞ of 100 is a factor of more than two larger that what could have been run on the IBM 4381 for the same case, yet it is clear from the plots that the series has converged by L=70. For still larger values of Pe, values of L of order 100 become necessary, a capability we have for the plots. But for L very much beyond 100 we encounter memory limitations on the HP 9000. Thus, although these computations are CPU intensive, they are not inordinately so, as measured against our local capabilities. The computations are above all memory intensive. With the 128 MB of RAM available on the cluster of IBM RS6000 and HP9000 machines which are used as campus-wide, multi-user computers, we typically have difficulty with runs projected to require ~ 100 MB.

4. SIGNIFICANCE OF AND PERSPECTIVES ON THE RESEARCH

There are several features of the past, present, and future research that are noteworthy.

(i) For the practicing engineer, but no less for the scientist and engineering scientist, the overall dimensionless interphase transfer coefficients Nu and Sh are of the greatest importance and will be used in

applications, both widely and immediately.

(ii) But the local Nusselt and Sherwood numbers $Nu(\theta)$, $Sh(\theta)$ are also of direct utility, as well as being of fundamental significance, for the local properties $Nu(\theta)$ and $Sh(\theta)$ offer insight into the manner in which the macroscopic properties Nu and Sh arise.

(iii) The temperature and concentration fields $\theta(r,\theta)$ and $c(r,\theta)$ are too often considered to be only a means to an end by the practicing engineer, the end being the dependence of Nu and Sh, or perhaps $Nu(\theta)$ and $Sh(\theta)$, on Pe (and on Da_{II} for reacting systems). Yet $\theta(r,\theta)$ and $c(r,\theta)$ in turn provide insight into the nature of $Nu(\theta)$ and $Sh(\theta)$, just as they provide insight into Nu and Sh.

An understanding of the character of the passive additive field and especially its boundary layer(s) and its wake(s) enables one to develop intuition and to make engineering estimates of the manner in which and the extent to which, for instance, chemical reactions affect the concentration field and thereby enhance mass transfer. In a multiparticle system, knowledge of the passive additive boundary layers and wakes enables one to make estimates of the interparticle distance at which interphase transfer could no longer be assumed to be an additive property of the individual particles, even in the absence of meaningful hydrodynamic interactions.

(iv) Finally, in the same manner in which Fourier modes and spectra offer an alternative way of thinking about

fields, so also do radial functions provide a deeper insight into $\theta(r,\theta)$ and $c(r,\theta)$. And the number of coupled modes in a given modal differential equation that arise from different convective, diffusive, or reactive contributions has the same sorts of advantages in offering completely different perspectives that Fourier modes and modal coupling have in wave and other physical and chemical phenomena, right up to the level of complexity of pattern and chaotic behavior.

Overriding all of the above, however, are the completely different classes of flow fields and concomitant new classes of forced convective diffusion and diffusion-reaction problems attacked in this research program.

There are a number of different research directions in this general area which could be pursued. For forced convective diffusion about a solid particle, with or without chemical reaction, in addition to the convective to diffusive effect as measured by the Peclet number Pe, there is the inertial to viscous effect characterized by the Reynolds number Re. Because Pe = Re Sc or RePr, an increase in Re has a profound effect on forced convection. This is true not solely in the sense of increasing Pe parametrically, but in modifying the very nature of the velocity field; with increasing Re the velocity first manifests fore-aft asymmetry of the streamlines, then a clear momentum boundary layer-wake, and eventually a closed, recirculating wake (e.g. Batchelor, 1967). This research

direction, in which Re is increased, has been taken earlier by A. Acrivos, B. T. Chao, D. W. Moore, B. A. Finlayson, and others. The present research is predicated in the first instance on an (r,θ)-separable velocity field, and in the second on the θ-dependence being exactly expressible in terms of a finite number of Legendre functions. The PI's approach does not preclude velocity fields that do not satisfy the linearized Navier-Stokes equations. And although there are approximate solutions to the full Navier-Stokes equations for uniform streaming flow past a solid sphere for $Re > 1$, there are no such exact solutions.

The PI has elected to limit the present stage of his research program to $Re < 1$ — physically, to small particles or very viscous fluids — but to consider other flows than the uniform streaming flow, and for the case of a uniform streaming flow to incorporate reactions into the flowing medium or at the surface of the solid sphere.

The PI's overriding concern was thus not to simply extend the range of the values of the parameters for a given class of problems, but to carry the research into entirely new classes of problems, either by dent of new classes of convecting velocities or by dent of incorporating homogeneous second order reactions, rather than just bringing first order ones into the ambit. The classes of problems would also be enlarged to include consideration of heterogeneous reactions, both linear and nonlinear.