limit $a_c \to 0$, $Bi_{eq} \to -1$ satisfies the linearized equations with $\theta_f' = A_1(1-z)$ with A_1 being any arbitrary constant.

In conclusion, we have examined the onset of natural convection induced by an exothermic surface reaction with the hope of capturing the phenomena of generation of spatial thermal structures along the solid fluid interface. In so doing, we have revealed an interesting connection between the bifurcation behavior of a stirred tank reactor with an exothermic reaction and the bifurcation behavior of the classical Benard problem.

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The asymmetric Graetz problem in a radial capillary gap cell

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INTRODUCTION

THE RADIAL capillary gap cell (RCGC) was originally developed by Beck and Guthke [1]. The RCGC consists of two or more circular parallel plates with the electrolyte entering the cell through a central inlet and flowing outward in the radial direction as shown in Fig. 1. This cell finds application in electro-organic syntheses where the electrolyte has low conductivity and the electrodes must be placed close together to minimize ohmic resistance losses. The typical gap width for these cells range from 0.1 to 1 mm. The RCGCs are also used as coulometric cells for adsorption studies and coulometric metal detectors [2].

Dworak and Wendt [3] solved the convective diffusion equation for the mass transfer to the electrodes in an RCGC. A parabolic velocity profile was assumed which was true for creeping flow. Several other assumptions were made for convenient mathematical treatment which limited the utility of the work to the symmetric Graetz problem and for thin, non-interacting boundary layers. The local mass transfer coefficient was calculated using a Leveque type approximation. Burgi *et al.* [2] analyzed the mass transfer for an RCGC electrochemical detector. Nondimensionalization was used to transform the convective diffusion equation for the RCGC into that of the Graetz problem in rectangular ducts. Eigenvalues and eigenfunctions obtained by Brown [4] were then used to solve the problem of mass transfer in the electrochemical detector, with symmetric boundary conditions at the electrodes.

The objective of this work was to analyze the mass transfer in an RCGC with creeping flow for the asymmetric Graetz problem. Nondimensionalization was used to extend the solutions for the asymmetric Graetz problem in rectangular ducts developed by Edwards and Newman [5] to the RCGC. The variation of the local Sherwood number, as a function of Reynolds and Schmidt number, for various cases has been presented. The analysis was also extended to laminar flow with a non-parabolic velocity profile.

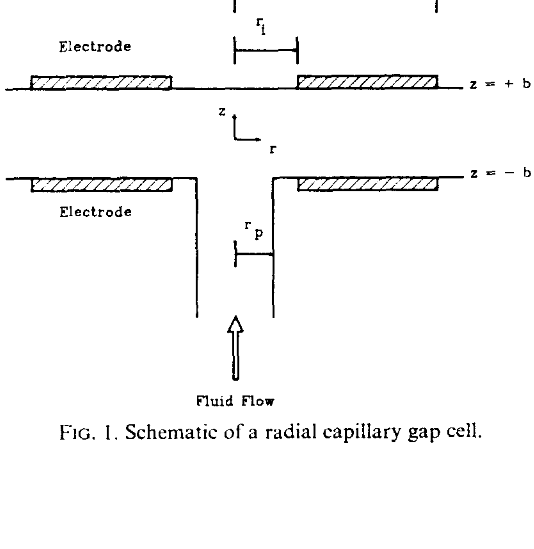

FIG. 1. Schematic of a radial capillary gap cell.

MODEL STATEMENT

The convective diffusion model for an RCGC has been discussed in detail by Dworak and Wendt [3] and Burgi *et al.* [2]. The convective diffusion equation for the RCGC, where radial diffusion is neglected, is given by

$$v_r \frac{\partial C}{\partial r} = D \frac{\partial^2 C}{\partial z^2} \tag{1}$$

where for creeping flow

$$v_r = \frac{3Q}{8\pi b r}\left(1 - \frac{z^2}{b^2}\right).$$

The model equation is nondimensionalized to the following form:

$$(1-\xi^2)\frac{\partial \Theta}{\partial \zeta} = \frac{\partial^2 \Theta}{\partial \xi^2} \tag{2}$$

NOMENCLATURE

A	coefficient in eigenfunction expansion of concentration
b	half cell width [cm]
C	concentration of species i [mol cm^{-3}]
D	diffusion coefficient of species i [cm^2 s^{-1}]
h	cell width [cm]
r	radial coordinate [cm]
Re	Reynolds number, Q/hv
Sc	Schmidt number, v/D
Sh	local Sherwood number
Pe	Peclet number, $Re\,Sc = Q/hD$
Q	flow rate [cm^3 s^{-1}]
v	velocity [cm s^{-1}].

Greek symbols

δ	boundary layer thickness [cm]
ζ	dimensionless coordinate in radial direction
Θ	dimensionless concentration
λ	eigenvalue
ν	kinematic viscosity [cm^2 s^{-1}]
ξ	dimensionless coordinate in transverse direction.

Subscripts

b	concentration in the bulk
k	summation index in eigenfunction expansion
o	concentration at the surface
r	radial component of velocity
z	transverse component of velocity.

by using the dimensionless quantities

$$\Theta = \frac{C-C_b}{C_o-C_b};\quad \xi = \frac{z}{b};\quad Re = \frac{Q}{2b\nu};\quad Sc = \frac{\nu}{D}$$

$$\zeta = \frac{4\pi D}{3Qb}(r^2-r_i^2) = \frac{8\pi}{3Re\,Sc}\frac{(r^2-r_i^2)}{h^2}.$$

The inlet boundary condition is

$$\Theta = 0 \quad \text{at} \quad \zeta = 0, \quad -1 < \xi < 1. \tag{3}$$

The other boundary conditions are dependent on the particular application of the RCGC. In this work, the asymmetric Graetz problem is considered and thus the boundary conditions at the circular plates are

$$\Theta = 0 \text{ at } \xi = 1 \quad \text{and} \quad \Theta = 1 \text{ at } \xi = -1 \quad \text{for } \zeta \geqslant 0. \tag{4a, b}$$

RESULTS AND DISCUSSION

Boundary layer thickness

Since electrolytic solutions have large Schmidt numbers, a Lighthill transformation [6–8] was used to determine the boundary layer thickness. The boundary layer thickness near the leading edge of the electrode with a step change in concentration was calculated to be

$$\frac{\delta}{b} = \left(\frac{6\pi D}{Qb}(r^2-r_i^2)\right)^{1/3}. \tag{5}$$

The concentration profile in the mass transfer boundary layer (in terms of the similarity variable, η) would be

$$\Theta = \frac{1}{\Gamma(4/3)}\int_0^\eta \exp(-x^3)\,dx \quad \text{where} \quad \eta = (1-\xi)\left(\frac{2}{9\zeta}\right)^{1/3}. \tag{6}$$

The profile has been tabulated by Abramowitz and Stegun [9]. Equations (5) and (6) provide a more accurate description of the boundary layer thickness and the concentration profile than the equations presented by Dworak and Wendt [3]. Equation (5) presents the boundary layer thickness as an explicit function of the flow rate, diffusivity and the cell dimensions as opposed to the one obtained by Dworak and Wendt [3].

Sherwood number correlations

The local Sherwood number for the asymmetric Graetz problem differs at the two walls. There is one set of correlations for the Sherwood number at the wall with a step change in concentration ($\xi = -1$ and Θ steps in value from 0 to 1 in this case) and another set of correlations for the Sherwood number at the wall without a step change in concentration ($\xi = 1$ and Θ remains equal to 0). The correlations for the Sherwood number at the wall with a step change in concentration are also applicable to the symmetric Graetz problem.

Sherwood number at wall with a step change

In the entrance region when the diffusion layers are sufficiently thin and do not interact, a Leveque series solution [10] for the local Sherwood number can be generated. Edwards and Newman [5] generated the Leveque series to solve the convective diffusion equation for the flow in a rectangular channel. Since the non-dimensionalized form of the convective-diffusion equation for the RCGC, equation (2), is the same as that of flow in the rectangular channel, the solution of Edwards and Newman can be used for the RCGC. The local Sherwood number in terms of the Reynolds and Schmidt numbers and the dimensions of the RCGC is

$$Sh = -2\left.\frac{\partial\Theta}{\partial\xi}\right|_{\xi=-1} = 0.66795\left(\frac{h}{r^2-r_i^2}\right)^{1/3} Re^{1/3}Sc^{1/3} - 0.2 - 0.1233485\left(\frac{h^2}{r^2-r_i^2}\right)^{-1/3} Re^{-1/3}Sc^{-1/3} + O[(Re\,Sc)^{-2/3}]. \tag{7}$$

Equation (7) is valid in the entrance region for $\zeta \leqslant 0.11$. The local Sherwood number predicted by Dworak and Wendt [3] was approximately equal to the first term of the series. The above correlation, obtained using a Leveque series, is more accurate than the one by Dworak and Wendt [3] and it also provides an estimate of the truncation error.

In the downstream region, $\zeta > 0.11$, the Graetz approach, of solving the convective diffusion equation by separation of variables, should be used. A three-term Graetz series is sufficient to describe the Sherwood number

$$Sh = 1 + 2\sum_{k=1}^{3}|A_k|\,e^{-\lambda_k^2\zeta}. \tag{8}$$

The eigenvalues and coefficients necessary for the use of the above equation have been computed by Edwards and Newman [5].

Sherwood number at wall without a step change

The Leveque approach cannot be used at the wall without the step change in concentration. To predict the local Sherwood number at the wall without the step change in concentration, the Graetz approach of solving the convective diffusion equation by separation of variables should be used. In the entrance region, $\zeta \leqslant 0.18$, instead of using a large number of terms from the Graetz series, an empirical correlation developed by Edwards and Newman [5] can be used

$$Sh = -2\frac{\partial\Theta}{\partial\xi}\bigg|_{\xi=1} = \exp\left(0.9594-0.6069\frac{1}{\zeta}-0.4512\,\mathrm{e}^{-0.276\zeta}\right). \tag{9}$$

In the downstream region, $\zeta > 0.18$, a three-term Graetz series solution is sufficient

$$Sh = 1-2\sum_{k=1}^{3} A_k\,\mathrm{e}^{-\lambda_k^2\zeta}. \tag{10}$$

The eigenvalues and coefficients have been computed by Edwards and Newman [5].

Laminar flow in RCGC

The parabolic velocity profile is valid only for creeping flow. Savage [11] used perturbation to obtain a series expansion for a laminar flow velocity profile. The first term in the series expansion for the radial component of the velocity profile is the same as the creeping flow solution. The subsequent terms in the series expansion come from the inertial term in the equation of motion. The laminar velocity profile, consists of a non-zero transverse component whereas the transverse component in the creeping flow velocity profile is equal to zero. When Savage's velocity profile is used in the convective diffusion equation, the solution of the differential equation by separation of variables, as in the case of Graetz problems, is not possible. A numerical solution of the convective diffusion equation is necessary.

Since the boundary layer thickness, calculated using the Lighthill transformation, used only the first-order term in the Taylor series expansion of the velocity profile, equation (5) is valid even for laminar flow (non-parabolic velocity profile). The Leveque approach, which linearizes the velocity profile in the diffusional boundary layer can be used in the entrance region to determine the Sherwood number at the wall with a step change in concentration. Thus, in the entrance region, the Sherwood number at the wall with a step change in concentration is equal to the first term in the series presented in equation (7). The other Sherwood number correlations presented here in equations (8)–(10), which were obtained by using the Graetz approach of separation of variables, would show some deviation from the actual Sherwood number because the Graetz approach is not strictly valid for the non-parabolic velocity profile in the laminar flow region.

The convective diffusion equation with Savage's velocity profile was solved numerically using finite differences. Experimental and theoretical investigations involving mass transport as well as the electrochemical aspects of the RCGC are underway in the authors' laboratory and the results of these investigations will be reported in subsequent papers.

SUMMARY

The mass transfer in an RCGC with creeping flow was analyzed for the asymmetric Graetz problem. Nondimensionalization was used to extend the solutions for the asymmetric Graetz problem in rectangular ducts to the radial capillary gap cell. The variation of the local Sherwood number, as a function of Reynolds and Schmidt number, for various cases has been presented.

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