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DEVELOPED MASS TRANSFER AT HIGH PECLET NUMBER IN CREEPING FLOW

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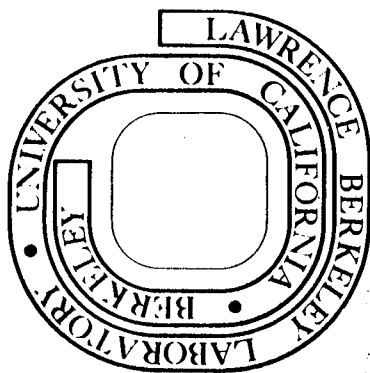
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Simulation of a Packed Bed as an Array of
Periodically Constricted Tubes

II: Developed Mass Transfer at High
Péclet Number in Creeping Flow

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Abstract

The convective diffusion equation for a limiting reactant with a large Péclet number has been solved for the asymptotic Sherwood number in a periodically constricted tube (PCT). The analysis yields a Graetz-like eigenvalue problem which is valid for any arbitrarily shaped PCT. Calculations have been made for an effective Sherwood number for creeping flow in a packed bed modeled as an array of sinusoidal PCT. The results depend upon two ratios of period length, average radius, and sinusoidal amplitude.

Scope

Recently, Payatakes et al. (1973a,b) have introduced a new model for the flow channels in a packed bed. These authors consider the bed to consist of an array of periodically constricted tubes. This is a higher order approximation to the true nature of the flow in a bed than that of the straight tube capillary model. In part I of this series we have shown how to calculate the velocity profiles in any periodic, piecewise continuous PCT. These velocity profiles can be used in solving the convective diffusion equation.

In the limit of high Péclet number, diffusion becomes negligible in the streamwise direction, and for the deep sections of the bed the Sherwood number becomes the same in successive periods. These two conditions enable us to pose a Graetz-like eigenvalue problem for the asymptotic Sherwood number of a limiting reactant. Since the asymptotic Sherwood number is always smaller than that of the entry region, a lower limit of performance of the bed can be expected a priori.

Conclusions and Significance

At a high Péclet number and in the fully developed mass-transfer region, the convective diffusion equation for a limiting reactant in a PCT can be solved by a Graetz-like eigenvalue problem. This technique is valid for laminar flow in any piecewise continuous PCT.

Results are presented in figure 5 for the Sherwood number of a packed bed modeled as an array of sinusoidal PCT. The creeping-flow

velocity profiles calculated in part I have been assumed to be applicable. The results are correlated by the two dimensionless geometry parameters r_A and A/r_A (figure 2). The bed Sherwood number exhibits different behavior in the amplitude ratio A/r_A for small and large values of average radius r_A . For long skinny tubes (small r_A) the Sherwood number increases with A/r_A , whereas for larger r_A this trend reverses itself.

Introduction

The flow channels in a randomly packed bed defy an analytical expression. To predict a priori the transfer rates across a bed, it then becomes necessary to resort to empirical correlations or, alternatively, to a microscopic model for the flow channels. The appropriate rate equations can be solved within the framework of the model to predict the performance of a bed. Payatakes et al. (1973a,b) have advanced a new model for the flow channels in a packed bed. These workers suggest that the bed can be considered as an array of periodically constricted tubes.

In part I of this series we have shown how to calculate the creeping-flow velocity profiles for any continuous PCT. The purpose of this paper is to calculate the asymptotic Sherwood number for a mass-transfer-limited reactant in a packed bed modeled as an array of sinusoidal PCT. The reactant Péclet number is assumed to be large, and the creeping-flow velocity profile is assumed applicable.

For a deep bed, the effective mass-transfer coefficient becomes independent of the velocity. This is in contrast to the entry region where the transfer rate is proportional to the approach velocity to the 1/3 power. The entry region has an effective transfer coefficient larger than that for the deeper sections of the bed. Calculating the deep-bed asymptotic Sherwood number thus gives a lower limit to the expected behavior. The horizontal line of figure 1 shows the nature of this Sherwood number. The dashed lines indicate entry-region coefficients for two different sized beds. The left and right hand sides indicate schematically regions where axial diffusion and turbulent convection, respectively, become important.

No experimental correlations for packed beds have shown the mass-transfer rate to be independent of velocity. The Wilson and Geankopolis (1966) correlation is typical. Newman and Tiedemann (1976) discuss in detail possible explanations for this behavior.

Sørensen and Stewart (1974) have solved the creeping-flow hydrodynamics and the convective diffusion equation in an array of simple cubic packed spheres for an arbitrary Péclet number. Their results are an aid in predicting the behavior through a randomly packed bed. Also of relevance is the work of Young and Finlayson (1976). They present a collocation technique to calculate the asymptotic Sherwood number for an arbitrarily shaped, constant cross section duct. Sherwood numbers for various geometries and wall boundary conditions have been compiled by Shah and London (1971).

Convective Diffusion Equation at High Péclet Numbers

The bed is modeled as an array of sinusoidal PCT (figure 2). As shown in part I, the average velocity at the length averaged radius through each tube is related to the superficial approach velocity v_s as

$$\langle v_{Ad} \rangle = \frac{v_s}{\epsilon} \left[1 + \frac{1}{2} (A/r_A)^2 \right]. \quad (1)$$

The convective diffusion equation need be solved in only one period due to the assumed homogeneity and periodicity of the structure.

The dimensionless, steady-state convective diffusion equation for a limiting reactant can be written in generalized vector notation as

$$\vec{v} \cdot \nabla C = \frac{2r_A}{Pe} \nabla^2 C. \quad (2)$$

This equation is to be solved in the far downstream region of a PCT for the asymptotic solution as $Pe \rightarrow \infty$. Solving this equation in a straight tube after neglecting diffusion in the axial direction results in the well-known Graetz solution. At high Pe it is also valid to neglect diffusion parallel to the streamwise velocity in a PCT.

It is convenient to solve equation 2 in a transformed coordinate system (ψ, ξ, θ) (figure 3). The ψ coordinate is constant along streamlines and is found directly from the stream function. The ξ direction is parallel to the streamwise velocity

at all positions and is scaled such that $\xi = 0$ at the beginning of a period and $\xi = 1$ at the end. It is defined implicitly by $(\nabla\psi) \cdot (\nabla\xi) = 0$. The angular coordinate θ has its usual meaning. In this coordinate system, diffusion will be important in the ψ direction and negligible in the ξ direction, at high Péclet numbers.

With neglect of diffusion in the ξ direction, equation 2 can be written as

$$\frac{v_{\xi}}{h_{\xi}} \frac{\partial C}{\partial \xi} = \frac{2r_A}{Pe} \frac{1}{h_{\theta} h_{\psi} h_{\xi}} \frac{\partial}{\partial \psi} \left(\frac{h_{\theta} h_{\xi}}{h_{\psi}} \frac{\partial C}{\partial \psi} \right). \quad (3)$$

Explicit forms for two of the metric factors can be determined. By inspection $h_{\theta} = r$. Since the stream function represents the amount of fluid flowing in a stream tube between a point and the axis,

$$\psi = \frac{2}{r_A} \int_0^{\psi} v_{\xi} r h_{\psi} d\psi \quad (4)$$

after appropriate normalization. It follows that the metric factor h_{ψ} is related to the streamwise velocity v_{ξ} :

$$h_{\psi} = \frac{r_A^2}{2r} \frac{1}{v_{\xi}}. \quad (5)$$

Equation 3 now becomes

$$\frac{\partial C}{\partial \xi} = \frac{8}{r_A Pe} \frac{\partial}{\partial \psi} \left((r/r_A)^2 v_{\xi} h_{\xi} \frac{\partial C}{\partial \psi} \right) \quad (6)$$

which applies to any PCT.

Unfortunately, equation 6 cannot be solved by a separation of variables technique. One can, however, formulate a perturbation solution to equation 6 in the deep region of the bed where the entrance effects have been damped. Equation 6 suggests as a first approximation that

$$\frac{\partial C}{\partial \xi} = 0 \quad (7)$$

at large Pe . This would imply that the concentration is a function of ψ only and is constant along a streamline. Any function of ψ will suffice. The first order term in the perturbation solution should then be a function only of ψ . The second order term will then be a diffusive correction function to take into account that the concentration must also be changing in the ξ direction. Assume a solution of the form

$$C(\psi, \xi) = C_1(\psi) + C_2(\psi, \xi) . \quad (8)$$

Substitution of equation 8 into equation 6 yields

$$\frac{\partial C_2}{\partial \xi} = \frac{8}{r_A Pe} \frac{\partial}{\partial \psi} \left((r/r_A)^2 v_{\xi} h_{\xi} \frac{\partial C_1}{\partial \psi} \right) \quad (9)$$

after neglect of the diffusive term in C_2 .

In the far downstream region of a PCT, the fractional decrease of concentration through each period must be the same, that is

$$C(\psi, \xi + 1) = C(\psi, \xi) e^{-\beta \lambda} \quad (10)$$

where β is independent of position. If we set $C_2(\psi, 0) = 0$, this means that C_2 and C_1 are related:

$$C_2(\psi, 1) = C_1(\psi) (e^{-\beta \lambda} - 1) . \quad (11)$$

Equation 9 can now be integrated from $\xi = 0$ to $\xi = 1$, to obtain a Sturm-Liouville eigenvalue problem for the function $C_1(\psi)$.

$$\frac{d}{d\psi} \left(G(\psi) \frac{dC_1}{d\psi} \right) + \lambda C_1 = 0 \quad (12)$$

$$G(\psi) = \int_0^1 (r/r_A)^2 v_{\xi} h_{\xi} d\xi \quad (13)$$

$$\lambda = (1 - e^{-\beta \lambda}) \frac{r_A Pe}{8} \approx \beta \lambda \frac{r_A Pe}{8} . \quad (14)$$

The integral in equation 13 is carried out over the arc length for a constant value of ψ in the integrand. The second identification of λ to $\beta \lambda$ in equation 14 is possible since $Pe \rightarrow \infty$.

Equation 12 is to be solved subject to the conditions

$$C_1(0) = 1 \quad (15i)$$

$$C_1(1) = 0 \quad (15ii)$$

$$C_1'(0) = -\lambda/G'(0) . \quad (15iii)$$

Condition (i) is a normalization for the first order solution.

Condition (ii) satisfies the limiting reactant constraint of a zero

wall concentration. Condition (iii) results from the finite concentration on the centerline.

The first eigenvalue of equation 12 can be related to the effective Sherwood number for a deep porous bed which is modeled as an array of PCT. A macroscopic mass balance on the reactant over the length of the period can be written in terms of an effective mass-transfer coefficient k_m (Newman and Tiedemann, 1976; Bennion and Newman, 1972). The β in equation 11 can then be related to this coefficient as

$$\beta = k_m a/v_s . \quad (16)$$

With equation 16 and 14, the Sherwood number for a limiting reactant in a deep bed with creeping flow and high Péclet number can be written as

$$Sh_B = \frac{\epsilon}{a} \frac{k_m}{D} = \lambda \left[\frac{2\epsilon}{ar_{Ad} \sqrt{1 + 1/2(A/r_A)^2}} \right]^2 \quad (17)$$

Equations 17 and 12 are the main results of this paper. By means of the perturbation approach, we have demonstrated how the two-dimensional convective-diffusion equation in a PCT can be reduced to a Graetz-like eigenvalue problem at high Péclet numbers. The first eigenvalue of this problem is simply related to the bed Sherwood number as given in equation 17.

The eigenfunction $C_1(\psi)$ generated by the perturbation analysis is a first order approximation to the concentration distribution. It identically satisfies equation 7, and gives the correct integral properties to the correction function $C_2(\psi, \xi)$. The local transfer rate to the wall can be found by differentiation of this profile with respect to the normal distance from the wall. After a change in coordinate system (see next section), the analysis yields

$$\left. \frac{\partial C}{\partial n} \right|_w = - \frac{\sqrt{B(z) r_w(z)}}{2r_A} \left. \frac{dC_1}{d\rho} \right|_{\rho=1} \quad (18)$$

where

$$B(z) = \left. \frac{\partial v_\xi}{\partial n} \right|_w .$$

The local wall flux is thus proportional to the square root of the local shear rate. The integral of equation 18 over the surface area of a period is related to the eigenvalue.

The left side of equation 17 depends upon the macroscopic bed quantities a and ϵ . The right side is a function of PCT geometry and flow regime through λ . The eigenvalues of equation 12 are independent of the Péclet number in creeping flow. Thus, irrespective of curvature effects, the asymptotic Sherwood number is a constant independent of the Péclet number for a deep bed.

Method of Solution

The eigenvalue problem as posed in equations 12 thru 15 is ill suited numerically to the ψ coordinate. Equation 12 has two singular points, one at $\psi = 0$, the other at $\psi = 1$. The singularity at $\psi = 0$ presents no problems; however that at $\psi = 1$ does. An analysis of equation 12 near the point $\psi = 1$ indicates that the first derivative of C_1 approaches infinity. A change in coordinate will eliminate this singularity. Define a length-like transformation variable ρ as

$$\psi = 2\rho^2 - \rho^4 . \quad (19)$$

Equation 12 and its boundary conditions then transform as

$$\frac{d}{d\rho} \left(\frac{G(\psi(\rho))}{4\rho(1-\rho^2)} \frac{dC_1}{d\rho} \right) + 4\lambda\rho(1-\rho^2)C_1 = 0 \quad (20)$$

$$C_1(\rho = 0) = 1 \quad (21i)$$

$$C_1(\rho = 1) = 0 \quad (21ii)$$

$$\frac{d}{d\rho} C_1(\rho = 0) = 0 . \quad (21iii)$$

Equations 20 and 21 were solved by the method suggested by Newman (1973) for eigenvalue problems.

All calculations were done on a CDC 7600 computer. Further details of the analysis and numerical programs have been compiled by Fedkiw.

Results and Discussion

The analysis presented in this work can be used to calculate the high Péclet number asymptotic Sherwood number for any continuous, periodic tube. Only the stream function need be known. Calculated results are presented for the sinusoidal PCT of figure 2 in creeping flow. The results are a function of the two dimensionless geometry parameters r_A and A/r_A .

Figure 4 presents the first eigenvalue of equation 12 normalized with respect to the first eigenvalue of the straight-tube Graetz problem ($\lambda_G = 0.91419$).

Figure 5 presents the Sherwood number for a packed bed modeled as an array of sinusoidal PCT. The concentration drop across the bed can be written as

$$\ln c_o/c_L = Sh_B \frac{aL_B}{\epsilon} \frac{6}{Pe_B} \quad (21)$$

Figure 4 shows a monotonic behavior of the eigenvalues with r_A and A/r_A . However, the bed Sherwood number shows different trends for small and large r_A . For small r_A , Sh_B increases with A/r_A , whereas for larger r_A this trend reverses itself. This effect is caused by the geometrical term in equation 17.

The quantity $2\epsilon/a$ in equation 17 is the standard definition for the equivalent radius of the bed. This defines the bed in terms of a straight cylinder network of radius $r_{eq,d}$ having the same surface area to empty volume ratio. The quantity $r_{Ad}^2 [1 + 1/2(A/r_A)^2]$

in the denominator of equation 17 defines another equivalent radius $\tilde{r}_{eq,d}$. This is the radius of a straight cylinder network through which the average velocity is given by v_s/ϵ . For long skinny PCT (small r_A), the ratio $(r_{eq}/\tilde{r}_{eq})^2$ is greater than one and increases with A/r_A . Thus for a bed composed of these tubes, the Sherwood number increases as A/r_A is increased. However, as r_A becomes larger the ratio $(r_{eq}/\tilde{r}_{eq})^2$ becomes less than one and Sh_B decreases with A/r_A .

For most beds r_A will be bounded approximately by $0.3 < r_A < 0.5$ while the A/r_A ratio will be in the range $0.2 < A/r_A < 0.5$, perhaps close to 0.33. Payatakes et al. report these parameters for a randomly packed bed of glass spheres as $r_A = 0.3$, $A/r_A = 0.36$, and for a bed of sand as $r_A = 0.31$, $A/r_A = 0.41$.

Sørensen and Stewart (1974) have calculated the asymptotic value of the Sherwood number in a simple cubic packed bed of uniform sized spheres. Their results yield $Sh_B = 0.619$. This information can be used in conjunction with the friction factor, Reynolds number product calculated by these same authors. This suggests that the PCT parameters for a simple cubic packing of spheres are $r_A \approx 0.5$ and $A/r_A \approx 0.33$. We expect this r_A value to be an upper limit for uniform spheres since the simple cubic packing has the highest porosity of all sphere packing configurations.

Notation

- a specific interfacial area of bed, cm^{-1}
 A dimensionless wall oscillation amplitude, A_d/ℓ
 c_o concentration of limiting reactant entering bed, mole/cm^3
 c_L concentration of limiting reactant leaving bed, mole/cm^3
 C dimensionless reactant concentration, $(C_d - C_o)/(C_b - C_o)$
 D diffusion coefficient of reactant, cm^2/sec
 G function of ψ defined by equation 13
 h_ψ, h_ξ, h_θ dimensionless metric factors
 k_m effective mass transfer coefficient of a bed, cm/sec
 L_B length of bed, cm
 ℓ length of PCT period, cm
 Pe reactant Péclet number in a PCT, $2r_{Ad}\langle v_{Ad} \rangle/D$
 Pe_B bed Péclet number, $6v_s/aD$
 r dimensionless radial coordinate, r_d/ℓ
 r_A dimensionless average PCT radius, r_{Ad}/ℓ
 r_w dimensionless wall radius, r_{wd}/ℓ
 $r_{eq,d}$ equivalent radius, $2\epsilon/a$
 $\tilde{r}_{eq,d}$ equivalent radius, $r_{Ad}\sqrt{1 + 1/2(A/r_A)^2}$
 Re_B bed Reynolds number, $6v_s/av$
 Sh_B bed Sherwood number, $\epsilon k_m/aD$
 v_s superficial approach velocity, cm/sec
 $\langle v_{Ad} \rangle$ average velocity in a tube of constant radius r_{Ad} , cm/sec
 v_ξ dimensionless streamwise velocity, $v_{\xi d}/\langle v_{Ad} \rangle$
 z dimensionless axial coordinate, z_d/ℓ

Greek

- β constant defined by equation 10, cm^{-1}
 ϵ bed porosity
 ν kinematic viscosity, cm^2/sec
 ξ streamwise coordinate
 ρ transformation coordinate of equation 17
 θ polar coordinate
 λ eigenvalue of equation 12
 ψ dimensionless normalized stream function, $-2\psi_d/r_{Ad}^2 \langle v_{Ad} \rangle$

Subscripts

- d dimensional quantity
b bulk

Acknowledgment

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References

- Bennion, D. N., and J. Newman, "Electrochemical removal of copper ions from very dilute solutions," J. Appl. Electrochem., 2, 113 (1972).
- Fedkiw, P.S., Ph.D. thesis, in preparation.
- Newman, J., "The Fundamental Principles of Current Distribution and Mass Transport in Electrochemical Cells," in Electroanalytical Chemistry, Marcel Decker, Inc., N.Y., 6, 187 (1973).
- Newman, J., and W. Tiedemann, "Flow-Through Porous Electrodes," in Advances in Electrochemistry and Electrochemical Engineering, John Wiley and Sons, Inc., 1976.
- Payatakes, A. C., C. Tien, and R. M. Turian, "A New Model for Granular Porous Media: Part 1. Model Formulation," AICHE J., 19, 58 (1973).
- , "Part 2. Numerical Solution of Steady State Incompressible Newtonian Flow Through Periodically Constricted Tubes," AICHE J. 19, 67 (1973).
- Shah, R. K., and A. L. London, "Laminar Flow Forced Convection Heat Transfer and Flow Friction in Straight and Curved Ducts -- A Summary of Analytical Solutions," Tech. Rep. No. 75, Dept. of Mech. Eng., Stanford Univ., Calif. (1971).
- Sørensen, J. P., and W. E. Stewart, "Computation of Slow Flow through Ducts and Packed Beds - III Heat and Mass Transfer in a Simple Cubic Array of Spheres," Chem. Eng. Sci., 29, 827 (1974).

Greek

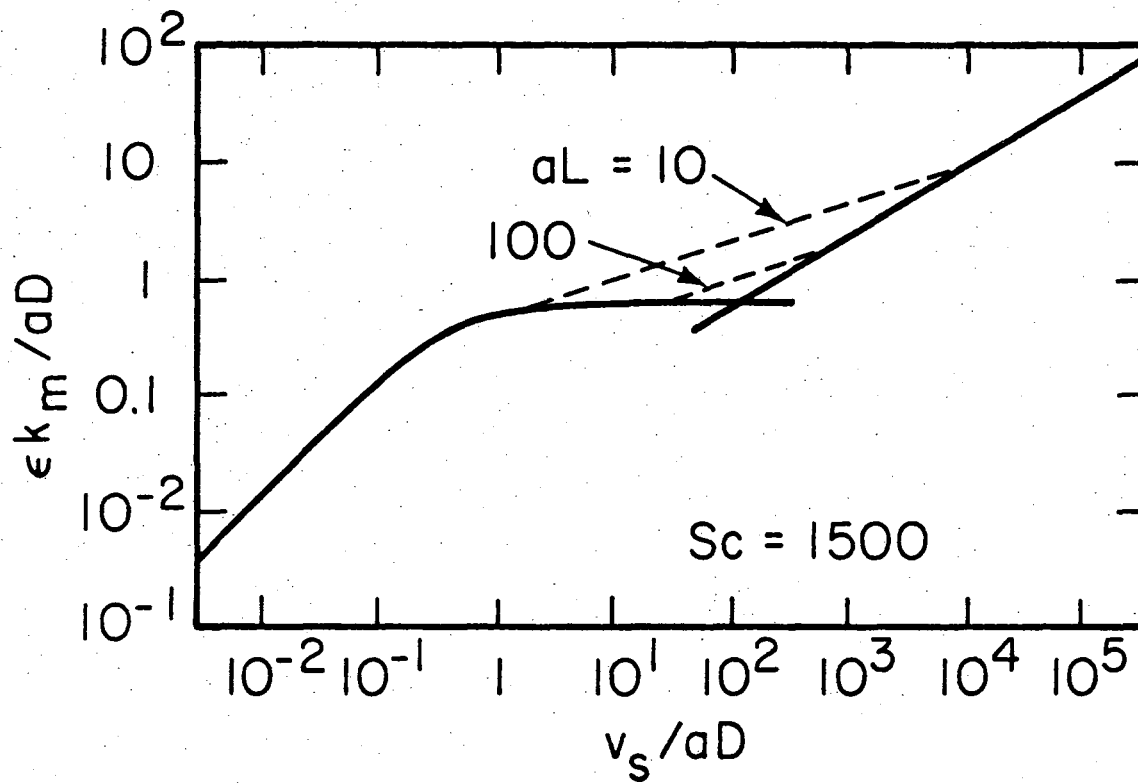
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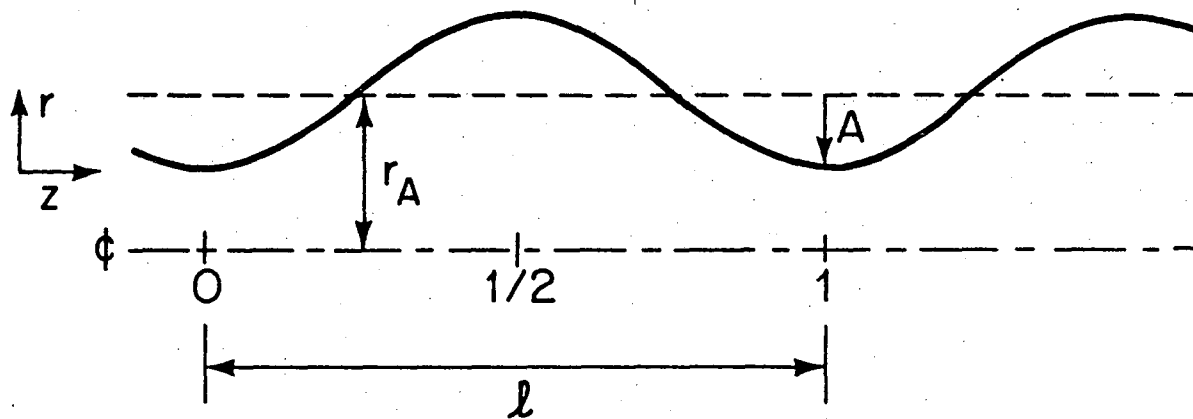
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Figure 1. Expected behavior of bed Sherwood number.



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Figure 2. The wall of a PCT generated by $r_w(z) = r_A - A \cos(2\pi z)$.
 All lengths are dimensionless with respect to the period
 length l .

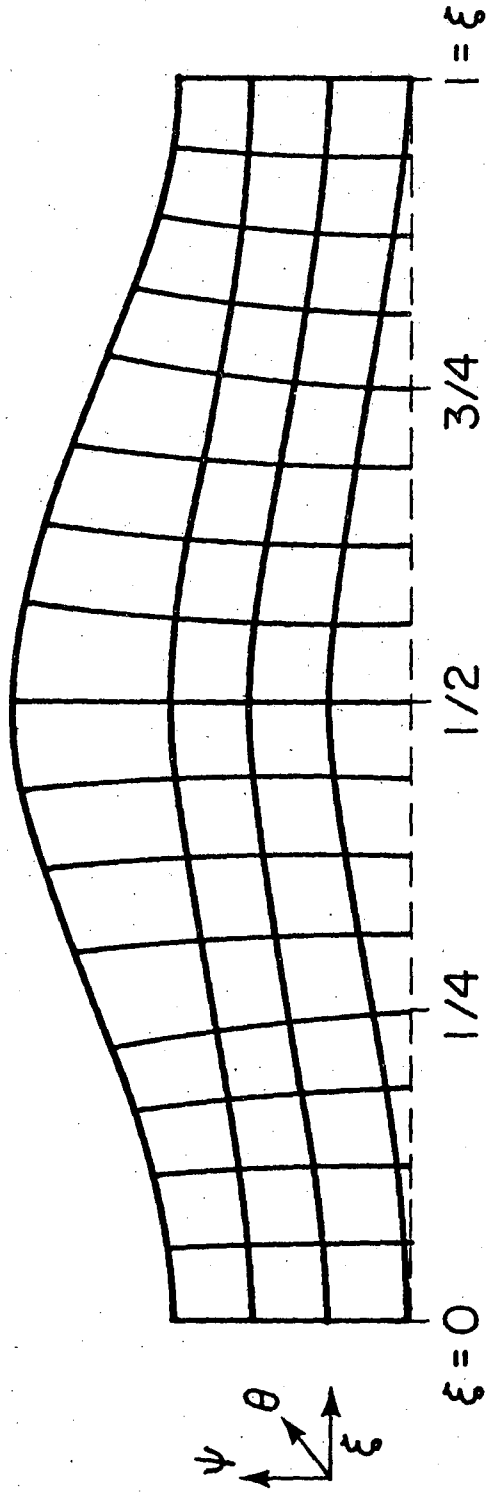
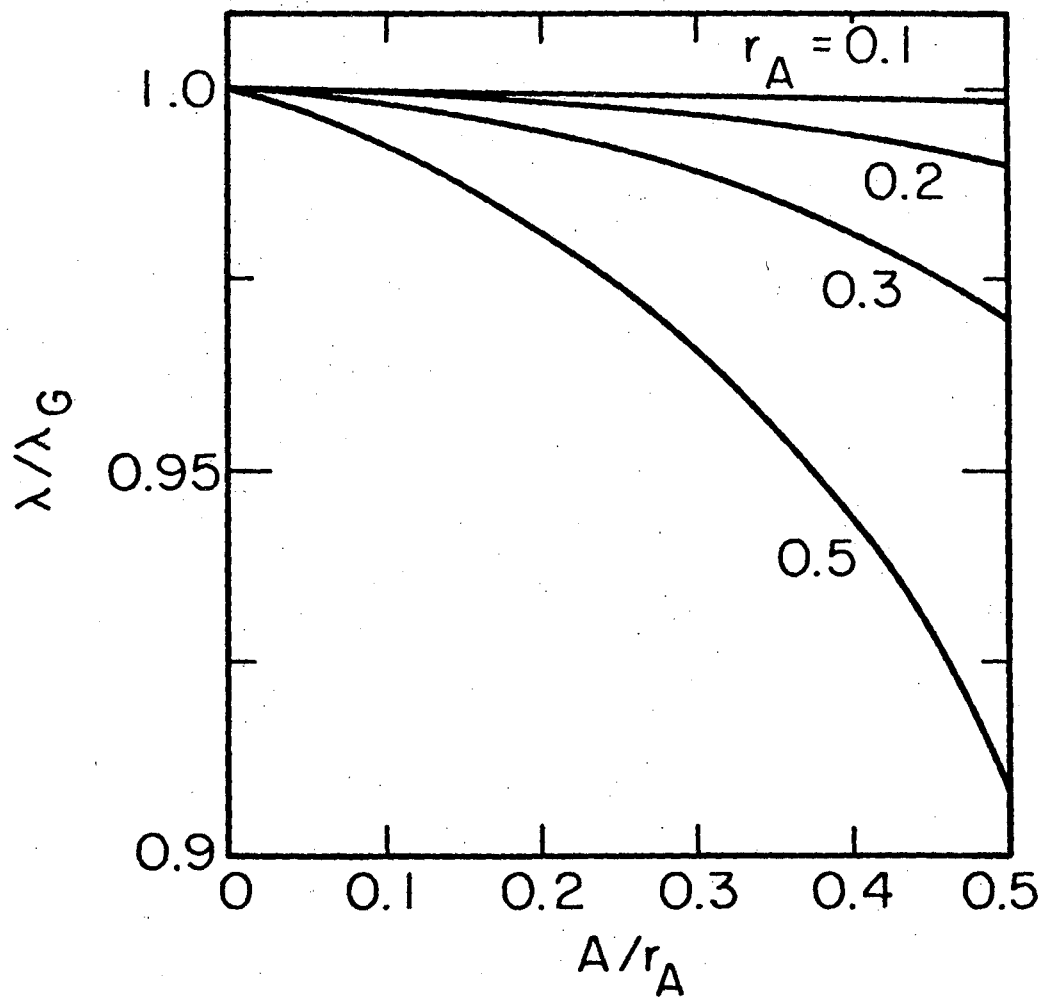


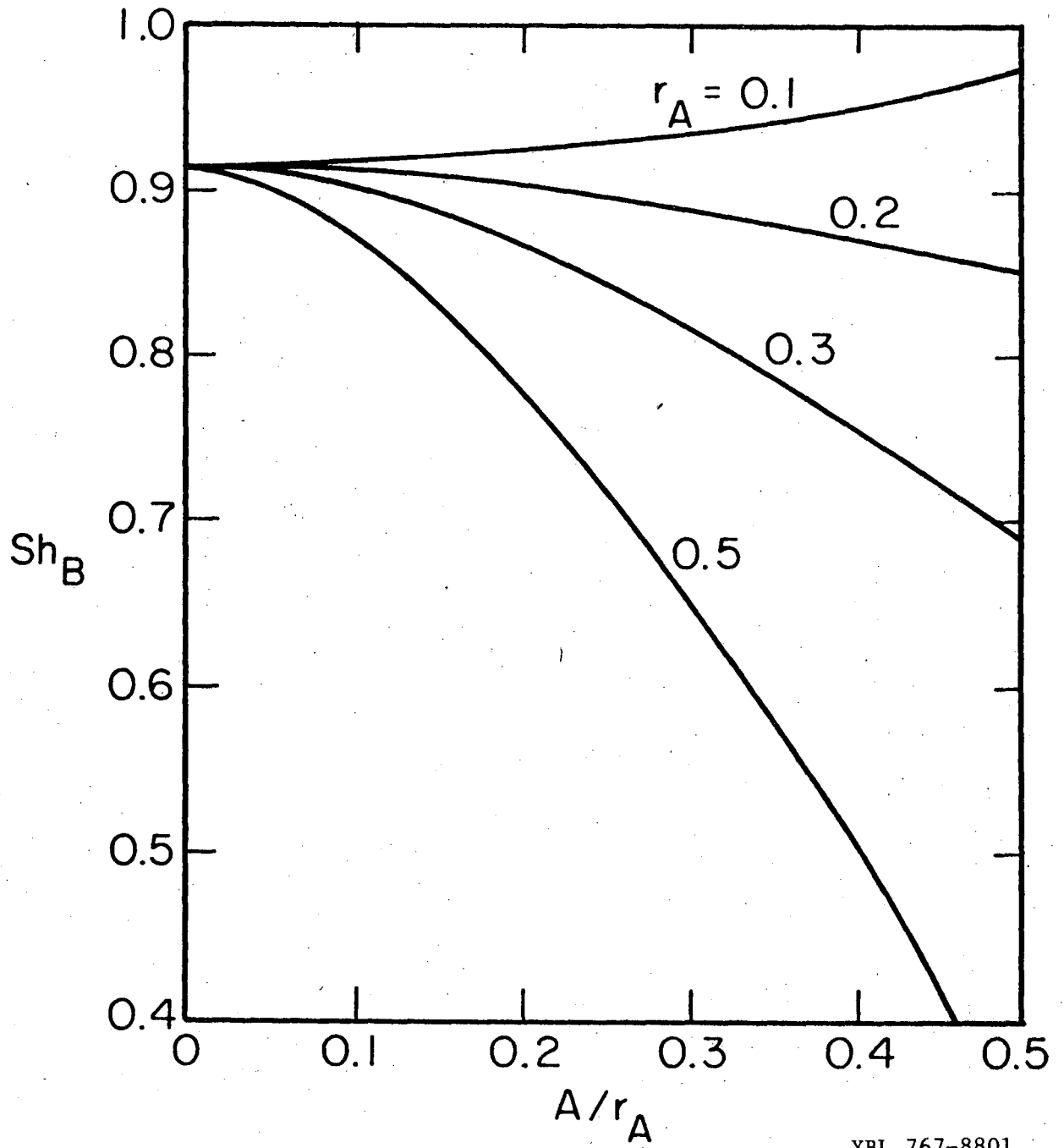
Figure 3. The (ψ, ξ, θ) coordinate system.

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Figure 4. Eigenvalues for the mass transfer problem in a PCT normalized with respect to the Graetz problem.



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Figure 5. Sherwood number for a packed bed modeled as an array of PCT.

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