SIMULATION OF NONLINEAR FIXED-BED ADSORBERS BY ORTHOGONAL COLLOCATION WITH THE AID OF FAST FOURIER TRANSFORM

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Introduction

The fast Fourier transform (FFT), which is capable of inverting some Laplace transforms, has been successfully applied to the prediction of the elution curves of fixed-bed adsorbers with linear [1] or nonlinear kinetics [2]. Recently, it has been furhter extended to the simulation of slurry adsorbers with pore diffusion and nonlinear kinetics [3]. The most attractive advantages of the FFT technique are its computing speed and accuracy. However, there is one major limitation in this method when dealing with relatively complicated systems: the memory space needed for the data may become very large. In addition, for such systems, the mathematical manipulations to obtain workable transfer functions may not be feasible.

The orthogonal collocation method is frequently used to simulate fixed-bed absorbers because of its computing speed [4]. After the partial differential

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equations are primarily treated by the orthogonal collocation method, the resulting ordinary differential equations can be solved numerically by any standard integration method. The mathematical manipulation needed is little and the programming is easy. However, this method is not so powerful as one might foresee for difficult problems in which the temporal integrations require much computation time. Moreover, modern integration routines such as RKGS (subroutine in SSP) and DGEAR (subroutine in IMSL) proceed with changing step size, and introduce inconvenience in the performance of parameter estimation.

The main objective of the present study is to establish a method that combines the FFT method and orthogonal method to overcome the shortcomings of these two methods. A fixed-bed adsorber with nonlinear kinetics and axial dispersion is chosen as an example in this work.

1. Development of the Method

Consider a fixed-bed adsorber with nonlinear adsorption kinetics. The flow pattern in the adsorber is assumed to be plug flow with axial dispersion. For ease of explanation, an adsorption-controlled process was assumed in this study. Under these circumstances the dimensionless governing equations can be written as

$$\frac{\partial y}{\partial \tau}=\frac{1}{\mathrm{Pe}}\frac{\partial^2 y}{\partial x^2}-\frac{\partial y}{\partial x}-\beta\frac{\partial \theta}{\partial \tau} \tag{1}$$

$$\frac{\partial \theta}{\partial \tau}=\phi_1 y(1-\theta)-\Phi_2\theta \tag{2}$$

$$y(\tau=0, x)=\theta(\tau=0, x)=0 \tag{3}$$

$$\frac{1}{\mathrm{Pe}}\frac{\partial y}{\partial x}=y-y_{in} \qquad \text{at } x=0 \tag{4}$$

$$\frac{\partial y}{\partial x}=0 \qquad \text{at } x=1 \tag{5}$$

By applying orthogonal collocation with respect to the axial coordinate, Eqs. 1–5 can be reduced to a set of ordinary differential equations that are initial-value problems and can be solved by using general methods of integration. In the present study, however, these equations are further reduced to algebraic equations by taking Laplace trasnformation. The resulting set of equations can be written as:

$$\bar{\theta}_i=\frac{\Phi_1}{\Phi_2+s}(\bar{y}_i-\overline{y_i\theta_i}) \qquad i=2, \ldots, M+1 \tag{6}$$

$$\begin{pmatrix} 1-A_{1,1}/Pe & -A_{1,2}/Pe & \cdots & -A_{1,M+2}/Pe \\ \mu_{2,1} & \mu_{2,2} & \cdots & \mu_{2,M+2} \\ \cdot & \cdot & \cdots & \cdot \\ \cdot & \cdot & \cdots & \cdot \\ \mu_{M+1,1} & \mu_{M+1,2} & \cdots & \mu_{M+1,M+2} \\ A_{M+2,1} & A_{M+2,2} & \cdots & A_{M+2,M+2} \end{pmatrix} \times \begin{pmatrix} y_1 \\ y_2 \\ \cdot \\ \cdot \\ y_{M+1} \\ y_{M+2} \end{pmatrix} = \begin{pmatrix} y_{in} \\ \delta y_1\theta_1 \\ \cdot \\ \cdot \\ \delta y_{M+1}\theta_{M+1} \\ 0 \end{pmatrix} \tag{7}$$

where

$$\mu_{i,j}=\begin{cases} s+\delta+A_{i,j}-\dfrac{B_{i,j}}{Pe} & i=j \\ A_{i,j}-\dfrac{B_{i,j}}{Pe} & i\neq j \end{cases} \tag{8}$$

$$i=2, \ldots, M+1; j=1, 2, \ldots, M+2$$

$$\delta=\frac{\beta\Phi_1 s}{\Phi_2+s} \tag{9}$$

where M is the number of interior collocation points; $A_{i,j}$ and $B_{i,j}$ represent the first and second derivatives of orthogonal polynomials at the zeros, respectively. The desired time-domain solution can then be obtained by using the following iterative procedure:

(1) Calculate $A_{i,j}$ and $B_{i,j}$.
(2) Obtain $\bar{y}_i$ and $\bar{\theta}_i$ by setting $\overline{y_i\theta_i}$ equal to zero, i.e. a linear approximation.
(3) Invert $\bar{y}_i$ and $\bar{\theta}_i$ into the time domain.
(4) Calculate $y_i\theta_i$ numerically with the resulting y_i and θ_i.
(5) Transfer $y_i\theta_i$ to the Laplace domain.
(6) Substitute $\overline{y_i\theta_i}$ into Eqs. 6 and 7 to obtain new $\bar{y}_i$ and $\bar{\theta}_i$.
(7) Repeat steps 3–6 until convergence is achieved.

2. Result and Discussion

In this study, the breakthrough curves of an adsorption column were obtained by integrating the elution data of pulse input with respect to time. Because a Dirac's delta pulse input was used for the calculation, $\bar{y}_{in}$ is equal to unity. The breakthrough curves obtained from various numbers of iterations were evaluated and are shown in **Fig. 1**. It is important to check whether the iterative procedure is rapidly convergent. If rapid convergence is not assured, the present method may not give a correct solution. For the present case, only two iterations were needed to reach a good convergence. After the number of iterations needed was determined, data of break-

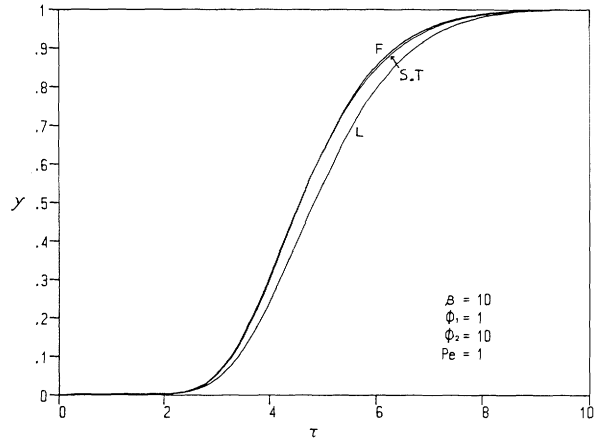

Fig. 1. Convergence of breakthrough curves. L: linear approximation, F: 1st iteration; S: 2nd iteration; T: 3rd iteration

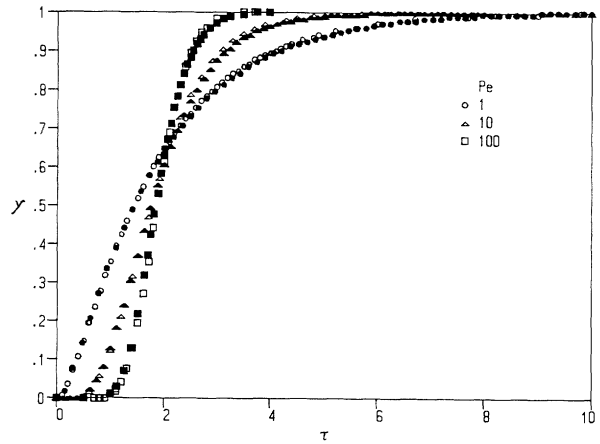

Fig. 2. Comparison of breakthrough data. Solid symbols: the present method; open symbols: the conventional orthogonal method. $\beta=10$, $\Phi_1=1$, $\Phi_2=10$.

through curves with different N were calculated and compared. It was found that $N=32$ is sufficient to give a satisfactory result.

It is known that, to avoid oscillation or divergence, the conventional orthogonal collocation method should be manipulated with an adequate number of collocation points and sufficiently small step sizes in the time integration. Therefore, the computation time may be large. However, when the time integration is replaced by the FFT method (i.e. the present method), the execution time can be reduced because the computations needed are fewer. It is also found that, even for a difficult system, the number of sample points N used in the present method has little effect on the accuracy of the result.

To compare the computing speed of the present method with that of the conventional orthogonal collocation method, the breakthrough curves for various sets of parameters were evaluated. A comparison between these two methods is shown in **Fig. 2** and in **Table 1**. It can be seen that the breakthrough data obtained by the two methods correspond with each other, but the computation times required for the conventional collocation method are longer and vary with the parameters employed. The longer the process time involved, the greater is the computation time needed by the conventional method—but not in the present method.

In summary, the present method, combining orthogonal collocation and the FFT technique, has been shown to be adequate for calculation of the breakthrough curves of nonlinear adsorption systems. In addition to high computing speed, easy programming and few mathematical manipulations needed, the selectable time interval makes it convenient for parameter estimations. On the basis of these advantages, we believe that this method is elegant in application to nonlinear fixed-bed adsorption problems.

Table 1. Comparison of Computing Speeds of Present Method and Conventional Orthogonal Collocation Method

Method	*Pe*	*N*	Execution time on CDC Cyber-840, sec
OC+FFT	1, 10, 100	32	0.82
		64	1.60
		128	3.16
OC+DGEAR	1		49.74
	10		8.27
	100		5.82

OC: orthogonal collocation
Process parameters: $\Phi_1=1$, $\Phi_2=10$, $\beta=10$
$M=10$ for all cases

Nomenclature

C	=	adsorbate concentration in fluid phase	[mol/cm^3]
C_0	=	inlet concentration of adsorbate	[mol/cm^3]
$C_{in}(t)$	=	column input function	[—]
D_L	=	axial dispersion coefficient	[cm^2/s]
k_1	=	adsorption rate constant	[cm^3/mol, s]
k_2	=	desorption rate constant	[s^{-1}]
L	=	column length	[cm]
M	=	interior collocation points	[—]
N	=	number of sample points used in FFT	[—]
Pe	=	VL/D_L	
q_m	=	adsorption concentration at complete coverage of surface	[mol/g]
R	=	radius of packed particles	[cm]
s	=	Laplace variable	[—]
t	=	time	[s]
τ	=	$t/(L/V)$	
V	=	interstitial velocity	[cm/s]
x	=	z/L	
y	=	C/C_0	
y_{in}	=	C_{in}/C_0	
z	=	axial distance	[cm]
β	=	$(1-\varepsilon)\rho_{pqm}/\varepsilon C_0$	
δ	=	parameter, Eq. 9	
ε	=	bed void fraction	
ε_p	=	porosity of particles	
θ	=	fraction of surface coverage	

$\mu_{i,j}$ = parameter, Eq. 8
ρ_p = density of solid particle [g/cm^3]
Φ_1 = $k_L C_0(L/V)$
Φ_2 = $k_2(L/V)$

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