Mathematical Modelling of Displacement Washing of Packed Bed of Cellulosic Fibers

Abstract: Mechanism of displacement washing of packed bed of porous, compressible, cylindrical particles such as cellulosic fibers (pulp) is presented with the help of two parameters namely, Peclet number (Pe) and Biot number (Bi). Bulk fluid solute concentration, intra-pore solute concentration and the concentration of solute adsorbed on fiber surface are taken to be the function of washing time and position in the bed. Concentration of solute adsorbed on the fiber surface and intra-pore solute concentration are related by Langmuir adsorption isotherm. Model equations are solved using orthogonal collocation on finite elements. Key industrial parameters such as wash yield, displacement ratio and % efficiency of washing are calculated from model. Model predicted values are compared with the experimental values and a good correlation is found between the two.

Key Words: Peclet number, Biot number, Displacement ratio, %Efficiency, Wash Yield, Wash ratio.







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Introduction

Problem of heat and mass transfer during flow through packed beds of porous particles is of great interest in the field of engineering and science. For the past few decades researches on these problems have also gained momentum in the field of mathematical modelling. A variety of models related to adsorptiondesorption (Raghvan & Ruthven, 1983; Sridhar et. al., 1994; Liu & Bhatia, 1999) and diffusion-dispersion phenomenon (Grahs, 1974; Al-Jabari et. al., 1994; Eriksson et. al., 1996; Potucek, 1997) have been developed. Ding & Bhatia (2003) has considered the Knudsen and molecular diffusivity depending on the pore size of the particle, Dudukovic (1977), Bhatia (1987) and Liu & Bhatia

(2001) have discussed the radial diffusivity in gaseous system. Cavendish & Oh (1979), Sharma & Yortsos (1987), Immanuel et al., (2003) have considered the effective diffusion coefficient in the porous media. Balasubramanian et al. (1999) has considered the radial dispersion in the curved tubes. Arora et al. (2006) have considered the pore diffusion in particles using pore diffusion model.

In the present study the washing behavior of pulp which consists of cylindrical, compressible particles with porous structure is presented through a dynamic model using Peclet number (Pe) and Biot number (Bi). Peclet number is the ratio of advection (transport by bulk fluid motion) to dispersion whereas the Biot number signifies the mass transfer rate to dispersion rate in the packed bed of cellulosic fibers. It relates the mass transfer resistances inside and at the surface of the body. The black liquor solutes (lignin derivatives, Na, Mg, Ca and K ions) occupy the space (interstices) between the fibers (as displaceable solutes) and in the pores of the fibres (as non-displaceable solutes). These solutes are removed by water/weak wash liquor. The mechanism involved is the sum of displacement of the liquor by the movement of water plug controlled by fluid mechanics, dispersion due to back mixing, diffusion due to concentration gradient and adsorption-desorption due to relative affinity of various solutes towards the fiber surface. Grahs (1974) has divided the packed bed of fibers into three zones namely, flowing liquor, stagnant liquor and fibers as shown in Fig. 1. The mass transfer takes place between fibers to stagnant liquor and then from stagnant liquor to flowing liquor.



Figure 1: A packed bed of cellulose fibers

Mathematical Model of Displacement Washing of Bed of Pulp Fibers

In the washing process of brown pulp, solute present in the irregular void channels of the bed, associated with diffusion like dispersion, is removed by the introduction of a solvent. Due to porous nature of fibers, solutes present in the pores of particles diffuse out when it comes in contact with the external fluid. Since some solutes are adsorbed on fiber surface, mass transfer takes place through internal and external fiber surface and from fiber surface to the external fluid.

Flow of fluid through the bed is described by bulk fluid concentration c(z,t). Concentration of solute adsorbed on fiber surface and intrapore solute concentrations are described by n(z,t) and q(z,t), respectively. Fiber and bed porosities are described by β and ε , respectively and the consistency of fibers is described by C_F. Axial dispersion coefficient (D_L) is independent of the axial distance *L*. Mass transfer through the stagnant layer is controlled by film resistance mass

transfer coefficient (k_{p}) . Proposed model is based on the following simplified assumptions.

- 1. System is isothermal and the bed of fibers is macroscopically uniform.
- 2. Cellulose fiber particles are porous, compressible in nature having cylindrical geometry.
- Movement of solute within the fiber pores is described by Fick's law of molecular diffusion and dispersion.
- 4. Molecular diffusion is quantified by diffusion coefficient (D_v) whereas dispersion in fluid is measured by axial dispersion coefficient (D_i) .
- Nonlinear adsorption isotherm prevails. Thus Langmuir adsorption isotherm is assumed to be followed to relate the concentration of solute adsorbed on fiber surface and the intrapore solute concentration.

at z = L(6)

The differential mass balance for bulk fluid phase in packed bed of cellulose fibers is given in Fig 1A. The unsteady state partial differential equations describing the behavior of fluid flow through the packed bed (cake of fibers, called pulp mat) are given below:



Fig. 1A: Differential mass balance for bulk fluid phase in packed bed of cellulose fibers

Mathematical equations for pulp fiber particle phase

Adsorption isotherms

As already indicated Langmuir type adsorption isotherm has been followed to relate the concentration of solute adsorbed on fiber surface and intrapore solute concentration. Adsorption rate constant (k_1) is assumed to be of second order in forward direction and desorption rate constant (k_2) is of first order in backward direction:

At equilibrium, equation (2) reduces to Langmuir adsorption isotherm:

2.3 Mathematical equations for bulk fluid phase

$$u\frac{\partial c}{\partial z} + \frac{\partial c}{\partial t} + \frac{2(1-\varepsilon)k_f}{\varepsilon KR}(c-q) = D_L \frac{\partial^2 c}{\partial z^2}, \qquad (4)$$

Boundary conditions :

$$uc - D_L \frac{\partial c}{\partial z} = 0$$
, at $z = 0$(5)

$$\frac{\partial c}{\partial z} = 0 ,$$

Initial condition :

 $C = q = C_0$ and $n = N_0$. at t = 0(7)

The set of equations (1) to (7) are converted into following dimensionless form using non dimensional variables given in nomenclature.

Boundary conditions :

$$C - \frac{1}{Pe} \frac{\partial C}{\partial \xi} = 0$$
, at $\xi = 0$ (11)

Initially

$$C = Q = N = 1$$
 at $\tau = 0$ (13)

In equations (8) and (10) two dimensionless numbers are presented namely Peclet number $Pe = \frac{uL}{D_L}$ and Biot number $Bi = \frac{k_L L^2}{KRD_L}$. The physical significance of these two dimensionless numbers is that Peclet number (Pe) measures the extent of dispersion and Biot number (Bi) signifies the mass transfer resistances inside and on the surface of the fibers.

Solution of the model

There are many techniques available to solve a system of partial differential equations. Some important solution procedures are given below:

Laplace transform techniques mainly limited to linear differential equations. For problems of nonlinear type, there are two ways: through numerical methods or by using simulation software. The former includes finite difference method on an equidistant grid, Orthogonal Collocation or mixed collocation method, orthogonal collocation on finite elements (Arora et al., 2006, 2007), Spline collocation method, Cubic Hermite collocation, orthogonal collocation of mathematical software has become more popular now a days as it is the easiest way to get the solution. There are plenty of softwares available. For example, Kumar et al. (2013) employed MATHEMATICA, whereas Gupta et. al. (2011) used OSCM and MATLAB 'pdepe' solver.

Orthogonal collocation on finite elements

Method of orthogonal collocation on finite elements (OCFE) is followed to solve the system of partial differential equations (8) to (13). This method was first developed by Carey & Finlayson (1975) to solve the stiff system of ordinary differential equations involving Thieles modulus. Later on Sridhar (1996) and Liu & Bhatia (1999) have followed this technique in a modified form. However, their methods were influenced by collocation points instead of elements. In present technique, the axial domain is divided into small elements and then orthogonal collocation is applied. Present model is a special case of the model given in Arora et. al. (2006).

In OCFE, the global domain $0 \le \xi \le 1$ is divided into small elements of equal length ξ_{ℓ} . Within ℓ^{th} element, a new variable v is introduced in such a way that v varies from 0 to 1 as ξ varies from ξ_{ℓ} to $\xi_{\ell+1}$. Domain of v is called local domain and orthogonal collocation is applied on v. In Fig 2 and Fig 3, mathematical description of global and local domains is presented, respectively. The trial function and its first derivative are taken to be continuous at the nodal points.



Figure 3: Location of collocation points in tth element

3.2 Collocation points

Choice of collocation points is an important part of orthogonal collocation method. In present study, zeros of Legendre polynomials have been taken as collocation points, since these polynomials give good results on average:

$$(j-1)P_{j-1}(\eta) = (2j-3)\eta P_{j-2}(\eta) - (j-2)P_{j-3}(\eta); j = 2, 3, ..., m$$
.....(14)

 η 's lie in the domain of [-1,1] and can be transformed on to [0,1] using following relation:

$$x_{m+2-j} = \frac{\eta_j}{2} + \frac{1}{2}; j = 2, 3, ..., m$$
.....(15)

Application of OCFE

OCFE is applied on the set of partial differential equations (PDE's) (8) to (13) which convert these PDE's into a set of differential algebraic equations (DAE's) as follows:

$$\frac{\partial Q_j^{\ell}}{\partial \tau} + \frac{1-\beta}{\beta} N' \frac{\partial N_j^{\ell}}{\partial \tau} = \frac{Bi}{Pe} (C_j^{\ell} - Q_j^{\ell}); \ j = 2,3,\dots,m \text{ and } \ell = 1,2,3,\dots,r$$
(16)

$$\frac{\partial N_{j}^{\ell}}{\partial \tau} = P^{*} \left[C' Q_{j}^{\ell} (1 - N_{j}^{\ell}) - k^{*-1} N_{j}^{\ell} \right]; \ j = 2, 3, \dots, m \text{ and } \ell = 1, 2, 3, \dots, r$$

$$C_1^1 - \frac{1}{Pe} \sum_{i=1}^{m+1} A_{1i} C_i^1 = 0$$
, at $\xi = 0$ (19)

The required system is reduced into a stiff system of r(3m+1)+1 DAE's, which is solved using MATLAB with ode15s system solver. In this system the set of differential algebraic equations is not converted into the set of ordinary differential equations and hence saves the computational time.

Results and Discussion

Simulation using lab scale data

To validate the model, experimental data of a lab scale pilot plant is used and is given in Table-I (Kukreja et al, 1995). Experimental results are presented for lignin. These experiments were performed on a drum washer of width 30.48 cm and radius 23.46 cm. About 25-30 kg of blown unwashed pulp (mixture of Indian wood and non-wood pulp) with Kappa number 20 at 348K, was collected from a paper mill. It was cooled to room temperature. Pulp was diluted with water to different consistencies to carry out experiments. Cake was washed

using clean water. RPM of the drum, cake thickness and vacuum were measured during experiments. Solid content, density, viscosity, and surface tension of the black liquor, consistency and density of fibers were determined in the laboratory. It is observed in this study, that no single factor effects the whole washing process, rather, these parameters interrelated on each other. Both Peclet number and Biot number depend upon axial dispersion coefficient. Interstitial velocity depends upon bed porosity as given by Darcy's law.

Table - I : Experimental data of lab scale pilot plant

Parameter	Range	Unit
C ₀	$(77.3-159) \ge 10^{\circ}$	kg/ m ³
k_1/k_2	$(1.54-2.48) \ge 10^{-14}$	-
В	$(9.61-8.79) \ge 10^{-1}$	-
Е	(9.86-9.54) x 10 ⁻¹	-
u/L	$(1.398-2.460) \ge 10^{-1}$	1/s
k _f	$(5.580-8.029) \ge 10^{-7}$	m/s

Therefore, with the change in bed porosity, both the Peclet number and Biot number change which ultimately effect the whole process. Also film resistance mass transfer coefficient depends upon interstitial velocity (Shiraishi et al., 1996), which ultimately effects the Biot number.

The effect of Peclet number and Biot number on exit solute concentration is shown in Fig 4 with the help of breakthrough curves. It is observed from this figure that with the increase in Peclet number and Biot number, solution profiles converge to zero more rapidly as compared to smaller ones. It is because of the reason that with the increase in Peclet number and Biot number, axial dispersion coefficient decreases which results in decrease in

interstitial velocity. As a result, bed porosity increases resulting in the availability of more volume for flow, due to which less back mixing of solute occurs. Hence impurities adsorbed on fiber surface can be washed out effectively within small time period.

Effect of Peclet number and Biot number on Industrial parameters

The effect of Pe and Bi is expressed in terms of common parameters used by industry like displacement ratio (DR), wash ratio (WR) and % efficiency. These parameters have been calculated from exit and average solute concentration. The equations have been dealt in details in Arora et. al. (2006).

Displacement Ratio: It is the ratio of actual possible reduction in dissolved solids to maximum possible reduction at any stage. This factor is commonly used in industry to check the functioning of equipment and reduction in black liquor solids. Effect of sorption is ignored in displacement ratio. It is calculated using the formula:

$$DR = \frac{C_0 - c_d}{C_0 - C_S}$$
(22)

In Fig 5, displacement ratio is plotted versus time for different values of Peclet number and Biot number. It is observed from this figure that with the increase in Peclet number and Biot number, displacement ratio converges to steady state condition rapidly as compared to small values of Peclet number and Biot number.

It signifies that maximum possible reduction in black liquor solids can be achieved at large porosity levels with less back mixing and axial dispersion. Similar results have also been discussed by Grahs (1974), Crotogino et. al. (1987) and Trinh et. al. (1989).

% Efficiency: It is the key factor to check the efficiency of equipment. It signifies percentage of black liquor solids removed during washing operation. Higher % efficiency gives better percentage of removal of solutes which ultimately gives higher efficiency of equipment. Kukreja et. al., (1998) has calculated % efficiency using displacement ratio, inlet and outlet consistencies of pulp, respectively.



Figure 4: Effect of Peclet number and Biot number on exit solute concentration profile.



Figure 5: Effect of Peclet number and Biot number on displacement ratio.

% efficiency = $(1 - ((1 - DR)(100 - C_{yd})/(100 - C_{yi})))100$ (23)

Behavior of % efficiency for different values of Peclet number and Biot number is shown in Fig 6. It is observed from this figure that with the increase in Peclet number and Biot number, % efficiency increases. It supports the experimental fact of the study of Trinh & Crotogino

(1987) and Crotogino et. al. (1987) that better efficiency of the equipment can be achieved for large cake thickness and small flow rates which ultimately depend upon porosity level of the bed.

Cullinan (1991) has mentioned that local efficiency is governed by the rate of mass transfer, being operated with diffusion and displacement. Hence the overall performance of the wash stage is determined by the local efficiency.

Wash Ratio: It is the ratio of the wash liquor entering to the liquor leaving the pulp. Wash ratio can also be calculated using %efficiency and displacement ratio as given in following equation. In Fig 7, it is observed that wash ratio is effected more by Peclet number and Biot number than that of displacement ratio and %efficiency which ultimately effects the whole washing process. In practical situations, wash ratio should be greater than 1 for good recovery of solutes. WR = 1, refers to minimum effective wash ratio corresponding to ideal case when shower concentration is equal to the concentration of solute in unwashed cake.

$$WR = \frac{-100 \times \ln(1 - DR)}{\% efficiency}$$
(24)

Wash Yield: It is the ratio of dissolved solids removed to the dissolved solids entering with the unwashed



Figure 8: Effect of Peclet number and Biot number on wash yield.







Figure 7: Effect of Peclet number and Biot number on wash ratio.

pulp at any stage. If we assume that solute is split in same proportion as liquor, it is easy to calculate how much solute is left in thickened pulp. Crotogino et. al. (1987) has calculated the wash yield in terms of extraction step input and discharge consistency. Kukreja et. al. (1998) have calculated wash yield by relating it with % efficiency and wash ratio as:

$$WY = 1 - \exp(-\% \, efficiency \times WR) \qquad (25)$$

For wash liquor ratio equal to unity, wash yield is equivalent to displacement ratio. In Fig 8, the behavior of wash yield is shown for different values of Peclet number and Biot number. It is observed from this figure that wash yield is not significantly affected by change in Peclet number and Biot number. However, significant changes can be seen at $\tau > 0.8$ for Pe = 32.5 and Pe = 10, which signifies that higher wash yield can be obtained for large Peclet number and Biot number.

Values of displacement ratio, %efficiency and wash ratio are calculated experimentally using the samples obtained from industry and are compared with the values calculated from model. The percentage error is found to be less than 3% which is within the range of acceptable accuracy. Comparison of experimental and model predicted values is given in Table-II. Subscript E stands for experimental values and subscript C stands for model predicted values.

Model predicted values imply the exit and average solute concentration calculated at different values of Bi and Pe. Then these values have been taken as input values to calculate industrial parameters. Therefore, these parameters indirectly depend upon Pe and Bi.

Pe & Bi	DR _E	DR _C	Relative error*	%E _E	%E _C	Relative error*	WR _E	WR _C	Relative error*
32.5 & 12.8	0.76	0.78	-0.0263	76.76	78.39	-0.0212	1.13	1.1	0.0265
20.5 & 7.7	0.76	0.76	0	76.34	77.22	-0.0115	1.16	1.19	-0.0259
20.3 & 5.0	0.77	0.76	0.0130	77.68	77.34	0.0044	1.19	1.19	0
21.4 & 5.8	0.78	0.76	0.0256	78.68	77.71	0.0123	1.14	1.17	-0.0263
18.5 & 4.9	0.8	0.82	-0.0250	81.27	82.89	-0.0199	1.18	1.17	0.0085
17.5 & 4.8	0.77	0.75	0.0260	77.56	77.01	0.0071	1.3	1.3	0
19.4 & 6.6	0.75	0.76	-0.0133	76.06	77.46	-0.0184	1.22	1.29	-0.0574
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Table-II : Comparison of experimental and model calculated values

Relative error = $\frac{Experimental value - Model predicted value}{Experimental value - Model predicted value}$

Experimental value

Conclusions

In present work, a dynamic mathematical model has been developed and an attempt has been made to correlate it with industrial parameters. Effect of base case parameters such as bed porosity, cake thickness, interstitial velocity and axial dispersion coefficient has been lumped into Peclet number and Biot number. It is observed that these factors do not effect the washing process separately rather they simultaneously effect the washing behavior. Different industrial parameters such as displacement ratio, %efficiency, wash yield, wash ratio have been calculated through the mathematical model and numerical results have been presented in the form of breakthrough curves. A comparison between experimental and model predicted values has been made and the error is found to be less than 3%. It signifies the applicability and validity of model. It is also observed that higher efficiency of the equipment can be achieved for large Peclet number and Biot number and hence efficient washing operations can be obtained.

During washing the speed at which the diffusion of black liquor solids inside the fiber voids and the surrounding liquor takes place is dependent on the concentration difference between the black liquor solutes inside and outside of the fibers, temperature, turbulence around the fibers and the size of the molecule. The larger the concentration difference, the higher the temperature and turbulance and the smaller the molecule size, the faster the diffusion will be and hence faster the equilibrium will be reached (16).

Nomenclature

Bi	=	Biot number, $\frac{k_f L^2}{KRD_r}$
c	_	Concentration of solute in the liquor kg $/m^3$
C _d	=	Concentration of solute in the discharged pulp, kg /m ³
С	=	Dimensionless concentration, c/C_0
C'	=	Dimensionless parameter, C_0 / C_F
Co	=	Inlet solute concentration in the vat, kg /m3
C _F	=	Fiber consistency, kg /m ³
Cs	=	Solute concentration in the wash liquor, kg /m3
C _{vd}	=	Discharged consistency of pulp, kg of fibers / kg of liquor
C _{vi}	=	Inlet vat consistency of pulp, kg of fibers / kg of liquor
DL	=	Axial dispersion coefficient, m ² /s
k.	_	Dimensionless parameter k_1/k_2
k.k.	=	Mass transfer coefficient, 1/s
k.	=	film resistance mass transfer coefficient. m/s
ĸ	=	Volume equilibrium constant, dimensionless
L	=	Thickness of the bed, m
n	_	Concentration of solute adsorbed on the fibers, kg /m ³
	_	
N	=	Dimensionless concentration of solute in fibers, n/N_0
N'	=	Dimensionless parameter, N_0 / C'
N ₀	=	Initial concentration of solute adsorbed on the fibers, kg $/m^3$
P.	=	Dimensionless parameter, $\frac{k_1L}{u}$
Pe	=	Peclet number, $\frac{uL}{D_r}$
q	=	Intrapore solute concentration, kg /m ³
Q	=	Dimensionless intrapore solute concentration, q/C_0
R	=	Particle radius, m
t	=	Time, s
u	=	Interstitial velocity through bed, m /s
z	=	Distance from point of introduction of solvent, m
0		
Greek S	sym	DOIS
β	=	Particle porosity, dimensionless
3	=	Porosity of cake, dimensionless
θ	=	Dimensionless parameter, $2(1-\varepsilon)/\varepsilon$
ξ	=	Dimensionless axial distance, z/L
τ	=	Dimensionless time, tu / L

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