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ORIGINAL PAPER

Wavelet method to film-pore diffusion model for methylene blue adsorption onto plant leaf powders

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Abstract In this paper, we have developed an accurate and efficient Haar wavelet method to solve film-pore diffusion model. Film-pore diffusion model is widely used to determine study the kinetics of adsorption systems. To the best of our knowledge, until now rigorous wavelet solution has been not reported for solving film-pore diffusion model. The use of Haar wavelets is found to be accurate, simple, fast, flexible, convenient, and computationally attractive. The power of the manageable method is confirmed. It is shown that film-pore diffusion model satisfactorily describes the kinetics of methylene blue adsorption onto three low-cost adsorbents, Gauva, teak and gulmohar plant leaf powders, used in this study.

Keywords Methylene blue · Adsorption kinetics · Film-pore diffusion model · Low-cost adsorbents · Haar wavelet method

Abbreviations

A_s Total surface area of all the particles, m^2dm^{-3}
Bt Biot number ($Bt = k_f d_p/D_{eff}$), −
C_t Bulk concentration at time t, mg dm^{-3}

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C_s	Surface concentration, mg dm^{-3}
C_o	Initial bulk concentration, mg dm^{-3}
d_p	Diameter of particle, m
D_{eff}	Internal effective diffusivity, m^2 s^{-1}
K_L	Langmuir adsorption constant, $dm^3 mg^{-1}$
r	Radial position in the particle, m
R	Radius of the particle, m
q_e	Solid phase dye concentration at equilibrium, mg g^{-1}
q_i	Solid phase dye concentration at grid i at time t, mg g^{-1}
t	Time, s or min
V	Volume of solution, dm^3
ε	Particle porosity, –
ρ_p	Particle density, kg m^{-3}
k_f	External film transfer coefficient, m s^{-1}

1 Introduction

Adsorption process has been proven to be one of the highly efficient methods for removal of colors, odors, and organic and inorganic pollutants emanating from various industrial processes. Large amounts of dyes are used by textile industry and a significant portion of these dyes are not consumed in the process and therefore let out with the effluent. As the cost of commercial adsorbents is too high, interest for using low-cost adsorbents for removal of dyes from textile effluents is continuously growing. A recent survey indicates that, in India, on an average fresh water consumed and effluent generated per kg of finished textile are 175 and 125 L respectively [1]. The presence of dyes in aqueous effluents is highly objectionable as this affects the photosynthetic activity in receiving water body by reducing/preventing light penetration. As the dyes are recalcitrant in nature it is difficult to treat them in conventional biological treatment plant [2,3]. Various researchers have worked on biological degradation of dyes. But, very often, the metabolic intermediates are found to be more toxic than the original compound [4]. Therefore, identification of low-cost adsorbents is given more attention by the researchers recently as commercial adsorbents like activated carbon are too costly. Few recent studies investigating application of low cost adsorbents are: jackfruit peel [5], pine apple stem [6], Ashoka (Saraca asoca) leaf powder [7], pomelo peel [8], Chara contraria [9], groundnut shell powder [10], broad been peels [11] etc.

Adsorption of dye is complex process involving one or more of the following consecutive steps (i) diffusion of dye molecules across the external liquid film surrounding the solid particles, (ii) adsorption and desorption on the external surface of the particle, (iii) internal diffusion of dye within the particle either by pore diffusion, or surface diffusion or both and (iv) adsorption and desorption on the internal surface of the particle. Since adsorption is a surface phenomena and majority of the adsorbents used are porous, external and internal resistances to the mass transfer of the solute play major role in controlling the rate of adsorption. In order determine the rate controlling step and to understand the adsorption mechanism it is necessary to determine external mass transfer coefficient and internal pore diffusivity. Simplified single resistance models are available to predict external film transfer coefficients. These are robust models, efficient for quick estimation of mass transfer parameters mentioned above. However, accurate values of the parameters can only be obtained using more rigorous two resistance models. Film pore diffusion model (FPDM) was employed successfully to describe the kinetics of methylene blue adsorption onto GLP, TLP and GUL. Diffusion based kinetic models are too complex and require rigorous solution methods. For many of the diffusion models pure analytical solution are not possible. In our previous paper we had employed method of lines to solve film-pore diffusion model and had shown that Film-pore model could describe the kinetics of adsorption of MB onto GLP, TLP and GUL [1]. In this work, we have proposed a Haar solution to film-pore diffusion model.

As a powerful mathematical tool, wavelet analysis has been widely used in image digital processing, quantum field theory, numerical analysis and many other fields in recent years. The Haar transform is one of the earliest examples of what is now known as a dyadic, compact, orthonormal wavelet transform. The Haar function is an odd rectangular pulse pair. Therefore, it is the simplest and oldest orthonormal wavelet with compact support. Several definitions of the Haar functions and various generalizations have been published in literature. Haar functions appear to be highly attractive in variety of applications including image coding, binary logic design and edge extraction.

Chen and Hsiao [12] first derived a Haar operational matrix for the integrals of the Haar function vector and demonstrated the application of Haar analysis in dynamic systems. Then Hsiao [13], who first proposed a Haar product matrix and a coefficient matrix, laid down the pioneer work in state analysis of linear time delayed systems via Haar wavelets. In order to take the advantages of the local property, several authors had used the Haar wavelet to solve the differential and integral equations [14–18]. Lepik [19–21] had solved higher order as well as nonlinear ODEs and some nonlinear evolution equations by Haar wavelet method. Hariharan et al. [22–25] have introduced the solution of Fisher's equation, Cahn-Allen equation, Convection-diffusion equations and some nonlinear parabolic equations by the Haar wavelet method.

The fundamental idea of Haar wavelet method is to convert the problem of solving for the one-dimensional differential equation with constant coefficients, which satisfies the boundary conditions and initial conditions in to a group of algebraic equations, which involves a finite number of variables.

2 Materials and methods

Detailed development of FPDM is described earlier by McKay et al. [26,27]. Solution of FPDM by method of lines is described in our previous paper [1]. In the present paper development of Haar solution is described in detailed and the results are compared with our previous solution.

2.1 Haar wavelet and its properties

2.1.1 Haar wavelet

Haar wavelet was a system of square wave; the first curve was marked up as $h_0(t)$, the second curve marked up as $h_1(t)$ that is

$$h_0(x) = \begin{cases} 1, & 0 \le x < 1 \\ 0, & otherwise \end{cases} \tag{1}$$

$$h_1(x) = \begin{cases} 1, & 0 \le x < 1/2, \\ -1, & 1/2 \le x < 1, \\ 0, & otherwise, \end{cases} \tag{2}$$

where $h_0(x)$ is scaling function, $h_1(x)$ is mother wavelet. In order to perform wavelet transform, Haar wavelet uses dilations and translations of function, i.e. the transform make the following function.

$$h_n(x) = h_1\left(2^j x - k\right), \quad n = 2^j + k, j \ge 0, 0 \le k < 2^j. \tag{3}$$

2.1.2 Function approximation

Any square integrable function $y(x) \in L^2\,[0, 1)$ can be expanded by a Haar series of infinite terms

$$y(x) = \sum_{i=0}^{\infty} c_i h_i(x), \quad i \in \{0\} \cup N, \tag{4}$$

where, the Haar coefficients c_i are determined as, $c_0 = \int_0^1 y(x)h_0(x)dx$, $c_n = 2^j \int_0^1 y(x)h_i(x)dx,\ i = 2^j + k, j \ge 0, 0 \le k < 2^j, x \in [0, 1)$ such that the following integral square error ε is minimized:

$$\varepsilon = \int_0^1 \left[y(x) - \sum_{i=0}^{m-1} c_i h_i(x) \right]^2 dx, \quad m = 2^j, j \in \{0\} \cup N. \tag{5}$$

Usually, the series expansion contains infinite terms for smooth$y(x)$. If $y(x)$is piecewise constant by itself, or may be approximated as piecewise constant during each subinterval, then $y(x)$ will be terminated at finite m terms, that is

$$y(x) = \sum_{i=0}^{m-1} c_i h_i(x) = c_{(m)}^T h_{(m)}(x) \tag{6}$$

where the coefficients $c_{(m)}^T$ and the Haar function vector $h_{(m)}(x)$are defined as $c_{(m)}^T = [c_0, c_1, \ldots, c_{m-1}]$ and $h_{(m)}(x) = [h_0(x), h_1(x), \ldots, h_{m-1}(x)]^T$ where 'T' means transpose and $m = 2^j$.

The first four Haar function vectors, which $x = n/8$, $n = 1, 3, 5, 7$ can be expressed as follows

$$h_4(1/8) = [1, 1, 1, 0]^T, \quad h_4(3/8) = [1, 1, -1, 0]^T,$$
$$h_4(5/8) = [1, -1, 0, 1]^T, \quad h_4(7/8) = [1, -1, 0, -1]^T,$$

which can be written in matrix form as

$$H_4 = [h_4(1/8), h_4(3/8), h_4(5/8), h_4(7/8)]$$

$$H_4 = \begin{bmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & -1 & -1 \\ 1 & -1 & 0 & 0 \\ 0 & 0 & 1 & -1 \end{bmatrix},$$

In general, we have

$$H_m = [h_m(1/2m), h_m(3/2m), \ldots, h_m(2m-1)/2m],$$

where $H_1 = [1]$, $H_2 = \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}$. The collocation points are identified as $x_l = (2l-1)/2m$, $l = 1, 2, \ldots, m$. In application, in order to avoid dealing with impulse function, integration of the vector $h_m(x)$ given by

$$\int_0^x h_m(t)dt \approx P_m h_m(x), \quad x \in [0, 1], \tag{7}$$

where P_m is the $m \times m$ operational matrix and is given by $P_{(m)} = \frac{1}{2m}\begin{pmatrix} 2mP_{(m/2)} & -H_{(m/2)} \\ H^{-1}_{(m/2)} & O \end{pmatrix}$

where O is a null matrix of order $\frac{m}{2} \times \frac{m}{2}$.

$$P_1 = [1/2],$$

$$P_2 = \frac{1}{4}\begin{pmatrix} 2 & -1 \\ 1 & 0 \end{pmatrix}, \quad P_4 = \frac{1}{16}\begin{bmatrix} 8 & -4 & -2 & -2 \\ 4 & 0 & -2 & 2 \\ 1 & 1 & 0 & 0 \\ 1 & -1 & 0 & 0 \end{bmatrix},$$

$$P_8 = \frac{1}{64}\begin{bmatrix} 32 & -16 & -8 & -8 & -4 & -4 & -4 & -4 \\ 16 & 0 & -8 & 8 & -4 & -4 & 4 & 4 \\ 4 & 4 & 0 & 0 & -4 & 4 & 0 & 0 \\ 4 & 4 & 0 & 0 & -4 & 4 & 0 & 0 \\ 1 & 1 & 2 & 0 & 0 & 0 & 0 & 0 \\ 1 & 1 & -2 & 0 & 0 & 0 & 0 & 0 \\ 1 & -1 & 0 & 2 & 0 & 0 & 0 & 0 \\ 1 & -1 & 0 & -2 & 0 & 0 & 0 & 0 \end{bmatrix}$$

Table 1 Comparison between Haar wavelet method (HWM) and method of lines (MOL) by obtaining the mass transfer coefficients using film-pore diffusion model adsorption of MB onto GLP and $m = 16$, $t = 10$ s

Temperature (K)	C_0(mg dm^{-3})	k_f(ms^{-1})		D_{eff} (m^{2}s^{-1})		Error	
		MOL (M)	HWM (H)	MOL (M)	HWM (H)	E_M	E_H
303	50	1.00×10^{-6}	3.00×10^{-6}	1.74×10^{-13}	2.54×10^{-13}	1.197	0.938
	100					0.140	0.129
	150					0.935	0.824
	200					1.610	1.102
313	50	1.71×10^{-6}	3.45×10^{-6}	6.46×10^{-13}	9.26×10^{-13}	1.462	1.221
	100					1.120	0.927
	150					1.267	1.016
	200					7.570	5.112
323	50	4.27×10^{-6}	4.75×10^{-6}	3.11×10^{-13}	5.31×10^{-13}	0.856	0.284
	100					0.160	0.000
	150					2.168	1.208
	200					3.164	1.016

E_M Error by method of lines
E_H Error by Haar wavelet method

It should be noted that calculations for $P_{(m)}$ and $H_{(m)}$ must be carried out only once; after that they will be applicable for solving whatever differential equations.

The fast capability of HT should be impressive (See Refs. [12,19–25]). Table 1 shows the numbers of additions and multiplications for these three transforms.

In practical applications, a small number of terms increases the calculation speed and saves memory storage while a large number of terms improve resolution accuracy. Therefore, it is essential to have a trade-off between calculation speed, memory saving, and the resolution accuracy and this has been considered in the analysis.

2.1.3 Method of solution

Before, discussing the method a solution a brief introduction of film-pore diffusion model is given. Film-pore diffusion model assumes that both external film and internal pore diffusion resistances are significant and play a role in controlling the mass transfer. Thus, the governing equations are:

(i) Assuming linear driving force the rate of external mass transfer is given by:

$$\frac{dC_t}{dt} = -k_f \frac{A_s}{V} (C_t - C_s) \tag{8}$$

(ii) Within the pore diffusion of solutes follows Fick's law of diffusion. Following equation is attained by making a mass balance of dye in a spherical:

$$\varepsilon \frac{\partial C_i}{\partial t} + \rho_p \frac{\partial q_i}{\partial t} = D_{eff} \left[\frac{\partial^2 C_i}{\partial r^2} + \frac{1}{r} \frac{\partial C_i}{\partial r} \right] \tag{9}$$

Corresponding initial condition and boundary conditions are:

$$\text{I.C.: At t} = 0,\ \mathrm{C_i} = 0 \quad \text{for } 0 \le \mathrm{r} \le \mathrm{R} \tag{10}$$

$$\text{B.C.1: } \frac{\partial C_i}{\partial r} = 0 \quad \text{at r} = 0. \tag{11}$$

$$\text{B.C.2: } k_f(C_t - C_s) = D_{\mathit{eff}} \left.\frac{\partial C_i}{\partial r}\right|_{r=R} \tag{12}$$

(iii) Solid phase concentration at any radial location may be expressed as function of aqueous phase concentration at that location as follows:

$$\mathrm{q_i} = \mathrm{f(C_i)} \tag{13}$$

Assuming equilibrium within the pore eq. (13) is described by relevant isotherm expression of the system. Substituting Eq. 13 in Eq. 9 we get:

$$\varepsilon\frac{\partial C_i}{\partial t} + \rho_p\frac{\partial f(C_i)}{\partial t} = D_{\mathit{eff}}\left[\frac{\partial^2 C_i}{\partial r^2} + \frac{1}{r}\frac{\partial C_i}{\partial r}\right] \tag{14}$$

Since the system follow Langmuir isotherm [1],

$$q_i = f(C_i) = \frac{q_e K_L C_i}{1 + K_L C_i} \tag{15}$$

Following dimensionless variables were defined to convert the above equations into dimensionless form,

$$\left.\begin{array}{l} Z = \frac{r}{R}; \quad \overline{C_i} = \frac{C_i}{C_o}; \quad \overline{C_t} = \frac{C_t}{C_o}; \quad B_i = \frac{k_f R}{D_{\mathit{eff}}}; \end{array}\right\} \tag{16}$$

After substituting the dimensionless variables in Eq. (14) can be rewritten as follows:

$$\frac{\partial \overline{C_i}}{\partial \tau} = A(\overline{C_i})\left[\frac{\partial^2 \overline{C_i}}{\partial Z^2} + \frac{1}{Z}\frac{\partial \overline{C_i}}{\partial Z}\right] \tag{17}$$

where,

$$A(\overline{C_i}) = \frac{1}{\left(\varepsilon_p + \left(\frac{q_h \rho_p}{C_o}\right)\left(\frac{1+bC_o}{(1+bC_o\overline{C_i})^2}\right)\right)} \tag{18}$$

Consider the equation

$$\dot{\bar{C}}_i\,(Z,\tau) = A(\bar{C}_i)\left[\bar{C}_i'' + \left(\frac{1}{Z}\right)\bar{C}_i'\right] \tag{19}$$

Let us divide the interval (0, 1] into N equal parts of length $\Delta\tau = (0, 1]/N$ and denote$\tau_s = (s-1)\Delta t,\, s = 1, 2, \ldots N$. We assume that $\dot{C}''(Z,\tau)$can be expanded in terms of Haar wavelets as formula

$$\dot{\bar{C}}_i''(Z,\tau) = \sum_{i=0}^{m-1} c_s(i)h_i(Z) = c_{(m)}^T h_{(m)}(Z) \tag{20}$$

where . and ′ means differentiation with respect to t and x respectively, the row vector $c_{(m)}^T$ is constant in the subinterval $\tau \in (\tau_s, \tau_{s+1}]$

Integrating formula (20) with respect to τ from τ_s to τ and twice with respect to Z from 0 to x, we obtain

$$\bar{C}_i''(Z,\tau) = (\tau-\tau_s)c_{(m)}^T h_{(m)}(Z) + \bar{C}_i''(Z,\tau_s) \tag{21}$$

$$\begin{aligned}\bar{C}_i(Z,\tau) = (\tau-\tau_s)c_{(m)}^T Q_{(m)}h_{(m)}(Z) + \bar{C}_i(Z,\tau_s) - \bar{C}_i(0,\tau_s)\\ +Z[\bar{C}_i'(0,\tau) - \bar{C}_i'(0,\tau_s)] + \bar{C}_i(0,\tau)\end{aligned} \tag{22}$$

$$\bar{C}_i'(Z,\tau) = (\tau-\tau_s)c_{(m)}^T P_{(m)}h_{(m)}(Z) + \bar{C}_i'(Z,\tau_s) - \bar{C}_i(0,\tau_s) + \bar{C}_i'(0,\tau) \tag{23}$$

$$\dot{\bar{C}}_i(Z,\tau) = c_{(m)}^T Q_{(m)}h_{(m)}(Z) + Z\dot{\bar{C}}_i(0,\tau) + \dot{\bar{C}}_i(0,\tau) \tag{24}$$

with the boundary conditions, we obtain

$$\begin{aligned}\bar{C}_i(0,\tau_s) = g_0(\tau_s), \quad \bar{C}_i(1,\tau_s) = g_1(\tau_s)\\ \dot{\bar{C}}_i(0,\tau) = g_0'(\tau), \quad \dot{\bar{C}}_i(1,\tau) = g_1'(\tau)\end{aligned}$$

Putting $x = 1$in formulae (21)–(24), we have

$$\begin{aligned}\bar{C}_i'(0,\tau) - \bar{C}_i'(0,\tau_s) = -(\tau-\tau_s)c_{(m)}^T P_{(m)}h_{(m)}(Z) + g_1(\tau)\\ -g_0(\tau) - g_1(\tau_s) + g_0(\tau_s)\end{aligned} \tag{25}$$

$$\dot{\bar{C}}_i'(0,\tau) = g_1'(\tau) - c_{(m)}^T Q_{(m)}h_{(m)}(Z)f - g_0'(\tau) \tag{26}$$

where the vector f is defined as

$$f = [1, \underbrace{0, \ldots, 0}_{(m-1)elements}]^T$$

Substituting formulae (25) and (26) into formulae (21)–(24), and discretizising the results by assuming $Z \to Z_l$, $\tau \to \tau_{s+1}$ we obtain

$$\bar{C}_i''(Z_l, \tau_{s+1}) = (\tau_{s+1} - \tau_s)c_{(m)}^T h_{(m)}(Z_l) + \bar{C}_i''(Z_l, \tau_s) \tag{27}$$

$$\begin{aligned}\bar{C}_i'(Z_l, \tau_{s+1}) = {} & (\tau_{s+1} - \tau_s)c_{(m)}^T Q_{(m)}h_{(m)}(Z_l) + \bar{C}_i(Z_l, \tau_s) - g_0(\tau_s) + g_0(\tau_{s+1}) \\ & + Z_l\big[-(\tau_{s+1} - \tau_s)c_{(m)}^T P_{(m)} f + g_l(\tau_{s+1}) \\ & - g_0(\tau_{s+1}) - g_1(\tau_s) + g_0(\tau_s)\big]\end{aligned} \tag{28}$$

$$\dot{\bar{C}}_i(Z_l, \tau_{s+1}) = c_{(m)}^T Q_{(m)}h_{(m)}(Z) + Z\dot{\bar{C}}_i(0, \tau) + \dot{\bar{C}}_i(0, \tau) \tag{29}$$

$$\begin{aligned}\bar{C}_i(Z_l, \tau_{s+1}) = {} & c_{(m)}^T Q_{(m)}h_{(m)}(Z) + g_0'(\tau_{s+1}) + Z_l\Big[-c_{(m)}^T P_{(m)} f \\ & + g_1'(\tau_{s+1}) - g_0'(\tau_{s+1})\Big]\end{aligned} \tag{30}$$

In the following the scheme

$$\dot{\bar{C}}_i(Z_l, \tau_{s+1}) = A(\bar{C}_i)\left[\bar{C}_i''(Z_l, \tau_{s+1}) + \frac{1}{Z}\bar{C}_i'(Z_l, \tau_{s+1})\right] \tag{31}$$

which leads us from the time layer τ_s to τ_{s+1} is used.

Substituting formulae (27)–(30) into the formula (31), we gain

$$\begin{aligned}& c_{(m)}^T Q_{(m)}h_{(m)}(Z_l) + Z_l\left[-c_{(m)}^T P_{(m)} f + g_1'(\tau_{s+1}) - g_0'(\tau_{s+1})\right] + g_0'(\tau_{s+1}) \\ & = A(\bar{C}_i)\begin{bmatrix}(\tau_{s+1}-\tau_s)c_{(m)}^T h_{(m)}(Z_l)+(\tau_{s+1}-\tau_s)c_{(m)}^T Q_{(m)}h_{(m)}(Z_l) \\ +\bar{C}_i(Z_l, \tau_s)-g_0(\tau_s)+g_0(\tau_{s+1}) \\ +Z_l[-(\tau_{s+1}-\tau_s)c_{(m)}^T P_{(m)} f+g_l(\tau_{s+1}) - g_0(\tau_{s+1})-g_1(\tau_s)+g_0(\tau_s)]\end{bmatrix}\end{aligned}$$

From the above formula, the wavelet coefficients $c_{(m)}^T$ can be successively calculated.

Here $A(\bar{C}_i)$ are constants (linear) and $\in = \mathbf{0.5}$, $\rho = 500$.

Table 1 gives a comparison of Haar wavelet solutions and method of lines. It is evident that Haar wavelet solutions are better than that of the method of lines. Value of absolute error decreased when m was increased. The results show that combining with wavelet matrix, the method in this paper can be effectively used in numerical calculus for constant coefficient differential equations, and that the method is feasible. At the same time with the sparse nature of Haar wavelet matrix, compared with the method of lines [1], using the above method can greatly reduce the computation and from the above results, we can see that the numerical solutions are in good agreement with exact solution. The power of the manageable method is thus confirmed.

All the numerical experiments presented in this section were computed in double precision with some MATLAB codes on a personal computer System with Processor Intel(R) Core$^{(TM)}$ 2 Duo CPU T5470 @ 1.60 GHz (2CPUs) and 1 GB RAM.

3 Conclusion

In this paper FPDM model equations had been solved by the Haar wavelets method. The external film transfer coefficients and internal pore diffusivities were obtained. Comparing the magnitude of the errors, it can be seen that the Haar wavelet method could predict the concentration decay curve for the adsorption of methylene blue onto TLP, GUL and GLP very closely. The sparseness in Haar wavelets based operational matrices gives precise accuracy in solving numerical equations by Haar wavelet method. According to this scheme the spatial operators are approximated by the Haar wavelet method and the time derivation operators by the finite difference method. We transform the FPDM model equations into a linear system of algebraic equations which is easily and efficiently to solve. We found that Haar wavelets method had good approximation effect by comparing with method of lines of the FPDM model equations at the same time. It is worth mentioning that Haar solution provides excellent results even for small values of $m (m = 16)$. For higher values of m(i.e., $m = 32, m = 64, m = 128, m = 256$), we can obtain the results closer to the real values. The method with far less degrees of freedom and with smaller CPU time provides better solutions than classical ones.

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