Numerical Modeling of Industrial-Scale Pulsed Bed Adsorber for Colorant Removal in Sugar Refining

Terasut Sookkumnerd

1 School of Chemical Engineering, Suranaree University of Technology, Nakhon Ratchasima 30000, THAILAND

Corresponding author: terasut@gmail.com

Abstract. Because pulsed bed adsorbers are preferred when the high fluid flow rate is expected, pulsed bed adsorbers are used in many industries; for example, waste water treatment and food industries. However, few attempts have been focused on modeling of pulsed bed adsorbers. In this paper, the mathematical model of an industrial-scale pulsed bed adsorber has been developed. By using the data on adsorption equilibrium constant, mass transfer and axial dispersion in a lab-scaled fixed-bed adsorber, the numerical analysis of an industrial-scale pulsed bed adsorber for colorant removal in sugar refining process is performed and the numerical results are analyzed to gain the better understanding of this unit operation. The optimum cycle time depended on the fraction of adsorbent being removed and replaced in each pulse; the optimum cycle time increases from 4 hours at 5 % pulse to 12 hours at 14% pulse. In addition, the fluctuation of feed flow rate would affect the performance of pulsed bed adsorbers; for example, the 10% increase of flow rate would make the colorant in the syrup at the outlet higher than its specification. The finding of this paper would allow engineers to optimize an industrial-scale pulsed bed adsorber more efficiently.

1. Introduction

Pulsed-bed adsorbers are adsorbers that the liquor is fed at the bottom of the bed of adsorbent similarly to the fixed-bed but the spent adsorbent is periodically removed from the bottom while the new or on-site regenerated adsorbent is added to the top at the specified time interval so that the height of bed remains unchanged. This type of operation is referred as the pulse operation and the period of the adsorbent removal and the replacement of new adsorbent is called the cycle time [1-3]. This pulse operation simulated the countercurrent movement of spent adsorbent and liquor; hence, the efficiency of mass transfer is greater when comparing with the fixed-bed adsorbers. Usually, the pulsed-bed adsorbers are preferred when the usage rate of adsorbent and the liquor throughput are relatively high. In order to make the pulsed bed adsorbers cost competitive, the spent adsorbent is usually regenerated on-site with a limited feedback control, if any, on the effectiveness of regeneration [2, 4]. Presently, pulsed-bed adsorbers have been used in wastewater treatment and recently been applied in sugar refining where the pulsed bed adsorbers of granulated activated carbon (GAC) are applied to remove the colorants in sugar syrup. Among several operating parameters, cycle time of pulse and percentage of the bed removed is frequently used as the operating parameters for controlling pulsed-bed adsorbers. Generally, 5-15% of adsorbent in the bed is removed and replaced with new adsorbent in
order to minimize its effect on hydrodynamics and dynamics of the adsorbers while the cycle time is set at 12 - 24 hours in order to minimize the abrasion of adsorbent [1]. Although the pulsed bed adsorbers, which are widely used in industrials, the information on the effect of cycle time, liquor flow rate and regeneration efficiency of adsorbent on the performance of pulsed bed adsorbers is relatively scarce. Consequently, it is the aim of this article to explore the effects of operating parameter on the operation of industrial-scale pulsed bed adsorbers by developing a mathematical model of industrial-scaled pulsed bed adsorbers and then applying the model to investigate the effect of operating parameters; namely, cycle time and flow rate on the performance of an industrial scale pulse bed adsorber, numerically. It is worthwhile to note that, according to the author’s knowledge, there is no research article published in the literature that focus on the mathematical modeling of pulsed-bed adsorbers.

In this article, the adsorption equilibrium constant or K and the overall mass transfer coefficient according to the linear driving force model or k was first evaluated from the breakthrough curve of a laboratory GAC fixed-bed adsorber published in the literature [5]. Secondly, the tortuosity of GAC (τ) was calculated from the overall mass transfer coefficient measured and the correlation for film mass transfer (k_f) and axial dispersion coefficient (D_L) obtained from the fitting of breakthrough curve. Thirdly, the mathematical model of an industrial-scale pulsed bed adsorber was developed and solved based on the assumption of ideal plug flow of adsorbent during the pulse. Finally, the effect of fraction of adsorbent removed and replace during the pulsed operation and liquor flow rate on the pulsed bed adsorber was investigated.

2. Theory and method.

2.1. Breakthrough of fixed-bed Adsorber

Based on the assumptions of linear adsorption isotherm, Ruthven showed that the breakthrough of a fixed-bed adsorber can be correlated from the following equation [6]

\[
\frac{C}{C_0} = \frac{1}{2} \text{erfc} \left( \frac{L}{\sqrt{8} \sqrt{\tau - \sqrt{8}}} \right) \quad (1)
\]

where

\[
\tau = k \left( \frac{L}{v} \right) \quad (2)
\]

and

\[
\xi = \frac{kk_f(1+\varepsilon)}{\varepsilon} \quad (3)
\]

C was the concentration at the outlet, C_0 was the feed concentration, t was the time, L was the height of adsorption bed, v was the liquor interstitial velocity, ε was the bed porosity and τ and ξ are dimensionless variables.

2.2. Modeling of pulsed-bed Adsorbers

By modifying the initial conditions of the mathematical model of a fixed-bed adsorber for plug flow and linear driving force model [6], the mathematical model for an industrial-scale pulsed bed adsorber are:

\[
\frac{\partial (c/c_0)}{\partial t} = \left( 1+\varepsilon \right) \frac{\partial (\tilde{q}/c_0)}{\partial t} + \frac{\partial (c/c_0)}{\partial x} \quad (4)
\]

\[
\frac{\partial (\tilde{q}/c_0)}{\partial t} = kK \left( C/C_0 - C_{eq}/C_0 \right) \quad (5)
\]

The initial condition at the first cycle are;

\[
C(t=0,x)=0 \quad (6)
\]

\[
\tilde{q}(t=0,x)=0 \quad (7)
\]

\[
C(t,x=0)=C_0 \quad (8)
\]
For the other cycle, the initial condition at the beginning of each cycles are:

\[
C(t=Nt_{cycle}, z) = \begin{cases} 
0 & \text{if } z \geq (1-X_{\text{pulse}})L \\
C(t=Nt_{cycle}, z-X_{\text{pulse}}L) & \text{if } z < (1-X_{\text{pulse}})L
\end{cases}
\]

\[
\bar{q}(t=Nt_{cycle}, z) = \begin{cases} 
0 & \text{if } z \geq (1-X_{\text{pulse}})L \\
\bar{q}(t=Nt_{cycle}, z-X_{\text{pulse}}L) & \text{if } z < (1-X_{\text{pulse}})L
\end{cases}
\]

where \( k \) was the overall mass transfer coefficient, \( K \) was the adsorption equilibrium constant, \( \bar{q} \) was the average concentration in the adsorbent, \( C_{eq} \) was the concentration in liquid at equilibrium with \( \bar{q} \), \( N \) was the number of cycle, \( t_{cycle} \) was the cycle time, \( z \) was the distance from the inlet, \( X_{\text{pulse}} \) was the fraction of bed that was removed and replaced by the new adsorbent in each cycle and \( L \) was the height of the pulsed bed.

### 2.3. Numerical analysis of pulsed-bed Adsorbers

By fitting equation (1) with the breakthrough curve of sugar colorants of a fixed-bed Reactor reported by Cortes [5], the adsorption equilibrium constant (\( K \)) and the overall mass transfer coefficients were obtained. The fitting was carried on by Solver in Microsoft Excel.

Once \( k \) was obtained, the correlation of the external (film) mass transfer coefficient or \( k_f \) [6] and axial dispersion coefficient or \( D_L \) [7] were used to estimate the pore diffusion of sugar colorants (\( D_p \)) and totoursity (\( \tau_p \)) from [6]

\[
D_p = \frac{k_f^2/15 \epsilon_p}{\frac{1}{k} \frac{D_M (1-\epsilon_p)}{v^2 \epsilon_p}}
\]

\[
\tau_p = D_p / D_M
\]

where \( R_p \) was the radius of particle, \( \epsilon_p \) was the particle porosity and \( D_M \) was the molecular diffusivity of sugar colorants in water. Note that the molecular diffusivity of pullulan [8] was used to estimate the molecular diffusivity of sugar colorants since both are similar in origin and the particle porosity was estimated from the total porosity of bed reported by Cortes [5] assuming the typical value of bed porosity of 0.38 [9].

### 2.4. Estimating parameters of fixed-bed Adsorber from breakthrough curve

The typical of pulsed bed adsorber with the diameter of 4 m and the height of 15 m was used in the numerical analysis. The typical flow rate of sugar syrup of 87.90 m³/hr was assumed. The superficial velocity at the flow rate was 0.001943 m/s which was comparable to the experiment run by Cortes [5] that the experimental results was used to estimate the parameters; hence, the mechanism in both cases should be the same. The color of feed syrup was typically 700 ICUMSA and the color of sugar syrup at the outlet was controlled at 100 ICUMSA. The ratio of the color at the outlet to that at the inlet will be used as the ratio of concentration at the outlet to that at the inlet or \( C/C_0 \).

The numerical method of lines was used to solve equation (4) and equation (5) with the initial conditions shown in equation (6) to equation (10). The 5-point biased upwinding method was used to approximate the spatial derivative [10] and the resulted ordinary differential equations were solved by ode solver in Scilab which is the open source alternative to Matlab. The step sizes in spatial direction of 0.008442 m and of 0.25 m were appeared to generate the numerical results with acceptable accuracy.

### 3. Results and discussions

#### 3.1. Fitting of breakthrough curve and evaluate parameters from breakthrough curve

Based on the breakthrough reported by Cortes [5], Equation (1) was used to estimated \( k \) and \( K \) for the flow rate from 0.023 ml/min to 0.043 ml/min as illustrated in table 1.
Table 1. Adsorption Equilibrium Constant and Overall Mass Transfer Coefficient from Breakthrough Curve.

| Flow rate (ml/min) | k (min)    | K          |
|--------------------|------------|------------|
| 23                 | $6.03 \times 10^{-3}$ | 63.41      |
| 33                 | $6.35 \times 10^{-3}$ | 63.41      |
| 43                 | $8.27 \times 10^{-3}$ | 63.41      |

From the values of overall mass transfer coefficient, the correlation of $k_f$ [6] and the correlation of $D_L$ [7], the effective diffusivity in porous activated carbon can be evaluated as shown in table 2.

Table 2. Film Mass Transfer Coefficients and Pore Diffusivity of Colorants in Sugar Syrup.

| Flow rate (ml/min) | $k_f$ (min) | $D_p$ (m$^2$/s) |
|--------------------|-------------|------------------|
| 23                 | $8.06 \times 10^{-2}$ | $1.80 \times 10^{-10}$ |
| 33                 | $9.07 \times 10^{-2}$ | $1.89 \times 10^{-10}$ |
| 43                 | $9.90 \times 10^{-2}$ | $2.46 \times 10^{-10}$ |

From the average value of $D_p$ and the value of $D_M$, the value of totuosity of activated carbon was approximated to be 2.012 which was in the range of 2 – 10 reported for activated carbon [4].

Figure 1. The Experimental and Correlated Breakthrough Curve.

From the values of $D_p$, $D_L$ and $K$, the breakthrough curves of sugar colorants from a fixed-bed adsorber at different flow rate was shown in figure 1. From figure 1, it was obvious that the parameters obtained can describe the experimental results adequately especially at 33 and 43 ml/min.

3.2. Numerical analysis of an industrial-scale pulsed-bed Adsorber

From the values of parameters obtained from the experimental results of a fixed-bed adsorber, the breakthrough of a fixed-bed adsorber with the same dimension as a pulsed bed was shown in figure 2 along with the concentration profile in a pulsed bed adsorber in figure 3.
Figure 2. The Predicted Breakthrough Curve from a Fixed-bed Adsorber with the same Size as an Industrial-scale Pulsed Bed Adsorber.

Figure 3. The Concentration Profile in a Fixed-bed Adsorber with the same Size as an Industrial-scale Pulsed Bed Adsorber at different time.

Then, the mathematical model of an industrial-scale pulsed bed adsorber was solved and illustrated as shown in figure 4 to 8. From figure 4 to 8, it can be seen that the value of $C/C_0$ would reach the pseudo-steady state value after 40 cycles. Furthermore, if $t_{cycle}$ was less than the optimum cycle time, $C/C_0$ will be higher than the specification of processed syrup set at 0.1429. The optimum cycle time would depend on $X_{pulse}$. When $X_{pulse}$ was 0.05, the optimum cycle time was 4 hours and, when $X_{pulse}$ was 0.14, the optimum cycle time was 12 hours. Comparing with figure 3, the utilization of activated carbon will be more efficient if $X_{pulse}$ is less.

Figure 4. Predicted Performance of an Industrial-scale Pulsed Bed Adsorber for $X_{pulse} = 0.05$ and $t_{cycle} = 12$ hours.

Figure 5. Predicted Performance of an Industrial-scale Pulsed Bed Adsorber for $X_{pulse} = 0.05$ and $t_{cycle} = 6$ hours.

Figure 6. Predicted Performance of an Industrial-scale Pulsed Bed Adsorber for $X_{pulse} = 0.05$ and $t_{cycle} = 4$ hours.

Figure 7. Predicted Performance of an Industrial-scale Pulsed Bed Adsorber for $X_{pulse} = 0.14$ and $t_{cycle} = 15$ hours.
Figure 8. Predicted Performance of an Industrial-scale Pulsed Bed Adsorber for $X_{\text{pulse}} = 0.14$ and $t_{\text{cycle}} = 12$ hours.

Then, the effect of flow rate on the optimum cycle time for this industrial-scale pulsed bed adsorber was investigated. It is assumed that the flow rate varied from 70.32, 79.11, and 87.90 m$^3$/hr or -20% and -10% from the typical flow rate of 87.90 m$^3$/hr. The analysis showed that if $X_{\text{pulse}}$ was still at 0.05, the optimum cycle time would be changed to 13 hours when the flow rate decreased by 10% and to 15 hours when the flow rate decreased by 20%.

From figure 9 and figure 10, it was obvious that the increase in syrup flow rate would disturb the original pseudosteady state and make the process reach the new pseudosteady state. Consequently, the cycle time set for the industrial-scale pulsed bed adsorber must be adjusted so that it can tolerate the fluctuation of syrup feed flow rate. Alternatively, the feed forward model based predictive control can be implemented so that the utilization of adsorbent is maximized.

Figure 9. Dynamic Response of an Industrial-scale Pulsed Bed Adsorber for $X_{\text{pulse}} = 0.14$ and $t_{\text{cycle}} = 15$ hours. Originally, the flow rate was set at 70.32 m$^3$/min; then the flow rate was suddenly increased by 10% for 120 min.

Figure 10. Dynamic Response of an Industrial-scale Pulsed Bed Adsorber for $X_{\text{pulse}} = 0.14$ and $t_{\text{cycle}} = 15$ hours. Originally, the flow rate was set at 70.30 m$^3$/min; then the flow rate was suddenly increased by 20% for 120 min.
4. Conclusions
In conclusion, the mathematical model for an industrial-scale pulsed bed adsorber was developed. By using the data of breakthrough curve of sugar colorant published in the literature, the adsorption and mass transfer parameters of a fixed-bed adsorber were evaluated; the obtained values agreed with the reported ranges in the literature. The parameters were then used in the numerical analysis of an industrial-scale pulsed bed adsorber. In the analysis, it was found that the optimum cycle time depended on the fraction of adsorbent being removed and replaced in each pulse. Generally, the higher the fraction being pulsed, the longer the optimum cycle time. When the fraction being pulsed was kept constant, the decreasing flow rate would yield the longer optimum cycle time. Hence, the feed forward of an industrial-scale pulsed bed on flow rate should maximize the utilization of adsorbent. Further studies on this aspect will be published subsequently.

5. References
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