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## Study of Nonlinear Wave Propagation Theory. II. Interference Phenomena of Single-Component Dye Adsorption Waves

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### ABSTRACT

Yellow acid dye was adsorbed from aqueous solution by granular activated carbon packed in a column. The feed concentration was kept constant for a period of time and then switched to another level to simulate the concentration variation in wastewater treatment processes. The resulting breakthrough curves were experimentally measured and theoretically predicted. Explicit equations and an algorithm were developed based on the wave interference theory to predict the column breakthrough curves of single-component adsorption processes with step change in the feed concentration. The experimental results show that the wave interference theory predicts the column breakthrough curves satisfactorily.

*Key Words.* Wave propagation; Fixed-bed dynamics; Dye; Activated carbon; Adsorption; Wave interference

### INTRODUCTION

Fixed-bed operations are widely used in chemical processes and pollution control processes such as separating ions by ion-exchange bed or removing toxic organic compounds by carbon adsorption bed. A powerful tool that can be used to predict fixed-bed column dynamics without tedious column tests and sophisticated mass-transfer modeling is the nonlinear wave propagation theory (1, 2). The concept of “wave” was first introduced by Klotz (3), and the

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wave sharpening tendency, self-sharpening or nonsharpening, was first discussed by Glueckauf and Coates (4). Following these pioneer studies, the chromatography theory was successfully established and applied for chemical analysis and preparation purposes. The initiation of the "coherence" concept by Helfferich (5, 6) made a great contribution to the multicomponent chromatography theory. The development history of the nonlinear wave propagation theory can be found in two excellent review papers (7, 8). This paper is not meant to reinvent the chromatography theory, but to develop more explicit expressions from the theory to analyze the experimental breakthrough data of the activated carbon adsorption process with feed composition change.

The textile industry discharges wastewater with high color, high suspended solids, and dissolved organics. Due to its variation in pH, temperature, flow rate, and contained pollutants, textile wastewater usually requires physico-chemical as well as biological treatments to meet the more stringent effluent standards. Besides chemical oxygen demand, color removal from textile wastewater is of primary concern. An effective method to remove color is to use an adsorbent such as activated carbon to adsorb the dye molecules and then remove the color from the wastewater (9–17). Although this direct addition of powdered activated carbon is quite efficient, the carbon loss as wasted sludge is rather significant. Granular activated carbon is usually used in the fixed-bed adsorption process because of ease of operation and no carbon loss problem. To design and operate a fixed-bed adsorption process successfully, the column dynamics must be understood; that is, the breakthrough and desorption curves under specific operating conditions must be predictable.

The design procedure for a fixed-bed process is quite straightforward and is well documented (18, 19). Using the nonlinear wave propagation theory, one can easily calculate the breakthrough volume for a specific feed concentration and design the fixed-bed volume with a proper choice of safety factor. However, the operating strategy of a fixed bed is rather complicated, especially when the feed concentration varies with operating time. Although the equalization tank in a wastewater treatment process can even the flow-rate variation, concentration variation is still inevitable. Therefore, the feed dye concentration to an activated carbon bed can vary with operating time. This variation of the feed concentration makes the column dynamics more complicated than the case of constant feed concentration.

In the language of wave propagation theory, the concentration wave velocity is defined as the variation of the traveling distance with time for a given concentration. Therefore, different concentrations possess different traveling velocities. If the traveling velocity for the first feed concentration is slower than that of the second feed concentration, then the first feed concentration will be caught by the second and this will result in wave interference phenomena. The general wave interference phenomena of multicomponent sys-



tems were examined in detail in a recent paper (20). This study focuses on applying the wave interference concept, developing more explicit expressions to predict the effluent histories of single-component adsorption systems, and verifying the predicted results with dye adsorption data.

## WAVE INTERFERENCE THEORY

The theoretical basis of this study is directly built upon the nonlinear wave propagation theory developed by Helfferich and his coworkers (1, 20). Consider a carbon bed presaturated with a very low dye concentration,  $C_P$ . At time zero, a feed stream with dye concentration  $C_{F1}$  enters the bed at linear velocity  $u_0$ . After operating for a while at  $t = t_0$ , the feed dye concentration switches to  $C_{F2}$  while the flow velocity remains at  $u_0$ . The governing equation to describe the column dynamics is

$$\rho \frac{\partial q}{\partial t} + \varepsilon \frac{\partial C}{\partial t} + u_0 \varepsilon \frac{\partial C}{\partial Z} = 0 \quad (1)$$

where  $C$  is the dye concentration in the mobile phase,  $q$  is the dye concentration in the stationary phase,  $\rho$  is the carbon bed density,  $\varepsilon$  is the void fraction of the bed,  $u_0$  is the linear velocity of the carrier fluid,  $t$  is the operating time, and  $Z$  is the distance from the inlet of the mobile phase. The initial condition for Eq. (1) is

$$\text{At } t = 0, \quad C = C_P \quad \text{and} \quad q = q_P$$

The boundary condition for Eq. (1) is

$$\begin{aligned} \text{At } Z = 0, \quad C = C_{F1} \quad \text{and} \quad q = q_{F1} \quad \text{for} \quad 0 < t < t_0 \\ C = C_{F2} \quad \text{and} \quad q = q_{F2} \quad \text{for} \quad t \geq t_0 \end{aligned}$$

The assumptions associated with Eq. (1) are:

- (1) The adsorption reaches local equilibrium.
- (2) No chemical reactions occur in the column.
- (3) Only mass transfer by convection is significant.
- (4) Radial and axial dispersions are negligible.
- (5) The flow pattern is ideal plug flow.
- (6) The temperature in the column is uniform and invariant with time.
- (7) The flow rate is constant and invariant with the column position.

The dye concentration in the stationary phase is related to that in the mobile phase by the Redlich–Peterson adsorption isotherm model (21):

$$q = \frac{AC}{B + C^M} \quad (2)$$



Although Eqs. (1) and (2) associated with the initial and boundary conditions can be solved for  $C = C(Z, t)$  by numerical methods, the cause-and-effect relation between the column dynamics and the feed concentration change cannot be clearly figured out. To illustrate the cause-and-effect relation, simple expressions based on the wave interference theory will be developed. Depending upon the relative magnitude of  $C_P$ ,  $C_{F1}$ , and  $C_{F2}$ , there are six different wave interference cases as shown in Fig. 1, but only two of them will be considered in this paper.

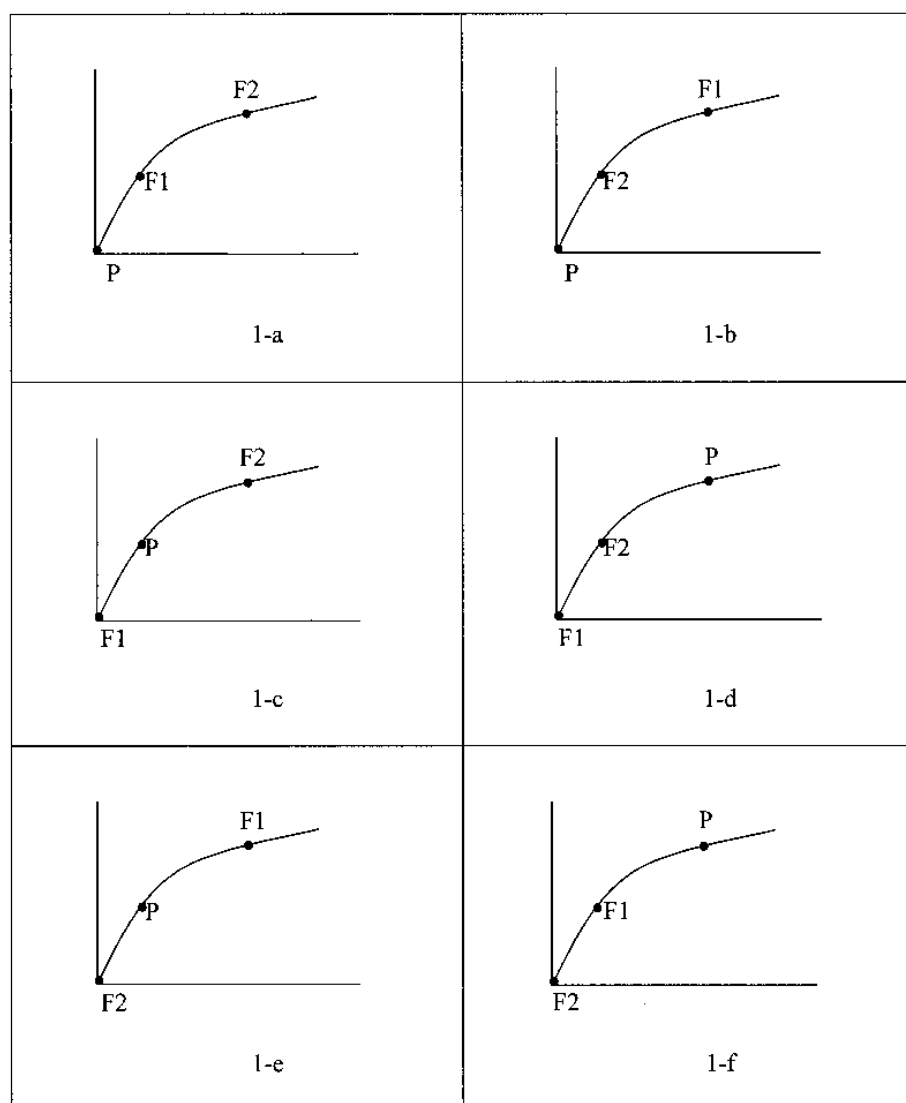


FIG. 1 Six different wave interference cases.



**Case 1.  $C_P < C_{F1} < C_{F2}$**

According to the wave propagation theory (1, 7), the introduction of the first feed results in a self-sharpening wave with the wave velocity

$$u_{F1 \rightarrow P} = \frac{u_0}{1 + \frac{\rho}{\varepsilon} \left( \frac{q_{F1} - q_P}{C_{F1} - C_P} \right)} \quad (3)$$

This concentration wave velocity is the slope of the straight line originating at  $t = 0$  and separating concentration P from F1 in the distance–time ( $Z-t$ ) diagram as is shown schematically in Fig. 2-a. The switch to the second feed concentration at  $t = t_0$  also results in a self-sharpening wave with the wave velocity

$$u_{F2 \rightarrow F1} = \frac{u_0}{1 + \frac{\rho}{\varepsilon} \left( \frac{q_{F2} - q_{F1}}{C_{F2} - C_{F1}} \right)} \quad (4)$$

This concentration wave velocity is the slope of the straight line originating at  $t = t_0$  and separating concentration F1 from F2 in the  $Z-t$  diagram as is also shown schematically in Fig. 2-a. One can easily see from Fig. 2-b that

$$\frac{q_{F2} - q_{F1}}{C_{F2} - C_{F1}} < \frac{q_{F1} - q_P}{C_{F1} - C_P}$$

Therefore,

$$u_{F2 \rightarrow F1} > u_{F1 \rightarrow P}$$

Because the second feed concentration wave is faster than the first one, it will catch the first feed concentration wave at  $t = t_1$ , which can be obtained from solving the following equations simultaneously:

$$\begin{cases} Z_1 = u_{F1 \rightarrow P} t_1 \\ Z_1 = u_{F2 \rightarrow F1} (t_1 - t_0) \end{cases} \quad (5)$$

Thus,

$$t_1 = \frac{u_{F2 \rightarrow F1}}{u_{F2 \rightarrow F1} - u_{F1 \rightarrow P}} t_0 \quad (6)$$

The corresponding position at which two waves meet each other is

$$Z_1 = \frac{u_{F2 \rightarrow F1} u_{F1 \rightarrow P}}{u_{F2 \rightarrow F1} - u_{F1 \rightarrow P}} t_0 \quad (7)$$

Once the second feed concentration wave catches the first one, wave interfer-



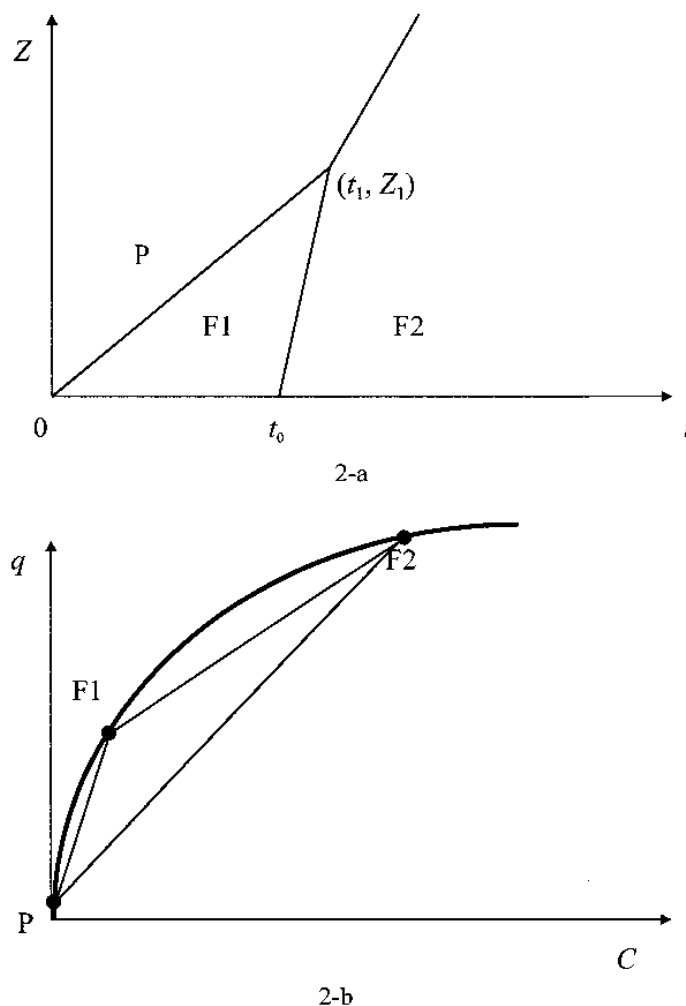


FIG. 2 Schematic diagrams showing wave interference for  $C_P < C_{F1} < C_{F2}$ . 2-a: Distance-time diagram. 2-b: Adsorption isotherm.

ence phenomenon occurs and results in a self-sharpening wave with the wave velocity

$$u_{F2 \rightarrow P} = \frac{u_0}{1 + \frac{\rho}{\varepsilon} \left( \frac{q_{F2} - q_P}{C_{F2} - C_P} \right)} \quad (8)$$

This concentration wave velocity is the slope of the straight line originating at the point  $t = t_1$ ,  $Z = Z_1$ , and separating composition  $F2$  from  $P$  in the  $Z-t$  diagram as is shown schematically in Fig. 2-a. As shown in Fig. 2-b,

$$\frac{q_{F2} - q_{F1}}{C_{F2} - C_{F1}} < \frac{q_{F2} - q_P}{C_{F2} - C_P} < \frac{q_{F1} - q_P}{C_{F1} - C_P}$$



Therefore,

$$u_{F2 \rightarrow F1} > u_{F2 \rightarrow P} > u_{F1 \rightarrow P}$$

This suggests that the first slow P|F1 cut wave will be accelerated, the second fast F1|F2 cut wave will be slowed down, and the waves will merge into a P|F2 cut wave with velocity in between the two waves.

The effluent history can be easily obtained by plotting a horizontal line  $Z = L$  in the  $Z-t$  diagram as is shown in Fig. 3. Depending upon the bed length,  $L$ , the effluent history or the so-called breakthrough curve may have different shapes. Figure 3-a shows the case for  $L < Z_1$ . In this case the bed length is shorter than the position where wave interference occurs; the first feed concentration wave leaves the bed before it can be caught by the second one. There are therefore two breakthrough times, one for the first feed concentration and the other for the second. Figure 3-b shows the case for  $L > Z_1$ . In this case the bed length is longer than the position where wave interference occurs; the first feed concentration wave is caught and merged by the second one. There is therefore only one breakthrough time for the second feed concentration.

### Case 2. $C_P < C_{F2} < C_{F1}$

In this case the introduction of the first feed also results in a self-sharpening wave with the wave velocity calculated by Eq. (3). This concentration

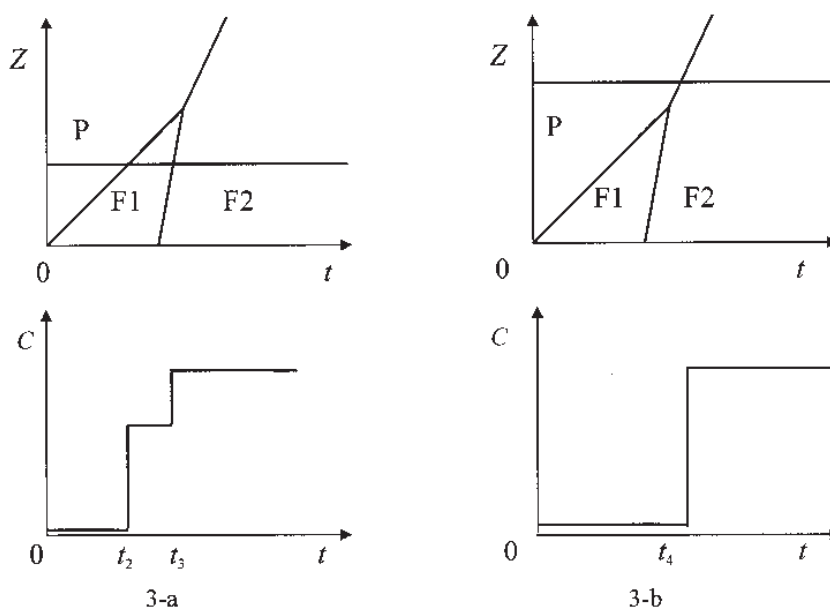


FIG. 3 Schematic diagrams showing wave interference for  $C_P < C_{F1} < C_{F2}$ . 3-a:  $L < Z_1$ . 3-b:  $L > Z_1$ .





wave velocity is the slope of the straight line originating at  $t = 0$  and separating concentration P from F1 in the  $Z-t$  diagram as is shown schematically in Fig. 4-a. The switch to the second feed concentration at  $t = t_0$  results in a non-sharpening wave with the wave velocity

$$u_C = \frac{u_0}{1 + \frac{\rho}{\varepsilon} \left( \frac{dq}{dC} \right)}, \quad \text{for } C_{F2} \leq C \leq C_{F1} \quad (9)$$

The concentration wave velocities are the slopes of the straight lines all originating at  $t = t_0$  and separating concentration F1 from F2 in the  $Z-t$  diagram as is shown schematically in Fig. 4-a. Theoretically, there are infinite numbers

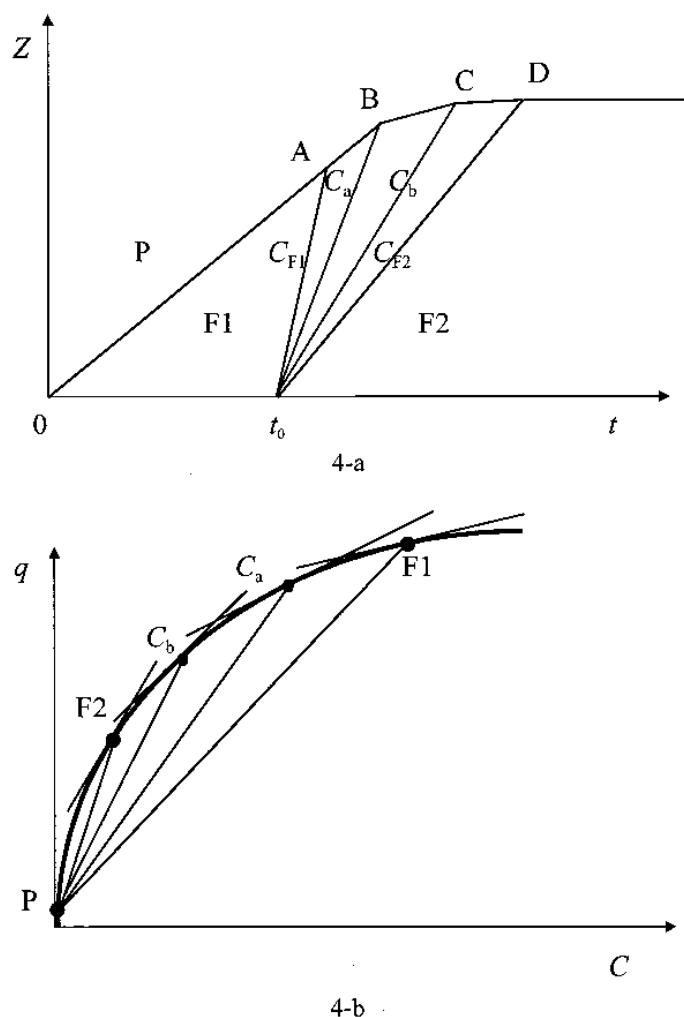


FIG. 4 Schematic diagrams showing wave interference for  $C_P < C_{F2} < C_{F1}$ . 4-a: Distance-time diagram. 4-b: Adsorption isotherm.



of straight lines representing the continuous variation of the concentration from the first feed to the second. Shown in Fig. 4-a are only four lines to illustrate the procedure to obtain the  $Z-t$  curve after wave interference occurs.

As shown in Fig. 4-b, the nonsharpening F1|F2 cut wave velocity of the first feed concentration is greater than the self-sharpening P|F1 cut wave velocity of the first feed concentration because the tangent slope at F1 is less than the chord slope of  $\overline{F1P}$ . Therefore, the nonsharpening F1|F2 cut wave will catch the self-sharpening P|F1 cut wave and this results in another self-sharpening wave, separating the presaturation concentration P from the first feed concentration F1. As is shown in Fig. 4-b, the nonsharpening wave velocity of concentration  $C_a$  is greater than the self-sharpening P|F1 cut wave velocity because the tangent slope at  $C_a$  is less than the chord slope of  $\overline{F1P}$ . Therefore the self-sharpening P|F1 cut wave will be caught by the nonsharpening wave of concentration  $C_a$  and this results in another self-sharpening P| $C_a$  cut wave.

As in Case 1, the first wave interference Point A, shown in Fig. 4-a, locates at  $t = t_A$  and  $Z = Z_A$ , which can be calculated by Eqs. (6) and (7), respectively. The second wave interference Point B locates at  $t = t_B$  and  $Z = Z_B$ , which can be obtained from solving the following simultaneous equations:

$$\begin{cases} Z_B = u_{F1 \rightarrow P} t_B \\ Z_B = u_{C_a}(t_B - t_0) \end{cases} \quad (10)$$

Thus,

$$\begin{cases} t_B = \frac{u_{C_a}}{u_{C_a} - u_{F1 \rightarrow P} t_0} \\ Z_B = \frac{u_{C_a} u_{F1 \rightarrow P}}{u_{C_a} - u_{F1 \rightarrow P}} t_0 \end{cases} \quad (11)$$

In Eq. (11) the nonsharpening wave for the concentration  $C_a$  is calculated by the following equation:

$$u_{C_a} = \frac{u_0}{1 + \frac{\rho}{\varepsilon} \left( \frac{dq}{dC} \right)} \quad (12)$$

So far, the first two interference points, A and B, are determined and the line segment  $\overline{AB}$  representing the P|F1 cut can be plotted. Similarly, other interference points such as C and D can be determined by the same procedure and the line segments  $\overline{BC}$  and  $\overline{CD}$  representing the P| $C_a$  and P| $C_b$  cuts, respectively, can be obtained.

Once the whole  $Z-t$  diagram is obtained, the effluent history can be easily obtained by plotting a horizontal line  $Z = L$  in the  $Z-t$  diagram as is shown in Fig. 5. Again, depending upon the bed length,  $L$ , the effluent history may have



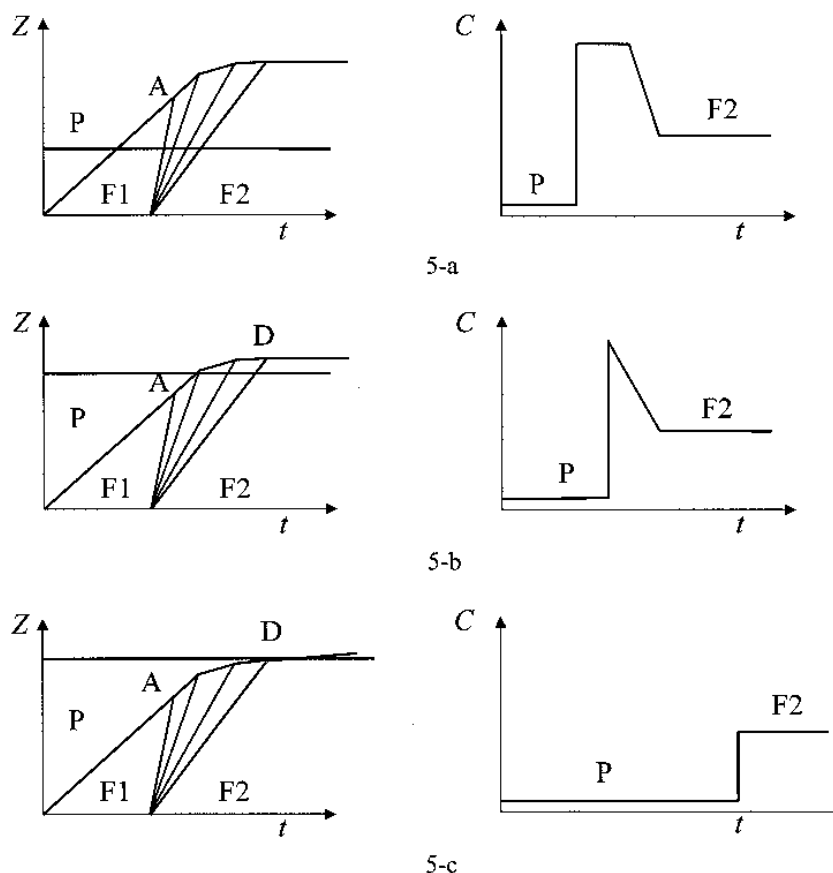


FIG. 5 Schematic diagrams showing wave interference for  $C_P < C_{F1} < C_{F2}$ . 5-a:  $L < Z_A$ . 5-b:  $Z_A < L < Z_D$ . 5-c:  $L > Z_D$ .

different shapes. Figure 5-a shows the case for  $L < Z_A$ . In this case the bed length is shorter than the position where the first wave interference point occurs; the first feed concentration wave leaves the bed before it can be caught by the second one. Therefore, at the first breakthrough time, the effluent concentration jumps from the presaturation concentration to the first feed concentration. Then the effluent concentration remains at the first feed concentration for a while until the first feed concentration wave is caught by the second feed one. Once the second feed concentration wave catches the first one, wave interference phenomenon occurs and makes the effluent concentration decrease from the first feed one to the second feed one. Finally, the second feed concentration breaks through and the effluent concentration remains at that concentration.

Figure 5-b shows the case for  $Z_A < L < Z_D$ . In this case the effluent concentration jumps from the presaturation concentration to a concentration in between the first feed one and the second feed one, and then decreases to the



second feed concentration. Finally, the second feed concentration will also break through.

Figure 5-c shows the case for  $L > Z_D$ . In this case the bed is so long that the first feed concentration wave is totally run over by the second one, therefore the effluent concentration jumps from the presaturation concentration to the second feed one and then remains at that concentration.

## EXPERIMENTAL

Yellow acid dye (Sumitomo Chemical Corp., Japan) with the main ingredient triphenylmethane was used to prepare the synthetic wastewater for the column tests in this study. Granular activated carbon (Taipei Chemical Corp., Taiwan) with a size range between 30 and 32 mesh was used. The column tests were carried out in a water-jacketed glass column with an inside diameter of 1.20 cm. A fixed volume of the yellow dye solution was fed to the top of the column by a metering pump (Watson-Marlow, 302S/RL), flowed through the carbon bed, and then exited from the bottom of the column. After the fixed volume of the dye solution flowed through the bed, a second feed with a different dye concentration was introduced to the column. Other experimental details can be found in the previous paper (21). The presaturation and feed compositions of the column tests are summarized in Table 1.

## RESULTS AND DISCUSSION

The adsorption isotherm of the yellow dye at 25°C was determined in the previous study (21) and was best represented by the Redlich–Peterson model with the parameters shown in Table 2. Using these parameters and the equations developed above, the effluent concentrations for all the column test runs can be predicted.

In practice, one can plot the in-bed volume versus cumulative effluent volume diagram with the slope being the dimensionless concentration wave velocity,  $1/[\varepsilon + \rho(dq/dC)]$  or  $1/[\varepsilon + \rho(\Delta q/\Delta C)]$ , to replace the distance–time

TABLE 1  
Presaturation and Feed Composition of Column Tests

	Run 1	Run 2	Run 3	Run 4
Presaturation concentration (mg/L)	0	0	0	0
First feed concentration (mg/L)	200	200	492	492
Volume of first feed (L)	3.5	3.5	1.5	1.2
Second feed concentration (mg/L)	486	486	20	20



TABLE 2  
Redlich–Peterson Adsorption Model Parameters for the  
Yellow Dye at 25°C

Parameter	Value	Unit
A	842	$\text{mg/g}/(\text{mg/L})^{0.087}$
B	0.266	$(\text{mg/L})^{0.913}$
M	0.913	—

diagram with the slope being the concentration wave velocity. The in-bed volume is the volume traveled by the concentration wave. The feed-switch time  $t_0$  is correspondingly replaced by the total volume of the first feed flowing through the column.

The  $V_{\text{in-bed}}$  vs  $V_{\text{eff}}$  diagram for Run 1 is shown in Fig. 6 with the interference point being  $V_{\text{eff}} = 3375 \text{ BV}$  and  $V_{\text{in-bed}} = 1.6 \text{ BV}$ . Since the interference in-bed volume is greater than unity, the first feed concentration will break through before it is caught by the second one. There are therefore two breakthrough volumes, one for the first feed concentration and the other for the second, as shown in Fig. 6. The experimental and predicted

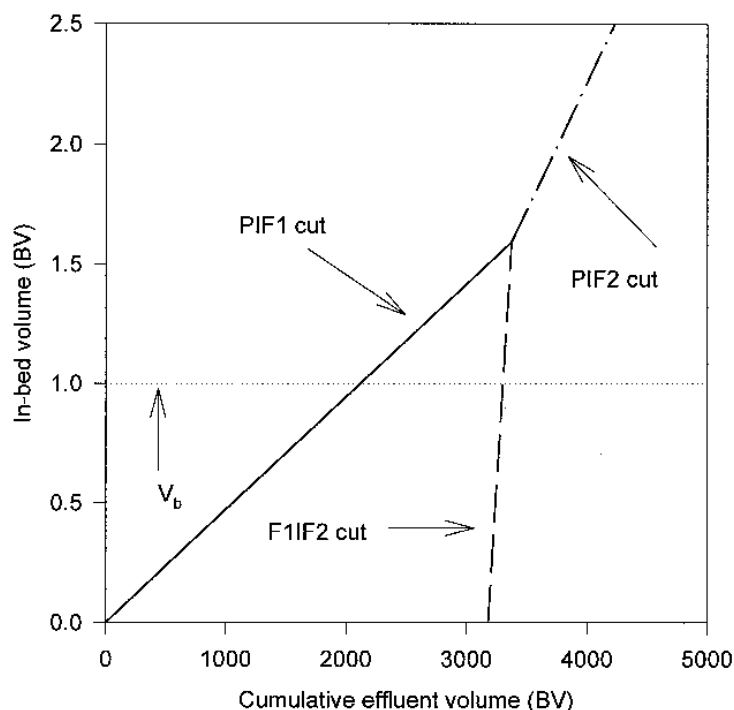


FIG. 6 In-bed volume versus cumulative effluent volume diagram for column test Run 1.

breakthrough curves for Run 1 are shown in Fig. 7. As one can see from Fig. 7, the wave interference theory gives satisfactory prediction of the breakthrough curve.

The  $V_{\text{in-bed}}$  vs  $V_{\text{eff}}$  diagram for Run 2 is shown in Fig. 8 with the interference point being  $V_{\text{eff}} = 1954$  BV and  $V_{\text{in-bed}} = 0.9$  BV. Since the interference in-bed volume is less than unity, the first feed concentration will be caught and merged by the second one. There is therefore only one breakthrough volume for the second feed concentration, as shown in Fig. 8. The experimental and predicted breakthrough curves for Run 2 are shown in Fig. 9. As one can see from Fig. 9, the wave interference theory also gives quite satisfactory prediction of the breakthrough curve.

The  $V_{\text{in-bed}}$  vs  $V_{\text{eff}}$  diagram for Run 3 is shown in Fig. 10 with the first interference point being  $V_{\text{eff}} = 1265$  BV and  $V_{\text{in-bed}} = 1.35$  BV. Since the interference in-bed volume is greater than unity, the first feed concentration will break through before it is caught by the second one. This wave interference phenomenon results in the breakthrough curve shown in Fig. 11. The  $V_{\text{in-bed}}$  vs  $V_{\text{eff}}$  diagram for Run 4 is shown in Fig. 12 with the first interference point being  $V_{\text{eff}} = 940$  BV and  $V_{\text{in-bed}} = 1$  BV. In this run, as soon as the first feed concentration leaves the column, the wave interference phenomenon occurs. The resulting breakthrough curves are shown in Fig. 13.

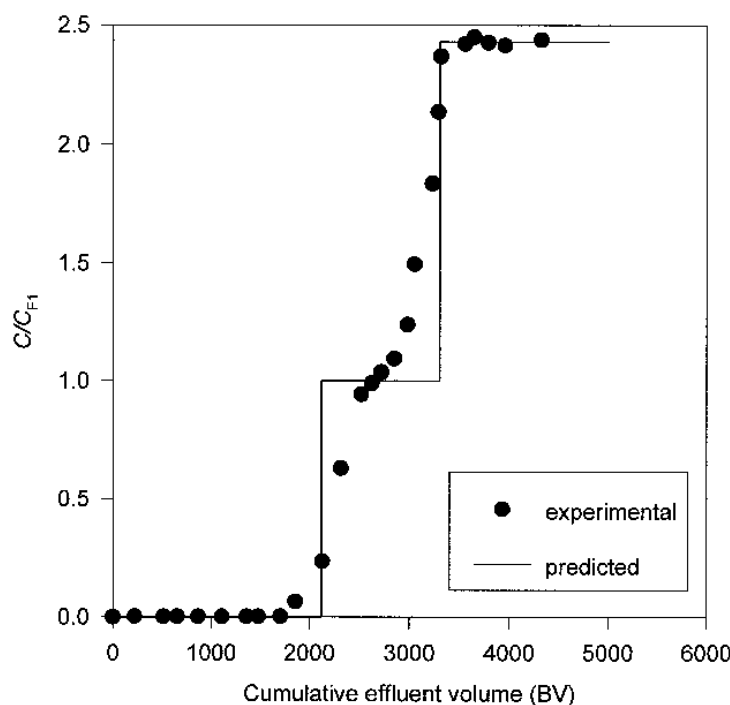


FIG. 7 Breakthrough curve for column test Run 1.



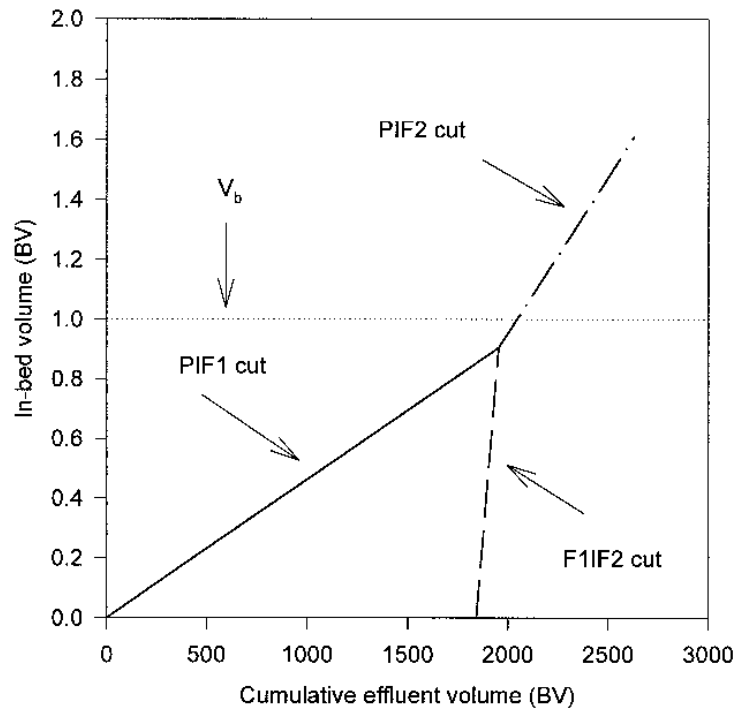


FIG. 8 In-bed volume versus cumulative effluent volume diagram for column test Run 2.

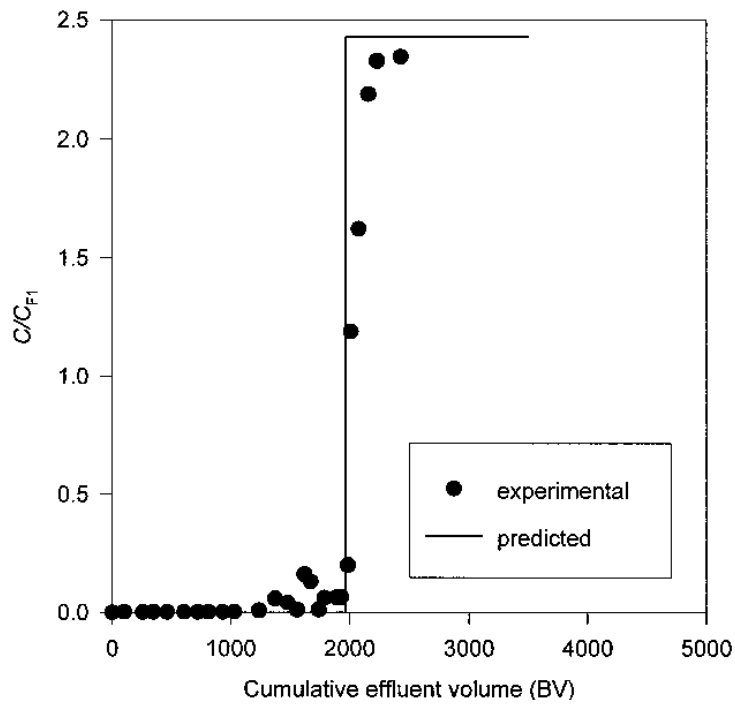


FIG. 9 Breakthrough curve for column test Run 2.



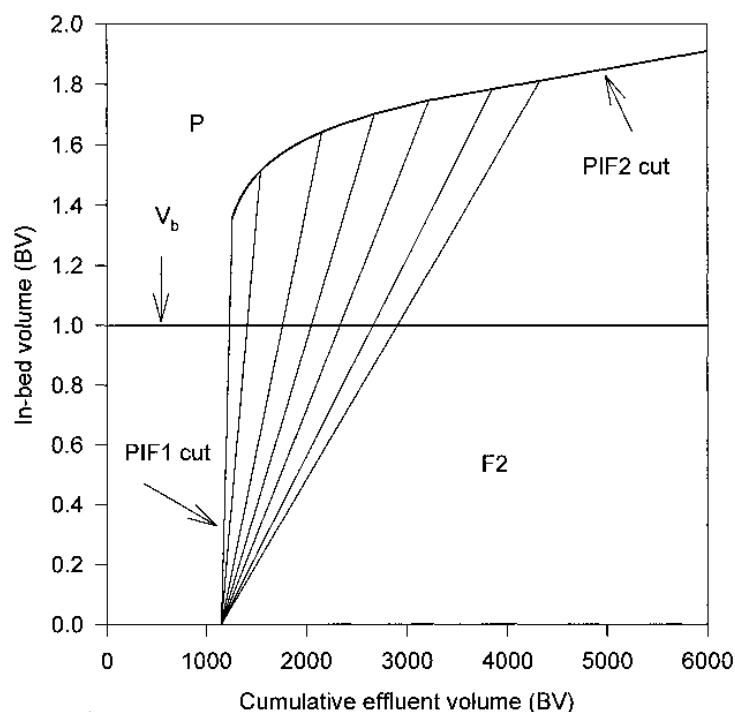


FIG. 10 In-bed volume versus cumulative effluent volume diagram for column test Run 3.

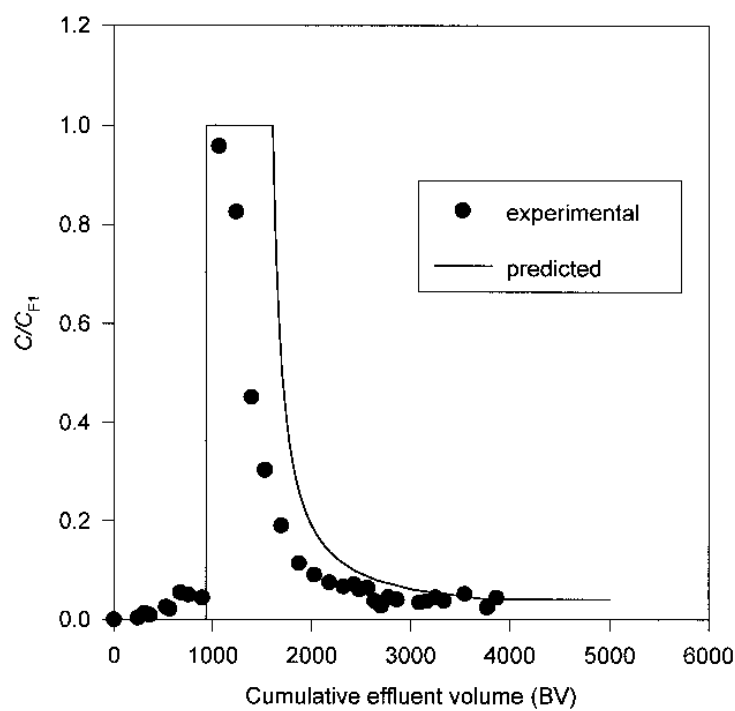


FIG. 11 Breakthrough curve for column test Run 3.





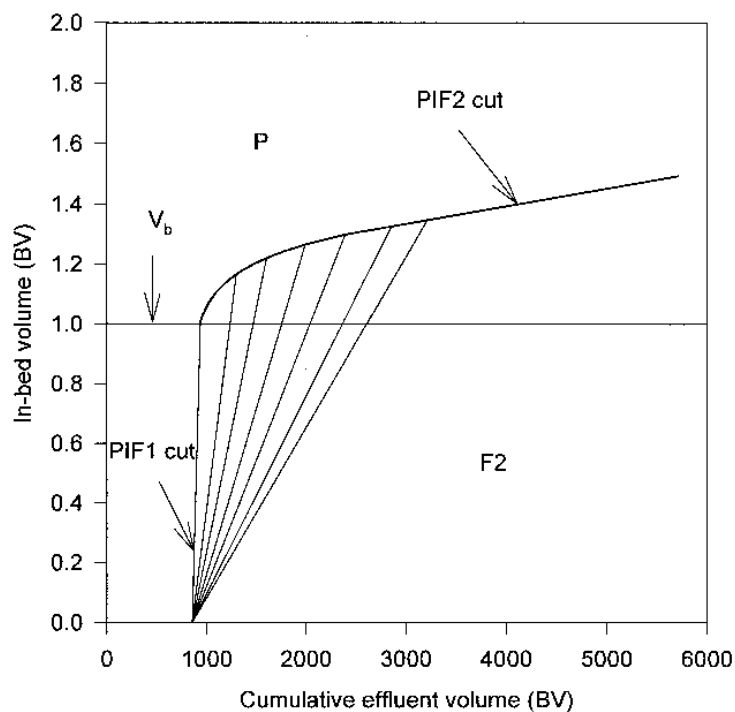


FIG. 12 In-bed volume versus cumulative effluent volume diagram for column test Run 4.

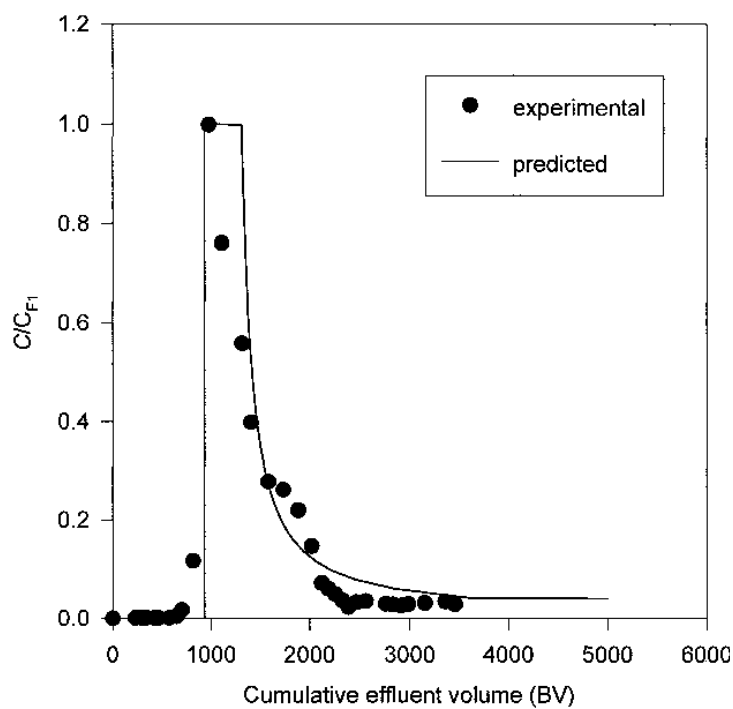


FIG. 13 Breakthrough curve for column test Run 4.



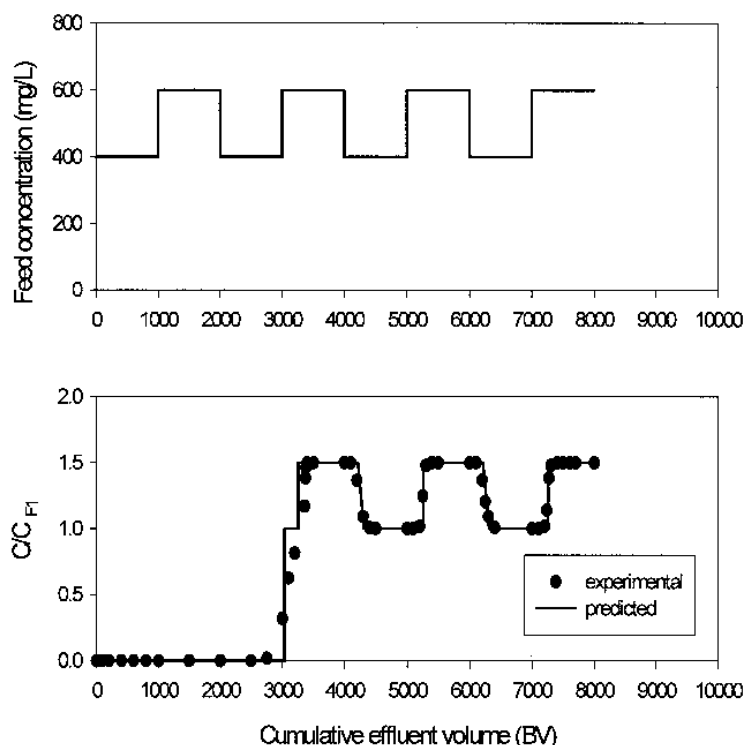


FIG. 14 Breakthrough curve for column test run with cyclic feed concentration change.

In addition to the above four test runs whose breakthrough curves can be satisfactorily predicted by the wave interference theory, another column test run with a cyclic feed concentration change was performed to further validate the applicability of the theory. The column was initially packed with fresh activated carbon, and the 400- and 600-mg/L yellow dye solutions were cyclically fed to the column. The feed concentration and the breakthrough curve for this test run are shown in Fig. 14. The wave interference phenomenon of this test run contains the two interference cases discussed above; both self-sharpening and nonsharpening waves result from feed concentration interference. Again the wave interference theory predicts the breakthrough curve satisfactorily.

## CONCLUSIONS

The effluent histories of yellow dye adsorption by an activated carbon bed are experimentally measured and theoretically predicted by the wave interference theory. Explicit expressions have been developed from the wave interference theory to predict the single-component adsorption column dynamics with the feed concentration change. Five column tests of yellow dye adsorp-



tion by an activated carbon bed are performed, and the results show that the dye effluent histories predicted by the wave interference theory are in good agreement with the experimental ones.

This study provides a handy methodology to understand the cause-and-effect relations of wave interference phenomena. Without solving the governing equation for the column dynamics, the theory uses the concept of wave propagation and simple calculations to construct the distance–time diagram. Then the effluent histories can be easily obtained if the bed length is specified.

## NOMENCLATURE

$A$	Redlich–Peterson model constant $[\text{kg/kg}-(\text{kg/m}^3)^{1-M}]$
$B$	Redlich–Peterson model constant $[(\text{kg/m}^3)^M]$
$C$	dye concentration in mobile phase ( $\text{kg/m}^3$ )
$C_P$	presaturation dye concentration in mobile phase ( $\text{kg/m}^3$ )
$C_{F1}$	first-feed dye concentration in mobile phase ( $\text{kg/m}^3$ )
$C_{F2}$	second-feed dye concentration in mobile phase ( $\text{kg/m}^3$ )
$M$	Redlich–Peterson model constant
$q$	dye concentration in stationary phase ( $\text{kg/kg carbon}$ )
$q_P$	presaturation dye concentration in stationary phase ( $\text{kg/kg carbon}$ )
$q_{F1}$	dye concentration in stationary phase equilibrium with first feed ( $\text{kg/kg carbon}$ )
$q_{F2}$	dye concentration in stationary phase equilibrium with second feed ( $\text{kg/kg carbon}$ )
$t$	time (h)
$u_0$	linear velocity of carrier fluid (m/h)
$u_{F1 \rightarrow P}$	concentration wave velocity of F1 P cut (m/h)
$u_{F2 \rightarrow F1}$	concentration wave velocity of F2 F1 cut (m/h)
$Z$	distance from the inlet of mobile phase (m)
$\varepsilon$	void fraction of bed
$\rho$	carbon bed density ( $\text{kg/m}^3$ )

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