Chapter

Immobilization of Powdered Coal Fly Ashes (CFAs) into CFA Beads and Column Studies on Color Removal from Pulp Mill Effluents Using These CFA Beads

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Abstract

In this study, immobilization process of the three (3) powder CFAs was studied. The major results on immobilization process were briefly presented. A total number of fifteen (15) column studies from the combination of the five (5) types of CFAs beads and the three (3) PMEs samples were performed. In each column study, a set of aggregate parameters of flow rate, empty bed contract time, operational time, and throughput volume was studied, and the data was fitted to existing modeling of breakthrough curves. The overall operational time was 12–24-hour, color removal efficiencies were 40–90%, and throughput volume of treated PMEs was 10–14 bed volume. For the column study, the correlation coefficient R^2 value for each combination indicated that the Thomas model had a better fit with the observed data than the Adams-Bohart model, and the color adsorption capacities of CFA beads varied in a wide range of 0.31–28.23 mg/g.

Keywords: coal fly ash (CFA), pulp mill effluent (PME), color removal, immobilization, column study

1. Introduction

As one of the dominant industries in Georgia, USA, the pulp and paper industries consume huge amounts of fresh water and a wide variety of chemicals during pulp production processes. A significant amount of water and these chemicals are released as high pollutant load with intense color effluent into surface water bodies [1]. Pulp and paper mill effluents (PPMEs) transport high concentrations of organic/inorganic pollutants and color compounds like lignocellulosic compounds, tannins, hemicelluloses, pectin, resin acids, unsaturated fatty acids, carboxylic acid, and other substances [2]. These untreated effluents are responsible for increasing the levels of chemical oxygen demand (COD), biochemical oxygen demand (BOD), total organic carbon (TOC), adsorbable organic halides (AOXs), toxic contaminants, and heavy metals in the water ecosystem [3]. Therefore, PPMEs must be treated before they are discharged into receiving water bodies.

Physical treatment can efficiently remove 80% of suspended solids (SS) from pulp mill effluents (PMEs) [4]; Aerobic lagoon and anaerobic treatment studies showed potentially removing 50% color and 60% COD from PMEs [4, 5]. However, these fungal treatment studies are still ongoing and in the early stage of industrial implementation [6]. The coloration of receiving water bodies from PMEs causes negative impacts including an unpleasing esthetic appearance, reduction of dissolved oxygen (DO) level, and reduction of sunlight transmission into bodies of water which may adversely affect aquatic life. Additionally, insufficient sunlight reduces photosynthetic activity, therefore, making it difficult to remove organic pollutants and causing an increase of water/wastewater treatment costs downstream [7, 8]. Hence, tertiary treatment is essential for the further treatment of PMEs.

Adsorption, a proven and widely used treatment process, removes a variety of contaminants from industrial wastewater. Activated carbon, the most common adsorbent, is widely used to remove contaminants from industrial wastewater for a long time [9, 10]. However, the high production costs of activated carbon have motivated researchers to explore alternative low-cost adsorbents like coal fly ash (CFA), rice husk ash (RHA), bagasse fly ash (BFA), etc. industrial for wastewater treatment.

Energy sector is another dominant industry in Georgia, USA. It produced 6.1 million tons of CFA as byproducts in 2011 and 60% of CFA were disposed of in ash ponds. Investigating other beneficial uses of CFA in environmental engineering is necessary. The results from our Batch Studies have shown that CFA can effectively remove color from PME as a low-cost adsorbent [11]. However, these results were achieved in a batch operation using the powdered CFA samples, which are rarely used in a column in practice for a continuous operation due to their low permeability.

Therefore, the objectives of this study were to explore the cost-effective immobilization processes of powdered CFAs with water and addition of binders, to produce the CFAs beads with strength and high adsorption capacity for color removal from PME, and to use these CFA beads in column studies to remove color from the PME under a continuous operation.

2. Material and methods

2.1 Coal fly ash (CFA)

Three (3) raw CFA samples were obtained from three individual coal-based power plants units of Georgia Power Company and labeled as CFAs #1 - #3. The first one was Class "C" subbituminous ash, whereas the last two were Class "F" bituminous ashes. These CFAs were stored in plastic containers at room temperature (20 ± 10°C) in the Water and Environmental Research Lab (WERL) at Georgia Southern University (GSU) for further study. The chemical composition of CFA#2 is summarized in **Table 1**.

2.2 Pulp mill effluent

For this study, three (3) biologically treated secondary PMEs samples were collected from three different pulp mill factories from surrounding areas in Georgia, USA. They were labeled as PMEs #1 - #3, respectively. All these PMEs were collected from secondary clarified effluent outlets. These PMEs were stored in plastic container in a refrigerator at temperature of 4 ± 1°C in the WERL at GSU for further study. The primary properties of these PMEs are summarized in **Table 2**.

Chemical compositions	% w/w
Silicon dioxide (SiO_2)	50.96
Aluminum oxide (Al_2O_3)	21.00
Titanium dioxide (TiO_2)	1.11
Iron oxide (Fe_2O_3)	14.32
Calcium oxide (CaO)	4.39
Magnesium oxide (MgO)	0.9
Potassium oxide (K_2O)	2.49
Sodium oxide (Na_2O)	1.07
Sulfur trioxide (SO_3)	1.94
Loss on ignition	1.82
Total	100%

Table 1.
The chemical composition of CFA#2.

PMEs	Received date	Number of buckets	Color	pH	COD	TOC
			(mg/L (Pt-Co) Color Units)		(mg/L)	(mg/L)
PME 1	16-Mar-15	One (1) 5 – gallon	157	6.92	255	
	9-Jun-16	Two (2) 5 – gallon	182	5.62	222	164.9
PME 2	26-Aug-15	One (1) 5 - gallon	355	8.21	86.6	
	17-Jun-16	Two (2) 5 – gallon	834	6.39	135.5	90.8
PME3	1-Jan-16	Three (3) 1 – gallon	723	7.51	248	
	12-Oct-16	Three (3) 1 – gallon	968	5.89	298.5	110.5

Table 2.
The primary properties of the three (3) PMEs.

2.3 Chemical reagent

For immobilization process, N type hydrated lime was collected from the Material Laboratory at GSU.

2.4 Immobilization process

The immobilization study of powdered CFAs was started by operating the key equipment of pelletizer (DP-14 Agglo-Miser Disc Pelletizer supplied by Mars Mineral) with each of the three CFA samples and water only without adding any binders. Multiple immobilizing parameters, such as RPM and the vertical angle of the pan of the pelletizer, and the ratio of CFA sample to water, as well as optimum curing conditions on humidity and duration, were investigated. The optimal rotational speed of pelletizer and vertical angle of the pan were found 32 RPM and 45 degree, respectively, and they were maintained the same during the whole immobilization process. For each batch, 2500 gm of CFA powder was pelletized by adding hydrated lime or Class "C" type CFA directly while water was added using spraying bottle. After palletization, the fresh pellets had to be cured with thin layer covered by wet cloth at room temperature (20 ± 1°C) in the WERL at GSU for two weeks and exposed to atmosphere for air dry for one week before being used in column studies (12).

2.5 Fixed bed column

For each column study, the fixed-bed column was set-up using a glass column with a 15 mm internal diameter and 750 mm bed height and a peristaltic pump with medium flow range (1 mL/min). The schematic diagram of the fixed-bed column system is illustrated in **Figure 1**. For each column study, the column was packed by 2–4 mm diameter CFA beads, one type at a time. The mass of the CFA beads typically varied in a range of 50–75 g with a varied height of the bed of CFA beads in a range of 505–655 mm, which is dependent on the size distribution of the CFA beads produced from the above immobilization process. A layer of washed sand with a height of 25 mm was placed at the bottom of the CFA beads to protect any loss of beads as well as give mechanical support in the fix-bed column system. The top of the bed of CFA beads was submerged at least 50 mm below the water surface of the PME sample in the column by maintaining a high position of the outlet in treated effluent tubing (inverse "U" shape) shown in **Figure 1**.

All of these column studies were performed at room temperature (20 ± 1°C) at the WERL at GSU and the treated PME samples were collected at different intervals: every 1 h from the 1st hour to 12th hour, every 2 hours from the 13th hour to 24th hour until the column of the CFA beads was exhausted. These samples were analyzed for color. Meanwhile, all the treated PME samples were collected in the treated PME container as an accumulated sample and its color was tested in the end of a column study. The treated PME sample was separated by vacuum-filtering the mixture through a Supor-450 47 mm 0.45 μm membrane paper. Immediately after filtration, the filtrated samples were collected to test color. Duplicate flasks with identical CFA and effluent mixtures were used to represent each sample. The average reading of color and pH measurements between the two duplicate samples were documented. The color of PME samples was measured using HACH DR 5000 Spectrophotometer and HACH Method 8025.

Color removal efficiency of CFA can be calculated using following equation:

$$Removal\ Efficiency\ (\%) = \frac{(C_i - C_t) * 100}{C_i} \tag{1}$$

where E = Removal Efficiency (%), C_i is the initial color concentration (mg/L), C_t is the color concentration at time t (mg/L).

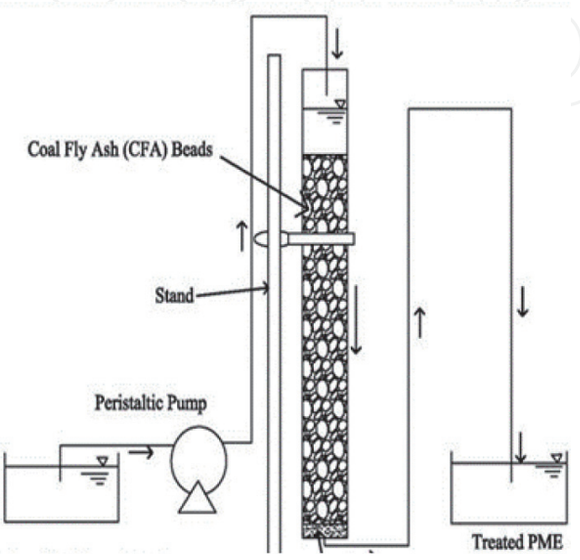

Figure 1.
A schematic diagram of column.

The effluent volume (V_{eff}) can be expressed as:

$$V_{eff} = Q \times t_{total} \tag{2}$$

Where, Q stands for volumetric flow rate (mL/min) and t_{total} stands for the time of exhaustion (min).

The area under the breakthrough curve (A) can be determined by integrating color adsorbed concentration (C_{Ad}) vs. time (t). For a given volumetric flow rate (Q) and initial color concentration (C_0) of PMEs, the total amount of color adsorbed (q_{total}) can be calculated by the following equation:

$$q_{total} = \frac{Q}{1000} \int_{t=0}^{t_{total}} C_{Ad} dt \tag{3}$$

Total amount of color, M_{total} (mg) passed through the fixed-bed column system is determined by following equation:

$$M_{total} = \frac{C_0 \times Q \times t}{1000} \tag{4}$$

Where, C_0 is denoted as initial color concentration (mg/L) of PME, Q is the volumetric flow rate (mL/min) of PMEs.

The fixed-bed column system color removal efficiency with respect to the flow volume can be calculated by following equation:

$$Removal\ Efficiency = \frac{q_{total}}{M_{total}} \times 100 \tag{5}$$

3. Results and discussions

3.1 Immobilization of powder CFAs

In each batch trial during the immobilization process, CFA beads were produced with small spherical shape and maintained CFA grayish original color. It was observed that the RPM and inclined vertical angle of the pan of pellitizer, and water mixing ratio with CFAs were important factors that affected beads size distribution. In the immobilization process, at high RPM of the pan reduced the size of CFA beads while at low RPM beads diameters were increased. The optimal rotational speed of pelletizer and vertical angle of the pan were found to be 32 RPM and 45 degree, respectively, and were maintained the same during the whole immobilization process. It is also found out that CFA1 (Class "C") or hydrated lime can be used as binder to produce CFA beads. The combination of hydrated lime and CFAs required more water while the combination of CFA1 with CFA2or CFA3 required less.

For each batch of immobilization process, cost effective blinders were used to produced relatively uniform and consistent size and shape. The newly produced beads were put into large foil tray with thin layer and covered with a soaked wet cloth at room temperature of $20 \pm 1°C$ for two weeks. The cloth was kept wet during these two weeks and then was removed. The beads were continually exposed to the atmosphere for air dry at room temperature of $20 \pm 1°C$ for one week. In this study only the amount of water used for immobilization was documented while the water used during 2-week "wet" curing period was not documented. It was observed that about 10–20% of water contents could produce the CFA beads with a

high strength. In general, it was observed that the strength of the beads increased with in increased weight percentage of hydrated lime, adsorbed more water during curing period, and much more pores formed. It was observed that about 10–20% of water content could produce the beads with a high strength. In this study, it was particularly important for the CFA beads to hold their shape in PMEs. Those CFA beads, which could hold their shape in PMEs at least 24 h, were considered to be beads with a high strength. A summary table for total five (5) types of CFA beads is shown in **Table 3**.

The five (5) types of CFA beads produced from the three (3) CFA samples are shown in **Figure 2**.

3.2 Column studies

3.2.1 Type of beads of CFA1 only with PMEs

In these column studies, the raw PMEs samples were passed through 75 g of CFA1 only beads within the column. The flow rates of Q = 0.78–0.89 mL/min, empty bed contact time (a measure of the time during which water to be treated is in contact with CFA beads in a contact column), EBCT = 125 min with an operation time t = 12–24 h. During this operational period, almost 5–13 bed volumes of PMEs were treated in fixed bed column. **Figure 3** shows the physical properties of PMEs and fixed bed column. At the end of each column study, the apparent color ratio of C/C_0 reached up to 0.546, which corresponded to 45.4% apparent color removal.

3.2.2 Types of CFA beads of CFA2 + Lime and CFA2 + CFA1 with PMEs

In these column studies, the raw PMEs sample were passed through 50 ~ 75 g of CFA2 + Lime (Mass Ratio of CFA2 to Lime = 4:1) and CFA2 + CFA1 beads with a

Type No.	Name of CFA beads	CFA samples	Cost-effective binders	Mass ratio of CFA to binder
1	CFA3 + Lime	CFA3	Hydrated Lime	4:1
2	CFA3 + CFA1	CFA3	CFA1	1:2
3	CFA1	CFA1	Water Only	8.1:1
4	CFA2+ Lime	CFA2	Hydrated Lime	4:1
5	CFA2+ CFA1	CFA2	CFA1	1:2

Table 3.
Summary of the finalized cost-effective binders and a Total of five (5) types of CFA beads produced.

Figure 2.
The five (5) types of CFA beads produced from the three (3) CFA samples (from left to right: 1 CFA3 + 2 CFA1; 1 CFA2 + 2 CFA1; 4 CFA3 + 1 Lime; 4 CFA2 + 1 Lime; and CFA1. Note: the numbers of 1, 2, and 4 are referred to mass ratios).

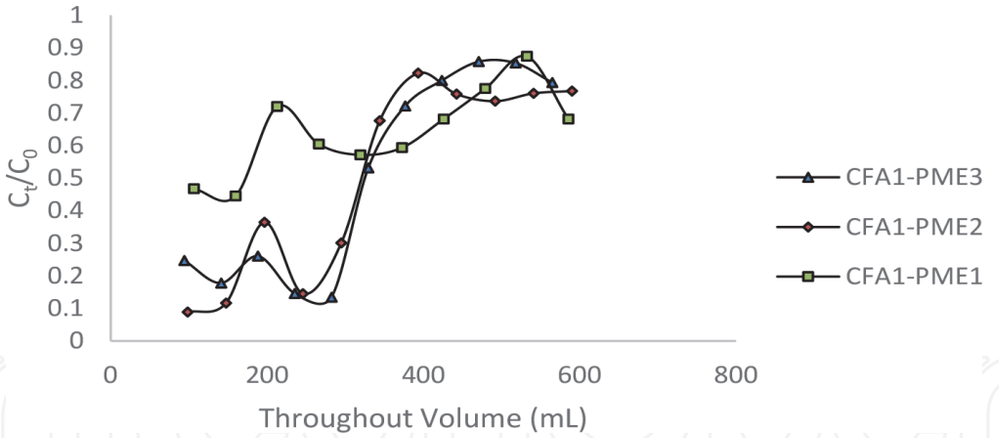

Figure 3.
Breakthrough of color column experiments using three different PME and CFA1 only beads. Experimental setup: Initial color concentration: PME1: 182; PME2: 834 and PME3: 968 (mg/L Pt-Co); bed height: 51 ~ 61 cm; flow rate: 0.78 ~ 0.89 mL/min; at room temperature.

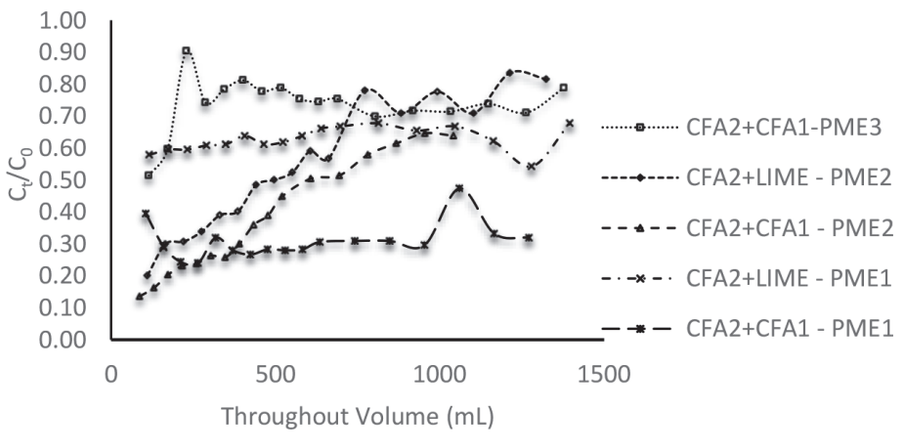

Figure 4.
Breakthrough of color column experiments using three different PMEs, CFA2 and additive materials CFA beads. Experimental setup: initial color concentration: PME1: 182; PME2: 834 and PME3: 968 (mg/L Pt-Co); bed height: 56 ~ 63 cm; flow rate: 0.72 ~ 0.96 mL.

height of 562–660 mm in the column, respectively. The flow rates of Q = 0.72–0.97 mL/min, EBCT = 125 min with an operation time t = 24 h. **Figure 4** shows the physical properties of PMEs and fixed bed column. During this operational period, almost 6–12 bed volumes of PMEs were treated in fixed bed column. At the end of each column study, the apparent color ratio of C/C_0 reached up to 0.27, which corresponded to 73% apparent color removal. The curve of CFA2 + Lime - PME1 shows a different characteristic compare with others. Color might be added due to leaching out of chemicals from the CFA beads and breakdown some chemical reagents present in PME1.

3.2.3 Type of CFA beads of CFA3 + Lime and CFA3 + CFA1 with PMEs

In these column studies, the raw PMEs sample was passed through 50 ~ 75 g of CFA3 + Lime, and CFA3 + CFA1 beads with a height of 582–645 mm in the column, respectively. The flow rates of Q = 0.75–0.96 mL/min, EBCT = 125 min with an operation time t = 24 h. **Figure 5** shows the physical properties of PMEs and fixed bed column. During this operational period, almost 6–14 bed volumes of PMEs were treated in fixed bed column. At the end of each column study, the apparent color ratio of C/C_0 reached up to 0.473, which corresponded to 52.7% apparent color removal.

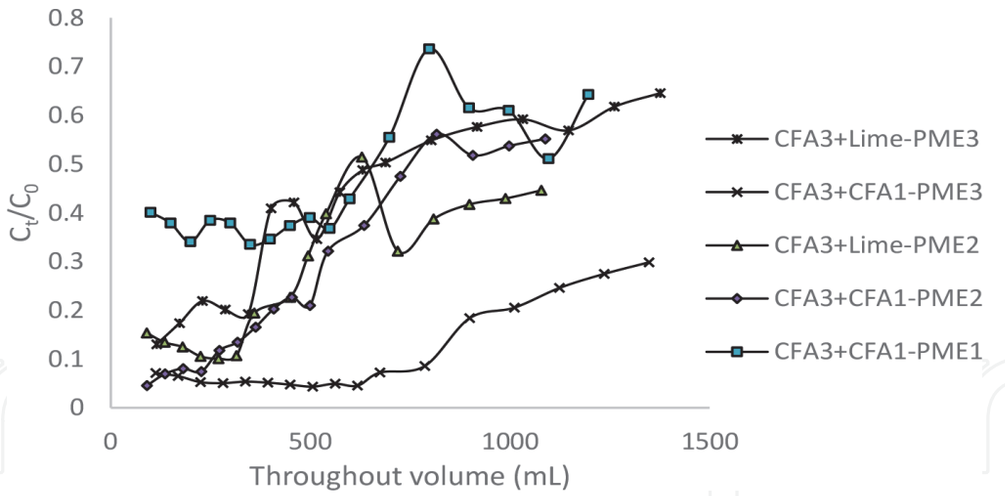

Figure 5.
Breakthrough of color column experiments using three different PME and CFA3 and additive materials CFA beads. Experimental setup: initial color concentration: PME1: 182; PME2: 834 and PME3: 968 (mg/L Pt-Co); bed height: 58 ~ 65 cm; flow rate: 0.75 ~ 0.96.

3.3 Modeling of breakthrough curves from column studies

3.3.1 Adams-Bohart model

The Adams-Bohart model is typically applied to check the dynamic behavior of the column which describes the relationship between $\frac{C_t}{C_0}$ and t in a continuous fixed-bed column system. This model is eminent to predict the initial phase of the breakthrough curve. The linear equation is expressed as:

$$\ln\left(\frac{C_t}{C_0}\right) = K_{AB} \times C_0 \times t - K_{AB} \times N_0 \times \left(\frac{Z}{U_0}\right) \tag{6}$$

Where, C_0 and C_t are denoted as the color concentration (mg/L Pt-Co) of influent and effluent of column system; K_{AB} represents the kinetic constant (L/mg. min); N_0 is the saturation concentration (mg/L); Z is denoted as bed depth of the fixed-bed column and U_0 stands for superficial velocity (cm/min). By plotting $\ln\left(\frac{C_t}{C_0}\right)$ vs. t, N_0 is obtained from the intercept and K_{AB} can be calculated from slope of the graph.

The column studies experimental data obtained from the five different combinations of CFA beads with PME1 were used for linear regression analysis and their corresponding parameters kinetic constant K_{AB} and saturation constant N_0 were obtained along with the correlation coefficient (R^2) given in (Eq. (6)). The model is shown in **Figure 6**. At initial color concentration, C_0 = 182 mg/L, the R^2 value of Adams-Bohart model for different combinations were varied in a range of 0.54–0.80. Under the similar condition, this model corresponding parameters kinetic constant K_{AB} values were varied 2.20–4.40 (10^{-6} L/mg.min), and N_0 = 10.23–0.78 (10^3 mg/L) respectively.

The column studies experimental data obtained from the four different combinations of CFA beads with PME2 were used for linear regression analysis and their corresponding parameters kinetic constant K_{AB} and saturation constant N_0 were obtained along with the correlation coefficient (R^2) given in (Eq. (6)). The model is shown in **Figure 7**. At initial color concentration, C_0 = 834 mg/L, the R^2 value of Adams-Bohart model for different combinations varied in a range of 0.70–0.94.

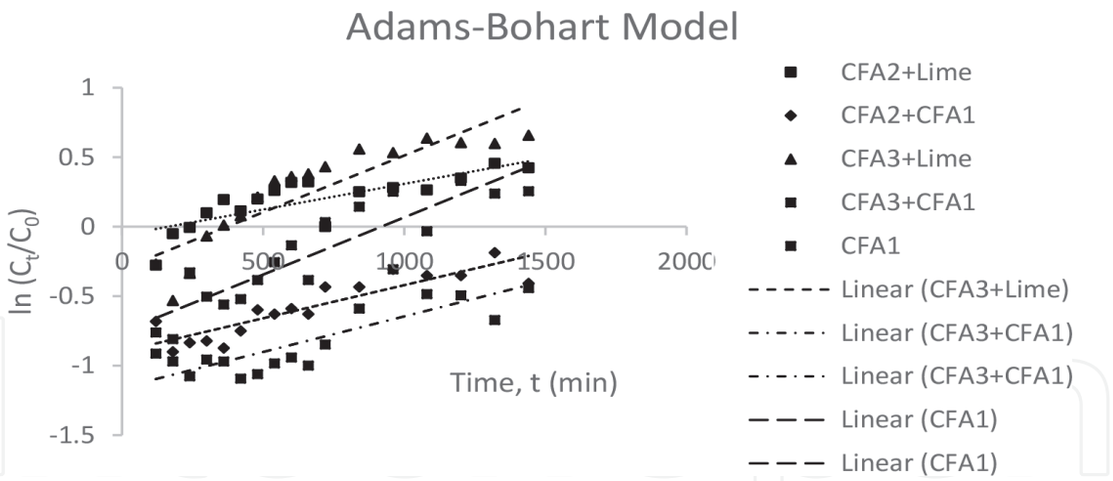

Figure 6.
Application of the Adams-Bohart model to the experimental data from column study for PME1.

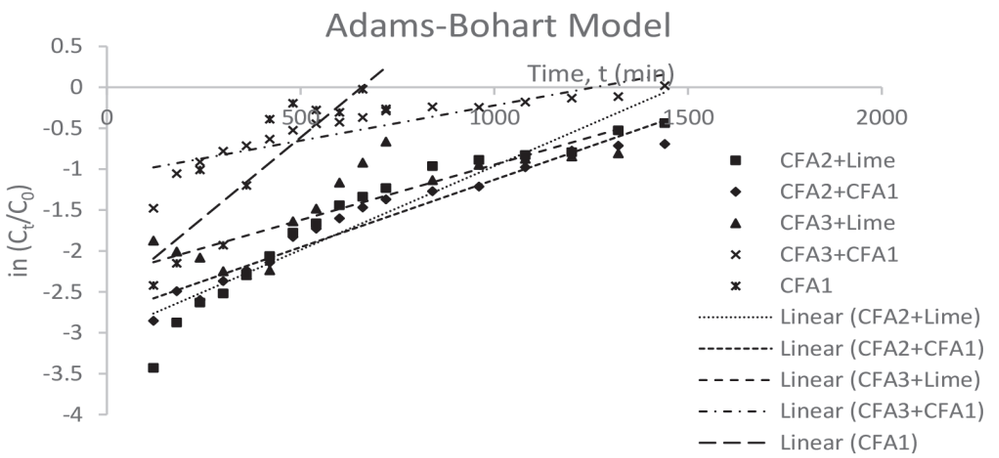

Figure 7.
Application of the Adams-Bohart model to the column study experimental data for PME2.

Under the similar condition, this model's corresponding parameters kinetic constant K_{AB} values was varied 1.68–4.68 (10^{-3} L/mg.min). N_0 = 12.63–4.23 (10^3 mg/L) respectively.

The column studies experimental data obtained from the four different combinations of CFA beads with PME3 were used for linear regression analysis and their corresponding parameters kinetic constant K_{AB} and saturation constant N_0 were obtained along with the correlation coefficient (R^2) given in (Eq. (6)). The model was shown in **Figure 8**. At initial color concentration, C_0 = 968 mg/L, the R^2 value of Adams-Bohart model for different combinations varied in a range of 0.67–0.85. Under the similar condition, this model's corresponding parameters kinetic constant K_{AB} values was varied 1.14–3.2 (10^{-3} L/mg.min). N_0 = 55.22–16.06 (10^3 mg/L) respectively.

3.3.2 Thomas model

Thomas model is one of the widely used models in fixed-bed continuous column operation. This model based on three assumptions: i) follows Langmuir Isotherm model; ii) obeys the second order reversible reaction kinetics; and iii) there is no axial depression of the adsorbent. The linear equation is expressed as follows

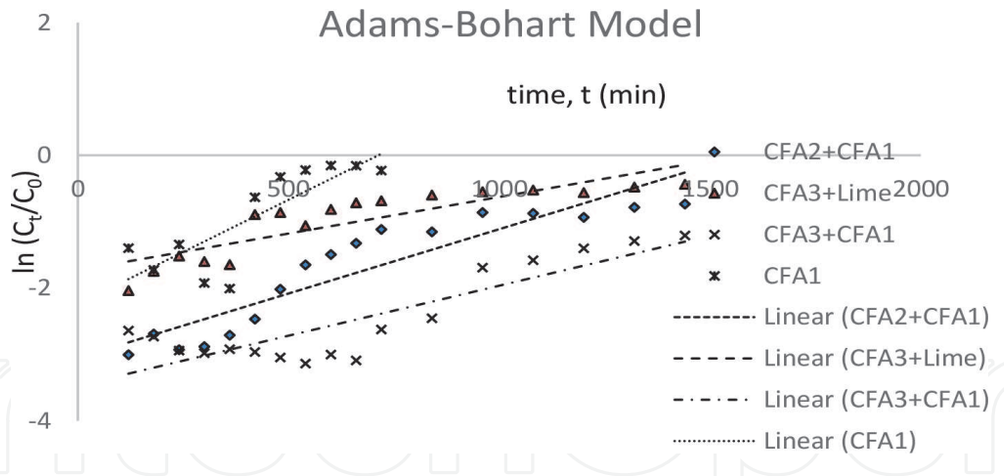

Figure 8.
Application of the Adams-Bohart model to the column study experimental data for PME3.

$$\ln\left(\frac{C_0}{C_t}-1\right)=\frac{K_{Th}\times q_0\times M}{Q}-K_{Th}\times C_0\times t \tag{7}$$

Where, C_0 and C_t are denoted as the color concentration (mg/L Pt-Co) of influent and effluent of column system; K_{Th} stands for Thomas rate constant (L/(min.mg)); q_0 is the maximum color adsorption capacity for CFA beads (mg/g), M is the mass of CFA beads (g), Q is the flow rate (mL/min). By plotting $\ln\left(\frac{C_0}{C_t}-1\right)$ versus t, Thomas rate constant K_{Th} can be obtained from the slope and q_0 can be calculated from the interception of the plot.

The experimental data obtained from the three selected combinations of three types of CFA beads with PME1 column studies were used for linear regression analysis and their corresponding Thomas rate constant K_{Th} and maximum color adsorption capacity q_0 were calculated along with the correlation coefficient (R^2) given in (Eq. (7)). The model is shown in **Figure 9**. At initial color concentration C_0 = 182 mg/L, the R^2 value of Thomas model for different combination varied in a range of 0.54–0.79. The Thomas rate constants were calculated for different combinations and were 6.593, 14.286 and 5.495 (x10^{-6} L/min.mg) for the CFA2 + CFA1, CFA3 + CFA1 and CFA1 only beads, respectively. The maximum color adsorption capacity q_0 was obtained from the plot and their values were in a range of 0.31–1.72 mg/g. On the other hand, the color adsorption capacities after 20 hours' operation of pump started were obtained and they varied in a range of 0.72–1.67 mg/g, which

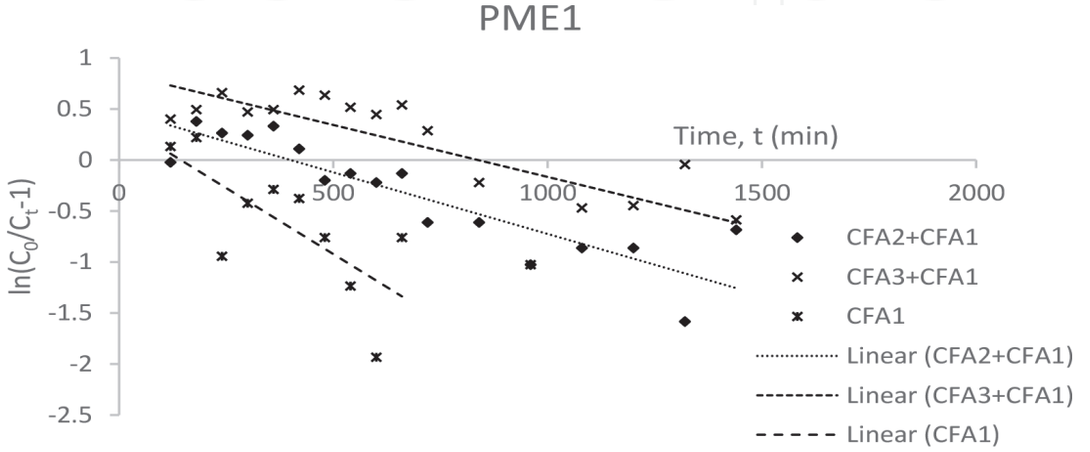

Figure 9.
Application of Thomas model to the column study experimental data for PME1.

were very close to the range of the Thomas model adsorption capacities. However, these values were very low compared with the results from the batch studies. For the CFA beads (Class "F" type CFA + binder), the maximum color removal capacity increased with decreased the volumetric flow rate of column operation.

The experimental data obtained from the five selected combinations of CFA beads with PME2 column studies were used for linear regression analysis and their corresponding Thomas rate constant K_{Th} and maximum color adsorption capacity q_0 were calculated along with the correlation coefficient (R^2) given in (Eq. (7)). The model is shown in **Figure 10**. At initial color concentration C_0 = 834 mg/L, the R^2 value of Thomas model for different combination varied in a range of 0.71 to 0.97. Thomas rate constants were calculated for different combinations and were 3.36, 2.64, 9.71, 2.16 and 3.16 (x 10^{-6} L/min.mg) for the CFA3 + Lime, CFA3 + CFA1, CFA1 only, CFA2 + Lime, CFA2 + CFA1 beads, respectively. The maximum color adsorption capacity q_0 was obtained from the plot and their values were in a range of 3.65 to 14.02 mg/g. On the other hand, the color adsorption capacities after 20 hours' operation of pump started were obtained and they varied in a range of 4.04–12.95 mg/g which were very close to the range of the Thomas model adsorption capacities. However, these values were quite high compared with the results from the batch studies. For the CFA beads (Class "F" type CFA + binder), the maximum color removal capacity increased with decreased the volumetric flow rate of column operation.

The experimental data obtained from the four selected combinations of CFA beads with PME3 were used for linear regression analysis and their corresponding Thomas rate constant K_{Th} and maximum color adsorption capacity q_0 were calculated along with the correlation coefficient (R^2) shown in (Eq. (7)). The model is shown in **Figure 11**. At initial color concentration C_0: 968 mg/L, R^2 value of Thomas model for different combination were varied in between 0.76 to 0.85. Thomas rate constant were calculated for different combination 23.1, 1.86, 1.76 and 35.71 ($x10^{-6}$ L/min.mg). The maximum color adsorption capacity q_0 was obtained from the plot and their values were varied in between 0.8 ~ 28.23 mg/g. Where, color adsorption capacity after 20 hours' operation of pump started were obtained and their value varied in a range of 4.4 ~ 12.71 mg/g that had some difference with Thomas model constant values. Again, these values were quite high compared with the batch studies results. For the CFA beads (Class "F" type CFA + binder), the maximum color removal capacity increased with decreased the volumetric flow rate of column operation.

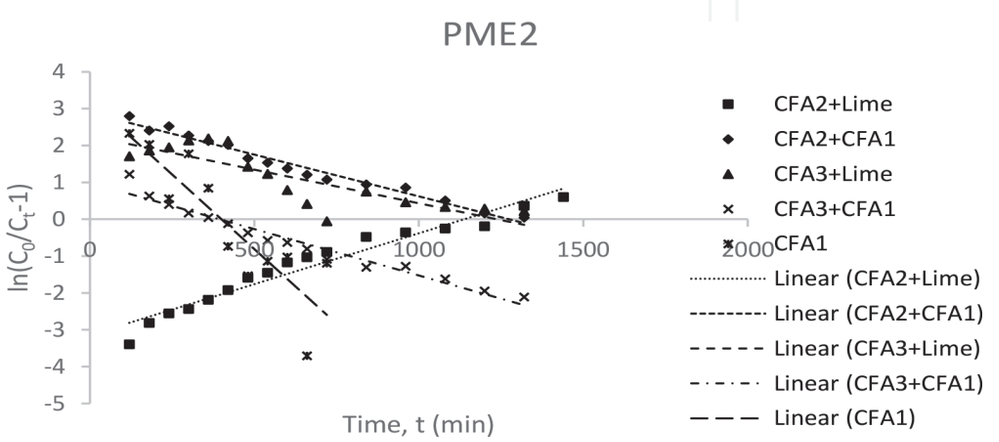

Figure 10.
Application of Thomas model to the column study experimental data for PME2.

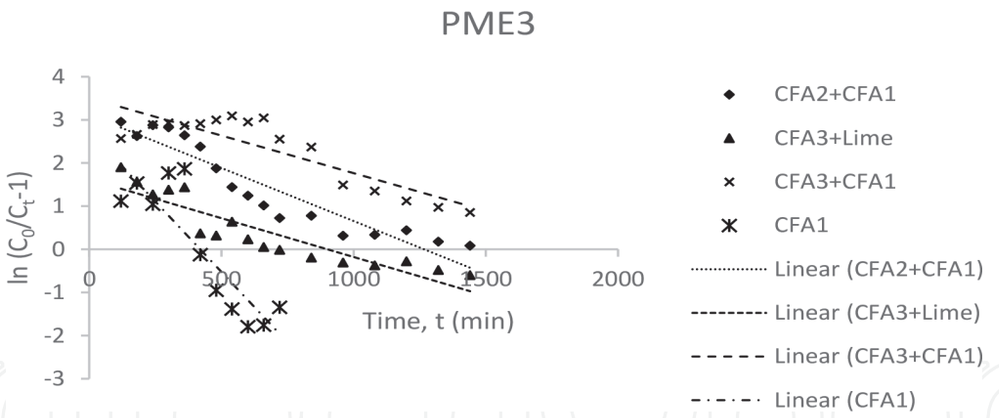

Figure 11.
Application of Thomas model to the column study experimental data for PME3.

4. Conclusions

Based on the significant results obtained from the column studies, the following conclusions can be reached:

- From the immobilization studies, both hydrated lime and the Class "C" of CFA1 were found to be cost-effective binders for Class "F" CFA2 or CFA3 in addition to water, while water only was the cost-effective binder for Class "C" CFA1.

- To produce CFA beads with strength and capability to removal color, the effective mass ratio of Class "F" CFA2/CFA3 sample to hydrated lime was 4:1 while that to Class "C" CFA1 was 1:2.

- From column studies, the CFA beads were able to be packed into columns and used to remove color from the PMEs under continuous operation. They could hold the good shape after they contacted and submerged in PME samples for 12–24 h. The overall color removal efficiencies were in a range of 45–90% while the general throughput volume of treated PME was in a range of 10–14 bed volumes of CFAs beads in the columns.

- From column studies, based on true color removal, for the three PMEs, the CFA3 + CFA1 and CFA2-CFA1 beads were good ones with high adsorption capacities.

- The correlation coefficient of R^2 value for each column study indicated that the Thomas model had a better fit with the observed data than the Adams-Bohart model did both for PME2 and PME3, whereas, the Adams-Bohart model fitted better than the Thomas model for PME1.

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