Evaluation of Musa Acuminata (Banana Peels) as a Biomass Adsorbent in Treating Water Contaminated with Chromium (Cr^{6+})

Sthembile Mzimela, Paul Musonge, and Babatunde F Bakare

Abstract— Banana peels (BPs) are one of the agricultural waste peels that are a burden to the ecology as they accumulate in environmental spaces with no definite use for them. However, BPs have been identified as a potential biomass adsorbent for the removal of heavy metals (HM) due to their lignocellulosic properties containing functional groups such as hydroxyls and carboxylic groups that aid in the removal of HM from water. BPs can be a cheap and abundant resource to obtain and use in treating water as an alternative to other technological methods that are expensive to purchase and maintain. Therefore, BPs were used in this study to evaluate their performance as a biomass adsorbent in the adsorption process for the removal of hexavalent chromium (Cr^{6+}) in water. Batch studies were conducted in jar tests for the evaluation of equilibrium and kinetic studies for the adsorption process. Equilibrium isotherms studied were the Langmuir, Freundlich, and Elovich isotherms and the process favored the Freundlich isotherm with and coefficient of determination (R^2) of 0.99. Therefore, adsorption of Cr^{6+} occurred on a heterogeneous surface of BPs with a surface heterogeneity factor (n) of 1.77. The pseudo-first, pseudo-second, and intra-particle diffusion order kinetic studies were applied and the process favored the pseudo-second order kinetic with an R^2 of 0.9986 and K$_2$ of 0.8g/mg.min. This indicated that the process went through chemisorption with some sharing of electrons between the surface of the BPs and Cr^{6+} ions in water. In a fixed-bed column, the height was varied for the removal of Cr^{6+} using BPs. An increase in bed height significantly increased the breakthrough time of the bed during adsorption from 10 min at 5cm to 420 min at 30cm. The Thomas, Adams-Bohart, and Yoon-Nelson models were used to evaluate the dynamic behavior of the system. The process followed the Adams-Bohart model with R^2 ranging between 0.94 and 0.98 as well as the Yoon-Nelson model with R^2 ranging between 0.93 and 0.97. The study showed that BPs has the potential to be an alternative biomass adsorbent in treating water with Cr^{6+}.

Keywords— Adsorption, Biomass Adsorbent, Chromium, Banana Peels

I. INTRODUCTION

Water contaminated by heavy metals (HMs) in South Africa is a big problem as it impacts the quality and availability of drinkable water to humans due to the toxic effects HMs such as chromium (Cr) has on humans when accumulated within the bloodstream. HMs are described as chemical elements that are five times greater than the specific gravity of water, but are soluble in water due to the high content of sulfates present in water making it easier for metals to dissolve at low pH [1-3]. HMs are not biodegradable and high HM content accumulated in the bloodstream of human beings is toxic and can cause adverse effects such as mental illness, skin rashes, a suppressed immune system, and many other health issues [4-6]. The most common HM found in South Africa are Co, Cd, As, Hg, Ni, Pb, Fe, Mn, Mg, and Zn including Cr.

Chromium (Cr) is listed in the most common HMs found in water bodies and the metal exists in two oxidation states, Cr^{3+} and Cr^{6+} [7]. Cr^{3+} is used in the system for blood mechanism maintenance at required amounts, however, Cr^{6+} is a poisonous form of Cr for humans. The ingestion and accumulation of Cr^{6+} in the bloodstream can cause blood diseases, cancers, and fatalities over a period of time [4]. A study done by [8] in 2014 detected concentration levels of Cr^{6+} in the water of 219.6 µg/L, which was greater than the limit set by WHO and DWAF of <50µg/L. Recent studies done by [9] and [10] detected concentrations ranging between 0.005 − 0.15 µg/L and 0.001 − 0.40µg/L, respectively, from water boreholes around the Muledene village in Limpopo as well as from surface and groundwater between the South African and Mozambican borders.

Many authors have emphasized the advantages and disadvantages of the common technological methods used for remediating water containing HMs such as ultrafiltration, reverse osmosis, chemical precipitation, coagulation-flocculation, and ion exchange. The main issues that are most prevalent with these current methods are their affinity for

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certain metals, low removal efficiencies, large sludge production, operational cost, and maintenance [1, 2, 4, 5, 11-15]. Therefore, many research studies have been done in the adsorption of HMs using biomass adsorbents as an inexpensive and available resource that is biodegradable and more effective in removing toxic HMs in water such as banana peels.

Bananas in South Africa are one of the most abundant fruits available with an estimated farming production in 2016 of 402'053 tons with an additional 114'913 tons of fruit being imported [16]. Due to the abundance of the banana fruit, several tons of banana peels (BPs) are produced as waste mainly from marketplaces and households as garbage, which accumulates as environmental solid waste [12, 17]. However, BPs like most agricultural waste peels has lignocellulosic characteristics which contain polymeric groups such as cellulose, hemicellulose, and lignin that have been identified to contain groups that aid in the removal of HMs in water [14, 17-19].

Preliminary studies have been done previously for the characterization of BPs using analytical methods such as FTIR, SEM, EDS, XRD, and BET. From the results obtained BP was identified to contain functional groups of hydroxyl, carbonyl, amines, and carboxylic acid groups. The surface of the peels was recorded to be uneven and mesoporous with mainly an amorphous structure containing very few crystals and pore size of 2.9 nm.

Therefore, this paper presents the kinetic studies conducted from batch experiments and the dynamic studies conducted from column experiments for the removal of Cr^{6+} in water.

II. MATERIALS AND METHODS

A. Materials

Banana peels were collected from a local fruit and vegetable store and were washed from dirt and impurities twice with tap water followed twice by deionized water. The washed peels were then placed on a tray and dried in a convection air dryer at a constant air speed of 1.6 m/s and temperature of 60°C for 48 hours. The dried peels were then crushed in a blender and reduced to powder and washed again from impurities obtained while crushing. The resultant cake was dried in an oven at a constant temperature of 120°C for 48 hours until dry. The resultant powder was sieved in a balance shaker for 10 minutes and a particle size distribution of 710μm (8%), 500 μm (16%), 355 μm (24%), 250 μm (32%), and 180 μm (20%) was obtained. The powder was stored for use.

B. Chemicals

Water was synthesized to represent drinkable water contaminated by Cr^{6+}. Water was synthesized by using sodium chromate tetrahydrate ($Na_2CrO_4.4H_2O$) with de-ionized water to prepare a water solution of 1000 mg/L of Cr^{6+} for the kinetic studies and column experiments.

C. Equilibrium Adsorption Isotherm Studies

Isotherm studies were done to determine the relationship between the adsorbate and adsorbent as well as the degree to which adsorption has taken place at equilibrium conditions [15].

The Langmuir isotherm model was developed by Irvin Langmuir and the model is based on the assumption that there is a monolayer or single-layer binding of molecules onto the surface of the adsorbent with even distribution sites leading to the surface being chemically saturated [20]. The model also assumes that the surface of the solid is homogenous such that the sites on the surface have equal active sites with no movement or migration of adsorbate molecules on the surface [20].

The Langmuir isotherm model is described by the following linear equation (Eq. 1),

$$\frac{C_e}{q_e} = \frac{1}{K_L q_L} + \frac{C_e}{q_L} \tag{1}$$

Where q_e is the adsorption capacity of an adsorbent (mg/g), C_e is the equilibrium concentration (mg/L), K_L is the Langmuir constant (L/mg) and q_L is the Langmuir monolayer adsorption capacity (mg/g) [21].

The Freundlich isotherm is an empirical expression that gives an empirical relation between the concentration of the solute on the surface of an adsorbent to the concentration of the solute in the gas/liquid phase [22]. The isotherm is based on the assumption that adsorption can occur on a heterogeneous surface of an adsorbent with non-uniform distribution of energy levels [23].

The Freundlich isotherm model is described by the following linearized equation (Eq. 2),

$$\log q_e = \log K_f + \frac{1}{n}\log C_e \tag{2}$$

Where K_f is the Freundlich constant known as the sorption capacity [24]. The slope, $1/n$, is the adsorption intensity or surface heterogeneity, where the slope ranges between 0 to 1 for a process that undergoes physical adsorption. If $1/n$ is above 1 then the process has undergone chemisorption [25].

The Elovich isotherm model is based on the Elovich kinetic principle which assumes that adsorption sites increase exponentially with adsorption which is based on chemisorption of gas molecules onto the surface of solids, leading to multilayered adsorption [26]

The Elovich isotherm equation can be expressed as follows (Eq. 3),

$$\ln\frac{q_e}{C_e} = \ln K_E q_E - \frac{q_e}{q_E} \tag{3}$$

Where K_E (L mg^{-1}) is the Elovich constant and q_E is the Elovich monolayer adsorption capacity (mg/g).

D. Adsorption Kinetic Studies

Kinetic studies were done to determine the mechanism of the adsorption process taken place as well as the potential rate controlling or limiting steps such as mass transfer and chemical reaction processes [27].

The pseudo-first model is based on the assumption that the rate of change of solute uptake with time is directly proportional to the difference in saturation concentration and the amount of solute uptake with time is usually applicable at the initial stages of adsorption [28].

The model is based on the capacity of adsorption and it is described by the following linearized equation 4 (Ho 2006),

$$\ln(q_e - q_t) = \ln q_e - \frac{K_1}{2.303} t \qquad (4)$$

Where q_e and q_t (mg/g) are the adsorption capacities at equilibrium and time t (min) respectively, and K_1 (min^{-1}) is the pseudo-first-order rate constant.

The pseudo-second model is based on the assumption that adsorption may be second-order and that the rate-limiting step is chemisorption involving valent forces through sharing or exchanging of electrons between the biosorbent and divalent metal ions in an aqueous solution [28].

The linearized form of the pseudo-second equation is (Eq. 5),

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \qquad (5)$$

The intra-particle diffusion model (Eq. 6) is based on the solute uptake which is almost proportional to the half time, $t^{(1/2)}$, rather than contact time [29]. When applied, the model involves a multi-step approach involving the transport of solute molecules from the aqueous solution phase to the surface of the biomass adsorbent particles, which is followed by diffusion of the solute molecules into the interior of the pores, which is likely to be a slow process [30].

$$q_t = K_{id} t^{1/2} \qquad (6)$$

K_{id} is the intraparticle diffusion rate constant (mg.g^{-1} min$^{-1/2}$).

E. Dynamic Column Studies-Bed Height

The height of the column bed in a fixed-bed column was varied at 5, 10, 15, 20, and 30 cm at constant parameters of pH, influent flow rate, and metal concentration of 4, 4mL/min, and 5 mg/L, respectively. Results from the experiment were fitted with mathematical models to determine the dynamic behavior of the adsorption process between the uptake of Cr^{6+} by banana peels. The mathematical models used were the Thomas model, the Yoon-Nelson model, and the Adams-

Bohart model.

F. Mathematical Models

Mhematical models were developed to determine the dynamic response of the column. The mechanism by which adsorption takes place in a column may be through mass transfer of solute from liquid to the surface of the solid, diffusion and/or reaction on the solid surface (chemisorption) [31, 32].

The Thomas model is based on the assumption that the adsorption process is derived from the Langmuir isotherm model for adsorption-desorption and that the rate driving force follows the pseudo-second-order reversible reaction kinetics with no dispersion and with a predominating film diffusion resistance [31, 33].

The linearized form of the Thomas model can be expressed as (Eq. 6),

$$\ln\left[\frac{C_o}{C_T} - 1\right] = \frac{K_T q_T M}{Q} - K_T C_o t \qquad (6)$$

Where K_T is the Thomas model constant (L/ mg min), q_T is the adsorption capacity (mg/g), Q is the volumetric flow rate (mL/min), M is the mass of adsorbent (g), C_o initial metal concentration (mg/L), and C_T is the effluent concentration (mg/L).

The Yoon-Nelson model was developed as a relatively simple model to address adsorption and the breakthrough behavior of adsorbate in a gas phase with respect to the adsorbent [34, 35].

The following equation below is the linearized equation of the Yoon-Nelson model (Eq. 7),

$$\ln\frac{C_t}{C_o - C_t} = k_{YN} t - \tau k_{YN} \qquad (7)$$

Where k_{YN} and τ are the Yoon-Nelson rate constant (min^{-1}) and the time required for 50% adsorbate breakthrough (min).

The Adams-Bohart model was developed with a basis on surface reaction theory where the relationship between the adsorption rate is proportional to the residual capacity of the adsorbent and the concentration of the species with an adsorption equilibrium that is not instantaneous [11].

This model describes the initial part of the breakthrough curve, as well as the relationship between C_t/C_o and t during continuous operation – see Eq. 8 [36].

$$\ln\left(\frac{C_t}{C_o}\right) = K_{AB} C_o t - K_{AB} N_o\left(\frac{Z}{U_o}\right) \qquad (8)$$

Where K_{AB} is the Adams-Bohart kinetic constant (L/mg min), N_o is the saturation concentration (mg/L), Z is the adsorbent bed depth (cm), and U_o is the linear velocity defined as the ratio of volumetric flowrate in (mL/min) to the cross-

sectional area in (m^2) of the bed (cm/min).

III. RESULTS AND DISCUSSION

A. Equilibrium Adsorption Isotherm Studies

Metal concentrations were varied between 10 and 60 mg/L for 120 minutes at constant pH, biomass dose, and agitation speed of 4, 3 g, and 180 rpm, respectively, and the results after sorption of Cr^{6+} onto BPs was fitted into 3 isotherm models (Langmuir, Freundlich, Elovich). Isotherm parameters from the results obtained in the models from figures 1 to 3 were recorded in Table 1.

The coefficient of determination, R^2, and the calculated adsorption capacity for the adsorption of Cr^{6+} were used to determine the best fit model for the process. The Freundlich isotherm had the highest R^2 of 0.98 as seen in Fig. 2, however, the adsorption capacity, K_f, from the model was not close to the experimental value obtained of 8.10 mg/g. The Langmuir adsorption capacity, q_m, was close to the experimental capacity as seen in Table 1 which also gave a good fit for the process. The separation factor, R_L, was between 0 and 1, showing that adsorption was favorable.

The Elovich isotherm did not fit well with the process, therefore, adsorption was not multilayered.

From the observations made, it was noted that adsorption was a homogeneous monolayer process with similar active sites as well as heterogeneous with multi-layered active sites on the surface of the BPs where adsorption occurred. Studies done by researchers using several biomass adsorbents including BPs found that adsorption can be described by more than one isotherm where the surface of an adsorbent may have different responses or behaviors in adsorbate uptake from an aqueous solution [17, 37-39].

B. Adsorption Kinetic Studies

Contact time was varied from 5, 10, 20, 30, 50, 80, and 120 minutes during batch experiments at constant pH, agitation speed, dosage, and metal concentration of 4, 180 rpm, 5 g, and 10 mg/L, respectively. Results obtained from the experiments were fitted to the pseudo-first, pseudo-second order kinetic, and intra-particle diffusion models.

The adsorption process for Cr^{6+} favored the pseudo-second order kinetic reaction model with an $R^2 \sim 1$. This showed that the rate-limiting step in the process was chemisorption of which, where, adsorption may have been through ion exchange. The adsorption capacity calculated by the pseudo-second kinetic order model, q_e, was similar to the experimental q_e of 2.43 mg/g. Table 2 shows the parameters obtained from Fig(s). 4 to 6.
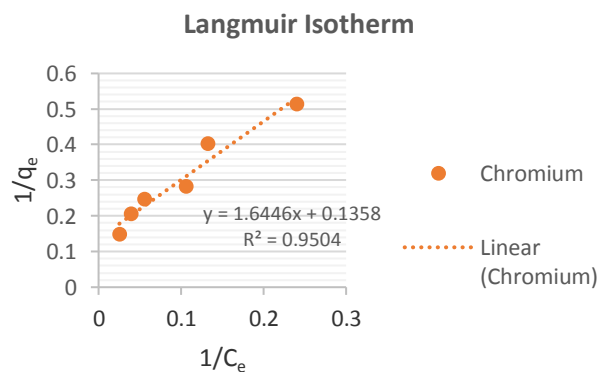

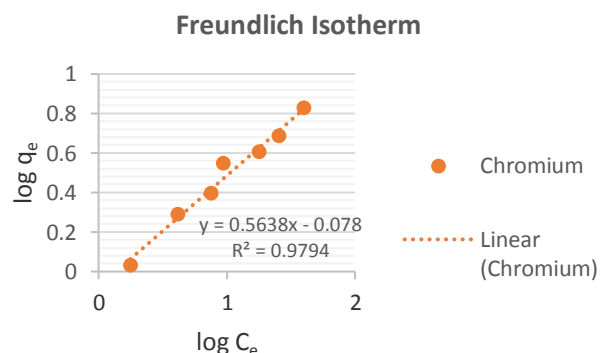Fig 1. Plot of Langmuir isotherm graph for Cr^{6+}

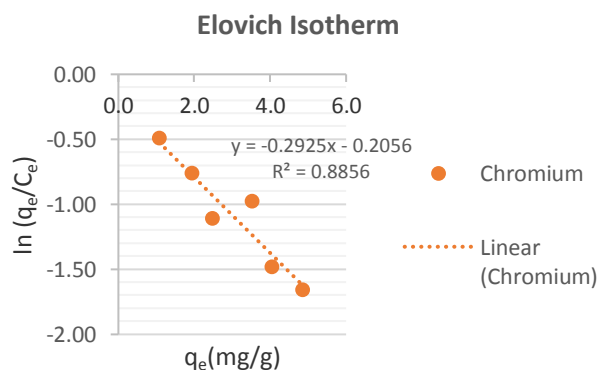

Fig 2. Plot of Freundlich isotherm graph for Cr^{6+}

Fig 3. The plot of Elovich isotherm graph for Cr^{6+}

TABLE I
EQUILIBRIUM ADSORPTION ISOTHERM PARAMETERS FOR Cr^{6+}

Langmuir		Freundlich		Elovich	
q_m (mg/g)	7.36	K_f (mg/g)	0.83	$\ln(K_E q_m)$	0.206
K_L (mg/g)	0.083	1/n	0.56	$1/q_m$ (g/mg)	0.293
R_L	0.548-0.168	n	1.77	q_m (mg/g)	3.42
R^2	0.95	R^2	0.99	K_E (L/mg)	0.36
				R^2	0.89

TABLE II
KINETIC PARAMETERS FOR THE ADSORPTION OF Cr^{6+}

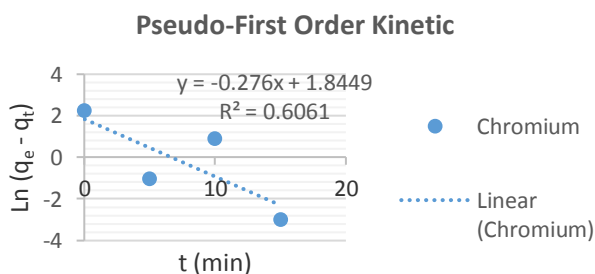| Pseudo-first		Pseudo-second		Intraparticle diffusion	
$q_{e\text{-calc}}$ (mg/g)	6.3	$q_{e\text{-calc}}$ (mg/g)	2.66	$q_{e\text{-calc}}$ (mg/g)	1.92
K_1 (min^{-1})	0.636	K_2 (g/mg.min)	0.8	K_{id} (mg/g.min$^{1/2}$)	0.063
R^2	0.606	R^2	0.9986	R^2	0.71

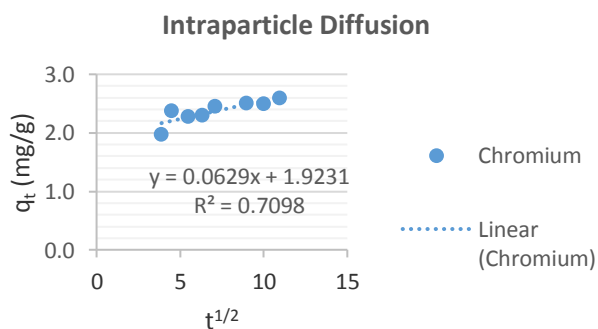Fig. 4 Pseudo-first order kinetic graph for Cr^{6+}

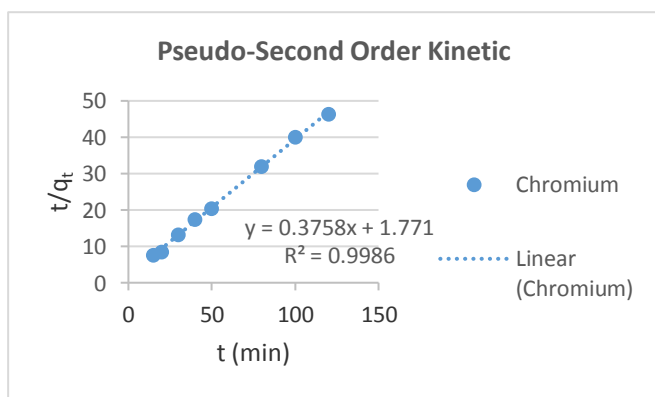Fig. 6 intra-particle diffusion graph for Cr^{6+}

Fig. 5 Pseudo-second order kinetic graph for Cr^{6+}

C. Dynamic Column Studies-Bed Column

Bed height was varied in a fixed bed column from 5, 10, 15, 20 to 30 cm, and parameters that were kept constant were volumetric flow rate, initial metal concentration, and water pH during constant operations. The increase in bed height generally increased breakthrough time and saturation time as seen in Figure 7. The longest breakthrough time reported was between 7 and 8 hours of continuous operation until saturation was reached after 30 hours at a bed height of 30 cm. Table 3 also shows how the column performed during operation. From the table, the used bed height, H_B, also known as the mass transfer zone increased with bed height along with the adsorption capacity, q_B, with the highest at 20 cm of 0.296 mg/g.
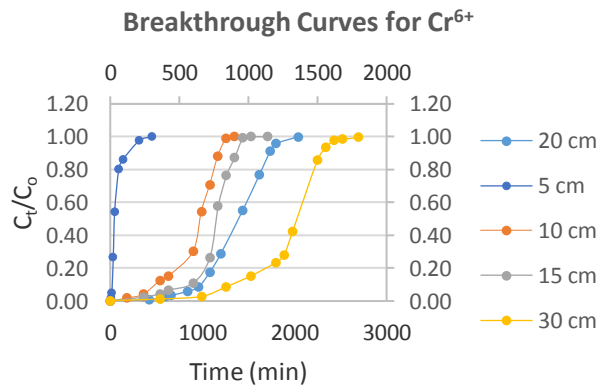
126

Fig. 7 Breakthrough curves at different bed-height

D. Study of Mathematical Models for the Adsorption of Cr^{6+}

Results from the bed-height experiments were fitted with the Thomas, Yoon-Nelson and Adams-Bohart models and the results are shown in Table 4. The R^2 value was used to determine the best fit model for the process. From the table it was noted that the Yoon-Nelson and Adams-Bohart models that gave the best fit for the adsorption of Cr^{6+} by BPs. The adsorption capacity q_T increased from bed height of 5 cm, but the increase was random with no specific trend, whereas k_T decreased at increasing bed height. Both increase and decrease in q_T and k_T, respectively, are in line with other research works done on the adsorption of toxic substances from aqueous solution using biomass materials [11, 32, 36, 40]. The Yoon-Nelson model is based on the assumption that the decrease in the rate of adsorption is directly proportional to the

adsorbate adsorption and breakthrough on the adsorbate. The model does not take in to any account the properties of the adsorbate, type of adsorbent used and physical features of the adsorption bed [31]. From Table 4, the parameters given by the model showed that k_{YN} decreased and τ increased as bed height increased. The increase in τ can be attributed to the increased availability in the biosorbent where the residence time of the passing influent stream is increased. The Adams-Bohart model is based on the surface reaction theory and it describes the relationship between C_t/C_o in relation to the time during continuous operation. The model assumes that equilibrium is not instantaneous and that it is used for describing the initial part of the breakthrough curve. The constant k_{AB} showed a decrease in value as height increased, while N_o was random with no specific trend. The R^2 value showed a good fit at bed heights of 10 cm, 15 cm, and 20 cm, therefore indicating that there was a reaction between the surface of the banana peels with the Cr^{6+} ions during continuous operation. A study done by (García-Sánchez et al. 2016) on the removal of fluoride from well water found that the Adams-Bohart model fitted well with their experimental data with k_{AB} decreasing as bed height increased from 7.0 to 13.5 cm and with an R^2 of 0.96 at 10 cm bed height.

IV. CONCLUSION

This study aimed to evaluate the use of banana peels as a biomass adsorbent in removing Cr^{6+} from water and to understand the kinetics of the process, the adsorption equilibrium, and the performance of BPs in a fixed-bed column. The Langmuir and Freundlich isotherms both fitted well with the experimental data for the process, indicating that adsorption is not only by a homogeneous equilibrium,

TABLE III: PARAMETERS OF COLUMN PERFORMANCE

C_o (mg/L)	C_B (mg/L)	H_T (cm)	t_B (min)	t_t (min)	t_U (min)	H_{UNB} (cm)	H_B (cm)	q_B (mg/g)
5	0.05	5	2.5	60	2.48	4.79	0.206	0.011
5	0.05	10	60	840	59.40	9.29	0.707	0.122
5	0.043	15	90	960	89.23	13.61	1.394	0.121
5	0.043	20	300	1620	297.42	16.33	3.672	0.296
5	0.0495	30	450	1800	445.55	22.57	7.426	0.291

TABLE IV: PARAMETERS FROM MATHEMATICAL MODELS FOR THE ADSORPTION OF CR^{6+}

Height, Z (cm)	Thomas Model Parameters			Yoon-Nelson Parameters			Adams-Bohart Parameters		
	K_T (mL/ min mg)	q_T (mg/g)	R^2	K_T (min^{-1})	τ (min)	R^2	K_{AB} (mL/ min mg)	N_o (mg/L)	R^2
5	0.0062	161.87	0.74	0.0296	67.20	0.77	0.0018	205777.30	0.47
10	0.0018	1178.83	0.91	0.0080	627.94	0.97	0.0011	388191.75	0.98
15	0.0017	959.96	0.88	0.0073	776.32	0.93	0.0011	301570.61	0.97
20	0.0012	1192.83	0.94	0.0060	1201.80	0.94	0.0008	383884.70	0.98
30	0.0011	827.71	0.95	0.0051	1345.80	0.97	0.0008	263620.93	0.94

TABLE V: GLOSSARY

Symbol	Description
C_o (mg/L)	Influent concentration
C_B (mg/L)	Effluent concentration
H_B (cm)	Height of used bed
H_T, Z (cm)	Total bed height
H_{UNB} (cm)	Height of unused bed
q_B (mg/g)	the adsorption capacity of adsorbent in a fixed-bed column
t_B (min)	Time equivalent to the breakthrough point
t_t (min)	Time equivalent to saturation point
t_U (min)	Time equivalent to a usable capacity of the bed up to the break-point

but also through a heterogeneuous multilayer process. The process favored the pseudo-second order kinetic and from column experiments, while the breakthrough point increased as bed height increased. The Yoon-Nelson and Adams-Bohart models fitted well with the column experiments.

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