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**MODELLING OF THE ADSORPTION KINETICS OF CHROMIUM (VI) USING WASTE BIOMATERIALS**

**MODELADO DE LA CINÉTICA DE ADSORCIÓN DEL CROMO (VI) UTILIZANDO BIOMATERIALES DE RESIDUOS**

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**Abstract**

The presence of heavy metals in bodies of water is an environmental problem, due to its toxicity and bioaccumulation in ecosystems. The adsorption kinetics of Cr (VI) was studied in a batch system using plantain peels and oil palm bagasse, evaluating the effect of temperature and amount of adsorbent on the process. The bioadsorbents were placed in contact with the solution of Cr (VI) at pH 2 and 100 ppm, taking samples at different times until equilibrium. The kinetics were adjusted to the pseudo-first order, pseudo-second order and Elovich models. It was established that: the increase in temperature favours the process for the plantain peel and decreases the capacity of adsorption for oil palm bagasse; and that a decrease in the amount of adsorbent favours the kinetics for both biomasses studied. The Elovich model describes the behaviour for the plantain peel, while the pseudo-first and pseudo-second-order models do it for the oil palm bagasse when the temperature varies. The Elovich model better approximates the kinetic data by varying the adsorbent dose of both biomasses, suggesting that the process of adsorption of Cr (VI) was controlled by chemisorption.

*Keywords:* biomass, chromium, plantain peel, oil cane bagasse, remotion.

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**Resumen**

La presencia de metales pesados en cuerpos de agua es un problema ambiental, por su toxicidad y bio-acumulación en los ecosistemas. Se estudió la cinética de adsorción de Cr (VI) en sistema por lotes usando cáscaras de plátano y bagazo de palma aceitera, evaluando el efecto de la temperatura y cantidad de adsorbente sobre el proceso. Se colocó en contacto los bioadsorbentes con la solución de Cr (VI) a pH 2 y 100 ppm tomando, muestras a diferentes tiempos hasta el equilibrio. Se ajustó la cinética a los modelos de pseudo-primer orden, pseudo-segundo orden y Elovich. Se estableció que: el aumento en la temperatura favorece el proceso para la cáscara de plátano y disminuye la capacidad de adsorción para el bagazo de palma aceitera; y que una disminución de la cantidad de adsorbente favorece la cinética para ambas biomásas estudiadas. El modelo de Elovich describe el comportamiento para la cáscara de plátano, mientras que los modelos de pseudo-primer y pseudo-segundo orden lo hacen para el bagazo de palma aceitera al variar la temperatura. El modelo de Elovich aproxima mejor los datos cinéticos variando la dosis de adsorbente de ambas biomásas, lo que sugirió que el proceso de adsorción de Cr(VI) estuvo controlado por quimisorción.

*Palabras clave:* biomasa, cromo, cáscara de plátano, bagazo de palma aceitera, remoción.

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## 1 Introduction

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The contamination of water with heavy metal ions has become a global environmental problem, due to the high toxicity exhibited by metal ions and their tendency to bioaccumulate in different ecosystems and living organisms, with the possibility to be transferred to the population human via the food chain (Marimón *et al.*, 2018, Wang *et al.*, 2017). According to the World Health Organization (WHO), the metals that cause most concern include lead, cadmium, zinc, nickel, cobalt, chromium, copper and mercury (Shah *et al.*, 2015). Being the chromium compounds of great industrial importance, and in turn highly toxic, the Cr (VI) ion stands out for its health effects, it can cause acute tubular and glomerular damage, be corrosive and cause chronic ulceration and perforation of the nasal septum, as well as the chronic ulceration of other skin surfaces (Paredes-Carrera, 2015). Occupational exposure to Cr (VI) can be a cause of asthma and exposure to it, particularly in the production and industries of chromium pigment, is associated with cancer of the respiratory tract (Memon *et al.*, 2009; Marichelvam & Azhagurajan, 2018).

The conventional method of chromium treatment consists of four steps: Reduction of Cr (VI) to Cr (III), precipitation of Cr (III) as Cr (OH)<sub>3</sub> at high pH, settlement of insoluble metal hydroxide, elimination of dehydrated sludge. The main shortcomings of conventional treatment include the high cost of safe disposal of the sludge, the expensive chemicals needed for the reduction of Cr (VI) and the incomplete reduction of Cr (VI) (Wen *et al.*, 2018; Memon *et al.*, 2009).

Nowadays, the scientific community has focused on the development of new processes and materials, which can be used to eliminate metal ions and contaminants from aqueous effluents or industrial waste, so adsorption is highlighted using agricultural materials for this purpose (Interiano-López *et al.*, 2019; Suárez-Vázquez *et al.*, 2019), among these we find, the orange peel, the melon peel, the lemon peel, the cassava peel, among others. Adsorbents of agricultural origin have polymeric groups that serve as active centres for the capture of metals (Marimón *et al.*, 2018, Bhatnagar *et al.*, 2015, Ramrakhiani *et al.*, 2016; Abdolali *et al.*, 2016). Plantain peels and oil palm bagasse can be obtained quickly and cheaply in markets, agricultural industries, chip factories, hotels, houses, farms, among others (Mahindrakar & Rathod.,

2018). Thus, the objective of the present article was the study of the adsorption kinetics of Cr (VI) in a batch system, evaluating the effect of temperature and quantity of material in such process using oil palm bagasse and plantain peels.

## 2 Materials and methods

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### 2.1 Preparation of the biosorbent

The plantain peels were supplied by local businesses in the city of Cartagena de Indias (Colombia). These were washed with abundant water to remove the impurities and pulp residues that could contain. Then a manual size reduction was made to facilitate its handling in the later stages. A final wash with distilled water was made to eliminate dirt or other compounds, and peels were dried in an oven for 8 h at 60 °C (Temesgen *et al.*, 2018). This material was ground and sieved to different sizes to choose those between 0.15 and 1 mm.

The palm bagasse was obtained as a waste product from the palm oil extraction process in María La Baja (Bolívar-Colombia). The fibril was subjected to three washes. In each wash, 3 litres of water per kilogram of biomass were used. The first one with water at room temperature; the second and third washes were done with hot water to remove the most considerable amount of oil retained in the fibre. The wet bagasse was placed in plates for drying in an oven for 8 h at 60 °C. After drying, the biomass was subjected to a cleaning process to remove husks and seeds. The reduction of the size of the palm bagasse was carried out using an electric mill. Then sieving was carried out, thus allowing to classify the particle sizes of interest.

### 2.2 Adsorption kinetics as temperature and dose of biomass changing

The experiments were carried out by putting in contact the studied biomass (plantain peel and palm bagasse) with 100 mL solution of Cr (VI) at a concentration of 100 ppm. The temperature was set at 30, 55 and 80 °C with 0.325 grams of biomass. Also, the amount of biomass was set at 0.6193, 0.325 and 0.0306 g using constant temperature (30°C), with an agitation of 200 rpm. Samples of metal remaining in solution were taken each hour until reaching 7h in order to know the behaviour of the kinetics of Cr (VI) removal.

The measurement of the metal in solution after the adsorption process was done by UV-Vis spectrometry at 540 nm (Neolaka *et al.*, 2018). The obtained data were adjusted to the experimental models using non-linear regression in the OriginPro software, having as an adjustment criterion the correlation coefficient  $R^2$ .

### 2.3 Adjustment to kinetic models

The adjustment of the kinetic data describing the rate at which the contaminant is removed from the treated effluent was carried out using the kinetic models of pseudo-first-order, pseudo-second-order and Elovich.

#### 2.3.1 Kinetic model of pseudo-first-order

Equation 1 describes the pseudo-first-order model (Tien & Ramarao, 2017).

$$q_t = q_e(1 - e^{-k_1 t}) \quad (1)$$

Where  $q_e$  and  $q_t$  are the adsorption capacities at equilibrium, at time  $t$ , respectively expressed in milligram of metal/gram of adsorbent (mg/g);  $k_1$  is the constant of pseudo-first order ( $\text{min}^{-1}$ ).

#### 2.3.2 Kinetic model of pseudo-second-order

Equation 2 describes the pseudo-second-order model (Simonin, 2016)

$$q_t = \frac{t}{1/(k_2 q_e^2) + t/q_e} \quad (2)$$

Where  $k_2$  is the second-order adsorption constant ( $\text{g}^{-1} \text{min}^{-1}$ ). This constant is obtained from the graph of  $t/(q_t \text{ vs } t)$ . Also,  $q_e$  and  $q_t$  are the adsorption capacities at equilibrium, at time  $t$ , described in Equation 1.

#### 2.3.3 Elovich model

Equation 3 describes the Elovich model (Largitte & Pasquier, 2016).

$$q_t = \frac{1}{\beta} \ln(\alpha \beta t) \quad (3)$$

Where  $\alpha$  is the initial adsorption rate of the Elovich model ( $\text{mg/g} \cdot \text{min}$ ), and  $\beta$  is the constant related to the extent of the surface coverage and the activation energy in the chemisorption ( $\text{g/mg}$ ).

## 3 Results and discussion

### 3.1 Kinetics varying temperature

Figures 1 and 2, show the adsorption kinetics and fitting to models, varying temperature for the plantain peel and oil palm bagasse, respectively.

The adsorption of the plantain peel for the Cr (VI) ions shows that it is considerably affected by the variation in temperature. The increase in adsorption capacity is distinguished as the temperature of the aqueous solution increases. This behaviour suggests that the active sites available for adsorption increased with temperature. Therefore, the adsorption of Cr (VI) by plantain peel seems to be an endothermic phenomenon (Kim & Kim., 2019). In addition, the increase in temperature can lead to some changes in the size of the pores that become larger, and to a relative increase in the diffusion of the Cr (VI) ions due to the decrease in the viscosity of the solution, which improves its exposure to adsorption in difficult access sites (Ben-Ali *et al.*, 2017). It is also established that equilibrium is achieved at approximately 200 min of operation for all the conditions studied.

From Figure 2, it is observed that an increase in temperature decreases the adsorption capacity. The trend suggests that adsorption of Cr (VI) with oil palm bagasse is exothermic (Mahindrakar & Rathod., 2018). The negative effect may be due to the damage of the biosorbent active sites, that is, the biosorption process that uses the biomass at a higher temperature may lead to physical damage to the biosorbent.

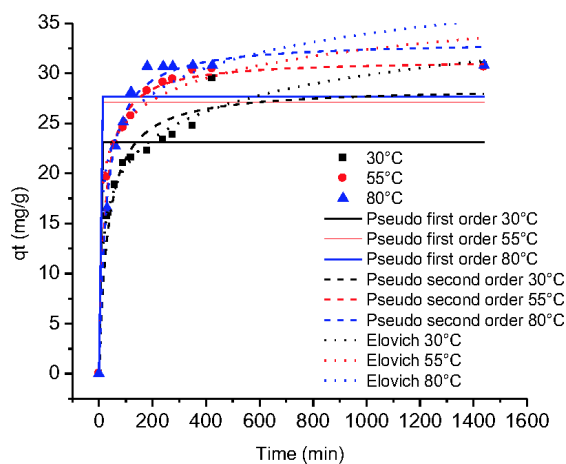


Fig. 1. Fitting of adsorption kinetics models of plantain peel changing the temperature.

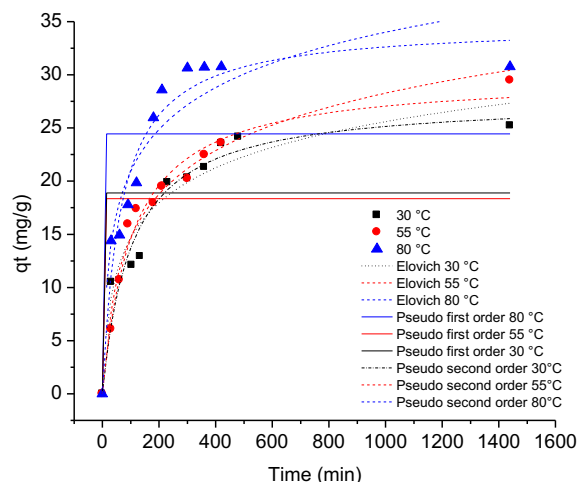


Fig. 2. Adjustment of adsorption kinetics models of palm bagasse changing temperature.

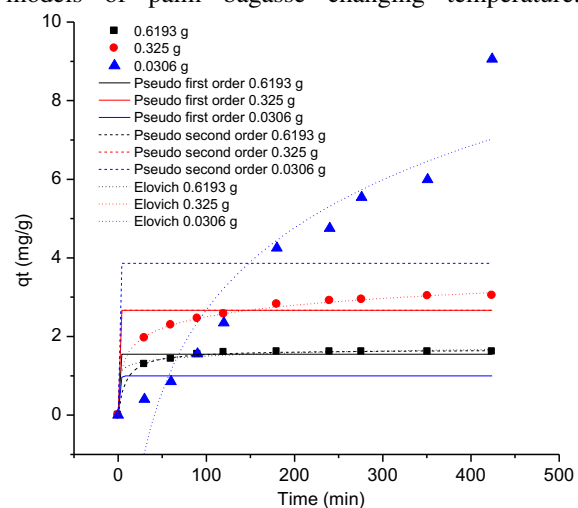


Fig. 3. Kinetic models adjustment of adsorption using plantain peel changing the dose of biomass.

The elimination of metal ions through the process of biosorption at room temperature was more advantageous (Rangabhashiyam & Balasubramanian., 2018).

### 3.2 Kinetics varying amount of biomass

Figures 3 and 4 show the adsorption kinetics and adjustment to models, varying the amount of biomass for the plantain peel and oil cane bagasse, respectively.

From the results, it is evidenced how the amount adsorbed per unit mass of adsorbent is considerably reduced in higher amounts of adsorbent, this may be due to the unsaturated biosorbent sites at a fixed

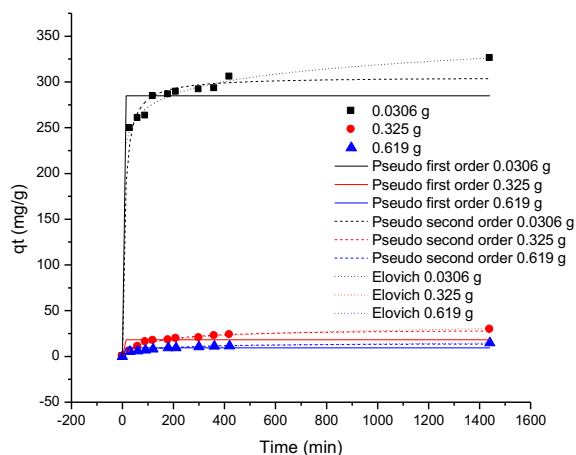


Fig. 4. Kinetic modelling of sorption of the palm bagasse, changing biomass amount.

concentration of Cr (VI) and the aggregation of biosorbents that leads to the decrease of the total surface area of it. In addition, an increase in the length of the diffusion path, makes some biosorption sites can overlap, decreasing the amount of biosorbed metal per unit mass, and therefore, it becomes more difficult to penetrate the Cr ions (VI) at the adsorption sites (Rangabhashiyam *et al.*, 2018, Denardin *et al.*, 2018, Herald *et al.*, 2018).

### 3.3 Kinetic model adjustment

In Table 1 the adjustment parameters made to the different biomasses from adsorption models by varying the temperature is shown; also Figures 2 and 3 show the fitting to the kinetics models, finding that the Elovich model is the one that best fits the plantain peel at the different temperatures studied ( $R^2$  higher than 0.97). The value of the initial biosorption rate ( $\alpha$ ) and the value of  $\beta$  were higher at lower solution temperatures, indicating that the biosorption performance coincides with a chemical biosorption nature and with adsorption with heterogeneous sites (Pandiarajan *et al.*, 2018). There is also evidence of relatively high values in the correlation coefficients in the pseudo-first and pseudo-second-order models. On the other hand, it is established that the pseudo-second-order and Elovich models present a better precision with the experimental data for oil palm bagasse at different temperatures, indicating that chemical adsorption is the speed limiting step. Besides, the values decrease with the increase of the temperature of the aqueous solution, which confirms that the biosorption capacity is disadvantaged at higher

Table 1. Adjustment parameters to adsorption models of Cr (VI) considering the change in temperature for plantain peel and palm bagasse.

Biomass	Temperature	Pseudo-first-order model	Pseudo-second-order model	Elovich model
Plantain peel	30 °C	$k_1: 0.02$	$k_2: 0.72$	$\alpha: 2.41$
		$q_e: 2.43$	$q_e: 2.71$	$\beta: 0.61$
		$R^2: 0.91$	$R^2: 0.95$	$R^2: 0.97$
	55 °C	$k_1: 0.02$	$k_2: 5.41$	$\alpha: 2.35$
		$q_e: 2.87$	$q_e: 2.66$	$\beta: 1.55$
		$R^2: 0.96$	$R^2: 0.83$	$R^2: 0.99$
	80 °C	$k_1: 0.02$	$k_2: 1.31$	$\alpha: 1.68$
		$q_e: 3.05$	$q_e: 3.45$	$\beta: 0.42$
		$R^2: 0.99$	$R^2: 0.99$	$R^2: 0.98$
Palm bagasse	30 °C	$k_1: 0.01$	$k_2: 0.76$	$\alpha: 1.63$
		$q_e: 2.42$	$q_e: 2.94$	$\beta: 0.09$
		$R^2: 0.94$	$R^2: 0.96$	$R^2: 0.96$
	55 °C	$k_1: 0.01$	$k_2: 0.24$	$\alpha: 1.55$
		$q_e: 2.22$	$q_e: 2.75$	$\beta: 0.06$
		$R^2: 0.97$	$R^2: 0.98$	$R^2: 0.98$
	80 °C	$k_1: 0.04$	$k_2: 0.02$	$\alpha: 12.1$
		$q_e: 0.65$	$q_e: 0.71$	$\beta: 1.31$
		$R^2: 0.93$	$R^2: 0.97$	$R^2: 0.98$

temperatures, maybe due to the oxidation of free radical (Denardin *et al.*, 2018; ul Haq *et al.*, 2019).

From Table 2 and Figures 1 and 2, it is evident that the Elovich model presents a better accuracy on the kinetic data for plantain peel and oil palm bagasse with a correlation coefficient of 0.99 for all the biomass quantities studied. Based on the values of  $\alpha$  and  $\beta$ , it can be suggested that in the process of adsorption of Cr(VI), a chemisorption process was involved. However, the correct adjustment by the pseudo-first and pseudo-second-order models cannot be ruled out. In such a way that the adsorption capacity is favoured at smaller quantities, possibly due to the overcrowding of the biosorbent particles as a result of the excess of the amount of biosorbent that subsequently leads to the superposition of the biosorption sites of plantain peel and oil palm bagasse (Wang *et al.*, 2018; Guerrero *et al.*, 2015; Moyo *et al.*, 2015).

## Conclusions

From the kinetic study of adsorption of Cr (VI) in a batch system using plantain peel and oil palm bagasse, it was found that an increase in temperature favours the adsorption of Cr (VI) to the plantain peel. Which suggests that the process is governed by an endothermic nature, while the adsorption capacity is affected at higher temperatures using the oil palm bagasse, suggesting that the adsorption is exothermic. According to the adjustment to kinetic models of adsorption varying the temperature, it was determined that the Elovich model better describes the experimental data for the plantain peel, corroborating that at a lower temperature the parameter  $q_e$  was higher. As for the palm bagasse, it was found that there was a better adjustment of the pseudo-second-order models and Elovich, which shows a decrease in the values  $q_e$  with the increase in the temperature of the

Table 2. Adjustment parameters to adsorption models of Cr (VI) considering the change in the dose of biomass for plantain peel and palm bagasse.

Biomass	Dose of biomass	Pseudo-first-order model	Pseudo-second-order model	Elovich model
Plantain peel	0.6193 g	$k_1$ : 6.19	$k_2$ : 3.45	$\alpha$ : 6.91
		$q_e$ : 1.53	$q_e$ : 1.53	$\beta$ : 50.6
		$R^2$ : 0.95	$R^2$ : 0.95	$R^2$ : 0.99
	0.325 g	$k_1$ : 0.02	$k_2$ : 5.41	$\alpha$ : 2,35
		$q_e$ : 2.87	$q_e$ : 2.66	$\beta$ : 1.55
		$R^2$ : 0.96	$R^2$ : 0.83	$R^2$ : 0.99
	0.0306 g	$k_1$ : 0.09	$k_2$ : 5.82	$\alpha$ : 0.33
		$q_e$ : 2.25	$q_e$ : 3.86	$\beta$ : 0.07
		$R^2$ : 0.97	$R^2$ : 0.07	$R^2$ : 0.99
Palm bagasse	0.6193 g	$k_1$ : 0.01	$k_2$ : 0.03	$\alpha$ : 4.16
		$q_e$ : 0.98	$q_e$ : 1.17	$\beta$ : 0.05
		$R^2$ : 0.95	$R^2$ : 0.97	$R^2$ : 0.98
	0.325 g	$k_1$ : 0.01	$k_2$ : 0.24	$\alpha$ : 1.55
		$q_e$ : 2.22	$q_e$ : 2.75	$\beta$ : 0.06
		$R^2$ : 0.97	$R^2$ : 0.98	$R^2$ : 0.98
	0.0306 g	$k_1$ : 0.06	$k_2$ : 29.8	$\alpha$ : 0.49
		$q_e$ : 28.0	$q_e$ : 3699	$\beta$ : 1388
		$R^2$ : 0.97	$R^2$ : 0.99	$R^2$ : 0.99

aqueous solution, ratifying that the biosorption capacity is disadvantaged at higher temperatures. On the other hand, the Elovich model showed a better fit in the kinetics of both biomasses studied by varying the amount of adsorbent, which suggested that in the process of removal of Cr (VI) a chemisorption process was involved.

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### Nomenclature

$q_e$  adsorption capacities at equilibrium  
 $q_t$  adsorption capacities at time  $t$

$k_1$  constant of pseudo-first order  
 $T$  time  
 $k_2$  second-order adsorption constant  
 $A$  initial adsorption rate of the Elovich model  
 $B$  constant related to the extent of the surface coverage and the activation energy

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