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Determination of Kinetic Parameters in the Biosorption of Chromium (VI) in Aqueous Solution

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Abstract

The contamination of aquatic bodies by heavy metals is a growing environmental problem, making more critical the study and development of new technologies and materials that can be used for the removal of this type of pollutants. Thus, adsorption arises using residual materials as a sustainable alternative for the solution to this problem. In the present study, the use of plantain peels in the adsorption of Cr (VI) in a batch system is proposed, establishing the kinetics of the process at different temperature conditions, particle size and amount of adsorbent. The fit of the data was done using the theoretical models of pseudo-first-order, pseudo-second-order and Elovich. From the data, it is established that the pseudo-second-order and Elovich models show a better adjustment, so that the adsorption in the material occurs on two adsorption sites and that such process is related to chemical adsorption. The maximum

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adsorption capacity of Cr (VI) was found at a condition of 0.0306 g, 0.6775 mm and 55°C at a time of 420 min establishing the efficient use of plantain peels for the removal of the ion metallic in the studio.

Keywords: Kinetic models; metallic ion; peels; removal; Chromium (VI) .

Determinación de parámetros cinéticos en la biosorción de Cromo (VI) en solución acuosa

Resumen

La contaminación de cuerpos acuáticos por metales pesados es un problema ambiental creciente haciendo cada vez más importante el estudio y desarrollo de nuevas tecnologías y materiales que puedan ser usados para la remoción de este tipo de contaminantes. Así surge la adsorción usando materiales residuales como una alternativa sostenible para la solución de esta problemática. En el presente estudio se propone el uso de las cáscaras de plátano en la adsorción de Cr (VI) en un sistema por lotes estableciendo la cinética del proceso a diferentes condiciones de temperatura, tamaño de partícula y cantidad de adsorbente. El ajuste de los datos fue hecho usando los modelos teóricos de pseudo-primer orden, pseudo-segundo orden y Elovich. De los datos se establece que son los modelos de pesudo-segundo orden y Elovich los que muestran un mejor ajuste determinado así que la adsorción en el material se da sobre dos sitios de adsorción y que tal proceso está relacionado con una adsorción química. La máxima capacidad de adsorción de Cr (VI) fue encontrada a una condición de 0.0306 g, 0.6775 mm y 55°C a un tiempo de 420 min estableciendo el uso eficiente de cáscaras de plátano para la remoción del ion metálico en estudio.

Palabras clave: Modelos cinéticos; ion metálico; cáscaras; remoción; Cromo (VI).

1 Introduction

One of the main challenges many countries around the world are facing is the decrease in the supply of safe and clean drinking water. Similarly, the presence of heavy metals in high concentrations is an environmental problem due to the adverse effects for human health, as well as for ecosystems [1].Residues from industries such as mining, electroplating, tanneries, textiles, ceramics, photography, gravure and battery manufacturing, contain moderate to excessive amounts of

hexavalent chromium compounds beyond the conventional legal limit of 0.1 mg/L [2],[3],[4],[5].

Cr (VI) can exist in the form of dichromate ($Cr_2O_7^{2-}$), hydrochromate ($HCrO_4^-$) or chromate (CrO_4^{2-}) depending on the pH and redox potential of the medium [6]. The treatment of these effluents is necessary because, in humans, Cr (VI) causes lung cancer, ulcers, perforations of the nasal septum and damage to the kidneys and liver [7]. Various physicochemical methods are used for the removal of heavy metals from aqueous solutions such as solvent extraction [8], ion exchange [9], chemical precipitation [10], reverse osmosis [11], and membrane separation [12]. However, some methods require energy, chemical additives, and have a limitation against concentrations below 50 ppm [13]. So, the development of efficient and low-cost separation processes is, therefore, of great importance.

Accordingly, bio-adsorption is a viable alternative to achieve the desired objective, which again requires the development of new adsorbents with efficient adsorption capacities [4]. The use of biomass from agricultural residues as adsorbents has been widely researched, due to the low cost, immediate availability and high removal efficiency. Various bio-adsorbents such as plantain peels [6],[13], cassava shells [14], orange peels [15], rice husk [5], cocoa peels [16], tea waste [17], potato husks [1], lemon [18], coffee pulp [19], and other husks were tested for the removal of these contaminants [20],[21]. Therefore, in the present study, the adsorption kinetics of Cr (VI) in aqueous media were investigated using plantain peels, at different temperature conditions (°C), adsorbent dose (g) and particle size.

2 Methodology

2.1 Preparation of biomass and solution

The plantain peels came from farms in Santa Rosa and Villanueva (Colombia), being waste products in agro-industrial production processes. The plantain peels were subjected to a washing process, then, a manual size reduction was made for the subsequent stages and washed again with distilled water to remove tannins and resins. The husks were dried for 8h at 60 °C in an oven. After the drying process, the biomass was reduced

in size and classified in a shaker type sieve through a series of sieves to choose the appropriate particle size for the adsorption process. The sizes used were: 0.135,0.355,0.6775,1 and 1.219 mm, as shown in the 1 [22].

The synthetic solution of Cr (VI) was prepared using 0.2828 g of potassium dichromate ($K_2Cr_2O_7$) in 1 L of deionized water to obtain a concentration of 100 ppm of the solution, which pH was 2 [23],[24].

2.2 Kinetic essays

The design of experiments was carried out using Statgraphics Centurion XVI.II Software for Windows. The design used was a continuous linear factor on the response surface (2^2-star central compound). The essays were carried out in a batch system with a solution of 100 mL of Cr (VI) and a fixed agitation of 200 rpm, setting the initial concentration at 100ppm, varying the temperature of the adsorbent amount and particle size, according to the ranges established in Table 1, for a total of 15 experiments with a rotability factor of 1.6818. Samples were taken at different time intervals for 7 h.

Table 1: Experimental ranges and levels of variation for the independent variables.

Independent variable	**Units**	**Range and level**				
		$-\alpha$	**-1**	**0**	**1**	$+\alpha$
Temperature	°C	30	40	55	70	80
Adsorbent Dose	g	0.031	0.15	0.325	0.5	0.619
Particle size	mm	0.135	0.355	0.6775	1	1.219

The final concentration measurements were made on an ultraviolet and visible spectrometer (UV/VIS) at 540 nm [25]. The data obtained were adjusted to the adsorption models through the software Origins Pro 8®.

2.3 Fit to the kinetic models

In order to determine and interpret the mechanisms in the process of adsorption of metal ions and the main parameters that govern the sorption kinetics, the kinetic data obtained empirically were adjusted to the models of pseudo-first-order, pseudo-second-order and Elovich.

The pseudo-first-order kinetic model

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

Where q_e and q_t are the adsorption capacities at equilibrium, in a time t, respectively expressed in (mmol/g), while k_1 is the pseudo-first-order constant (min^{-1}) [26].

The pseudo-second-order kinetic model

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

Where k_2 is the adsorption constant of second-order ($g^{-1}min^{-1}$), this constant is obtained from the graphic t/qt vs t [27].

Elovich model

$$q_t = \frac{t}{\beta} ln(\alpha * \beta * t) \tag{3}$$

Where α is the initial adsorption rate of the Elovich model (mg/g min), and β is the constant related to the extent of surface coverage and the activation energy in chemisorption (g/mg) [28].

3 Results and discussion

The adsorbent dose is one of the crucial parameters studied while conducting batch mode studies. The Figure 1 show the kinetic at different conditions of biomass quantity and particle size, setting the temperature at 55°C; it was established that the highest capacity is reached using the least amount of biomass.

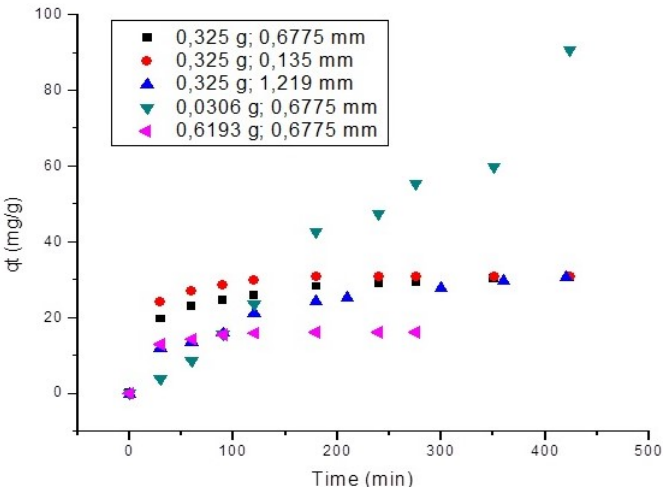

Figure 1: Adsorption kinetics of Cr (VI) with plantain peels at 55°C.

The adsorption of Cr (VI) was maximum at 0.03 g of adsorbent dose, and when the adsorbent dose was increased, the adsorption capacity decreased progressively. While having 0.03 g of adsorbent dose, 91.56 mg/g of adsorption was achieved, which was reduced to 17.79 mg/g for the adsorbent dose of 0.62 g. The low adsorption at higher doses may be due to the aggregation of adsorbent particles and the low driving force that leads to the adsorption process when the concentration of Cr (VI) decreases because an increasing number of adsorption sites cannot come into contact with the molecules of adsorbate [29],[30]. Additionally, the sites of unsaturated biosorbents at a fixed concentration of Cr (VI) and the aggregation of more adsorbent lead to a decrease in its total surface area [31].

The Figure 2 shows the kinetics at different conditions of biomass quantity and particle size, setting the temperature at 40°C. It was determined that all the materials reach equilibrium after 2 hours of processing.

From Figure 2 it is established that the best adsorption capacity of Cr (VI) is given when the amount of biomass is 0.15 g, achieving up to 36.4 mg/g of adsorption that was reduced to 19.7 mg/g for 0.5g of the adsorbent. As the phenomenon presented at 55°C, there is also a decrease

in the adsorption capacity when increasing the adsorbent dose. Which may be since an increasing number of active adsorption sites cannot come into contact with a low concentration of Cr (VI) ions, thus decreasing the total surface area of the adsorbent [31],[32].

As the adsorbent dose was increased the adsorption capacity decreased, and this may be due to the decrease in the total adsorption surface area available for the Cr (VI) ion resulting from the superposition or aggregation of the adsorption sites. Which, by increasing the mass of adsorbent, the amount of contaminant adsorbed per unit mass of the adsorbent is reduced. Consequently, it causes a decrease in the q_e value [6].

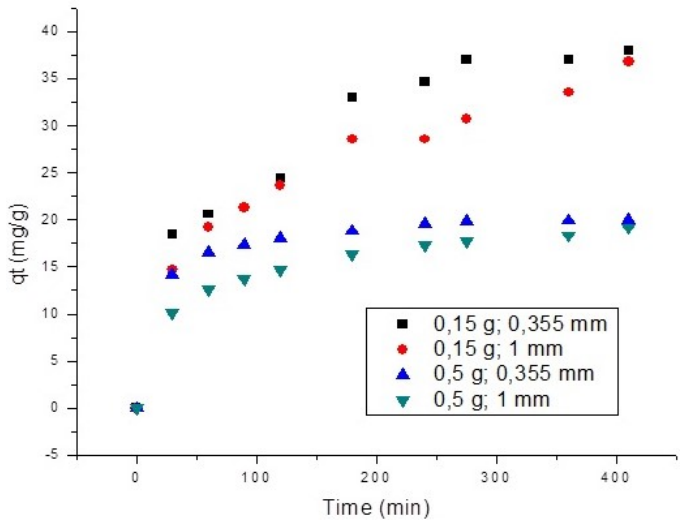

Figure 2: Adsorption kinetics of Cr (VI) with plantain peels at 40 °C.

The Figure 3 shows the kinetics at different conditions of biomass quantity and particle size, setting the temperature at 70°C. It was found that the particle size of the biomass does not affect the process, while the amount of biomass does.

The proportional increase in adsorption capacity with the dose of adsorbent, shown in Figure 3, is related to the increase in the total area available surface of contact, and therefore, the higher number of ion exchange sites available for the interaction with chromium ions [6],[15],[33].

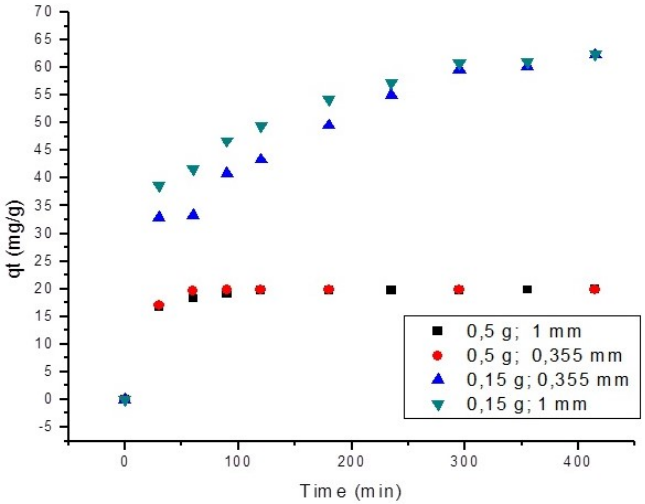

Figure 3: Adsorption kinetics of Cr (VI) with plantain peels at 70 °C.

The Figure 4 presents the kinetics at different conditions of biomass quantity and particle size varying the temperature at 80 and 30°C. It was found that the initial rate of adsorption is higher than at 80°C, making it easier for this condition to reach equilibrium [34]. However, after 7 hours of the process, both conditions achieve the same adsorption capacity.

For the conditions studied in the Figure 4 and from the Table 2, the research established that the pseudo-second-order and Elovich models have better precision in predicting the experimental data in the adsorption study of Cr (VI) using plantain peel. On the other hand, from the value of the constant velocity k_2, it is established that the limiting step of the velocity for the removal can be attributed to the chemical adsorption where the formation of the covalent bond is due through the exchange of electrons between the metal ions and the binding sites of the adsorbent.

This effect occurred through the complexation of the ions with the functional groups available in the plantain peel [35]. Also, the initial adsorption rate (α) is high, which implies the predominance of the adsorption of Cr (VI) when supposing adsorption in multilayers. In which each layer shows several energies of activation for chemisorption [36].

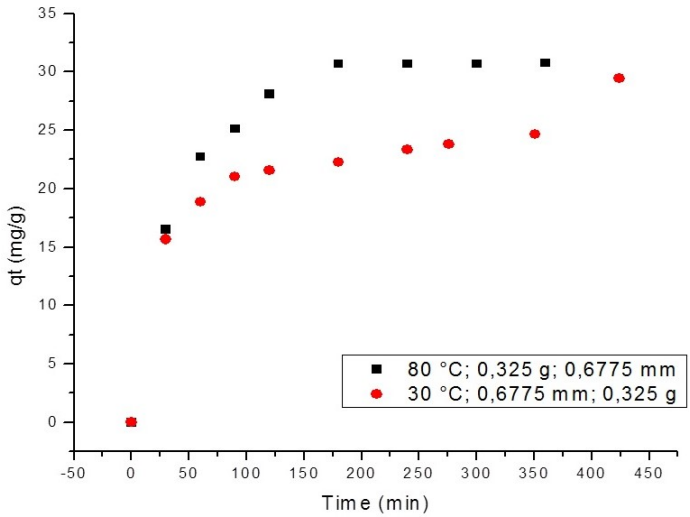

Figure 4: Adsorption kinetics of Cr (VI) with platain peels.

The Table 2 contains the adjustments to the kinetic models of pseudo-first-order, pseudo-second-order and Elovich in the adsorption of Cr (VI) with the plantain peels and the parameters found.

The fit between the experimental results and the standard parameters applied was compared with the correlation coefficients. From the data reported in the Table 2, it can be inferred that the Elovich and pseudo-second-order models are the ones that best fit the experimental data. This fact is because the reported R^2 is close to the unit; in the same sense, the q_e reported by the pseudo-second-order model is closer to the values obtained experimentally. The adjustment of these two models to the data indicates that the process is controlled by chemisorption or ion exchange. This phenomenon is caused by the presence of the different functional groups typical of biomasses of lignocellulosic origin [37],[38],[39].

Table 2: Fit to the kinetic models of adsorption.

Conditions	Pseudo-first order model	Pseudo-second orden model	Elovich Model
55 °C; 0.325 g; 0.6775 mm	k_1: 0.03 q_e: 28.80 R^2: 0.96	k_2: 1447.63 q_e: 31.49 R^2: 0.99	α: 15.511 β: 0.24 R^2: 0.99
55 °C; 0.325 g; 0.1351 mm	k_1: 0.05 q_e: 30.25 R^2: 0.99	k_2: 3241.55 q_e: 31.84269 R^2:0.99	α: 1887.61 β: 0.40 R^2:0.99
55 °C; 0.325 g; 1.219 mm	k_1: 0.01 q_e: 30.09 R^2: 0.97	k_2: 533.49 q_e: 37.31 R^2:0.98	α: 0.89 β: 0.13 R^2:0.98
55 °C; 0.0306 g; 0.6775 mm	k_1: 9.84E-6 q_e: 20250.31 R^2: 0.97	k_2: 8.02E17 q_e: 38.63 R^2:0.070	α: 0.733 β: 0.033 R^2:0.88
55 °C; 0.6193 g; 0.6775 mm	k_1: 0.051 q_e: 15.90 R^2: 0.99	k_2: 524.10 q_e: 16.86 R^2:0.99	α: 506.14 β: 0.69 R^2:0.99
40 °C; 0.15 g; 0.355 mm	k_1: 0.04 q_e: 19.20 R^2: 0.98	k_2: 600.44 q_e: 20.60 R^2:0.99	α: 58.17 β: 0.45 R^2:0.99
40 °C; 0.15 g; 1 mm	k_1: 0.02 q_e: 17.66 R^2: 0.96	k_2: 224.99 q_e: 19.94 R^2:0.99	α: 2.18 β: 0.23 R^2:0.99
40 °C; 0.5 g; 0.355 mm	k_1: 0.01 q_e: 36.65 R^2: 0.94	k_2: 1352.70 q_e: 42.89 R^2:0.96	α: 1.96 β: 0.12 R^2:0.98
40 °C; 0.5 g; 1 mm	k_1: 0.01 q_e: 32.97 R^2: 0.94	k_2: 901.26 q_e: 39.39 R^2:0.97	α: 1.44 β: 0.12 R^2:0.99
70 °C; 0.5 g; 1 mm	k_1: 0.06 q_e: 19.60 R^2: 0.99	k_2: 1361.31 q_e: 20.29 R^2:0.99	α: 479992.19 β: 0.94 R^2:0.99
70 °C; 0.5 g; 0.355 mm	k_1: 0.06 q_e: 19.91 R^2: 0.99	k_2: 1686.59 q_e: 20.53 R^2:0.99	α: 5.28E7 β: 1.17 R^2:0.99
70 °C; 0.15 g; 0.355 mm	k_1: 0.01 q_e: 58.51 R^2: 0.92	k_2: 5938.89 q_e: 67.62 R^2:0.96	α: 3.89 β: 0.079 R^2:0.98
70 °C; 0.15 g; 1 mm	k_1: 0.02 q_e: 58.02 R^2: 0.93	k_2: 9661.72 q_e: 64.37 R^2:0.98	α: 13.33 β: 0.10 R^2:0.99
80 °C; 0.325 g; 0.6775 mm	k_1: 0.03 q_e: 24.35 R^2: 0.92	k_2: 768.73 q_e: 27.12 R^2:0.96	α: 6.05 β: 0.24 R^2:0.97
30 °C; 0.325 g, 0.6775 mm	k_1: 0.023 q_e: 30.58 R^2: 0.99	k_2: 1320.01 q_e: 34.42 R^2: 0.99	α: 4.56 β: 0.17 R^2: 0.98

4 Conclusions

The effect of temperature, adsorbent dose and particle size of the Cr (VI) removal process was evaluated using plantain peel as a biosorbent on the adsorption kinetics; it was found that the adsorption of the ions is rapid in the first minutes. The research also concluded that the particle size has no significant effect on the process, at 40 and 55°C greater contaminant elimination capacity is achieved as the amount of adsorbent decreases, and at 70°C the adsorption capacity increases as the biosorbent dose is increased. Additionally, from the fit to the kinetic models, it was found that Elovich and pseudo-second-order adjust the experimental data, so the removal is attributed to the chemical interaction between the metal ions and the functional groups on the surface of the biosorbent. Finally, from the experimental results, the plantain peel can be used as an effective adsorbent for the removal of Cr (VI) from the aqueous solutions.

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