Journal of Engineering and Technology for Industrial Applications

ITEGAM-JETIA

Manaus, v.6 n.24, p. 21-26, July/Aug., 2020

DOI: https://doi.org/10.5935/jetia.v6i24.682

ISSN ONLINE: 2447-0228

RESEARCH ARTICLE OPEN ACCESS

ADSORPTION KINETICS OF PHENOL FROM AQUEOUS SOLUTION USING SUGARCANE BAGASSE ASH AS LOW-COST ADSORBENT MATERIAL

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ARTICLE INFO

Article History
Received: July 27th, 2020
Accepted: August 11th, 2020
Published: August 31th, 2020

Keywords:
Kinetic,
Adsorption,
Phenol,
Sugarcane bagasse ash.

ABSTRACT

The sugarcane bagasse fly ash was used to evaluate its adsorption behavior for phenol removal from aqueous solution at three different temperatures. Adsorption tests were performed in batch reactors and also in fixed-bed columns. Pseudo-first order, pseudo-second order and intraparticle diffusion kinetic models were applied to describe adsorption kinetics in batch systems. The pseudo-second model fitted appropriately the obtained experimental data at the three different temperatures tested. Thomas, Yoon-Nelson, Adams-Bohart and Dose-Response mathematical models were tested for describing phenol adsorption in dynamic systems (fixed bed columns). Experimental data were well-fitted to the non-linear form of all these models with high regression coefficients.

I. INTRODUCTION

Phenols are organic compounds produced and used in different industry such as coke, oil refineries, pharmaceutical and phenolic resin plants [1]. Phenols are widely used and, consequently, they are frequently detected in water. They are some of the most dangerous pollutants and pose an important threat to both environment and human health, even at low concentrations [2-4]. The discharge of phenolic waste into watercourses affects to the flora and fauna and the ingestion of small amounts beings may cause affections human health [5]. For all these reasons, phenols are pollutants classified as pollutants of high priority. Consequently, their treatment is very important for safe discharge to the environment. Various methods have been used to remove phenol from aqueous solutions including adsorption, ion exchange, oxidation, precipitation and solvent extraction [6]. Especially adsorption is an interesting technique since it has low cost and is greatly efficient [7], in wastewater treatment. In addition, the cost of operation of the adsorption procedure can be significantly decreased with the use of solid waste adsorbents.

Sugarcane bagasse is a solid waste produced as a consequence of sugar production in industries. The high amounts of sugarcane bagasse generated after the extraction of sucrose from sugarcane has to be treatment and safely disposal. Sugarcane bagasse has been reported as suitable adsorbent material for wastewater treatment and particularly for metal cations removal from wastewaters [8-10]. For example, recently, [10] prepared an activated carbon from sugarcane bagasse and investigated kinetics and mechanisms of Cr(VI) adsorption onto the carbonaceous material.

Sugarcane bagasse is frequently used as fuel in combustion systems generating new solid waste, mainly two types of ashes: the sugarcane bagasse bottom ashes (SBBA) and the sugarcane bagasse fly ashes (SBFA). Some researchers have analyzed the use of SBFA in civil engineering [11-14]. However, finding other recycling processes to give value to this waste is of great interest. Researchers as [15] performed a complete characterization of sugarcane bagasse ash for its use as adsorbent material. Now, in the present study, the application of SBFA as adsorbent material for the removal of phenol from wastewater was studied. Previously, [16] already used SBFA as adsorbent for

the removal of phenol from aqueous solution. The previous study showed that the SBFA was an effective adsorbent for the removal of phenol from aqueous solution. However, in the work of [16] only adsorption tests in batch systems were carried out.

In this work, the sorption kinetics of phenol in aqueous solution, in batch and column systems, using SBFA was investigated. Authors consider that the study of kinetics in adsorption processes is important since it clarifies the mechanism of adsorption.

II. DEVELOPMENT

II.1 MATERIAL

Sugarcane bagasse fly ash was collected from the particulate collection devices attached to the boiler furnaces in a Sugar Factory from Villa Clara, Cuba. Then, the ash was sieved using a 1 mm sieve, dried at 110 °C for 6h and stored in vacuum desiccator, without any pretreatment, for further analyses. A complete characterization of SBFA was performed in a previous work [15] and a summary of the main composition and physical properties was reported in Table 1.

Table 1: Chemical and physical properties of SBFA.

Chemical properties									**Physical properties**		
Si (%)	**Al (%)**	**Fe (%)**	**Ca (%)**	**Mg (%)**	**K (%)**	**Organic matter (%)**	**Inorganic matter (%)**	**pH_{pzc}**	**Bulk density (g/cm^3)**	**Real density (g/cm^3)**	**Surface area (m^2/g)**
29.54	0.36	0.60	7.01	1.14	5.81	18.02	81.98	7.40	0.53	2.26	79.11

Source: Adapted from [15].

II.2 BATCH ADSORPTION EXPERIMENTS

The pH of the aqueous solution of phenol was 7.0, this value has been used in other studies [17] and the work temperatures were 27; 40 and 60 °C. For each experimental test, 50 mL of aqueous solution with a phenol concentration of 50 mg/L was taken in a 250 mL flask containing 5g of adsorbent; the solution was stirred at a constant shaking rate for 50 minutes and the supernatant liquid was collected at different contact times and analyzed for the residual concentration of phenol using the method ASTM Designation: D 1783 – 01 Standard Test Methods for Phenolic Compounds in Water [18], using a spectrophotometer GENESYS 10S UV-VIS, Thermo Scientific.

Pseudo-first-order, pseudo-second-order and intraparticle diffusion models, were used for analyzing characteristic kinetic parameters of phenol adsorption onto SBFA. A summary of the equations of the models is presented in Table 2.

Table 2: Summary of the equations of the kinetic models applied in this work for describing phenol adsorption onto SBFA in a batch system.

	Pseudo-first-order model	**Pseudo-second-order model**	**Intraparticle diffusion model**
Equation	$\frac{dq_t}{dt} = k_1(q_e - q_t)$	$\frac{dq_t}{dt} = k_2(q_e - q_t)^2$	$q_t = k_{(id)} t^{1/2} + C$
	$\mathrm{Ln}\left(\frac{q_e - q_t}{q_e}\right) = -k_1 t$	$\frac{t}{q_t} = \frac{1}{k_2 \cdot q_e^2} + \frac{t}{q_e}$	
Parameters	where q_e and q_t are the amounts of adsorbate uptake per mass of adsorbent, mg/g, at equilibrium and at any time t (min), respectively, and $k_1(min^{-1})$ is the rate constant of the pseudo-first order equation.	where k_2 is the pseudo-second-order rate constant, g/(mg·min).	where, $k_{(id)}$ is the intra-particle diffusion rate constant ($mg/g \cdot min^{1/2}$) and C (mg/g) is a constant that gives idea about the thickness of the boundary layer.
Reference	[19]	[20-21]	[22]

Source: [19-22].

II.3 COLUMN ADSORPTION EXPERIMENTS

Column adsorption studies were developed in a glass column having an internal diameter of 2.3 cm and 44 cm height at 27 ºC. Experiments were carry out, changing the absorbent mass (47.5 y 65.0 g), from these masses was obtained a bed height of 25.0 and 32.5 cm, respectively.

Inlet phenol concentration was 5.0 mg/L and the flow rate was 0.012 L/min. The phenol samples were collected every 10 min and were prepared for concentration determination analyses by spectrophotometry.

Different column kinetic models were used in this study to describe the dynamic behavior of adsorption in the fixed-bed column. A summary of the equations of the models is presented in Table 3.

Table 3: Summary of the equations of the kinetic models applied in this work for describing phenol adsorption onto SBFA in a fixed-bed column.

Equation	Parameters	Reference
$\frac{C}{C_0} = e^{K_{AB}C_0 t - \frac{K_{AB}N_0 Z}{v}}$	K_{AB}: kinetic constant, $L \cdot mg^{-1} \cdot min^{-1}$ N_0: volumetric adsorption capacity, $mg \cdot L^{-1}$ v: linear flow rate, $cm \cdot min^{-1}$ Z: height in the column, cm	Bohart and Adams [23]
$\frac{C}{C_0} = \frac{1}{1 + \exp\left(\frac{K_{Th}}{Q}(q_0\, m - C_0\, V_{eff})\right)}$	K_{Th}: rate constant, $mL\ min^{-1}\ mg^{-1}$ q_0: maximum concentration of solute in the solid phase, $mg \cdot g^{-1}$	Thomas [24]
$\frac{C}{C_0} = \frac{1}{1+\exp(K_{YN}(\tau - t))}$	K_{YN}: rate constant, min^{-1} τ: time required to retain 50% of the initial adsorbate	Yoon and Nelson [25]
$\frac{C}{C_0} = 1 - \frac{1}{1+\left(\frac{C_0\, V_{eff}}{q_0\, m}\right)^a}$	a: empiric parameter	Yan et al. [26]

Source: [23-26].

III. RESULTS AND DISCUSSIONS

III.1 BATCH ADSORPTION STUDY

The relation between the adsorption capacities in function of contact time for different temperatures is shown in Figure 1. The adsorption capacity increased 1.4 times as the contact time increased from 5 to 15 minutes. Also, in the first 5 minutes about 65%, 78% and 65% of the equilibrium adsorption capacity was reached for 27, 40 and 60 ºC, respectively. Qadeer and Rehan reported the phenol adsorption with activated commercial coal and found that the time to reach the equilibrium was five minutes [27-29]. This can be explained by high availability of active sites on the SBFA surface at the beginning of the process [7].

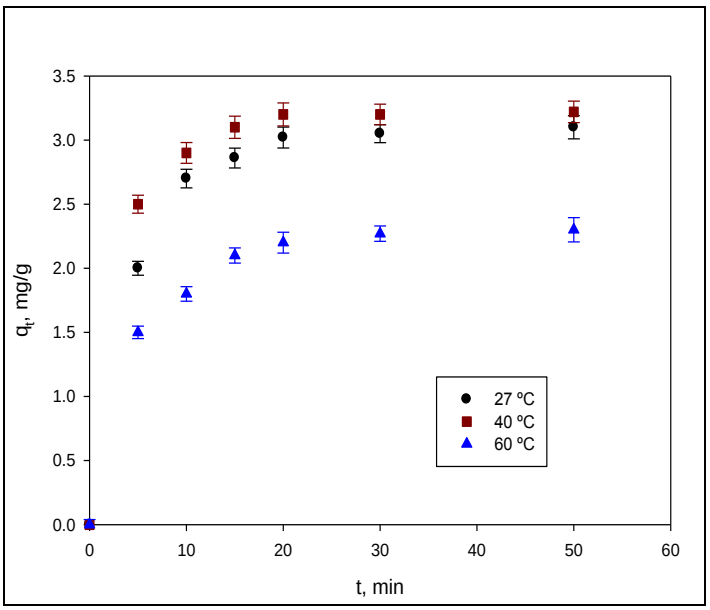

Figure 1: Experimental adsorption capacities in function of contact time at different temperatures.
Source: Authors, (2020).

The experimental results provided in Figure 1 have been fitted by linear regression to the three kinetic models considered in material and methods section: pseudo-first order, pseudo-second order and intraparticle difussion models. Figures 2, 3 and 4 show the results of the fit at the three temperatures tested.

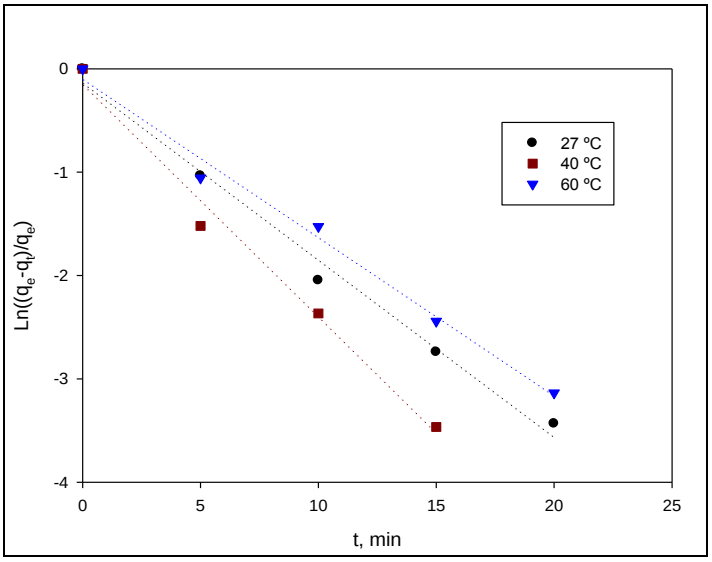

Figure 2: Pseudo-first-order adsorption kinetics of phenol on SBFA.
Source: Authors, (2020).

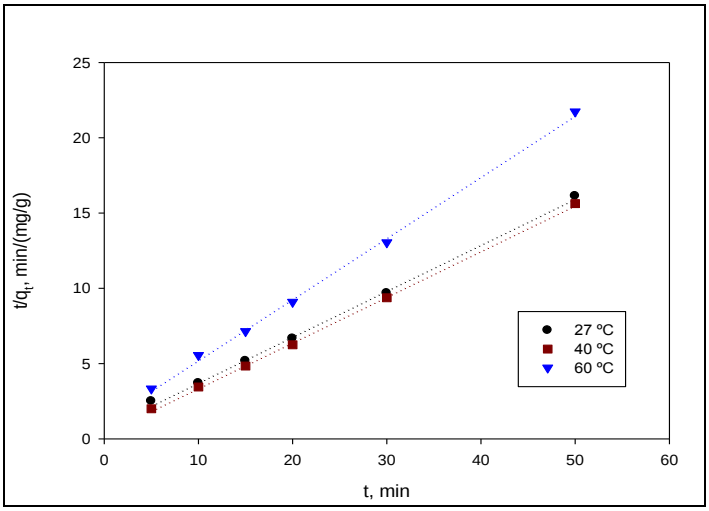

Figure 3: Pseudo-second-order adsorption kinetics of phenol on SBFA.
Source: Authors, (2020).

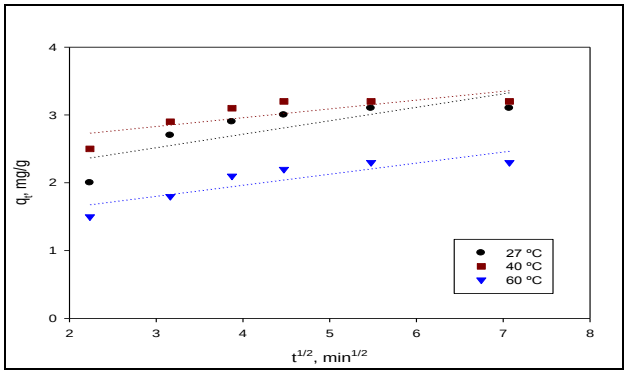

Figure 4: Intraparticle diffusion adsorption kinetics of phenol on SBFA.
Source: Authors, (2020).

Table 4 reports the values of the model parameters calculated from the linear fitting. Results show that, in pseudo-first-order and pseudo-second-order models, higher values of R^2 were found and the calculated values of q_e were comparable to those of experimental values. Also, data exposed that, in general, the pseudo-second-order model was the more adequate to reproduce the experimental data. It indicates chemical adsorption onto phenol removal by SBFA [30]. Intraparticle diffusion model did not fit well to the process of adsorption of phenol due to the low values of R^2 obtained, especially for temperatures of 27 and 40 ºC. Intraparticle diffusion model fitted the experimental results in a lower degree.

Table 4: Kinetic parameters for the adsorption of phenol on SBFA.

Models	**Parameters**	**Temperature**		
		27 °C	**40 °C**	**60 °C**
Pseudo-first-order	$k_1(min^{-1})$	0.21	0.30	0.19
	$q_{e\ Calc.}(mg/g)$	3.08	3.17	2.26
	R^2	0.998	0.988	0.968
Pseudo-second-order	$k_2(g/mg{\cdot}min)$	0.13	0.26	0.14
	$q_{e\ Calc}(mg/g)$	3.27	3.30	2.46
	R^2	0.999	0.999	0.998
Intraparticle diffusion	$k_{id}(mg/g{\cdot}min^{1/2})$	0.20	0.13	0.16
	$C(mg/g)$	1.93	2.45	1.32
	R^2	0.665	0.641	0.771

Source: Authors, (2020).

According to some previous works, temperature has an evident influence on adsorption of both gas and liquid substances The maximum sorption capacity q_e calculated from pseudo-first and pseudo-second order models decreased as the temperature increased, mainly when temperature changed from 27-40 to 60 ºC. Some authors [31-33], reported that, when the temperature increased, the attraction between the adsorbate and the active groups of the solid surface weaken and there was a greater inclination of the adsorbate to discharge from the solid surface towards the liquid phase, which causes a drop in adsorbate adsorption.

III.2 COLUMN ADSORPTION STUDY

Studies of adsorption in fixed-bed or packed-bed columns are indispensable for the scale-up of the adsorption processes [34-35]. In this sense, to determine the kinetic parameters that characterizes dynamic adsorption is important carry out research that lets determinate the corresponding breakthrough curves.

In the Figures 5 and 6, the breakthrough curves show saturation times from 110 and 150 min for 47.5 and 65.0 g respectively.

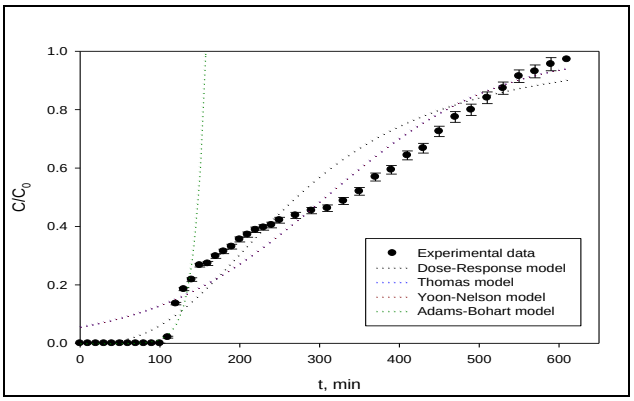

Figure 5: Non-linear plot of Thomas, Yoon-Nelson, Adams-Bohart and Dose-response models for the adsorption of phenol using 47.5 g of SBFA.
Source: Authors, (2020).

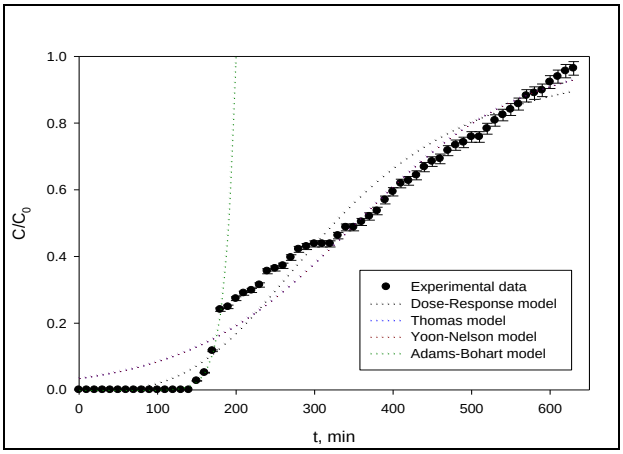

Figure 6: Non-linear plot of Thomas, Yoon-Nelson, Adams-Bohart and Dose-response models for the adsorption of phenol using 65.0 g of SBFA.
Source: Authors, (2020).

Table 5 shows the parameters calculated from the kinetic models, applied to the breakthrough curves, for both experiments performed with different weight of adsorbent.

Table 5: Kinetics parameters in dynamic conditions for the adsorption of phenol on SBFA.

Models	**Parameters**				
Dose-response	***m**(ash)(g)*	***A***	***q_0** (mg/g)*	***R^2***	
	47.5	2.72	0.34	0.867	
	65.0	3.26	0.30	0.908	
Thomas		***k_{TH}** (mL/min·mg)*	***q_0** (mg/g)*	***R^2***	
	47.5	0.00184	0.39	0.896	
	65.0	0.00187	0.32	0.917	
Yoon-Nelson		***k_{YN}** (min^{-1})*	***τ** (min) calc.*	***R^2***	***τ** (min) exp.*
	47.5	0.0092	306.3	0.896	330
	65.0	0.0094	351.5	0.917	360
Adams-Bohart		***k_{AB}** (L/mg·min)*	***N_0**(mg/L)*	***R^2***	
	47.5	0.0146	90.62	0.765	
	65.0	0.0148	88.55	0.998	

Source: Authors, (2020).

According to the results obtained for Dose-response and Thomas models, as the bed height increased, the amount of phenol removed by gram of SBFA, q_0, decreased. In addition, if the values obtained of q_0 for Dose-response and Thomas models are compared, very similar values are found.

The Thomas and Yoon-Nelson fitted better the full experimental breakthrough curve. Adams–Bohart model also represents the sorption of phenol accurately for the initial part of the breakthrough curve and fitted better for the highest mass of adsorbent studied. In all the cases, R_2 and the rate constant values increased with an increase in the sorbent mass, due to increase of the height of the mass transfer zone. Regards the parameter τ of Yoon-Nelson model (the half-life of adsorbate breakthrough) it did not show important differences between the calculated values and the experimental ones.

IV. CONCLUSIONS

From the results obtained it can be concluded that the sorption process occurs quickly, reaching the maximum sorption capacity before 20 minutes of operation, being slightly more favorable at temperatures between 27 and 40 ºC.

The kinetics of adsorption of phenol using SBFA as adsorbent in batch systems was well described by pseudo-first order and pseudo-second order models with R^2 values higher than 0.99 in both models, which suggests that adsorption process was well defined by chemisorption.

The kinetic models used to predict the adsorption of phenols in fixed-bed columns, Thomas, Yoon-Nelson, Adams-Bohart and Dose-response models, fitted well to the experimental breakthrough curves. Although, Adams-Bohart model was only able to predict the initial part of the curve.

V. ACKNOWLEDGMENTS

The authors are thankful to the Chemical – Pharmacy Faculty of the Central University “Marta Abreus” of Las Villas, UCLV and to the “Centro de Estudios Ambientales de Cienfuegos”, for providing all necessary technologies and infrastructure for carrying out this work.

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