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Submission date: 31-Jul-2023 04:27PM (UTC+0700)

Submission ID: 2139438375

File name: Artikel_SICETO.pdf (609.34K)

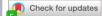
Word count: 4886

Character count: 24841

RESEARCH ARTICLE | JULY 21 2023

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AIP Conference Proceedings 2689, 060013 (2023) https://doi.org/10.1063/5.0115018





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Removal and Isotherm Adsorption of COD and Turbidity in Wastewater from Oil and Gas Industry onto Water Hyacinth (*Eichhornia Crassipes*) Activated Carbon

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Abstract. This research was conducted to determine the removal percentage and isotherm adsorption model of the oil and gas industry wastewater treatment using activated Carbon made from water hyacinth (*Eichhornia crassipes*). The method of making the adsorbent was carried out by impregnation of dry water hyacinth using H3PO4 followed by 7 rbonisation in a furnace at a temperature of 600°C for 3 hours. The processed wastewater feeds the oil and gas industry's primary effluent treatment-secondary effluent treatment (PET-SET). Variations in this study were adsorbent dose (1, 3, and 5 g) and contact time (0, 15, 1, 45, 60, 90, 120, 180, 240 minutes). Parameters reviewed in the study were COD levels and turbidity in wastewater. The research was carried out in a batch system on a laboratory scale. COD was analysed with MERCK Spectroquant Prove 600 No. 335 and turbidity with Turbidimeter of Hanna H1 98703. The results showed that the highest percent of COD removal reached 54.65% when the contact time was 240 minutes with an adsorbent dose of 5 g. The highest turbidity removal reached 73.82% at a contact time of 240 minutes with 1 g of adsorbent. Almost all variations resulted in regression over 0.9 both Langmuir and Freundlich isother 2, whether for COD and turbidity. Using error analysis calculations, the adsorption process of wastewater from the oil and gas industry onto water hyacinth activated Carbon followed the Langmuir isotherm adsorption model with a maximum adsorption capacity of 8.46 mg/g for COD and 6.65 mg/g for tt 2 idity. It concluded that the activated Carbon made from water hyacinth was an effective adsorbent for application in wastewater treatment from the oil and gas industry.

INTRODUCTION

The wastewater produced from oil and gas industry petroleum must be treated first before re-entering water bodies. Pollutants included oil and fat, dissolved sulfide (H2S), ammonia (NH3-N), phenol, hydrocarbon compounds, and others. These pollutants disturb the biota that lives in water bodies around the industry. [1] stated that the content of pollutants could be alighatic and aromatic compounds with low to high molecular components. It encourages researchers to research wastewater from the oil and gas industry, which aims to reduce the pollutant content in the was water.

Treatment of wastewater from the oil and gas industry by adsorption is one effective option, and it can be done on a small or large scale. The polluted wastewater is contacted with adsorbents, generally inactivated Carbon, which filters and absorbers various pollutants such as pathogenic compounds, dissolved solids, synthetic dyes, heavy metals, etc. In general, the adsorption process onto adsorbents, including Activated Carbon, occurs through several basic stages, starting from the process of absorbing pollutant particles on the surface of the adsorbent, then entering the pores, and finally being adsorbed on the inner wall of the adsorbent so that the pollutants number in the wastewater reduced.

Water hyacinth (*Eichhornia crassipes*) was an aquatic weed plant group. Water hyacinth grows in shallow ponds, wetlands and swamps, slow-flowing water, lakes, reservoirs, and rivers. These plants can tolerate extreme changes in water level, water flow, and changes in nutrient availability, pH, temperature, and toxins in the water. Water hyacinth has fast vegetative reproduction, especially in the tropics and subtropics areas. This rapid growth of water hyacinth interferes with water transportation, reduces dissolved oxygen in the water, disrupts the life of aquatic biota, causes siltation, and reduces the aesthetics of the aquatic environment.

This water hyacinth thrives in the waters around the oil and gas industry. It disrupts jetty operations, damages aesthetics, and makes the oil and gas industry pay an extra for cleaning this water hyacinth. For one cleaning only, there are 5 (five) trucks of water hyacinth around the jetty. It encourages researchers to convert the water hyacinth into a more valuable product, such as biosorbent, activated Carbon, bio briquette, biogas and bioethanol.

Water hyacinth consists of 60% cellulose, 8% hemicellulose, and 17% lignin. This content supports the performance of water hyacinth as a raw material to produce adsorbent [2–9], a mixture to make biogas [10], bio briquettes [11] and bioethanol [12]. Direct use of water hyacinth (phytoremediation), namely planting water hyacinth in containers that containing an oil-water mixture, has been reduced the oil content in the mixture [13], Pb and Total Dissolved Solids (TDS) in groundwater [14], COD, BOD, TSS, pH, colour, and odour in tofu waste [15–17].

Water hyacinth is also used as an adsorbent of heavy metals, including Pb2+ [2], Ni (II) and Cu (II) [3], Mercury (II) [4], Cr and Cu [5], Cd [6], Pb [8] Cr(VI) [18], and electroplating waste [9]. Water hyacinth adsorbent also degraded the content of synthetic crude oil [13,19], diesel, Castrol, lubricant [20], and used cooking oil [21]. Another use of water hyacinth as an adsorbent was to absorb ammonia [22]. Reduced levels of BOD, COD, TSS, turbidity, and pH in well [23], in tofu waste [24,25], in tempe waste [26], in textile waste [27], in laundry waste [28], in laboratory waste [29], and oil and gas [30] and reduce Phenol levels in PLTU waste [31].

Research on the use of water hyacinth as an adsorbent has been carried out. The degradation in COD levels was 95.89%, Fe 95.8%, Pb 95.36%, Cd 99.44% in textile wastewater [27]. The reduction in COD levels was 47.74% in laundry waste [28]. The degression in COD levels was 64%, turbidity 92%, TSS 83%, and BOD 74% in laboratory waste [29]. COD and BOD levels were lowered by 40.91% and 73.33%, respectively, in tofu wastewater [25]. Research conducted by [30] reduced COD levels up to 7.93% in oil and gas wastewater.

This study aims 3 determine the effect of variations of the adsorbent dose on the percentage of removal and the adsorption isotherm in the oil and gas industry wastewater. The pollutant parameters reviewed in the study were COD and turbidity.

RESEARCH METHODOLOGY

Materials

The tools were glassware, analytical balance (Sartorius BSA 2244S), shaker (SWB30-Shaking Bath Water Type 50 CAT NUMBER 154555), furnace (Carblite Gero AAF 1100), oven (Memmert E07086), desiccator, sieve (20 mesh), bottle santes, plastic containers, stopwatches, masks, and gloves. Water hyacinth, wastewater from the oil and gas industry, aquadest, H₃PO₄ 1 M (technical grade), filter paper, and aluminium foil.

Procedures

Raw Material Preparation

Water hyacinth (E. crassipes) were taken from waters around PT. Kilang Pertamina International Refinery Unit (RU) III Plaju, Palembang, Indonesia. Then separated the roots from the stems and leaves. The parts used in this study were its stems and leaves. The washing of water hyacinth continued the preparation by using freshwater from a regional drinking water company. The washing repetition was three times and followed by three times washing with aquadest. The clean water hyacinth was reduced to 5 cm and dried using an oven at 110°C for 48 hours. This dried water hyacinth was reduced in size to 20 mesh.

Production of Activated Carbon

Dried water hyacinth was impregnated in 1 M $\rm H_3PO_4$ solution with a mass ratio of 1:3 for two days at room temperature. Next, it was carbonised in a furnace where the pyrolysis occurred with little or no oxygen at a temperature of 600°C for 3 hours. The resulting activated Carbon was cooled to room temperature, washed with distilled water to a neutral pH (6-7), and dried in an oven at 110°C for 3 hours. The resulting adsorbent is referred to as water hyacinth activated Carbon.

Application of the Adsorbent in Oil and Gas Industry Wastewater

The wastewater used was the primary effluent treatment-secondary effluent treatment (PET-SET) feed on PT. Kilang Pertamina International Refinery Unit (RU) III Plaju, Palembang, Indonesia. This wastewater was residual process water for the refinery. The adsorbent is applied to the wastewater in the batch system. The adsorbent dose varied, namely 1, 3, 5 g per 200 ml volume of wastewater. The contact time was 15, 30, 45, 60, 90, 120, 180, 240 minutes. The adsorbent was put into the wastewater and then stirred using a shaker at 120 rpm until the specified contact time. This application aimed to reduce COD and turbidity levels in oil and gas industry wastewater.

Data Analysis

Percentage removal for both COD and turbidity calculated based on equation (1), where C_o was the initial COD concentration or turbidity value, C_e was the COD concentration or turbidity value at time t.

$$\%removal = \frac{C_o - C_e}{C_o} \times 100\%$$
 (1)

The COD concentration or turbidity data calculate the adsorption capacity as presented in equation (2). Adsorption capacity (q_e) stated the amount of adsorbate adsorbed per adsorbent weight (mg/g), V stated the volume of wastewater (litre), and m stated the mass of adsorbent used (gram).

The data were also used to calculate the appropriate Langmuir and Freundlich adsorption isotherm model. The Langmuir isotherm equation is presented in equation (3), and the Freundlich isotherm is presented in equation (4) [32–35]. Where K represented the Langmuir adsorption equilibrium constant (L/mg), q_m represented the maximum (theoretical) adsorption capacity of the adsorbent (mg/g). K_f represented the Freundlich adsorption capacity of a system, whether the adsorption process favourable or not, namely at range 1-20 [(mg/g) (L/mg)n] and n indicated the fitness of the model with the adsorption process (n > 1) [32]. In determining the suitability of the isotherm model with the experiment, the analysis error equation was calculated, namely the Chi-square test (X2) and the Root Mean Square Error R [33] according to equations (5) and (6). Where $q_{e\,exp}$ represented the value of q_e under experimental conditions, $q_e\,_{cal}$ was the predicted q_e from the model, and n represented the amount of data in the experiment.

$$q_e = (C_o - C_e) x \frac{V}{m}$$
 (2)

$$\frac{C_e}{q_e} = \frac{1}{q_m K} + \frac{C_e}{q_m} \tag{3}$$

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \tag{4}$$

$$Chi - square\ test\ (X^2) = \sum_{i=1}^{n} \left(\frac{q_{e\ exp} - q_{e\ cal}}{q_{e\ exp}}\right)^2 \tag{5}$$

Root Mean Square Error (RMSE) =
$$\sqrt{\frac{\sum(q_{e\,exp} - q_{e\,cal})^2}{n}}$$
 (6)

RESULT AND DISCUSSION

Til treated wastewater was a feed for the primary effluent treatment-secondary effluent treatment (PET-SET) from PT. Kilang Pertamina International Refinery Unit (RU) III Plaju, Palembang, Indonesia. The wastewater source was from the rest of the process water due to petroleum processing in the refinery. However, now the wastewater in the PET-SET unit original treatment of the primary effluent treatment-secondary effluent treatment (PET-SET) was treatment (PET-SET) and treatment (PET-SET) and treatment (PET-SET) are treatment (PET-SET).

The initial analysis of wastewater samples from the oil and gas industry resulted from COD levels of 516 ppm and 7 bidity levels of 173 NTU. The maximum permitted level based on the Regulation of the Governor of South Sumatra No. 8 of 2012 concerning the quality standards for the disposal of process wastewater from petroleum processing activities for COD levels of 160 ppm, BOD 80 ppm, oil and fat 20 ppm, pH 6-9, sulfide levels 0.5 ppm, ammonia levels eight ppm, and phenol content of 0.8 ppm. Based on it, the level of COD in wastewater exceeded the predetermined waste quality standard threshold. The quality standard limits were not listed; the lower the turbidity value was, the better.

Effect of Adsorbent Dosage and Time on COD Removal

Figure 1 shows the decreasing COD levels in wastewater after the adsorption process using water hyacinth activated Carbon. In this study, the COD concentration decreased from the initial waste concentration of 516 ppm to 234 ppm. The decrease occurred when the adsorbent dose used was 5 g with a time of 240 minutes. Based on the graph, the COD value continued to decrease throughout the contact time, indicating the adsorption process. The higher the adsorbent dose and the longer the contact time, the lower the COD value obtained. The standard quality value of COD had not been achieved by looking at this downward trend. It could be stated that additional contact time and adsorbent dose would be an effective adsorbed of water hyacinth activated Carbon to remove COD in wastewater.

Figure 2 described the percentage of COD removal, which showed an increase in COD removal each contact time. In this experiment, the highest removal of COD was 54.65% at a dose of 5 g adsorbent and a time of 240 minutes.

Based on Figures 1 and 2, the higher the adsorbent dose and contact time, the absorption of COD levels was not too high but tended to increase. At the contact time of 120 minutes, there was a decrease in the COD removal percentage. It happened because, at the first of 60 minutes, the adsorption occurred very fast. At 90 minutes, the adsorbent started to saturate so that the adsorption took place slowly. That indicated after 90 minutes, adsorption went on and tended to increase while the step-up was not significant.

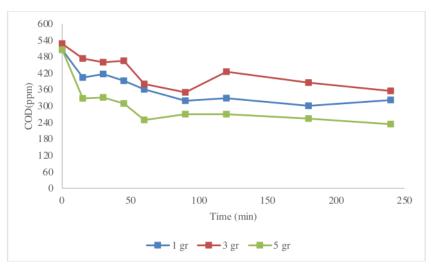


FIGURE 1. The degradation of COD levels in wastewater

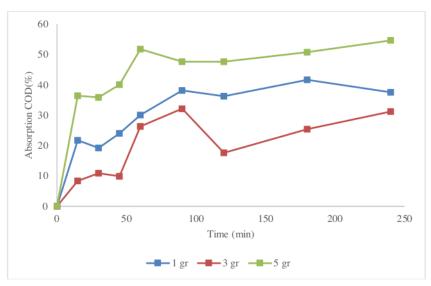


FIGURE 2. Percentage of COD removal in wastewater

In this study, the performance of water hyacinth is activated. Carbon on COD removal resulted from better performance than water hyacinth adsorbent in the treatment of laundry waste [28] and tofu liquid waste [25]. Compared to research results of textile industry wastewater [27] and laboratory wastewater [29], the water hyacinth activated Carbon in this study showed a lower performance. Compared to the research of [29] that used oil and gasliquid waste as feed, the decrease in the percentage of COD obtained was 7.9% within 30 hours by using an activated carbon sourced from cassava peel. It proves that activated Carbon from water hyacinth showed a better performance to treat wastewater from the oil and gas industry than cassava peel. This result is also supported by the research of [19], which stated that activated Carbon of water hyacinth had a better effect on COD removal in well water than adsorbents from coconut shells.

Effect of Adsorbent Dosage and Time on Turbidity Removal

Figure 3 showed a decress in turbidity levels in wastewater after the adsorption process was carried out. Based on the initial parameter test of wastewater of the oil and gas industry, the value of the turbidity of the waste was 173 NTU. The highest turbidity reduction was 45.3 NTU that occurred at the adsorbent dose used was 1 gram, and the contact time was 240 minutes.

Figure 4 showed that the highest turbidity removal efficiency was 73.82% that occurred at a contact time of 240 minutes with a dose of 1 gram of adsorbent used. Based on the results of research of [36] that reviewed the turbidity of industrial seaweed waste using nano adsorbents, the percentage of turbidity was 41%. The value of the decrease in turbidity obtained from this study was greater than that nano adsorbent. By utilising water, hyacinth activated Carbon, the turbidity removal in this research showed lower performance than the laboratory wastewater treatment [29].

Overall, this turbidity result represented a similar trend as the decrease in COD value. Where adsorption took place quickly in the first 60 minutes, the next process occurred slowly. It was because the pores of the adsorbent had been filled with wastewater pollutants, thereby reducing its ability to absorb pollutants. In other words, there was a decrease in the adsorption rate.

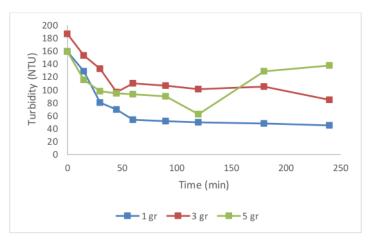


FIGURE 3. The degradation of turbidity level in wastewater

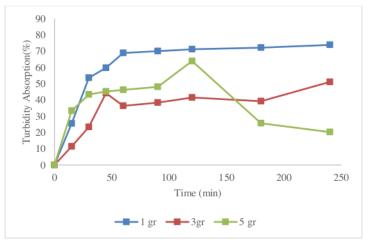


FIGURE 4. Percentage of turbidity removal in wastewater

Effect of Adsorbent Dosage on Adsorption Isotherms

Based on [35], the suitable isothern 9 dsorption model in this study could be proven through the correlat 9 n coefficient (R²). The regression value of the linear graph of each equation should be close to one that concluded the type of adsorption isotherm followed the isotherm equation.

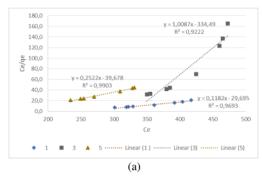
TABLE 1. Langmuir isotherm adsorption

A 314		COD			Turbidity	
Adsorbent dose (g)	\mathbb{R}^2	q _m (mg/g)	K (l/mg)	\mathbb{R}^2	q _m (mg/g)	K (l/mg)
1	0.9693	8.46	0.0040	0.9440	6.65	0.0256
3	0.9222	0.99	0.0030	0.8621	0.69	0.0116
5	0.9903	3.97	0.0065	0.8909	0.90	0.0160

TABLE 2. Freundlich isotherm adsorption

Adsorbent		COD			Turbidity	
dose (g)	\mathbb{R}^2	n	$\mathbf{K_f}$	\mathbb{R}^2	n	$\mathbf{K_f}$
1	0.9486	0.42	0.88	0.9706	1.03	0.48
3	0.9869	0.23	1.09	0.9174	0.40	0.75
5	0.9912	0.81	0.60	0.9049	0.70	0.52

Based on Table 1 and 2, the regression value obtained was almost above 0.9, indicating the data fit with the isotherm model approach. Figure 5 showed Langmuir isotherm adsorption of COD and turbidity using water hyacinth activated Carbon with doses of 1, 3, and 5 g plotted between C_e versus C_e/q_e . The linear regression ob and for COD was good, which R^2 above 0.9, while for turbidity only at a dose of 1 g showed a good correlation. The maximum adsorption capacity value of the Langmuir model was 0.69-8.46 mg/g. The experimental adsorption capacity of water hyacinth adsorption to COD ranged from 2.87 to 39.4 mg/g. The value of the adsorption capacity of water hyacinth in absorbing wastewater from the oil and gas industry was smaller than the results of [18] and [7], which absorb Cr and Pb, respectively. For the Langmuir isotherm equilibrium constant value obtained was also relatively small ranging from 0.0040-0.0256 l/mg.



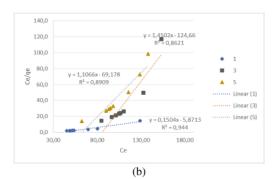
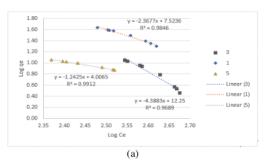


FIGURE 5. Langmuir isotherm adsorption: (a) COD (b) turbidity

For the Freundlich regression calculation, the linearisation line was obtained from the log Ce versus q_e . Figure 6 showed the Freundlich isotherm graph for COD, and turbidity using water hyacinth activated Carbon at doses 1, 3, and 5 g. All regression values showed a very good correlation with values above 0.9. Kf value ranged 0.48-1.09 (mg/g) (L/mg)ⁿ, which meant that almost all were not in the favourable adsorption range. Only adsorption data of COD with a dose of 3 g met the favourable adsorption for the value of n ranged 0.23-1.03. Only the Freundlich isotherm for the turbidity parameter at a dose of 1 gram had a good fit with the model. The heterogeneity factor value or n, which is less than 10, indicated that the reaction was reversible [37].



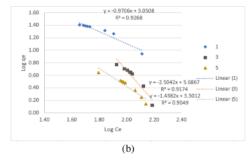


FIGURE 6. Freundlich isotherm adsorption: (a) COD (b) turbidity

TABLE 3. Compation of Error analysis from isotherm adsorption model by using Chi-square test (X2) dan Root mean square

error (RMSE) approaches								
Adsorbent dose (g)	Langmuir				Freundlich			
	COD		Turbidity		COD		Turbidity	
	X^2	RMSE	X^2	RMSE	X^2	RMSE	X^2	RMSE
1	4.15	25.02	3.60	15.66	7.54	32.27	7.60	21.56
3	5.40	6.75	5.17	3.68	5.18	6.66	4.95	3.62
5	2.63	5.62	3.40	2.11	6.99	8.92	5.07	2.47

The comparison of Figures 5 and 6 showed that the Langmuir and Freundlich model approach could describe the adsorption process that occurred. This approach was suitable because the value of the regression was close to 1. Error analysis was used, such as equations (5) and (6), to ensure the results of the calculation of the model approach. The results of the error analysis calculation from the adsorption process that developed were presented in Table 3. The error analysis calculation using the chi-square test (X²) showed a better result than the root mean square error (RMSE). Based on Table 3, overall errors obtained from the Langmuir 3 dsorption isotherm model were smaller than Freundlich's, so it can be concluded that the adsorption process for wastewater from the oil and gas industry followed the Langmuir isotherm model. The adsorption process of wastewater from the oil and gas industry showed a homogeneous surface adsorption process assuming that all active adsorbent sites had the same affinity for the adsorbate and formed a monolayer.

CONCLUSION

The results showed that the adsorption method using activated Carbon from water hyacinth was effectively used in the wastewater treatment process from the oil and gas industry. The highest COD removal achieved 54.65% at the contact time of 240 minutes and an adsorbent dose of 5 grams, 3 d turbidity removal achieved 73.82% at the contact time of 240 minutes and 1 gram of adsorbent. This wastewater adsorption process followed the Langmuir adsorption model with a maximum adsorption capacity of 8.46 mg/g for COD and 6.65 mg/g for t2 bidity. It concluded that the activated Carbon made from water hyacinth was an effective adsorbent for application in wastewater treatment from the oil and gas industry.

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