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Adsorption Of Metal Lead (Pb) In Batik Industrial Wastewater Using Cellulose-Based Adsorbent: Literature Review

ABSTRACT

This literature review aims to determine characteristics of the batik industrial wastewater, the type of adsorbent activation method most widely used to adsorb lead (Pb) in the batik industry wastewater, and the correlation between source of cellulose and parameters on the adsorption ability of lead metal (Pb) with variations in adsorbent mass, pH and contact time in batik industrial wastewater. This literature review was carried out with 7 steps of literature review consisting of exploring topics, searching, storing and organizing information, selecting the required information, expanding the search, analyzing and evaluating. information and present the results. The results of this literature review show that 1) Batik industrial wastewater contains BOD, COD, TSS and heavy metals. From the literature review, it was obtained that the BOD and COD values came from the batik industrial wastewater of Jetis Sidoharjo with a value of 1775.5 mg/L and 16654.80 mg/L, the largest TSS value came from the batik industrial wastewater Gedhog with a value of 449 mg/L. The largest metal content of lead came from the batik industrial wastewater of Wiradesa with a value of 7.654 mg/L. 2) The most widely used activation method for the treatment of adsorbents is the chemical activation method with strong acids such as HCl, HNO_3 and H_2SO_4. 3) There is a correlation between the source of cellulose and parameters in adsorption of lead metal in batik industry wastewater. Different sources of cellulose and parameters resulted in different adsorption capacities. Based on the literature review, the highest percentage of cellulose was found in sawn teak (60%), corn cobs (41%), rice straw (37.71%), rice husks (34%), and kapok seeds (21.83%). Maximum adsorption lies in the adsorbent with a mass of 0.1 – 1g, pH 5-7, and contact time of 30-45 minutes.

Key word: Adsorbent, Cellulose, Batik Industry Wastewater, Lead (Pb)

INTRODUCTION

The increase of batik industry in Indonesia has led to an increase in the liquid waste of the batik industry. The most batik-producing areas are still attached to the island of Java, which is spread across Central Java, West Java, East Java, Yogyakarta, Banten and Jakarta, while outside Java, the largest batik industry is in the province of Jambi [1] Waste is the result or residual waste generated from a production process, both industrial and domestic (household) which is better known as waste [2].

The increase in the batik industry will cause the wastewater load to be even greater. The wastewater generated in Indonesia in 2017 was 883 tons/day, of which 29% of the load came from the textile industry[3].

The textile industry usually produces liquid waste that is thick in color and contains high and fluctuating BOD (Biochemical Oxygen Demand), COD (Chemical Oxygen Demand), pH, temperature, turbidity, salinity, and toxic chemicals. Sources of heavy metals chromium (Cr) and lead (Pb) which is toxic, can come from dyes ($CrCl_3$, $K_2Cr_2O_7$) or from mordant which is a binder for dyes including $Cr(NO_3)_2$ and $PbCrO_4$[4], other sources of heavy metals such as Cu come from premet dyes and acids [5]

The increasing amount of batik industrial waste causes environmental pollution. One of the heavy metals contained in the batik industry wastewater is lead metal. Lead is one of the heavy metals that has high toxicity and is non-biodegradable so that its presence persists in nature, lead or what is known as lead is symbolized by Pb having atomic number 82, the presence of lead in the human body can cause neurological disorders [6]

If the batik industry liquid waste generated from each batik production process is directly discharged into the waters without being processed first, it will cause changes in the physical and chemical characteristics of the water and can threaten aquatic ecosystems. For this reason, waste needs to be processed first to comply with wastewater quality standards with parameters BOD, COD, TSS and heavy metals. The maximum limit is BOD (60 mg/L), COD (150 mg/L), TSS (50 mg/L) and heavy metals (Pb; 0.1 mg/L) [7]. One of the waste treatment that can be applied is by adsorption.

Adsorption was chosen because the required cost is relatively cheap, the process is relatively simple, efficient and has high effectiveness [8]. One of the compounds that acts as an absorbent or adsorbent is cellulose, this can be found from a variety of natural materials. Cellulose is a compound with functional groups that can bind with metal ions, the functional groups are carboxyl and hydroxyl groups [9]

This literature review aims to determine the characteristics of the batik industry wastewater, the most frequently used activation method and to determine the correlation between the source of cellulose and different parameters on lead adsorption.

METHODS

This literature review method consists of seven steps, namely exploring topics, conducting searches, storing and organizing information, selecting the information needed, expanding the search, analyzing and providing information and presenting the results of a literature review [10]

RESULTS AND DISCUSSION

Based on the results of the literature review, it was obtained the results of a review of the characteristics of the batik industry wastewater, the most frequently used activation method as well as the source of cellulose and its parameter.

1. Characteristics of indurstrial batik wastewater

COD, BOD, TSS and heavy metals can be found in the wastewater of the batik industry. BOD (Biochemical Oxygen Demand) is a characteristic that indicates the amount of dissolved oxygen required by microorganisms (usually bacteria) to decompose or decompose organic matter. BOD can be interpreted as a description of organic matter that is easily decomposed in waters. While COD (Chemical Oxygen Demand) is the amount of oxygen needed to break down all organic matter contained in water. TSS (Total Suspended Solid) is a suspended solid that causes water turbidity, does not dissolve, and cannot settle [11].Heavy metals contained in the batik industry wastewater include lead metal and chromium metal. The liquid waste content of the batik industry is different from one another. This is caused by the use of chemicals, especially in the process of dyeing or coloring, pelorodan and washing in the batik production process. Chemicals used in the batik-making process include: dyes as the main chemical and auxiliary chemicals, namely caustic soda (NaOH), soda ash (Na_2CO_3), baking soda ($NaHCO_3$), sulfuric acid (H_2SO_4), sulfites, and nitrites[4].

Besides being caused by the chemicals used, the content of the batik industry wastewater also depends on the amount of batik production and the presence of a wastewater treatment plant (WWTP). WWTP in the batik industry wastewater treatment process serves as a place to treat wastewater before being discharged into the waters. Based on the regulation of the Minister of the Environment, the quality standard of batik wastewater for the content of BOD, COD, TSS, lead and chromium is 60, 150, 50, 0,1 and 1 mg/L[7].

The liquid waste content of the batik industry in the Banaran Sukoharjo area is 615 mg/L COD and the color intensity is 7000 PCU (Platina Cobalt Unit). This batik industry wastewater sample was taken from the batik dyeing process using dyes[3]. In the batik industry wastewater there are 350 mg/L BOD, COD of 424 mg/L and TSS of 375 mg/L. The wastewater sample used was taken from the UKM Batik Tulis Amali CH Sidoarjo with the condition that it had just come out of the rest of the batik coloring process[12]. One of the batik industries in Laweyan has COD and BOD values of 660.28 mg/L, and 1600 mg/L. This batik industry wastewater sample was taken at the WWTP location in the Laweyan batik village, which is located on Jalan Radjiman Laweyan, Surakarta City (behind the Laweyan village office) in an equalizing tank[13]. The liquid waste of the Yogyakarta Krebet batik industry has a COD value of 216.8 - 3845.5 mL. This sample of wastewater from the Yogyakarta Krebet batik industry was obtained from each batik-making process[14] The batik industry can produce liquid waste of 305 – 533 L/month and 7,712 m^3/month solid waste. The following is a table of characteristics of the batik industry wastewater obtained based on a literature review:

Table 1. Characteristics Batik Industrial Wastewater

Sources	BOD (mg/L)	COD (mg/L)	TSS (mg/L)	Pb (mg/L)	Cr (mg/L)	Zat Warna	*Ref*
Laweyan Batik Industry	660,28	1600	-	-		-	[13]
Plenthong Yogyakarta Batik Industry	101,665	-	-	1,122		-	[15]
Banyuurip Ageng Batik Industry	-	-	-	3,015		-	[16]
Sokaraja Batik Industry	-	-	-	0,014		-	[17]
Krebet Batik Industry	-	216,8-3845,5	-	1-4	3,89	-	[14]
Jetis Sidoharjo Batik Industry	1775,5	16.654,8	208	-	<0,02	-	[18]
Jenes Batik Industry	-	-	-	0,215	1,517	-	[4]
Banaran Sukoharjo Batik Industry	-	615	-	-	-	7000 PCU	[3]
Batik Industry	-	-	-	0,252	-	-	[19]
Wiradesa Batik Industry	-	-	-	7,629	-	-	[20]
Tulis Batik Industry	-	9014,6	-	2,366	-	-	[21]
Gedhog Tuban Batik Industry	-	4951,75	448	-	7	-	[22]
Amali CH Batik Industry	350	424	375	-	-	-	[12]

In the wastewater of the Jetis batik industry there are BOD, COD and TSS in the amount of 1775.5 mg/L, 16,654.80 mg/L, 208 mg/L. This batik industry wastewater sample was obtained from the batik coloring process. At the location of the Kampung Jetis batik industry, there is no WWTP due to the unavailability of sufficient land for the construction of the WWTP at Kampung Jetis Sidoarjo. Based on the research conducted, batik industrial wastewater contained COD of 9014.6 mg/L.

Based on Table 1, batik industrial wastewater contained BOD, COD, TSS and heavy metals. The largest BOD value is in the batik industry wastewater from Jetis, Sidoharjo with the amount of 1775.5 mg/L. The largest COD value is in liquid batik waste originating from Jetis, Sidoharjo with a total of 16654.80 mg/L. Meanwhile, the largest TSS was found in the batik industry wastewater, where COD was 4951.75 mg/L and TSS was 448 mg/L. This liquid waste sample was obtained from one of the batik production sites in Tuban with the name batik gedhog. batik industry originating from the Tuban gedhog batik industry with the amount of 449 mg/L. The highest levels of lead are found in the liquid waste of the Wiradesa batik industry with a level of 7.654 mg/L. Meanwhile, the largest concentration of chromium metal is found in the wastewater of the batik industry which is located on the banks of the Jenes Laweyan river with an amount of 1.516 mg/L.

The difference in content in the batik industry wastewater is caused by the use of chemicals, especially in the processdyeing or coloring, pelorodan and washing in the batik production process. Chemicals used in the batik-making process include: dyestuffs as the main chemical and auxiliary chemicals, namely caustic soda (NaOH), soda ash (Na_2CO_3), baking soda ($NaHCO_3$), sulfuric acid (H_2SO_4), sulfites, and nitrites [4]. Apart from being caused by chemicals used, the content of the batik industry wastewater also depends on the amount of batik production and the presence of a wastewater treatment plant (WWTP). WWTP in the batik industry wastewater treatment process serves as a place for treat waste water before it is discharged into the environment.

2. Activation Method On Adsorbent

Activation is a treatment of adsorbent which aims to enlarge the pores by breaking the hydrocarbon bonds or oxidizing the surface molecules so that the charcoal undergoes changes in properties, both physical and chemical, namely the surface area increases and affects the adsorption power [23].

The interaction between the activating agent and the structure of the carbon atoms resulting from the carbonization is the mechanism of the activation process. Physical activation can be done using heat, steam and CO_2 gas, while chemical activation uses chemicals called activators [24]. Meanwhile, for physico-chemical activation, it is done by combining physical and chemical activation.

The activation of the adsorbent is divided into two types, namely hardwood and softwood. Hardwoods contain more cellulose and extractives, and less lignin, than softwoods; For pulp purposes, softwood fibers are, on average, more than three times the length of those contained in hardwoods (thus the origin of the terms "long-fiber pulp" and "short-fiber pulp"). And since increasing fiber length usually translates into more inter-fiber bonding, softwood pulps impart greater strength to the products into which they are made, than do hardwood pulps manufactured by the same process.

The example of hardwood adsorbents are rubber sawdust, pine sawdust, merbau wood sawdust, kapok seed dan cocoa husk. Meanwhile, softwood adsorbent such as rice husk, pomelo leaves, kepok's banana, jengkol skin and dami jackfruit.

The following is a table of activation methods for hardwood adsorbents consisting of rubber sawdust, pine sawdust, merbau wood sawdust, kapok seed charcoal and cocoa husks.

Table 2. Activated Methods On Hardwoods Adsorbent

Adsorbent	Activation Methods	Treatment	Result	Ref
Rubber Wood Sawdust	Physic-Chemistry	Heated in an oven at 110 ^{0} C for 24 hours, then soaked in 11 ml 98% H_2SO_4 and 6.6 ml 65% concentrated HNO_3.	Activated carbon with a size of 0.5 - 1 mm at a temperature of 110 ^{0}C, the average pore size of the adsorbent is 42.6 A and the adsorption capacity is 38.56 mg/g	[25]
Pine sawdust	Chemistry	Activated with 0.1 N NaOH for 24 hours	Pine sawdust adsorbent with efficiency 99,84%	[26]
Merbau Wood Sawdust	Chemistry	Activated with HCl 2N for 24 hours	Merbau wood sawdust adsorbent activated with capacity adsorption 1,925 mg/L.	[27]
Kapok seed charcoal	Chemistry	Soaked in H_3PO_4 85% for 24 hours	Kapok seed charcoal activated with capacity adsoprion 0.05 mg/g.	[28]
Cocoa husk	Physics	Cocoa husk carbonated with 600 ^{0}C for 15 minutes	Charcoal cocoa husk with percentage of adsorption 98,715%	[29]

Research using Rubber Wood Sawdust adsorbent using physico-chemical activation method by heating at 110 ^{0}C in a 24 hour oven, then soaking in 11 ml 98% H_2SO_4 and 6.6 ml 65% HNO_3 obtained carbon with an active size of 0.5 – 1mm with an average pore size of 42.6 A for adsorbent and an adsorption capacity of 38.56 mg/g. In the adsorbent derived from pine sawdust chemically activated using 0.1 N NaOH for 24 hours, sawdust was obtained with an adsorbent efficiency of 99.84%.

Activation using NaOH solution aims to cause delignification of the adsorbent. The function of delignification is to dissolve compounds such as lignin which can inhibit the adsorption process. This is because the presence of lignin will hinder the ion transfer process to the active side of the adsorbent. NaOH solution will break the bonds of cellulose with lignin. This is because the OH- ions from the NaOH solution will break the bonds from the basic structure of liglin. The following is a mechanism for breaking the bonds of cellulose derived from rice straw with lignin.

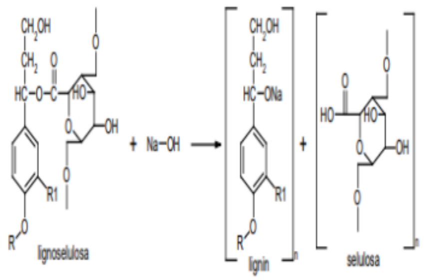

Figure 1. The mechanism of breaking the bond between lignin and cellulose by NaOH.

Lignin degradation begins with the attack of the H atom bound to the phenolic OH group by hydroxide ions (OH^-) from NaOH. The H atom in that section is acidic because it is bonded to an O atom which has a large electronegativity. The more electronegative O atom will attract electrons to the H atom, so the H atom will be positively charged and easy. released into H^+ ions. Acidity is also influenced by the resonance effect of the alkyl group at the para position, so that the H atom in the phenolic group will be more acidic [30]

Research using adsorbent derived from kapok seed charcoal activated using chemical methods through immersion with 85% H_3PO_4 for 24 hours obtained kapok seed activated charcoal adsorbent with adsorption capacity of 0.05 mg/g. The charcoal activation process by immersion in 150 ml of 85% H_3PO_4 solution for 24 hours has the aim of opening the pores of the charcoal surface by breaking hydrocarbon bonds or oxidizing surface molecules so that the activated charcoal of kapok seeds undergoes physical and chemical changes, namely the surface area increases and increase the absorption of activated charcoal. The following is an activation reaction of charcoal with H_3PO_4:

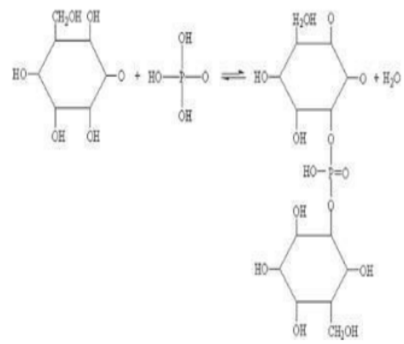

Figure 2. Activated reaction between charcoal and H_3PO_4

In the figure, it is known that H_3PO_4 reacts with the formed charcoal and then forms micropores on the surface which serve as a place for adsorption to take place. This makes the surface of the adsorption on the charcoal wider [31]

Activation methods on softwood adsorbents derived from rice husk, pomelo leaves, dami jackfruit, kepok banana peel and jengkol peel are divided into physical, chemical and physico-chemical activation methods. In a study using an adsorbent derived from jengkol skin, it was chemically activated through immersion in 5N nitric acid for 90 minutes and then sieved on a 100 mesh sieve to obtain an adsorbent of jengkol skin.

The following is a table of activation methods for softwood adsorbents:

Table 3. Activated Methods On Softwood Adsorbents

Adsorbent	Activated Methods	Treatment	Result	Ref
Rice Husk	Physic-Chemistry	Carbonized at 400 - 600 ^{0}C for 90-150 minutes, soakan on HCl, filter with100 mesh sieve	Activated carbon with 4.86% humidity, 30.04% Ash, and 15.76% volatile matter	[32]
Pomelo Laves	Physics	Dried on oven at 65 ^{0}C and filter	Pomelo Leaves adsorbent with size <355µm	[33]
Dami Jacfruit	Physisc	Carbonized at 250 ^{0}C, 2.5 hours	Jackfruit dami charcoal with adsorbent absorption efficiency of 96.67%.	[34]
Kepok's Banana Peel	Chemistry	Soaked for 90 minutes in 5M concentrated nitric acid, sieved through a 100 mesh sieve	Activated kepok banana peel adsorbent with a size of 100 mesh	[35]
Jengkol Skin	Chemistry	Soaked in 5N nitric acid for 90 minutes, sieved on sieve 100 mesh	Jengkol peel adsorbent is activated with size 100 mesh	[36]

In a study using an adsorbent derived from jengkol skin, it was chemically activated through immersion in 5N nitric acid for 90 minutes and then sieved on a 100 mesh sieve to obtain an activated jengkol skin adsorbent with a size of 100 mesh. Nitric acid is used as an activating agent because it has H^+ ions that can dissolve metal impurities by exchanging metal ions bound to the biosorbent. This process causes an increase in the number of active biosorbent sites that have H^+ ions. Soaking with nitric acid also serves to break lignin from the lignocellulosic complex. The process of breaking lignin is necessary because lignin can cover the hydroxyl groups of cellulose in biosorbents by forming covalent bonds with lignin chromophores.

Based on Table 2 and Table 3, the activation method that is most widely used to activate the adsorbent is the chemical activation method. The most widely used compounds in this activation method are acidic compounds in the form of strong acids such as nitric acid (HNO_3), hydrochloric acid (HCl), sulfuric acid (H_2SO_4) and phosphoric acid (H_2SO_4). This acid compound is widely used because it has H^+ ions which can dissolve ions that pollute the surface of the adsorbent, and is used to open the pores of the adsorbent surface by breaking hydrocarbon bonds or oxidizing surface molecules so that they undergo physical and chemical changes, namely the surface area increases. large and increase the adsorption capacity of the adsorbent.

3. Sources of Cellulose and Parameters

7 Cellulose is a polysaccharide that has a very high relative molecular mass, cellulose is composed of 2000 - 3000 glucose. The existence of cellulose is very abundant in nature because it can be found on every part of the plant such as roots, stems and twigs. Cellulose is the main constituent of plant cell walls which is very abundant. The following is a table sources of cellulose and parameters

Table 4. Sources of Cellulose and ParametersSumber Selulosa dan Parameternya

Sources	Modes	Efficiency of Adsorption (%)	% Cellulose	[Pb] (ppm)	Varians			Ref
					Mass adsorbent (g)	Contact Time	pH	
Corncob raw	*batch*	97,29	41	58,86	0,02 ; 0,04 ; - 0,06 ; 0,08 ; 0,12 **(0,08)**	-	-	[37]
Kapok seed	batch	97,76	21,83	118	0,3 ; 0,6 ; 0,9 ; 1,2 **(1,2)**	30 menit	-	[28]
Rice husk	batch	88,765	37,71	5 ; 10 ; 15 ; 20 ; 25 **(10)**	0,5	30 menit	4; 5; 6; 7; 9 **(5)**	[38]
Jati wood sawdust	batch	99,98	60	61,44	0,1; 0,2 ; 0,3 ; 0,4 ; 0,5 **(0,4)**	-	3 ; 4 ; 5 ;6 ; 7 ; 8 **(7)**	[39]
Agave baggase	batch	93,14	-	-	0,4	5-120 **(15)**	3-6.5 **(5.5)**	[40]
Mangifera Indica	batch	69	-	100	0,1	240	5	[41]
Kakao skin	batch	98,715	17,27	100	0; 0,5; 1; 1,5**(0,5)**	30; 40;50; 60; **(40)**		[29]

Based on research with corncob adsorbent raw material with 41% cellulose content, it has an adsorption efficiency of 97.29% to adsorb lead (Pb) with the most effective adsorbent mass of 80 mg. In this study using variations in the form of adsorbent weight, namely 20, 40, 60, and 80 mg. The weight of the adsorbent increased at 20, 60, 80 and 120 mg, causing an increase in the percentage of lead adsorbed. This shows that the weight of the adsorbent affects the adsorption process because as the weight of the adsorbent increases, the value of % Pb adsorbed to ions also increases and reaches equilibrium. However, the adsorbent weight of 40 mg decreased. The small lead absorption is due to the contact time between the adsorbate and adsorbent that exceeds the optimum time which can cause desorption. Then at 120 mg the absorption decreased. This is due to the saturated adsorbent.

Based on the description and Table 4, the 5 largest percentages of cellulose are found in teak saws with a percentage of cellulose 60%, corn cobs 41%, rice straw 37.71%, rice husks 34% and kapok seeds 21.83%. Based on the mass range of the adsorbent, the maximum adsorption lies in the adsorbent with a mass range of 0.1 - 1 g, Total adsorbent affect the adsorption process, where the increasing weight will cause the adsorbent to reach the saturation point if the surface has been filled with adsorbate and there will be a decrease in adsorption capacity after reaching the maximum value due to the desorption process.

Based on the pH range in Table 4, the pH range for maximum adsorption of batik liquid waste lies at pH 5-7. At high pH, precipitation of Pb ions occurs which reduces the solubility of Pb ions in solution, this causes a decrease in the number of Pb ions that can be absorbed by the cell surface. The high pH causes the presence of more OH^- ions so that metal ions begin to precipitate. This causes the absorption to be reduced [39]. Absorption is also low if the pH occurs below the optimum range, because the H^+ concentration is too high it will be able to block the interaction between metal ions and the adsorbent.

Based on the contact time in Table 4 to adsorb lead metal, the maximum results are in the range of 30-45 minutes. The adsorption of ions from a solute will increase if the contact time is longer, the time to reach an equilibrium state in the metal adsorption process by the adsorbent ranges from several minutes to several hours. If the adsorbent is contacted in a relatively short time span, it will result in not many functional groups of the adsorbent interacting with the metal in the solution so that there are not many functional groups that play a role in adsorption of lead metal.

CONCLUSION

Batik industrial wastewater contains BOD, COD, TSS and heavy metals. The largest BOD and COD values are in the batik industry wastewater from Jetis, Sidoharjo with the amount of 1775.5 mg/L and 16654.80 mg/L. The largest TSS is found in the liquid waste of the Gedhog Tuban batik industry with the amount of 449 mg/L. The highest levels of lead are found in the liquid waste of the Wiradesa batik industry with a concentration of 7 ppm. The largest concentration of chromium metal is found in the wastewater of the batik industry which is on the banks of the Jenes Laweyan river with an amount of 1.516 mg/L.

The most widely used activation method for the treatment of adsorbents is the chemical activation method using acidic compounds in the form of strong acids, namely HCl, HNO_3, and H_2SO_4. There is a relationship between the source of cellulose and its parameters in the adsorption process of lead metal in the batik industry wastewater, different sources of cellulose and parameters will result in different adsorption capacities in adsorption of lead metal.

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