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Characterization of Activated Carbon from Coal and Its Application as Adsorbent on Mine Acid Water Treatment

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Abstract

Anthracite and Sub-bituminous as activated carbon raw material had been utilized especially in mining field as adsorbent of dangerous heavy metal compound resulted in mining activity. Carbon from coal was activated physically and chemically in various temperature and particle sizes. Characterization was carried out in order to determine the adsorbent specification produced hence can be used and applied accordingly. Proximate and ultimate analysis concluded anthracite has fixed carbon 88.91% while sub-bituminous 49.05%. NaOH was used in chemical activation while heated at 400–500°C whereas physical activation was conducted at 800–1000°C. Activated carbon has high activity in adsorbing indicated by high iodine number resulted from analysis. SEM-EDS result confirmed that activated carbon made from coal has the quality in accordance to SNI and can be used as adsorbent in acid water treatment.

Keywords:

Abstrak (Indonesian)

Batubara Antrasit dan Sub Bituminus sebagai bahan baku karbon aktif dimanfaatkan khususnya di bidang pertambangan sebagai adsorben senyawa logam-logam berat yang berbahaya yang dihasilkan oleh aktivitas pertambangan. Pada penelitian ini aktivasi dilakukan secara fisika dan kimia dengan variasi suhu dan ukuran partikel. Karakterisasi dilakukan untuk melihat spesifikasi dari adsorben yang dihasilkan sehingga dapat digunakan dan diaplikasikan pada penelitian ini. Pengujian *proximate dan ultimate* dimaksudkan untuk mengetahui spesifikasi bahan baku. Hasil pengujian didapat nilai *fixed carbon* Antrasit sebesar 88,91 % sedangkan Sub Bituminus 49,05%. Pada aktivasi kimia pereaktif yang digunakan adalah NaOH dengan suhu 400-500^{o}C dan aktifasi fisika dilakukan pada suhu 800-1000^{o}C. Aktivasi karbon aktif dilihat dari analisa tingginya bilangan iodine dan hasil analisa SEM-EDS yang disimpulkan bahwa karbon aktif dari batubara memilki kualitas yang sesuai standar SNI dan dapat dimanfaatkan sebagai adsorben.

Keywords:

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INTRODUCTION

Indonesia has coal resource approximately 124,796.74 million tones with production level at 435 million tones [1] which is highly potential to produce activated carbon. Recently, coal is used to provide electricity by burning it in power plant. Coal utilization, as raw materials for activated carbon, not only provides the use of coal in different ways but also increase industrial competitiveness and comparative advantage of natural resource. Coal has relatively high carbon content particularly high rank coal such as anthracite and sub-bituminous hence suitable for activated carbon materials.

Activated carbon is a type of charcoal resulted from carbonization process and activation. In carbonization process, organic materials are break down into carbon (char form) at 300–600°C. The resulting charcoal has approximately 70–80%. In order to be used as adsorbent, charcoal need to activate so its surface area is expanding and pore size is widened [2].

Activated carbon typically has 15% water content, ~3% ash and ~83% carbon. Granular and powder are particle shape designed for conventional use of activated carbon nowadays. Carbon particle normally has limited porosity and surface area around 10–15 m^2/g. Upon activation, the surface area enlarge and reach 700–1200 m^2/g [3]. High surface area of activated carbon must be accompanied high flexibility to liquid and gas. High flexibility indicates good adsorption capacity of the carbon.

Several researches had been conducted on activated carbon synthesis and its application as adsorbent. Among raw materials has been used are coconut and palm shell. The activation process involved sulfate acid and phosphate acid as activating agents, soaked for 10 hours and used to adsorbed phenol compound. Activation process using phosphate acid provide charcoal with better water content, ash content and adsorption capacity compared to activated by sulfate acid [4]. Corn stalks had been processed into activated carbon using potassium hydroxide at various temperatures. The activated carbon obtained was able to adsorbed butanol up to 410 m/g [5]. Another researcher reported activated carbon synthesis from coconut shell using phosphate acid as activating agent. Its application on adsorption of red dye Procyon showed the activated carbon has adsorption capacity 278.197 mg/g [6]. Palm oil shell was also used to produce activated carbon using $ZnCl_2$ and physical activation. Good result was obtained when the activated carbon used to adsorbed methane in laboratory experiment [7]. Activation process using 4 M HNO_3 also reported at 180°C and applied to adsorbed creatinine. Activated carbon can adsorb the adsorbate up to 29% [8].

Figure 1. Coal result before and after physical and chemical activation

MATERIALS AND METHODS

Activated Carbon Preparation

Coal as raw materials was taken from Air Laya Mine of PTBA Tanjung Enim. Sample was prepared through size reduction using Jaw Crusher Restech followed by grinding in Baldor grinder and Sieve Shaker to obtained size of 100, 140 and 200 mesh. The resulting fine particle coal was subjected for carbonization process to produce activated carbon.

Carbonization Process

Carbonization was carried out in a muffle furnace at 300°C for 2 hours. The process will remove not only all water contained in coal but also volatile matter to formed charcoal. The solid material produced was porous with open pore structure and surface area bigger than initial materials.

Physical Activation

Activated carbon result from carbonization was activated physically through heating process at higher temperature 800, 900 and 1000°C in muffle furnace for 4 hours. Activated carbon prepared in various particle sizes was heated in the same temperatures. Heating in muffle furnace provide anaerobic atmosphere particularly when sample was place on a porcelain cup with cover. Minimizing oxygen supply prevented surface structure of activated carbon from damage or pore collapse upon heating. After 4 hours heating, furnace was cooled gradually until it reaches temperature below 100°C.

Chemical Activation

NaOH 2.5 M solution was prepared from initial concentration 40 M through dissolution using aquadest and the result used as activating agent. The procedure of chemical activation is as follows, 100 g NaOH dissolved in 1 L aquadest to obtain NaOH solution. 10 g charcoal was weighted, soaked in 100 mL NaOH solution and stirred for 18 hours. Activated carbon resulted was than precipitated and washed. Washing process was conducted to remove activating agent remains and indicated by neutral pH of solution. Activated carbon had cleaned was dried in oven at 120°C for 2 hours followed by calcination process in muffle furnace at various temperature (400, 450 and 500°C) for 4 hours.

Mine acid water tests

Activated carbon was used to remove heavy metals (Fe and Mn) in water as well as TSS and to increase pH so it fulfills quality threshold required for waste water from commercial and coal mine activity (Ministry of Environment provisioned no. 113/2003). The test procedure was carried out by placed activated carbon of weight 1, 2 and 3 g into Erlenmeyer which is embedded in a jar test. 200 mL of water was poured into each variation of carbon weight and then the Jar test run at 700 rpm for 1 hour without heat treatment. After 1 hour of running test, activated carbon was filtered and the filtrate was analyzed for remaining Fe and Mn. Activated carbon recovered from filtration was subjected for surface area analysis and SEM-EDS to provide information of surface structure changes and compound contained in its pore.

RESULT AND DISCUSSION

Proximate and ultimate analysis was conducted to confirmed coal specification of anthracite and sub-bituminous. Figure 1 shows coal result before and after preparation by physical and chemical activation. Analysis result of proximate and ultimate are display on Table 1 and 2.

Table 1. Proximate analysis result of coal

Parameter	Unit	Sample code	
		Anthracite	Sub Bituminous
Total Moisture	%, ar	4,47	22,97
Inherent Moisture	% ,adb	1,23	5,96
Ash Content	%,adb	1,23	1,98
Volatile Matter	%, adb	8,63	43,01
Fixed Carbon	% ,adb	88,91	49,05
Total Sulfur	% ,adb	1,17	0,36
Caloric Value	Cal/gr	8352	6998

Table 2. Ultimate analysis result of coal

Parameter	Unit	Kode Sampel	
		Anthracite	Sub Bituminous
Inherent Moisture	% ,adb	1,23	5,96
Ash Content	% ,adb	1,23	1,98
Total Sulfur	%, adb	1,17	0,36
Carbon (C)	% ,adb	77,35	60,82
Hydrogen (H)	% ,adb	3,75	6,72
Nitrogen (N)	% ,adb	1,70	1,08
Oxygen (O)	% ,adb	13,57	23,08

Fixed carbon contains in the samples according to Table 1 are relatively high. Anthracite has 88.91% fixed carbon and 8.63% volatile matter while sub-bituminous has 49.05% fixed carbon and 43.01% volatile matter. Both coal samples have ash content less than 5%. According to reference [2], coal with less than 5% ash content can provide iodine number between 600 and 700 mg/g. High content of fixed carbon made anthracite potentially as raw material for activated carbon which can be used for wide purposes such as adsorbent on impurity removal in waste water.

Carbonization process result

Figure 2 shows carbonization result for anthracite gave higher yield of activated carbon compare to sub-bituminous. The reason for this is mass shrinkage of sub-bituminous bigger than anthracite due to high volatile matter (43.01%) and hydrogen (6.72%) contained. High content of volatile matter and hydrogen can cause internal porosity evolved on sub-bituminous hence produced high result of carbon (70-90%). Sub-bituminous on application, potentially give more adsorption process compare to anthracite in consequence of higher internal porosity. In order to obtain even larger absorption, carbon resulted was subjected to chemical activation so it can open more pores and produce bigger pore size.

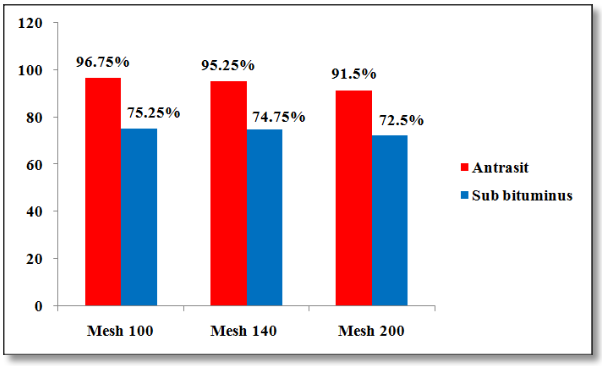
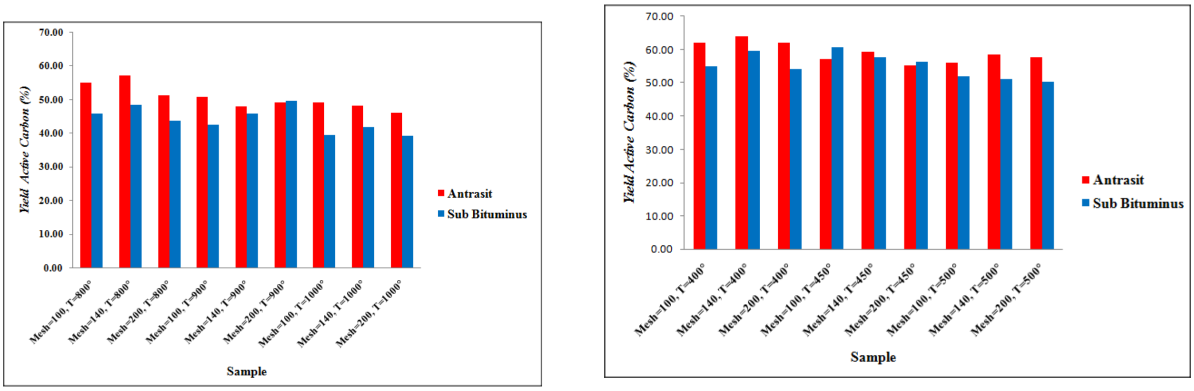


Figure 2. Coal mass reduction during carbonization process

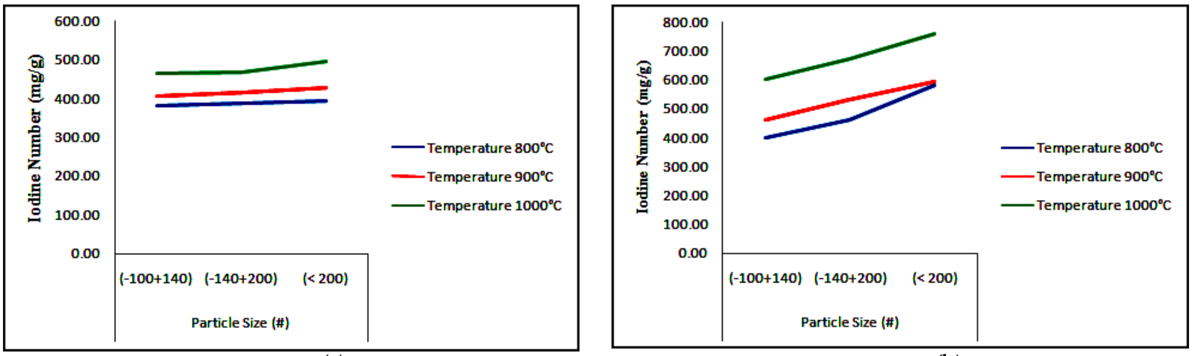
Result of activation process

Mass reduction measurement during activation process indicated chemical activation is smaller than physical activation. From the Figure 3, activated carbon yield for anthracite is between 46.54% and 66.35% whereas yield for sub-bituminous is 50.41% to 53.43%. Coal mass was loosed during activation due to decomposition and evaporated into air.



(a) (b)

Figure 3. Yield of Active Carbon for: (a) mass reduction during physical activation and (b) chemical activation



(a) (b)

Figure 4. Iodine number of activated carbon produced from (a) anthracite (b) sub-bituminous by physical activation

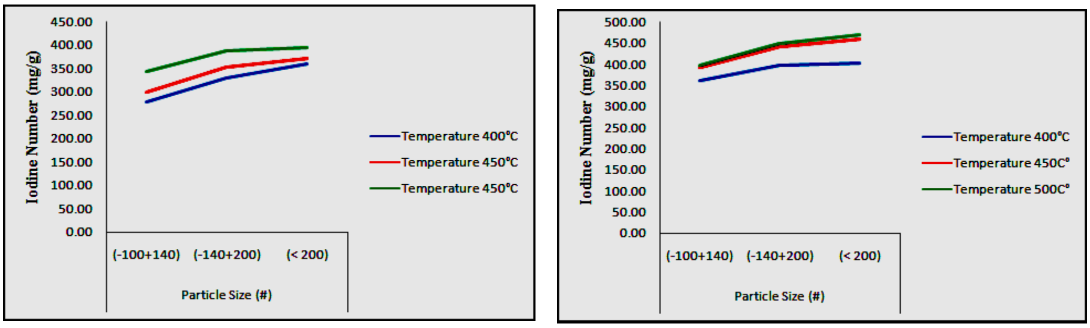


Figure 5. Iodine number of activated carbon produced from (a) anthracite (b) sub-bituminous chemical activation

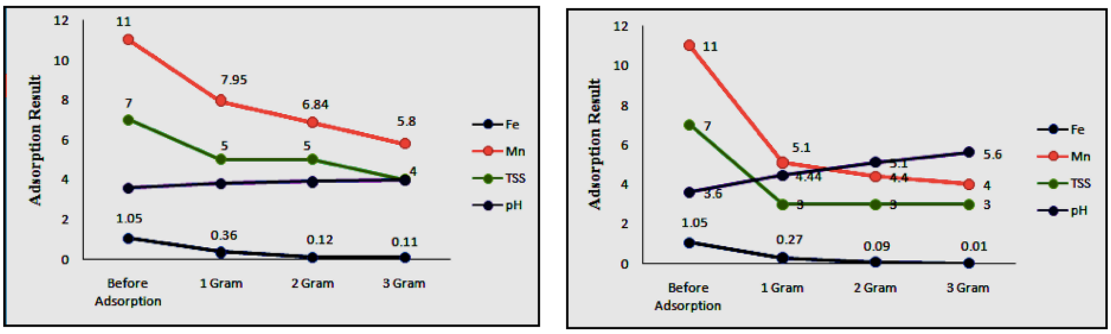


Figure 6. Result of heavy metal removal using activated carbon from (a) anthracite and (b) sub-bituminous by physical activation

Iodine number analysis result

Iodine number is one of several parameters that indicate activated carbon quality. The number represents amount of particular adsorbate uptake by activated carbon. The larger iodine number it has, the higher adsorption capacity of adsorbent.

According Figure 4, iodine number of activated carbon resulted from sub-bituminous is between 400.57 to 761.58 mg/g which was higher than anthracite with iodine number between 383.08 to 494.70 mg/g. Average value of iodine number calculated for sub-bituminous is 562.52 mg/g whereas anthracite is 426.90 mg/g. Result also found out that higher temperature of calcination gave better iodine number due to volatile matter released. This event caused pore widening of activated carbon.

In addition from Figure 5, Carbon from anthracite which was activation chemically gave iodine number test between 279.29 to 394.97 mg/g having average at 346.96 mg. Sub bituminous on the other hand provide carbon with iodine number between 361.28 to 470.74 mg/g having average at 418.79 mg/g. Compare to previous result, chemical activation gave iodine number smaller than physical activation of activated carbon result. Chemical compound used in activation process which is NaOH 4M was not able to provide higher iodine number both anthracite and sub-bituminous.

Iodine number also reveals higher when carbon from sub-bituminous rather than anthracite was activated both chemically and physically. Sub-bituminous appears to has volatile matter higher than anthracite 43.01% compare to 6.63% (adb). Upon carbonization, volatile matter release caused carbon product to evolve pore on its surface hence increase the adsorption capacity on application. Volatile matter hence plays important role in pore formation of activated carbon from coal.

Application result of activated carbon on Mine acid water

Heavy metal contained in mine acid water is quite large. Adsorption process using appropriate adsorbent can provide potential method for removing hazardous content. A porous material as shown by activated carbon is able to reduce heavy metal contamination in water. Result of heavy metal adsorption using activated carbon is shown on Figure 6.

The figure shows that heavy metal was able to be adsorbed by both adsorbents prepared. Activated carbon prepared from anthracite reduced Fe by 89.52%; Mn 47.2%; TSS 42.86% while pH increased from 3.6 to 3.97. Activated carbon from sub-bituminous on the other hand is able to reduced Fe by 99.04%; Mn 63.64%; TSS 57.14% while pH increased from 3.6 to 5.6.

Figure 7 depicted parameters measured reduction using activated carbon prepared by chemical activation. Both anthracite and sub-bituminous appears to reduced moderately compare to physical activation result. Anthracite was able to reduced Fe by 36.19%; Mn 26.72%; TSS 42.85% while pH increases from 3.6 to 4.22. Sub-bituminous reduced Fe by 86.66%; Mn 44.90%; TSS 42.86% while pH increased from 3.6 to 5.9.

Three parameters have been reduced significantly using activated carbon prepared from anthracite and sub-bituminous i.e. Fe, Mn and TSS. Activated carbon with iodine number >200 mg/g report previously to be able to adsorbed heavy metal approximately 70–80% [9] but cannot increased significantly pH. The reason for this is caused by gas which has acid property contained inside internal pore of adsorbent generated from pyrite (FeS_2). Gas from internal pore undergoes releasing process more effective by physical compare to chemical activation. Chemical activation according to the result cannot neutralize acid property of activated carbon pore.

SEM-EDS micrograph result

Coal based activated carbon requires two aspects to be concerned in order to obtain good result i.e. compound homogenization technique and special growing condition. Phase growth can be studied by using image result from SEM-EDS method [10]. The method not only provides micro structure image but also compositional analysis result of the growing phase with approximately 10% error factor. Analysis using SEM-EDS also does not cause damage on sample and no thin layer preparation needed. Figure 8 shows SEM micrograph of sub-bituminous coal under certain magnification.

Layered structure of sub-bituminous coal can be observed base on Figure 8. Fine particles of coal along with pore with medium radii also can be seen under larger magnification. Physical activation at 1000°C disrupt the structure and created pore with size 1.23–3.67 μm. Further analysis using SEM-EDS on activated carbon after being used for adsorption as shown by Figure 9 reveals metal particles at the adsorbent surface.

EDS identified the metal particles being adsorbed on carbon surface are Na, Ca, Mg and heavy metal Cu and Zn. Other researcher found out elemental composition of activated carbon made from coal is carbon, magnesium, potassium as well as iron [2].

CONCLUSION

Activated carbon is successfully prepared from anthracite and sub-bituminous coal. Iodine number of activated carbon from sub-bituminous is larger than anthracite via physical activation gave 761.58 compare to 494.70 mg/g. Chemical activation provide iodine number smaller i.e. 394.97 mg/g for anthracite and 470.74 mg/g for sub-bituminous. Application on heavy metal adsorption indicates activated carbon prepared from sub-bituminous is able to adsorb more than anthracite. Sub-bituminous seems to be potential candidate with adsorption capacity for removing Fe up to 90%.

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