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Comparison of Alum and Coal-Based Activated Carbon for the Treatment of Raw Water

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Abstract

The availability of clean water is an essential element for supporting the life cycle and sustainable development. The efficiency of providing raw water sources is an important factor for adequate water quantity and quality. This study examines the effectiveness of alum compared with coal-based activated carbon in the removal of Fe, Mn, and turbidity in raw water from the Lematang River, Muara Enim Regency. Activated carbon in this study was produced using a KOH activator with varying activation temperatures (700 °C, 800 °C, and 900 °C). The research results showed that the quality of the third activated carbon met the active carbon quality standards (moisture, ash content, Methylene blue dye and iodine adsorption capacity), but the activated carbon with activation at a temperature of 800 °C had the largest surface area. Activated carbon was proven more effective in reducing Fe and Mn concentrations than alum, while alum was superior in reducing turbidity. After three cycles of activated carbon regeneration, the reduction in Fe and Mn remained below 5%. These results show that activated carbon is a cost-effective and eco-friendly option for raw water treatment.

Keywords: Coal, activated carbon, alum, raw water, regeneration

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Abstrak (Indonesian)

Ketersediaan air bersih merupakan elemen penting dalam mendukung siklus kehidupan dan pembangunan berkelanjutan. Efisiensi pengolahan sumber air baku menjadi faktor penting untuk memastikan kuantitas dan kualitas air yang memadai. Penelitian ini mengevaluasi efektivitas alum dibandingkan dengan karbon aktif berbahan dasar batubara dalam mengurangi kandungan Fe, Mn, dan kekeruhan pada air baku yang berasal dari Sungai Lematang, Kabupaten Muara Enim. Karbon aktif dalam penelitian ini diproduksi menggunakan aktivator KOH dengan variasi suhu aktivasi (700 °C, 800 °C, dan 900 °C). Hasil penelitian menunjukkan bahwa kualitas ketiga karbon aktif memenuhi standar kualitas karbon aktif (kadar air, kadar abu, daya serap terhadap zat warna Metilen biru dan iodium), namun karbon aktif yang diaktivasi pada suhu 800 °C memiliki luas permukaan terbesar. Karbon aktif terbukti lebih efektif untuk menurunkan konsentrasi Fe dan Mn dibandingkan alum, sementara alum lebih unggul dalam menurunkan kekeruhan. Setelah dilakukan regenerasi karbon aktif hingga 3 siklus, penurunan efisiensi removal Fe dan Mn di bawah 5%. Dengan hasil ini menunjukkan karbon aktif merupakan pilihan yang ekonomis dan ramah lingkungan untuk pengolahan air baku.

Kata Kunci: batubara, karbon aktif, alum, air baku, regenerasi

INTRODUCTION

The Lematang River, located in the Muara Enim district, serves as a supply of raw water to drinking water according to the Regulation of the Governor of South Sumatra No. 16 of 2005. Various activities such as household activities, industry, agriculture and natural processes cause erosion of river quality to decrease and do not meet water quality for consumption and sanitation. This is a challenge for companies tasked with supplying drinking water, especially in developing countries [1,2]. Using the Lematang River for raw water is problematic due to its high turbidity value, Fe and Mn content, particularly during the wet season.

Coagulation and flocculation methods using alum (aluminum sulfate) are commonly used for raw water treatment [3,4]. The use of alum in water purification is effective in precipitating suspended substances and reducing turbidity. Moreover, it generates substantial quantities of sludge, necessitating additional management. The presence of alum in drinking water contributes to health issues, including presenile dementia, osteomalacia, and Alzheimer's disease [5,6]. Additionally, alum has been employed as an organic and inorganic compound adsorbent [7]. Alum has an adsorption capability of 1.974 mg/g for phosphate [8] and 22.10 mg/g for arsenic [9].

Adsorption is the most common conventional technology used to remove hazardous pollutants. This is due to the ease of application, low cost compared to other methods with modern technology, and abundant availability [10,11]. Various adsorbents have been used for river water purification, such as zeolite [12], activated carbon [13], bentonite [14], and iron oxide [15].

Activated carbon is an amorphous carbon-based material with a high surface area and porous structure [16]. Activated carbon is often used as an adsorbent for waste treatment due to its excellent adsorption capacity, thermal stability, ease of recycling, and strong chemical stability [17]. Raw materials and production processes influence the surface area. To increase the specific surface area, activation is carried out on the surface of activated carbon. Low-calorie coal has the potential as a raw material for activated carbon. This coal is accessible in substantial amounts as an underexploited natural resource. Its application in the synthesis of activated carbon enhances the value of resources that were once deemed less economically viable.

Chemical activation can use materials such as KOH [17,18], NaOH [19], H_3PO_4 [20], and Na_2SiO_3 [21]. Several studies have shown that KOH is an effective and commonly used activator. Physical activation enlarges pores, increases specific surface area, and removes volatile compounds so that activated carbon has a high adsorption capacity [22]. The results conducted by Bakara *et al.* [23] demonstrated that temperature activation affects the surface area of activated carbon from bamboo. At low temperatures, the activation process is not fully effective, while at very high temperatures, it causes the pore structure to be damaged. Previous research has made activated carbon from coal with KOH activator and an activation temperature of 600 °C, obtaining a surface area of 265.443 mg/m^2 [24].

This study aims to make activated carbon from coal with variations in activation temperature. Activated carbon with the largest surface area removes contaminants from raw air to reduce Fe, Mn, and turbidity compared to alum.

MATERIALS AND METHODS

Materials and Instruments

The material used in this research was coal from coal mines in the Tanjung Enim district. Raw water from the Lematang River is at the inlet section of raw water processing. Chemical ingredients, namely $Al_2(SO_4)_3$ (alum), KOH, NaOH, HCl, Methylene blue dye, and iodine are sourced from Merck, Germany.

Methods

Preparation activated carbon

Activated carbon was synthesized using the procedure of Subroto *et al.* [24] and Mi *et al.* [25]. After being purified of impurities, the coal was dried for 24 hours at 105 °C in an oven. Coal is crushed to a size of 140 mesh. Activation using a 1 M KOH solution was carried out by immersing coal with a mass ratio of 1:2. The mixture was placed in the oven heated at a temperature of 105 °C for 24 h. Next, calcination was carried out at different temperatures (700 °C, 800 °C, and 900 °C) with the names AC1, AC2 and AC3. The resulting activated carbon is washed with demineralized water until the pH is neutral. Finally, it is dried again in the oven using the same procedure. Moisture, ash content, Methylene blue dye, and iodine capacity are determined according to the Indonesian National Standard [SNI 06-3730-1995].

Characterization activated carbon

X-ray Diffraction (XRD Rigaku Miniflex 300 Japan) was used to analyze the crystal structure of activated carbon. Brunauer-Emmett-Teller (BET) surface area (NOVA Touch 4LX) was used to measure the specific surface area of activated carbon based on the principle of gas adsorption. Morphology and element composition were determined using Scanning Electron Microscopy - Energy Dispersive X-ray Spectroscopy (SEM-EDX JEOL JSM-6510LA). Fe and Mn concentration analysis using Atomic Absorption Spectrophotometers (Shimadzu AA-7800). Turbidity analysis using a turbidimeter (Turbidity Meter Turb® 430 T).

Treatment of raw water

In the raw water treatment process, 2 adsorbents are alum and activated carbon from coal. Three variables are used to optimize raw water processing, namely activated carbon/alum dosage (0.2 - 0.7 g/L), stirring duration (30 - 180 minutes), and stirring speed (100 - 400 rpm). The research used the bath method with a raw water Removal of Fe, Mn, and turbidity reduction is calculated based on the equation (1):

$$Removal\ (\%) = \frac{(C_i - C_e)}{C_i}\ x\ 100\ \% \qquad (1)$$

C_i dan C_e are the initial concentration and pollutant concentration at equilibrium

RESULTS AND DISCUSSION

Characterization of activated carbon

Table 1 displays the quality of activated carbon derived from coal at various temperatures, as well as the parameters of moisture, ash content, adsorption capacity for Methylene blue dye, and iodine levels. The results were compared to the quality standards for activated carbon in powder (SNI No. SNI 06-3730-1995), and all parameters satisfied the requirements. The initial concentration and pollutant concentration at equilibrium are determined by the maximal moisture and ash content of 15 and 10%, respectively, and the adsorption capacity of iodine dyes and Methylene blue dye of at least 120 mg/g and 750 mg/g, respectively.

Table 1. Activated carbon quality

Parameters	AC1	AC2	AC3
Moisture (%)	11.34	10.34	9.65
Ash content (%)	2.10	1.85	1.65
Methylene blue capacity (mg/g)	345.5	365.8	356.4
Iodine capacity (mg/g)	1835.4	1885.4	1960.6

Moisture and ash content are parameters that influence the performance of activated carbon. Water and ash in the activated carbon pores hinder the adsorption process, so the absorption capacity decreases. The adsorption capacity of activated carbon for iodine appears to be very large (> 750 mg/g). This indicates that activated carbon is of high quality and has better adsorption capabilities for small molecules. The adsorption capacity of Methylene blue dye indicates the ability of activated carbon to adsorb large molecules.

Figure 1 depicts the XRD pattern of activated carbon. The peak of activated carbon at an activation temperature of 900 °C (AC3) is more pronounced than at 700 °C (AC1) and 800 °C (AC2). The identical phenomenon was noted in the findings of Bakara *et al.* [20], where the peak intensity of the XRD spectra is augmented with the elevation of the activation temperature of bamboo-derived activated carbon

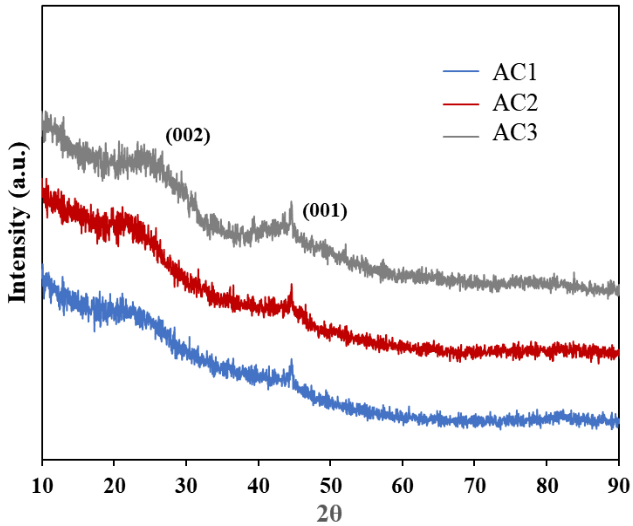

Figure 1. XRD patterns of activated carbon

According to the Scherer equation, which measures the crystallite size of activated carbon, AC3 is bigger than AC1 and AC2. Because more atoms are distributed periodically, the crystallite size is larger and has a higher peak intensity. The crystallite sizes are 16.8 nm, 21.84 nm, and 31.4 nm in order to increase activation temperature.

Table 2. Parameters of surface area

Parameters	AC1	AC2	AC3
Surface area (m^2/g)	279.235	321.325	316.260
Average pore diameter (nm)	3.133	3.436	3.611
Pore volume (cm^3/g)	0.217	0.312	0.286

Physical and chemical activation affects the surface area of activated carbon. Several studies show that KOH is an effective activator in the synthesis of activated carbon [27,28]. The mechanism that occurs using the KOH activator is as follows [29,30]:

$$6KOH + 2C \rightarrow 2K + K_2CO_3 + 3H_2 \qquad (2)$$

$$K_2CO_3 \rightarrow K_2O + CO_2 \qquad (3)$$

$$K_2CO_3 + 2C \rightarrow 2K + 3CO \qquad (4)$$

$$K_2O + C \rightarrow 2K + CO \quad (5)$$

$$CO_2 + C \rightarrow 2CO \quad (6)$$

The surface area of activated carbon at a temperature of 800 °C has the largest surface area (**Table** 2). Increasing the activation temperature causes the opening and formation of micropores in the activated carbon, thereby increasing the specific surface area. However, high temperatures cause small pores (micropores) to merge into larger pores, even closing the micropores. Apart from that, the porosity network can collapse, and the pore volume decreases, so the surface area decreases [30]. Research by Wang et al [31] shows that activated carbon from lignite coal and bituminous coal activated at a temperature of 900 °C has a higher surface area than that activated at 1100 °C. Activated carbon with activation at a temperature of 800 °C was used in further research.

The outcome of the SEM-EDX investigation of the morphology and element composition of AC2 is shown in **Figure** 2. It seems that the pore sizes of activated carbon are not uniform. These pores form as a result of chemical or physical activation during the formation of activated carbon. These pores enable molecule adsorption, which is essential for pollution removal.

The results of EDX analysis show that the main constituent of activated carbon is carbon. A high carbon content (generally >80%) indicates success in the raw material carbonization process. SEM-EDX results reveal that activated carbon is suited for applications requiring large surface area and excellent adsorption.

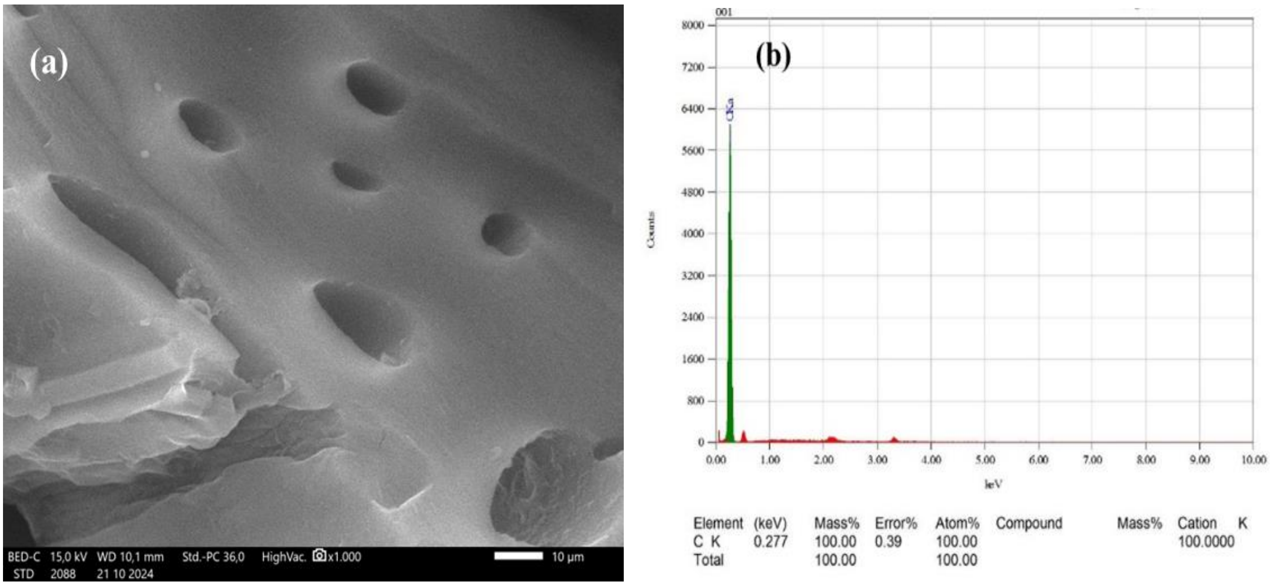

Figure 2. (a) SEM image and (b) EDX of activated carbon (AC2)

Treatment of Raw Water

Figure 3 presents the removal of Fe, Mn, and turbidity in raw water using alum and activated carbon. The previous results of the analysis of raw water from the Lematang River contain Fe = 5.68 mg/L, Mn = 3.29 mg/L, and turbidity = 51.60 mg/L [24]. The ability of activated carbon to reduce Fe and Mn is better than that of alum. Activated carbon has a porous structure with a large surface area and numerous micro, meso, and macro pores, making it effective in the adsorption of Fe and Mn ions through an adsorption mechanism.

Alum works by forming floc through reactions with Fe and Mn ions. This floc then settles so that the Fe and Mn levels in the water decrease. Activated carbon can work at various pH levels and different water conditions, making it more flexible than alum. However, alum is more effective in reducing turbidity, and its coagulation and flocculation processes are very efficient in removing suspended particles that cause turbidity, even at low concentrations. When alum is added, aluminum ions (Al^{3+}) bind to the particles and neutralize the charge, allowing the particles to combine. Next, the small particles combine to form larger flocs through the flocculation process. These flocs have sufficient weight to settle to the bottom of the container, reducing turbidity significantly [32,33].

The use of activated carbon and alum is influenced by dosage, stirring time, and stirring speed. Turbidity initially decreases as the dose increases until it reaches an optimal point but then increases again if more doses are added. If the coagulant dose continues to be added beyond the optimal dose, the excess coagulant molecules can cause the formed floc to become dispersed again due to the excess charge on the particles. As a result, turbidity begins to increase again, even though the amount of coagulant/adsorbent is greater. This phenomenon also decreases the turbidity of nano-banana peels [34].

Under optimum conditions, namely, a dosage of 0.5 g/L, stirring time of 120 minutes, and stirring speed of 250 rpm, the reduction in Fe, Mn, and turbidity using activated carbon was 74.33%, 83.91%, and 64.98%, while using alum it was 69.80%, 73.34%, and 93.08 %. The decrease in turbidity with different chemicals indicates that Poly Aluminum Chloride (PAC) at a dosage of 3.5 mg/L can diminish turbidity by 88%, while $FeCl_3$ at 30 mg/L can reduce turbidity by 77% [35].

The primary function of activated carbon is adsorption, which works better for dissolved organics but less well for eliminating big particles that produce turbidity. The application of chemicals like alum, PAC, and $FeCl_3$ seems to yield a higher percentage reduction in turbidity compared to activated carbon. However, the sludge coagulation process generates by-products. If this mud is not managed properly, it can pollute the surrounding environment, such as rivers, and reduce overall water quality

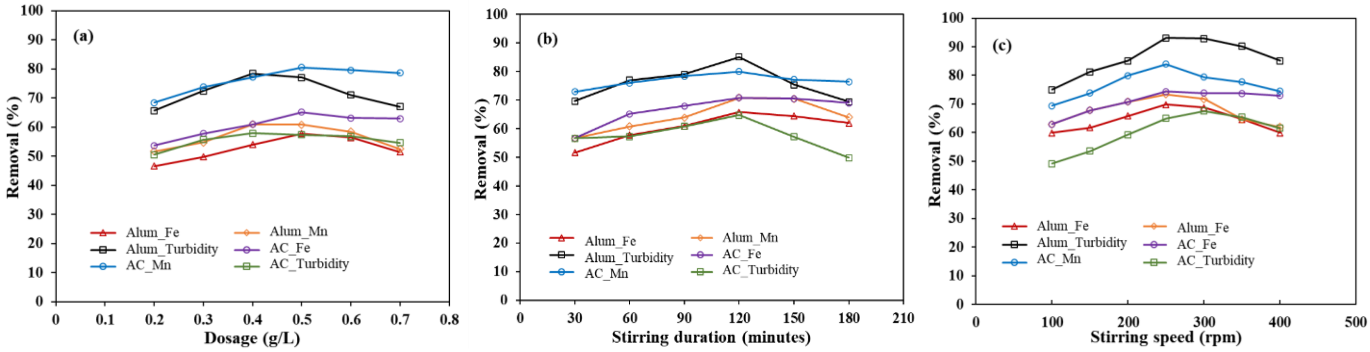

Figure 3. Treatment of raw water using alum and activated carbon

One of the advantages of using activated carbon for water purification is that it can be regenerated. Activated carbon can be regenerated because it can absorb and release molecules through physical or chemical processes. Activated carbon regeneration is economically profitable. Regeneration methods can be carried out thermally and non-thermally [36]. Non-thermally, this can be done using chemicals. In this study, regeneration was carried out using a 0.1 M HCl solution [15,24]. The acid solution dissolves the adsorbed, then rinsed with distilled water and dried for reuse.

Figure 4 is the result of three cycles of activated carbon regeneration. After three regeneration cycles, the reduction in Fe and Mn removal efficiency was under 5%, whereas turbidity above 5%. The decrease in Fe and Mn removal efficiency of less than 5% signifies that the activated carbon exhibits robust performance. Nonetheless, this does not align with turbidity. Adsorbed particles are not completely removed during regeneration. Materials such as high molecular weight organic compounds and colloids can adhere tightly to the surface of activated carbon.

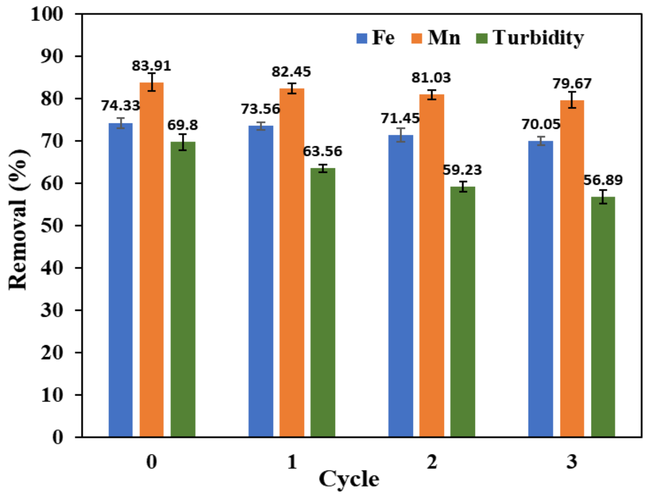

Figure 4. Regeneration of activated carbon for three cycles

CONCLUSION

This study examined the generation of activated carbon from coal using KOH as an activator at various activation temperatures (700 °C, 800 °C, and 900 °C). Activated carbon with an activation temperature of 800 °C has the largest surface area. Application of activated carbon to reduce Fe, Mn, and turbidity of raw water from the Lematang River by 74.33%, 83.91%, and

64.98%, respectively. The effectiveness of activated carbon in reducing Fe and Mn is greater than that of alum, namely 69.80% and 73.34%. For turbidity, alum is more effective, namely 93.08%. However, activated carbon has the advantage of being regenerated and reused.

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