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Powdered Activated Carbon (PAC)-Ceramic Composite Adsorbent for Iron and Aluminum Cations Removal from Acid Mine Drainage

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Highlights:

- This composite adsorbent was able to remove metallic cations of Fe (iron) and Al (aluminum) contained in acid mine drainage. The best removal results in this study were 60.87% for Fe and 52.13% for Al with a PAC:clay ratio of 45:55 (w/w) in 10 hours contact time.
- Increasing the PAC:ceramic ratio led to an increase in contaminant removal.
- The analysis results showed the effect of contact time on the contaminant removal, i.e., a longer contact time resulted in a higher contaminant removal.

Abstract. Acid mine drainage has become a serious problem globally, polluting groundwater with heavy metals. Adsorption is considered a simple and effective approach to addressing this emerging issue. A commonly used adsorbent is powdered activated carbon (PAC), but this is susceptible to being washed into the waste stream, either during or after the adsorption process due to its low density. This research combined PAC with clay that was molded into small clay balls (~1 cm in diameter) then baked at a very high temperature of 1000 °C to create a ceramic adsorbent. The adsorbent activation used NaOH 48% alkali solution to improve its capability in binding metallic cations. This research demonstrated that the PAC-ceramic composite is an efficient adsorbent for the removal of Fe (iron) and Al (aluminum) cations from acid mine drainage. The results showed that the most favorable contaminant removal was 60.87% for Fe and 52.13% for Al, using a PAC:clay ratio of 45:55 (w/w) in 10 hours contact time.

Keywords: *acid mine drainage; adsorbent; ceramic; composite; contaminant removal; powdered activated carbon (PAC).*

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1 Introduction

Acid mine drainage is a well-known environmental problem in many sulfidic mines. It is characterized by low pH resulting in an acidic environment and high concentrations of various heavy metals [1,2]. Heavy metal contamination is a critical environmental issue; therefore, appropriate steps must be taken to reduce heavy metals and metalloids in water to acceptable levels [3]. Several treatment methods have been developed recently to remove these pollutants, including adsorption, ion exchange, coagulation, and flocculation. Adsorption with ion exchange and filtration using membrane technology are the most commonly reviewed methods in the literature [4].

With fast developments in metal plating, mining, tanning, and other industries, heavy metal contamination from wastewater has been rising, particularly in developing nations. Heavy metal pollutants are not biodegradable, unlike organic pollutants. The accumulation of heavy metals is associated with hazardous effects on living organisms because these ions tend to be toxic and/or carcinogenic. Due to its effectiveness and simplicity, adsorption is frequently used for heavy metal removal in industrial wastewater [5]. The adsorption process has various engineered operations and mainly produces well-treated effluent. In addition, adsorption is known as a reversible process. Therefore, some adsorbents can be regenerated using a suitable desorption process [6].

Activated carbon is widely used as an adsorbent to remove heavy metal pollutants. Numerous researchers have studied the utilization of activated carbon to remove heavy metals [7]. Recently, decreasing coal-based commercial activated carbon sources has caused their price to increase. In order to improve the ability of activated carbon in heavy metal adsorption without additional costs, additives and activated composites can be used as valuable options. Such additive agents, for example, alginate [8], tannic acid [9], magnesium [10], and surfactants [11], have been proved to be effective as heavy metal adsorbents.

Various efforts have been done to find activated carbon from inexpensive and abundant alternative sources. It has been suggested that carbonaceous materials can be converted into activated carbon for heavy metal remediation [12]. Activated carbon can also be prepared from waste materials such as eucalyptus bark; its adsorption of Cu (II) and Pb (II) components has been reviewed [13]. The results showed that the highest contaminant removal for Cu (II) was 0.45 and 0.53 mmol/g for Pb (II), respectively. The major removal mechanism for both heavy metals is adsorption. Activated carbon manufactured from poultry litter to treat heavy metal-contaminated water has also been studied [14], revealing that it has better affinity in the adsorption process and higher capacity than activated carbons made from coconut shells or coals sold commercially.

Scientists have been searching for inexpensive and abundantly available adsorbents, especially for the removal of heavy metal ions. Many studies have been done on inexpensive adsorbents that have been developed, such as natural modified montmorillonite and kaolinite, which were studied as adsorbents for some types of heavy metals [15]. The kaolinite contained in clay from Longyan, China, has been studied as an adsorbent for heavy metal ions of Pb^{2+} , Cd^{2+} , Ni^{2+} , and Cu^{2+} from liquid waste. Adsorption within 30 minutes was observed to determine the maximum adsorption [16]. Clay containing kaolinite can be utilized to treat liquid waste containing Pb^{2+} . It is also able to reduce contamination from 160.00 mg/L to 8.00 mg/L using brine sediments to treat aqueous solutions to remove zinc and copper. The same adsorption evaluation was conducted using sawdust and a mixture of both sawdust and clay with a ratio of 1:40, resulting in an optimum contaminant removal of 5.59 mg/g for zinc and 4.69 mg/g for copper, respectively [6].

The activation of carbon prepared from bamboo using a physiochemical process with potassium hydroxide (KOH) and carbon dioxide (CO_2) as activating agents at 850 °C for 2 hours could adsorb methylene blue dye at 30 °C, resulting in an optimum monolayer contaminant removal of 454.2 mg/g [17]. The ability of activated carbon to adsorb contaminants is quite impressive, but the contaminant removal standard of activated carbon still needs to be improved. In order to increase the contaminant removal, sodium dodecyl sulfate (SDS) as anionic surfactants, dioctyl sulfosuccinate sodium (DSS) or sodium dodecylbenzene sulfonate (SDBS) are utilized to impregnate the activated carbon. Surfactant-impregnated activated carbon could remove 0.198 mmol/g of Cd (II) [11].

Activated carbon is usually crushed into a powder known as powdered activated carbon (PAC) to enhance its specific surface area and create more adsorption sites. However, the powdered structure of activated carbon has a low density and tends to be washed into the waste stream. Therefore, PAC needs to be bound with other heavier material and the material used for binding must also be a good adsorbent that does not reduce PAC adsorption capacity. This study chose clay as the binding material for activated carbon because it is inexpensive, abundantly available and a good adsorbent for metal ions [15-16]. This study investigated a technique of combining and modifying activated carbon with clay into a new kind of composite ceramic adsorbent to remove heavy metal ions contained in acid mine drainage wastewater.

2 Material and Method

The clay used in the adsorbent material is a kind of natural clay from South Sumatera, Indonesia. The clay and the activated carbon must be in powder form. The clay was made into powder by removing its moisture content and then

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pulverizing the dry remains. The powdered clay was rehydrated by adding water for molding purpose. Sample preparation was conducted by varying the adsorbent PAC:clay ratio (5:95, 10:90, 15:85, 20:80, 25:75, 30:70, 35:65, 40:60, and 45:55) (g/w). The clay and carbon powders were mixed thoroughly. A small amount of deionized water was added to the mixture so it could be molded into small clay balls ~1 cm in diameter. Then, these clay balls were calcined in a furnace at temperatures of 1000 °C for 12 hours. In order to enhance the contaminant removal, activation of the adsorbent needs to be carried out to remove any impurities in the cavities of the pore system in the adsorbent surface. Impurities cause the pores to become thinner and/or clogged. Activation of the adsorbent was done by immersing the ceramic balls in NaOH 48% solution for an hour. Then, the ceramic balls were washed three times with deionized water. Afterwards, they were put in an oven for another hour at 150 °C for the final drying process.

The adsorbent dosage for each experiment was 700 g for 500 ml acid mine drainage wastewater, the range of contact times used was 1 to 10 hours. The adsorbent was studied to analyze its adsorptive ability in removing heavy metal pollutants contained in acid mine drainage wastewater. The contact between activated carbon-ceramic adsorbent and acid mine drainage occurred within a column made of glass with 5.2 cm diameter and 50 cm height. The experiments were conducted in batches.

The concentrations of Fe and Al contained in the liquid samples were analyzed before and after contact with the adsorbent using an atomic absorption spectrophotometer (AAS). The contaminant removal was calculated using the following Eq. (1):

$$\text{Re} = \frac{(C_x - C_y)V}{W} \quad (1)$$

The contaminant removal percentage was calculated using the following Eq. (2):

$$\% \text{Re} = \frac{C_x - C_y}{C_x} \times 100\% \quad (2)$$

where Re is the contaminant removal (mg/g), C_x and C_y are the concentrations of the cations before and after the adsorption process, V is the volume of the sample solution (mg/L), V is the volume of the sample solution (L), and W is the weight of the adsorbent (g).

3 Results and Discussion

Table 1 and Figure 1 show that at each adsorbent ratio, the contaminant removal increased. Before the adsorption process, the initial metallic cation concentrations were 3.50 mg/L for Fe and 10.12 mg/L for Al. The adsorbent ratio has a crucial effect on the contaminant removal. The Fe removal rose from 1.06 mg/g to 2.98 mg/g, and the Al removal rose from 2.92 mg/g to 7.39 mg/g with adsorbent ratios ranging from 5:95 to 45:55 (w/w). The best contaminant removal results were achieved with ratios of 45:55 with 29.71% for Fe and 25.59% for Al. The addition of PAC mass to the adsorbent ratio contributed to an increase the pore volume, which resulted in greater contaminant removal.

Table 1 Contaminant removal of various adsorbent ratios in 5 hours contact time.

PAC:Clay Ratio	Contaminant removal (mg/g)	
	Fe	Al
5 : 95	1.06	2.92
10 : 90	1.29	3.00
15 : 85	1.32	3.21
20 : 80	1.49	3.69
25 : 75	1.72	4.32
30 : 70	2.06	5.18
35 : 65	2.44	6.00
40 : 60	2.75	7.00
45 : 55	2.98	7.39

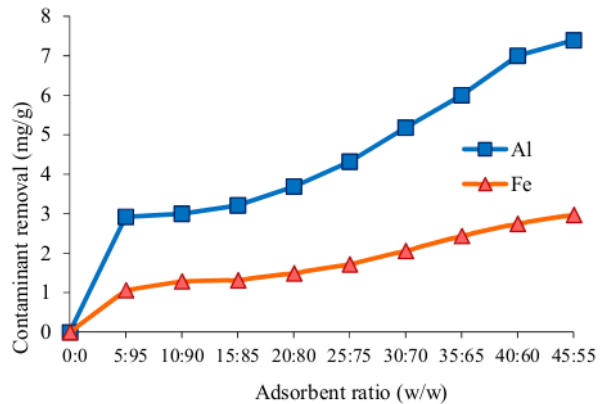


Figure 1 Contaminant removal of various adsorbent ratios in 5 hours contact time.

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The performance characteristics for different contact times in binding heavy metal cations can be seen in Table 2 and Figure 2, i.e., higher removal of metallic cations, resulting from a longer contact time. The observed Fe adsorption increased from 0.92 mg/g to 6.10 mg/g, and the Al adsorption increased from 1.89 mg/g to 28.90 mg/g with increasing contact times from 1 to 10 hours. The best contaminant removal results were 60.87% for Fe and 52.13% for Al in 10 hours contact time. The adsorption data from Table 2 and Figure 2 were derived from the adsorbent adsorption ability, which is strongly related to contact time. The longer the contact time, the higher the amount adsorbed. The contact time may have allowed enough opportunity to create binding sites between the adsorbent and the metal cations of Fe and Al on the adsorbent surface.

Table 2 Contaminant removal in various contact times using 45:55 (w/w) adsorbent ratio.

Contact time (hours)	Contaminant removal (mg/g)	
	Fe	Al
1	0.92	1.89
2	1.68	3.56
3	2.21	4.34
4	2.77	5.19
5	2.98	7.39
6	3.04	12.09
7	3.16	18.61
8	4.02	22.75
9	5.46	27.65
10	6.10	28.90

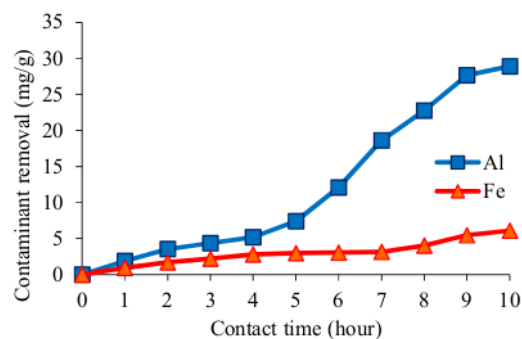


Figure 2 Contaminant removal in various contact times using 45:55 (w/w) adsorbent ratio.

A large surface area of the adsorbent usually has a large number of pores with unique shapes and different sizes. The International Union of Pure and Applied Chemistry (IUPAC) defines three types of well-known pores: macropores, mesopores, and micropores. The pore types of an adsorbent that are most relevant to the mass transfer to the inside of the material are macropores and mesopores. At the same time, the micropore volume mainly determines the internal surface size as well as the adsorbent removal capability. As mentioned, the surface area increases internally due to the increase of the micropore volume. In principle, the larger the micropore volume, the more significant the amount of adsorbate that can be adsorbed. The chemistry on the surface area affects the adsorbate and adsorbent interaction concerning the adsorbent types. This usually happens during the adsorption of ions into engineered adsorbents relevant to the activated carbon adsorption process.

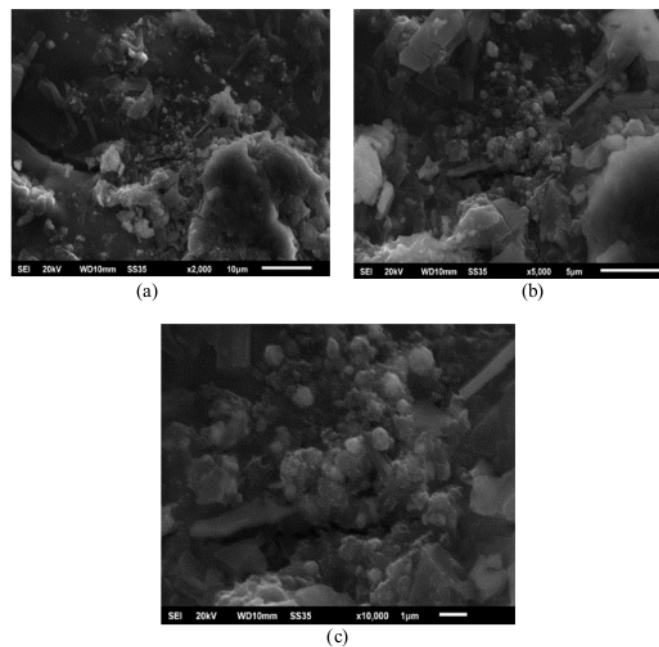


Figure 3 (a-c) SEM images of 45:55 w/w PAC-ceramic composite adsorbent before the adsorption process with 2000, 5000, and 10,000x magnification.

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By comparing Figures 3(a)-(c) with Figures 4(a)-(c) it can be seen that the characteristic surface area in Figure 3 is entirely different from that in Figure 4. This is due to the smaller amount of material attached to the surface in Figure 3. In contrast, the surface images in Figure 4 seem to be covered entirely by attached material. The adsorbed ions are usually bound firmly to the first outer surface compact layer of the adsorbent. Figures 3 and 4 show that the ion adsorption may have caused some changes in the complexity of the adsorbent surface, but this depends on the adsorption process type.

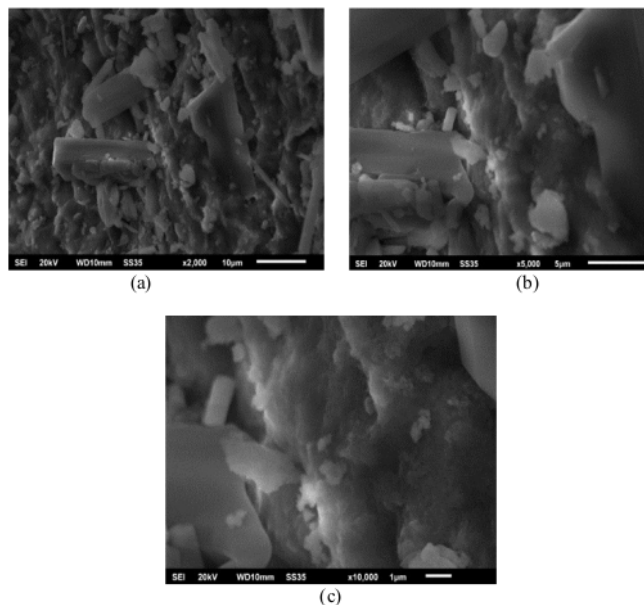


Figure 4 (a-c) SEM images of used 45:55 w/w PAC-ceramic composite adsorbent after the adsorption process with 2000, 5000, and 10,000x magnification.

The corresponding EDS spectra analysis in Figures 5 and 6 show that the PAC-ceramic composite adsorbent was composed of C, O, Na, Al, Si, Ti, K, and Fe. When the mass percentages in Table 3 are compared with those in Table 4, it can be seen that there were increases in the composition percentages from 3.71% to 7.75% for Al and from 0.54% to 1.38% for Fe. This shows the addition of Al and Fe into the adsorbent surface after the adsorption process, proving that some Al

and Fe cations in wastewater were bound to the surface and were adsorbed into the adsorbent pores.

It has been shown that a longer contact time enhances the adsorption process. The alkaline groups on the surface of the activated carbon bind the cations from the wastewater to the adsorbent surface during the adsorption process in an aqueous solution [18,19]. Fe and Al cations must have interacted with the adsorbent during the adsorption process. In this case, the surface layer of the activated carbon is basic. This is the result of the activation process in which the adsorbent was immersed in NaOH 48% solution.

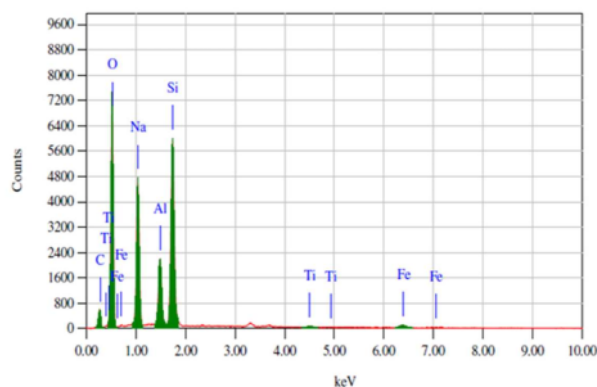


Figure 5 Energy-dispersive X-ray spectroscopy (EDS) spectra analysis of 45:55 w/w PAC-ceramic composite adsorbent materials before the adsorption process.

Table 3 Elemental mass analysis of adsorbent materials before the adsorption process.

Element	(keV)	Mass %	Atom%
C	0.277	13.75	19.46
O	0.525	58.34	62.00
Na	1.041	12.90	9.54
Al	1.486	3.71	2.34
Si	1.739	10.66	6.46
Ti	4.508	0.10	0.03
Fe	6.398	0.54	0.16

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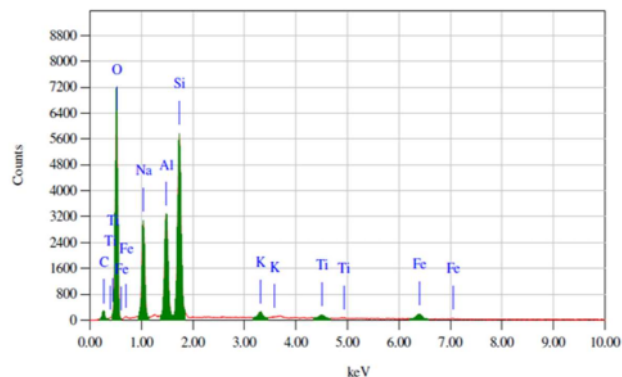


Figure 6 EDS spectra analysis of 45:55 w/w PAC-ceramic composite adsorbent after the adsorption process.

Table 4 Elemental mass analysis of adsorbent materials after the adsorption process.

Element	(keV)	Mass %	Atom %
C	0.277	2.44	3.76
O	0.525	61.62	71.33
Na	1.041	11.48	9.25
Al	1.486	7.75	5.32
Si	1.739	14.33	9.45
K	3.312	0.56	0.25
Ti	4.508	0.46	0.19
Fe	6.398	1.38	0.46

The reaction occurs at or below room temperature via initial activation of the carbon head groups C-O⁻ with water adsorbed on the hydrophilic surface to form reactive hydroxyl groups. Subsequent reaction with surface-bound hydroxyl groups yields stable bonds between the outer layer and the metal cations material. The descriptions are as follows [20]:



The following formulas explain the sorption of Fe and Al:



4 Conclusion

Chemical and physical properties of ceramic adsorbent ratios between activated carbon and clay greatly affect the removal performance of metallic cations of Fe and Al. The greater the mass of activated carbon in the adsorbent material, the higher the removal of Fe and Al cations. At the same time, the analysis results showed that the contact time affected the contaminant removal; the contaminant removal was increased when the contact time was extended. This study revealed that this adsorbent was effective in reducing metallic cations of Fe and Al contained in acid mine drainage; its most favorable results were 60.87% for Fe and 52.13% for Al using PAC:clay ratio 45:55 (w/w) in 10 hours contact time.

Nomenclature

Re	=	contaminant removal
C_x	=	cations concentration before adsorption process
C_y	=	cations concentrations after adsorption process
v	=	tested solution volume
W	=	adsorbent weight

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