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Application of Coal Fly Ash as Adsorbent for Removing Lead (Pb) Metal

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Abstract. *Coal fly ash (CFA), a major byproduct of coal combustion, poses significant environmental challenges due to its hazardous constituents, including heavy metals. This study explores the potential of CFA sourced from PT PLN Nusantara Power UPK Nagan Raya as a low-cost adsorbent for Pb removal from aqueous solutions. The research investigates the influence of adsorbent dosage and stirring time on adsorption efficiency and capacity, aiming to optimize conditions for practical application in wastewater treatment. Characterization of CFA revealed a composition dominated by silica and alumina, with crystalline phases such as quartz and mullite, which provide active sites for adsorption. Batch experiments were conducted at varying CFA doses (2–8 g) and contact times (30–90 minutes), with pH fluctuations monitored throughout the process. Results indicate that adsorption efficiency increases with higher adsorbent dosage and extended stirring time, achieving a maximum Pb removal of 99.684% at 8 g and 90 minutes. Conversely, adsorption capacity (mg/g) was highest at lower doses, peaking at 0.945 mg/g for 2 g at 90 minutes, reflecting the trade-off between efficiency and capacity due to particle agglomeration and site saturation at higher doses. The findings confirm CFA's dual mechanism of Pb removal through surface complexation and precipitation, influenced by pH and mineral composition. This study demonstrates that CFA can serve as an effective, sustainable adsorbent for heavy-metal remediation, offering a circular-economy solution for coal combustion residues. Future work should address regeneration strategies and safe disposal of spent adsorbent to prevent secondary contamination.*

Keywords: *Coal Fly Ash, Adsorption, Lead Removal, Wastewater Treatment, Adsorption Efficiency*

1. Introduction

Fly ash is a material derived from coal combustion that has a grayish color and fine grain size. Fly ash contains chemical elements including silica (SiO_2), alumina (Al_2O_3), ferrous oxide (Fe_2O_3), and calcium oxide (CaO). It also contains additional elements such as magnesium oxide (MgO), titanium (TiO), alkaline earth (Na_2O), and others (Fatimah et al., 2021). Coal combustion produces approximately 75%-80% fly ash. Therefore, fly ash generated from coal combustion requires storage. The dominant SiO_2 and Al_2O_3 content gives fly ash its adsorption capacity, which is used to remove pollutants from liquid waste (Ambia, 2021). However, currently, based on Government Regulation Number 22 of 2021 concerning the Implementation of Environmental Protection and Management, fly ash and bottom ash are no longer categorized as B3 waste because the company has already managed B3 waste to make it non-hazardous. However, the coal-fired power plant (PLTU) still needs to elaborate its policies to reduce the area of storage and its reuse capacity.

Coal fly ash (CFA) as a coal combustion residue (CCR) comprising fly ash, bottom ash, boiler slag, and FGD gypsum. These materials can contain trace toxicants (Pb, As, Hg, Se), necessitating careful handling. The U.S. EPA's 2015 CCR Rule established National impoundment criteria for landfills and impoundments—location restrictions, liners, groundwater monitoring, corrective action, and structural integrity—because uncontrolled disposal risks groundwater contamination, dust emissions, and catastrophic failures. This policy backdrop underscores the environmental imperative for beneficial reuse strategies like adsorption, which simultaneously address waste and water quality. One of the effects of non-biodegradable lead (Pb) is its bioaccumulation in living organisms (Mahardhika et al., 2016). Lead is highly toxic and can cause acute and chronic lead poisoning, such as digestive problems. Lead (Pb) is a weak carcinogen that can cause lung cancer, stomach cancer, and glioma. (Adhani *et al.*, 2017).

Several studies have shown that fly ash and bottom ash can be used to neutralize acid mine drainage at certain concentrations. Research by Telaumbanua (2017) investigated the use of fly ash and palm oil bottom ash to absorb color in industrial wastewater. The results showed the highest color and COD reduction with fly ash adsorbent, with a color value of 96.96% and a COD concentration of 83.26%. Meanwhile, color reduction using bottom ash and fly ash was 53.93% and COD reduction was 62.82%. Furthermore, bottom ash adsorbent resulted in a color reduction of 53.03% and COD reduction of 71.08%. Study done by Ambia (2021) found that the optimum mass of 100 g/L resulted in an adsorption effectiveness of 98.09% for Mn, 97.41% for Zn, 97.20% for Fe, 78.39% for BOD, and 14.80% for COD.

This research focuses on utilizing CFA waste as an absorbent to remove lead (Pb) metal in solution. In wastewater treatment, to enhance the ability of coal fly ash to absorb the heavy metal in wastewater, modification of carbon CFA must be done. Study by Myllymäki et al., 2018) state that the surface properties of an adsorbent influence the amount of fluid that can be adsorbed onto the adsorbent. This leads to adsorbent modification to make it more effective in removing Pb. The adsorbent surface is modified through activation.

In Indonesia (and Aceh specifically), CFA arises from coal-fired power plant (PLTU) such as in Nagan Raya; its characterizations show silica- and alumina-rich ash with quartz and mullite phases, spherical particles, and sometimes notable CaO content (Darmayanti et al, 2018). Those traits matter because higher Ca can raise pH and favor surface precipitation of $Pb(OH)_2$ or $PbCO_3$ in carbonate-bearing systems, whereas silica/alumina glass supports ion exchange and complexation at deprotonated surface sites. Converting CFA from a disposal liability to a water-treatment asset amplifies circular economy benefits and reduces environmental footprints when engineered safely (e.g., preventing secondary leaching).

Based on the background, the research aims to use the Nagan Raya CFA as an adsorbent for removal of Pb(II) from aqueous solution. The core design variables that control adsorption outcomes focused on the adsorbent dosage and stirring/contact time and assess efficiency (%) and capacity (mg/g). The urgency of this study was to compare the literature that consistently shows: (i) increasing dose raises % removal (more sites) but often lowers capacity (mg/g) because the same solute mass is distributed over more adsorbent; and (ii) longer contact time improves removal up to equilibrium, beyond which changes are minimal. We hope that the use of coal fly ash from PLTU Nagan Raya will strongly align with sustainability principles by transforming a local plant waste stream into a solution for local wastewater quality challenges.

2. Method

The research conducted on the laboratory scale is based on the steps below (Sopiah dkk., 2017):

a) Preparation for adsorbent
   a. Collect coal fly ash (CFA) from the PLTU Nagan Raya power plant.
   b. Dry the CFA to remove moisture and heat it to reduce unburned carbon.
   c. Grind and sieve the CFA to obtain a uniform particle size of 100 mesh.

b) Preparation of Lead solution
   a. Prepare a synthetic Pb(II) solution at a known concentration using lead nitrate dissolved in distilled water.
   b. Measure and adjust the initial pH of the solution as required.

c) Batch Adsorption Experiments
   a. Add a specific mass of CFA (e.g., 2 g, 4 g, 6 g, 8 g) into separate beakers containing a 500 ml fixed volume of Pb solution.
   b. Stir the mixture at a constant speed for predetermined contact times (30, 60, and 90 minutes).

d) Monitoring Parameters
   a. Record pH changes during the adsorption process.
   b. After the contact time, filter the solution to separate the adsorbent.
   c. Collect the filtrate for Pb concentration analysis using Atomic Absorption Spectroscopy (AAS) or ICP.

e) Data Analysis
   a. Calculate the water content and ash content of adsorbent, pH fluctuation during the adsorption process, and adsorption efficiency (%) and adsorption capacity (mg/g) using standard formulas.
   b. Compare results across different doses and contact times to determine optimal conditions.

3. Results and Discussions

3.1 Characteristics of CFA

Fig 1. presented the visual comparison of CFA pre- and post-heating. The color/lightness of CFA changes as unburned carbon is reduced, and physically adsorbed moisture is removed. Table 1. Showed the water and the ash content characteristics compared to SNI of technical charcoal. The lower moisture ensures a consistent effective dose (mass of dry CFA per liter), while ash content provides a proxy for residual carbon, which can influence adsorption via oxygenated functional groups and hydrophobic domains. Prior studies note that unburned carbon can be beneficial for certain adsorptive interactions, though excessive carbon may mask mineral sites or complicate pH behaviour.

Figure 1. Coal fly ash (CAF) before (left) and after heating (right).

Table 1. Analysis of water and ash content compared to Indonesian standard.

No	Parameter	Results (%)		Limits of National Technical Charcoal	SNI 06-3730-1995
		Before Treatment	After Treatment		
1	Moisture content	11	2.25	Max 15%	Meet
2	Ash content	7	3.9	Max 10%	Meet

Heating (mild calcination) can also alter surface hydroxyl density and reduce volatile contaminants, marginally affecting initial pH upon contact with water. For Ca-rich ash, thermal history influences free lime and anhydrite phases, which in turn shape slurry alkalinity. These changes help explain the pH trends you document later and the efficiency differences across doses. CFA from Nagan Raya has been shown to contain significant Si/Al, with quartz and mullite detectable by XRD—even in derivative materials used for silica extraction—confirming the structural basis for adsorption.

Table 1. presents the moisture and ash content of fly ash before it became adsorbent, with a moisture content of 11, which is still below the quality standard, and an ash content of 7, which is below the quality standard. Based on the analysis of the activated carbon characteristics above, it can be explained that the moisture content of coal fly ash before it became an adsorbent was 11%, while after it became an adsorbent, the moisture content decreased to 2.25%. The characteristics of the resulting fly ash adsorbent meet SNI 06-3730-1995 standards, which are below 15%. Analysis results show that the water content before and after adsorption is 11% and 2.25%, while the ash content before and after adsorption is 7% and 3.9%. The use of coal fly ash is an alternative method for absorbing the heavy metal Pb. The characteristics of coal fly ash within the Indonesian standard made it the CFA reliable to be used as one of local adsorbent. The advantages of using fly ash include its easy availability and relatively low cost as a replacement for previously used adsorbents. It also reduces the solid waste generated by the Nagan Raya PLTU.

3.2 Effect of Dosage and Stirring time on the Pb Removal

Figure 2. illustrates the effect of adsorbent dosage (2–8 g) and mixing time (30, 60, and 90 minutes) on the residual concentration of Pb in solution. The initial Pb concentration was approximately 3.8 mg/L before treatment. By increasing the adsorbent dosage significantly reduced the Pb concentration in solution across all mixing times. At a dosage of 2 g, a substantial decline in Pb concentration was observed, particularly at 60 and 90 minutes, where the residual concentration decreased to approximately 0.04 mg/L and 0.02 mg/L, respectively. In contrast, at 30 minutes, the concentration remained higher (1.1 mg/L), indicating that contact time plays a critical role in achieving adsorption equilibrium. As the adsorbent dosage increased from 2 g to 8 g, the residual Pb concentration further decreased, reaching values close to zero at longer mixing times (60 and 90 minutes). The lowest Pb concentration was achieved at 8 g dosage and 90 minutes of mixing (approximately 0.012 mg/L). The results indicates that both higher adsorbent dosage and longer contact time enhance removal efficiency.

The reduction in Pb concentration with increasing adsorbent dosage can be attributed to the greater availability of active adsorption sites and increased surface area, which facilitate metal ion binding (Foo & Hameed, 2010; Bhatnagar & Sillanpää, 2010). At lower dosages, adsorption sites may become saturated quickly, limiting removal efficiency. Conversely, higher dosages provide more binding sites, increasing the probability of Pb ion adsorption. The effect of mixing time suggests that adsorption occurs rapidly during the initial stage (0–30 minutes), followed by a slower approach toward equilibrium. This behavior is consistent with typical adsorption kinetics, where rapid surface adsorption is followed by gradual intra-particle diffusion into porous structures (Ho & McKay, 1999). The near-complete removal observed at 60–90 minutes indicates that equilibrium conditions were likely reached within this time range.

Interestingly, slight fluctuations at higher dosages (e.g., minor increases at 6 g for 30-minute contact) may be associated with particle aggregation or overlapping of adsorption sites, which can reduce effective surface area at excessive dosages (Crini & Badot, 2008). However, the overall trend confirms improved Pb removal performance with optimized dosage and sufficient contact time. These findings align with previous studies reporting that heavy metal adsorption efficiency is strongly influenced by adsorbent dosage and contact time. For example, Gupta and Nayak (2012) demonstrated that increasing adsorbent mass significantly enhanced Pb(II) removal due to increased surface functional groups. Similarly, Babel and Kurniawan (2003) reported that equilibrium adsorption of heavy metals typically occurs within 60–120 minutes depending on adsorbent characteristics.

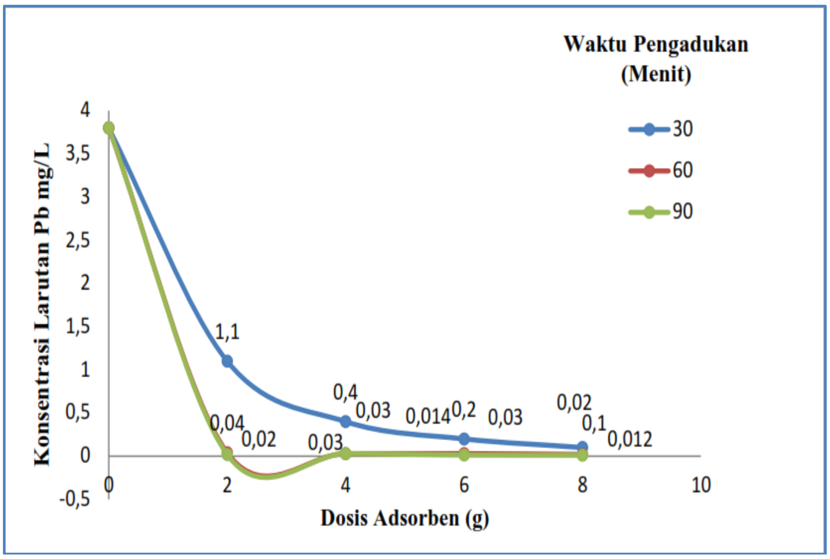

Figure 2. Effect of dosage of adsorbent and stirring time on Pb removal (mg/L)

3.3 pH Fluctuation at Different Dosage and Stirring Time

Table 2. showed the pH fluctuation during the adsorption process at variated stirring time. Based on the results of the pH test after adsorption, the initial pH ranged from 5.9 to 7.2. At 2 grams, the pH increased to 7 in 30 minutes, 7.1 in 60 minutes of stirring, and 7 in 90 minutes. The highest increase was at 6 grams, with an increase of 7.2 in 90 minutes, 7 in 60 minutes, 7 in 30 minutes, and 9 grams with an increase of 7.2 in 90 minutes, 7.1 in 60 minutes, and 7.1 in 30 minutes. The pH elevated with increasing fly ash adsorbent mass and stirring time. When metal elements dissolve in water, they dissociate into metal ions and other ions. Under certain conditions, these metal ions can be absorbed by the adsorbent through Van der Waals forces, the attractive forces that occur between the adsorbent particles and the metal ions.

Table 2. Fluctuation of pH during the adsorption process at variation of dosage

and stirring time.

No	Parameter	Dosis Adsorben (gram)	Variasi Waktu (Menit)	Nilai Awal pH	Nilai Akhir pH	Baku Mutu
1	pH	2	30	5,9	7	6-9
2			60	5,9	7,1	6-9
3			90	5,9	7	6-9
4		4	30	5,9	7,1	6-9
5			60	5,9	7,1	6-9
6			90	5,9	7,1	6-9
7		6	30	5,9	7	6-9
8			60	5,9	7	6-9
9			90	5,9	7,2	6-9
10		8	30	5,9	7,1	6-9
11			60	5,9	7,1	6-9
12			90	5,9	7,2	6-9

Based on Tabel 2. In general, the longer the stirring time, the greater possibility interactions between the adsorbent and the ions in the solution, which can cause changes in pH (Wakkel et al., 2019). If the adsorbent could capture H+ ions, the addition of adsorbent will cause an increase in the solution's pH (Xie et al., 2017). However, changes in pH can also be influenced by the properties of the adsorbent and the ions in the solution (Siddiqui et al., 2019).

3.4 The Adsorption Capacity

Figure 3. showed the result of adsorption capacity at variation of dosage and stirring time using fly ash activated carbon.

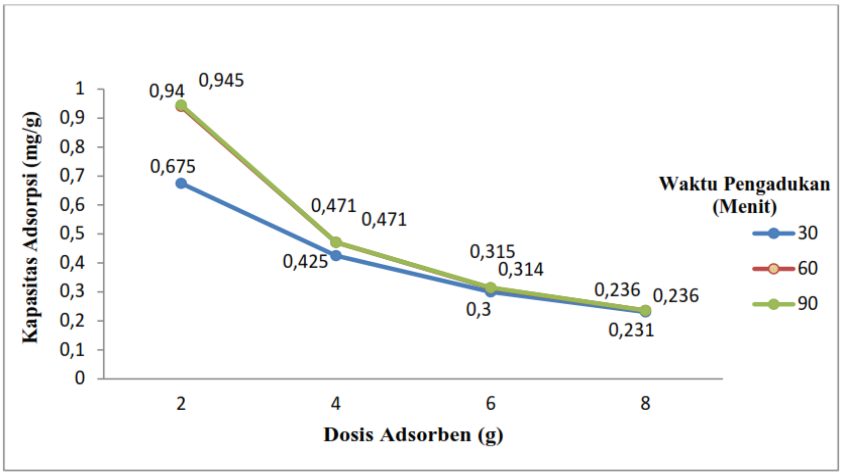

Figure 3. Adsorption capacity at variated dosage and stirring time

Based on Figure 3, the highest Pb adsorption capacity was obtained at an adsorbent dosage of 2 g with a stirring time of 90 minutes, reaching 0.945 mg/g. At the same dosage, the adsorption capacities were 0.675 mg/g and 0.94 mg/g for 60 and 30 minutes of contact time, respectively. In contrast, the lowest adsorption capacity was observed at the highest adsorbent dosage (8 g) with 30 minutes of stirring, yielding 0.231 mg/g, while similar values of 0.236 mg/g were recorded at 60 minutes. Adsorption capacity (mg/g) reflects the ability of the fly ash adsorbent to bind Pb ions per unit mass of adsorbent. The results indicate an inverse relationship between adsorbent dosage and adsorption capacity, where increasing the adsorbent dose led to a decrease in adsorption capacity per gram. This phenomenon can be attributed to the incomplete utilization of active sites at higher dosages. As the adsorbent mass increases, particle aggregation and site overlapping may occur, reducing the effective surface area available for interaction with Pb ions (Istighfarini et al., 2017).

In general, adsorption capacity represents the maximum amount of adsorbate that can be retained by an adsorbent under specific operating conditions, including temperature, pH, and contact time. Therefore, although higher dosages may enhance overall removal efficiency, the adsorption capacity per unit mass tends to decrease due to reduced adsorption site utilization efficiency. The influence of contact time further indicates that adsorption proceeds rapidly at the initial stage due to a high concentration gradient and abundant vacant sites, followed by a slower phase as equilibrium is approached. The higher adsorption capacity observed at 90 minutes suggests that sufficient time is necessary for Pb ions to diffuse into internal pores, consistent with intra-particle diffusion mechanisms described in adsorption kinetic models.

4. Conclusions

Based on the research findings, the optimal operational conditions for Pb removal using CFA adsorbent were achieved at a dosage of 8 g with a stirring time of 90 minutes, resulting in the lowest residual Pb concentration of 0.012 mg/L. In contrast, the least effective condition was observed at a dosage of 2 g with 30 minutes of stirring, where the residual Pb concentration remained at 1.1 mg/L.

The highest removal efficiency (99.684%) was obtained at 8 g dosage and 90 minutes of contact time. However, the maximum adsorption capacity (0.945 mg/g) was achieved at a lower adsorbent dosage of 2 g with 90 minutes of stirring. This indicates that while higher adsorbent dosages enhance overall removal efficiency, lower dosages provide better adsorption capacity per unit mass due to more efficient utilization of active sites.

From a practical perspective, the use of 8 g CFA with 90 minutes of contact time is recommended for maximum Pb removal in wastewater treatment applications, particularly when meeting regulatory discharge standards is the primary objective. However, for cost-efficient operation and optimal material utilization, lower dosages may be considered depending on the required treatment target.

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