

The Ability of Sub-Bituminous Coal Activated with NaOH in Removal of Mercury: Equilibrium and Isotherm Study

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Abstract—Improved utilization strategies for Sub-Bituminous Coal (SC) can be made using alkaline materials to increase soil productivity and can also reduce metal concentrations in aqueous solution. This study aims to examine the characteristics of SC activated with NaOH (SC-NaOH) and the mechanism of Hg²⁺ adsorption by SC-NaOH through an adsorption isotherm model approach developed for experimental equilibrium. The SC-NaOH was characterized by proximate composition (29.59% moisture; 76.02% volatile matter; 72.02% ash and 4.02% fixed carbon) and chemical properties (pH 12.60; EC >2.00 dS/m; CEC 141.60 cmol(+)/kg; OC 14.38% C and total N 0.10% N). The adsorption of Hg²⁺ on SC-NaOH increased with increasing Hg concentration and decreasing pH. The adsorption capacity and coefficient of Hg²⁺ by SC-NaOH were 262.51 mg/g and 131.12 l/kg at pH 1.68 and Hg²⁺ concentration of 100 mg/l with a removal efficiency of 65.63%. Hg²⁺ adsorption isotherms occurred in Freundlich model ($1/n = 1.03$: cooperative adsorption and $R^2 = 1$) = Langmuir model ($R_L = 0.94$: favorable and $R^2 = 1$).

Keywords—activation, equilibrium, geochemical, isotherm, mercury, NaOH, sub-bituminous coal

I. INTRODUCTION

Mercury (Hg) is one type of highly toxic heavy metal that is not necessary for human beings and the life cycle of biodiversity. Hg can damage human health even in low amounts [1, 2]. Inorganic Hg is the type of mercury most commonly found in the environment [3]. Hg gradually accumulates in water and soil solutions. Under normal conditions, mercury rapidly methylates with chemicals and organisms, producing more harmful forms of Hg such as methylmercury or dimethylmercury [4]. This is the main basis for selecting stable materials for heavy metals removal in soil and water systems. Heavy metals, such as Hg, can damage water and soil systems. Therefore, the selection of the right adsorbent material is very important. Based on US EPA regulations, it explains that the Hg threshold limit allowed to enter treated wastewater is 10 g/l and drinking water is 2 g/l [5]. Meanwhile, the threshold limit for Hg contamination in soil is only allowed to be 1 mg/kg [6]. Therefore, a suitable technology is needed to reduce the impact of Hg pollution in the environment through the adsorption process with the application of amelioration technology.

Adsorption technology through amelioration is a powerful effort to reduce water and soil pollution. One potential adsorbent that can be used in soil and water amelioration technology is low-rank coal such as lignite [7] and sub-bituminous coal [8]. The utilization of low-rank coal

starts with its potential as a humic substance with a higher content than other types of organic materials such as peat and compost. One type of low-rank coal used is sub-bituminous coal. Development of strategies to optimize the utilization of sub-bituminous coal in amelioration technology as a collaboration between soil amendments, fertilizers, and adsorbents. Amelioration technology has so far only focused on soil improvement. However, on the other hand, it can also be used as an adsorbent in the removal of heavy metals such as Hg in this study. Amelioration technology is an appropriate technology that is very applicable in overcoming contamination and pollution and at the same time can be used as a soil amendment and fertilizer from sub-bituminous coal as a humic material has been used on marginal lands such as Inceptisols, Ultisols [9], and Oxisols [10] and polluted soils [11].

Chemical activation using alkaline materials such as NaOH and chemical fertilizers is also used to optimize the use of low-rank coal as a soil ameliorant. Based on the research that has been conducted, shows an increase in soil pH, CEC, and SOM. The NaOH reagent was determined as the best alkaline material in the activation process of Sub-bituminous coal as a soil improver. The SC-NaOH activation improved the chemical characteristics of SC, such as pH (5.34–12.65 units) and CEC [24.39–148.20 cmol(+)/kg]. The increase in the number of O-H, C=O, and CH₃ functional groups can improve the chemical properties of Ultisols such as pH, CEC, organic C, available P, and total N by 49 units, 28.08 cmol(+)/kg, 1.63% C, 2.37 ppm P, and 0.06% N, compared to the control [12]. This proves that the potential of coal activated with alkaline materials can increase nutrient availability and metal sorption.

Adsorption technology is a very effective method to reduce water and soil pollution. Therefore, it is necessary to identify the right adsorbent material to use. However, the type of adsorbent will have varying effects on adsorption. This study focuses on the use of sub-bituminous coal activated with 10% NaOH to adsorb Hg in an aqueous solution. This study aims to determine the mechanism of Hg adsorption that occurs through the adsorption isotherm model approach developed to explain various experimental equilibrium results.

II. MATERIALS AND METHODS

This research was conducted from March to August 2023 at the Soil Laboratory of the Faculty of Agriculture and the Environmental Chemistry Laboratory of the Faculty of

Mathematics and Natural Sciences, Andalas University, West Sumatra.

A. Activation of Sub-Bituminous Coal with NaOH and Its Analysis

Sub-bituminous coal (SC) was obtained from the Bonjol area of Pasaman, West Sumatra at a depth of 1-2 m from the ground (Fig. 1A). The coal was cleaned with running water and dried at room temperature (25 °C) for 7×24 h at Greenhouse, Agriculture Faculty UNAND. After drying, the coal was oven-dried for 1×24 hours at a temperature of 40–70 °C. The coal was pulverized with a Disc Mill model FFC 23 and sieved for 10 m with an EMS-8 Electromagnetic Sieve Shaker sieve of 500 µm size (Fig. 1B). The SC is weighed based on the dosage and percentage of the formulation used (sample weight). Activation of SC is carried out by dissolving 10% NaOH as the activating agent with H₂O based on the field capacity of SC, and the calculations are shown in Eq. (1).

$$NaOH (g) = \frac{10}{100} \times coal\ weight \quad (1)$$

Furthermore, the composition is stirred evenly, allowed to stand for 1×24 hours in a 250 mL beaker, and covered with aluminum foil with plastic wrapping. The results of SC-NaOH are dried using an oven at a temperature of 70 °C for 1×24 hours to homogenize the water content in the ameliorant.

Coal-activated analysis includes pH H₂O (pH-active), pH KCl (pH-potential), pH PZC [(2·pH KCl) – pH H₂O], and ΔpH (pH H₂O – pH KCl), electrical conductivity (EC) by the Electrometric method by pH meter HANNA HI 9813-16 portable, cation exchange capacity (CEC) by the leaching of NH₄OAc 1N pH 7 method, organic C by the Walkley-Black method and total N by Kjeldahl methods [13–15]. Analysis Scanning electron microscope-energy dispersive X-ray (SEM-EDX) merk: JSM-6510A/ JSM-6510LA at LPPT UGM test as an electron microscope that can produce images by scanning the surface of the specimen with a focused electron beam. which is focused on a certain magnification and scale. The electron microscope has a resolution of 0.1–0.2 nm. EDX is a technique to analyze elemental chemical elements of coal. Coal analysis is also carried out through X-ray diffraction (XRD) using Bruker D8 Advance with Davinci X-ray Diffractometer with Cu radiation source in the range of 200 to 900, with a step size of 0.020. X-Ray Fluorescence (XRF) brand: PANalytical Type Minipal 4 and Fourier transform infrared (FT-IR) spectra were measured in the range of 600 to 4000 cm⁻¹ using an ABB MB-3000 Series FTIR Spectrometer instrument with diamond Internal Reflection Element (IRE) in absorbance mode. XRD, XRF, and FT-IR analyses were carried out at LPPT UNP.

The characteristics of SC-NaOH have the atomic composition of C (27.58%), O (52.61%), Si (10.48%), Al (4.98%), Na (2.40%), K (0.62%), Fe (0.44%), Mg (0.37%), Cu (0.28%), Ca (0.13%) and Ti (0.10%) with the mineral content of quartz (SiO₂); montmorillonite (Al₂Ca_{0.3}H₄O₁₃Si₄); muscovite (Al₃H₂KO₁₂Si₃) and illite (Al₂H₂₄K₄Mg₃O₆₈Si₂₄) and oxide composition [SiO₂ (64.38%); Al₂O₃ (22.64%); Fe₂O₃ (5.14%); K₂O (2.89%); CaO (1.87%); P₂O₅ (1.66%); ZnO; MnO and CuO (≤0.02%)] as well as O-H and N-H

functional groups; C-H≡C-H; C=C-H; C=O, C-H and C=C-H and minerals. SC-NaOH has a variety of functional groups, most of which are negatively charged, such as hydroxyl and carboxyl groups and also porous, with many surface gaps and binding sites for Hg ions

B. Determination of HgCl₂ Concentration

The Hg²⁺ concentration solution was dissolved from HgCl₂ reagent material in 0.1 M HCl solution with a set concentration level of 1, 10, and 100 mg/l. Determination of concentration by dissolving 0.13 g of HgCl₂ in 1000 mL of 0.1 M HCl, while the concentrations of 1 and 10 mg/l Hg²⁺ were made by dissolving the parent solution of 100 mg/l Hg²⁺ concentration [16].

C. Adsorption of Hg by Batch Equilibrium Method

Adsorption of Hg focused on isothermal adsorption with the Batch equilibrium method. Adsorbent was weighed 0.5 g of SC-NaOH and mixed with 20 ml of 1, 10, 100 mg/l concentration solution Hg in a 25 ml glass cylinder tube. Isothermal adsorption was carried out at a contact time of 1×24 hours using a rotary shaker at a speed of 300 rpm and a temperature of 25 °C in a Laminar flow chamber. Determination of adsorption pH was carried out after the contact time carried out at each concentration. Equilibrium filtrate concentrations of Hg were measured using CV-AAS [13]. The adsorbed Hg was calculated by Eqs. (2), (3), and (4) [17].

$$\%R = \frac{(Co-Ce)}{Ce} \times 100 \quad (2)$$

$$Qe (mg/g) = \frac{(Co-Ce)}{m} \times V \quad (3)$$

$$Kd (l/kg) = \frac{Ce}{Qe} \times 1000 \quad (4)$$

where:

Co (mg/l) = the initial concentration of the adsorbate; Ce (mg/l) = Equilibrium concentration; R (%) = removal efficiency; Qe (mg/g) = adsorption capacity; m (g) is the biochar's mass, and V (l) is the quantity solution of Hg; R = adsorption efficiency (%) and K_d = adsorption coefficient (l/kg).

D. Adsorption Isotherm Model

The adsorption isotherm models used consisted of one-parameter (Henry) and two-parameter (Freundlich and Langmuir) isotherm models. The adsorption isotherm models used linear models (Eqs. (5), (6), and (7)) [18].

Henry non-linear model:

$$Qe = K_{He}Ce \quad (5)$$

Freundlich non-linear model:

$$Qe = K_F Ce^{1/n} \quad (6)$$

Langmuir non-linear model:

$$Qe = \frac{Q_m K_L Ce}{1 + K_L Ce} \quad (7)$$

where:

Ce (mg/l) = Equilibrium concentration; R (%) = removal efficiency; Qe (mg/g) = adsorption capacity; Q_m (mg/g) = maximum adsorption capacity; K_{H_e/F/L} = Henry, Freundlich and Langmuir isotherm constant, and n = intensity constant of adsorption.

E. Experimental Data Modeling

The Batch Equilibrium Method was used to explain the mechanism in determining the applicability and stability of the adsorption process. Microsoft Excel 2016, SPSS 16, and OriginLab were used to process the data.

III. RESULT AND DISCUSSION

A. Morphology and Characteristics of Sub-Bituminous Coal Activated with NaOH

The morphology of SC (Fig. 1A) and SC-NaOH (Fig. 1B) shows a change in the color of NaOH-activated sub-bituminous coal powder to be darker, if identified with Munsell's soil color chart book is 5 Y 5/1 (gray) to 5 Y 4/3 (dark).

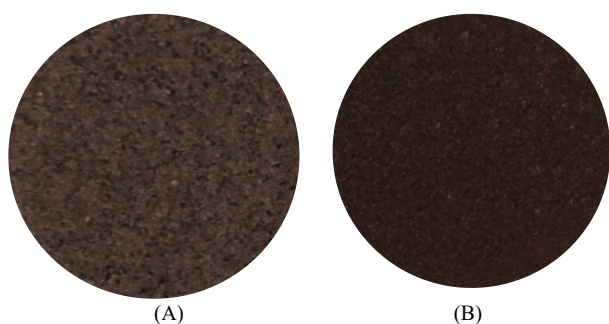


Fig. 1. Morphology of sub-bituminous coal (A) and activated with 10% NaOH (B) on passing 500 μm sieve.

Discoloration of coal after being reacted with NaOH is thought to be the decomposition of organic components such as lignin and humus which can cause decomposition or changes in the chemical structure of these organic components. This color change can be better understood through chemical and spectroscopic analysis to identify the compounds formed during the reaction between coal and NaOH. In addition, other factors that can affect this color change such as temperature, reaction time, and solution concentration can also affect the final result of the reaction [19].

1) Scanning Electron Microscope—Energy Dispersive X-Ray (SEM-EDX) analysis

Morphology The size and shape of sub-micron particles can be determined using SEM [20]. Fig. 2A shows the size and shape of the SC-NaOH particle morphology analyzed by SEM at 5000 times magnification. The results of SEM analysis show that the SC-NaOH particles are mostly wedge-shaped and aggregated with varying sizes and uneven numbers of particles. This difference arises from the decomposition process of chemical components in coal. NaOH pretreatment can improve the solubility of coal by finding an increase in the solubility of organic carbon and the release of low molecular weight aromatic fragments [21].

The EDX analysis can also identify the atomic composition contained in the material. The atomic composition of SC-NaOH particles is C (27.58%), O

(52.61%), Si (10.48%), Al (4.98%), Na (2.40%), K (0.62%), Fe (0.44%), Mg (0.37%), Cu (0.28%), and Ca (0.13%) (Fig. 2B). The process of treating coal with NaOH can result in complex changes to the composition and structure of the coal. Both a decrease and an increase in the number of certain atoms can occur depending on the treatment conditions, the initial nature of the coal, and the reaction mechanism that occurs. NaOH can contribute to changes in the carbon structure of coal due to the formation of new functional groups or modification of the aromatic structure [22].

The NaOH can form new oxygen groups on coal, such as hydroxyl (OH) groups or carboxyl (COOH) groups, leading to an increase in the number of oxygen atoms on the coal surface and NaOH can also be adsorbed on the coal surface, causing an increase in the number of sodium atoms in coal as well as interactions with other metals present in coal, causing an increase in the number of metal atoms [23, 24]. This is evident in the increase of O and Na atoms, 11.97% and 2.40% respectively, as well as the presence of metals such as Fe, Cu, and Ti. NaOH can also react with hydroxyl (OH) groups in coal or remodel the hydrocarbon structure, resulting in a decrease in hydrogen and nitrogen atoms in the form of soluble compounds. Meanwhile, NaOH can cause sulfur removal in the form of soluble compounds carried by the NaOH solution [25]. The N atom is no longer found in the atomic component of SC-NaOH, where previously the N atom in SC was 11.96%.

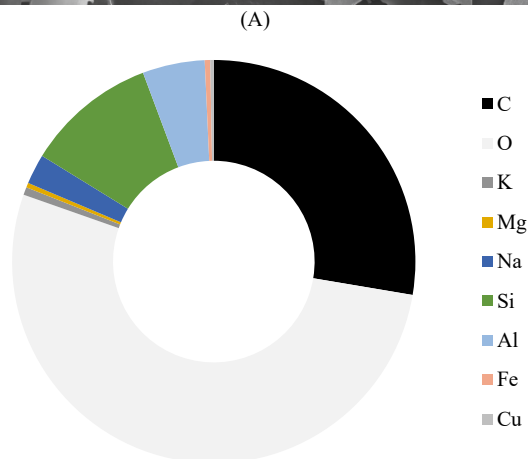
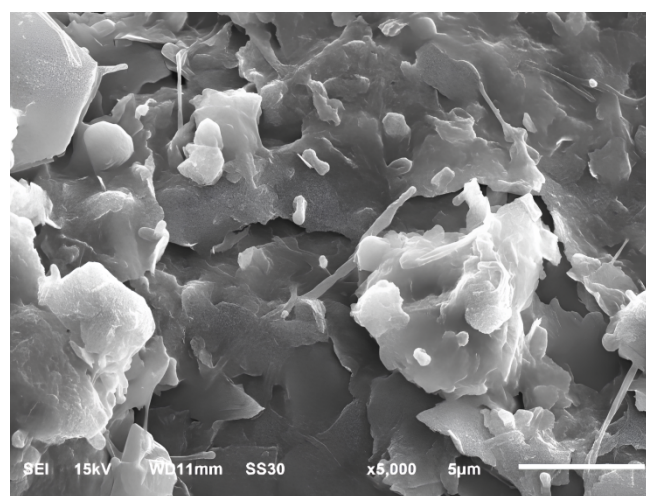


Fig. 2. (A) SEM at ×5000 magnification, and (B) EDX of sub-bituminous coal activated with 10% NaOH.

2) Fourier Transform Infrared (FT-IR), X-Ray Diffraction (XRD), and Fluorescence (XRF) analysis

The FT-IR analysis was performed to detect mineral components and monitor the changes in functional groups caused by SC-NaOH. The FTIR spectra obtained for coal at all peaks ranged from 4000-600/cm and contained 0-100% transmittance (Fig. 3A). The results of the FTIR spectrum of SC-NaOH have several main absorption band peaks, namely at 3619.59 and 3375.44/cm which characterize O-H and N-H bonds, 2916.59 and 2850.47/cm which characterize C-H stretching $-C\equiv C-H$ bonds; $C=C-H$.

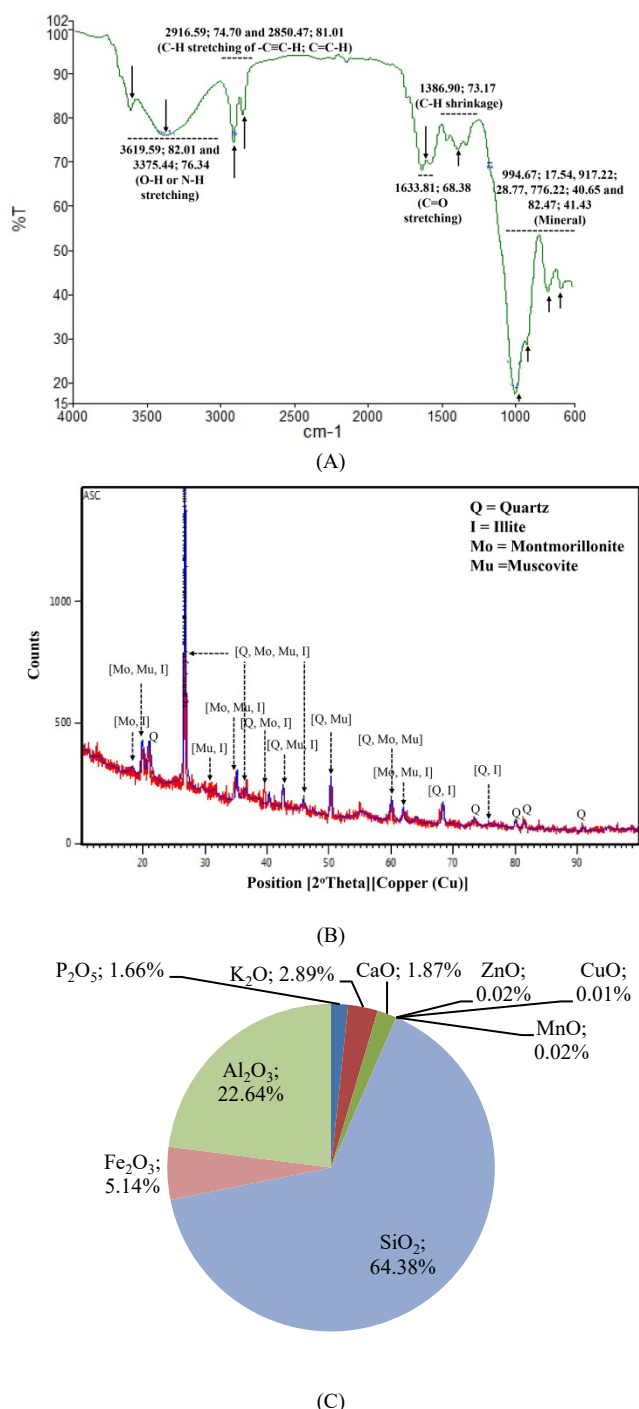


Fig. 3. XRD (A); FT-IR (B) spectrum and oxide composition by XRF (C) of sub-bituminous coal activated with 10% NaOH.

The absorption at 1633.81/cm characterizes C=O bonds, at wave numbers 994.67; 917.22; 776.22, and 682.47/cm there are C=C-H bonds and mineral bonds. The difference that

occurs in SC after activation with NaOH is the appearance of a new absorption at 1386.90/cm (C-H bond). Changes in functional groups in SC activated with NaOH are not much. In the range of $> 3600/cm$, high-frequency OH- vibrations occur. The degree of substitution of Al for Si in dioctahedral silicates is lower than in trioctahedral silicates. Absorption occurs at 413–529/cm due to perpendicular Si-O vibrations. Si-O-Si stretching produces two bands at 1594 and 2174/cm, while Si-O bending vibrations produce considerable absorption at 413 and 529/cm. In the presence of quartz in the coal sample, FT-IR spectra with absorption frequencies at 746 and 684/cm can be generated [26].

The chemical activation process can cause depolymerization, which is the breaking of long bonds in the coal molecular chain. This molecular weight reduction can cause the appearance of new absorption at certain wavelengths, such as at the -C-H bond. The reaction of coal with NaOH can cause dehydration and the formation of double bonds in the coal hydrocarbon chain. NaOH can react with functional groups present in coal, such as -OH and -COOH groups. This process can lead to the restructuring and formation of new bonds and cause changes in the methyl and methylene groups in coal.

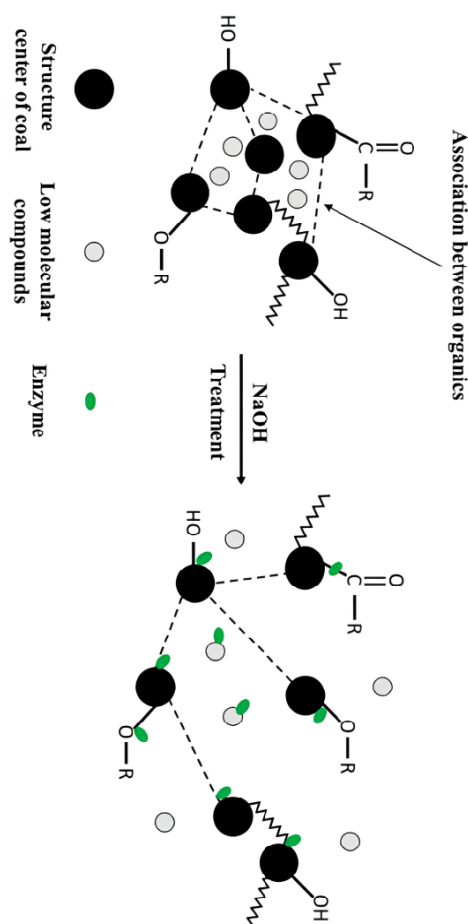


Fig. 4. Prediction of schematic from chemical structure degradation of sub-bituminous coal before and after depolymerization with alkaline compounds (e.g. NaOH) [21].

The XRD analysis shows that the minerals in SC-NaOH consist of quartz (SiO₂); montmorillonite (Al₂Ca_{0.3}H₄O₁₃Si₄); muscovite (Al₃H₂KO₁₂Si₃) and illite (Al₂H₂₄K₄Mg₃O₆₈Si₂₄) (Fig. 3B). Chemical activation of coal with NaOH can result in mineral alteration. NaOH is thought to cause stripping of

mineral layers and the release of metal ions, such as Al and Ca, into solution. This process may increase the porosity and surface liveliness of the coal. The reaction of coal with NaOH can cause the removal of K and Al ions and can also cause the removal of these ions, as well as affect the mineral structure and surface liveliness through changes in the molecular structure of the coal, especially the organic part [27, 28].

The oxide composition of SC-NaOH is SiO₂ (64.38%); Al₂O₃ (22.64%); Fe₂O₃ (5.14%); K₂O (2.89%); CaO (1.87%); P₂O₅ (1.66%); ZnO; MnO and CuO ($\leq 0.02\%$) (Fig. 3C). NaOH can cause changes in the oxide composition of coal. The reaction between NaOH and coal can result in the formation of certain oxides as well as changing the form of existing oxides. Chemical activation can lead to increased formation of carbon oxides, such as carbonates and bicarbonates. The organic fraction of coal and the resulting carbon oxides can contribute to an increase in pH in coal. This is evident from the increase in pH of SC-NaOH by 12.60 units. NaOH can cause the release of ions such as silicon, aluminum, and calcium into solution and affect the shape and solubility of iron oxides in coal [29].

The appearance of clusters and the intensity of absorption in each characteristic band of SC are the basis for the ability of SC as an adsorbent. SC has a macromolecular structure showing a highly aromatic character with a high carbon and low hydrogen content, in which aromatic groups with 2 to 5 condensation rings are cross-linked mainly by strong methylene and ether bridges. This transformation of lignite to SC suggests that a structural model for the development of

lignite to SC during the petrification process, which contains many fundamental processes, is depicted in Fig. 4. The main structural component of SC is the lignin precursor that loses hydroxyl and methoxyl groups sequentially through demethylation and dehydroxylation processes [30].

In addition to the shift in absorption bands and an increase in absorption intensity in each characteristic band from the activation process with NaOH. This indicates an increase in these groups and is an indication that there has been depolymerization (polymer disconnection) from the degradation of SC molecules to become more open and form more negative charges (Fig. 4). This is consistent with the increase in pH and CEC (Table 1), it appears that NaOH can activate SC to become more active in increasing negative charges. The disconnection of the bonding network in coal due to alkaline compounds begins with the disconnection of the aliphatic bridge which is the weakest bond. Furthermore, ether bridges have similar bond energy, so the disconnection of the bond network in coal is manifested by a transition into a plastic state, resulting in the formation of highly reactive bonds. The breaking of bonds between polyaromatic rings in the coal structure during the pyrolysis process with alkaline materials will cause the coal to become more reactive through ionic reactions at low temperatures. It also reduces van der Waals forces between coal molecules and cross-linking reactions, which explains the presence of water in coal. The breaking of coal molecules with calcium also produces anions in the coal that form organomineral bonds [31].

Table 1. Proximate and chemical characteristics of sub-bituminous coal activated with 10% NaOH

No	Analysis	Unit	Mean	SE	SD
1	Moisture	%	29.59	0.62	1.08
	Volatile Matter		76.02	0.62	1.08
	Ash		72.02	0.62	1.08
	Fixed Carbon		4.02	0.62	1.08
2	H ₂ O	unit	12.60	0.06	0.10
	KCl 1N		9.43	0.03	0.06
	PZC		6.27	0.09	0.15
3	Electrical Conductivity	dS/m	2.00	0.00	0.00
4	CEC	cmol(+)/kg	141.60	2.95	5.11
5	OC	%	14.38	0.15	0.26
6	Total N	%	0.10	0.01	0.02

Remarks: PZC = point of zero charge; CEC = Cation exchange capacity; OC = Organic C; SE = Standart error; SD = Standart deviation; n = 3 sample.

The activation process with alkaline compounds also forms complex bonds with organic compounds such as lignin, these bonds are thought to break the molecular bonds in lignin, thus forming new molecules and the formation of oxide bonds. This indicates that the formation of oxide bonds affects the reactivity in SC, it is suspected that these bonds will undergo neutralization and turn acid ions into more reactive because they are neutral. During the activation process, the carboxylic groups on the coal undergo CO₂ liberation and deposit inorganic species on the coal surface, resulting in carboxylate stabilization. The level of activity of alkaline compounds added to coal will depend on the alkalinity of the compound. Other factors that can affect the activity such as: (1) The method used to add the alkaline compound to the coal; (2) The nature of the anion bound to the alkali metal and (3) how much dispersion the alkaline compound has on the coal [31].

3) Proximate and chemical characteristics of sub-bituminous coal activated with NaOH

The proximate and chemical characteristics of SC-NaOH were analyzed (Table 1). The moisture and ash contents were 29.59% and 72.02%, respectively. Moisture has an impact on calorific value and the concentration of other elements, the relatively low moisture content of the coal sample indicates a considerable improvement in coal quality. Low ash concentration improves coke quality, therefore, it is an important criterion for coke production [32]. However, there was an increase in moisture and ash content after activation with NaOH. Chemical activation can increase the ability of coal to absorb water from the surrounding environment. Changes in the surface structure of coal make it more reactive to water and can also change the mineral composition in coal or lead to higher ash formation or deposition after the activation process [33].

The SC-NaOH has 76.02% volatile matter and 4.02%

fixed carbon. Apart from its use in coal ranking, volatile matter is one of the most important characteristics used in identifying the right application. Fixed carbon is the part of the coal that remains as a residue, but consists mostly of carbon, although it also contains small amounts of H, O, N, and S that are not carried away by the gas [34]. However, there is a decrease in volatile matter and fixed carbon of coal after activation with NaOH. Chemical activation can cause depolymerization or the breaking of long bonds in the coal molecular chain through the release of volatile components such as gas and steam from coal, which can result in a decrease in volatile matter and the release of carbon in the form of gas or steam can also reduce the fixed carbon content of coal [35].

The chemical characteristics of SC-NaOH based on pH H₂O; KCl 1N and PZC are 12.60; 9.43 and 6.27 units, respectively. This is due to the high organic acid content in SC-NaOH and is also influenced by various chemical components including sulfur, as well as interactions between solutions (H₂O and KCl 1N). During the activation process, NaOH reacts with the acidic components contained in the coal, producing alkaline compounds. Meanwhile, the PZC pH for coal ranges from 4 to 7.5 units. However, this value depends on the chemical composition of the coal used [36]. PZC can depend on the properties of the coal surface that can change during the activation process. A higher PZC indicates that the coal will tend to be alkaline under these conditions.

The SC-NaOH has an EC of 2.00 dS/m. NaOH, as a strong base, can remodel the coal structure and change the functional groups present on the coal surface. Increasing the ionic conductivity in coal can improve its ability to conduct electricity and the formation of more conductive functional groups such as pyrolytic carbon functional groups as well as NaOH can reduce the surface resistance of coal, making it more conductive. The EC value of coal usually varies from 10⁻³–10⁻⁸ S/m. Coal has a low EC and is a poor conductor. The EC value of coal is largely determined by its mineral composition (such as Si, Al, Fe, S, etc.) and non-carbonate components. This is because coal is a fossil fuel that is often used to generate heat, not electricity. Coal EC is not considered a relevant substance in the context of EC [37].

The CEC of SC-NaOH was 141.60 cmol(+)/kg. The increase in CEC after activation with NaOH may be due to

changes in the structure and nature of the coal surface, where hydroxide ions (OH⁻) from NaOH can react with various functional groups on the coal surface, including carboxylic acid groups and phenol groups. These changes can affect the coal's ability to exchange ions in solution. In general, the aromatic structure of SC is thought to be the site of ion exchange. In coal, there is very complex cation adsorption consisting of organic and inorganic compounds, so this is closely related to the CEC of coal as well as the presence of hydrogen ions in the aromatic structure of coal consisting of carbon bonds which are permanent ion exchange sites [38].

The organic C and total N contents of SC-NaOH were 14.38% and 0.10%, respectively. NaOH can remodel the coal structure and form new functional groups bound to carbon. These can include organic functional groups such as carboxylic acid groups, phenols, or others. The addition of hydroxyl (OH) groups to the coal structure, which can increase the organic carbon content, can also reduce the more complex structures in the coal and produce simpler compounds with organic functional groups. In general, SC has a low organic carbon content, usually ranging from 35 to 45% [39]. Although coal also contains small amounts of N in various compounds such as NH₃, organic nitrogen, and inorganic nitrogen, the N concentration in coal generally varies between 1-2.5% N [40]. The decrease in N after activation by 0.06% is because NaOH can react with nitrogen compounds, especially if there are amine groups (NH₂) in the coal, forming ammonia (NH₃). This process can cause nitrogen to escape from the coal in the form of ammonia gas.

B. Equilibrium and Isotherm Study

1) Adsorption of Hg²⁺ with sub-bituminous coal activated with NaOH

Adsorption of Hg in aqueous solution with SC-NaOH had a significant effect on the adsorption equilibrium concentration, which increased as the adsorption capacity of Hg increased. However, as the Hg concentration increased, the adsorption pH decreased (Table 2). At pH 1.68, the largest adsorption capacity was 262.51 mg/g at a concentration of 100 mg/l Hg²⁺. The adsorption process in acidic solution using 0.1 M HCl at different Hg concentration levels of 1, 10, and 100 mg/l, where the pH of the pure solution was 1.33, 1.32, and 1.30, respectively.

Table 2. Adsorption of Hg²⁺ at equilibrium by sub-bituminous coal activated with 10% NaOH

Co	Ce	pH solution Hg ²⁺	R	Qe	Kd	pH after adsorption
mg/l		unit	%	mg/g	l/kg	unit
1	0.31 c	1.33 a	68.67	2.75 c	114.41	1.71
10	3.29 b	1.32 ab	67.07	26.83 b	123.07	1.70
100	34.37 a	1.30 b	65.63	262.51 a	131.12	1.68
CV	8.00	0.84	3.33	4.16	10.04	0.8
Duncan's Test	**	*	ns	**	ns	ns
SE	0.83	0.01	1.82	3.31	10.07	0.01

Remarks: Co = the initial concentration of the adsorbate; Ce = Equilibrium concentration; R = removal efficiency; Qe = adsorption capacity; K_d = Coefficient adsorption; CV = Coefficient of variation; SE = Standard error; ** = significant at the 0.01 level; * = significant at the 0.05 level; ns = non-significant and n = 9 sample.

At increasing Hg²⁺ concentrations, the effectiveness of Hg removal decreased from 68.67% to 65.63% (Table 2). The higher the Hg injection concentration, the less effective the adsorbent is in adsorbing Hg. Adsorption efficiency increases at low concentrations because more adsorption sites on the adsorbent surface are available for Hg, whereas, at high

concentrations, the adsorption sites on the adsorbent become saturated and no longer have a place for adsorption, as well as because of the potential competition between Hg for the same adsorption sites on the adsorbent. The mass of adsorbent used, on the other hand, maintains a dynamic equilibrium as the concentration increases [41]. At the time of equilibrium, the

adsorbent has the greatest capacity to adsorb Hg ions. The peak capacity of the adsorbent in adsorbing Hg ions occurs at the equilibrium time, which affects the capacity and sorption coefficient of Hg. At a concentration of 100 mg/l, the sorption coefficient of Hg with SC-NaOH was 131.12 l/kg (Table 2).

The increase in Hg concentration is directly proportional to the adsorption coefficient. The adsorption coefficient can explain why a chemical attracts or adsorbs ions from a solution on the surface of a particular material [42]. More molecules or particles are expected to interact and adsorb on the adsorbent surface as the Hg concentration increases. Adsorption, on the other hand, may reach a saturation point at a certain concentration when all adsorption sites are occupied, and increasing the concentration no longer increases the adsorption coefficient. The pH value is very important during the adsorption process. It affects the adsorbent value and surface charge, as well as the speciation form of Hg in solution. At an acidic pH of 1.68, adsorption of Hg with SC-NaOH in a 0.1 M HCl solution occurred. Adsorption can fail at high pH levels due to competition for sorption sites in solution between H_3O^+ ions and Hg^{2+} cations, as well as a decrease in accessible surface area [43]. The adsorption capacity of Hg by SC-NaOH in 0.1 M HCl solution, on the other hand, was in the range of 2.75; 26.83; and 262.51 mg/g. At acidic pH values, the adsorption process remained effective.

2) Adsorption isotherm model of Hg^{2+} with sub-bituminous coal activated with NaOH

Isotherm models were used in Hg adsorption to assess the optimal adsorption process using high R^2 values [44]. Adsorption of Hg with SC-NaOH showed that the R^2 values of the linear isotherm models were all the same, Langmuir = Freundlich = Henry ($R^2 = 1$), or identical. This means that any isotherm model can be used. The Henry isotherm model describes the precision of the Hg adsorption process at different concentration levels. The partial pressure at a surface is related to the concentration (C_e). This model separates all molecules from their environment at the surface [45]. The equilibrium concentration of Hg in the adsorbed phase was calculated using Henry's linear equation (Fig. 5). The linear plot of the Henry isotherm model has the equation $y = 0.1314x - 0.1355$; $R^2 = 1$ (Table 3 and Fig. 2). This form explains why adsorbate-adsorbate interactions are stronger than adsorbate-adsorbent interactions.

Reversible adsorption processes are described by the Freundlich isotherm model. The Freundlich model, which

can be applied to multilayer adsorption, is also not limited to monolayer formation. The Freundlich isotherm model has two expressions: surface heterogeneity and the exponential distribution of active sites [45]. The ratio of Hg to solute varies with solution concentration, and the adsorbed amount indicates the total adsorption on each site. This shows that after the adsorption process is completed, Hg is bound to stronger binding sites, and the adsorption capacity decreases rapidly. In the heterogeneous system, the Freundlich isotherm model for Hg adsorption with SC-NaOH was applied. The linear equation of the Freundlich model is $y = 1.0302x - 0.9554$; $R^2 = 1$ (Fig. 6 and Table 3).

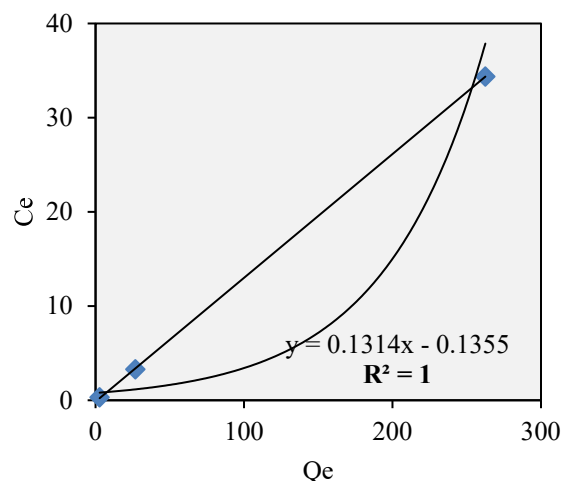


Fig. 5. Linear plot of Henry's isotherm model.

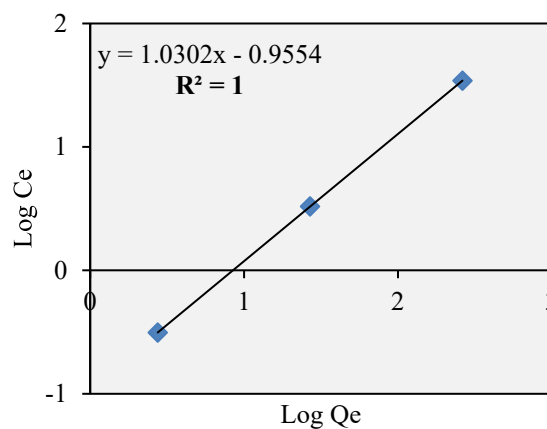


Fig. 6. Linear plot of Freundlich isotherm model.

Table 3. Isotherms adsorption of Hg with SC-NaOH

Linear Model	Isotherm parameters			
	n	K_F (mg/g) (l/mg) ⁿ	1/n	Linear Equation
Freundlich	0.97	9.02	1.03	$y = 1.0302x - 0.9554$; $R^2 = 1$
	Q_m (mg/g)	K_L (l/mg)	* R_L	Linear Equation
Langmuir	71.43	0.002	0.94	$y = 8.8016x - 0.014$; $R^2 = 1$

Remarks: Freundlich [$n = 1/b$; $K_F = 10^a$] and Langmuir [$Q_m = 1/a$; $K_L = a/b$]; * $R_L = 1/[1+(K_L \cdot C_0)]$

The n value indicates the type of isotherm where the K_F and n values fluctuate with temperature. Adsorption of Hg with SC-NaOH has n and K_F values of 0.97 and 9.02, respectively. Adsorption is favorable when 1/n is greater than

zero ($0 < 1/n < 1$); unfavorable when 1/n is greater than one; and irreversible when 1/n = 1 [44]. The 1/n ratio for Hg adsorption with SC-NaOH is 1.03, which indicates unfavorable adsorption or cooperative adsorption. A 1/n

value greater than one implies a cooperative adsorption phenomenon [46, 47]. The Freundlich isotherm model equation is empirical in the presence of surface heterogeneity, with adsorption energy distributed across the surface topography. This means that all surface sites and adsorption energies are in the same patch. The interaction between SC-NaOH and Hg generates adsorption energy.

The Langmuir isotherm model of adsorption on SC-NaOH is kinetic, with a contact time of 24 hours. The model also describes continuous intermolecular interactions at the surface with a zero accumulation rate [45]. The Langmuir isotherm model attempts to evaluate and compare the adsorption of Hg with SC-NaOH as a function of changes in surface chemistry and the structural geometry of SC-NaOH.

The Langmuir model explains the accumulation of layers in a single molecular layer (monolayer adsorption) as well as adsorption processes in similar and different environments. The Langmuir isotherm model also emphasizes homogeneous adsorption, with each Hg molecule covering a wide range of activation energies and enthalpy constants. There should be no movement of Hg within the development field, and each spot should have the same affinity for Hg with the increase in distance, the molecular properties will decrease rapidly. Table 3 and Fig. 7 show the linear equation of the Langmuir model with adsorption of Hg with SC-NaOH, linear equation $y = 8.8016x - 0.014$; $R^2 = 1$.

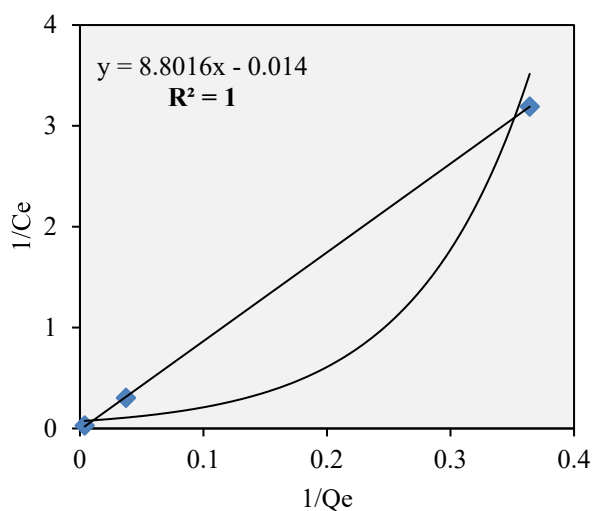


Fig. 7. Linear plot of Langmuir isotherm model.

The Langmuir model also defines the separation factor (R_L) as a dimensionless constant represented as a variation of the area and porosity of the relevant adsorbent. It also shows that a higher surface area and pore volume can increase the adsorption capacity. Based on the separation factor, adsorption is characterized as linear ($R_L = 1$), irreversible ($R_L = 0$), unfavorable ($R_L > 1$), or favorable ($0 < R_L < 1$) [44]. The R_L value for Hg adsorption with SC-NaOH was 0.94. This indicates that the adsorption of SC-NaOH on Hg is favorable. The Langmuir model assumes a homogeneous adsorbent surface with equal adsorption energy at all points. In contrast, the model and its coefficients were used to examine the distribution of contaminants in water and soil. To explain the variance of the adsorption surface and the site energy distribution (SED) presented, the Langmuir equation can only compare different adsorbents. Adsorbents explain the

adsorption site area distribution as evidence of isothermal variation.

3) Prediction mechanism of Hg^{2+} adsorption with sub-bituminous coal activated with NaOH

The adsorption process deposits a layer of adsorbate (Hg ions) on the surface of SC-NaOH. Adsorption of Hg on solid SC-NaOH involves three main steps: (1) transport of Hg from aqueous solution to the SC-NaOH surface; (2) adsorption onto the solid surface; and (3) transit within the SC-NaOH particles. Electrostatic attraction causes charged pollutants to adsorb on differentially charged adsorbents because heavy metals have a high affinity for hydroxyl (OH^-) or other surface functional groups [48]. When the adherence of the adsorbate to the adsorbent surface is non-specific, physical adsorption occurs. Chemisorption occurs when chemical bonds combine to form a strong force of attraction. Adsorption of Hg on SC-NaOH can occur through physical adsorption, chemisorption, electrostatic interaction, simple diffusion, intra-particle diffusion, hydrogen bonding, redox interaction, complexation, ion exchange, precipitation, and pore adsorption. Fig. 8 illustrates some potential pathways for Hg adsorption in SC-NaOH.

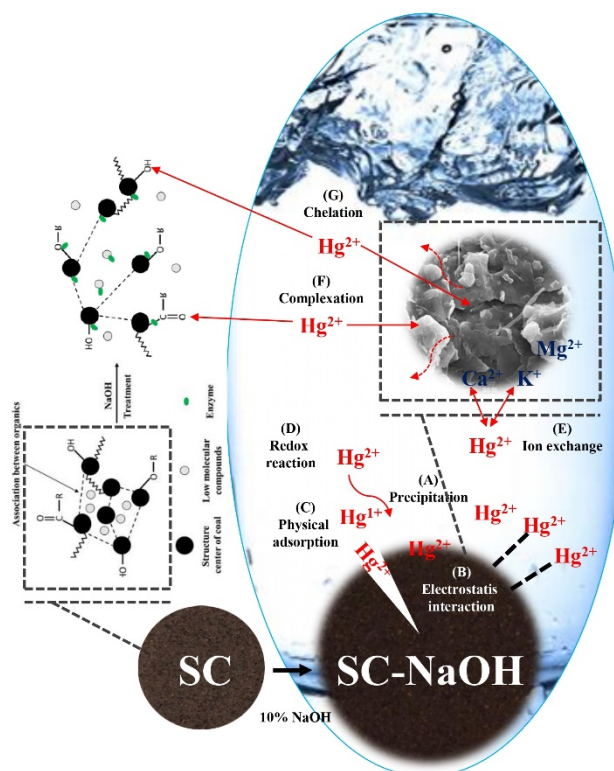


Fig. 8. Prediction mechanism of Hg adsorption with SC-NaOH.

The presence of these pores and gaps increases the surface area available for adsorption. SC-NaOH provides more possibilities for Hg ions to interact and be retained. This porous nature offers a high capacity for ion adsorption and is useful in soil and water system management applications. Adsorption of Hg by SC-NaOH involves (a) dispersion of Hg molecules, (b) electrostatic attraction to the surface, (c) electrostatic attraction between positively charged Hg ions, and (d) electrostatic attraction between negatively charged SC-NaOH functional groups. This effectively increases the adsorption capacity. The adsorption of Hg on the SC-NaOH surface can be affected by attraction forces such as van der

Waal and hydrogen bonding. The two main adsorption mechanisms are complexation and chelation. In general, complexation is the interaction of two or more metals, while chelation is a type of complexation that results in ring formation [49]. Hg plays an important role in the complexation process, is surrounded by ligands, and forms a mononuclear complex. Polynuclear complexes are formed when two or more Hg atoms are bound together at a central location by a ligand. Polydentate ligands can also be used in chelation to help form stable structures through different linkages.

IV. CONCLUSION

The characteristics of sub-bituminous coal activated with 10% NaOH have the atomic composition of C (27.58%), O (52.61%), Si (10.48%), Al (4.98%), Na (2.40%), K (0.62%), Fe (0.44%), Mg (0.37%), Cu (0.28%), Ca (0.13%) and Ti (0.10%), as well as O-H and N-H functional groups; C-H≡C-H; C=C-H; C=O, C-H and C=C-H and minerals of quartz (SiO₂); montmorillonite (Al₂Ca_{0.3}H₄O₁₃Si₄); muscovite (Al₃H₂KO₁₂Si₃) and illite (Al₂H₂₄K₄Mg₃O₆₈Si₂₄) and oxide composition [SiO₂ (64.38%); Al₂O₃ (22.64%); Fe₂O₃ (5.14%); K₂O (2.89%); CaO (1.87%); P₂O₅ (1.66%); ZnO; MnO and CuO (≤0.02%)]. The SC-NaOH was characterized by proximate composition (29.59% moisture; 76.02% volatile matter; 72.02% ash and 4.02% fixed carbon) and chemical properties [pH 12.60; EC >2.00 dS/m; CEC by 141.60 cmol(+)/kg; OC by 14.38% C and total N by 0.10% N]. The adsorption of Hg²⁺ on SC-NaOH increased with increasing Hg concentration and decreasing pH. The adsorption capacity and coefficient of Hg²⁺ by SC-NaOH were 262.51 mg/g and 131.12 l/kg at pH 1.68 and Hg²⁺ concentration of 100 mg/l with a removal efficiency of 65.63%. Hg²⁺ adsorption isotherms occurred in Freundlich model (1/n = 1.03: cooperative adsorption and linear equation: y = 1.0302x - 0.9554; R² = 1) = Langmuir model (R_L = 0.94: favorable and linear equation: y = 8.8016x - 0.014; R² = 1). The development of amelioration technology based on sub-bituminous coal activated with NaOH has been proven to be used in the removal of Hg in solution, so it has great potential in its wide application in overcoming Hg contamination and pollution in the community.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

Amsar Maulana conducted the entire research, data analysis, and writing process. Prof. Herviyanti, Dr. Teguh Budi Prasetyo, and Dr. Mimien Harianti facilitated the research and approved the final revision of the paper.

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