Efektivitas Kombinasi Koagulasi-Flokulasi dan Filtrasi-Adsorpsi Menggunakan Surfaktan Anionik Terhadap Penyisihan COD Pada Air Sumur yang Tercemar

The Effectiveness of Coagulation-Flocculation and Filtration-Adsorption Combination Using Anionic Surfactant for COD Removal on Contaminated Well Water

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ABSTRAK

Kualitas air sumur yang tercemar hidrokarbon umumnya ditandai dengan tingginya nilai COD dan kondisi pH yang asam sehingga tidak memenuhi standar baku mutu air bersih. Penelitian ini bertujuan untuk mengevaluasi efektivitas kombinasi surfaktan anionik Linear Alkylbenzen Sulfonat, peran koagulan ferri klorida dan kapur pada proses koagulasi dan flokulasi, serta proses filtrasi-adsorpsi dalam meningkatkan kualitas air. Eksperimen dilakukan dengan variasi konsentrasi LAS (200–1000 ppm) melalui tahapan *pre-treatment*, koagulasi-flokulasi, dan filtrasi menggunakan media pasir silika, mangan, dan karbon aktif. Hasil penelitian menunjukkan bahwa penambahan surfaktan LAS berperan dalam meningkatkan pH menuju kondisi netral. Nilai pH optimum diperoleh pada konsentrasi surfaktan 400 ppm dengan pH 7. Kombinasi proses koagulasi-flokulasi dan filtrasi-adsorpsi mampu menurunkan COD secara signifikan. Efisiensi penurunan COD tertinggi diperoleh pada perlakuan Variasi 2 dengan filtrasi-adsorpsi sebesar 85,3% pada konsentrasi 400 ppm, sehingga memenuhi baku mutu (<10 mg/L). Namun, peningkatan konsentrasi LAS yang berlebih menurunkan efisiensi pengolahan akibat kejenuhan adsorben dan pembentukan *micelle*. Secara keseluruhan, metode kombinasi ini terbukti efektif dan aplikatif untuk pengolahan air sumur tercemar hidrokarbon.

Kata kunci: Surfaktan anionik, Koagulasi, Flokulasi, Filtrasi, Adsorpsi

ABSTRACT

Well water contaminated with hydrocarbons is generally characterized by high COD levels and acidic pH conditions, making it fail to meet clean water quality standards. This study aims to evaluate the effectiveness of a combination of anionic surfactant Linear Alkylbenzene Sulfonate, the role of ferric chloride and lime as coagulants in the coagulation and flocculation processes, and the filtration-adsorption process in improving water quality. Experiments were conducted with varying LAS concentrations (200–1000 ppm) through the stages of pretreatment, coagulation-flocculation, and filtration using silica sand, manganese, and activated carbon media. The results showed that the addition of LAS surfactant helped raise the pH toward neutral conditions. The optimum pH was achieved at a surfactant concentration of 400 ppm, with a pH of 7. The combination of coagulation-flocculation and filtration-adsorption processes was able to significantly reduce COD. The highest COD reduction efficiency was achieved in Treatment Variation 2 with filtration-adsorption at 85.3% at a concentration of 400 ppm, thereby meeting water quality standards (<10 mg/L). However, an excessive increase in LAS concentration reduced treatment efficiency due to adsorbent saturation and micelle formation. Overall, this combined method proved to be effective and practical for treating hydrocarbon-contaminated well water.

Keywords: *Anionic surfactants, Coagulation, Flocculation, Filtration, Adsorption*

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INTRODUCTION

Clean water is a primary means for the survival of humans and ecosystems. However, the availability of clean water, especially groundwater as the main source, is increasingly threatened by various forms of pollution. This pollution can originate from industrial, agricultural, or household activities that release harmful contaminants into the environment. One type of pollution that often occurs is hydrocarbon contamination, which can come from industrial activities, oil spills, or leaks from underground storage tanks (Mawarni et al., 2023).

Hydrocarbon contamination in well water not only alters the physical characteristics of the water but also poses significant health risks to humans and the surrounding environment (Wahyuni, 2019). Hydrocarbons are organic compounds that require a large amount of oxygen to chemically decompose, thereby increasing the COD value of water beyond the established clean water quality standards. Additionally, some types of hydrocarbons and their degradation products can affect the acidity of water, causing the pH value of the water to not meet the clean water quality standards based on the Regulation of the Minister of Health of the Republic of Indonesia Number 2 of 2023. Water conditions with COD and pH values that do not meet quality standards are very dangerous for consumption or use in daily activities.

Surfactant or surface active agent is a substance added to liquids to enhance spreading properties by reducing surface tension (Hari et al., 2019). The active substances in surfactants have a high concentration compared to the surface of the solution. This causes the surface tension of the surfactant to decrease, allowing particles adhering to the washed object to detach and dissolve in water (Rusdi et al., 2020).

The treatment of oil-containing water usually has two stages: first, pre-treatment to reduce the levels of oil, grease, and suspended solids. Second, the advanced treatment stage is used to degrade and reduce pollutants to an acceptable discharge value. The complexity of wastewater composition and strict discharge limits requires a combination of several treatment methods (Kuyukina et al., 2020). Coagulation and flocculation are methods aimed at reducing pollutants in the form of colloids in water bodies. This method is carried out by adding coagulants that act as binders for the particles contained in the water (Windhono et al., 2023). Each type of coagulant has a different optimum concentration, as each coagulant has varying abilities to reduce pollutants in water, resulting in different levels of clarity in the treated water (Aini et al., 2021). The use of coagulant mass ferric chloride 3g and lime 3g, with a coagulant contact time of 15 minutes and rapid stirring rotation of 100 rpm, slow stirring of 40 rpm, and sedimentation for 30 minutes, showed a COD reduction efficiency of 34.4% (Santoso & Apriani, 2024). Research by Rajabi et al. (2025) explains in their journal that there is a significant interaction between coagulant dosage, pH, stirring time, and temperature on the reduction of COD.

One of the most cost-effective methods for mitigating water pollution is adsorption. Adsorption has high potential for removing color, odor, oil, and toxic pollutants because it has good absorption capacity. Adsorbents from coconut shell charcoal are efficiently used to reduce the levels of COD, BOD, TSS, and pH in wastewater containing LAS surfactants, with a maximum adsorption capacity of up to 37% (Septiani et al., 2023). Activated carbon has a small pore diameter, high abrasion resistance, and a sufficiently large surface area to be used in the adsorption process (Fadlilah et al., 2023). Silica sand is inert, helping to filter out large particles and solid materials from water.

Although various water treatment methods have been developed, there is still not much specific research on evaluating the effectiveness of method combinations for treating hydrocarbon-contaminated water. Previous research tends to focus on single methods or different combinations. Therefore, this study aims to analyze the combination of adding anionic surfactants LAS, lime coagulant, and ferric chloride, on the reduction of COD values and pH neutralization in contaminated well water. This approach will involve coagulation-flocculation, filtration, and adsorption methods using silica sand, manganese sand, and activated coconut shell carbon media, which are expected to provide innovative and effective solutions to address the problem of well water pollution by hydrocarbons.

MATERIALS AND METHODS

Materials

The material used was a water sample from a well contaminated with hydrocarbons located in the Central Bangka Islands. Other materials used are anionic surfactants of the type Sodium Linear Alkylbenzene Sulfonate (LAS) ECONAT N-65, coagulant Ferric Chloride ($FeCl_3$) (Supelco Merck Germany), and Lime (CaO) (ROFA 84%).

Pre-treatment

The pre-treatment process begins with emulsifying hydrocarbons in the sample by adding LAS surfactant. The variations in LAS surfactant concentration in this study are 200, 400, 600, 800, and 1000 ppm. The determination of this concentration follows the initial TPH value of the sample, which is 1 mg/L. The volume of the stock solution added (4, 8, 12, 16, and 20 ml) to 1L of sample water was then homogenized by rapid stirring at 100 rpm for 5 minutes (Fadila et al., 2024). To ensure accuracy and facilitate mixing into 1L of the sample without significantly altering the volume, a Stock Solution with a concentration of 50,000 mg/L (5%) was prepared.

Coagulation and Flocculation

The coagulation process is carried out by adding the coagulant ferric chloride 2.5 g/L and lime 3 g/L to the mixture, accompanied by rapid stirring at 100 rpm for 15 minutes. Subsequently, flocculation is performed with stirring at 40 rpm for 30 minutes. Then let it sit for 60 minutes to allow the formed flocs to settle and filter using filter paper. Then measure the chemical parameters, namely pH and COD, to observe the reduction in their levels.

Filtration and Adsorption

The filtration process uses filter paper and layered filter media made of silica sand, manganese sand, and activated carbon. The function of filtration is to remove materials such as sand, floc, or organic substances from water to enhance the effectiveness of hydrocarbon and organic compound removal. The adsorption process occurs when there is direct contact between the mixture of well water samples contaminated with hydrocarbon-surfactant LAS-coagulant ferric chloride and lime on the surface of the adsorbent or sorbent pores, which consist of silica sand, manganese sand, and activated carbon. The adsorption process that occurs is a physical adsorption process. In this study, two variations were conducted: Variation 1 Without Filtration-Adsorption and Variation 2 With Filtration-Adsorption.

Data Analysis

This research is an experimental study conducted continuously, starting from planning, preparation of tools and materials, design of research methods, experimentation, processing, and data analysis. testing the water quality parameters, namely pH (Acidity Level) and Chemical Oxygen Demand (COD), which will later be presented in the form of tables and graphs and compared with the quality standard based on the Regulation of the Minister of Health of the Republic of Indonesia Number 2 of 2023. The processing was carried out using 2 different variations to determine the most effective treatment. The results of the laboratory tests will be calculated using the formula for calculating the efficiency of COD reduction.

$$\text{Efficiency (\%)} = \frac{\text{Initial COD Concentration} - \text{Final COD Concentration}}{\text{Initial COD Concentration}} \times 100\% \quad (1)$$

RESULTS AND DISCUSSION

Sample Characteristics

Initial analysis of the contaminated well water raw material was conducted before processing to determine the baseline water quality and the level of contamination, as shown in Table 1.

Table 1. Test Result of Polluted Well Water Samples

Parameter	Unit	Quality Standard	Result
pH	-	6,5-8,5	5,7
COD	mg/L	<10	65,3

Table 1 shows that the well water is acidic because it is below the minimum pH threshold of 6.5, which means it does not meet the clean water quality standards according to Minister of Health Regulation Number 2 of 2023. In hydrocarbon pollution, the decrease in pH occurs due to the biodegradation process by microorganisms. Hydrocarbon-degrading bacteria produce organic acid compounds such as carboxylic acids as byproducts of metabolism, resulting in a decrease in the pH of the surrounding water. Low pH conditions can also be caused by the oxidation of hydrocarbon compounds or the activity of microorganisms that decompose organic matter into weak acids.

The results of the COD parameter test on the contaminated well water do not yet meet the clean water quality standards according to Permenkes Number 2 of 2023. A high COD value indicates pollution by hydrocarbons or oil and its derivatives. A high COD level indicates a large oxygen demand to chemically oxidize the organic material. This condition reflects the presence of petroleum residues or complex organic chemicals that are not easily degraded naturally.

The Effect of Variations in Surfactant Concentration, Coagulation-Flocculation, and Filtration-Adsorption on pH Parameters

Based on the chemical properties of the surfactant sodium linear alkylbenzene sulfonate, which is generally a weak base in the form of sodium salt, the addition of LAS to hydrocarbon-contaminated water can reduce interfacial tension and promote the formation of micelles, making it easier for hydrocarbons to be emulsified and dispersed in the aqueous phase (Zhao et al., 2021). This causes a gradual increase in pH, as the acidic contaminated well water binds some hydrogen ions (H^+), resulting in the water's pH rising toward neutral and slightly basic.

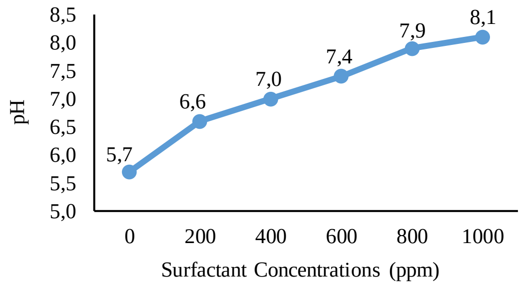

Figure 1. The Effect of Surfactant Concentration Variation on pH Parameters

Figure 1 shows that the addition of sodium linear alkylbenzene sulfonate (LAS) surfactant to hydrocarbon-contaminated well water at 1 mg/L affects the increase in pH value. The initial pH of the contaminated water was 5.7, which increased to 6.6 at a concentration of 200 ppm, 7.0 at 400 ppm, 7.4 at 600 ppm, 7.9 at 800 ppm, and 8.1 at 1000 ppm. The data shows a pattern of relationship between surfactant concentration and final pH value, meaning that the higher the concentration of added LAS, the higher the resulting final pH.

All variations of LAS concentration resulted in a final pH within an acceptable range for water quality. A concentration of 400 ppm was able to restore the pH to a neutral condition, while concentrations of 600-1000 ppm produced a slightly more alkaline pH. Thus, the addition of LAS within that concentration range is considered effective in improving the pH of hydrocarbon-contaminated well water.

The Effect of Variations in Surfactant and Coagulant Concentrations and Adsorption on COD Parameters

The addition of the coagulant ferric chloride is acidic because its hydrolysis process releases H+ ions, causing a drastic decrease in water pH (pH <5) [8], necessitating the addition of lime to raise or neutralize the water pH. The role of adding lime is as a neutralizing agent and provider of alkalinity. A relatively high dose of lime (3 g/L) has been proven to drastically raise the pH to slightly basic levels (pH 7-8), thereby optimizing the coagulation process and achieving the optimal pH range for floc formation. An agitation process at 100 rpm for 15 minutes ensures that the coagulant and lime are homogeneously mixed, while slow stirring at 40 rpm for 30 minutes facilitates the stable growth of flocs.

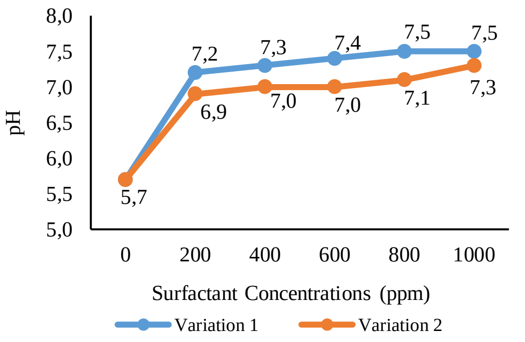

Figure 2. The Influence of Surfactant Concentration Variation, Coagulant, and Processing Variation on pH Parameter

Figure 2 shows a significant increase in pH across all variations of surfactant concentrations, with pH values meeting the clean water quality standards according to Permenkes Number 2 of 2023, which stipulates a pH range of 6.5-8.5. In the surfactant concentration variation of LAS 200 ppm, the pH increased to 7.2 from an initial pH of 5.7, reaching a pH of 7.5 at a concentration of 1000 ppm. This indicates that the high concentration of LAS surfactant in contaminated well water affects the chemical interaction with the added coagulant and lime, which has the potential to leave residual alkalinity in the treated water.

The increase in pH value is caused by the interaction of LAS, which is a weak base, with $FeCl_3$ and lime (3 g/L), leaving residual alkalinity (Santoso & Apriani, 2024). $FeCl_3$ is optimal at pH 4–9, according to initial conditions, thus supporting the destabilization of colloids into flocs (Hutabarat et al., 2022). The hydrolysis of $FeCl_3$ releases H^+ ions, causing a decrease in pH. $FeCl_3$ plays a role in binding negatively charged colloidal particles and forming easily sedimentable flocs.

$$FeCl_{3(s)} + 3H_2O_{(l)} \rightarrow Fe(OH)_{3(s)} + 3H^+_{(aq)} + 3Cl^-_{(aq)} \quad (2)$$

Lime (CaO) functions to raise the pH, producing OH^- ions and lowering the concentration of H^+ ions in water, through the reaction:

$$CaO_{(s)} + H_2O_{(l)} \rightarrow Ca(OH)_{2(s)} \quad (3)$$

The filtration and adsorption process using silica sand, manganese, and activated carbon functions as a buffer that brings the water's pH to a neutral condition. The effect of filtration-adsorption is observed from the comparison between the two treatments. Variation 2 shows that with filtration-adsorption, it consistently produces a lower pH value (approaching neutral) compared to variation 1 (without filtration-adsorption).

It can be seen from the pH values in variation 1 that the LAS surfactant concentration of 200 ppm has a pH value of 7.2 (slightly basic), at a concentration of 400 ppm, the water pH is 7.3, and at LAS surfactant concentrations of 800 and 1000 ppm, the pH value is the highest at 7.5. Thus, the higher the concentration of LAS surfactant used, the higher the pH value. In variation 2, the LAS surfactant concentrations of 200 and 400 ppm have already reached a neutral pH of 7.0, and at concentrations of 800-1000 ppm, the pH value is weakly basic and close to neutral at 7.1 and 7.3. The increase in pH from acidic conditions (5.7) to neutral and weakly basic (7.0-7.5) is a direct result of the chemical treatment process carried out. In addition to the influence of LAS surfactant concentration, the role of coagulants ferric chloride and lime, as well as manganese sand and activated carbon, also affects the pH value of the water. In each treatment of variation 1 and variation 2, a pH range of 7.0-7.5 was obtained, which still meets the clean water quality standards according to Minister of Health Regulation Number 2 of 2023 with a pH quality standard of 6.5-8.5.

The effect of surfactant concentration variation and filtration-adsorption on COD parameters

In Figure 3, it can be seen that the increase in LAS surfactant concentration from 200 to 1000 ppm shows a decrease in COD removal efficiency in both variation 1 and variation 2. In treatment variation 1, the efficiency decreased from 78.0% to 76.1%, while in variation 2, the efficiency decreased from 84.5% to 82.6%. This indicates that the high concentration of LAS surfactant in contaminated well water affects the chemical interaction with the added coagulant and lime.

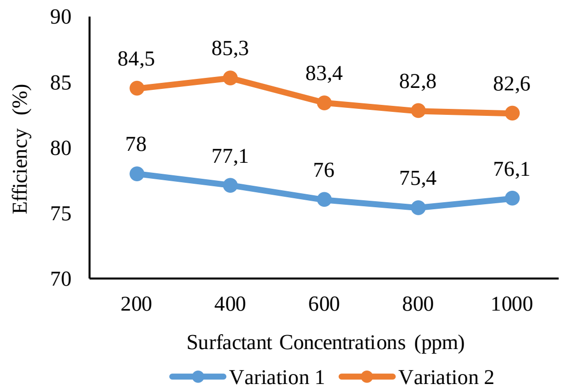

Figure 3. Test Results on the Effect of Surfactant Concentration Variation and Filtration Adsorption on the COD Parameter

Anionic surfactant LAS has a negative charge, and colloidal particles in water have the same negative charge, causing them to repel each other and remain suspended. $FeCl_3$ releases ions Fe^{3+} (iron (III)), which are positively charged; these ions neutralize the negative charge of LAS particles and colloids, making them unstable. The formation of hydroxide flocs occurs when Fe^{3+} reacts with water to form a precipitate like $Fe(OH)_3$, which traps LAS molecules and other colloidal particles within it. Flocs can bond to form larger flocs through a slow stirring process (Santoso & Apriani, 2024). This is due to the flocs containing organic materials, so the more flocs settle, the lower the concentration of COD in the water.

Research by Nugti et al. (2020) state that coagulants are used to facilitate separation during sedimentation. The addition of lime together with ferric chloride, aims to optimize the process and increase the efficiency of COD reduction. The acidic nature of ferric chloride causes a drastic decrease in pH, so lime is added to neutralize the acidity and maintain the pH within the optimal range because $Fe(OH)_3$ is the least soluble and most effective in forming flocs. The addition of lime can bind colloidal particles in the water, which then form a precipitate (Santoso & Apriani, 2024). Calcium ions (Ca^{2+}) from lime also act as co-coagulants that play a role in binding LAS surfactant molecules, especially at high concentrations.

Very high surfactant concentrations affect the COD results, where LAS molecules saturate the active sites on the adsorbent, thereby reducing the medium's capacity to remove other COD contaminants. This result is supported by previous research by Yusuf et al. (2024), which states that the saturation of the active sites on the adsorbent surface and the repulsive forces between the adsorbed organic pollutant molecules on the adsorbent surface hinder the diffusion of organic substances into the pores. This results in lower COD efficiency at surfactant concentrations of 800 and 1000 ppm. The increase in LAS concentration can reduce adsorption efficiency due to competition between surfactant molecules and other organic compounds in the water. Excess surfactants form micelles that bind some organic substances in the liquid phase, making them unable to be adsorbed by the medium.

Data shows that the treatment with filtration-adsorption results in higher efficiency, namely 82.6–85.3%, compared to without filtration-adsorption 75.4–78%. The water treated with filtration-adsorption at concentrations of 200 and 400 ppm has COD values of 9.8 and 9.3 mg/L, which meet the clean water quality standards according to Permenkes Number 2 of 2023. At concentrations of 800-1000 ppm, COD values above 10 mg/L were obtained, exceeding the quality standard (<10 mg/L). The treatment without filtration-adsorption with surfactant concentrations of 200-1000 ppm also resulted in values above the clean water quality standards. This indicates that there is an optimal influence between the solubility capacity of the surfactant and the adsorption capacity of the adsorbent.

The combination of silica sand, manganese, and activated carbon media can significantly enhance COD removal through physical filtration and chemical adsorption mechanisms. Silica sand acts as an initial filter for suspended particles that pass through the sedimentation stage and filter paper, and it can decompose organic materials present in the water (Abdillah & Mirwan, 2024). Manganese, as an oxidation catalyst to remove dissolved metals, helps in the oxidation of organic substances. Activated carbon provides a large surface area for the adsorption of organic compounds (Fasihah et al., 2022). Activated carbon media play an important role in removing dissolved organic compounds, including residual LAS and non-settled hydrocarbons. This increase in efficiency is also influenced by the ability of activated carbon to adsorb anionic surfactants like LAS through hydrophobic interactions and Van der Waals forces.

CONCLUSION

This study shows that the combination of the anionic surfactant LAS, the coagulant ferric chloride, and lime, as well as the filtration-adsorption process, is effective in improving the quality of hydrocarbon-contaminated well water. The addition of LAS was able to increase the pH from acidic conditions to neutral and near-neutral, while the combination of coagulation-flocculation and filtration-adsorption significantly reduced the COD value to meet the quality standards at an optimal concentration of 400 ppm. The best treatment was obtained in variation 2 with a LAS concentration of 400 ppm, a neutral pH of 7, and a COD reduction efficiency of 85.3%.

It is recommended that further research expand the surfactant dosage concentration to obtain more comprehensive results on the treatment of hydrocarbon-contaminated water. Furthermore, comparative studies using other types of surfactants outside the nonionic group are needed to evaluate differences in effectiveness and selectivity, thus obtaining more in-depth information regarding the influence of surfactant chemical structure on the treatment of hydrocarbon-contaminated water.

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