

UTILIZATION OF FLY ASH IN THE FERTILIZER INDUSTRY AS AN ALTERNATIVE TO SYNTHETIC COAGULANTS WITH THE ADDITION OF H_2SO_4

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ABSTRACT

Utilization of Fly Ash in the Fertilizer Industry as an Alternative to Synthetic Coagulants with the Addition of H_2SO_4 . This study aimed to utilize fly ash, a solid waste generated from coal combustion in the fertilizer industry, as an alternative coagulant for wastewater treatment. Fly ash was activated using H_2SO_4 at varying concentrations (2%, 4%, 6%, and 8%) and coagulant dosages (2 mL, 4 mL, and 6 mL). The effectiveness of fly ash as a coagulant in degrading suspended particles and Chemical Oxygen Demand (COD), as well as in reducing wastewater pH, was analyzed using the jar test method. The results demonstrated that fly ash coagulant was effective in reducing Total Suspended Solids (TSS) and COD, with the most significant reduction observed at a dosage of 4 mL and an H_2SO_4 concentration of 8%. Under these conditions, TSS decreased by 82.35% (from 0.79019 kg/ton to 0.13945 kg/ton), while COD decreased by 72.72% (from 0.02045 kg/ton to 0.00558 kg/ton). Correlation analysis indicated a strong relationship between H_2SO_4 concentration, coagulant dosage, and the reduction of TSS and COD. Although effective in reducing TSS and COD, the fly ash coagulant caused a decrease in wastewater pH due to its acidic nature. Further research is required to improve wastewater pH after the addition of fly ash coagulant.

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INTRODUCTION

In its development, the energy used to support production and transportation has shifted from reliance on human and animal power to the use of fossil energy sources such as petroleum and coal. Fossil energy requires millions of years to form, and at present, humans are highly dependent on these non-renewable energy resources⁽⁶⁾.

In addition to being non-renewable, the use of fossil energy—particularly coal—generates waste during industrial processes that can pollute the environment⁽¹¹⁾. In the Gresik area, a fertilizer and chemical industry operates its own power plant that utilizes coal as the primary energy source. Coal combustion in this power plant produces residual wastes in the form of fly ash and bottom ash⁽²⁴⁾. Currently, fly ash waste management is largely handled by third parties at a high cost, while some fly ash is disposed of in fly ash dumps. Such practices may cause environmental pollution, including air, soil, and groundwater contamination, and can also lead to social problems.

Coal combustion waste consists of approximately 80% fly ash and 20% bottom ash⁽¹⁾. Most coal combustion ash waste is still managed using landfill methods⁽¹⁷⁾, which have the potential to damage the environment. It is estimated that global fly ash production from coal

combustion has reached approximately 500 million tons per year. Coal is considered the most polluting fossil fuel; therefore, with the increasing use of coal as an energy source, its environmental impacts require serious attention.

Fly ash contains significant amounts of Al_2O_3 and FeO , which are essential elements for the development of alternative coagulants⁽⁴⁾. Aluminum- and iron-based coagulants have been widely applied in water and wastewater treatment processes due to their ability to provide positively charged ions that interact with negatively charged particles to form larger colloidal aggregates⁽⁸⁾. Consequently, fly ash can be managed using a reverse logistics approach to reduce its negative environmental impacts⁽²¹⁾.

Previous studies utilized HCl as an activator to enhance the coagulation properties of fly ash, as acid activation has been shown to effectively activate the constituent compounds in fly ash⁽¹⁾. Such studies examined the utilization of fly ash from the Ombilin Steam Power Plant as an alternative to artificial coagulants.

This study aims to synthesize coagulants derived from coal fly ash for application in wastewater treatment units within the fertilizer industry in the Gresik area, using H_2SO_4 as an activator. This approach offers a viable solution for managing coal fly ash waste while fulfilling the demand for coagulants by recycling fly ash for reuse in other process units. Previously, this waste was disposed of in landfills, posing risks of environmental pollution and social issues. Through this strategy, a closed-loop flow within the company's production process can be established. In addition to providing economic benefits, this approach enables companies to uphold environmental ethics in their business practices, thereby supporting the achievement of sustainable industrial development.

MATERIALS AND RESEARCH METHODS

The equipment used in this study included a furnace, a 100-mesh sieve, a mortar, an Erlenmeyer flask, a magnetic stirrer, a volumetric pipette, a volumetric flask, beakers, a measuring cup, a funnel, an analytical balance, an oven, a pH meter, a jar test apparatus, and filter paper. The materials used consisted of fertilizer industry wastewater, fly ash, distilled water (aquades), H_2SO_4 , and Whatman No. 41 filter paper.

In the initial stage of the experimental procedure, 20 g of fly ash samples were prepared and evenly placed in a porcelain crucible. The samples were then heated in a furnace at 400 °C for 1 h to remove moisture content and to promote pore formation in the fly ash. The dried fly ash was subsequently sieved using a 100-mesh sieve to obtain particles with a uniform and fine size distribution.

The sieved fly ash was mixed with an H_2SO_4 solution and continuously stirred at 80 °C for 60 min using a magnetic stirrer to extract aluminum components from the fly ash. The resulting mixture was filtered using filter paper to separate the insoluble fly ash residue from the formed coagulant solution. The filtrate, containing the fly ash-based coagulant, was collected and stored in a suitable container for further analysis or application. The application of the coagulant was subsequently conducted according to the dosage variations specified in the research variables using the jar test method.

RESEARCH RESULTS AND DISCUSSION

The results of the utilization of fly ash waste from the fertilizer industry in the form of coagulant products with the extraction of Al_2O_3 content have physical characteristics thicker than water and make water have increasingly acidic properties when reacted. The reaction that occurs produces coagulant in the form of aluminum sulfate solution (Al_2SO_4). The reaction that occurs is:



Based on the utilization results, chemical calculations were performed to determine the mass of $Al_2(SO_4)_3$ formed in each coagulant. The results indicated that increasing the H_2SO_4 concentration and the mass of fly ash used in coagulant preparation led to higher amounts of Al_2O_3 and $Al_2(SO_4)_3$ being formed. The results of the mass calculations for Al_2O_3 and $Al_2(SO_4)_3$ formation are presented in Table 1.

Table 1. Mass calculation results of Al_2O_3 and $Al_2(SO_4)_3$ formation.

No	Concentration H_2SO_4 (%)	Rate Fly Ash (kg)	Mass Al_2O_3 (kg)	Mass $Al_2(SO_4)_3$ (kg)
1	2	0,1	0,0069	0,0232
2	4	0,2	0,0139	0,0465
3	6	0,3	0,0208	0,0697
4	8	0,4	0,0278	0,093

Then laboratory analysis of coagulants from fly ash with coagulants that have a concentration of 8% H_2SO_4 and compared with the quality standard requirements for liquid aluminum sulfate in accordance with SNI 0032: 2011. It is known that the coagulant from fly ash has met some of the requirements of liquid aluminum sulfate according to SNI 0032: 2011 for liquid aluminum sulfate.

Table 2. Fly Ash Coagulant Analysis Results and Comparison with SNI 0032:2011 Quality Requirements for Liquid Aluminum Sulfate

No	Parameters	Unit	Result	Quality Requirements for Aluminum Sulfate SNI 0032: 2011	Conclusion
1	Specific gravity 20°C	-	1,42	Min. 1,3	Meets Standard
2	pH	-	1,2	Min. 3,0	Does Not Meet Standard
3	Water insoluble part	% (b/b)	0,19	Maks. 0,25	Meets Standard
4	Al_2O_3	% (b/b)	7,63	Min. 8	Does Not Meet Standard
5	Iron, Fe	% (b/b)	0,0008	Maks. 0,01	Meets Standard
6	Lead, Pb	mg/kg	8,7	Maks. 10	Meets Standard
7	Arsenic, As	mg/kg	1,5	Maks. 2	Meets Standard

Based on the analysis results, it was found that the fly ash-derived coagulant met several requirements for liquid aluminum sulfate according to SNI 0032:2011. However, some parameters, including pH and Al_2O_3 content, did not meet the standard. Therefore, further research is needed to optimize the coagulant preparation methods to achieve the required pH and Al_2O_3 levels in fly ash-based coagulants.

The appearance of fertilizer industry effluent before treatment was characterized by turbid water containing fine suspended particles. After the treatment process, noticeable changes in water quality were observed. Flakes or flocs formed and settled at the bottom of the beaker, resulting in clearer water. The high Total Suspended Solids (TSS) content in fertilizer industry wastewater is attributed to undissolved solid substances, both organic and inorganic, which include organic particles, suspended solids, fertilizer components, and lime residues ⁽²⁾.

This observation illustrates the physical changes in wastewater before and after the addition of fly ash coagulant using the coagulation-flocculation method. Furthermore, the test results demonstrate the effectiveness of fly ash coagulants in influencing pH and reducing TSS and Chemical Oxygen Demand (COD) in fertilizer industry wastewater.

Table 3. Relationship of Coagulant Dose Addition to pH, TSS, and COD Parameters

Concentration H_2SO_4	Dosage Coagulant (mL)	pH			TSS (kg/ton)			COD (kg/ton)				
		BM	HPA	HSPS	BM (kg/ton)	HPA	HSPS	ER (%)	BM (kg/ton)	HPA	HSPS	ER (%)
2%	2			4,5			0,278892	64,71			0,016734	18,17
	4			4,3			0,23241	70,59			0,013015	36,36
	6			4,2			0,185928	76,47			0,009296	54,54
4%	2			4,3			0,255651	67,65			0,013015	36,36
	4			3,7			0,185928	76,47			0,007437	63,63
	6			3,6			0,162687	79,41	0,3	0,020452	0,005578	72,72
6%	2	6-9	4,90		0,15	0,790194	0,209169	73,53			0,011156	45,45
	4			3,9			0,185928	76,47			0,009296	54,54
	6			3,6			0,162687	79,41			0,007437	63,63
8%	2			3,8			0,185928	76,47			0,007437	63,63
	4			3,5			0,139446	82,35			0,005578	72,72
	6			3,1			0,302133	61,76			0,013015	36,36

Effect of Coagulant on pH Parameters

The test results indicate that higher H_2SO_4 concentrations used in the preparation of fly ash coagulant lead to a significant decrease in wastewater pH. This is attributed to the inherent acidic nature of the fly ash-based coagulant. As the H_2SO_4 concentration increases, the resulting coagulant becomes more acidic, causing a greater reduction in the pH of fertilizer industry wastewater, particularly at higher coagulant concentrations and dosages.

Several mechanisms contribute to this pH decrease. First, increasing the coagulant concentration enhances the hydrolysis process in water, producing more H^+ ions and consequently lowering the pH. Second, the reaction of $Al_2(SO_4)_3$ generates sulfuric acid, which further reacts with the alkalinity of the wastewater, leading to an additional decrease in pH⁽⁸⁾.

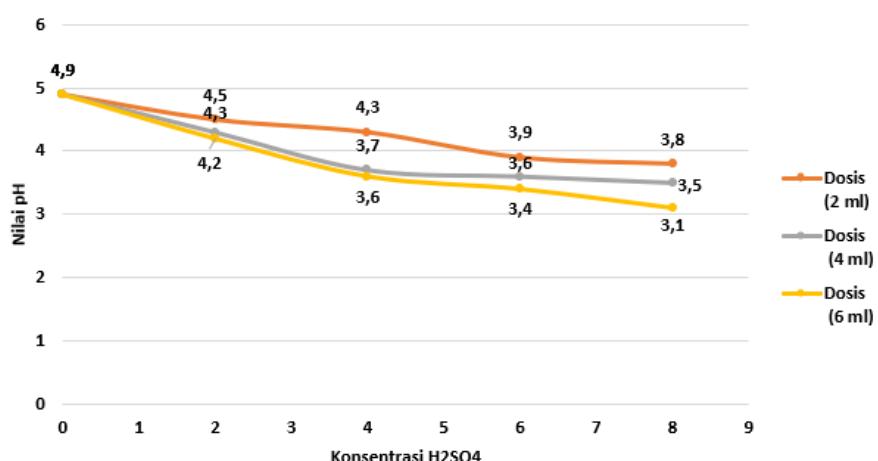


Figure 1. Graph of Effect of Coagulant Concentration and Dose on pH Parameters

Effect of Coagulant on pH Parameters

Data analysis from the graph demonstrates a clear downward trend in pH as both the H_2SO_4 concentration and coagulant dose increase. The most significant pH decrease occurred at an H_2SO_4 concentration of 8% with a coagulant dose of 6 mL, where the pH dropped from 4.9 to 3.1. Correlation analysis revealed a strong relationship between H_2SO_4 concentration, coagulant dose, and pH reduction, with a correlation coefficient of 89.31%. A P-value of 0.000 confirms that H_2SO_4 concentration and coagulant dosage have a significant effect on pH reduction.

However, the use of fly ash coagulant alone in the wastewater treatment process has not achieved optimal results in increasing pH. This is attributed to the high initial acid content in the wastewater and the already acidic condition of the effluent. The addition of coagulants at

high H_2SO_4 concentrations and dosages can have an opposite effect to the intended goal. Therefore, further studies are recommended to combine coagulant treatment with pH-increasing agents, such as quicklime (CaO), to neutralize acidic wastewater conditions⁽¹⁹⁾.

Effect of Coagulant on TSS Parameters

The addition of fly ash coagulant was effective in reducing Total Suspended Solids (TSS) in fertilizer industry wastewater. The optimum condition, achieved with a coagulant dose of 4 mL at an H_2SO_4 concentration of 8%, resulted in an 82.35% reduction in TSS, from 0.79019 kg/ton to 0.13945 kg/ton, meeting quality standards. This TSS reduction was accompanied by a clearer appearance of the wastewater and the formation of flocs at the bottom of the beaker. The formation of flocs occurs because the Al^{3+} ions from the coagulant bind to negatively charged particles in the wastewater, facilitating agglomeration and subsequent sedimentation.

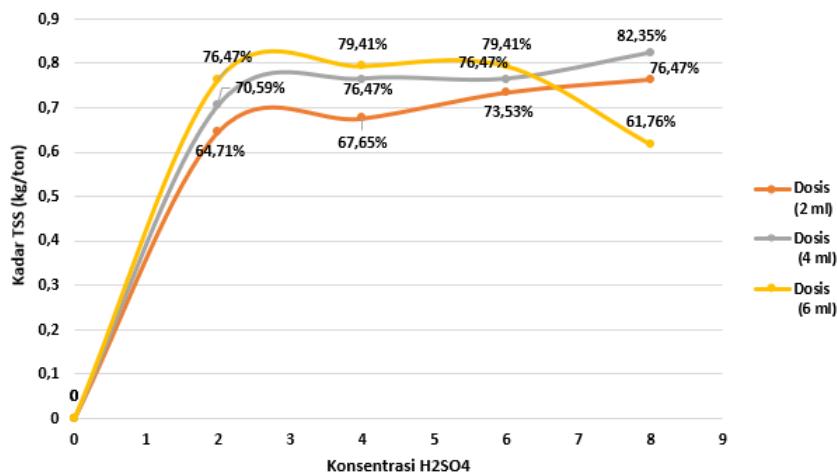


figure 2. Graph of Effect of Coagulant Concentration and Dose on TSS Parameters

Further analysis demonstrated that both coagulant dose and H_2SO_4 concentration had a significant impact on TSS reduction. The optimum condition, achieved at a coagulant dose of 4 mL with an H_2SO_4 concentration of 8%, showed the most pronounced effect. However, increasing the coagulant dose to 6 mL, beyond the optimum point, resulted in a decreased percentage of TSS removal. This is attributed to an excess of coagulant, which overcompensates the surface charge of the particles, rendering the flocs unstable and prone to re-dispersion⁽¹⁸⁾.

Multiple linear regression analysis confirmed the significant influence of coagulant dose and H_2SO_4 concentration on TSS reduction, with each variable showing a significance value of 0.000 (<0.05). The independent variables (coagulant dose and H_2SO_4 concentration) collectively explained 89.85% of the variation in the dependent variable (TSS reduction). Therefore, the optimum dose of fly ash coagulant for reducing TSS in fertilizer industry wastewater is 4 mL at an H_2SO_4 concentration of 8%.

Effect of Coagulant on COD Parameters

The addition of fly ash coagulant was also effective in reducing Chemical Oxygen Demand (COD) in fertilizer industry wastewater. The optimum condition, with a coagulant dose of 4 mL at an H_2SO_4 concentration of 8%, resulted in a 72.72% reduction in COD, from 0.02045 kg/ton to 0.00558 kg/ton. These results indicate that fly ash coagulant is effective in degrading COD in wastewater.

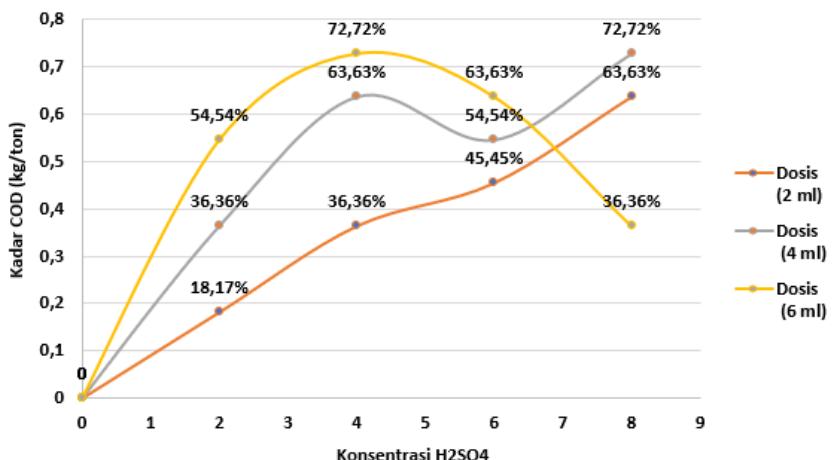


figure 3. Graph of Effect of Coagulant Concentration and Dose on COD Parameters

Effect of Coagulant on COD Parameters

The graph indicates a significant increase in COD removal efficiency with increasing H₂SO₄ concentration and coagulant dosage. Although the initial COD levels of the wastewater already met quality standards, testing was conducted to evaluate the effect of adding fly ash coagulant. A decrease in COD removal efficiency was observed at an H₂SO₄ concentration of 8% with a coagulant dose of 6 mL. This decline is likely due to the presence of excess organic matter from the coagulant, which cannot be fully degraded, thereby increasing the COD levels in the effluent ⁽⁹⁾.

Multiple linear regression analysis confirmed a strong correlation between H₂SO₄ concentration, coagulant dose, and COD reduction, with a correlation coefficient of 84.68%. The P-value of 0.000 indicates that both H₂SO₄ concentration and coagulant dosage have a significant influence on COD reduction.

Potential Cost Savings

The fertilizer industry examined in this study generates 2,800 m³ of wastewater per day and typically requires 4,600 liters of alum for treatment, meaning 1 liter of alum can treat approximately 0.6 m³ of wastewater. The industry also produces 20 tons of fly ash per day, which, if treated by a third party, costs IDR 350,000 per ton. Therefore, the total cost for fly ash treatment per day amounts to IDR 7,000,000.

This study demonstrates that fly ash can be utilized as a coagulant. To produce 900 mL of fly ash coagulant, 400 g of fly ash is required. The efficient coagulant dosage is 4 mL per 500 mL of wastewater. By utilizing the entire daily fly ash production (20 tons), approximately 450 liters of fly ash coagulant can be produced. This volume is sufficient to treat 56 m³ of wastewater. Consequently, the remaining wastewater requiring alum treatment is 2,744 m³, reducing the alum requirement to 392 kg. Prior to fly ash utilization, the industry spent IDR 8,600,000 per day for alum purchase (IDR 1,600,000) and fly ash treatment (IDR 7,000,000). After implementing fly ash-based coagulant, the total daily treatment cost decreased to IDR 1,568,000, resulting in daily savings of IDR 7,032,000.

Environmental Impact of Fly Ash Coagulant Use

The innovation of utilizing fly ash as a coagulant represents a significant advancement in sustainable wastewater management. Previously considered industrial waste, fly ash is now transformed into an effective solution for mitigating water pollution. The use of fly ash reduces dependence on conventional coagulants, which are often costly and may have negative environmental impacts.

Fly ash-based coagulants provide several advantages in wastewater treatment. The chemical properties of fly ash enable efficient coagulation and flocculation, allowing suspended particles to aggregate and settle. This is evident from the high removal efficiency achieved for TSS⁽⁵⁾. Similarly, the reduction in COD demonstrates that fly ash coagulants can improve wastewater quality, making effluent safer for environmental discharge or reuse within the company.

Beyond environmental benefits, this approach also offers significant economic advantages. The use of fly ash as a coagulant substantially reduces wastewater treatment costs by minimizing the need for commercial coagulant chemicals. Furthermore, the utilization of fly ash creates opportunities for new business ventures in the production and distribution of fly ash-based coagulants.

CONCLUSIONS AND RECOMMENDATIONS

The conclusions of this study indicate that the concentration of H_2SO_4 and the dosage of fly ash coagulant significantly affect the reduction of pH, Total Suspended Solids (TSS), and Chemical Oxygen Demand (COD) in fertilizer industry wastewater. This finding is supported by statistical analysis, which demonstrates a strong correlation between these variables. The optimum condition was achieved at an H_2SO_4 concentration of 8% with a coagulant dosage of 4 mL, resulting in TSS and COD removal efficiencies of 82.35% and 72.72%, respectively. Although the fly ash coagulant was effective in reducing TSS and COD levels, its application caused a decrease in wastewater pH. Therefore, additional treatment is required to neutralize the pH before the treated effluent is discharged into receiving water bodies. The application of this innovation is projected to reduce wastewater treatment costs by approximately IDR 7,032,000 per day.

Based on the results of this study, further research is recommended to develop effective methods for pH neutralization following coagulation. In addition, large-scale testing of the fly ash coagulant is necessary, along with further variations in stirring speed and initial wastewater pH, to determine the optimal operating conditions and pH range for practical application.

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