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Waste Utilization for Neutralization of Acid Mine Drainage Using Fly Ash, Bottom Ash, and Goat Manure Granules

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Abstract: Acid mine drainage (AMD) is one of the most serious environmental impacts of mining, characterized by low pH and high metal content. This study aimed to evaluate the neutralization efficiency of waste-based granules made from fly ash, bottom ash (FABA), and goat manure for AMD treatment under laboratory conditions. Two types of granules were produced: (i) 45% fly ash, 45% bottom ash, and 10% cement; and (ii) 45% fly ash, 30% bottom ash, 15% goat manure, and 10% cement. Each type was applied at different volumes (10%, 30%, and 50%) and positions (bottom and suspended) for 48 hours. Results showed that granule type, volume, and placement significantly influenced pH neutralization and metal removal. The best performance was achieved by the suspended granule without manure at 50%, raising AMD pH from 2.45 to 9.17 within 3 hours, with 88.7% Fe and 66.5% Mn removal. XRD and Micro-XRF analyses confirmed that neutralization occurred through dissolution of Ca-based minerals and formation of calcite and gypsum. The study demonstrates that FABA granules offer a low-cost, sustainable option for AMD treatment and can serve as a contextual learning model in environmental and chemistry education, linking waste utilization with real-world applications of acid-base reactions and environmental sustainability.

Keywords: AMD; FABA; Granules; Manure; pH neutralization

Introduction

Acid mine drainage (AMD) is a persistent environmental problem associated with mining activities and represents a complex geochemical process governed by acid–base reactions, oxidation–reduction reactions, and metal solubility equilibria (Jiao, Zhang, Su, Tang, Huang, & Ma, 2023). From a scientific perspective, understanding AMD formation and treatment provides an important context for applying fundamental chemistry concepts, including mineral oxidation, alkalinity generation, metal precipitation, and pH regulation (Skousen, Zipper, Rose, Ziemkiewicz, Nairn, McDonald, & Kleinmann, 2017). Integrating real-world AMD case studies into science education has also been shown to strengthen contextual and inquiry-based learning in environmental chemistry and sustainability studies (Araripe & Zeidler, 2024).

Mining has become a key part of economic development in many countries, mainly because it supplies the raw materials needed for industry and energy (Pavolova, Čulková, Šimková, Seňová, & Kudelas, 2022; Amegboleza & Ülkü, 2025). Over the years, the scale of mining has kept growing, and large areas of land continue to be opened for mineral and coal extraction (Jasansky, Lieber, Giljum, & Maus, 2023). In Indonesia, for instance, mining concessions already cover about 9.1 million hectares, which include both

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operating sites and areas that have been left after mining (Muliawati, 2024). While the sector contributes significantly to the economy, the environmental impact is hard to ignore. One of the most persistent problems faced in mining regions is AMD, which is still widely regarded as a major source of long-term pollution (Mukherjee, Paramanik, Paramanik, Dasmodak, Rajak, & Ganguly, 2024). AMD usually develops when rocks that contain sulfide minerals are exposed to air and water during excavation, drilling, or when stored as waste (Simate & Ndlovu, 2021). This exposure sets off oxidation reactions that produce acidic, metal-rich water, and this can occur in waste rock piles, tailings, and even in the exposed rock walls of mine pits (Pope, Christenson, Gordon, Newman, & Trumm, 2018; Wright, Paciuszkiewicz, & Belmer, 2018).

AMD is basically very acidic water with a really low pH, and it often ends up carrying a lot of dissolved metals and sulfate too (Yuan, Ding, Bi, Li, Wen, & Bai, 2022). This usually starts from rocks that contain sulfide minerals (Otunola & Mhangara, 2024). Pyrite (FeS_2) is the one most people mention (Saputra & Ramli, 2023; Parbhakar-Fox & Lottermoser, 2016; Susanto, Soedjono, & Titah, 2024). When these rocks are opened up and get in contact with air and water during mining, the reaction slowly produces sulfuric acid (Parbhakar-Fox & Lottermoser, 2016; Skousen, Ziemkiewicz, & McDonald, 2019). After that, the acidic water dissolves the metals from the rocks, like Fe, Al, Mn, Cu, Zn, Cd, Pb, and As, and they flow out into the mine water (Durães, Bobos, & Da Silva, 2017; Plante, Schudel, & Benzaazoua, 2021). Once the metals are dissolved like that, they move around more easily in the environment. This is worrying because organisms can absorb them (Chan, Routh, Luo, Dario, Miao, Luo, & Wei, 2021), and the effects don't always show right away. Over time, streams near mines, soil, and even plants and animals around the area start to show toxic impacts from this metal-rich and acidic runoff (Venkateswarlu, Nirola, Kuppusamy, Thavamani, Naidu, & Megharaj, 2016; de Paiva Magalhães, da Costa Marques, Baptista, & Buss, 2015).

In Indonesia, AMD problems have been found in many of the main coal-producing areas, such as East Kalimantan, South Kalimantan, Central Kalimantan, and Sumatra (Oktariani, Putri, & Situmorang, 2025). Some of the post-mining voids there have extremely acidic water, with pH recorded as low as 3.31 (Kusdarini, Sania, & Budianto, 2024). These acidic lakes are not only a chemical issue, but they have also become a safety problem for nearby communities. There were at least 168 deaths reported between 2014 and 2020 because people, often children, fell or drowned in abandoned mining pits (Laia, 2021). Cases like this show how serious the situation is and why AMD treatment and better mine-water management are urgently needed. This is also in line with Government Regulation No. 22/2021, which states that the parties responsible for pollution must work to restore the water quality in the affected areas.

Conventional AMD treatment is usually carried out by adding alkaline materials, for example limestone, lime, or sodium hydroxide, to increase the pH so that the dissolved metals can precipitate as metal hydroxides (Zvinowanda & Caliphs, 2023). Although this method is quite effective, it often requires high operational costs and is not suitable for long-term treatment, especially in large post-mining areas (Festin, Tigabu, Chileshe, Syampungani, & Odén, 2019). Because of this limitation, passive treatment systems have received more attention (Skousen et al., 2019). These systems make use of industrial waste materials or natural materials that have buffering capacity, and they are considered more practical and low-cost to operate (Dold, 2017).

Fly ash and bottom ash (FABA), which are by-products from coal combustion, contain alkaline oxides such as CaO, MgO, and Al_2O_3, and several studies have shown that they have good potential to neutralize AMD (Qureshi, Jia, Maurice, & Öhlander, 2016; Weinberg, Coyte, Wang, Das, & Vengosh, 2022; Saidy, Lestari, & Wulandari, 2024). When FABA is mixed with a binding material like cement, it can be processed into granule form so that the reactivity and stability of the material in water can be improved (Agusta, 2017).

In this study, AMD taken from a gold mining site in Banten, Indonesia was treated using granules made from FABA and goat manure. Two types of granules were prepared and tested by applying them in different positions and with different volume ratios to see their ability in neutralizing AMD and removing dissolved metals.

The purpose of this research is to provide basic data that can support the development of passive treatment systems for AMD management, especially for post-mining areas in Indonesia. In addition, this study offers valuable educational relevance by demonstrating the application of chemical principles such as neutralization, precipitation, and pH regulation in solving real environmental problems. The findings can serve as contextual learning material for environmental and chemistry education, fostering students' understanding of how scientific concepts are applied in sustainable waste management and pollution control.

Method

This study used a completely randomized design with three factors and three repetitions. In total, there were 36 treatment units, plus three units that were kept as controls without any treatment. The first factor (A) was the granule type. Granule 1 (A1) was made with

45% fly ash, 45% bottom ash, and 10% cement, while Granule 2 (A2) used 45% fly ash, 30% bottom ash, 15% goat manure, and 10% cement. The second factor (B) was the placement of the granules, either put at the bottom of the container (B1) or placed near the surface (B2) by hanging them with a mesh. The third factor (C) was the amount of granules added, which was 10% (C1), 25% (C2), or 50% (C3) of the 200 mL volume, and the rest of the space was filled with AMD. Each combination was repeated three times, making 36 units, and another three units were used as untreated controls.

The AMD used in this experiment was taken from a gold mining site in Banten. The samples were kept in clean polyethylene jerrycans that had been washed before use. The FABA was collected from PLTU Lontar 3 in Banten, and the goat manure was bought from a local fertilizer shop, then air-dried and sieved before using it. Ordinary Portland cement was used as the binder for making all the granule types.

Two types of granules were made based on the formulas mentioned earlier. Each material was first air-dried for about 2–3 days, then crushed and sieved to get a particle size of less than 1 mm. The materials were then mixed according to the ratio needed and sprinkled with a bit of water to help form the granules. The granules produced were around 5–10 mm in size. They were air-dried for 48 hours and then left to cure at room temperature for another two days before being used.

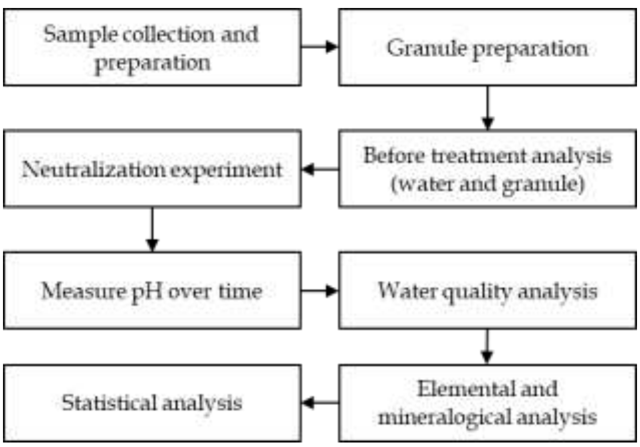

Figure 1. Research flow chart

The neutralization tests were done in 200 mL plastic measuring cups at room temperature (around 25 ± 2 °C). Each cup was filled with the granules based on the treatment design, and then AMD was added. The initial pH of the AMD was checked before adding the granules, and the pH was then recorded at 3, 6, 9, 12, 24, and 48 hours using a calibrated pH meter (Mediatech 3in1 Portable Multifunction TDS Temp pH Meter 9901). The experiment was kept in static conditions to represent a passive treatment setup.

Before the treatment, the AMD samples were tested for total suspended solids (TSS) using the APHA 24th 2540-Solids D method, and for dissolved metals (Fe and Mn) using Atomic Absorption Spectrophotometry (AAS, APHA 24th 3111B). After the experiment, the best treatment (the one that reached the highest pH within 3 hours) and the least effective treatment (the one that reached the lowest pH above 6 after 48 hours) were tested again for the same parameters using the same methods. The granules before and after the reaction were also analyzed using Micro-XRF to see the elemental composition, and XRD (PANalytical Empyrean) to check the crystal phase changes related to the AMD neutralization process.

The pH values at 48 hours were compared between the treatment groups and the control using a one-way ANOVA to see how effective the treatments were. For the pH changes over time (0–48 hours) within the treatment groups, a three-way repeated measures ANOVA was used to look at the main effects of granule type, placement, and volume, as well as their interactions. If there were significant differences ($p < 0.05$), a Bonferroni post hoc test was done to find out which treatment combinations were better. All statistical analysis was carried out using JASP version 0.95.30.

Result and Discussion

Initial characteristics of acid mine drainage

The AMD taken from the gold mining site had a very acidic condition with an initial pH of 2.45. The water quality test showed that the TSS was 16 mg/L, dissolved Fe was 32.42 mg/L, and dissolved Mn was 1.97 mg/L. The Fe and Mn levels were much higher than the water quality limits for most mining activities (except for Mn), based on the Regulation of the Minister of Environment and Forestry (MoEF) No. 5/2022, which sets the limits at 5–7 mg/L for Fe and 1–4 mg/L for Mn. This shows that treatment is needed. The TSS was still below the maximum limit (200 mg/L), but the very low pH (standard is 6–9) already gives a serious risk to aquatic life and also makes the water not suitable for reuse. Another point found in this early test is that Fe and Mn are actually not listed as standard parameters for gold mining wastewater in the current regulation. This makes the research even more relevant, as it turns out that the Fe and Mn content in post-gold mining water is high.

Treatment and controls comparison

The one-way ANOVA showed that there was a significant difference in the pH at 48 hours between the treated groups and the control ($p < 0.001$). The untreated AMD stayed almost the same in pH during the whole observation, while all the treatments showed an increase in pH with different levels. This result confirms that using FABA and goat manure granules, no matter the type or setup, was able to reduce the acidity of the AMD better than leaving it untreated (see Figure 2).

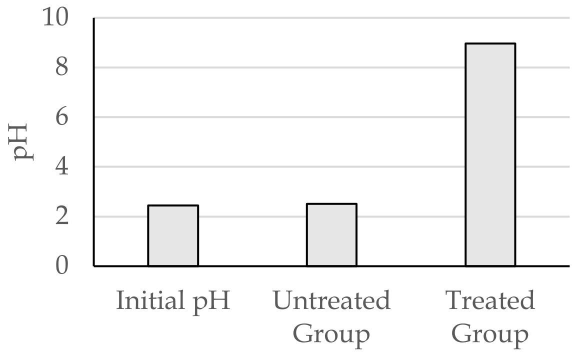

Figure 2. Average pH comparison between treated groups and untreated group in 48-hour

Similar to previous studies, treatments using FABA and combinations with organic matter significantly increased the pH of AMD compared to untreated controls, which remained consistently acidic (Husada, Fauzi, Mansur, & Suharyono, 2025). Laboratory experiments have shown that FABA applications can elevate AMD pH from strongly acidic toward neutral, and combined FABA-organic treatments further improve pH and metal removal relative to untreated AMD (Raletsena & Mongalo, 2024; Saidy, Priatmadi, Septiana, & Mulyawan, 2021).

Temporal dynamics pH neutralization

The three-way repeated measures ANOVA (Table 1) showed that all the main factors and their interactions with time had a very significant effect on pH. Time gave the strongest effect, which means the pH increased a lot during the 48 hours for all treatments. The Greenhouse–Geisser correction was used because the sphericity assumption was not met.

All the three-way and two-way interactions between time and the treatment factors were also significant ($p < 0.001$). This means that the speed of pH increase was affected by the granule type, placement, and volume, both on their own and when combined. The interaction between time and granule position had the strongest effect size ($\omega^2 = 0.950$), followed by the three-way interaction of time, position, and volume ($\omega^2 = 0.924$).

Main effects of treatment factors

The between-subjects results also showed that all three factors had a significant effect (Table 1). The placement of the granules gave the biggest influence on the overall pH ($\omega^2 = 0.996$), followed by the granule volume ($\omega^2 = 0.985$) and then the granule type ($\omega^2 = 0.890$).

Table 1. Result of Three-Way Repeated Measures ANOVA

Cases	Sum of Squares	df	Mean Square	F	p	ω^2
Within Subjects Effects						
Time	921.467	3.237	284.654	3274.78	< .001	0.991
Time ✻ Granule_Type	32.54	3.237	10.052	115.64	< .001	0.797
Time ✻ Granule_Position	157.411	3.237	48.626	559.42	< .001	0.95
Time ✻ Granule_Volume	78.07	6.474	12.058	138.72	< .001	0.835
Time ✻ Granule_Type ✻ Granule_Position	10.689	3.237	3.302	37.99	< .001	0.558
Time ✻ Granule_Type ✻ Granule_Volume	15.572	6.474	2.405	27.67	< .001	0.496
Time ✻ Granule_Position ✻ Granule_Volume	185.462	6.474	28.646	329.56	< .001	0.924
Time ✻ Granule_Type ✻ Granule_Position ✻ Granule_Volume	27.687	6.474	4.277	49.2	< .001	0.64
Residuals	6.753	77.691	0.087			
Between Subjects Effects						
Granule_Type	19.378	1	19.378	404.29	< .001	0.89
Granule_Position	596.596	1	596.596	12447	< .001	0.996
Granule_Volume	230.496	2	115.248	2404.46	< .001	0.985
Granule_Type ✻ Granule_Position	1.093	1	1.093	22.81	< .001	0.304
Granule_Type ✻ Granule_Volume	5.629	2	2.815	58.72	< .001	0.606
Granule_Position ✻ Granule_Volume	70.773	2	35.386	738.28	< .001	0.952
Granule_Type ✻ Granule_Position ✻ Granule_Volume	6.263	2	3.132	65.34	< .001	0.632
Residuals	1.15	24	0.048			

The Bonferroni post hoc test showed that Granule 1 (without manure) gave higher pH values than Granule 2 (with manure) at all time points and for all setups, with a mean difference of 0.555. Granules that were hung or suspended under the water surface also increased the pH much more than those placed at the bottom of the container, with a mean difference of 3.077.

For the volume factor, all pairwise comparisons showed significant differences. The 50% volume gave the highest pH increase, with a difference of 0.638 units compared to 30% volume and 2.271 units compared to 10% volume. The 30% volume treatment also increased the pH much more than the 10% volume, with a mean difference of 1.633.

Optimal treatment combination

Figure 3 shows how the pH changed over time for the different treatment combinations. The best result was from the granule without manure, placed in the suspended position with 50% volume. This treatment increased the pH from 2.45 to above 7.0 in less than 3 hours. This combination showed a very fast neutralizing effect, reaching a pH of 9.17 within 3 hours. On the other hand, the least effective setup (granules with manure, placed at the bottom, 50% volume) took 24 hours just to reach pH 4.58 and only reached a final pH of 8.58 after 48 hours.

The rapid pH increase observed in the suspended granules at high dosage is consistent with previous studies reporting fast alkalinity release from FABA during AMD treatment. FABA only systems have been shown to neutralize acidic mine water within hours, whereas the addition of organic matter can delay early pH response due to slower reaction kinetics. Improved contact between alkaline materials and AMD, such as suspended placement, further enhances neutralization efficiency (Qureshi et al., 2016; Skousen et al., 2019; Weinberg et al., 2022).

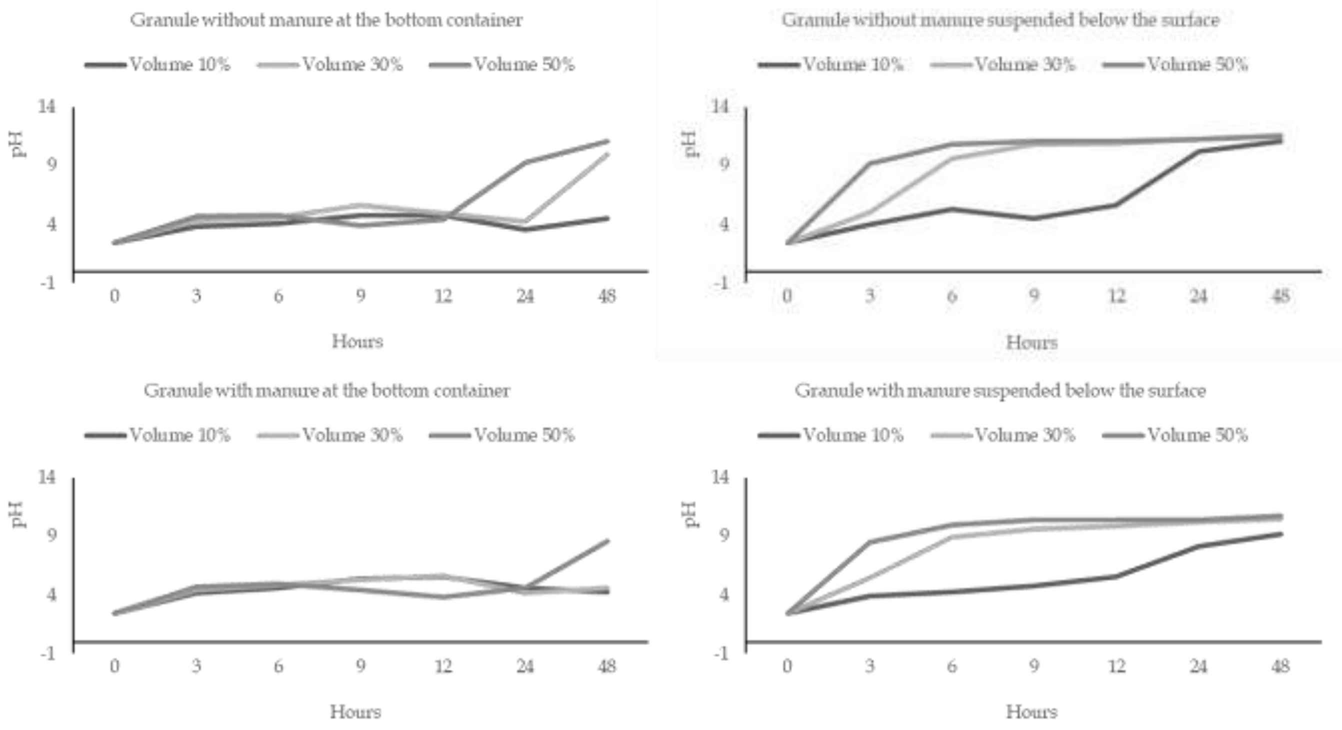

Figure 3. Temporal progression of pH across different combinations

Characterization of granules and metal removal

The Micro-XRF results (Table 2) showed clear differences in the composition of the two granule types, both before and after the treatment. Granule 1 (without manure) was dominated by Si (34.61%), Al (18.37%), Ca (20.25%), and Fe (18.31%), representing the aluminosilicate and CaO phases characteristic of FABA. Granule 2 (with manure) contained slightly lower Si (28.56%) and Al (14.75%) but higher Ca (30.32%), reflecting partial incorporation of Ca-bearing minerals and organic residues from goat manure.

After 48 hours of AMD neutralization, notable elemental shifts occurred. In Granule 1, which was positioned in a suspended configuration below the water surface after treatment and exhibited the most rapid and highest pH increase, Si and Al decreased to 33.39% and 16.44%, respectively, while Ca and S increased to 24.15% and 0.97%. These changes indicate partial leaching of aluminosilicate glass phases under acidic conditions and formation of Ca–S secondary minerals, likely gypsum or ettringite, through reaction with sulfate ions from AMD. Fe decreased slightly from 18.31% to 17.55%, suggesting that part of the Fe removed from solution precipitated as free flocs rather than adhering to the granule surface.

Conversely, in Granule 2, which was placed at the bottom of the container after treatment and showed the slowest neutralization performance in 48 hours, Ca decreased from 30.32% to 27.66%, while Fe increased from 16.75% to 17.58%. The increase in surface Fe demonstrates enhanced adsorption and precipitation of Fe(III) species on manure-amended granules, possibly aided by functional groups in the organic matrix. The lower Ca fraction suggests greater Ca consumption during buffering, in agreement with the slower pH increase observed in bottom-placed treatments. These results confirm that AMD neutralization involved dissolution–precipitation and surface sorption mechanisms, where Ca-based minerals provided alkalinity, and Fe and Mn were immobilized through precipitation and adsorption.

Table 2. Elemental Composition of Granule Before and After Treatment using Micro-XRF

Element (%)	Granule 1 (Without manure)		Granule 2 (With manure)	
	Before	After	Before	After
Silicon	34.6	33.39	28.56	30.12
Calcium	20.25	24.15	30.32	27.66
Aluminium	18.37	16.44	14.75	15.01
Iron	18.31	17.55	16.75	17.58
Magnesium	1.96	1.87	1.92	2.18
Potassium	1.58	1.35	2.2	1.83
Titanium	1.35	1.28	1.26	1.25
Sodium	1.17	1.05	1.18	0.98
Sulfur	0.69	0.97	0.96	1.11
Palladium	0.68	0.92	0.7	0.75
Strontium	0.4	0.43	0.51	0.45
Manganese	0.28	0.26	0.28	0.3
Others*	0.37	0.33	0.64	0.78
Total	100	100	100	100

Note: *Others include cobalt, phosphorus, zinc, vanadium, copper, rubidium, chromium, rhodium, chlorine, and zirconium

Previous studies have demonstrated that the configuration and contact regime of reactive media significantly influences AMD treatment efficiency (Thisani, Kallon, & Byrne, 2021). Laboratory investigations showed that flow configuration and contact time affect pH increase and metal removal performance by altering the extent of interaction between reactive media and AMD, with more effective contact conditions improving neutralization outcomes (Nordstrom, Blowes, & Ptacek, 2015; RoyChowdhury, Sarkar, & Datta, 2015). Reviews of passive AMD treatment systems also emphasize that system design and media positioning determine treatment effectiveness by modifying residence time and water–substrate contact (Skousen et al., 2017).

XRD diffraction patterns (Figure 4) further verified the mineralogical transformations inferred from the μ-XRF results. Before AMD exposure, both granule types exhibited strong peaks at 2θ ≈ 26.6° (quartz, SiO_2) and minor peaks around 29.4–29.6° (calcite, $CaCO_3$), along with faint reflections attributed to hematite (Fe_2O_3) and anhydrite ($CaSO_4$). After AMD treatment, new or intensified peaks appeared at 20.9° and 29.5°, corresponding to gypsum ($CaSO_4 \cdot 2H_2O$) and calcite, while the relative intensity of the quartz peak remained stable. The emergence of these secondary phases indicates formation of Ca–S minerals and carbonate reprecipitation during AMD neutralization.

For granule without manure, suspended below the water surface, the calcite and gypsum/ettringite peaks became dominant after treatment, confirming that neutralization occurred primarily through rapid dissolution of CaO and $Ca(OH)_2$ followed by reprecipitation of Ca–Al–S phases. This process stabilized the pH at neutral to alkaline levels within 48 hours.

Neutralization of AMD via calcium hydroxide addition increases pH by consuming H^+ and releasing Ca^{2+}, which subsequently precipitates as carbonate (e.g., calcite) and sulfate minerals such as gypsum once sulfate ions are available in solution. Studies on AMD neutralization have shown that calcite dissolution/precipitation is a dominant mechanism as pH rises with FABA input, and gypsum commonly forms as Ca^{2+} reacts with dissolved sulfate in treated waters (Iakovleva, Mäkilä, Salonen, Sitarz, Wang, & Sillanpää, 2015; Akinwekomi, Kefeni, Maree, & Msagati, 2016). Moreover, under high-alkaline conditions with sufficient aluminum, ettringite and other calcium aluminate sulfate phases can also precipitate, capturing sulfate and stabilizing the pH (Pratinthong, Sangchan, Chimupala, & Kijjanapanich, 2021).

For the granule with manure that was placed at the bottom of the container, the calcite peak became a bit lower, and the wider Fe-related peaks showed that amorphous Fe oxyhydroxides (such as goethite or ferrihydrite) were formed. These Fe-bearing phases matched the Micro-XRF results, which showed more Fe on the granule surface, and also supported the decrease of dissolved Fe in the water tests. From these mineral changes, it can be seen that the granule without manure that was suspended in the water mainly worked through fast Ca-based neutralization, while the granule with manure placed at the bottom helped to retain Fe more through sorption and redox-related precipitation under lower oxygen conditions.

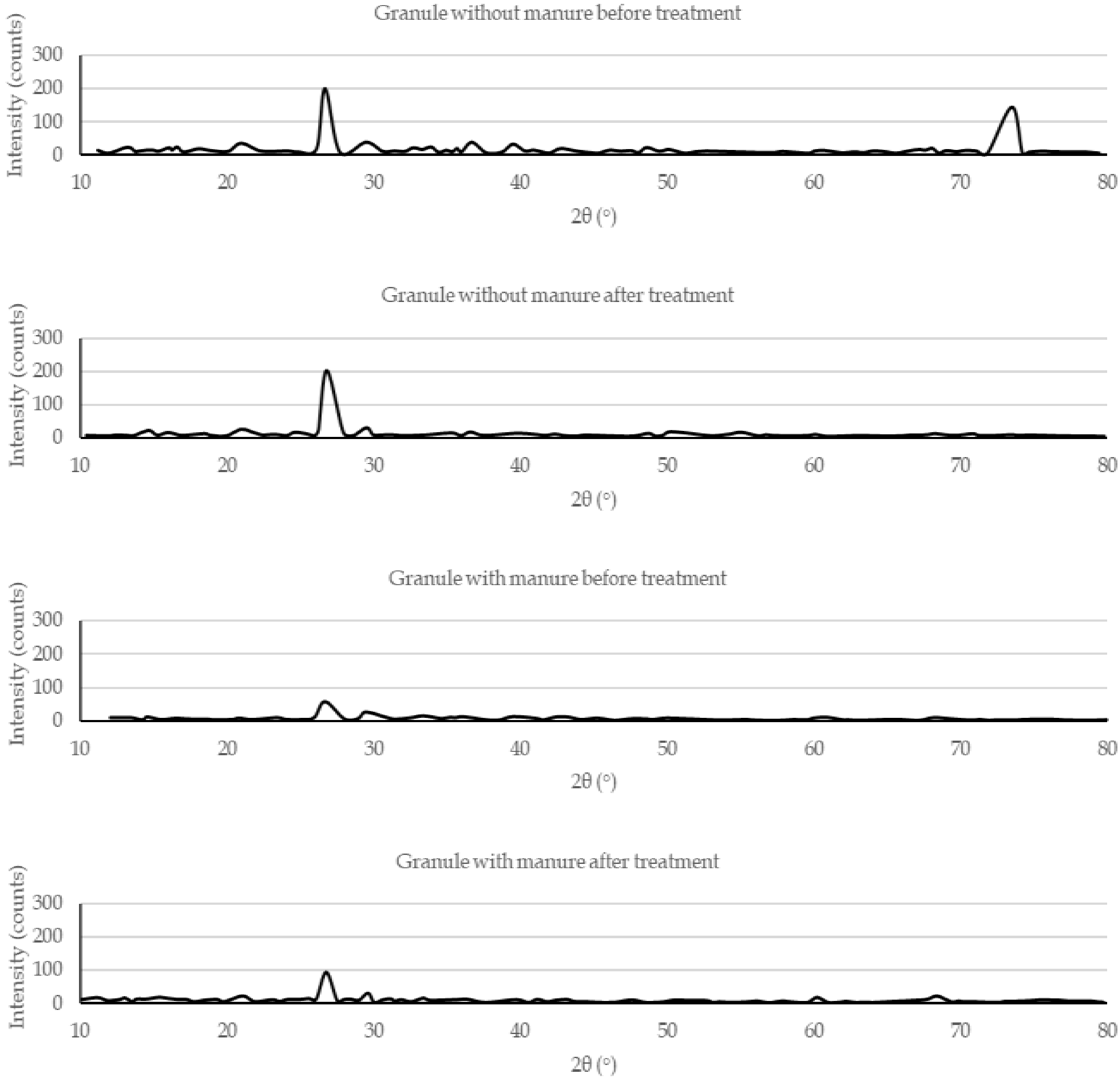

Figure 4. X-ray diffraction (XRD) patterns of granules before and after 48 hours of acid mine drainage (AMD)

Water quality analysis

The chemical quality of AMD improved markedly after treatment with both granule types. The initial pH of 2.45 increased to 9.17 in 50% volume of granule without manure suspended below the water surface treatment within 3 hours, surpassing the neutralization threshold. This is the faster increased of pH in all of the recorded experiment. In contrast, granule with manure placed at the bottom of the container in 50% volume achieved a pH of 8.58 after 48 h, indicating slower alkalization due to reduced Ca reactivity and limited aeration at the bottom position.

Dissolved Fe concentration decreased sharply from 32.42 mg/L to 3.66 mg/L in the best-performing treatment, achieving 88.7% removal efficiency, while Mn decreased from 1.97 mg/L to 0.66, achieving 66.5% removal efficiency. Meeting regulatory standards under Regulation of the MoEF of the Republic of Indonesia No. 5/2022. The manure-amended granule also meeting the regulatory standards by achieving Fe and Mn concentrations of 3.6 mg/L and 0.77 mg/L, respectively. Total suspended solids (TSS) remained low at 60 mg/L and 16 mg/L, respectively. Confirming that the neutralization process did not introduce excessive particulate matter into solution.

The fast increase in pH and the high removal of Fe and Mn show that the Ca- and Al-containing oxides in the FABA worked well as sources of alkalinity, and the formation of secondary Ca–S and Fe-hydroxide phases helped to trap the dissolved metals. These results are also in line with the mineral changes seen in the XRF and XRD analysis.

Conclusion

This study demonstrated that FABA granules are effective in neutralizing AMD, with granule composition, placement configuration, and treatment volume significantly influencing the rate and magnitude of pH improvement, metal removal, and mineral transformations. The best-performing treatment, granules without manure, applied in a suspended setup and at 50% volume, successfully increased pH from 2.45 to above 7.0 within three hours and removed 88.7% Fe and 66.5% Mn. Micro-XRF and XRD analyses confirmed that neutralization was driven by the dissolution of Ca- and Al-rich oxides and the formation of secondary minerals such as gypsum and calcite, which contributed to pH stabilization and metal sequestration. Although manure addition enhanced surface adsorption capacity, it slowed pH elevation, indicating suitability only for systems requiring gradual neutralization. Overall, these findings show that optimized FABA granules, particularly in suspended configurations, offer a low-cost and environmentally beneficial option for passive AMD treatment, with further field-scale evaluation recommended.

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Place acknowledgments, including information on grants received, before the references, in a separate section, and not as a footnote on the title page

Author Contributions
Conceptualization, I.M.; methodology, D.P.R.; formal analysis, D.P.R.; writing—original draft preparation, D.P.R.; writing—original draft preparation, D.P.R.; visualization, D.P.R.; supervision, I.M. and H. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest
The authors declare no conflict of interest.

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