**JOURNAL OF DEGRADED AND MINING LANDS MANAGEMENT**
Volume 11, Number 3 (April 2024):6009-6015, doi:10.15243/jdmlm.2024.113.6009
ISSN: 2339-076X (p); 2502-2458 (e), www.jdmlm.ub.ac.id

**Research Article**

# Influence of limestone as pre-treatment for sustainable acid mine drainage water quality management in Tanzania

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*Article history:*
Received 23 January 2024
Revised 7 March 2024
Accepted 14 March 2024

*Key words:*
acid mine drainage
heavy metals
limestone
sulphate

## Abstract

Acid Mine Drainage (AMD) is a major environmental problem due to low pH, high sulphate content, and dissolved metals. The aim of this study was to investigate the potential of limestone materials for the primary treatment of AMD water. Different doses of limestone were used in this study in the ratio of 0 g/1,000 mL as control, 20 g/1,000 mL, 40 g/1,000 mL, 60 g/1,000 mL, 80 g/1,000 mL, 100 g/1,000 mL, 200 g/100 mL (limestone/AMD) in a batch experiment of 1,500 mL plastic container. The results showed that the pH of AMD water improved to 8.6 at a dose of 100 g/1,000 mL, and the sulphate concentration decreased from 2,277 mg/L to 506 mg/L at a dose of 200 g/1,000 mL. The removal of Fe, Cu, Zn, Mn, and Ni at 200 g/1,000 mL was 99%, 92%, 68%, 96%, and 99%, respectively. The results of this study also showed that the Mn content decreased slowly as the limestone dosage increased. In addition, this study showed that limestone is a good neutralizing agent for increasing the pH of AMD water and reducing sulphate and dissolved metal concentrations. The results of this study indicated that limestone is suitable as a primary treatment agent for AMD water treatment in gold mines in Tanzania.

**To cite this article:** Marwa, A. and Sweya, L.N. 2024. Influence of limestone as pre-treatment for sustainable acid mine drainage water quality management in Tanzania. Journal of Degraded and Mining Lands Management 11(3):6009-6015, doi:10.15243/jdmlm.2024.113.6009.

## Introduction

Environmental problems related to mine wastes, especially waste rocks and mine drainage, continue to increase (Cole et al., 2001; Tabelin et al., 2021). One of the emissions recognized as serious in mining is AMD from mining, the quality of which contains dangerous heavy metals with a higher sulphate content (Park et al., 2019). Most sources of AMD are abandoned mines, and this situation can continue for years if appropriate control and prevention measures are not implemented (Rezaie and Anderson, 2020).

AMD water can develop under a variety of conditions, including exposure to sulphide minerals in the presence of oxygen and water, as well as other factors such as moisture levels, pH, and the chemical activity of iron (Aggarwal, 2017). A variety of techniques and approaches are available to treat AMD, including chemical treatments and biological treatment. However, most of these technologies are more maintenance-intensive and require ongoing operational costs (Doshi, 2006). Other researchers, Turingan et al. (2022) reported that the use of lime as an active treatment for AMD required continuous monitoring, which does not seem to be feasible in the long term since AMD can persist long after mine cessation. However, the use of limestone can replace other limes because it is very cheap compared to lime (Maree et al., 1992).

The use of limestone can be alternative and sustainable in the long term because the materials are cheap and locally available (Hedin et al., 1994; Jage et al., 2001; Lopez-Fernandez et al., 2003; Turingan et al., 2022). Using limestone reduces reagent costs, clarifier retention time and sludge compared to other treatment methods (Dempsey and Jeon, 2001; Sibrell et al., 2003). The use of limestone can be one solution for sulphate sorption, especially in neutral mine water, simply because the available calcium ions can bind sulfate ions (Silva et al., 2012). Using limestone, the removal efficiency was 100% Cu, 47.8% Ni, 36.8% Zn, and the optimum pH was 8.9-9.1(Ya et al., 2009). In addition, a study by Fuchida et al. (2020) reported that an increase in pH after using limestone resulted in precipitation of other metals (Cu, Zn, and Pb) as hydroxide/or carbonate. In other studies, Turingan et al. (2022) reported that the best result was observed when limestone was used; the pH increased from 1.35 to 8.08, and the metal content decreased by 39% (Fe), 94% (Ni) and 52% sulphate. In addition, sulphate removal from mine drainage occurs through a process called gypsum precipitation, which involves the use of limestone (INAP, 2003). However, this method is only effective when the pH levels are alkaline (Ferreira et al., 2011). Limestone application can be one of the best options for sulphate removal in neutral mine water because available calcium ions can bind to sulphate ions (Silva et al., 2012). The authors reported that sulphate was reduced from 588 mg/L to 87 mg/L in a continuous stirred tank test with limestone. However, a study published by Stumm and Morgan (2017) showed that the pH of limestone dissolution cannot be sufficient to precipitate chemical divalent metals such as Zn, Mn and Ni. In the limestone process, the rate of $CaCO_3$ neutralization depends on the $CaCO_3$ dose (Maree et al., 1992). Limestone can be used as an alternative for the removal of sulphate in mine waters having 1,200-2,000 mg/L (Silva et al., 2012). Brahaita et al. (2017) and Davis et al. (2000) reported in their study that limestone is composed of $CaCO_3$ when used as a means for AMD treatment due to its cheaper and can also reduce heavy metals, but the materials cannot treat AMD with high iron content because they can scale and prevent the solubility process (Skousen et al., 2017).

A study by Turingan et al. (2022) suggested that future research is recommended to evaluate the ability of limestone and how much AMD can be treated before treatment materials achieve breakthroughs. The aim of this study was to investigate the possible use of limestone as a locally available material for pre-treatment of AMD with different doses of limestone. The specific objective of this study was to determine the effectiveness of different limestone ratios as a pre-treatment for AMD, as well as the effect of retention time on treatment efficacy.

## Materials and Methods

### *Materials*

Limestone used in the experiment was obtained from Athi River Mining Limited in Kange, Tanga region of Tanzania. It was crushed into powder form, sieved and classified into size fractions at the African Minerals and Geosciences Centre. Its composition is shown in Table 1. Limestone crushing and sieving were carried out to ensure an increase in surface area for contact time (Jha et al., 2015). The fresh AMD was obtained from North Mara Gold Mine (1°28.416'S and 34°30.992'E), Tarime, Tanzania. The AMD contained various chemical compositions, as shown in Table 2.

Table 1. Characteristics of limestone used in the experiment.

| Parameter | (% w/w) |
|---|---|
| CaO | 57.16 |
| $SiO_2$ | 1.28 |
| MgO | 0.16 |
| $Fe_2O_3$ | 0.16 |
| $K_2O$ | 0.13 |
| $CeO_2$ | 0.11 |
| $V_2O_5$ | 0.05 |
| CuO | 0.06 |
| $TiO_2$ | 0.10 |
| $SO_3$ | 0.06 |
| SrO | 0.03 |
| LOI | 40.71 |

Table 2. Characteristics of actual AMD used for experimental testing.

| Parameter | Acid Mine Drainage |
|---|---|
| pH | 3.1 |
| Ec (µs/cm) | 2,950 |
| Sulphate (mg/L) | 2,277 |
| Copper (mg/L) | 0.49 |
| Iron (mg/L) | 2.3 |
| Manganese (mg/L) | 55 |
| Nickel (mg/L) | 2.5 |
| Zinc (mg/L) | 7.7 |

### *Limestone and AMD dosing*

A batch experiment using limestone to treat AMD used different weight ratios of limestone: 20 g, 40 g, 60 g, 80 g, 100 g and 200 g. The dose of limestone was added to 1,000 mL of fresh AMD in a container of 1,500 mL. The ratios used in the experimental test (limestone: AMD) were as follows: 20 g/1,000 mL, 40 g/1,000 mL, 60 g/1,000 mL, 80 g/1,000 mL, 100 g/1,000 mL, and 200 g/1,000 mL. This study used an experimental design as adopted by Jha (2013).

### *Experimental set-up and design*

The experiment was done with manual mixing (stirring) because the limestone needs to dissolve properly to allow a good treatment of AMD. In this process, AMD was mixed in the first step and allowed to settle in the second step. The container volume used in the experimental set-up was 1,500 mL (for the settling and clarification tanks). In this experiment, doses of 0 g (without mixed with limestone), 20 g, 40 g, 60 g, 80 g, 100 g and 200 g of limestone were placed in six separate containers containing 1,000 mL of fresh AMD, and one container without limestone

(0 g) was used as a control. For the mixing process, the containers were stirred immediately after the addition of limestone for 10 minutes and then every six hours for 24 hours, after which the mixture was transferred to a separate container for settling and measurement.

### *Water sampling and analysis*

A batch experiment using limestone to treat AMD was conducted for 30 days. Water samples were taken once a week for physical-chemical analysis. All samples were collected and sent to the Environmental Engineering Laboratory of Ardhi University for analysis. Analysis was performed according to APHA methods (1998). pH and electrical conductivity (EC) were measured using a potentiometric method using Sension 378. Nickel, zinc and copper were analyzed using an AAnalyst 100 and PerkinElmer Instrument (Atomic Absorption Spectrometer). Iron was analyzed using the 1-1-Phananthroline Method (DR/4000U spectrophotometer. Sulphate was analyzed using the turbidimetric method with the DR/4000U spectrophotometer. Manganese was also analyzed using the HACH product of the periodate oxidation method, which was also performed with the spectrophotometer DR/4000U. The selection of heavy metals was based on the actual chemical compositions of AMD (Table 2).

### *Statistical analyses*

One-way ANOVA test was used for the statistically significant difference of F-limestone used in AMD treatment, considering $p<0.05$ as the statistically significant difference.

## Results and Discussion

### *Effectiveness of using limestone in acid mine drainage treatment*

The study used limestone as pre-treatment to raise the pH of acid mine water because many metals are found at low pH. This study was able to show how the pH of water can be increased, and the metal content of AMD can be reduced. The experiment using limestone was tested for 63 days at different doses. Table 2 shows the general quality of AMD water after treatment with different doses of limestone. AMD water pH improved from 3.1 to 8.6. AMD pH was 3.1 before limestone dosing, but after limestone dosing, it increased to 8.5 at 20 g/1,000 mL, 8.1 at 40 g/1,000 mL, 8.5 at 60 g/1,000 mL, 8.4 at 80 g/1,000 mL and 8.6 for 100 g /1,000 mL and to 7.9 at 200 g/1,000 mL. Figure 1 shows the change in pH in different usage ratios of limestone. The variation showed that the pH of most limestone doses increased from the initial pH until day 56 when the pH changes remained unchanged in all doses until the end of the experiment (Figure 1).

This study showed that AMD pH increased at all dose ratios (Figure 1), suggesting that limestone has a high potential to neutralize AMD. However, the lowest pH increase was observed at the dose of 200 g/1,000 mL, which is probably due to the precipitation and coating of the limestone grains. This result is consistent with Hammarstrom et al. (2003), who confirmed that when limestone was used to treat AMD, the pH of AMD water increased from 2.9 to above 7, but the pH dropped below 4 within two days. Hammarstrom et al. (2003) and Rotting et al. (2005) also emphasized the development of scale in the limestone grains, which slowed down the reaction and prevented the limestone from producing more alkali.

Other studies, Skousen et al. (2017) reported that scales developed in the treatment of and can prevent limestone dissolution processes. However, to reduce the scale problem, other researchers, Watten et al. (2005 and Rotting et al. (2008), reported the use of carbon dioxide to accelerate the dissolution of limestone in AMD treatment.

In Table 2, the level of manganese reduction at 200 g/1,000 mL after the 63-day experiment was low compared to other dosage ratios. Similarly, this study showed results that were observed with a dosage ratio of 200 g/100 mL, where the electrical conductivity increased compared to the other ratios. This is supported by Hammarstrom et al. (2003), who pointed out that electrical conductivity indicates the ions present in the sample. In this study, a high reduction of sulphate, Zn, Cu, Fe, Mn and Ni was observed at 200 g/1,000 mL (Table 3). A statistically significant difference tested by ANOVA indicated that limestone doses had a statistically significant $p<0.05$. This study showed that manganese was removed from 2277 mg/L to 506 mg/L at 200 g/1,000 mL, corresponding to approximately 78%. A study by Silva et al. (2012) reported a reduction of sulphate from 588 mg/L to 87 mg/L using limestone, which was about 85%.

Table 3. AMD performance results with different limestone doses after 63 days of experiment.

| Limestone Dosing | pH | Ec (µs/cm) | $SO_4$ (mg/L) | Mn (mg/L) | Zn (mg/L) | Cu (mg/L) | Ni (mg/L) | Fe (mg/L) |
|---|---|---|---|---|---|---|---|---|
| 0 g/1,000 mL | 3.1 | 2,430 | 2,277 | 55 | 7.7 | 0.49 | 7.7 | 2.3 |
| 20 g/1,000 mL | 8.5 | 2,087 | 754 | 1.4 | 3.5 | 0.07 | 1.3 | 0.009 |
| 40 g/1,000 mL | 8.1 | 3,009 | 702 | 0.7 | 3.5 | 0.08 | 0.9 | 0.01 |
| 60 g/1,000 ml | 8.5 | 2,093 | 678 | 1.3 | 3.2 | 0.05 | 0.2 | 0.006 |
| 80 g/1,000 mL | 8.4 | 2,085 | 670 | 1.2 | 3.2 | 0.06 | 0.3 | 0.01 |
| 100 g/1,000 mL | 8.6 | 2,088 | 641 | 0.9 | 3.1 | 0.04 | 0.2 | 0.01 |
| 200 g/1,000 mL | 7.9 | 3,001 | 506 | 2.3 | 2.5 | 0.04 | 0.005 | 0.02 |

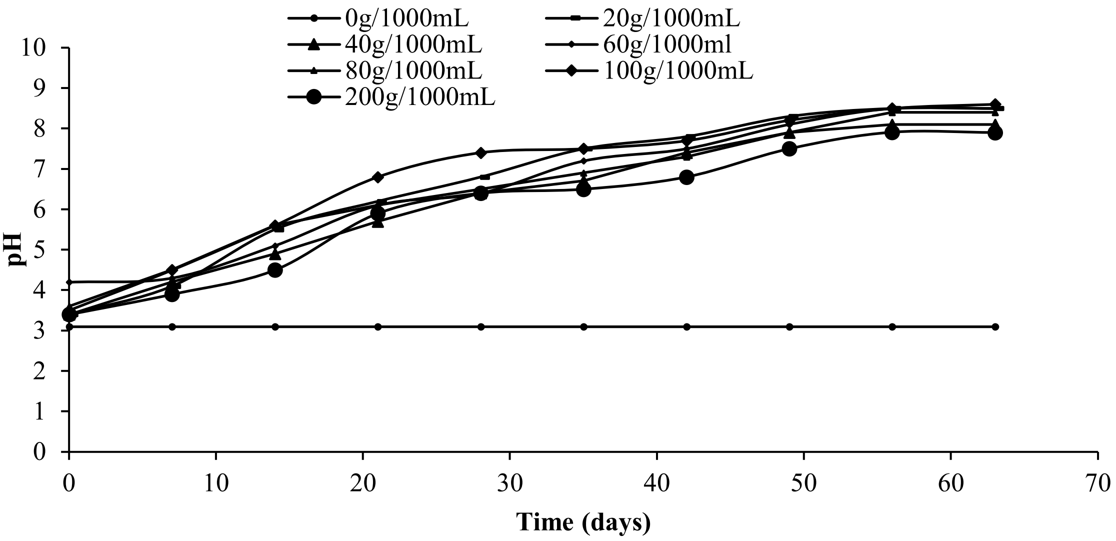
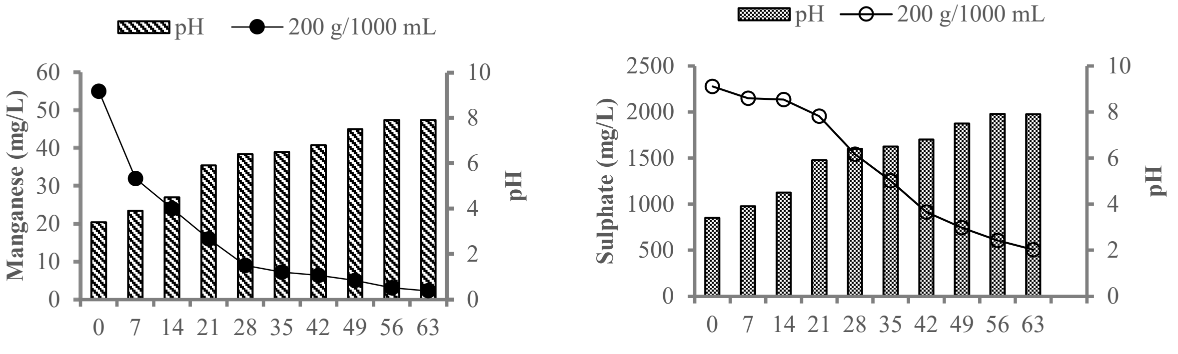


Figure 1. Variation of pH in batch experiments with different doses of limestone for the treatment of AMD.

***Effect of time on sulphate, manganese removal, and pH at optimal limestone dosage***

The effect of time on sulphate and manganese reduction is shown in Figure 2. Manganese reduction was observed from day 28 until the end of the experiment. The relationship between the removal of manganese and sulphate was confirmed by the increase in the pH of the system (Figure 2a and Figure 2b). At the end of the experiment, a manganese reduction of 2.3 mg/L was observed, which corresponds to a pH value of 7.9 at the optimal dosage ratio (200 g/1,000 mL). In other studies, Silva et al. (2012) reported that manganese concentration reached 0.8 mg/L at pH 8.8 and concluded that the efficiency of manganese removal depends on pH above 8.5. Other researchers, Tan et al. (2010) and Moodley et al. (2018) have examined the effects of limestone on the removal of manganese from AMD water. Their findings indicated that an increase in pH resulting from the use of limestone can significantly enhance the removal of manganese from AMD water.



(a) (b)

Figure 2. Effect of time on (a) sulphate, and (b) manganese removal and pH with optimal limestone dosage (200 g/1,000 m/L) in AMD treatment.

***Trends in sulphate, heavy metal concentration and efficiency in a batch experiment with different limestone dosages for AMD treatment***

In Table 3, at the optimal dose of 200 g/1,000 mL, sulfate removal was higher, reaching 78%. However, the iron removal efficiency observed was 99% removal at all dosage ratios, but the zinc removal efficiency was different, which was 68% at 200 g/1,000 mL. The highest concentrations of Fe, Zn and Mn were observed on day 20 of the studies (Figures 3b, 3c and 3d). This study showed that sulphate removal increased by 78% at a concentration of 200 g/1,000 mL (Table 3 and Figure 3a). A study by Jha (2013) found significant sulphate removal in AMD of up to 70% from an initial concentration of 10 g/500 mL of limestone usage. It also showed that increasing limestone resulted in more sulphate reduction in AMD (Table 3). However, in the study of Turingan et al. (2022), sulphate removal is 52% when used with limestone to treat AMD. In other studies, Silva et al. (2012) reported that sulphate sorption into limestone grains was the only way to reduce sulphate in mine water. In addition, this study showed that metals such as Cu, Ni, and Fe were reduced by 92%, 99%, and 99%, respectively, at the optimal dose of limestone.

Table 3. Sulphate and heavy metal removal in the treatment of AMD using different doses of limestone after 63 days of the experiment.

| Limestone Dosing | % Improved | | | | | |
|---|---|---|---|---|---|---|
| | $SO_4$ | Mn | Zn | Cu | Ni | Fe |
| 20 g/1,000 mL | 67 | 98 | 55 | 86 | 50 | 99 |
| 40 g/1,000 mL | 69 | 99 | 55 | 84 | 64 | 99 |
| 60 g/1,000 ml | 70 | 98 | 58 | 90 | 92 | 99 |
| 80 g/1,000 mL | 70 | 98 | 59 | 88 | 90 | 99 |
| 100 g/1,000 mL | 72 | 98 | 60 | 92 | 92 | 99 |
| 200 g/1,000 mL | 78 | 96 | 68 | 92 | 99 | 99 |

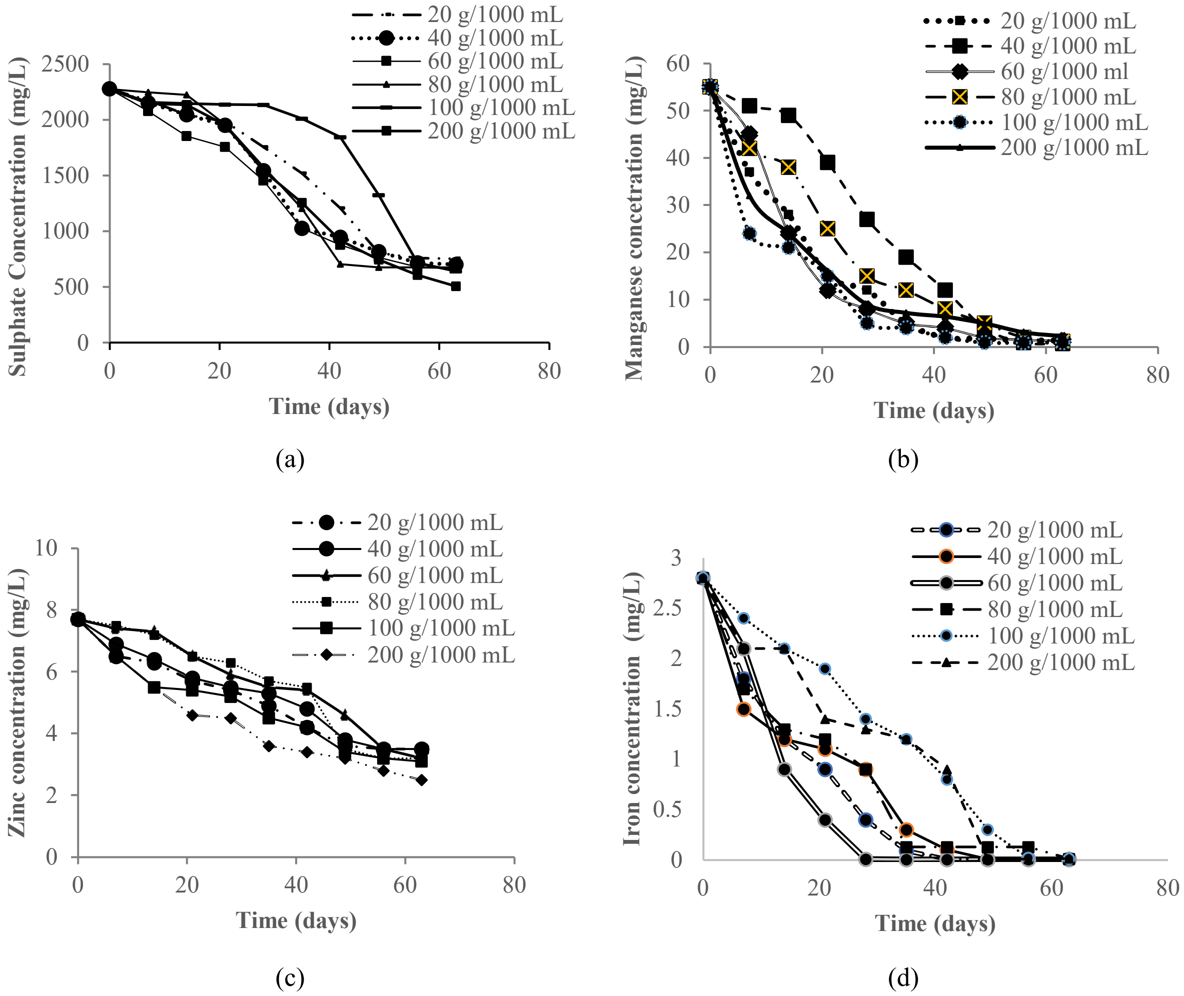


Figure 3. Effect of time on the removal of sulfate (a), manganese (b), zinc (c) and iron (d) in a batch experiment with different doses of limestone in AMD treatment.

Other researchers, Turingan et al. (2022), reported Cu, Ni, and Fe reductions of 99%, 63%, and 93%, respectively. Fuchida et al. (2020) reported that the increase in pH after applying limestone was due to the precipitation of other metals, such as Cu, Zn, and Pb, as hydroxides and/or carbonates. Ya et al. (2009)

reported the removal efficiency when the limestone used was 100% Cu, Ni (47.8%) and Zn (36.8%) at pH 9.1. Some researchers have reported that the removal of metals by limestone is contributed to by its sorption capabilities (Davis et al., 1987; Leppert, 1990; Pardas et al., 1994; Yao and Gao, 2007). However, in a study by Wu et al. (1999), it was also found that the exchange reaction on the surface of limestone can also influence the removal of metals.

## Conclusion

This study investigated the use of limestone as a pre-treatment for AMD water, and the objectives of this study were achieved. AMD water quality improved its pH, and most heavy metals decreased. In this study, it was found that an increased dose of limestone caused an increase in pH and precipitation of metals. A significant reduction of sulphate was observed at 200 g/1,000 mL from 2,277 mg/L to 506 mg/L with 78% removal. A limestone dosage of 100 g/1,000 mL produced the highest pH value for AMD water quality. This study showed promising results, showing that limestone can be used as a primary treatment for advanced AMD to raise pH levels and reduce metal concentrations. However, further research with other alkaline materials is emphasized to improve AMD water treatment.

### Acknowledgments

The authors would like to thank North Mara Gold Mine for the initial funding for this research. Also, thanks to Ardhi University at the School of Engineering and Environmental Studies for providing the necessary space for laboratory work.

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