



Journal of Environmental Science and Technology

ISSN 1994-7887

science
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Treatment for Acid Mine Drainage Utilizing Dimension Stone Waste

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ABSTRACT

When large quantities of rocks containing sulphide minerals are excavated from mines, they react with water and oxygen to create sulphuric acid. When this water reaches a certain level of acidity it also starts leaching traces of metals, this wastewater is termed as Acid Mine Drainage (AMD), affects the surface and subsurface hydrology and extends its impact to the surrounding land. Although, several AMD treatment techniques and methods exist, lime treatment is the most common approach which proves to be highly expensive. In this study dimension stone (Kotastone) waste, generated by the stone quarrying/cutting industries was used for the detoxification of AMD. The selection criteria of treatment agent was its free of cost availability. pH increased from 2.97 to 6.21, net acidity dropped from 1196.6 to 150 mg L⁻¹ of CaCO₃. Concentrations of metals like Fe²⁺, Cu²⁺, Zn²⁺, Ni²⁺, Co²⁺ and Mn²⁺ also reduced remarkably from 19.69 to 99.24%. The experiments were carried out in 500 mL cylinders with different waste doses ranging from 0.5 to 10 g. The cost comparison with hydrated lime and quick lime was also in favour of Kotastone waste for the AMD treatment. Now, major efforts are required by the mining industry and the research laboratories to convert this prototype product into commercial ventures so that this waste can become a utility material instead of nuisance.

Key words: Acid mine drainage, dimension stone waste, chemical treatment, kotastone waste, metal removal

INTRODUCTION

Acid Mine Drainage (AMD) is produced when iron sulfide-aggregated rocks minerals are exposed to oxygen and water. Although this process occurs naturally, mining can promote AMD generation simply through increasing the quantity of sulfides exposed (Skousen *et al.*, 1996). Naturally-occurring bacteria are also equally responsible for propagating AMD production by assisting in the breakdown of sulfide minerals. This water renders the soil and water bodies acidic by blocking sunlight and covering the streambed with a thick red blanket. Acid mine drainage becomes hazardous to the environment, affecting the surface and subsurface hydrology, thus disrupting the cycle of nature (Hanif *et al.*, 2005).

Three major mining projects located in the state of Rajasthan are having problem of AMD: Khetri Copper Project is an important mining cum metallurgical project, secondly Zawar group of mines of Hindustan Zinc Limited along with its processing plants located close to Udaipur city and thirdly Rampura Agucha project of Hindustan Zinc limited. Mine effluents containing bulk

quantity of toxic heavy metals from these projects, are being discharged in the surrounding environment affecting both the terrestrial and aquatic ecosystem (Farid *et al.*, 2002; Javed, 2005; Ekosse, 2008). These projects are carrying out active chemical treatment technology using lime/hydrated lime perpetually (Gangal and Zutshi, 1990; Gupta *et al.*, 1994).

Traditionally acid mine water is treated with various alkaline chemicals like limestone, lime, hydrated lime, soda ash, caustic soda and ammonia to increase the pH and concomitantly precipitate heavy metals. A characteristic of each chemical makes it more or less appropriate for a specific condition (Mudder *et al.*, 2005). Combination of lime and limestone was recommended by Maree *et al.* (1998) and Geldenhuis *et al.* (2001). Other agents investigated are blast furnace slag (Feng *et al.*, 2004) and fly ash (Petrik *et al.*, 2003). Feng *et al.* (2004) reported that the pH of acid mine water increased to neutral and most of the heavy metals were removed successfully by the use of slag as a potential sorbent. Petrik *et al.* (2003) and Potgieter-Vermaak *et al.* (2006) reported that the fly ash or fly ash leachate is effective alternative for neutralization of various sources of acid mine water, without the additional use of liming agents.

The dimension stone mining and processing waste generated at Kota (Rajasthan state) is quite substantial, being in the range of 1.5-2 million tones annually. It is a common practice to dump the waste in the form of slurry generated while finishing/polishing of Kotastone slabs on the barren government waste lands. As this waste cannot be used in construction or any other purpose; it not only degrades the quality of surrounding environment but also reduces the aesthetic value of such sites (Rajasthan, 2010).

Kotastone is a flaggy lime stone; its major constituents are nearly the same as lime stone except high percentage of silica content. If this waste can be used in the treatment of AMD, it will not only solve the problem of AMD treatment but also consume the waste, thus making the environment cleaner and safer to live in. On other hand it is cheap in comparison to other alkaline chemicals so the prime objective of this study was to examine a better and effective alternative for the AMD treatment at a relatively cheaper cost. In this study laboratory scale experiments were conducted to determine the feasibility and potential application of Kotastone waste for the treatment of acid drainage.

MATERIALS AND METHODS

Materials: An artificial solution, with a low pH and elevated metals concentrations, was mixed to simulate Acid Mine Drainage (AMD). The simulated AMD permit better control throughout the experiment as it is stable for longer period. The simulated AMD was stored in 50 L PVC vessel and its characteristics are presented. This study was conducted in the last week of the June 2010 (Table 1).

Methods

Following steps were adopted for the study: Acid mine drainage was simulated in the laboratory by mixing different chemicals in distilled water in a 50 L jar. This acidic water was measured and put in different bottles in pre-determined volume of 500 mL. Kotastone waste was accurately weighed ranging from 0.5 to 10 g and mixed in the acidic water kept in separate bottles. These sample mixtures were kept for 96 h for the chemical reactions to take place. Observations of pH, acidity, alkalinity and sulfate were taken after 15 min, 12, 24 and 96 h. Metals concentrations were observed after 96 h when the pH was almost stable.

Table 1: Characterization of simulated AMD for the experiment

Component	Concentration (mg L ⁻¹)	Source
Fe ²⁺	98.252	FeSO ₄ .7H ₂ O
Zn ²⁺	25.043	ZnSO ₄ .7H ₂ O
Cu ²⁺	26.546	CuSO ₄ .5H ₂ O
Ni ²⁺	17.369	NiSO ₄ .6H ₂ O
Co ²⁺	1.1920	CoSO ₄ .7H ₂ O
Pb ²⁺	0.1183	PbSO ₄
Mn ²⁺	33.344	MnSO ₄ .H ₂ O
SO ₄ ²⁻	869	-
pH	2.97	-

Instrumentation: For the examination of the performance of Kotastone to treat AMD, the following procedure was adopted. The pH of water sample was determined using pH meter (Make-RI, New Delhi, Model 151R). Buffer solutions of 4, 7 pH were used for calibration purpose. Acidity and alkalinity were measured using methods 2310 B, 2320 B titration methods as given in the standard methods for the examination of water and waste water (APHA, 1992; El-Fadaly *et al.*, 1999). An Atomic Absorption Spectrometer (Make-Thermo Electron Corporation, S series) was used to determine the metals ion concentration.

Chemicals used for treatment purpose: Kotastone waste was used for treating AMD. Different doses of fine grained Kotastone waste ranging from 0.5, 1, 2, 5 and 10 g were used in 500 mL polythene jars as treating agents. The chemical composition of Kotastone waste is given in Table 2.

Kotastone waste was analyzed at chemical laboratory of Rajasthan State Mines and Minerals Ltd, Jodhpur. Kotastone is a flaggy limestone found with low CaO and high SiO₂%.

Cost economics: For comparative cost analysis the commercial cost of hydrated lime was compared with the quick lime and kotastone waste (available at free of cost). The amount of base required for acid neutralization is dependent on the molecular weight of the neutralization compound Table 8.

$$\text{Acidity} \times \text{Volume} = \text{Total Acidity (\# moles CaCO}_3 \text{ equivalent acidity)}$$

Total molecular weight of 1000 amount of neutralizing:

$$\text{Acidity} \times \text{Neutralizing Compound} \div \text{Conversion Factor} = \text{Compound Needed}$$

(moles) (g mol⁻¹) (grams to kilograms) (kg)

Examples: 1 m³ (1000 L) tank with 1000 mg L⁻¹ acidity.

Moles of CaCO₃ equivalent acidity:

$$\left(\frac{1000 \text{ mg}}{\text{L}}\right) \left(\frac{1000 \text{ L}}{1 \text{ m}^3}\right) \left(\frac{1 \text{ g}}{10^3 \text{ mg}}\right) \left(\frac{1 \text{ mol CaCO}_3}{100 \text{ g}}\right) (1 \text{ m}^3) = 10 \text{ moles}$$

Table 2: Chemical composition of Kotastone waste

Minerals	% in Kotastone waste
SiO ₂	37.08
CaO	28.00
MgO	1.21
Al ₂ O ₃	1.89
Fe ₂ O ₃	1.01
LOI	23.86

Neutralization amounts:

- Hydrated lime:

$$10 \text{ mol equiv. CaCO}_3 \left(\frac{74 \text{ g}}{1 \text{ mol Ca (OH)}_2} \right) \left(\frac{1 \text{ kg}}{10^3 \text{ g}} \right) \left(\frac{100}{96.57} \right) = 0.77 \text{ kg Hydrated Lime}$$

- Quicklime:

$$10 \text{ mol equiv. CaCO}_3 \left(\frac{56 \text{ g}}{1 \text{ mol CaO}} \right) \left(\frac{1 \text{ kg}}{10^3 \text{ g}} \right) \left(\frac{100}{76.63} \right) = 0.73 \text{ kg Quicklime}$$

- Kotastone waste:

$$10 \text{ mol equiv. CaCO}_3 \left(\frac{100 \text{ g}}{1 \text{ mol CaCO}_3} \right) \left(\frac{1 \text{ kg}}{10^3 \text{ g}} \right) \times \left(\frac{100}{50} \right) = 2.00 \text{ kg Kotastone waste}$$

Cost of chemical per 10 mol acidity removal (assuming Transportation cost 700 Rs./tones):

- Hydrated lime:

$$0.77 \text{ kg} \times (2.9 + 0.7) = \frac{2.77 \text{ Rs.}}{10 \text{ mol acidity}}$$

- Quicklime:

$$0.73 \text{ kg} \times (2.4 + 0.7) = \frac{2.26 \text{ Rs.}}{10 \text{ mol acidity}}$$

- Kotastone waste:

$$2.00 \text{ kg} \times 0.7 = \frac{1.40 \text{ Rs.}}{10 \text{ mol acidity}}$$

RESULTS AND DISCUSSION

The efficiency of neutralizing agent depends on several factors; usually cost and health considerations limit the choice. Other selection criteria of neutralizing agents include reaction rate, sludge production and disposal, safety and ease of handling, total cost and the effect of an over-dosage.

Table 3: pH change with different doses of Kotastone waste

Retention time	1.0	2.0	4.0	10.0	20.0
	-----(g L^{-1})-----				
Initial	2.97	2.97	2.97	2.97	2.97
15 min	3.02	3.19	3.82	4.26	4.44
12 h	3.85	4.40	5.18	6.00	6.01
24 h	4.07	4.55	4.87	5.77	6.07
96 h	4.08	4.38	5.25	5.93	6.21

Retention time and pH: The increases in the pH of AMD with different doses are given in Table 3. It was observed that during initial period of treatment pH increased rapidly and after 12 h it stabilized and further increase in the retention time does not cause any significant increase in pH. It was observed that a minimum dose of 1 g L^{-1} with 12 h contact time was required to reach a pH of 3.85. Larger doses of Kotastone waste ensured an increase in pH with similar retention time. After 12 h retention time pH reaches 4.4 with 2 g L^{-1} , 5.18 with 4 g L^{-1} , 6 with 10 g L^{-1} and 6.01 with 20 g L^{-1} . pH with different doses of Kotastone waste in 12 h retention time reaches almost near to its final values 4.08, 4.38, 5.25, 5.93, 6.21 for 96 h retention time. The pH changes significantly from 2.97 to 5.93 and 6.21 with doses quantity 10 and 20 g L^{-1} . The results obtained for are in agreement with those reported by Maree *et al.* (1998) for limestone.

Metals removal: The amounts of residual metal concentration were investigated by reporting the Fe^{2+} , Cu^{2+} , Zn^{2+} , Ni^{2+} , Co^{2+} , Mn^{2+} ion concentrations before and after treatment with the different quantity of dosing agent. Concentrations of metals are dropped up to 0.201, 11.205, 61.306, 26.771, 0.842 and 13.242 mg L^{-1} for Cu^{2+} , Zn^{2+} , Fe^{2+} , Mn^{2+} , Co^{2+} and Ni^{2+} , respectively (Table 4). The oxidation of ferrous ions results in formation of ferric ions in acid mine water, imparting a yellow-brown colour to the solution. Ferric ions are removed as a ferric hydroxide precipitate in the presence of hydroxide, carbonate or oxide containing materials, up to a pH of minimum solubility (pH value about 4.0). Ferric ions are relatively easy to remove from acid mine water because they start to precipitate at a pH as low as 3.0. This might have been caused by the fact that ferric hydroxide precipitate at a lower pH range (Bhattacharya *et al.*, 2008; Ji *et al.*, 1997; Maree *et al.*, 1992). The percentage removal of Fe^{2+} ranging from 1.57, 10.51, 23.44, 36.61 and 37.6 were achieved with 1, 2, 4, 10, 20 g L^{-1} doses, respectively (Table 5).

Manganese hydroxide precipitation is variable due to its many oxidation states but will generally precipitate at a pH of 9.0 to 9.5. However, a solution pH of 10.5 is sometimes necessary for complete removal of Mn^{2+} in some cases, complete Mn^{2+} removal is very difficult to attain. This high pH for Mn^{2+} removal can cause Al^{3+} to enter solution again. For waters with high Mn^{2+} and Al^{3+} , a two phase treatment system may be required. As per this discussion, the appropriate chemical treatment may depend on both the oxidation state and concentrations of metals in the AMD (Kalin *et al.*, 2006; USEPA, 1994).

Interactions among metals also influence the rate and degree to which these metals precipitate. In the present investigation Mn^{2+} removal reaches maximum 19.69% with 20 g L^{-1} dose; show that Mn^{2+} is not removed effectively with Kotastone waste. This removal is due to co-precipitation but only because the Fe^{2+} concentration in water is much greater than Mn^{2+} content (about 4 times more or greater). For example, ferric hydroxide precipitation will largely remove Mn^{2+} from the water at pH 8 due to co-precipitation but only if the Fe concentration in the water is much greater than the Mn^{2+} content (about 4 times more or greater). If the Fe concentration in the AMD is less than

Table 4: Metal concentrations (mg L⁻¹) with different doses of Kotastone waste (After 96 h retention)

Doses (g L ⁻¹)	Copper	Zinc	Iron	Manganese	Cobalt	Nickel
AMD	26.546	25.043	98.252	33.344	1.192	17.369
1.0	22.911	24.876	96.706	33.142	1.011	15.330
2.0	7.755	22.131	87.920	32.477	0.981	15.065
4.0	0.240	16.135	75.219	32.188	0.890	13.962
10.0	0.211	11.421	62.274	29.466	0.863	13.539
20.0	0.201	11.205	61.306	26.777	0.842	13.423

Table 5: Percentage of metal removal with different doses of Kotastone waste (After 96 h retention)

Doses (g L ⁻¹)	% Cu removal	% Zn removal	% Fe removal	% Mn removal	% Co removal	% Ni removal
1.0	13.69	0.66	1.57	0.60	15.14	11.73
2.0	70.78	11.62	10.51	2.60	17.65	13.26
4.0	99.09	35.57	23.44	3.46	25.29	19.61
10.0	99.20	54.39	36.61	11.36	27.55	22.05
20.0	99.24	55.25	37.60	19.69	29.31	22.72

four times the Mn²⁺ content, Mn²⁺ may not be removed by co-precipitation and a solution pH of 9 is necessary to remove the Mn²⁺. AMD contains multiple combinations of acidity and metals and each AMD is unique and also its treatment by these chemicals varies widely from site to site (Skousen *et al.*, 1990, 1993, 1996).

Metal oxides, hydroxides and oxy-hydroxides, especially ferric iron and aluminum hydroxides can scavenge trace metals such as Cu²⁺, Ni²⁺, Co²⁺ and Zn²⁺ from the free water column in a process called co-precipitation (Ahmad and Afzal *et al.*, 2001; Nkegbe, 2005). During co-precipitation trace metal cations adsorb to negatively charged surface sites of metal hydroxide and oxy-hydroxide precipitates (Al-Jlil, 2010). Since co-precipitation usually occurs on metal hydroxides and oxy-hydroxides, co-precipitation primarily occurs under aerobic conditions.

Cu²⁺ removal was very significant, almost 100% with doses of more than 4 g L⁻¹ of kotastone waste due to co-precipitation and low metal solubility of copper in the range of pH 5. Different doses of, 2, 4, 10, 20 g L⁻¹ of kotastone waste successfully removed copper up to 13.69, 70.78, 99.09, 99.2 and 99.24%, respectively. Other trace metals such as Ni²⁺, Co²⁺ and Zn²⁺ were removed less effectively than Cu²⁺ due to their higher metal solubility at low pH. Zn²⁺ was removed from 0.66 to 55.25% with 1 to 20 g L⁻¹ doses. Cobalt removal was from 15.14 to 29.31%, Nickel removal was 11.73 to 22.72% from various doses of Kotastone waste in increasing order (Table 5). The lower removal percentage was possibly due to co-precipitation only, because their metal solubility at pH 6.21 is almost very high (Table 3).

In addition to the removal of dissolved metals through hydrolysis reactions and co-precipitation, metals may precipitate as metal carbonate complexes. Carbonates are present in acidic drainage from the dissolution of Kotastone which are, either added to acidic drainage waters during passive pre-treatment methods or during treatment. Metal carbonates generally require higher pH values to precipitate out of solution than metal hydroxides.

Net acidity removal: In study to neutralize acid mine drainage, total acidity of the water body must be calculated. First, acidity and alkalinity is determined from acid-base titrations of water samples collected from an acid drainage affected water body (Langmuir and Whittemore, 1971). Then, net acidity of the water sample is calculated from difference of acidity and alkalinity. Initial

Table 6: Net acidity (mg L^{-1}) with different doses of Kotastone waste (Initial net acidity 1196.6 mg L^{-1} of CaCO_3)

Retention time	1	2	4	10	20
15 min	945.7	729.4	560	478.8	458.8
12 h	868.5	665.3	498.5	362.3	322.6
24 h	810.6	607.4	363.4	304.4	207.9
96 h	723.2	451.9	228.3	169.3	150

Table 7: Percentage acidity removals with different chemical doses of Kotastone waste (initial net acidity 1196.6 mg L^{-1} of CaCO_3)

Retention time	1	2	4	10	20
15 min	20.96	39.04	53.20	59.99	61.66
12 h	27.42	44.40	58.34	69.72	73.04
24 h	32.26	49.24	69.63	74.56	82.62
96 h	39.56	62.23	80.92	85.85	87.46

Table 8: Neutralizing compounds and their cost comparison

Neutralizing compound	Chemical formula	Molecular weight (mol g^{-1})	% of CaO	Efficiency (Based on % of CaO)	Commercial cost Rs/Tones	Cost (10 molL acidityremoval)
Gotan hydrated lime	Ca(OH)_2	74	73.08	96.57	2900	Rs 2.77
Gotan quicklime	CaO	56	75.32	76.63	2400	Rs 2.26
Kotastone	CaCO_3	100	28.00	50.00	-	Rs 1.40

(Assuming transportation cost 700 Rs./tones) up to 500 km

net acidity was 1196.6 mg L^{-1} of CaCO_3 . Removal efficiency of acidity was calculated for various doses with different retention time Net acidity was removed from 20.96 to 61.66% within 15 min of treatment by respective chemical doses. After 96 h retention time net acidity was removed from 39.56 to 87.46% from 1.0 to 20.0 g L^{-1} doses, respectively (Table 7). Net acidity remains 723.2, 451.9, 228.3, 169.3, 150 mg L^{-1} of CaCO_3 with increased doses of kotastone waste, although the acidity removal rates decreased with increased quantity of doses of kotastone waste (Table 6). This is possibly due to incomplete neutralization of greater doses up to 96 h. This statement is also supported by the chemical composition of kotastone waste which shows the presence of 28% CaO. Kotastone waste was 50% efficient comparative to pure limestone.

Cost analysis: Cost comparison of Kotastone waste with quick lime and hydrated lime clearly show that chemical cost of Kotastone waste including transportation cost for 500 km is more economic than above mentioned conventional chemicals. Kotastone waste is an alternative pre-treating agents for treatment of AMD. It is also cost effective for treating for mining projects located in the state of Rajasthan having a severe problem of AMD: Khetri Copper Project, Zawar group of mines of Hindustan Zinc Limited along with its processing plants located close to Udaipur city in Rajasthan and thirdly Rampura Agucha project of Hindustan Zinc limited. Metals removal percentage also appreciate the use of kotastone waste which remove copper 100% and Zn more than 55%.

Skousen *et al.* (1996) reported the neutralization efficiency of hydrated lime to be 90% and each chemical has its own characteristics that make it more or less appropriate for a specific condition. The best choice among alternatives depends on both technical and economic factors.

The technical factors include the quality and quantity of acid mine drainage and the desired final water quality. The economic factors include prices of reagents and transportation cost.

The experimental data reveals that addition of Kotastone waste to the AMD increased its pH around 6.21 with the neutralization efficiency 50%.

The basic concept of this study is to treat a waste by another waste or to resolve one of the environmental problems being faced by the mining sector (Hegazy *et al.*, 2007). This method can be employed to treat the acidic effluents effectively and as well as solving the problem of waste disposal. The maximum pollutant removal is attained within 12 h of the contact time. The comparative cost analysis displayed no economic constraints rather utilization of one industrial pollutant to treat another metal pollutant and proved to be highly effective.

CONCLUSIONS

From the laboratory study, it is concluded that the treatment of AMD is achieved through application of acid neutralizing capacity of kotastone waste. Kotastone waste being freely available, only transportation charges are required which proved to be much more economic than conventional chemicals (lime, hydrated lime). Further research may help mining industries facing the problem of AMD to reap the benefits of using waste to treat another waste.

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