



Research article

Acid mine water treatment using neutralizer with adsorbent material

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Abstract. One of the biggest issues in the mining sector is due to acid mine drainage, especially in those abandoned mining operations and active ones that fail to adequately control the quality of their water discharge. The removal degree of copper, iron, lead, and zinc dissolved metals in acid mine drainage was investigated by applying different proportions of mixtures based on neutralizing reagent hydrated lime at 67 % calcium oxide (CaO), with adsorbent material – natural sodium bentonite, compared to the application of neutralizing reagent without mixing, commonly used in the neutralization of acid mining drainage. The obtained results show that the removal degree of dissolved metals in acid mine drainage when treated with a mixture of neutralizing reagent and adsorbent material in a certain proportion, reaches discharge quality, complying with the environmental standard (Maximum Permissible Limit), at a lower pH than when neutralizing material is applied without mixing, registering a net decrease in the consumption unit of neutralizing agent express on 1 kg/m³ of acid mine drainage. Furthermore, the sludge produced in the treatment with a mixture of the neutralizing reagent with adsorbent material has better characteristics than common sludge without bentonite, since it is more suitable for use as cover material, reducing the surface infiltration degree of water into the applied deposit.

Keywords: bentonite; acid mine drainage; adsorption of metals; water treatment; cover material

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Introduction. The most ecologically harming effluents are those that come from mine discharges with low pH and elevated metal concentrations [1-3]. These conditions lead to ground degradation and hydrology system modification [4, 5], which plays a main role in life development where ecosystems and people are strongly affected by its shortage [6, 7].

In that sense, mining companies face a relevant challenge to run and raise their metal or energy production under a sustainable approach involving economic, social, and environmental spheres [8, 9]. To do so, lead with mine drainage, which mainly is divided into three types: basic, neutral, and acid. The main concern is linked to the acid ones, called acid mine drainage (AMD) due to its characteristic of a less than 3 pH and high concentration of heavy metals [10, 11] Furthermore, as [12, 13] states, AMD impacts can be classified into four groups: ecological, biological, physical, and chemical.

The treatment of AMD is mostly divided into passive or active technologies [14], the latter is commonly applied for effective and rapid toxic material removal reducing environmental impacts due to legal requirements [15], and where materials beneficiation-valorization under a circular economy understanding is trending [16, 17]. Nevertheless, a huge amount of sludge production is inevitable and management could represent a barrier in the active application process [18].



AMD treatment strategy must be based on its flow rate, chemistry, logistic capacity, and economic sources as well as legal discharge requirements and receiving body characteristics [19, 20]. Subsequently, AMD neutralization and element immobilizations are quite crucial [21].

There is evidence that companies have been using lime or unactivated attapulgite for AMD neutralization, heavy metal removal [22, 23] and secondary minerals formations in those affected lands by AMD [24].

It is also well documented by [25, 26] that nowadays, it is also important to incentivize production methods to reduce AMD formation at the source and predict environmental harm under a preventive approach, since AMD generation can passively continue over the years [27, 28] and that mining companies should not rely on single technologies to achieve environmental standards [29].

This research aims, despite obtaining good results in the neutralization and metals removal in acid mine drainage, which is measured by the quality of the produced effluent, to adapt the quality of the solid by-product known as neutralization sludge and, to improve its use properties as cover material for tailings and waste dumps, decreasing mine closure costs and environmental liabilities.

In the treatment of AMD, it is very common to use the neutralization process – alkaline precipitation followed by oxidation and sludge separation from the effluent, discharging treated water that reaches the environmental standard. The progress and optimization of the process are based on the sludge recirculation to take advantage of its remaining alkalinity and to increase the sludge sedimentation rate produced with the technology called “High-density sludge” and by the improvements in the flow neutralizing quality that depends on the lime hydration conditions to have the maximum proportion of hydroxyl ions free. Also, equipment works better at homogenizing flows, minimizing “dead zones” and reactor flow short circuits.

The applied concept in the study provides the neutralization process and alkaline precipitation an additional adsorption effect of dissolved metals using an adsorbent material that does not interfere with the main process. It also contributes to the removal of dissolved metals by fixing them on its surface, so that stoichiometrically, the requirement for hydroxyl ions is reduced, in other words, the consumption of lime should be lower.

Bentonite is a rock composed of crystalline clay-like minerals formed by devitrification. The chemical alterations that accompany its glassy igneous material, usually a tuff or volcanic ash often contain variable proportions of accessory glass beads that were originally phenocrysts in volcanic glass. Nevertheless, commercially, bentonites are defined exclusively on a mineralogical basis and they are generally classified according to their cations interlayers and their corresponding capacity for a swell in water as sodium, calcium, or potassium. The study applied sodium bentonite.

Many studies on the simple adsorption of ions in bentonite [30] investigated the capacity of activated bentonite as an adsorbent to remove components under experimental conditions [31] evaluated the adsorption relation of norfloxacin (NOR) and copper Cu^{2+} on bentonite compound elements. The binary adsorption of Cd(II) – Ni(II) on bentonite revealed that both metals presented a very strong antagonism similar to the adsorption of another metal [32]. On the other hand, information on simultaneous adsorption on bentonite multi-component systems or other mineral clay is very rare.

According to [33], using natural bentonite, Cd(II) , Cu(II) , Ni(II) , and Pb(II) ions were removed in systems of one and multiple components under various conditions, with maximum adsorption at 20 min. The pH rise favored the removal of metallic ions. The heavy metal adsorption followed the Langmuir isotherm in both systems of one and multiple components, indicating that Sorption mechanisms do not change under competence conditions. Due to strong antagonism between cations, the solution cation's coexistence reduced adsorption capacity compared to the individually obtained with single metal systems. The adsorption order selectivity was: $\text{Cu} > \text{Ni} > \text{Pb} > \text{Cd}$.



Also, [34] investigated the adsorption of Fe(II) acid drainage from abandoned mine coal in Enugo Okpara – Nigeria, using clay from bentonite. The study was based on an initial concentration of 1308, 62, 49 and 24 mg/l of Fe(II), Cu(II), Zn, and Pb(II), respectively in a 100 ml bottle. In addition, incremented bentonite doses were added from 0 up to 7 g one by one stabilizing the solutions at a constant pH of 2.7 finding that the biggest adsorption occurs under a dose of 4 g in 100 ml. The metal concentration at the end of the process with orbital shaking at 200 rpm for 4 h was 78.50, 0.075 and 0.053 mg/l of Fe(II), Cu(II), and Zn(II), respectively.

Comparably, [35] states about different activations of natural bentonite, including acid activation showing that bentonite compounds have a high potential for pollutant removal in mine wastewater. Linked to acid activation, when the acid concentration is more than 30 %, the bentonite-specific surface area will not increase significantly and bentonite sodium acidification requires a lower concentration than calcium bentonite. As can be seen, there is scientific evidence that sodium bentonite is an abundant and relatively cheap material that can be used in the treatment of AMD.

The main objective of the current study is to reduce the neutralizing reagent consumption, and improve the produced sludge quality by neutralization, making it more suitable for later use as cover material for tailings dumps, clearing, or liability closure state in mining activity.

Different mixing ratios of neutralizing reagent with adsorbent material were applied to identify the most effective, both in metal removal and sludge quality. The investigation is complemented by a sludge waterproofing test as a cover material on the exposed material surface.

Furthermore, the current study is located in Cerro de Pasco, Pasco, Peru at 4300 meters above sea level, and produced by mine dumps with accumulated pyrite belonging to a closed mine after its exploitation phase in an open pit way.

Methods. *Hydrated lime and sodium bentonite.* Hydrated lime was used as neutralizing reagent (NR), sampled from an acid mine drainage neutralization industrial plant which, analyzed in the laboratory results with 66.73 % calcium oxide (CaO), 1.8 % moisture and granulometry of 96 % – 200 mesh.

Bentonite was used as adsorbent material (BE), natural sodium whose chemical name is a silicate of hydrated aluminum – high montmorillonite content white-cream colored powder contained in bags of 25 kg; 94-98 % bentonite, and quartz or silicon oxide of 2-6 % with 2.52 specific gravity and 881 kg/m³ at 20 °C relative density.

The adsorption capacity of soluble metals, waterproofing, and sealing are the most relevant properties of bentonite for this study.

Acid Mine Drainage. The sample collected from acid mine drainage of 25 ± 0.25 l, was taken following the acid drainage treatment plant sampling procedure, from where the aliquots were taken for the experimental tests: pH (*in situ*) 1.91-1.96; Cu 14.94 mg/l; Fe 836.75 mg/l; Pb 0.32 mg/l; Zn 60.72 mg/l.

Reagent Preparation. As a solid reagent for neutralization and precipitation tests of metals, three powdered mixtures of hydrated lime with bentonite in different proportions were prepared evidenced in Table 1.

Then, 150 g was mixed in 1 l of distilled water for each case. In addition, hydrated lime was prepared by mixing 150 g of it in 1 l of distilled water.

Experimental Tests of Neutralization and Precipitation. The experimental tests were carried out in jar-test equipment of six jars, with a two liter capacity for each one, with adjustable speed agitators and time control. Due to sanitary restrictions, the tests were performed in an isolated environment inside the laboratory located in the environmental operations unit of the Universidad Continental in Huancayo.



Table 1

Mixtures characteristics, %

Mixture	Hydrated lime (NR)	Bentonite (BE)
M1	95	5
M2	90	10
M3	80	20

Table 2

Reagents consumption in the neutralization and precipitation test, g/l

Reagent	Mix	NR	BE
NR 100 %	6.45	6.45	0.00
NR 95 % – BE 5 %	6.35	6.03	0.32
NR 90 % – BE 10 %	6.04	5.44	0.60
NR 80 % – BE 20 %	7.12	5.70	1.42

The used test standard was 1 l of the acid drain, agitator at 300 rpm, a reaction time of 20 min, and the addition of reagent with a graduated milliliter syringe. During the test, the reagent was contained in a beaker with a magnetic stirrer to keep the mixture homogeneous.

Table 2 shows the reagent dosages used in the four neutralization and precipitation tests. The reagents were fed as a solid-liquid mixture.

Additionally, the obtained sludge in the neutralization tests for each dosage with five repetitions was collected and stored in four containers separately, one for each dosage.

Analytical methods. The metals in liquid solutions were analyzed by the following methods: copper (Cu) – SMEWW-APHA-AWWA-WEF. Part 3500-Cu B, 23rd Ed. 2017; zinc (Zn) –

SMEWW-APHA-AWWA-WEF. Part 3500-Zn B, 23rd Ed. 2017; iron (Fe) – SMEWW-APHA-AWWA-WEF. Part 3500-Fe B, 23rd Ed. 2017; lead (Pb) – SMEWW-APHA-AWWA-WEF. Part 5210-Pb B, 23rd Ed. 2017.

Experimental coverage tests. The four percolation columns were prepared using 4-inch (10.16 cm) internal diameter PVC pipes, a padding height of 50 cm ± 0.5 cm. A 4-inch PVC cover was placed at the bottom with 1/8 inch drilled from inside to outside to collect percolating liquid by gravity. At the bottom of each column, transparent containers were placed to collect the percolated. Also, the lateral parts were covered with transparent polyethylene to minimize losses by evaporation.

The sludge from each of the four test groups was combined due to its origin similarity, high molecular weight soluble polymer flocculant was added and shaken for 10 min at 60 rpm, then by separating the supernatant liquid obtained a stable thickened sludge whose content of solids is detailed in Table 3.

Table 3

Characteristics of the test group

Test group	Sludge volume, ml	Solids content, %	Column number
NR 100 %	280	32	1
NR 95 % – BE 5 %	275	28	2
NR 90 % – BE 10 %	270	29	3
NR 80 % – BE 20 %	271	27	4

In this first coverage stage of the percolation columns, after 45 min in none of the columns, liquid leakage was observed from the bottom. It was observed that on the surfaces the coloration was turning reddish and without liquids above it as shown in Table 4.

Percolation Experimental Tests. After 18 h of percolation columns were covered with the respective sludge, and cool water was gradually added to the column's surface: 200 ml at 0 min, 150 ml at 10 min, and 100 ml at 20 min.

Volume of supernatant and percolated liquid after 25 min since added last cool water is detailed in Table 5.



Table 4

Coverage data from the percolation columns with thickened sludge

	Column, cm			
	1	2	3	4
Fill Height	50	50	50	50
Sludge Height	3.4	3.5	3.6	3.6
Initial Height	53.4	53.5	53.6	53.6
Reduction 10 min	2.8	2.6	2	1.8
Height at 10 min	50.6	50.9	51.6	51.8

Table 5

Volume of liquid after added water

Column number	Height, cm	Volum of liquid, ml	
		Supernatant	Percolate
1	0.3	24	145
2	1.2	94	142
3	1.6	126	105
4	2.2	177	65

Result and Discussion. Metals Neutralization and Removal. Neutralization with unmixed hydrated lime is within the range of consumption (6.45 g/l) that is handled in the industry for acid water with pH between 1.91 and 1.96 as well as the content of analyzed dissolved metals.

In the tests with a mixture of hydrated lime with bentonite, the pH decreases with the increase in the proportion of bentonite – also the concentration of dissolved metals in treated water increases slightly. Nevertheless, still, at lower pH, the removal of dissolved metals occurs while complying with the environmental maximum permissible limit (MLP) standard already mentioned.

It should be noted that to reach the pH range of 7.6 to 8.0 using only hydrated lime as a reagent, although the consumption is low, the concentration of dissolved metals is above the MPL standard (pH 6 to 9, Pb 0.16 mg/l; Zn 1.20 mg/l; Cu 0.40 mg/l and Fe 1.6 mg/l): pH 7.8; Pb 0.18 mg/l; Zn 5.18 mg/l; Cu 1.23 mg/l; Fe 177.78 mg/l.

Furthermore, bentonite has a dissolved metal adsorption effect, decreasing its concentration in the liquid (Table 6) especially iron with which it would have an effect of pretty good adsorption, as mentioned by [34].

Table 6

Liquid characteristics obtained by the acid drainage treatment under different proportions of neutralizer and adsorbent, mg/l

Reagent	pH	Pb	Zn	Cu	Fe
NR 100 %	8.93-9.27	0.03	0.16	0.05	1.21
NR 95 % – BE 5 %	8.72-8.89	0.03	0.28	0.09	1.54
NR 90 % – BE 10 %	7.89-8.15	0.09	0.75	0.25	1.55
NR 80 % – BE 20 %	7.61-8.0	0.11	0.97	0.32	1.58

Coverage and percolation tests. From the results of coverage with thickened sludge, it is observed that at a higher proportion of bentonite in the mixture, the formed layer thickness on the surface is greater 10 min after its application. Also, for up to 45 min, there was no evidence of percolation at the bottom of the column.

By adding fresh water to the top surface of the columns, it is observed that when the coverage is of the sludge from the hydrated lime treatment (column 1), the height of the supernatant is lower and the volume of the percolated is higher. When the proportion of bentonite increases the percolation speed and the percolate volume decrease evidencing that water remains caught in the column.



For purposes of applying sludge mixtures with bentonite contents, it remains to investigate the sequence of its use on mineral deposits, tailings, or dismounts, depending on the characteristics that should eventually have the exposed layer exposed.

Conclusions. Acid water treatment with hydrated lime mixed with bentonite removes dissolved metals at a lower pH than when only hydrated lime is used. Sludge from acid water treatment with hydrated lime mixed with bentonite acquires additional properties of reducing percolation speed directly proportional to the content of bentonite. The use of hydrated lime neutralization sludge mixed with bentonite must be adapted to the type of coverage that is required for the closure of tailings, clearings, or exposed deposits in general.

It is highly expected to reduce AMD effluents at the source to reduce treatment costs, environmental damage, and tedious sludge management under a preventive approach.

Acid water neutralization sludge, currently considered a waste of no commercial value, which on the contrary represents handling and disposal costs, can be used as cover material.

The use of cover material based on neutralization sludge and bentonite on dumps of acid-generating material will gradually decrease the potential for acid generation of the dump by reducing aeration and water infiltration, both essential factors for the oxidation of sulfides and generation of acids and dissolved metals.

Coverage with a mix of bentonite neutralization sludge, unlike geosynthetic coverage, has the advantage of being better incorporated into the landscape environment in the post-closure of deposits generated by mining.

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