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Acid mine drainage treatment using by-products from quicklime manufacturing as neutralization chemicals



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HIGHLIGHTS

• By-products from quicklime manufacturing were investigated for AMD treatment.

• All tested by-products removed over 99% of Al, As, Cd, Co, Cu, Fe, Mn, Ni, Zn from AMD.

• All tested by-products removed approximately 60% of sulphate from AMD.

• The neutralization capacity of the by-products varied.

• Results indicate that two of the by-products could be used for AMD treatment.

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ABSTRACT

The aim of this research was to investigate whether by-products from quicklime manufacturing could be used instead of commercial quicklime (CaO) or hydrated lime (Ca(OH)₂), which are traditionally used as neutralization chemicals in acid mine drainage treatment. Four by-products were studied and the results were compared with quicklime and hydrated lime. The studied by-products were partly burnt lime stored outdoors, partly burnt lime stored in a silo, kiln dust and a mixture of partly burnt lime stored outdoors and dolomite. Present application options for these by-products are limited and they are largely considered waste. Chemical precipitation experiments were performed with the jar test. All the studied by-products removed over 99% of Al, As, Cd, Co, Cu, Fe, Mn, Ni, Zn and approximately 60% of sulphate from acid mine drainage. However, the neutralization capacity of the by-products and thus the amount of by-product needed as well as the amount of sludge produced varied. The results indicated that two out of the four studied by-products could be used as an alternative to quicklime or hydrated lime for acid mine drainage treatment.

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1. Introduction

Exposure of sulphide minerals in rocks to oxygen and water results in the production of environmentally harmful mine water that is acidic and has high concentrations of sulphates and metals (Brown et al., 2002; Bowell, 2004; Akcil and Koldas, 2006). The term used for this kind of mine water is acid mine drainage (AMD). AMD is produced both in active mines and also after mine closure (Johnson and Hallberg, 2005).

http://dx.doi.org/10.1016/j.chemosphere.2014.07.090 0045-6535/© 2014 Elsevier Ltd. All rights reserved. AMD may contain several thousands of mg L^{-1} of sulphate (Brown et al., 2002; Bowell, 2004). Sulphate has been considered quite harmless to the environment and more attention has been paid to limiting metal containing water emissions (Bowell, 2004; Silva et al., 2012). Not until recent years have authorities in Finland set limits for sulphate emissions for mine discharge water. In Finland these limits are mine specific and are defined in environmental permissions. High concentrations of sulphate can form sediments at the bottom of rivers and lakes (Benatti et al., 2009) in which thick layers of sediment can cause anoxic conditions and toxicity via the formation of hydrogen sulphide (Benatti et al., 2009). AMD typically contains elevated concentrations of toxic metals and therefore causes a threat to the environment and health (Brown et al., 2002; Akcil and Koldas, 2006).



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Gray (1997) has summarized the main pollutant factors of AMD as acidity, salinization, metal toxicity and sediment toxicity.

AMD treatment can be divided into active and passive methods. Passive methods include biological treatment with constructed wetlands and chemical treatment with limestone drains. Active treatment methods include chemical precipitation with alkaline chemicals, biological treatment with bioreactors, and other techniques such as adsorption, ion exchange and reverse osmosis (Brown et al., 2002).

The traditional treatment for mine water is chemical precipitation with quicklime or hydrated lime for neutralization and removal of metals and sulphates (Brown et al., 2002). Addition of alkaline slurry which is typically made of quicklime slaked with water precipitates metals primarily as hydroxides. Sulphate removal occurs through precipitation of gypsum, but only partial sulphate removal is possible due to the solubility of gypsum (INAP, 2003). The solubility of gypsum depends on the composition and ionic strength of the solution and is between 1500 and 2000 mg L⁻¹ (INAP, 2003). In addition to the traditional treatment, an integrated limestone/lime process for the neutralization of AMD has also been studied (Geldenhuys et al., 2003; Maree et al., 2004).

Precipitation with quicklime or hydrated lime is effective for the treatment of highly acidic mine waters, barely affected by temperature fluctuations and the process is relatively simple (Brown et al., 2002). However, lime precipitation has some problems (Matlock et al., 2002; INAP, 2003; Chen et al., 2009) such as large amount of sludge produced, high water content in the produced sludge, difficulties in dewatering of the sludge, high sludge disposal costs and plugging of equipment and pipelines. In recent years there has been growing interest in the utilization of industrial by-products that would otherwise be treated as waste. EU waste legislation encourages applying the following waste hierarchy (1) prevention, (2) preparing for re-use, (3) recycling, (4) other recovery and (5) disposal (Directive 2008/98/EC). Re-use of waste is both ecological and economical due to reduced amount of waste and reduced chemical and waste disposal costs. Utilization of paper and pulp mills by-products as alkaline chemicals for AMD treatment have been studied by Alakangas et al. (2013) and cement kiln dust utilization by others(Mackie et al., 2010a,b; Mackie and Walsh, 2012).

In this research by-products from quicklime manufacturing are investigated for AMD treatment and the results are compared with commercial quicklime and hydrated lime. Neutralization as well as metals and sulphate removal capacities are used to measure the chemical precipitation efficiency of the by-products. Characterization is performed with particle size distribution and scanning electron microscope (SEM) for the by-products and with X-ray diffraction (XRD) for sludge formed by the by-products.

2. Experimental

2.1. By-products from quicklime manufacturing

Samples of four by-products denoted as BP A to BP D from quicklime manufacturing were collected from two lime plants located in Finland. BP A is partly burned lime stored outdoors whilst BP B is also partly burnt lime but stored in a silo. Partly burnt lime which is a mixture of limestone and quicklime is produced at fault situations and during kiln power-up. BP C kiln dust is the dust collected with fabric filter during quicklime manufacturing whereas BP D is a mixture of BP A and dolomite. Present application options for these by-products are limited and they are mainly stored onsite. The particle size distribution of BP B differed radically from the other by-products, quicklime and hydrated lime. In this research the coarsest material of BP B was excluded from the experiments. Table 1 represents properties of the byproducts compared to quicklime (CaO) and hydrated lime $(Ca(OH)_2)$ reported by the manufacturer. Free lime is the amount of CaO available for reacting and is measured by titration according to the standard ASTM C-25 (ASTM International, 2011). Total lime includes free lime but also CaO that is bonded to calcium carbonate (CaCO₃). BP A and BP C had the smallest amount of total lime as well as free lime. On the other hand, the amount of total and free lime for BP B, were bigger than those for hydrated lime.

2.2. Mine water

Mine water samples were taken from an operating copper-zinc mine located in Finland. The collected water was AMD which was pumped from the underground workings. Mine water, process water from the concentrating plant and tailings are compiled and treated simultaneously in the industrial process at the mine site. In the industrial process onsite hydrated quicklime as a 12% by weight slurry is used to raise the pH of mine water above 10 to ensure the precipitation of heavy metals as hydroxides.

2.3. Experimental procedures

Experiments were performed with the jar test (Kemira Kemwater, Flocculator 2000) in which an AMD sample volume of 800 mL was used for each jar. Experiments were conducted at room temperature (20 ± 2 °C). 10% by weight slurries were made for each by-product, quicklime and hydrated lime by dissolution into ultrapurified water. The appropriate amount of slurry was then added to raise the AMD sample pH from 2.6 to 9.5. Next, the sample was rapid mixed at 150 rpm for 1 min, followed by slow mixing at 50 rpm for 5 min and then left to settle for 30 min. The pH and mixing and settling parameters were chosen with minor alterations according to literature (Mackie et al., 2010b). After settling water samples from the supernatant were taken for sulphate and metal analysis. The sludges were filtered with Schleicher & Schuell 589 Blue ribbon quantitative low ash filter paper by vacuum filtration, air dried and weighed prior to analysis.

2.4. Analytical methods

Sulphate analyses to water samples were performed in an accredited laboratory with ion chromatography (IC) according to SFS-EN ISO 10304-02 (Finnish Standards Association SFS, 1997) standard and metal analyses with inductively coupled plasma optical emission spectrometry (ICP–OES) according to SFS-EN ISO 11885 (Finnish Standards Association SFS, 2009) standard. The sludge samples were analysed with PAN analytical XRD equipment with Cu K α at 40 mA and 45 kV. SEM images of by-products, quick-lime and hydrated lime were obtained using a Zeiss Ultra plus field emission SEM equipment at the Centre of Microscopy and Nanotechnology in the University of Oulu operated at 5 kV. The amount of sludge was estimated from the graduation marks of the jar test jars. Particle size distributions of by-products, quicklime and hydrated lime were determined with an Alpine Air Jet Sieve

T	able 1
Р	roperties of by-products reported by the manufacturer

Sample	Total lime (%)	Free lime (%)
BP A BP B BP C BP D Quicklime	54 87 48 Not reported 94	14 75 10 Not reported 91
Hydrated lime	73	/1

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