



Research paper

Acid rock drainage treatment by clinoptilolite with slurry bubble column: Sustainable zinc removal with regeneration of clinoptilolite



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ABSTRACT

Clinoptilolite is shown to be capable of removing zinc from ARD, while the zinc in loaded clinoptilolite is displaced by NaCl as regenerant. Repeated capture–regeneration cycles were tested, showing the evolution of uptake capacity over multiple cycles. Fresh and reused NaCl as regenerant were compared to explore a possible means of minimizing the waste volume. The results prove that uptake/removal cycles can be repeated several times while maintaining reasonable uptake and removal capacities. Three removal-only cycles after 10 full cycles continuously decreased the accumulated zinc in clinoptilolite, allowing the uptake capacity for further upcoming cycles to be recovered. When 10 full cycles were tested after the three removal stages, results were similar to those for the first 10 cycles. By reusing NaCl as regenerant, only one-quarter of the regenerant volume was required to achieve a similar total zinc uptake from ARD compared with fresh NaCl.

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1. Introduction

Concentrations of heavy metals in natural surface waters increase significantly after excavation and subsequent weathering of sulfide-rich minerals in certain mining sites and construction areas (Gray, 1998; Valente and Leal Gomes, 2009). The resulting aqueous solution is known as acid mine drainage (AMD) or acid rock drainage (ARD). Water contaminated by ARD, often containing elevated concentrations of metals, can be toxic to aquatic organisms, leaving receiving streams devoid of most living creatures. Fish in the receiving waters are exposed indirectly to metals through ingestion of contaminated sediments and food. Iron hydroxides may physically coat the surfaces of stream sediments and streambeds, threatening the spawning habitat (Kimmel, 1983). The specific effects of low pH and heavy metals on growth and reproduction may be related to calcium metabolism and protein synthesis (Fromm, 1980). Due to the toxic effects of heavy metals (He et al., 1997, 1998), it is necessary to treat ARD before it is discharged to the receiving environment. ARD water remediation is therefore in demand around the world (Akcil and Koldas, 2006; Gazea et al., 1996; Johnson and Hallberg, 2005). The use of relatively simple, yet effective, technologies utilizing low cost abundant natural resources would be advantageous to help establish more sustainable treatment strategies.

Clinoptilolite particles can be effectively used in ion exchange processes as sorbent materials for heavy metal removal applications in wastewater, owing to their favorable ion exchange capacities and low cost (Barrer et al., 1978; Breck, 1974; Curkovic et al., 1997; Inglezakis

et al., 2004). Several aspects of metal removal via clinoptilolite, such as determination of removal capacity, effect of particle size, temperature, other cations and pretreatment, have been investigated over the years (Doula, 2006; Inglezakis et al., 2001, 2004; Oren and Kaya, 2006; Ouki and Kavannagh, 1997; Petrus and Warchol, 2003, 2005; Semmens and Martin, 1988). However, very few studies have been carried out using natural ARD (Cui et al., 2006; Li et al., 2008; Xu et al., 2011). The results of these studies show less satisfactory treatment performance of clinoptilolite, due to the complex composition of ARD, than for artificial acidic solutions with similar metal concentrations of only one or two metals (Motsi et al., 2009, 2011).

Limited studies have been performed on the regeneration of clinoptilolite after Zn removal. Blanchard et al. (1984) sought optimum regeneration conditions for removing NH₄ and mixed metals (including Zn) from solution in clinoptilolite packed beds. Zamzow et al. (1990) demonstrated effective removal and complete elution of Zn for 27–37 removal/regeneration cycles from a clinoptilolite packed column for water treatment, attempting to meet the US EPA drinking water standards, with NaCl as the regenerant. Vasylechko et al. (2003) tested HCl-modified clinoptilolite for Zn removal with potassium salts for successful regeneration. However, regenerant recycling over repeated uptake/removal cycles has not been previously investigated. Successful regeneration of clinoptilolite would enable the recovery of the sorbent and concentration of regenerant solution to a volume significantly less than for the original solution. Studies of this possibility are essential for future practical application of this natural material in ARD treatment.

Most previous research on adsorption capacity has investigated artificial solutions. Batch desorption tests have been based on spiked artificial feed, with single (Turan et al., 2005; Vasylechko et al., 2003)

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or multiple (Blanchard et al., 1984; Zamzow et al., 1990) metals in distilled water, where the performance tends to be much better than for ARD. In our previous studies, freshly prepared regenerant was introduced in each cycle (Li et al., 2008; Xu et al., 2011). No study has been reported on recycling the regenerant. Cyclic regeneration was limited to only 1–3 cycles in artificial solution (Blanchard et al., 1984). Only a single study (Li et al., 2008) has been reported for ARD treatment. In that case, a small batch rotating column provided multiple regenerations.

The present study aims to minimize the solid waste (used-clinoptilolite) and liquid waste (loaded-regenerant) resulting from ARD treatment. To achieve this, tests were performed in a slurry bubble column on Zn uptake by clinoptilolite, followed by the removal of zinc from the resulting partially-loaded clinoptilolite by contacting with regenerant. In this paper, we use the terms “uptake” and “removal”, rather than “sorption” and “regeneration”. We do this because the mechanism of loading may involve ion exchange, as well as sorption (Xu et al., 2011).

In the first set of tests, denoted as FRESH, fresh NaCl was used for each removal to extract Zn from the partially loaded-clinoptilolite. In the second set, fresh NaCl regenerant was used only for the initial cycle, after which the same regenerant was used repeatedly for 9 further cycles. To examine the recovery of the uptake capacity of the loaded-clinoptilolite, three removals with fresh NaCl solution were carried out next, followed by 10 cycles of uptake/removal following the same procedures as for the previous 10-cycle sequence. The work is novel as this is the first time that natural ARD has been tested with multiple uptake/removal cycles up to 10 cycles. No previous study has been conducted on reused regenerant to determine the recovery of Zn uptake capacity and the service life of the clinoptilolite. The results are helpful in developing a more sustainable treatment system to minimize liquid and solid wastes commonly associated with ARD treatment processes.

2. Materials and methods

A slurry bubble column (Cui et al., 2006; Xu et al., 2010, 2011) of inside diameter 0.090 m and overall height 1.4 m was employed for the experiments. A schematic is shown in Fig. 1. Air was introduced into the bottom of the column through a ‘wind box’ before passing through a distributor plate containing 57 uniformly-distributed orifices of diameter 6.3 mm, with a screen underneath to prevent loss of particles. The bubble formation and coalescence caused by the air flow created strong liquid circulation, often called “Gulf-streaming” (up in the core and down at the walls).

The clinoptilolite tested in the experiments was from Bear River Zeolite, USA. Key properties are listed in Table 1. This material was first oven-dried (105 °C for 24 h) to equalize the moisture content of the various size fractions. Based on Xu et al. (2010), the Zn uptake by raw clinoptilolite is ~12–15% lower than that of the NaCl pre-treated clinoptilolite. For consistency, the batches of clinoptilolite for all regeneration tests in this paper were pre-treated with NaCl (Xu et al., 2010) to homogenize the ions on its adsorption sites.

ARD from Highway 97C in British Columbia (Coquihalla Merritt-to-Peachland connector) with a pH of 3.2 was used in the experiments. Background concentrations are given in Table 1. Our major focus was zinc, as it is the contaminant of major concern in this ARD.

Based on previous work (Cui et al., 2006; Xu et al., 2010), slurries were prepared to provide concentrations of 100 g clinoptilolite/kg liquid. 2.0 kg ARD was added for each test. One-hour agitation time was adopted for each uptake test and also for each removal test. Operational parameters examined were particle size 600–850 µm, sorbent dosage 100 g kg⁻¹ ARD, and initial aqueous pH 3. The zinc ion concentration in the ARD could vary due to seasonal changes. The ARD was spiked with zinc ions in the absorption batch experiments by adding zinc chloride (ZnCl₂) solution to give Zn concentrations of

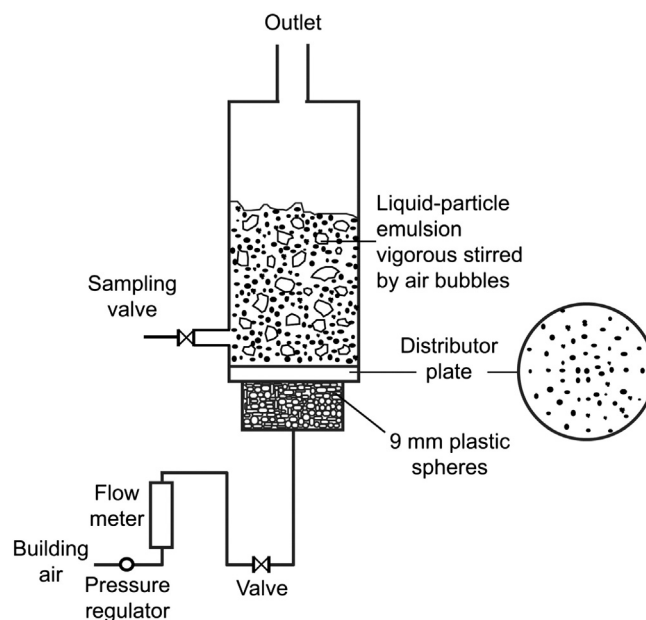


Fig. 1. Schematic of slurry bubble column.

100 mg kg⁻¹, to match the highest seasonal zinc concentrations at the contaminated site. After contacting for 60 min in the slurry bubble column, the supernatant liquid was separated from the clinoptilolite samples by filtration using Whatman No. 2 filter paper. The Zn-loaded clinoptilolite was then drained in the column prior to the removal test.

The metal ion concentrations of the supernatant were determined by an atomic adsorption spectrophotometer (AAS) (Thermo Jarrell Ash Video 22, TJA Solutions, US) after acidification with 0.5% HNO₃ and passage through a 0.45 µm filter (Clesceri et al., 1998). Aqueous zinc removal was determined by comparing concentrations before and after each sorption test (USEPA, 1987).

Based on previous studies (Xu et al., 2011), 20 g L⁻¹ NaCl solution at pH 3 was adopted for the removal of the Zn from the loaded clinoptilolite. Studies were carried out in batch mode in the slurry bubble column with 100 g of loaded clinoptilolite/kg regenerant. 2.0 kg regenerant was used for each test. The removal of Zn is calculated from the Zn concentrations in the initial regenerant and final regenerant, based on the same principle as for adsorption tests (USEPA, 1987).

Table 1
Clinoptilolite and ARD metal composition.

Mineral components ^a	85% clinoptilolite; balance opaline silica	ARD				
		Metal	(mg kg ⁻¹)	(mmol kg ⁻¹)		
Chemical composition ^a	Cations	K	3.47%	Zn	14.4	0.220
		Ca	1.60%	Mn	10.6	0.193
	Na	0.44%	Al	21.9	0.812	
	Other elements	Al	6.1%	Ca	198	4.94
		Fe	1.3%	K	4.5	0.115
		Ba	1200 ppm	Na	179	7.79
		Sr	560 ppm	Si	40.4	1.44
		Zr	480 ppm			
		Ce	130 ppm			
		Rb	120 ppm			
		La	55 ppm			
		Y	55 ppm			
		Nd	45 ppm			
		Nb	40 ppm			
		Zn	35 ppm			
		Cu	25 ppm			
		Pb	20 ppm			

^a Source: Provided by the supplier.

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