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Research article

Comparison of the efficiency of chitinous and ligneous substrates in metal and sulfate removal from mining-influenced water

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ABSTRACT

Mining-influenced water (MIW) remediation is challenging, not only due to its acidity and high metal content, but also due to its presence in remotely located mine sites with difficult surrounding environments. An alternative to common remediation technologies, is the use of sulfate-reducing bacteria (SRB) to achieve simultaneous sulfate reduction and metal removal in on-site anaerobic passive systems. In these systems, the organic carbon source (substrate) selection is critical to obtaining the desired effluent water quality and a reasonable treated volume. In this study, we evaluated the use of two different substrates: a chitinous product obtained from crushed crab shells, and a more traditional ligneous substrate. We put the substrates, both with and without water pretreatment consisting of aeration and pH adjustment, in anaerobic experimental columns. The treatment with the chitinous substrate was more effective in removing metals (Al, Cu, Fe, Cd, Mn, Zn) and sulfate for a longer period (458 days) than the ligneous substrate (78 days) before suffering Zn breakthrough. The reactors fed with pretreated water had longer operational periods and lower metals and sulfate concentrations in the effluent than those with untreated influent water. Zn was consistently removed to levels < 0.3 mg/L for 513 days in the chitinous substrate columns, while levels < 0.3 mg/L were maintained for only 140 days in the ligneous substrate pretreated column. The highest sulfate removal rates achieved in this study were in the range of 5–6 mol/m³/d for the chitinous substrate and 1–2 mol/m³/d for the ligneous substrate. Overall, the chitinous substrate proved to be more efficient in the removal of all the aforementioned metals and for sulfate when compared to the ligneous substrate. This could be the determinant when selecting a substrate for passive systems treating acidic MIW, particularly when Zn and Mn removal is necessary.

1. Introduction

Acidic mining-influenced water (MIW), which is formed due to the biochemical reaction of sulfide minerals in active and inactive mining sites, has been widely found to increase metals and sulfate contaminants in streams and groundwater (Hiibel et al., 2011; Klein et al., 2014; Pinto et al., 2011). Several metals (e.g., Fe, Cu, Al) found in MIW can be removed by precipitation induced by pH neutralization and aeration, but other metals (e.g. Zn, Mn) are more difficult to remove by these processes (Medírcio et al., 2007; Nuttall and Younger, 2000), and may require alternative treatments, such as membrane filtration, ion exchange, adsorption, etc. (Fu and Wang, 2011). These alternative treatments are usually more expensive than traditional treatment systems, and demand the input of chemicals, energy, and supervision for the effective removal of contaminants. Therefore, we pursued the use of a passive system that requires fewer monetary and material resources

for metal removal.

Anaerobic bioremediation is one of the passive treatment technologies commonly used to precipitate and separate metals as metal sulfides using sulfate reducing bacteria (SRB) (Al-Abed et al., 2017). Under anaerobic bioremediation, metals removal occurs by 1) precipitation as sulfides, 2) precipitation as carbonates or hydroxides (a consequence of the increase of pH), and 3) adsorption onto the substrates and onto the biomass. In addition to metal adsorption, substrates play other important roles: as carbon and nitrogen sources for biomass growth, as an inoculum source, as air/water exchanging porous medium, and as a neutralizing agent (Neculita and Zagury, 2008). Hence, the substrate amount and composition directly affect the removal efficiency and lifetime of a bioreactor.

The selection of simple and easily degradable substrates, those that have short-chain low molecular weight (e.g., methanol, ethanol, lactate, acetate, etc.), has been successfully tried for sulfate and metals

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removal. But not all SRB species are able to oxidize lactate or ethanol to CO₂ (Zagury and Neculita, 2007), and they usually demand high amounts of substrate in an active system. Hence, these are unattractive for passive systems, which typically utilize complex substrates such as sawdust, wood chips, hay, alfalfa, manure, or combinations of these. In order to oxidize these complex substrates and to produce short-chain carbon compounds (e.g., acetate, etc.), SRBs rely on acidogens and methanogens as they cannot oxidize them by themselves (Neculita et al., 2007).

The use of a mixture of materials, rather than a single material substrate, usually yields better efficiencies due to synergism (Zagury and Neculita, 2007). Manure is frequently used in these mixtures because of its nutrient content, matrix complexity, degradability, and low cost, but it is not efficient as a single substrate material because it tends to generate clogs in the system as it is compacted during the operation. Hence, combinations of easily available carbon (as manure) and ligneous materials (e.g., wood chips, hay, sawdust, etc.) are popular choices of substrate to obtain a combination of degradability and porosity in Sulfate Reducing Bioreactors (SRBRs).

Substrates based on ligneous materials have been found by some investigators to be ineffective for Mn removal. For example, ligneous materials combined with calcium carbonate and urea were reported to be ineffective in removing Mn (Zagury et al., 2006; Zagury and Neculita, 2007). However, Mn removal by precipitation and adsorption in SRBRs has been reported at a pH range of 7–10 using a ligneous substrate with manure (Logan et al., 2005) and a combination of ligneous substrate, spent mushroom compost, and manure (Vasquez et al., 2016).

Due to these findings, we looked to emerging substrates, such as chitin-based products (crushed crab shells, etc.), as a viable option in SRBRs (Daubert and Brennan, 2007; Robinson-Lora and Brennan, 2010; Venot et al., 2008). Specifically, we looked at crushed crab shells, a chitin-based product, because of its composition: 40% calcium carbonate (neutralizing agent), 30% protein (carbon source), 20% chitin (N-acetylglucosamine polymer that serves as a solid support for the biomass), 7% moisture, and 3% ash (Pinto et al., 2011). The physical, particulate form of crushed chitin needs to be addressed for effective use in SRBRs, as it tends to mat and have very low permeability/hydraulic conductivity after being wetted. Therefore, it needs to be mixed with sand to obtain greater porosity and have the desired range of hydraulic conductivity in SRBRs (Al-Abed et al., 2017; Robinson-Lora and Brennan, 2009).

In pilot-scale chitinous substrate bioreactors with metal and sulfate-laden influent, Venot et al. (2008) found successful removal of Cu (0.03 in the influent to 0.002 mg/L in the effluent), Zn (4.67–0.02 mg/L), and Mn (19.3–3.77 mg/L). Robinson-Lora and Brennan (2010) compared a crab shell product with lactate to spent mushroom compost in sacrificial batch microcosms. They concluded that crushed crab shells were more efficient in metal and SO₄²⁻ removal and in acidity neutralization. Both substrates removed Al, but only the chitinous substrate removed Fe and Mn (Mn removal reached a 73% removal rate likely precipitating as rhodochrosite).

Neculita and Zagury (2008) evaluated maple wood chips, maple sawdust, composted poultry manure, and leaf compost as substrates in anaerobic batch reactors reporting effective removal rates (92–99.8%) for Fe, Ni, Cd, Zn, and Mn. The authors also found that higher C/N ratios and dissolved organic carbon (DOC)/SO₄²⁻ ratios provided greater sulfate and metal removal. Since it is difficult to compare substrate efficiencies with different variables, Zn and Mn removal efficiencies in chitinous and ligneous substrates should be compared using similar bioreactors and influent water.

The primary objective of this study was to evaluate the long-term effectiveness of a chitinous substrate (crushed crab shells) compared to a traditional ligneous substrate (wood chips, hay, and manure) on Zn, other metals (Al, Cu, Fe, Cd, Mn), and sulfate removal in MIW under anaerobic column bioreactor conditions. The secondary objective

Table 1
pH, metals, sulfate, and dissolved oxygen content in the Untreated Formosa Water (UFW) and Pretreated Formosa Water (PFW).

Parameter	Units	UFW	PFW
pH	pH units	2.94	6.67
Al	mg/L	17.8	0.057
As	mg/L	0.019	0.004
Ba	mg/L	0.010	0.05
Ca	mg/L	72.7	75.9
Cd	mg/L	0.267	0.242
Cu	mg/L	16.8	0.690
Fe	mg/L	106	0.357
Mg	mg/L	18.6	17.3
Mn	mg/L	1.53	1.42
Ni	mg/L	0.043	0.049
Pb	mg/L	0.078	< 0.017
Zn	mg/L	73.9	57.3
Sulfate	mg/L	2620	770
Dissolved Oxygen	mg/L	1.43	4.47

includes the evaluation of aeration and neutralization water pretreatment on the removal of the mentioned contaminants.

2. Materials and methods

2.1. Mine water collection and pretreatment

Water collected by the investigators from the Formosa Superfund site in Oregon was used in this investigation. The Untreated Formosa Water (UFW) was collected at the mine site, transported, and stored under a nitrogen blanket at room temperature using the methods described by Al-Abed et al. (2017). The mine water was pretreated in the laboratory in 15 L batches using mechanical stirring (IKA Model RW20 stirrers, Wilmington, NC) and continuous air purging for 30 min. At the 15th minute, ~14.3 mL of 10 N NaOH was added to the bucket for neutralization. After those 30 min, the water was allowed to settle for 15 min, sand filtered, and labeled as Pretreated Formosa Water (PFW). The metal content was measured before and after the pretreatment (see Table 1).

2.2. Columns fill materials

The chitinous substrate filling material was comprised of 140 g SC-20 (crushed crab shells product Chitorem SC-20[®], JRW Bioremediation LLC, Lenexa, KS) and 460 g quartz sand (Global Drilling Suppliers, Inc., Cincinnati, OH). The lignocellulose substrate filling material was comprised of 252 g ash tree wood chips (locally collected), 17 g hay (locally collected), and 4 g cattle manure (obtained from an Ohio farm). Each component of filling material was characterized for moisture (ASTM Method D2216-10) and elemental composition by Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES) analysis (EPA Method 6010B) after acid digestion using EPA Method 3051. Total Organic Carbon (TOC) was measured using a Shimadzu TOC-VCPH analyzer equipped with a SSM-5000A solid sample module.

2.3. Column bioreactor design, set-up and operation

Six plexiglass columns (length 101 cm, inner diameter 3.81 cm, volume 1.15 L) were operated in parallel (Fig. S11) with upward flow configuration. The column fillings were selected to compare the substrate performance: Columns 1, 3, and 5 (chitinous columns) contained a sand/SC-20 (3:1) mixture, while Columns 2 and 4 (lignocellulosic columns) contained a wood chips, hay, and cattle manure mixture. Column 6 was filled with sand only (Inert Control). The substrate amounts were determined to keep similar total carbon levels (32 g) in Columns 1–5 (see carbon content of these materials in Table S11).

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