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Evaluation of waste materials for acid mine drainage remediation

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ABSTRACT

Laboratory scale tests were conducted to assess the efficiency of two different types of waste materials to remediate acid mine drainage (AMD). The waste materials used in the current study were recycled concrete aggregates (RCAs), and fly ashes. Four different RCA materials and three different fly ash materials were evaluated. Column leach tests (CLTs) were conducted to determine the effects of the remediation materials on pH, electrical conductivity, alkalinity, oxidation reduction potential (Eh), and concentrations of sulfate (SO_4^{2-}), chromium (Cr), iron (Fe), manganese (Mn), copper (Cu), and zinc (Zn) in AMD. Results of the CLTs suggest RCAs and one of the highly alkaline fly ash can effectively raise pH of the AMD and reduce concentrations of Cr, Cu, Fe, Mn, and Zn in AMD. In addition, sulfate concentrations of AMD decreased significantly after being treated by RCAs while sulfate concentrations of the AMD increased when it was remediated by fly ashes. It is speculated that leaching of sulfate from fly ash samples during treatment may decrease the metal sorption capacity of fly ashes. X-ray fluorescence spectroscopy quantified the impact of CaO and loos on ignition (LOI) in the remediation materials on sorption capacity of metals from the AMD. Sorption capacity for Cr, Cu, Fe, and Zn was found to be greater in materials with high CaO and LOI content, and low unburned carbon.

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1. Introduction

Acid mine drainage is a caustic by-product of hard rock mining that can terminally impair waterways and alter landscapes. It occurs as a result of mining operations when sulfide-bearing rock oxidizes to form sulfuric acid in the presence of atmospheric oxygen and water (precipitation, groundwater, or surface water) [1–3]. The sulfuric acid dissolves surrounding rock, releasing metal ions into solution. The resulting leachate is referred to as acid mine drainage (AMD). It is highly acidic (pH typically < 3) and can contain high concentrations of trace metal ions including iron (Fe), zinc (Zn), manganese (Mn), nickel (Ni), cadmium (Cd), cobalt (Co), copper (Cu), and aluminum (Al) [4]. When AMD enters surface water, groundwater, and soil it can significantly lower pH and raise metal concentrations [5].

When feasible it is of best practice to prevent the formation of AMD using source control measures such as sealing or flooding of underground mines, solidification of mine tailings, and disposal of mine wastes in sealed waste heaps [1]. Unfortunately, these treatment methods can be extremely costly, requiring continuous chemical input and large volumes of virgin material, such as lime and limestone. In an effort to reduce the high costs of AMD treatment, interest has been developed in various applications of low-cost waste products, such as fly ash and recycled concrete aggregate (RCA), for AMD remediation. Both RCA and fly ash are highly alkaline, exhibiting unique binding properties that could make them effective alternatives to costly, lime and limestone treatment.

The application of fly ash for AMD remediation is a sustainable approach that could be advantageous to coal mines and power plants generating fly ash as a means to repurpose waste. Coal combustion by-products, including fly ash, are one of the nation's most widely produced industrial waste products. In 2012, the United States coal industry produced over 53.4 million tons of fly ash [6]. Unfortunately, less than 50% of fly ash is reused, and the remaining is disposed of in landfills or retained in ponding facilities [6].

Recent studies have experimented with mixing fly ash with mine tailings to improve pH and reduce metal concentrations in AMD. Shang et al. [7] found that co-disposing of fly ash and mine tailings neutralized the pH of pore fluid, and reduced effluent concentrations of heavy metals to meet local regulatory requirements for leachate quality. Similarly, Mohamed et al. [8] found that mixing mine tailings with a mixture of aluminum, lime, and fly ash (ALFA) effectively reduced leaching of heavy metal from the mine tailings. Siriwardane et al. [9] and Bulusu et al. [10] investigated

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the use of fly ash grouts to reduce mine flow rates and serve as permeable reactive barriers (PRBs) in abandoned mines. Siriwardane et al. [9] found when injected into an abandoned mine fly ash grout could successfully reduce mine flow rates by 95% while exceeding laboratory strength requirements for subsidence control. Furthermore, Bulusu et al. [10] used unconfined compressive strength, slump, and modified flow tests to demonstrate that fly ash grout acted as a permeable reactive barrier by effectively reducing the production of AMD in field applications.

In addition to fly ash applications, the application of recycled concrete aggregate (RCA) for remediation of AMD could conserve landfill space while also providing effective, low-cost treatment for AMD. Currently, only about 50% of the over 200 million metric tons of RCA produced annually in the U.S. is reused [11]. The primary reuses for AMD are as road base, components of new concrete or asphalt mixes, high-value riprap, and as low-value fill products [12]. Studies have shown that RCA is strong, porous, and composed of an assortment of alkaline materials (including limestone, calcium-bearing minerals, and Portland cement) with high neutralization and sorption capacities that could make it an effective material for AMD treatment. Indraratna et al. [13] revealed that when used as a reactive material in PRBs, recycled concrete removed Fe and Al from groundwater and improved its pH from acidic to mildly alkaline. In addition, Bestgen et al. [14] and Chen et al. [15] found that fine particles of RCA (less than 0.075 mm) had greater acid neutralization capacities than coarse particles, and Cu and Zn experienced the lowest leaching at pH less than 5 and highest leaching at pH of 2.

The objective of this study, through laboratory testing and analysis, was to evaluate the low-cost construction waste products, recycled concrete aggregate and fly ash, as remediation materials for AMD treatment. Column Leach Tests (CLTs) were conducted to assess the impact of fly ash and RCA on pH, electrical conductivity, alkalinity, oxidation reduction potential (Eh); and concentrations of Ca, Cu, Cr, Fe, Mg, Mn, and Zn in AMD. Following the CLTs, additional analytical methods were conducted to better understand the treatment process on a molecular basis. X-ray fluorescence spectroscopy (XRF) was used to evaluate the impact of oxide, alkalinity, and unburned carbon content of the remediation materials on their capacity to sorb metals from AMD. In addition, scanning electron microscopy (SEM) was utilized to observe the effects of fly ash and RCA treatment on the physical properties of the remediation materials.

This article includes the following sections. (1) The Materials section covers a description of the physicochemical properties of the mine waste, four different RCAs, and three different fly ashes used in this study. (2) The Methods section discusses the experimental test procedures used in the current study, including (a) preparation of samples for CLTs, (b) collection of effluent solutions from CLTs, (c) measurements of Ca, Cu, Cr, Fe, Mg, Mn, and Zn concentrations by atomic absorption spectrometry, (d) measurements of pH, electrical conductivity (EC), and oxidation-reduction potential (Eh), (e) measurement of alkalinity values and sulfate (SO_4^{2-}) anion concentrations, and (f) XRF and SEM analytical methods to investigate the surface characteristics of materials tested in the current study. (3) The Results section discusses the results, including: (a) the change in pH and Eh in AMD after remediation, (b) the change in metal concentrations in AMD and its correlation with EC, sulfate concentrations and explanation of changes on the particle surface characteristics via XRF and SEM analyses, (c) the change in alkalinity in AMD and its correlation with leached Ca concentrations, (d) impact of CaO content on AMD remediation, and (e) impact of LOI content on AMD remediation. (4) The Conclusions section provides a summary of the results section and recommendations of utilization of RCA and fly ash for such applications for future field studies.

2. Materials

Mine waste recovered from the Homestake Mine in Lead, SD was used to generate acid mine drainage. Before closing in 2002, the Homestake Mine served as the largest and deepest gold mine in North America [16]. Table 1 provides the percent composition of oxides and metals in the mine waste from Homestake Mine. XRF and ICP-MS analysis of the mine waste revealed it is primarily composed of SiO_2 (54.9%) and Al_2O_3 (16.3%). It also contains high Cu, Fe, Zn, and Mn contents which could pose a significant risk to the environment and human health if leached from the material.

Four RCAs collected from various highway surfaces and recycled concrete plants in South Dakota were used in the study. The RCAs are referred to as RCA1, RCA2, RCA3, and RCA4. RCA1 and RCA2 consist of pulverized concrete from former highway pavement surfaces in Philip and Pierre, South Dakota. RCA3 and RCA4 were obtained from stockpiles in Sioux Falls and Rapid City, South Dakota.

Fig. 1 provides a particle size distribution of the RCAs. The particle size distribution shows that RCA2 and RCA3 are predominately (<40% by weight) composed of gravel particles (>4.75 mm), while RCA1 and RCA4 are primarily (>40% by weight) composed of smaller sand particles (75 μm to 4.75 mm). The physical properties of the RCAs are summarized in Table 2. According to the Unified Soil Classification System (USCS), RCA1, RCA3, and RCA4 are

Table 1
Chemical composition and total metal content of mine waste, fly ash, and RCA's.

Material	MW	FA1	FA2	FA3	RCA1	RCA2	RCA3	RCA4
pH	2.02	6.1	6.6	9.5	10.85	12.52	12.36	11.69
LOI (%)	4.9	6.2	6.8	8.1	25.2	12.0	7.3	32.7
Major oxide content (% by weight)								
Al_2O_3	16.3	23.1	26.9	25.5	0.44	3.2	2.0	1.4
CaO	0.11	1.07	0.70	12.5	37.9	45.1	52.5	55.8
Cr_2O_3	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Fe_2O_3	7.8	3.2	5.5	13.7	4.6	6.1	4.5	4.2
MgO	0.37	0.60	0.20	1.9	2.6	2.0	1.3	2.0
MnO	0.01	0.06	0.06	0306	0.05	0.08	0.04	0.03
SiO_2	54.9	45.1	50.8	50.4	32.3	37.9	48.3	35.1
Metal content (mg/kg)								
Cr	43	15.5	68	42	22	16	14.7	11
Cu	36.1	59.6	33	36	16	9.3	8.7	12.4
Mn	35	33.9	215.6	207.7	355	579	330	244
Zn	38	53.94	28.78	83.96	28	36	38	53

Note: LOI: loss on ignition. MW: mine waste, FA: fly ash, RCA: recycled concrete aggregate.

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