

Management of coal processing wastes: studies on an alternate technology for control of sulfate and chloride discharge

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Abstract Management of coal mining and coal processing wastes, particularly of high sulfur coals, can generate excessive amounts of sulfate (SO_4^{2-}) and chloride (Cl^-) in mine drainage that are known to negatively impact quality of both surface and ground water. The U.S. Environmental Protection Agency provides guidance to states on allowable SO_4^{2-} and Cl^- discharges from mine sites. This research evaluates the hypothesis that co-disposal of CCPW and FCPW with appropriate compaction can result in improved geochemical and geotechnical environments that will minimize acid mine drainage formation and SO_4^{2-} and Cl^- discharges. Addition of ground limestone (ag-lime) to the mix was also evaluated as a drying agent and for improvement in overall geochemistry by buffering higher pH values within the coal waste. These objectives were to develop and implement innovative concepts for engineered co-management of CCPW and FCPW at coal mining sites. The authors performed long-term field column leaching studies to analyze improvement in SO_4^{2-} and Cl^- in water quality. Requirements for stricter standards in some states led to the need for development of potentially improved environmental practices. This paper presents the overall encouraging results of the field kinetic studies.

Keywords Kinetic testing · Acid mine drainage · Coal chemistry · Interior Basin · Refuse co-disposal

1 Introduction

A typical underground coal mining complex in the mid-continental USA consists of a combination of mine shafts and slopes to access the coal seam/s along with coal preparation and refuse disposal facilities located on the surface as shown in Fig. 1. The term “coal refuse” in this paper refers to reject rock from crushers prior to run-of-mine (ROM) coal entering the processing plant and other waste rock derived from the coal

cleaning processes. The reject rock from the crushers is typically a small portion of the coal refuse. The ROM coal is generally wet processed to remove mineral matter and sulfur and to improve the quality of coal shipped to power plants. The marketable coal recovery typically varies between 50% and 60% with the remaining fraction disposed as waste on-site. Coal refuse typically consists of two size fractions: (1) coarse coal processing waste (CCPW), which is generally larger than 150 microns (100 mesh) in size and in some cases, is larger than 3.2 mm (1/8-inch) in size; and (2) fine coal processing waste (FCPW), which is generally a slurry ($\pm 15\%$ solids content) with solids being less than 150 microns (100 mesh) in size. CCPW refers to reject from heavy media vessels, jigs, cyclones, and spirals, while FCPW refers to reject from flotation columns and cells and effluent from filter presses, screen-bowl centrifuges, and de-sliming cyclones, all of which is typically concentrated in a thickener. The FCPW constitutes about 5%–10% of the ROM coal processed from the Interior Basin coal mines in the USA.

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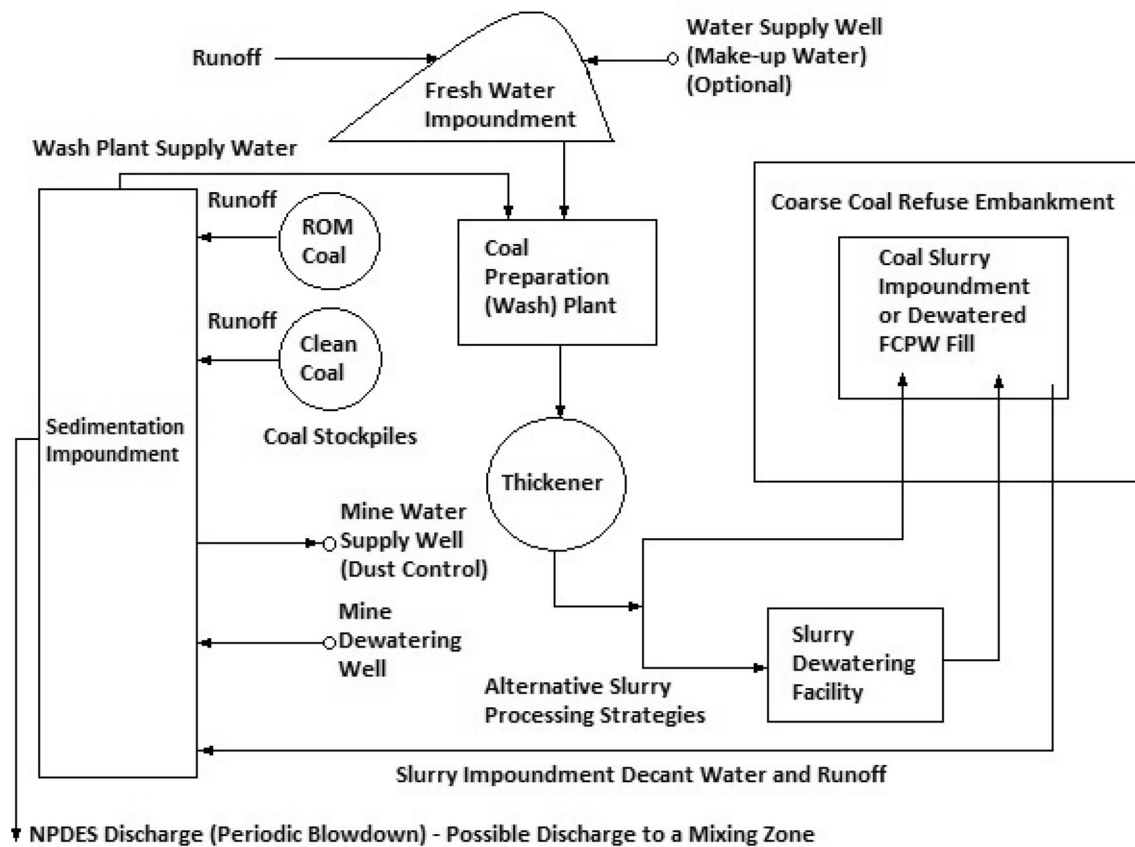


Fig. 1 Schematic of coal processing and water handling operations on the surface at a typical underground coal mine complex

FCPW is typically disposed as: (1) a slurry contained within an embankment constructed of CCPW (conventional practice) or incised ponds, (2) dewatered FCPW (50%–70% solids), contained within CCPW embankments as illustrated in Fig. 1, or potentially (3) a mono-fill constructed of a blend of CCPW and dewatered FCPW. The sizes of coal refuse disposal areas have been increasing in the Interior Basin due to the recent trends of mining thinner coal seams with larger equipment capable of cutting roof and floor rock. This has increased the percentage of the out-of-seam dilution (OSD) material to 50% or more of the ROM coal. Figure 1 also shows that underground coal mine complexes within the region typically operate a closed loop water-handling system where onsite water resources are continuously recycled and any regulated surface water discharges are from sedimentation basins, but typically only during larger precipitation events when either less stringent regulations is applied or there is a considerable amount of dilution water available.

2 Problem statement

Coal waste disposal facilities have been identified as a principal source of elevated SO_4^{2-} discharges by both state and federal environmental regulatory agencies (The Advent Group 2005;

Chugh et al. 2007a, b). Infiltration of oxygen- and ferric iron-bearing water into coal waste stockpiles contributes to the oxidation of pyrite and other sulfide minerals with the subsequent release of SO_4^{2-} and metals into mine drainage (Moses et al. 1987; Moses and Herman 1991; Evangelou 1995). Development of good management practices (GMPs) for refuse disposal and water management to reduce pyrite oxidation and SO_4^{2-} and chloride levels in these discharges are necessary to meet compliance levels for regulated constituents in mine drainage. Such practices should: (1) reduce water treatment costs during mining and subsequent reclamation of the facility, (2) reduce liability associated with maintaining and abandoning a large permanent impoundment structure, and (3) allow release of reclamation bonds in a timely fashion.

Weathering of pyrite can be controlled if water rich in oxygen and ferric ions is prevented from contacting sulfide minerals inherent in the waste rock (Evangelou 1995; Nordstrom 1982). Dissolved oxygen concentration, temperature, pH of water, reactive surface area of pyrites, form of pyrite, catalytic agents, flushing frequencies, and time allowed for contact are some of the variables that control the rate of pyrite oxidation and the associated SO_4^{2-} discharge. The inherent pyrite content within the coal and the associated strata is an important factor in determining the

amount of sulfate and metals in mine drainage. Although abiotic chemical conversion of Fe (II) to Fe (III) is an extremely slow reaction, the presence of iron-oxidizing bacteria increases reaction rate by an order of magnitude and, as a result, the production of SO_4^{2-} during the weathering process (Singer and Stumm 1970; Kleinmann and Crerar 1979). Within the Interior Basin, several researchers have provided noteworthy contributions to the control of acid mine drainage (AMD), including Nawrot and Gray 2000; Naumann and Wiram 1995; Wiram 1984, which has emphasized the advantages of adding alkalinity producing materials during coal mine reclamation. In China, the largest coal production country in the world, coal waste management and AMD are also very important. More than 350 mt of coal waste is produced each year, and is disposed as coal waste piles. Since most coal waste containing high sulfur content is located in arid or semi-arid regions, AMD issue has not drawn much attention to date. The focus of research has been on the spontaneous combustion because of the subsequent potential for oxidation of pyrite in coal wastes. The spontaneous combustion of coal waste piles has resulted in serious environmental problems, with several reclaimed coal waste piles re-igniting after establishing vegetation. Chinese scholars have developed new techniques for dealing with these problems (Hu et al. 2009a, b). The key new techniques are monitoring and mapping of the surface temperature by infrared instrument and photogrammetry, putting out fire by deep grouting, controlling oxidation process and acid generation by chemical and biological treatments and soil cover system with compaction (Hu et al. 2008, 2009b; Chen et al. 2014; Xu et al. 2014). However, more attention should be paid to AMD treatment, when spontaneous combustion is controlled.

3 Research hypothesis and overall research

This research has evaluated the hypothesis that co-disposal of CCPW and FCPW with or without addition of alkaline materials, and with appropriate compaction can improve geochemical and geotechnical parameters of coal waste that will minimize AMD formation and SO_4^{2-} discharges. This hypothesis was developed based on data available in the literature and also collected as part of a previous study (Chugh et al. 2007a) that concluded that the different-size CCPW fractions have a higher acid-forming potential (potential acidity or PA) and lower neutralization potential (NP) as compared to FCPW. In current practice, CCPW is typically disposed separately as a structural fill in surface disposal areas, while FCPW is disposed as a dilute slurry (about 15% solids) in impoundments that provide an overall lower disposal cost (NRC 2002). Previous research

also indicates that, in coal, the minerals which provide NP (e.g. calcite) weather faster than minerals that are acid-producing such as pyrite (e.g. Nawrot and Gray 2000; Hossner and Doolittle 2003). Therefore, drainage from coal waste, which is initially alkaline, remains alkaline for a certain period of time and then tends to become acidic. This period can be extended by addition of alkaline materials which is a widely accepted mine reclamation practice (Dreher et al. 1994; Nawrot and Gray 2000; Stewart et al. 2001). Chugh et al (2007a, b) and others have suggested that differential AP and NP rates could be used advantageously in waste management operations to allow for improved placement and compaction practices, and reduction of SO_4 discharges. There is a potential improvement in the overall NP and a reduction in the PA in a blend of CCPW and FCPW because the FCPW has a net neutralization potential (NNP; $\text{NNP} = \text{NP} - \text{PA}$) that in some cases is near neutral or, at least, a level slightly above ($-$) 5 tons of CaCO_3 equivalent per 1000 tons of material. Most permitting agencies consider a NNP less than ($-$) 5 tons of CaCO_3 equivalent per 1000 tons of material (in this case coal waste) potentially acid-forming and requiring management practices to control problematic mine drainage. Co-disposal of CCPW and FCPW offers an interesting alternative concept and is discussed in the next section.

Most Illinois mine operators would consider co-disposal if FCPW could be dewatered inexpensively to about 60%–65% solids and the long-term stability of a co-disposal fills can be ensured. As it is a relatively few number of U. S. mining operations currently practice refuse co-disposal. However, there remains a need to minimize disposal area space requirements and to eliminate the need for slurry disposal areas, which are becoming increasingly difficult and more expensive to permit. None of the operators interviewed in the preceding studies (Chugh et al. 2007a, b), have considered the potential improvements in geochemical and geotechnical environments as the result of co-disposal. A judicious addition of limestone or other alkaline materials to the co-disposal blend can provide additional advantages by buffering drainage pH, reducing high levels of total dissolved solids, and minimizing SO_4^{2-} discharge (Nawrot and Gray 2000). Both hypotheses presented above were evaluated in a field setting by constructing six large-size leaching columns—two each of 100% CCPW (the control sample); 90% (by volume) CCPW and 10% FCPW; and 84% CCPW, 8% FCPW, and 8% limestone. Material in each column was compacted to approximately 50% of the maximum Proctor density. All columns were equipped with a lysimetric and gravity drainage ports for collecting water samples.

This research was motivated by the Illinois Environmental Protection Agency (IEPA) policy to implement

lower standards for SO_4^{2-} and Cl^- discharge from coal mining and processing operations. Historically, Illinois allowed compliance level was 3500 mg/L for SO_4^{2-} discharges and 1000 mg/L for Cl^- discharges, whereas a “General Use” standard applicable to other industries had been set at 500 mg/L for both of these anions (Chugh et al. 2007a, b). The IEPA and the USEPA agreed that this difference was not legally defensible and implemented more restrictive SO_4^{2-} levels (USEPA 2009). These IEPA regulations also established a SO_4^{2-} discharge levels in a range of 500–2000 mg/L based on the hardness and Cl^- levels in the receiving stream; Cl^- discharge levels were also set at 500 mg/L.

3.1 Overall research

The overall goal of this study was to investigate the rate of SO_4^{2-} and Cl^- leaching from simulated kinetic tests in the field to demonstrate commercial potential for co-disposal technology. Co-disposal should enhance geotechnical stability through improved cohesion and compaction, while improving the geochemical environment. As a result, the overall refuse facility liabilities should be lowered. Addition of ground limestone (ag-lime) was also evaluated for use as a drying agent and as measure to improve in overall geochemistry by buffering higher pH values (< 8.0) inherent in fresh refuse from the basin. The study would aid mine operators in developing and implementing innovative concepts for engineered waste disposal. Although field testing was conducted at an Illinois mining operation, the concepts involved should be applicable throughout the Interior Basin and, with minor modifications, at other coal mines in the USA.

4 Research studies

These included: (1) physical and chemical characterization of CCPW, FCPW, and limestone materials; (2) characterization of geotechnical properties for identified waste disposal options; and (3) field-scale kinetic tests and data analysis.

4.1 Physical properties of coal waste

Both FCPW and CCPW were collected from a large coal preparation plant cleaning ROM coal from two commonly mined seams: Springfield (No. 5) and Herrin (No. 6). Most of this coal was produced from two underground mines. An approximately 18.9-liter (L) (5-gallon) sample of CCPW was collected and dried at low temperature (< 38 °C or < 100°F), and then screened for particle size distribution. The sampled materials were crushed to less than 10 mm (3/

8-inch) in size. Proctor compaction tests were conducted in 15 cm (6-inch) diameter molds to develop moisture-density data for optimizing compaction. The mold samples were also tested for uniaxial compressive strength for an undrained condition (Chugh et al. 2007a, b).

A similar sized sample of FCPW was also collected from the thickener underflow discharge. FCPW was decanted and air-dried before drying in a low temperature furnace. A 3.79-L (1-gallon) sample of agricultural ground limestone was also dried at low temperature and analyzed for particle size distribution.

4.1.1 Results

Table 1 shows average particle size distributions for CCPW and limestone samples. Since FCPW is uniformly less than 200 mesh (0.074 mm or 0.0029 inches), additional particle size analyses were not conducted on this fraction. A majority of CCPW (64.52%) is in the 4.75 to 50.8 mm (0.187–2.0 inch) size range. Agricultural ground limestone was a finer-grained material with a majority (72.97%) in the 1.70–19.05 mm (0.066–0.75 inch) size range. The initial moisture content of CCPW was 11.5%, whereas dewatered FCPW was 31.9% (68.1% solids content). In contrast, the initial moisture content of agricultural limestone was only 0.1%. Therefore, by blending limestone the moisture content could be significantly reduced.

4.2 Chemical properties of coal waste

The geochemical evaluation of CCPW, FCPW, and agricultural limestone included: (1) acid-base accounting, (2) determination of major and key trace elements in high temperature ash (HTA) product, and (3) x-ray fluorescence (XRF) analysis of all materials. Following SIUC laboratory determination of paste pH, the U.S. Geological Survey Energy Resources Program (USGS-ERP), the State of Illinois Office of Mines and Minerals (ILDNR-OMM), and

Table 1 Particle size distribution for CCPW and ground limestone

Sieve opening (mm)	Percent finer	
	CCPW	Ground limestone
50.8	94.31	100
19.05	62.20	100
4.75	29.79	78.09
1.7	15.09	27.03
0.425	4.44	13.17
0.075	0.53	4.58
< 0.075	0.52	3.58

a commercial testing laboratory determined the total and pyritic sulfur content along with an elemental analysis of all materials. Sulfur values were used to calculate potential acidity (PA) for CCPW and FCPW with respective values of 120.37 and 63.91 tons CaCO₃ per ton of coal waste.

Neutralization potential (NP) was also determined for all materials to determine the net neutralization potential (NNP). One CCPW and one FCPW coal waste sample was subjected to HTA analysis following heating in a 750 °C air-recirculating chamber furnace using ICP/MS and ICP-OES. Limestone elemental analysis was conducted using a Varian 725 ICP-OES for major elements (except SO₃, which was determined using a LECO sulfur analyzer) and presented in oxide form. A hand-held XRF analyzer was used to determine a large suite of elemental concentrations in whole dried and ground CCPW, FCPW and limestone samples.

4.2.1 Results

Acid-base accounting analyses results are presented in Table 2. The “mine permit” application provided representative data, while the remaining data was compiled from analyses performed in this study. Chugh et al. (2007a) presented additional acid-base accounting data for CCPW and FCPW samples collected from another similar preparation plant and found that: (1) the FCPW represented 10%–15% of the coal waste, and (2) the sulfur content is typically lower for FCPW as compared to CCPW. Elemental data and chemical analyses of HTA are presented in Tables 3 and 4. The second row of limestone data in Table 3 was provided by the quarry operator.

4.3 Geotechnical characteristics of different coal waste disposal options

Three waste disposal options were considered: (1) CCPW disposal in embankments or fills (the control)—Disposal Practice 1 or DP 1; (2) Co-disposal of blended CCPW

(90%) and FCPW (10%)—Disposal Practice 2 or DP 2; and (3) Co-disposal of blended CCPW (84%) and FCPW (8%) with limestone addition (8%)—Disposal Practice 3 or DP 3. Moisture-density relationships were developed using standard proctor tests for all disposal options. All tests were performed in a 15-cm (6-inch) mold using ASTM D698-12e1 Method C; ASTM (2013) prior to column construction (Behum et al. 2010). The mold size limited the top size of the material to about 20 mm (³/₄-inch).

4.3.1 Results

Particle size distribution for the materials used in Proctor tests are summarized in Table 5. These area based on three tests for DP 1 materials, two for DP 2 materials and two for DP 3 materials. The data is based on percent retained rather than percent passing.

The Proctor test results for all disposal options and tests are summarized in Fig. 2. For DP 1, the testing moisture contents varied between 2% and 17%. The field columns had maximum dry unit weight = 18.5 kN/m³ or 118 pcf. Similar data for DP 2 and DP 3 Proctor tests were 2%–9.5%, and 18.0 kN/m³ or 115 pcf; 2.5%–11.5% and 18.52 kN/m³ or 118 pcf. Moisture contents for maximum Proctor density values for DP 1, DP 2, and DP 3 mixes were 6.0%, 5.5%, and 7.5%, respectively. The addition of ground limestone advantageously increased the moisture content at which maximum density was achieved by about 2%.

4.4 Field kinetic testing

Field-scale kinetic tests were conducted at an active coal mining complex. During the first phase of field-scale testing, three sets of duplicate test columns (FC-1 through FC-6) were constructed using 200-L (55-gallon) barrels. These columns were monitored for 19 months. Unfortunately, a tornado damaged some columns. A new set of field-scale kinetic-leaching columns were assembled from relatively

Table 2 Geochemical properties of coal waste and ground limestone samples

Coal waste fraction	Mean sulfur content (%)		Median paste pH	Tons CaCO ₃ per 1000 Tons of waste		Net Neutralization potential (NNP)
	Total	Pyritic		Maximum potential acidity (PA)	Neutralization potential (NP)	
Mine permit (coarse)	5.70 (n = 2)	3.41 (n = 47)	7.12 (n = 47)	106.4 (n = 47)	23.8 (n = 47)	– 84.5 (n = 47)
Coarse	4.84 (n = 2)	3.90 (n = 1)	5.92 (n = 1)	121.88 (n = 1)	1.51 (n = 1)	– 120.37 (n = 1)
Fine	2.56 (n = 2)	2.13 (n = 1)	6.31 (n = 1)	66.53 (n = 1)	2.65 (n = 1)	– 63.91 (n = 1)
Limestone	NT	0.17 (n = 1)	8.35 (n = 1)	5.31 (n = 1)	58.17 (n = 1)	52.86 (n = 1)

n no. of samples, *NT* not tested

Table 3 Comparison of major elements in coal waste HTA and ground limestone

Sample ID	Al ₂ O ₃ (%)	CaO (%)	Fe ₂ O ₃ (%)	K ₂ O (%)	MgO (%)	Na ₂ O (%)	P ₂ O ₅ (%)	SiO ₂ (%)	SO ₃ (%)	TiO ₂ (%)
Coarse	17.9	3.24	10.7	3.59	1.40	0.817	0.249	59.3	2.33	0.699
Fine	18.2	4.30	8.12	3.50	1.35	0.732	0.135	58.0	2.39	0.721
Limestone	0.22	49.67	0.31	0.06	1.63	0.050	0.001	10.1	0.38	0.040
Limestone	0.50	54.52	0.30	0.05	0.64	0.030	0.043	1.07	0.11	0.0005

Table 4 Comparison of trace elements in coal waste HTA and ground limestone

Sample ID	Ba (ppm)	Cr (ppm)	Cu (ppm)	Li (ppm)	Mn (ppm)	Mo (ppm)	Ni (ppm)	Pb (ppm)	Sr (ppm)	V (ppm)	Zn (ppm)
Coarse	480	132	59.5	46.1	264	43.6	80.0	114	113	279	293
Fine	561	125	54.2	61.1	282	21.6	54.3	80.1	109	205	213
Limestone	294	25.8	10.9	NT	171	2.5	11.5	ND	532	ND	34.5

Note ND not detected, NT not tested

Table 5 Particle size distribution of materials used in Proctor tests

Sieve opening (mm)	Percent retained		
	DP 1	DP 2	DP 3
4.76	63	58	54
1.68	20	18	20
0.42	11	10	11
0.75	5	4	4
< 0.075	1	10	11

CCPW; two received a blend of 93.3% CCPW and 6.7% dewatered FCPW by volume; and two received a blend of 86.7% CCPW, 6.7% dewatered FCPW, and 6.7% ground limestone by volume. Assuming a normal porosity of 16%, approximately 300 kg (650 lb.) of CCPW was packed into each column. Sand cone tests were performed on four columns to determine dry unit weight and moisture content achieved during construction. Proctor dry unit weights of 21.55 kN/m³ (137.3 pcf) and 14.27 kN/m³ (90.92 pcf) were achieved for DP 1 and DP 3 mix materials, respectively. The kinetic test columns were leached in free-draining mode for 19.3 months (16 leach cycles; Fig. 3).

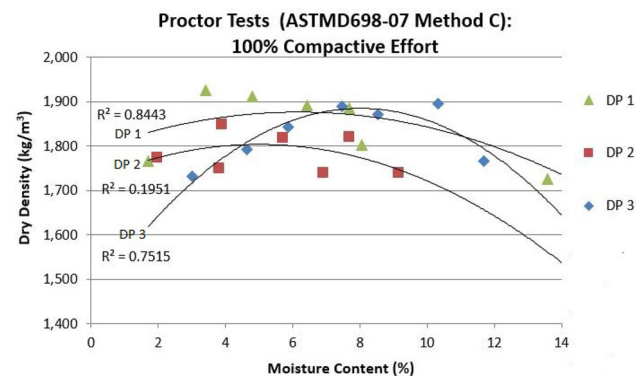


Fig. 2 Composite Proctor test results for DP 1, DP 2 and DP 3 options

inexpensive and readily available components; this second field experiment is the subject of this study. The revised field test columns were enlarged by using about 380-L (100-gallon), polycarbonate oval-tanks intended for live-stock activities. These stock tanks measured 113.4 cm (52.5 inches) long by 94 cm (37 inches) wide by 53.3 cm (21 inches) tall (Fig. 3). Two test columns received 100%

4.4.1 Leachate sampling and analysis

Pore-water and leachate samples were collected at about 38 days intervals using a 500 mL Nalgene® bottle. Field measurements of pH, temperature, and dissolved oxygen (DO) were conducted on each pore water and leachate sample with a professional-grade multi-parameter meter. In some cases, there was insufficient pore water sample volume to measure DO. Temperature, pH, and conductivity (specific conductance—SC) of pore water and leachate samples were measured immediately upon return to the laboratory. Alkalinity and ferrous iron were determined within 24 h after sample collection. All water sample quality analyses were statistically analyzed using spreadsheet-based models.

4.4.2 Anion analyses

Concentrations of SO₄²⁻, Cl⁻, F⁻, and nitrate (NO₃²⁻) anions were determined using an ion chromatograph (IC). For comparison, SO₄ concentration was also determined



Fig. 3 Field kinetic test columns (left); photograph of field test site (right)

Table 6 Average concentration of selected mine drainage parameters in leachate.*

refuse type	Interval (months)	pH	ORP	SO ₄	Cl	TDS	Alkalinity	Acidity	Fe	Mn	Al
CCPW-Only	≤ 7	8.02	0.132	3437	198.0	3865	236.6	9.0	0.76	0.89	1.22
	> 7	2.50	0.769	5449	10.8	10445	0.0	4909	1258	78.8	140.6
CCPW/FCPW blend	≤ 7	8.32	0.077	2490	197.2	2968	266.2	2.7	0.23	0.62	0.66
	> 7	3.56	0.621	4502	12.7	5253	18.2	1008	204.6	23.1	44.00
CCPW/FCPW/limestone blend	≤ 7	7.83	0.133	3093	126.2	3698	203.7	1.5	0.08	0.66	0.01
	> 7	5.82	0.454	3456	11.4	2549	35.9	100.2	12.43	7.10	5.14

* Data in mg/L except pH (median value), ORP (volts), acidity/alkalinity (mg/L CCE)

using colorimetric analysis (Hach Method 8051; Hach Company 2004).

4.4.3 Major and trace metal analyses

Major metals and a suite of trace metals were determined by either an atomic absorption mass spectrometer (AA-MS) or by an ion-coupled plasma optical emission spectrometer (ICP-OES). Additional metals analyses were performed by a commercial laboratory. These were supplemented by colorimetric tests for key mine drainage components: iron (Fe), manganese (Mn), and aluminum (Al) using the Hach FerroVer Method (Method 8008), Periodate Oxidation Method (Method 8034), and the Aluminon Method (Method 8012), respectively (Hach Company 2004).

Geochemical modeling was performed to predict reaction pathways and evaluate reaction kinetics. Models were used to better understand geochemical conditions such as pore water and leachate composition in column materials. Geochemical and modeling studies results are described in detail elsewhere (Behum et al. 2009, 2010; Lefticariu et al. 2014).

4.4.4 Kinetic studies results

4.4.4.1 Leachate chemistry Because this paper is focused more on assessing the overall geochemical advantages of the proposed DP 2 and DP 3 co-disposal practices over DP 1, only broad-based geochemical observations are made here. Table 6 shows the median pH of leachate collected from all columns during the initial leach period (≤ 7 months) ranged from 7.83 to 8.32, which was well above the carbonate buffer pH level of 6.37. However, there was a sharp decline in pH after that time for all DP 1 columns, and for three of the four blended refuse columns (DP 2 and DP 3). The median values of pH for DP 1, DP 2, and DP 3 during months 8–19 were 2.50, 3.56 and 5.82, respectively.

Table 6 also shows results for major anions monitored, including Cl⁻, HCO₃⁻ and SO₄²⁻. Of these, the HCO₃⁻ and Cl⁻ were the most easily leached anions. HCO₃⁻ is desired because it provides alkalinity needed to buffer the pH, but it declined after 7 months of testing. Cl⁻ declined from an average of 198.0 mg/L in the initial 7-month period to an average of 10.8 mg/L during the remaining test period for DP 1. Similarly, DP 2 and DP 3 showed

declines in Cl^- from 197.2 to 12.7 mg/L, and 126.2 to 11.4 mg/L, respectively. Thus, the discharge of the relatively easily leached and mobile anion Cl^- was relatively unaffected by the alternate disposal practices.

Conversely, the average SO_4^{2-} concentrations were overall greater and rose significantly after 7 months of leaching from an average of 3437–10445 mg/L in DP 12, 490–4502 mg/L in DP 2, and 3093–3456 mg/L in DP 3. Thus, the SO_4 concentrations in leachate from the blended columns were considerably lower especially in DP 3. Similar results were also obtained for major and trace cation (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Fe^{2+} , Mn^{2+} , Ni^{2+} , Zn^{2+} , Cd^{2+} , Sr^{2+} and Pb^{2+}) concentrations (Lefticariu et al. 2014). The principle cations during early leaching (< 7 months) were alkali metal ions sodium (Na^+) and potassium (K^+), and alkali earth elements calcium (Ca^{2+}) and magnesium (Mg^{2+}), which is consistent with the earlier laboratory leaching tests (Behum et al. 2009, 2010). Initial Na^+ concentrations were elevated in the initial 7-month period averaging 912.4, 839.1, and 809.1 mg/L for CCPW, CCPW/FCPW, and limestone blend leachate, respectively. Subsequently, Na^+ concentrations declined rapidly to relatively low levels in the final (> 7 months) leach period (203.5, 375.7, and 179.1 mg/L for DP 1, DP 2, and DP 3, respectively). Ca^{2+} and Mg^{2+} concentrations both increased during the study; however, Mg^{2+} increased at a much higher rate (from 30.8 to 253.4 mg/L in DP 1; similar, but less significant, increases in Ca^{2+} and Mg^{2+} concentrations were observed in DP 2 and DP 3 columns. Metals commonly associated with pyrite weathering (Fe, Mn, Al, Ni, Zn, Cu, and Cd) increased in the later leaching period (> 7 months), but were again lower in the DP 2 and DP 3 columns. These results are presented in detail in a research report by Lefticariu et al. (2014).

4.4.4.2 Pore-water chemistry Overall, the volume of pore water collected was much lower than the leachate volume. After 7 months of kinetic testing, all but two of the lysimeter ceramic sample cups ceased producing pore water samples because of Fe-rich precipitates. The remaining two lysimeters ceased producing pore water samples after 9 months of testing. This limited analysis of pore water chemistry during later leaching periods (> 7 months). During the kinetic testing, the average pore water sample collected was only 30.8 mL. The average SO_4^{2-} levels in pore water samples from DP 1, DP 2, and DP 3 columns were 2844, 3022 and 1995 mg/L, respectively, as compared to 3437, 2490, and 3093 mg/L, respectively, in leachate samples. Similar to the leachate sample observations, higher Cl^- concentrations were observed in pore water samples collected during the initial 7-month leach testing period. Again, additional data and discussion is provided in Lefticariu et al. (2014).

Elemental extraction in column leachate Data for most major and trace constituents were converted to a mass

(loading) basis by multiplying concentration values and leachate volume. This conversion allowed determination of the cumulative elemental extraction by calculating the percentage of leachate mass in each cycle, then comparing this to the original mass of the element within the column. Mass data was plotted as a function of time, which is represented by the leach cycle. The complete kinetic testing program consisted of 16 leach cycles completed in 568 days (19 months) with an average of 19813 mL of leachate collected for each cycle from all of the refuse columns. Leachate volume was then compared to the estimated pore volume of 54501 mL to yield an initial liquid-to-solids (L/S) ratio of 0.19. As a result, the average rate of pore volume flushing is approximately 0.36 volumes per leach cycle, which yields an approximate of 5.82 pore volumes that were leached over the course of the 19 month/16-cycle study. The cumulative extraction of S and Cl versus leach cycle is shown in Figs. 4 and 5. Although the extraction percentage was greater for Cl than for S during early leach cycles (< 7 months), overall amount of S extracted was higher, especially for CCPW (DP 1) columns. After 7 months of testing, S extraction increases in all leachate. However, only S extraction from DP 1 column leachate exceeded Cl extraction during this period.

Figure 6 plots the percentage extraction of major and selected trace elements during the 19-month kinetic test. S and Cl are the major anions in high total dissolved solids (TDS) discharges and Na and K are the major cations. Ca extraction was somewhat greater than Mg and Sr extraction; Mg extraction was elevated in the CCPW-only columns (DP 1). Mobility of Ca, Mg, and Sr elements commonly associated with calcareous elements were

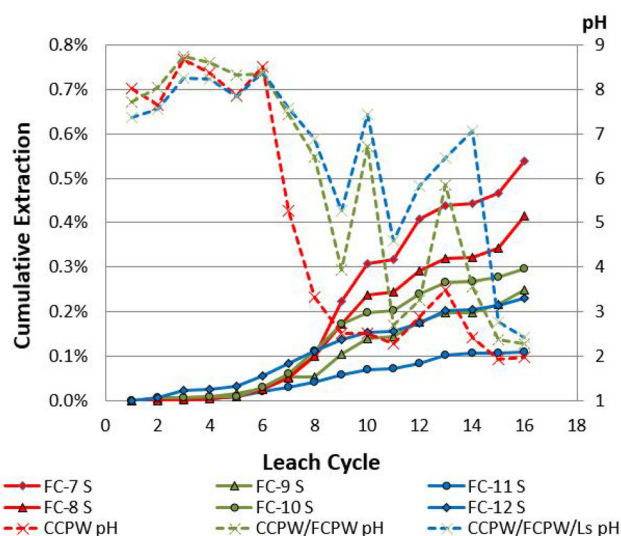


Fig. 4 Cumulative extraction of sulfur and pH (dashed lines) in different leach cycles

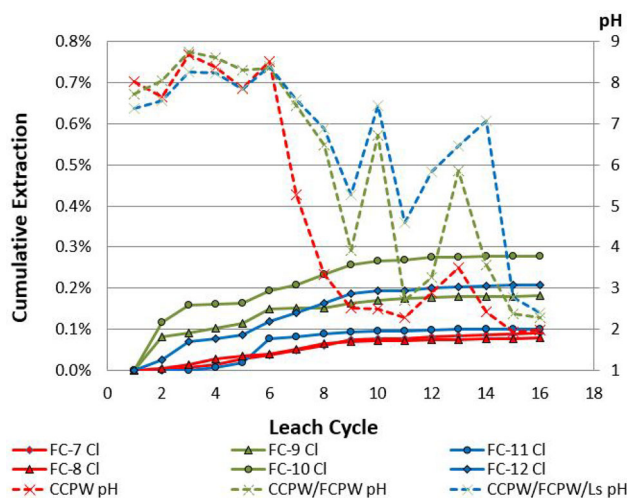


Fig. 5 Cumulative extraction of chloride and pH (dashed lines) in different leach cycles

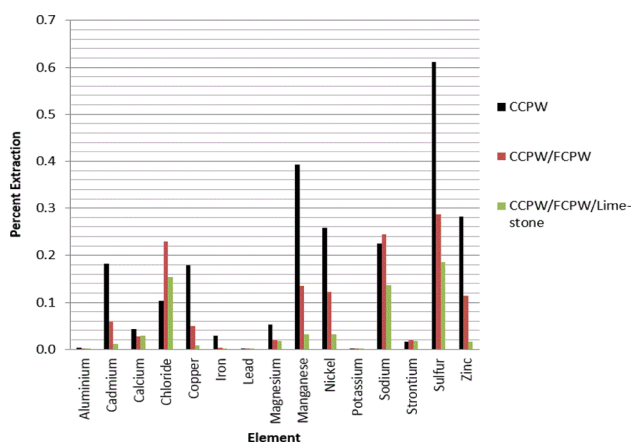


Fig. 6 Elemental extraction during 19-month kinetic test

relatively low during the 19-month kinetic test compared to Mn, Ni, and Zn trace elements that are commonly associated with pyrite (Fig. 6). Fe extraction from the DP 1 columns, and to a much later extent from CCPW/FCPW blend columns (DP 2), increased as pH dropped in later leaching periods (> 7 months). Mn, Ni, and Zn also had higher extraction rates than Fe suggesting considerable precipitation of Fe phases within the test columns. Only a very small amount of Fe extraction occurred in the limestone blend leachate (DP 3), which also maintained an overall higher pH (Lefticariu et al. 2014).

5 Conclusions and recommendations

5.1 Conclusions

- (1) The co-disposal practices of CCPW and FCPW with or without limestone addition are a significant

improvement over the current practice of disposing CCPW alone. The limestone addition practice further limits SO_4^{2-} release.

- (2) The proposed practice had minimal effect on release of chloride (Cl^-). Most of the discharge occurs soon after placement and must consider using water management practices of retention ponds, dilution, and discharge after large precipitation events.
- (3) The decline in HCO_3^- concentrations was much greater than sulfate (SO_4^{2-}) concentrations in alternate disposal practices. This suggests that either carbonate weathering rates are faster than pyrite weathering in a mine waste pile environment or that the alkalinity-producing minerals are being coated with mineral precipitates that are limiting dissolution.
- (4) Mobility of SO_4^{2-} was significantly lower in alternate disposal technology especially with limestone addition. The higher extraction of S compared to Ca suggests that the formation of calcium sulfate such as gypsum or anhydrite was relatively small.
- (5) Elements associated with alkalinity-producing minerals such as calcite and dolomite (Ca, Mg, and Sr) were leached to a greater extent than heavy metals typically associated with pyrite (Mn, Ni, and Zn) and lithophile elements (Al and K).
- (6) For the duration of experiments, the percentage of S extracted was smaller than Cl.
- (7) There was at least a 7-month period before additional fresh refuse or the final reclamation cover must be placed in order to avoid excessive release of SO_4^{2-} .

5.2 Recommendations

The encouraging results obtained in this study allow us to disseminate this alternate disposal concept to mine operators globally and seek input on how best to pursue commercialization of technology. Variables that might affect its success in the field need to be identified in cooperation with industry prior to wide-scale implementation of this technology. The authors have identified several areas for further studies that would aid commercialization: (1) a larger scale leaching study should employ commercially dewatered fine coal processing waste; (2) laboratory- and field scale leaching studies on DP 2 and DP 3 (co-disposal) alternatives should consider alternative alkaline amendment materials such as cement kiln dust (CKD), acetylene production waste lime, and steel slag, which may be available at a lower cost than agriculture ground limestone; (3) field demonstration of commercial dewatering technologies to dry coal waste to the necessary solids content of

about 65% is needed to develop an economic analysis; and (4) a comprehensive economic analysis of the alternative disposal technologies should be completed. The research team believes that the concepts presented here should be considered for developing other innovative concepts for management of coal waste materials to improve both the economics and environmental impact of disposal.

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