ABSTRACT

The objective of this study was to demonstrate through field-scale column leaching experiments that co-disposal of coarse and fine coal refuse provides both the geotechnical stability needed to lower refuse facility liabilities and the geochemical environment necessary to minimize sulfate and chloride discharges. Additions of limestone sand (aglime) were also evaluated as an improvement in overall geochemistry by buffering higher pH values within waste columns. The study was intended to aid mine operators in developing and implementing innovative concepts for engineered waste disposal at coal mine sites in Illinois. These concepts have the potential to greatly reduce long-term environmental liabilities by restricting the rate of coal and especially pyrite weathering thereby minimizing sulfate discharges to levels acceptable for performance bond release.

Field tests revealed a rapid decline in bicarbonate ($\text{HCO}_3^-$) concentrations as compared to sulfate ($\text{SO}_4^{2-}$) concentrations suggesting that either weathering rates are faster for carbonate than for pyrite in a mine waste pile environment or that alkalinity-producing minerals are being coated with mineral precipitates that are limiting dissolution. The mobility of sulfate was significantly lower in blended column leachate especially with limestone addition. Higher pH conditions in these columns favor precipitation of both oxy-sulfate minerals such as K- and Na-jarosite, which may be sequestering Fe and S. Both minerals were predicted to form during the late leaching period (> 7 months) in geochemical models. Conversely, greater mobility of Ca and Mg during the 19-month kinetic test was relatively low compared to elements that compose pyrite. Mn, Ni, and Zn showed a higher extraction rate than that of Fe suggesting that formation of multiple Fe-bearing minerals, hydroxides at higher pH (> 6), and sulfate minerals such as jarosite were combining to limit the mobility of Fe. Low pH in later leaching conditions lead to remobilization of metals previously sequestered within the column as secondary oxide and oxy-sulfate mineral weathering products. In this experiment, elements associated with alkalinity-producing minerals such as calcite and dolomite (Ca, Mg, and Sr) were extracted to a greater extent than heavy metals typically associated with pyrite (Mn, N, and Zn) and lithophile elements (Al and K). Also, S was extracted at considerably lower percentages than Cl. Additional Na in refuse and pore water samples could be derived from cation exchange (with Ca) in abundant clays associated with the ROM coal.
EXECUTIVE SUMMARY

This research was in response to the Illinois Environmental Protection Agency (IEPA) policy regarding implementation of lower standards for sulfate and chloride discharges from coal mining and processing operations. Historically, the allowed compliance level was 3,500 mg/L for sulfate discharges and 1,000 mg/L for chloride discharges, whereas “General Use” standards are set at 500 mg/L for both anions. More recently, the IEPA has agreed with the federal Environmental Protection Agency (USEPA) that this difference is not legally defensible and has implemented more restrictive sulfate levels, which vary due to chloride and hardness levels (USEPA, 2009). Current IEPA procedures establish sulfate discharge levels in a range of 500 to 2,000 mg/L based on hardness and chloride discharge levels at 500 mg/L.

The term “coal refuse” refers to coal processing waste. Coal refuse typically consists of two size fractions: 1) coarse coal processing waste (CCPW), which is generally larger than 150 micron (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 with solids 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, screenbowl centrifuges, and desliming cyclones, all of which is typically concentrated in a thickener. Coal refuse disposal facilities frequently associated with large underground mines have been identified as the principal sources of elevated sulfate discharges. Infiltration of oxygen- and ferric iron-bearing water into coal refuse stockpiles is believed to be a contributing factor in the release of sulfate from refuse materials as a result of pyrite weathering. Development of good management practices (GMPs) to restrict pyrite oxidation and thereby reduce sulfate levels in these discharges will meet present and future regulatory compliance levels for these constituents of mine drainage. These measures are intended to: 1) reduce water treatment costs during mining and subsequent complete reclamation of the mine facility, 2) reduce liabilities associated with maintaining and abandoning a large permanent impoundment structure, and 3) allow release of reclamation performance bonds in a timely fashion.

This research performed an evaluation of the hypothesis that co-disposal of CCPW and FCPW with appropriate compaction results in improved geochemical and geotechnical environments that will lead to minimization of acid mine drainage (AMD) formation and sulfate discharges. This hypothesis was based on extensive sampling data available in the literature and collected as part of a previous study requested by IEPA and funded by the ICCI (Chugh et al., 2007a). A preliminary 2-year laboratory column leaching study with 15.2 cm (6-inch) columns showed that co-disposal provided improvements in geochemical environment resulting in a 20% reduction in sulfate loading as compared to CCPW by itself. This study indicated that the net neutralization potential (NNP) for FCPW is generally near neutral and this material could be beneficially used to minimize AMD generation effects due to weathering of CCPW with a typically negative NNP (150-200 tons of calcium carbonate equivalent).
Discussions with most Illinois mine operators involved in the previous study indicated that they would like to co-dispose CCPW and FCPW if FCPW could be dewatered inexpensively and be shown to ensure embankment stability. Their objectives for co-disposal were to minimize space requirements for waste disposal and to eliminate slurry disposal areas that are very difficult to permit. None of the operators, however, referred to improvements in geochemical and geotechnical environments that would result from such practice.

A second hypothesis tested in this study was that addition of limestone to the co-disposal column can provide additional advantages by buffering drainage pH, thus reducing high levels of total dissolved solids and, more importantly, minimizing sulfate discharge. Both hypotheses were tested by constructing six waste 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 50% of Proctor density. All columns were equipped with a Lysimeter and a drainage port for collecting leachate, which was done during monthly sampling events.

Field column sampling results revealed a rapid decline in bicarbonate (HCO₃⁻) concentrations as compared to sulfate (SO₄²⁻) concentrations suggesting that either weathering rates are faster for carbonate than for pyrite in a mine waste pile environment or that alkalinity-producing minerals are being coated with mineral precipitates that are limiting dissolution. The mobility of sulfate was significantly lower in blended column leachate especially with limestone addition. Higher pH conditions in these columns favor precipitation of both oxy-sulfate minerals such as K- and Na-jarosite, which may be sequestering Fe and S. Both minerals were predicted to form during the late leaching period (> 7 months) in geochemical models. Conversely, greater mobility of Ca and Mg during the 19-month kinetic test was relatively low compared to elements that compose pyrite. Mn, Ni, and Zn showed a higher extraction rate than that of Fe suggesting that formation of multiple Fe-bearing minerals, hydroxides at higher pH (> 6), and sulfate minerals such as jarosite were combining to limit the mobility of Fe. Low pH in later leaching conditions lead to remobilization of metals previously sequestered within the column as secondary oxide and oxy-sulfate mineral weathering products. In this experiment, elements associated with alkalinity-producing minerals such as calcite and dolomite (Ca, Mg, and Sr) were extracted to a greater extent than heavy metals typically associated with pyrite (Mn, N, and Zn) and lithophile elements (Al and K). Also, S was extracted at considerably lower percentages than Cl. Additional Na in refuse and pore water samples could be derived from cation exchange (with Ca) in abundant clays associated with the ROM coal.

Using concepts tested in this project, the authors developed an alternate waste management system that uses a smaller mono-fill to replace the large traditional slurry impoundment. Although testing of this system was done at an Illinois mining operation, the concepts involved should be equally valid throughout the Illinois Basin as well as, with minor modifications, at other US coal mines.
OBJECTIVES

The main objective of the proposed study was to demonstrate through field-scale column leaching experiments that co-disposal of coarse and fine coal refuse provides both the geotechnical stability needed to lower refuse facility liabilities and the geochemical environment necessary to minimize sulfate and chloride discharges. Additional improvements by addition of limestone sand were also evaluated in this study. The study was intended to aid coal mine operators in developing and implementing innovative concepts for engineered waste disposal.

The specific goals of the study were to develop alternate coal processing waste management practices that will:

1. Minimize pyrite weathering and sulfate discharge;
2. Minimize the need for slurry ponds, which are increasingly difficult to permit;
3. Reduce water treatment costs during mining and upon completion of reclamation to allow for a timely release of performance bonds.

Meeting these goals will minimize environmental impacts to surface and ground water thereby complying with State of Illinois regulations promulgated under both the Clean Water Act (CWA) and the Surface Mining Control and Reclamation Act (SMCRA).

The project was divided into the following tasks:

Task I: Physical and Chemical Characterization of Refuse Materials

Physical (sieve analysis for particle size) and chemical (acid-base accounting for mineral content) properties of Illinois coarse coal processing waste (CCPW) and fine coal processing waste (FCPW) used in kinetic tests were determined.

Task II: Characterization of Geotechnical Properties for Refuse Disposal Options

Geotechnical properties of three refuse disposal alternatives were determined. Those options are: 1) CCPW disposal in embankments or fills (the control); 2) co-disposal of blended CCPW (90%) and FCPW (10%); and 3) co-disposal of blended CCPW (84%) and FCPW (8%) with limestone addition (8%). Geotechnical properties include moisture content, compaction potential, as well as compressive and/or shear strength.

Task III: Field-scale Kinetic Tests

Three sets of duplicate test columns (based on disposal alternatives described in Task II) were constructed using 100-gallon oval stock tanks. Columns were leached in free-draining mode for 19.3 months (16 leach cycles). Lysimeter and drainage samples were collected and analyzed to evaluate hydrologic conditions and geochemical environments with each column.
Task IV: Geochemical Modeling

Geochemical modeling was conducted 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.

Task V: Economic Modeling

Economic modeling was conducted to estimate the applicability of this research to full-scale coal processing and refuse disposal operations.

INTRODUCTION AND BACKGROUND

Coal refuse disposal facilities have been identified as a principal source of elevated sulfate discharges by both state and federal environmental regulatory agencies (The Advent Group, 2005; Chugh et al., 2007a; Chugh et al., 2007b). Infiltration of oxygen- and ferric iron-bearing water into coal refuse stockpiles is a contributing factor in the release of sulfate and metals from refuse materials as a result of weathering of the inherent sulfide mineral pyrite (Evangelou, 1995; Moses et al., 1987; Moses and Herman, 1991). There is a need to develop simple, reliable, and cost-effective management practices that restrict pyrite oxidation and inhibit the release of discharges with high concentrations of sulfate and metals. These measures or “good management practices” (GMPs) are intended to reduce water treatment costs during mining and prevent long-term problematic discharges which can hinder reclamation and bond release.

Weathering of pyrite can be controlled if water rich in oxygen and ferric ions access to sulfide minerals is restricted (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 are some of the parameters that control the rate of pyrite oxidation and the sulfate discharge process. The inherent amount of pyrite within the coal and the associated strata is an important factor in the amount of sulfate and metals in drainage from coal stockpiles and coal processing waste. Abiotic chemical conversion of ferrous iron to ferric iron is extremely slow; however, the presence of iron-oxidizing bacteria increases the rate of reaction by an order of magnitude and, as a consequence, the production of sulfates is much greater if weathering processes are mediated by microorganisms (Singer and Stumm, 1970; Kleinmann and Crerar, 1979). Within the Illinois Basin, researchers at the Cooperative Wildlife Research Laboratory (CWRL) at Southern Illinois University Carbondale (SIUC) have provided noteworthy contributions to the science and control of AMD generation (Nawrot and Gray, 2000; Naumann and Wiram, 1995; Wiram, 1984).

A typical Illinois Basin underground coal mining complex consists of a combination of mine shafts and slopes to access the coal bed along with coal preparation and refuse disposal facilities located on the surface as shown by the schematic in Figure 1. The term “coal refuse” as used in this report 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 process. Two size fractions of coal refuse are produced by the coal cleaning process: 1) coarse coal processing waste (CCPW) greater than 150 microns (100 mesh), and 2) fine coal processing waste (FCPW) less than 150 microns (100 mesh). In the Illinois Basin, FCPW has been disposed of as: 1) a slurry (± 15% solids) contained within an embankment constructed of CCPW (conventional practice), 2) dewatered FCPW (50-70% solids), which is also contained within CCPW berms as illustrated in Figure 1, or 3) a mono-fill constructed of a blend of CCPW and dewatered FCPW. The size of coal refuse disposal areas have been increasing in the Illinois Basin due to the recent trends of mining thinner coal seams with larger equipment capable of cutting roof and floor rock. This practice has increased the percentage of out-of-seam dilution (OSD) material mined to 50% or more of the ROM coal.

![Diagram](image.png)

**Figure 1**: Schematic of coal processing and water handling operations conducted on the surface at a typical underground coal mine complex.

Figure 1 also shows that these mine complexes typically operate a closed loop water-handling system where onsite water resources are continuously recycled and regulated. Surface water discharges typically occur from sedimentation basins, but only during larger precipitation events when less stringent regulations are applied and/or there is a considerable amount of dilution water available.
EXPERIMENTAL PROCEDURES

Physical Properties of Coal Refuse

Both fine and coarse refuse were collected from a large coal preparation plant cleaning ROM coal from two commonly mined seams: Springfield (No. 5) and Herrin (No. 6) both of the Carbondale Formation. Most of this coal is produced from two underground mines; however, a surface mine supplying one-fourth of the plant’s input may have also mined a small amount of ROM coal from the No. 5A and No. 7 coal beds. From this plant, an 18.9-liter (L) (5-gallon) sample of coarse refuse was collected and dried at low temperature (< 100°F), then screened for particle size analysis. A similar sized sample of fine refuse was collected from the preparation plant’s thickener underflow discharge line. This material was decanted and air dried before drying in the low temperature furnace. Finally, a 3.79-L (1-gallon) sample of agricultural ground limestone was also dried at low temperature and screened for particle size analysis. Several sieve analyses were performed on both CCPW and limestone samples.

After drying and particle size analysis, these materials were crushed to less than 3/8-inch in size. Standardized analyses using moisture-density curves were developed using 6-inch diameter molds, which were also tested for unconfined compressive strength to simulate un-drained conditions. By developing this series of moisture-density curves, the effect of the compaction effort on these physical properties was assessed and a maximum density and ideal moisture content were determined (Chugh et al., 2007a; Chugh et al., 2007b).

The term compaction or densification refers to reduction in void volume. It is achieved through energy applied as “compactive effort.” The effectiveness of this compactive effort depends upon the type of particles and the way the effort is applied. In cohesive materials, densification is primarily due to distortion and re-orientation of particles. With increasing moisture content, cohesion is reduced and the compactive effort becomes more effective. In a cohesionless soil or crushed rock, reduction in void ratio is achieved primarily through re-orientation and secondarily through fracturing of grains at points of contact. Again, with increasing moisture content (to a point), the effort becomes more effective. In coarse soils and broken rock, permeability may be large and pore water drains away easily. Limiting density is then determined by packing.

Chemical Analysis of Coal Refuse

Splits of CCPW and FCPW coal samples were sent to the US Geological Survey (USGS) Energy Resources Program (ERP) Energy Lab in Denver, Colorado for total sulfur content analysis and two additional samples were analyzed for pyritic sulfur content and total sulfur content by commercial laboratories. Total sulfur content was also determined on two samples of agricultural limestone used in the co-disposal refuse blend. Paste pH was determined for refuse samples in the SIUC geochemical laboratory. This data was then used to calculate potential acidity (PA) for coarse and fine refuse that yielded values of 120.37 and 63.91 tons CaCO₃, respectively, for every 1,000 tons of refuse. Neutralization potential (NP) was also determined for all column materials in a
commercial laboratory. By subtracting PA from NP, net neutralization potential (NNP) was determined.

In addition to conventional coal analysis, two Springfield (No. 5) coal refuse samples were subject to low temperature ash (LTA) preparation using a 750°C air-recirculating chamber furnace at the University of Kentucky. Both LTA samples were analyzed at the USGS-ERP Energy Lab.

**Experimental Design of Kinetic Tests**

Field-scale kinetic tests were conducted at an active coal mining complex operated by Peabody Energy between Equality and Eldorado in southern Illinois’ Saline County. Both surface and underground methods are used to mine the three seams with the largest reserves in the basin: Springfield (No. 5), Herrin (No. 6), and Danville (No. 7). Occasionally, the surface mine also recovers a small amount of a lower quality No. 5 rider coal called the No. 5A.

In the first phase of field-scale testing, three sets of duplicate test columns (FC-1 through FC-6) were constructed using 55-gallon barrels. These columns were monitored for 19 months and this project was to be a continuation of that monitoring. Unfortunately, in the first month of this project, a tornado severely damaged some of the columns. Consequently, the field-scale kinetic testing system had to be rebuilt. The new system was custom built and assembled from relatively inexpensive and readily available components. Columns were constructed with 100-gallon, polycarbonate, oval stock tanks measuring 113.4 cm (52.5 inches) long by 94 cm (37 inches) wide by 53.3 cm (21 inches) tall. Two columns received 100% coarse refuse; two columns received a blend of 93.3% coarse refuse and 6.7% dewatered fine refuse; and two columns received a blend of 86.7% coarse, 6.7% dewatered fine refuse, and 6.7% agricultural ground limestone. All percentages are by volume. These columns, shown in Figure 2, were leached in free-draining mode for 19.3 months (16 leach cycles).

![Figure 2: Schematic of kinetic test apparatus (left); photograph of field test site (right).](image-url)
The designated refuse mix was placed in each test cell over a 3.8-cm (1.5-inch) thick base layer of minus 1.25-cm (½-inch) chert gravel and compacted using a large electric-powered tamper. Embedded within the underlying chert gravel was a series of perforated 1.25-cm (½-inch) diameter PVC collection pipes that served as an underdrain. Geotextile fabric was used above the gravel to minimize underdrain clogging. Moisture analysis and laboratory compaction tests (Proctor analysis per ASTM D698-12e1 Method C; ASTM, 2013) were conducted prior to column construction to determine the amount of compaction for blended refuse columns (Behum et al., 2010). The compaction goal for all columns was to achieve 90% of the Proctor density. Sand cone tests were performed on four columns to empirically determine density and moisture content in those cells. After leaving a freeboard of about 5 cm (2.0 inches), approximately 45.7 cm (18 inches) of compacted coal refuse was placed in each test column.

**Sampling and Analysis**

Pore water and leachate samples were collected on average every 38 days using a 500 ml Nalgene® bottle. Volumes of both pore water samples and total collected leachate were measured with a graduated cylinder. Field measurements of pH, temperature, and dissolved oxygen (DO) were conducted on each pore water and leachate sample with a Hach Co. (Loveland, CO) Model HQ-40 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 SIUC geochemical laboratory using a Thermo Fisher Scientific (Waltham, MA) Orion 4-Star Plus laboratory-grade, multi-parameter meter. Alkalinity and ferrous iron were determined within 24 hours using a Hach Model AL-TL micropipette titration kit (Method 8203) and a Hach Model 890 colorimeter (Method 8146), respectively. All water sample analyses for this study were statistically analyzed using Excel 2013.

**Anion Analyses:** Concentrations of SO$_4^{2-}$, Cl$^-$, F$^-$, and nitrate (NO$_3^{2-}$) anions were determined using a Thermo Fisher Scientific/Dionex Model ICS-2000 ion chromatograph (IC). For comparison, SO$_4$ concentration was also determined by colorimetric analysis using the Hach SulfaVer Method (Method 8051).

**Major and Trace Metal Analyses:** Major metals and a suite of trace metals were determined by either a Teledyne Leeman Labs (Hudson, NH) Z-2000 Zeeman-corrected, atomic absorption mass spectrometer (AA-MS) at the SIUC geochemistry laboratory or by a Varian (Palo Alto, CA) Model 725 ion coupled plasma optical emission spectrometer (ICP-OES) at the USEPA-approved, Illinois Department of Natural Resources (IDNR), Office of Mines and Minerals (OMM) Analytical Laboratory in Benton, Illinois. Additional metals analysis was performed by the AcmeLabs commercial laboratory (http://acmelab.com). These metal analyses 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.
Geochemical Modeling

Geochemical modeling was conducted using the SpecE8 module of the Geochemist’s Workbench® program developed at the University of Illinois at Urbana-Champaign on each leachate where the analytical data set was complete. The “B-dot” equation option was implemented to calculate activity coefficients and whenever this value exceeded 15%, SpecE8 was allowed to adjust the balance by changes in $\text{SO}_4^{2-}$ levels.

Economic Modeling

This exercise compared co-disposal of blended CCPW and FCPW with and without limestone addition to the conventional practice of constructing large slurry impoundments with an enclosing coarse refuse embankment.

RESULTS AND DISCUSSION

Task I: Physical and Chemical Characterization of Refuse Materials

Physical Characterization of Refuse Materials: Data listed in Table 1 are average particle size distributions for CCPW and limestone samples. As FCPW is uniformly less than 200 mesh (0.074 mm or 0.0029 inches), particle size analyses were not conducted on this material. A majority (64.52%) of CCPW was in the 4.75 to 50.8 mm (0.187 to 2.0 inch) size range. Limestone was finer with a majority (72.97%) in the 1.70 to 19.05 mm (0.066 to 0.75 inch) size range. The initial moisture content of CCPW was 11.5% whereas dewatered fine refuse was 31.9% (68.1% solids content). Conversely, the initial moisture content of agricultural limestone was only 0.1%. Assuming a normal porosity of 16%, approximately 300 kg (650 lb) of CCPW was packed into each sample barrel.

<table>
<thead>
<tr>
<th>Sieve Size</th>
<th>CCPW Average (%)</th>
<th>Limestone Average (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>US (mesh)</td>
<td>Metric (mm)</td>
<td>English (inches)</td>
</tr>
<tr>
<td>&gt; 2 inches</td>
<td>&gt; 50.80</td>
<td>&gt; 2.000</td>
</tr>
<tr>
<td>&gt; 0.75 inches</td>
<td>&gt; 19.05</td>
<td>&gt; 0.750</td>
</tr>
<tr>
<td>&gt; 4</td>
<td>&gt; 4.75</td>
<td>&gt; 0.187</td>
</tr>
<tr>
<td>&gt; 12</td>
<td>&gt; 1.70</td>
<td>&gt; 0.0661</td>
</tr>
<tr>
<td>&gt; 40</td>
<td>&gt; 0.400</td>
<td>&gt; 0.0165</td>
</tr>
<tr>
<td>&gt; 200</td>
<td>&gt; 0.074</td>
<td>&gt; 0.0029</td>
</tr>
<tr>
<td>&lt; 200</td>
<td>&lt; 0.074</td>
<td>&lt; 0.0029</td>
</tr>
</tbody>
</table>

Chemical Characterization of Refuse Materials: A complete evaluation of CCPW and FCPW samples and samples of agricultural limestone included: 1) acid-base accounting, 2) determination of major and key trace elements, 3) chemical analysis of LTA derived from CCPW and FCPW samples, and 4) XRF analysis of dried, ground refuse and limestone samples. Data listed in Table 2 is from acid-based accounting analyses. The first row of data was extracted from mine permit applications submitted by the
cooperating mine with analysis done by IDNR-OMM. The remaining data was compiled from analyses done by USGS-ERP. Also, Chugh et al. (2007a) presented additional acid-based accounting data for coal refuse samples collected from separate preparation plant fractions and found that: 1) the fine coal fraction represented 10 to 15% of the waste rock, and 2) the sulfur content is typically lower for fine coal waste.

Table 2: Geochemical properties of blended No. 5 and No.6 coal refuse samples.

<table>
<thead>
<tr>
<th>Refuse Fraction ( # of Samples)</th>
<th>Mean Sulfur Content (%)</th>
<th>Median Paste pH</th>
<th>Tons CaCO₃ per 1000 Tons Refuse</th>
<th>Net Neutralization Potential</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Total (n=2)</td>
<td>Pyritic (n=47)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mine Permit (Coarse)</td>
<td>5.70</td>
<td>3.41</td>
<td>7.12</td>
<td>106.4 (n=47)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>23.8 (n=47)</td>
</tr>
<tr>
<td>Coarse</td>
<td>4.84</td>
<td>3.90</td>
<td>5.92</td>
<td>121.88 (n=1)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1.51 (n=1)</td>
</tr>
<tr>
<td>Fine</td>
<td>2.56</td>
<td>2.13</td>
<td>6.31</td>
<td>66.53 (n=1)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2.65 (n=1)</td>
</tr>
<tr>
<td>Limestone</td>
<td>0.17</td>
<td>8.35</td>
<td>5.31</td>
<td>58.17 (n=1)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>52.86 (n=1)</td>
</tr>
</tbody>
</table>

Elemental data and chemical analyses of LTA are presented in Tables 3 and 4. Both LTA samples were analyzed at the USGS-ERP Energy Lab using a Perkins Elmer Optima 5300 DV ICP-OES for major elements in oxide form in Table 3 and a Perkins Elmer/Sciex Elan 6100 ICP/MS for all trace elements in Table 4 (except Ba and Sr, which were done by the ICP-OES). Limestone analyses were done by the IDNR-OMM using a Varian 725 ICP-OES for major elements in oxide form shown in Table 3 (except SO₃, which was done by a LECO sulfur analyzer) and Mn, Ni, and Zn shown in Table 4. Other trace elements shown in Table 4 were analyzed by an OSMRE Niton XRF. The second row of limestone data in Table 3 was provided by a southern Illinois quarry.

Table 3: Comparison of major elements in coal refuse LTA and blended limestone.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Al₂O₃ (%)</th>
<th>CaO (%)</th>
<th>Fe₂O₃ (%)</th>
<th>K₂O (%)</th>
<th>MgO (%)</th>
<th>Na₂O (%)</th>
<th>P₂O₅ (%)</th>
<th>SiO₂ (%)</th>
<th>SO₃ (%)</th>
<th>TiO₂ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coarse</td>
<td>17.9</td>
<td>3.24</td>
<td>10.7</td>
<td>3.59</td>
<td>1.40</td>
<td>0.817</td>
<td>0.249</td>
<td>59.3</td>
<td>2.33</td>
<td>0.699</td>
</tr>
<tr>
<td>Fine</td>
<td>18.2</td>
<td>4.30</td>
<td>8.12</td>
<td>3.50</td>
<td>1.35</td>
<td>0.732</td>
<td>0.135</td>
<td>58.0</td>
<td>2.39</td>
<td>0.721</td>
</tr>
<tr>
<td>Limestone</td>
<td>0.22</td>
<td>49.67</td>
<td>0.31</td>
<td>0.06</td>
<td>1.63</td>
<td>0.050</td>
<td>0.001</td>
<td>10.1</td>
<td>0.38</td>
<td>0.040</td>
</tr>
<tr>
<td>Limestone</td>
<td>0.50</td>
<td>54.52</td>
<td>0.30</td>
<td>0.05</td>
<td>0.64</td>
<td>0.030</td>
<td>0.043</td>
<td>1.07</td>
<td>0.11</td>
<td>0.0005</td>
</tr>
</tbody>
</table>

Table 4: Comparison of trace elements in coal refuse LTA and blended limestone.

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

Note: ND = not detected; NT = not tested
Task II: Characterization of Geotechnical Properties for Refuse Disposal Options

During this study, moisture-density relationships were developed for: 1) CCPW by itself (baseline), 2) CCPW mixed with FCPW (solids concentration > 60%) at a ratio of 15:1, and 3) CCPW mixed with FCPW and agricultural limestone at a ratio of 15:1:1. This was done by way of standard Proctor tests. All Proctor tests were performed using the Modified Proctor Test in a 6-inch mold (ASTM D698 Method C). Method C limits the top size of the test sample to material that passes a ¾-inch screen. This is reflected in particle size charts for the three particle size tests performed (Tests 1, 2, and 5) on CCPW materials. Moisture content varied between 2% and 17%. Figure 3 is an example of the CCPW particle size distribution used in the geotechnical analysis. Proctor tests of CCPW produced a family of curves. Because these tests showed considerable variation, a composite was constructed as shown in Figure 4 for use in the field test calibration (Peak Dry Density = 118 psf).

**Figure 3:** Particle size analysis: Proctor Test #1.

**Figure 4:** Composite of all CCPW Proctor tests.
Two additional tests (Tests 3 and 4) were performed on blends of CCPW and FCPW materials without limestone addition. Moisture content varied between 2% and about 9.5%. Figure 5 shows one of these CCPW/FCPW blend particle size distributions. Figure 6 shows a composite plot constructed with all data from CCPW/FCPW blend Proctor tests for use in the field test calibration (Peak Dry Density = 115 psf).

![CCPW/FCPW Blend Particle Size Analysis: Proctor Test #3 (Limestone = 0%)](image1)

**Figure 5:** Particle size analysis: Proctor Test #3.

![Proctor Test #3 and 4 - CCPW/FCPW Blend (ASTMD698-07 Method C): 100% Compactive Effort](image2)

**Figure 6:** Composite of CCPW/FCPW blend Proctor tests.

Two final tests (Tests 6 and 7) were performed on blends of CCPW and FCPW materials with limestone addition. Moisture content varied between 2.5% and about 11.5%. Figure 7 shows one of these particle size distributions. Figure 8 shows a composite plot
of the family of curves for limestone blend Proctor tests (Peak Dry Density = 118 psf). Moisture content at Proctor densities was 6.0%, 5.5%, and 7.5% for CCPW, CCPW/FCPW blend, and limestone blend columns, respectively.

![CCPW/FCPW/Limestone Particle Size Analysis: Proctor Test #6](image)

**Figure 7:** Particle size analysis: Proctor Test #6.

![Proctor Tests 6 and 7 - CCPW/FCPW/Limestone (ASTMD698-07 Method C): 100% Compactive Effort](image)

**Figure 8:** Composite of CCPW/FCPW/limestone blend Proctor tests.

Sand cone tests were performed on four columns to empirically determine density and moisture content. Proctor densities of 2.199.3 kg/m³ (137.3 lb/ft³) and 1.456.4 kg/m³ (90.92 lb./ft³) were achieved for CCPW and CCPW/FCPW/limestone blend columns, respectively, by applying blows from an electric tamper to compact each 3-inch lift within the test column.
Task III: Field-scale Kinetic Tests

Leachate Chemistry: As shown in Table 5, the median pH of column leachate collected 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 dramatic decline in pH during subsequent testing in all CCPW and three of the four blended columns with CCPW dropping to a median value of 2.50 while the CCPW/FCPW blend dropped to a median of 3.56 and the limestone blend dropped to 5.82.

Also shown in Table 5 are major anions monitored during the 19-month field leach test including Cl⁻, HCO₃⁻ and SO₄²⁻. Of these, HCO₃⁻ and Cl⁻ were the most easily leached anions. In CCPW columns, Cl⁻ declined from an average of 198.0 mg/L in the initial 7-month period to an average of 10.8 mg/L in the remaining test period. Similarly, Cl⁻ declined from an average value of 197.2 mg/L to 12.7 mg/L in the CCPW/FCPW blend columns and from 126.2 mg/L to 11.4 mg/L in the limestone blend columns. HCO₃ levels declined more rapidly tracking the decline in pH with levels dropping to less than detection levels toward the end of testing when pH was less than 4.5. Average SO₄²⁻ concentrations were overall higher and rose significantly after seven months of leaching from 3,437 mg/L to 10,445 mg/L. The SO₄²⁻ increase was lower in blend columns where it increased from 2,490 mg/L to 4,502 mg/L in CCPW/FCPW blend columns and from 3,093 mg/L to only 3,456 mg/L in CCPW/FCPW/limestone blend columns.

Table 5: Average concentration of key mine drainage parameters in column leachate.

<table>
<thead>
<tr>
<th>Refuse Type</th>
<th>Interval</th>
<th>pH</th>
<th>ORP</th>
<th>SO₄²⁻</th>
<th>Cl⁻</th>
<th>TDS</th>
<th>Alkalinity</th>
<th>Acidity</th>
<th>Fe</th>
<th>Mn</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>CCPW/FCPW/Limestone Blend</td>
<td>≤ 7 months</td>
<td>7.83</td>
<td>0.133</td>
<td>3,093</td>
<td>126.2</td>
<td>3,698</td>
<td>203.7</td>
<td>1.5</td>
<td>0.08</td>
<td>0.66</td>
<td>0.01</td>
</tr>
<tr>
<td></td>
<td>&gt; 7 months</td>
<td>5.82</td>
<td>0.454</td>
<td>3,456</td>
<td>11.4</td>
<td>2,549</td>
<td>35.9</td>
<td>100.2</td>
<td>12.43</td>
<td>7.10</td>
<td>5.14</td>
</tr>
<tr>
<td>CCPW/FCPW Blend</td>
<td>≤ 7 months</td>
<td>8.02</td>
<td>0.132</td>
<td>3,437</td>
<td>198.0</td>
<td>3,865</td>
<td>236.6</td>
<td>9.0</td>
<td>0.76</td>
<td>0.89</td>
<td>1.22</td>
</tr>
<tr>
<td></td>
<td>&gt; 7 months</td>
<td>2.50</td>
<td>0.769</td>
<td>5,449</td>
<td>10.8</td>
<td>10,445</td>
<td>0.0</td>
<td>4,909</td>
<td>1,258</td>
<td>78.8</td>
<td>140.6</td>
</tr>
<tr>
<td>CCPW Only</td>
<td>≤ 7 months</td>
<td>8.32</td>
<td>0.077</td>
<td>2,490</td>
<td>197.2</td>
<td>2,968</td>
<td>266.2</td>
<td>2.7</td>
<td>0.23</td>
<td>0.62</td>
<td>0.66</td>
</tr>
<tr>
<td></td>
<td>&gt; 7 months</td>
<td>3.56</td>
<td>0.621</td>
<td>4,502</td>
<td>12.7</td>
<td>5,253</td>
<td>18.2</td>
<td>1,008</td>
<td>204.6</td>
<td>23.1</td>
<td>44.00</td>
</tr>
</tbody>
</table>

Note: Data in mg/L except pH (median value), ORP (volts), and Acidity/Alkalinity (mg/L CCE)

Table 6 shows measured major and trace cation (Ca²⁺, Mg²⁺, Na⁺, K⁺, Fe²⁺, Mn²⁺, Ni²⁺, Zn²⁺, Cd²⁺, Sr²⁺ and Pb²⁺) concentrations. As was observed in earlier laboratory leaching tests (Behum et al., 2009; Behum et al., 2010), the principle cations during early leaching (< 7 months) were alkalai metal ions sodium (Na⁺) and potassium (K⁺), and alkalai earth elements calcium (Ca²⁺) and magnesium (Mg²⁺). Initial Na⁺ concentrations were elevated in the initial 7-month average (912.4, 839.1, and 809.1 mg/L for CCPW, CCPW/FCPW, and limestone blend leachate, respectively), then declined rapidly to relatively low levels in the final (> 7 months) leach period (203.5, 375.7, and 179.1 mg/L for CCPW, CCPW/FCPW, and limestone blend leachate, respectively). Ca²⁺ and Mg²⁺
concentrations both increased during the study; however, Mg\(^{2+}\) increased at a much higher rate (from 30.8 mg/L to 253.4 mg/L in CCPW columns). Similar, but less extreme increases were observed in blended columns. Metals commonly associated with pyrite weathering (Fe, Mn, Al, Ni, Zn, Cu, and Cd) also increased in later leaching periods, but again to a lesser extent in blended columns.

**Table 6:** Average concentration of selected major and trace metals in column leachate.

<table>
<thead>
<tr>
<th>Refuse Type</th>
<th>Interval</th>
<th>Ca</th>
<th>Na</th>
<th>Mg</th>
<th>K</th>
<th>Sr</th>
<th>Zn</th>
<th>Ni</th>
<th>Cu</th>
<th>Cd</th>
<th>Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>CCPW-Only</td>
<td>≤ 7 months</td>
<td>260.3</td>
<td>912.4</td>
<td>30.8</td>
<td>15.45</td>
<td>0.511</td>
<td>0.116</td>
<td>0.715</td>
<td>0.064</td>
<td>0.018</td>
<td>0.041</td>
</tr>
<tr>
<td></td>
<td>&gt; 7 months</td>
<td>314.3</td>
<td>203.5</td>
<td>253.4</td>
<td>11.52</td>
<td>0.539</td>
<td>11.88</td>
<td>6.942</td>
<td>1.398</td>
<td>0.023</td>
<td></td>
</tr>
<tr>
<td>CCPW/FCPW Blend</td>
<td>≤ 7 months</td>
<td>81.7</td>
<td>839.1</td>
<td>18.8</td>
<td>14.31</td>
<td>0.345</td>
<td>0.020</td>
<td>0.017</td>
<td>0.002</td>
<td>0.032</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&gt; 7 months</td>
<td>368.3</td>
<td>375.7</td>
<td>97.4</td>
<td>17.03</td>
<td>0.920</td>
<td>24.37</td>
<td>2.223</td>
<td>0.598</td>
<td>0.029</td>
<td></td>
</tr>
<tr>
<td>CCPW/FCPW/Limestone Blend</td>
<td>≤ 7 months</td>
<td>276.8</td>
<td>809.1</td>
<td>48.1</td>
<td>16.85</td>
<td>0.772</td>
<td>0.061</td>
<td>0.192</td>
<td>0.002</td>
<td>0.050</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&gt; 7 months</td>
<td>355.9</td>
<td>179.1</td>
<td>102.7</td>
<td>16.93</td>
<td>1.071</td>
<td>5.574</td>
<td>0.619</td>
<td>0.169</td>
<td>0.039</td>
<td></td>
</tr>
</tbody>
</table>

Note: All data in mg/L

Pore Water Chemistry: Due to operational problems, the volume of pore water was much lower than the leachate volume. After seven months of kinetic testing, all but two of the lysimeter ceramic sample cups ceased producing pore water samples because of iron plugging. The remaining two lysimeters ceased producing pore water samples after nine months of testing. This restricted 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 from lysimeter sample cups with 200 ml capacity.

Tables 7 and 8 provide a compilation of average key parameter values and major and selected trace metal levels found in pore water samples collected only during the first seven months of kinetic testing. In most cases pore water concentrations were somewhat lower than leachate concentrations during this period. For example, average SO\(_4^{2-}\) levels in pore water samples from CCPW, CCPW/FCPW, and CCPW/FCPW/limestone columns were 2844, 3022 and 1995 mg/L, respectively, compared to 3437, 2490, and 3093 mg/L, respectively, in leachate samples. As with leachate samples, higher Cl\(^-\) concentrations were observed in pore water samples collected during the initial 7-month leach testing period; however, this may be the result of fewer samples being collected and the potential for dilution errors associated with the low volume of pore water samples.

**Table 7:** Average concentration of key mine drainage parameters in column pore water.

<table>
<thead>
<tr>
<th>Refuse Type</th>
<th>pH</th>
<th>SO(_4^{2-})</th>
<th>Cl(^-)</th>
<th>F(^-)</th>
<th>Acidity</th>
<th>Fe</th>
<th>Mn</th>
<th>Al</th>
<th>Ni</th>
</tr>
</thead>
<tbody>
<tr>
<td>CCPW-Only</td>
<td>7.65</td>
<td>2,844</td>
<td>849.5</td>
<td>4.92</td>
<td>1.07</td>
<td>0.787</td>
<td>0.054</td>
<td>0.025</td>
<td>0.212</td>
</tr>
<tr>
<td>CCPW/FCPW Blend</td>
<td>7.66</td>
<td>3,022</td>
<td>356.4</td>
<td>4.59</td>
<td>1.69</td>
<td>0.516</td>
<td>1.952</td>
<td>-0.001</td>
<td>0.378</td>
</tr>
<tr>
<td>CCPW/FCPW/Limestone Blend</td>
<td>7.76</td>
<td>1,995</td>
<td>515.6</td>
<td>4.41</td>
<td>3.02</td>
<td>1.555</td>
<td>1.182</td>
<td>-0.001</td>
<td>0.260</td>
</tr>
</tbody>
</table>

Note: Data in mg/L except pH (median value) and Acidity (mg/L CCE)
Table 8: Average concentration of selected major and trace metals in column pore water.

<table>
<thead>
<tr>
<th>Refuse Type</th>
<th>Ca</th>
<th>Mg</th>
<th>Na</th>
<th>K</th>
<th>Sr</th>
<th>Zn</th>
<th>Cu</th>
<th>Pb</th>
<th>Cd</th>
</tr>
</thead>
<tbody>
<tr>
<td>CCPW-Only</td>
<td>129.1</td>
<td>35.95</td>
<td>268.1</td>
<td>48.77</td>
<td>0.103</td>
<td>0.052</td>
<td>0.019</td>
<td>0.006</td>
<td></td>
</tr>
<tr>
<td>CCPW/FCPW Blend</td>
<td>321.4</td>
<td>52.32</td>
<td>449.1</td>
<td>13.12</td>
<td>0.448</td>
<td>0.766</td>
<td>0.113</td>
<td>0.049</td>
<td>0.008</td>
</tr>
<tr>
<td>CCPW/FCPW/Limestone Blend</td>
<td>178.1</td>
<td>32.87</td>
<td>362.9</td>
<td>11.46</td>
<td>0.175</td>
<td>0.283</td>
<td>0.083</td>
<td>0.113</td>
<td>&lt;0.001</td>
</tr>
</tbody>
</table>

Note: All data in mg/L

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 19,813 ml of leachate collected for each cycle from all of the refuse columns. Leachate volume was then compared to the estimated pore volume of 54,501 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 16-cycle study.

The cumulative extraction of S and Cl versus leach cycle is shown in Figures 9 and 10. Although the extraction percentage was greater for Cl than for S during early leach cycles (< 7 months), overall S extraction was higher, especially for CCPW-only columns. After seven months, S extraction increases in all leachate; however, only S extraction from CCPW-only column leachate exceeds Cl extraction during this period. Fe extraction from CCPW-only columns, and to a much later extent from CCPW/FCPW blend columns, increased as pH dropped in later leaching periods (> 7 months), as shown in Figure 11. Mn, Ni, and Zn had higher extraction rates then Fe as shown in Figures 12 and 13. Only a very small amount of Fe extraction occurred in the limestone blend leachate, which also maintained an overall higher pH (see Table 7 and Figure 11).

Figure 13 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 extraction except for the CCPW-only columns. Ca extraction was also considerably greater than Sr extraction. Mobility of Ca, Mg, and Sr elements commonly associated with calcareous elements were relatively low during the 19-month kinetic test compared to Mn, Ni, and Zn trace elements that are commonly associated with pyrite.
Figure 9: Cumulative extraction of sulfur and pH (dashed lines) vs. leach cycle.

Figure 10: Cumulative extraction of chloride and pH (dashed lines) vs. leach cycle.
Figure 11: Cumulative extraction of iron and pH (dashed lines) vs. leach cycle.

Figure 12: Cumulative extraction of manganese and pH (dashed lines) vs. leach cycle.
Task IV: Geochemical Modeling

Geochemical modeling was conducted on all leachate samples and shows that reaction pathways are predominantly controlled by the master parameter, pH. Modeling data provided the basis for an evaluation of reaction kinetics.

Changes in Mineral Speciation versus Cumulative Leachate Volume: Saturation indices (SI) were determined for all leachate samples and plotted against normalized wash volume (cumulative leachate volume per unit of refuse materials in columns) as shown in Figures 14-19. Lab pH is also shown in the plots. For SI > 0, minerals were supersaturated and tended to precipitate. For SI < 0, minerals were undersaturated and tended to dissolve. Geochemical models of early (< 7 months) leachate were supersaturated (SI > 0) with respect to manganese oxide minerals such as todorokite [(Na,Ca,K,Ba,Sr)\(_{1-3}\)(Mn,Mg,Al)\(_6\)O\(_{12}\)•3-4H\(_2\)O], pyrolusite [MnO\(_2\)], and birnessite [(Na\(_{0.3}\),Ca\(_{0.1}\),K\(_{0.1}\))(Mn\(^{4+}\),Mn\(^{3+}\))\(_2\)O\(_4\)•1.5H\(_2\)O], along with iron hydroxides such as goethite [FeO(OH)] and amorphous [Fe(OH)\(_{3\text{(ppd)}}\)], and carbonates such as calcite [CaCO\(_3\)], aragonite [CaC\(_2\)O\(_3\)], and rhodochrosite [MnCO\(_3\)]. All of these minerals except goethite become undersaturated when pH dropped. Conversely, undersaturated conditions were predicted in all column leachate models during the early leach period for both K-rich jarosite [KFe\(^{3+}\)\(_3\)(OH)\(_6\)(SO\(_4\))\(_2\)] and Na-rich jarosite [(Na,K)Fe\(^{3+}\)\(_3\)(OH)\(_6\)(SO\(_4\))\(_2\)]. Then, the solution became supersaturated when pH dropped. Early period leachate modeling suggested that Al was more likely to form precipitates of gibbsite [Al(OH)\(_3\)] rather than
alunite $[\text{KAl}_3(\text{SO}_4)_2(\text{OH})_6]$. Gypsum is at or near supersaturation throughout the latter part of the test (> 7 months) when sulfate content increases significantly (see Table 5).

**Figure 14:** CCPW SIs of carbonate and manganese minerals.

**Figure 15:** CCPW SIs of iron and sulfate minerals.
Figure 16: CCPW/FCPW blend SIs of carbonate and manganese minerals.

Figure 17: CCPW/FCPW blend SIs of iron and sulfate minerals.
Figure 18: CCPW/FCPW/limestone blend SIs of carbonate and manganese minerals.

Figure 19: CCPW/FCPW/limestone blend SIs of iron and sulfate minerals.
Task V: Economic Modeling

The Advent Group, Inc. (2005) performed a study to evaluate the cost impact of reducing the current 3,500 mg/L sulfate standard for Illinois mines to the 500 mg/L general use standard. The Advent Group data was collected from the IEPA and an attempt was made to analyze it; however, some of the data was found to be from non-active, but permitted sites. Analysis results indicated that available data is too limited and lacks consistency in reporting methods to be amenable to statistical analysis. Furthermore, analytical test procedures used to determine trace metal concentrations for reporting utilized varying detection limits that in some cases were not congruent with allowable surface discharge quality and reporting standards. Consequently, it was determined that the amount and quality of available data is insufficient to justify fully completing this task during the time period of this project. The project team updated the comparison of Gardner et al. (2003) to a 2011 cost basis. This was done to allow consideration of technological improvements and increased environmental and permitting costs.

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

A rapid decline in bicarbonate (HCO$_3^-$) concentrations was observed as compared to sulfate (SO$_4^{2-}$) concentrations. 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. This finding is consistent with Hossner and Doolittle (2003) who observed similar differential weathering rates in coal mine overburden materials.

The mobility of sulfate was significantly lower in blended column leachate especially with limestone addition. This suggests that higher pH conditions in these columns favor precipitation of both oxy-sulfate minerals such K-jarosite and Na-jarosite, which may be sequestering Fe and S as suggested in a similar study by Huggins et al. (2012). In geochemical models, both minerals were predicted to form during later leaching periods (> 7 months). Conversely, the mobility of Ca and Mg during the 19-month kinetic test was relatively low compared to elements that compose pyrite. The higher extraction of S compared to Ca suggests that the formation of calcium sulfates such as gypsum or anhydrite was relatively minor.

Small amounts of Mn, Ni, and Zn are present in the refuse and are likely associated with pyrite (Cravotta, 2008; Huggins et al., 2012). Kinetic tests and modeling showed the extraction rate of these minerals to be higher than that of Fe. This suggests that the formation of multiple iron-bearing minerals, hydroxides at higher pH (> 6), and sulfate minerals such as jarosite, which are relatively stable at lower pH (< 5) were combining to limit the mobility of Fe. Especially in CCPW-only columns, Fe extraction increased as pH decreased. This corresponded to an increased rate of pyrite weathering along with dissolution of iron hydroxide and sulfate minerals in the late leach period (> 7 months), which is consistent with other studies (Watzlaf, 1988) where low pH in later leaching
conditions leads to remobilization of metals that were previously sequestered within the column as secondary oxide and oxy-sulfate mineral weathering products.

A much lower mobility was found for the small amount of lead that is in the refuse. This suggests that either Pb may not be associated with pyrite in the test columns constructed for this project or that there is a greater retention of Pb by absorption onto Fe hydroxides.

In this project, elements associated with alkalinity-producing minerals such as calcite and dolomite (Ca, Mg, and Sr) were extracted to a greater extent than heavy metals typically associated with pyrite (Mn, N, and Zn) and lithophile elements (Al and K). It should be noted that a considerably lower percentage of S is extracted than Cl. Additional Na in refuse and pore water samples could be derived from cation exchange (with Ca) in abundant clays associated with ROM coal. In contrast, some Na could have been sequestered within test columns especially during the latest leach cycles (> 12 months) by substitution for K in jarosite.

Because the SI for gypsum is at or near supersaturation while sulfate content increases significantly throughout the latter part of the test (> 7 months), it is concluded that gypsum is in equilibrium with the leachate solution as suggested in other studies (Zhu and Anderson, 2002).

**Recommendations**

Simple batch dewatering was used to produce FCPW for co-disposal with CCPW in kinetic tests conducted for this project. A larger scale leaching study should employ commercially dewatered FCPW from an industrial process in place of the simulated FCPW used in this study.

Additional laboratory-scale leaching studies on CCPW/FCPW blends should utilize alternative alkaline amendment materials such as cement kiln dust (CKD), acetylene production waste lime, and steel slag that are lower in cost than agriculture lime.

Detailed studies are necessary to determine the future economic and geotechnical viability of co-disposal practices.
REFERENCES


DISCLAIMER STATEMENT

This report was prepared by Dr. Liliana Lefticariu of Southern Illinois University Carbondale (SIUC), with support, in part, by grants made possible by the Illinois Department of Commerce and Economic Opportunity through the Office of Coal Development and the Illinois Clean Coal Institute. Neither Dr. Lefticariu and SIUC, nor any of its subcontractors, nor the Illinois Department of Commerce and Economic Opportunity, Office of Coal Development, the Illinois Clean Coal Institute, nor any person acting on behalf of either:

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