



## A FIELD DEMONSTRATION OF AN ALTERNATIVE COAL WASTE DISPOSAL TECHNOLOGY – GEOCHEMICAL FINDINGS

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## **Conventional Practice: Fine Coal Processing Waste Placed in Coal Slurry Impoundments**



Photo courtesy Jack Nawrot, SIUC (ret.)

## **Challenges with Conventional Practice**

- Slurry impoundments are increasingly more costly and difficult to permit, and may have an extended liability due to slope stability concerns and the potential for a long-term sulfate discharge.
- Coal processing waste (CPW) has increased due to greater mechanization and more difficult mining conditions (increased Out-of-Seam Dilution - OSD).
- Regulatory requirements regarding discharges of sulfate and chloride have increased for Illinois Basin coal mines.

# **Problem Identification\***

- Weathering of the mineral matter in coal mine waste can release elevated amounts of Sulfate (SO<sub>4</sub><sup>2-</sup>) and Chloride (Cl<sup>-</sup>).
- Sulfate discharge tracks the rate of pyrite weathering.
- Chloride discharge levels increase with increased crushing in mining and processing.
- Sulfate and chloride anions are "conservative" in the environment.

\*Illinois Clean Coal Institute Project: DEV05-8, Chugh *et al.*, 2007 See: <u>https://icci.org/reports/DEV05-8Chugh.pdf</u>

# Hypothesis 1: Co-disposal of Fine and Coarse Waste to Minimize Sulfate

- Fine CPW (FCPW) will fill voids in coarse CPW (CCPW) saving space within the refuse pile structure.
- Compaction characteristics can be improved by a broader particle size distribution and increased moisture content.
- Lower permeability for compacted, co-disposed waste will lower the sulfate and chloride mass in mine discharge.
- The increased neutralization potential (NP) of the FCPW can improve the blended refuse acid-base account (ABA).

# **Hypothesis 2: Water Management**

- Chloride (Cl<sup>-</sup>) is a conservative ion and will leach readily from coal and coal waste.
- A good management practices for Cl<sup>-</sup> control from coal refuse areas is to to apply dilution and allow a controlled discharge during periods of higher precipitation.

## **Testing of Hypotheses: Goals and Objectives**

- Two laboratory-scale kinetic tests demonstrated that:
  - Effective management of coal stockpiles will minimize SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup> leaching in mine discharge waters.
  - Co-disposal of CCPW and FCPW will improve geochemistry and reduce SO<sub>4</sub><sup>2-</sup> in mine discharge waters.
- Two field-scale test columns validating laboratory results for coal refuse disposal and demonstrated a desirable level of structural stability.

## Initial Field Kinetic Testing: 55-gallon Experiment (operated May 6, 2011 – September 14, 2012)





- 6 Columns: 57 cm (22.5-in.) diameter by 85 cm (33.5 in.) tall.
  - Porosity =  $16\% \rightarrow 201$  kg of coal refuse.
  - Duplicates: CCPW, Blended CCPW and FCPW, and a CCPW/FCPW/Limestone Blend.
  - The initial moisture: coarse refuse was ~ 11%, dewatered fine refuse was ~ 50 %.
  - Compacted to 50% of the Proctor density.
  - Monthly sampling events over 18 months.

## Operational Problems: Field Test Columns Severely Damaged by the February 29, 2012 "Leap Day" tornado outbreak



Damage to SIU 55-gallon kinetic test cells.

EF-4 tornado damage to Harrisburg, IL.



https://en.wikipedia.org/wiki/2012 Leap Day tornado outbreak

# **Reconstructed Field Columns**

## Improved 100gallon test cells

Improved column study funded by the Illinois Clean Coal Institute (ICCI Project 12/4C-5).

## Geochemical properties of blended Springfield (No. 5) and Herrin (No. 6) coal refuse samples

Refuse	Sulfur Content Mean (%)		Paste pH	MT of CaCO <sub>3</sub> equivalent/ 1,000 MT of Material		
Fraction	Total	Pyritic	(median)	MPA	NP	NNP
Permit Data (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***	4.55	3.90	6.01	136.6	1.51	-135.1
Fine***	2.56	2.13	7.41	79.06	2.65	-76.41
Blend***	4.15	3.55	7.31	125.1	1.74	-123.3

Analysis by the US. Geological Survey and Illinois Dept. of Natural Resources; \*\* reported in permit documents for the cooperative mine complex for underground mining of the No. 5 coal; \*\*\* from this study (n = 2).



## Geotechnical Studies: Particle size and Proctor analysis

Limestone additions allows an important increase in the moisture content at the peak density.



# **Improved Column Results**

**Cumulative Extraction** 

## **Mineralogy:**

# Mineralogical composition of the initial material.



## **Elemental Extraction:**

Normalized elemental concentration data to yield elemental mass loading.

## **Leachate Chemistry:**

#### **Elemental Concentration Trends**



## SEM images: Minerals in the Springfield No. 5 coal



Massive Pyrite



#### **Pyrite Framboids**



Galena







Gypsum and Kaolinite

Kaolinite

Calcite and Gypsum

# Multiple Geochemical Processes Occur at Solid/Aqueous Solution Interfaces

### Processes:

- 1. Adsorption
- 2. Desorption
- 3. Precipitation
- 4. Dissolution
- 5. Incorporation

## **Species Produced:**

- A. Aqueous ions
- B. Outer-sphere complex
- C. Inner-sphere complex
- D. Multinuclear complex
- E. Surface precipitates
- F. Solid solution



Charlet and Manceau (1993) In: Environmental Particles, Vol. 2, 117

## **New Field Columns:**

#### **Temperature Variations**

Installation: November 16, 2012 Sampling: December 10, 2012 Experiment Ended: July 11, 2014 Total Duration: 19.3 months



## Advantages of Field Column Kinetic Testing:

- 1) Full-sized particles are used--The impact of a scale factor is minimized.
- 2) The materials are exposed to "real world" environmental conditions.
  - a) Temperature.
  - b) Precipitation.

## **New Field Columns:**

#### **Precipitation Patterns**

Installation: November 16, 2012 Sampling Initiated: December 10, 2012 Experiment Ended: July 11, 2014





#### Carbondale, IL Precipitation

- The materials are exposed to "real world" environmental conditions.
  - Temperature. a)
  - Precipitation. b)

## Variations in Leachate pH



- Leachate pH declined during the testing for all columns, but an improved pH buffering was evident with the blended refuse.
- Temperature and precipitation had an important effect on leachate pH values, with a step decrease during the spring and summer and higher values during the winter.

### Variations in the Conductivity (SC) of the Leachate Solution



Leachate SC increased during the testing for all columns, but to a lesser extent with the blended refuse.

Temperature and precipitation again had an effect on leachate SC values:

- 1) A step increase in SC during the summer.
- 2) Lower SC values during the winter.

### Variations in the Total Alkalinity of the Leachate Solution.



Alkalinity in leachate declined rapidly during the first 8 months of testing.
 Some alkalinity remained in the columns simulating co-disposal with limestone addition.

### **Chloride Concentrations in the Column Leachate.**



10/18/2012 1/26/2013 5/6/2013 8/14/2013 11/22/2013 3/2/2014 6/10/2014 9/18/2014

- Chloride, sulfate and bicarbonate were the major anions.
- Chloride was the most readily leached anion, rapidly flushing from the columns.
- Bicarbonate declined at a rate that matched total alkalinity.

### **Sodium Concentrations in the Column Leachate**



- The alkali metals Na<sup>+</sup> and K<sup>+</sup> were the principle counter ions to Cl<sup>-</sup> in the leachate.
- Na<sup>+</sup> declined at a by factor of 10 during the leaching tests.
- Na<sup>+</sup> was present as water-soluble compounds, such as halides (NaCl), sulfates (Na<sub>2</sub>SO<sub>4</sub>), and possibly nitrates (NaNO<sub>3</sub>).

## Sulfate Concentrations in the Column Leachate



10/18/2012 1/26/2013 5/6/2013 8/14/2013 11/22/2013 3/2/2014 6/10/2014 9/18/2014

- Sulfate concentrations varied similar to temperature.
- Sulfate concentrations were lower in leachate from the blended refuse columns.

### Manganese Concentrations in the Column Leachate



Manganese concentrations varied similar to temperature and  $SO_4$  concentration trends. Manganese concentrations were lower in leachate from the blended refuse columns.

### **Iron Concentrations in the Column Leachate**



- Iron concentrations remained low for most of the experiment except for the CCPW columns.
  (> 11 months of testing CCPW leachate iron also tracked changes in temperature and SO<sub>4</sub>).
- Iron likely precipitated within the CCPW columns during the earlier testing.

## **Weathered Coal Samples**





Behum et al., 2014

# Normalization of Concentration Data

• Variations in precipitation altered field column infiltration rates and as a result the leachate volume.

## • <u>Example</u>:

Cl Load (mg) = Cl Concentration (mg/L)/ Leachate Volume (L)

- The Cumulative % Extraction is then determined using the Cl Load and the original mass of for example Cl contained in the column to determine the % extracted.
- Cumulative % Extraction is then the % Cl load that has accumulated for each sample interval throughout the kinetic test. In this case the sample interval was:
   Sample Interval = 602 day duration/16 samples = 38 days.

## Chloride Extraction and pH Trends during Field Kinetic Testing



- Cl extraction was higher in the FCPW/CCPW blend due to the addition of FCPW; Cl is more readily leached from fine-grained materials.
- Cl extraction was lower in the FCPW/CCPW/limestone blend due to increased compaction and lower hydraulic conductivity.

### Sodium Extraction and pH Trends during Field Kinetic Testing



- Na<sup>+</sup> is the counter ion to Cl<sup>-</sup> in sodium chloride (NaCl).
- As with Cl, Na extraction was higher in the FCPW/CCPW blend due to the addition of FCPW; Na is more readily leached from finer grained materials.
- Na extraction was lowest in the FCPW/CCPW/limestone blend due to most likely due to an increased compaction and lower hydraulic conductivity.

### Sulfate Extraction and pH Trends during Field Kinetic Testing



- SO<sub>4</sub> extraction (actually S extraction!) was higher in the CCPW; after 8 months the S was more readily leached from CCPW.
- S extraction was the lowest in the FCPW/CCPW/limestone blend, possibly due to increased compaction and lower hydraulic conductivity.

### Iron Extraction and pH Trends during Field Kinetic Testing



- Fe extraction was higher in the CCPW but only after 8 months of leach testing; Fe was more readily leached from CCPW.
- Fe extraction was the lowest in the FCPW/CCPW/limestone blend, which is most likely due to increased compaction and lower hydraulic conductivity.

### Manganese Extraction and pH Trends during Field Kinetic Testing



- Mn extraction was higher in the CCPW, but again only after 8 months of leach testing.
- Mn was more readily leached from CCPW and to a lesser extent the CCPW/FCPW blend.
- Mn extraction was the lowest in the FCPW/CCPW/limestone blend, which may be due to increased compaction and lower hydraulic conductivity.

### **Calcium Extraction and pH Trends during Field Kinetic Testing**



- Ca extraction was initially higher in the limestone-amended CCPW/ FCPW blend.
- In the early test period Ca was more readily leached from CCPW and the CCPW/FCPW/Limestone blend.
- Ca extraction was overall lowest for the CCPW/FCPW blend.



#### CCPW/FCPW/Limestone Columns:

- Carbonate minerals were stable 1) though most of the testing where the pH > 6.0
- Carbonate minerals dissolved 2) whenever pH lowered to <4.5

\*The SI is compared to the average 0.36 pore volumes flushed from the columns every 38 day leach cycle per the average weight of the blend in the column (kg).

### **Geochemical Modeling Results**\*

#### **CCPW Columns:**

Pyrolusite

- 1) Carbonate minerals were stable early leach testing when pH > 6.0
- 2) Carbonate minerals dissolved as the pH lowered to <4.5



## Zinc Extraction and pH Trends during Field Kinetic Testing



- Zn extraction was higher in the CCPW, but again only after 8 months of leach testing.
- Zn was more readily leached from CCPW and to a lesser extent the CCPW/FCPW blend.
- Zn extraction was the lowest by far in the FCPW/CCPW/limestone blend, which may be due to an elevated pH and increased compaction and lower hydraulic conductivity.

## **Elemental Extraction during Field Kinetic Testing**



- Many elements were more readily leached from CCPW and to a lesser extent the CCPW/FCPW blend.
- Chloride and sodium were more easily leached from CCPW/FCPW blends.

# Conclusions

- Verified hypotheses that a for Cl<sup>-</sup> control is to apply dilution and to meter a controlled discharge during periods of greater precipitation.
- Good management practices for SO<sub>4</sub><sup>2-</sup> control are to:
  - Compact and cover CCPW within 8 months;
  - Additional improvements are expected with co-disposal of CCPW and dewatered FCPW;
  - Even smaller SO<sub>4</sub><sup>2-</sup> loading is anticipated with limestone additions to the CCPW/FCPW blend.

# Recommendations

- Testing is needed for refuse derived from mining other important coal seams and for the No. 6 seam in central Illinois.
- Additional field kinetic test improvements are suggested:
  - Test cells should be scaled up to > 20 tons and include blends of mechanically dewatered FCPW.
  - Alternative low-cost sources for adding alkalinity (e.g., drying agents such as CCR or CKD) should be explored.

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# **Questions?**

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For more information see: <u>https://icci.org/reports/12Lefticariu4C-5Final.pdf</u>

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