# **LABORATORY WEATHERING STUDIES OF COAL REFUSE\***

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### ABSTRACT

Coal refuse samples from Staunton, Illinois, were continuously leached in soxhlet extractors at 97, 65, 55 and 45°C for up to 192 hr. An identical refuse sample was leached once a week for II weeks using a humidity cell at approximately  $25^{\circ}$ C. Leachate collected from the soxhlet and humidity cell apparatus was analysed for pH, EC, SO<sub>4</sub>, Fe, Mn, Al and total acidity.

The data indicate that concentrations of  $SO<sub>4</sub>$ , Fe and Mn increased rapidly as the leaching temperature in the soxhlets increased. Soxhlet extractors removed virtually all soluble Fe and Mn species within 64 hr of leaching. A comparison of the leachate characteristics of the low-temperature soxhlet and humidity cell showed that the soxhlet removed  $50\%$  more total Fe and SO<sub>4</sub> for the complete leaching cycle. Pyrite oxidation in the soxhlct extractors was observed near the end of the extraction sequence, but pyrite oxidation in the humidity cell was masked by the release of soluble iron sulphates. Thus, the origin of leachate acidity in the humidity cell could not be distinguished between the soluble mineral phase and the acidity resulting from the oxidation of pyritic minerals.

The results of this experiment suggests that the use of the soxhlet extractor for leaching coal refuse may produce a faster and more accurate representation of water quality impacts than would the use of the humidity cell. Long-term leaching studies must be completed to further clarify the role of both these leaching methods for predicting water quality impacts.

#### INTRODUCTION

When coal refuse is stored on the earth's surface, it is subject to an oxidizing environment and to leaching. As a consequence, drainage from the refuse typically contains a high hydrogen ion concentration, a high concentration of total dissolved solids (TDS) and total suspended solids, and an increased trace metals content? $<sup>2,3</sup>$ </sup>

Acidity and increased trace metal loading have been identified as the most serious problems associated with refuse drainage.<sup>3</sup> However, the occurrence of high concentrations of cations and anions, expressed as TDS, can severely degrade water quality and should not be ignored.<sup>4</sup> Prediction of water quality problems associated with refuse composition is a complex function of the physical and chemical properties of the refuse. Even with this inherent complexity, some means of predicting potential water quality must be utilized. The most expedient method is the use of laboratory procedures.

According to Carroll<sup>5</sup> and Loughnan<sup>6</sup> the most important factor in natural chemical weathering is the quantity of water that leaches through the weathering material. Consequently, laboratory experiments simulating rock weathering must control the rate and volume of the leaching solution. The first apparatus to fulfill these requirements was the soxhlet extractor used by Pedro<sup>7</sup> to study the geochemical weathering of crystalline rocks. After this initial use of the soxhlet apparatus, Henin and Pedro<sup>8</sup> and Williams and Yaalon<sup>9</sup> continued experiments of rock weathering using the soxhlet. Renton and Hidalgo'" also used the soxhlet extractor to determine the acid potential of coal.<br>Singleton and Lavkulich<sup>11</sup> modified the soxhlet

Singleton and Lavkulich<sup>11</sup> extractor for geochemical weathering by  $(a)$  reducing the siphon height by half, and  $(b)$  removing the entire siphon to create a direct flow of the leachate to the solvent reservoir. These major modifications provided a greater zone of oxidation and rapid leaching that more accurately simulated geochemical weathering. In addition, the original ceramic thimbles were discarded in favor of cellulose thimbles that prevent soil particles from clogging the siphon and contaminating the leachate. However, no modifications were made in order to adjust the extraction temperature.

The problem of high leaching temperature is still a significant drawback to the use of the previous soxhlet extractor. Sobek *et oL ~'-* modified the soxhlet by moving the extraction chamber away from the upward path of the relluxing solvent. The primary features of this modification, as compared to the Singleton and Lavkulich<sup> $\text{H}$ </sup> and Pedro<sup>7</sup> soxhlets, are illustrated in Fig. 1. Through this modification, temperature of the extracting solvent can be controlled in order to simulate more accurately the conditions encountered in the natural weathering environment. Temperatures as low as 30°C can be used; this allows micro-organisms to be introduced into the weathering process.

Concerned with acid production from coal and associated overburden, Hanna and Brant<sup>13</sup> developed a sample-cell supplied with constantly saturated water vapour. Samples in the cell could be leached with water at a constant rate from an overhead separatory funnel. In a  $s$  similar method used by Carussio<sup>14</sup> to evaluate acid mine drainage production, samples were placed in humidified plastic containers (humidity cells.)and flushed at regular intervals with deionized water. This method provides simple control of air, temperature, moisture and microbes. Since its development, the humidity cell has been used in numerous studies to characterize leachate

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Fig. 1 Comparison of the Singleton and Lavkulich soxhlet (left) with the Sobek soxhlet (right)

from coal and overburden materials.<sup>[5-18</sup> In most of these papers, laboratory procedures for utilising the humidity cell are not well outlined. However, Sobek et al.<sup>19</sup> provides detailed laboratory procedures for the humidity cell.

Of these simulated weathering studies, only  $C$ aruccio<sup>15</sup> characterized the acid potential of coal refuse. The results of these studies show that as pyritic sulphur content increases in refuse composition, the total cumulative acidity increased. However, geochemical weathering and production of high iron, manganese, and total dissolved solids were not examined in Caruccio's  $1975$  study.<sup>15</sup> Because coal refuse contains a high concentration of impurities and weathering products, soxhlet extraction should rapidly remove most of the soluble components. The use of soxhlet apparatus to simulate geochemical weathering of refuse should require only a few days, while the humidity cells would require weeks of weathering.

Both the soxhlet and humidity cell mthods may be adequate to characterize refuse water quality. The objective of this research is to compare weathering methods for the same refuse sample. Thus, by simulating the weathering of coal refuse in the laboratory, rapid and effective methods of predicting water quality characteristics can be developed. With this information. refuse disposal sites can be appropriately designed to prevent water pollution.

## MATERIALS AND METHODS

The coal refuse selected for this weathering study was taken from the gob pile of the abandoned Consolidated Coal Company's Mine No. 14 near Staunton, Illinois. The mine was opened in 1904 and produced the Herrin No. 6 coal seam for approximately 19 years. The coal refuse generated at this site has been exposed to weathering for more than 50 years.

Refuse samples were air-dried and processed for

chemical laboratory analysis according to Sobek *et al.<sup>19</sup>* Soil pH was measured electrometrically in a paste and in a  $1:1$  refuse-to-water suspension.<sup>20</sup> The acid-base account and sulphur fractionation were done according to the procedures of Sobek *et al.<sup>19</sup>* Soluble bases, electrical conductivity, cation exchange capacity, and saturation percent were done by soil survey methods.<sup>20</sup> Total elemental analysis, except for carbon, was accomplished by lithium metaborate-lithium tetraborate fusion:<sup>21</sup> carbon was found by a Leco carbon furnace.

Refuse minerology was determined using standard North American Phillips x-ray equipment. Conditions included  $CuKa$  radiation,  $34kV$ ,  $20mA$  scanning rate  $2^{\circ}$ /min, and  $1^{\circ}$  exit and receiving slits. The analyses are based on x-ray diffraction patterns of random powdered sample material (scanning from  $4^\circ$  to  $60^\circ$  20). Quantitative estimates were made by adding synthetic corundum to the sample and using the diffraction peak at 2.08 A as a reference.

Clay mineral analyses were conducted on the <0.004-mm' fractions of refuse samples. Clay-sized fractions were separated by wet sedimentation techniques.<sup>20</sup> Oriented aggregates of the clay minerals were prepared by evaporating portions of the clay-water suspensions onto glass microslides for analysis by standard x-ray diffraction procedures. Analyses included x-ray diffraction traces for: (*a*) the air-dried material; (*b*) after solvation with ethyene glycol; and  $(c)$  after heating to  $300^{\circ}$ C and  $550^{\circ}$ C. Quantitative estimates of the clay minerals were made following a procedure of Schultz.<sup>22</sup>

## *Soxhlet and humidity (.ell procedures*

Leaching of the Staunton refuse was simulated by using two different modified soxhlets. Each soxhlet received 100 g of refuse  $(<2mm)$  placed in a single-thickness cellulose extraction thimble,  $43 \times 123$  mm, and covered with l-cm layer of acid-washed quartz to minimize mechanical erosion and channelling. Two additional soxhlets were fitted with a gas-dispersion tube (Coming 39533 coarse fritted glass cylinder) inserted to within 5 cm of the bottom of the sample to insure an oxidizing atmosphere. Compressed air was passed through the samples at the average rate of  $150 \text{ cm}^3/\text{min}$ . The combination of soxhlets and leaching conditions that were used in these experiments is given in Table I.

Table 1 Soxhlet type and leaching conditions

no.	Soxhlet Soxhlet type (see Fig. $  \rangle$ )	Leaching conditions
	Singleton and Lavkulich <sup>11</sup>	Maintained at $97^{\circ}$ C. drip rate of 3.2 ml/min, no dispersion tube
	Singleton and Lavkulich <sup>11</sup>	Maintained at 65°C, drip rate of 3.2 ml/min, with dispersion tube
	Sobek et al. <sup>12</sup>	Maintained at $55^{\circ}$ C. drip rate of $2.4 \text{ ml/min}$ , with dispersion tube
	Sobek et al. <sup>12</sup>	Maintained at 45°C, drip rate of 2.4 ml/min, no dispersion tube

The samples in all four soxhlets were leached with distilled water for four 16-hr periods. Leaching in three of the soxhlets was then continued for 128 and finally 192 hr. Leaching temperature was controlled using a Glas-col (Model KJ-502) electrical heating unit. At the end of each leaching period, the solvent in the reservoir was replaced with an equivalent volume of distilled water. This was necessary because the saturation limit of the solvent can be exceeded over the leaching period. If the leachate contained solids, it was filtered and the solids were dried, weighed, and dissolved in boiling distilled water and a few millilitres of ultra-pure  $HNO<sub>3</sub>$  and diluted to 500ml. All leachates and redissolved solids were preserved for elemental analysis with ultra-pure HNO<sub>3</sub>.

Two hundred grammes of refuse  $(< 2$  mm) was evenly spread out in a humidity cell and the lid sealed (for detailed construction and procedures, see Reference 19). Each week, 300 ml of distilled water was added to the humidity cell and allowed to equilibrate for one hour. The leachate was extracted and sediment that was removed with the leachate was centrifuged, filtered and returned to the ceil. Dry air was passed over the wet sample for three days. At the end of this time, humidified air was passed over the sample for four days. The sample was then leached again and the cycle of dry air and humidified air was continued. All leachates were preserved for elemental analysis with ultra-pure HNO<sub>3</sub>.

All leachate samples from the humidity cell and soxhlets were analyzed for pH, electrical conductivity, total acidity and alkalinity, Fe, Mn, AI and SO4. Precipitates filtered from the soxhlet leachates were sujected only to elemental analysis. Leachate pH was measured electrometrically, and electrical conductivity was determined using a Wheatstone bridge. Titratable acidity and alkalinity was determined by the Hach Water Test Kit Titration Method. Soxhlet and humidity-cell leachates were analysed for total Fe, Mn and AI using a Perkin-Elmer 306 atomic absorption spectrophotometer. Sulphate was determined colorimetrically using a Coleman 55 Spectrophotometer.

# RESULTS AND DISCUSSION

Saturation and 1:1 pH measurements (Table 2) clearly indicate acidic conditions. The increase in pH from the saturation paste to the 1:1 water suspension is a predictable result, given the high electrical conductivity. A negative neutralization potential and large percentage of pyritic sulphur indicate the potential for long-term acid production.<sup>1</sup> High concentrations of total iron and manganese also indicate the potential for long-term release of these metals to drainage water. In addition, high electrical conductivity and soluble basic cations suggest that drainage water from the refuse will contain significant levels of dissolved solids.

Like the chemical data, the mineralogical analysis (Table 3) shows a high concentration of pyrite. Associated with pyrite are possible weathering products such as gypsum and jarosite;  $KF_{3}(SO_{4})_{2}(OH)_{6}$  or  $Fe<sub>3</sub>(SO<sub>4</sub>)<sub>2</sub>(OH)<sub>3</sub>2H<sub>2</sub>O$  forms may be present. Iron is also present in the form of siderite. Total clay content is approximately equal to the sum of quartz plus pyrite. This is significant since the clay fraction has important exchange properties. Kaolinite (50% of total clay) can

**Table** 2 Chemical analysis of refuse

Parameter	Value
pH (saturation paste)	2.4
pH(1:1)	3.0
Neutralization	$-18.0$ tons
potential	$CaCO3/1000$ tons
Total S	7.5%
Sulfate sulphur	1.0%
Pyritic sulphur	4.7%
Organic Sulphur	$1.8\%$
Total C	7.2%
Total Fe	$7.6\%$
Total Mn	1.7%
Soluble K	$0.1$ meq/litre
Soluble Na	0.4 meq/litre
Soluble Mg	39.9 meg/litre
Soluble Ca	31.4 meq/litre
EC (saturation paste)	$7.2 \text{ mm}$ ho/cm
Cation exchange	28.1 meq/100 g
capacity	refuse

Table 3 Mineralogy



+0 denotes % of total clay.

have a cation exchange capacity (CEC) of 3 to 15 meq/100 g of clay, and illite (20% of total clay) l0 to 40 meq/100 g of clay (24). More important, however, is the large component of degraded (weathered) illite (30% of total clay) in the refuse. Weathered illite (removal of K+ and oxidation of  $Fe<sub>2</sub>$ +) can have a CEC of 120 meq/100 g of clay (25). This potentially high CEC value, plus that of the organic fraction, are reflected in the CEC of the refuse (28.1 meq/100 g of refuse). A CEC of this magnitude could play a significant role in producing prolonged acid drainage by the release of exchangeable acidity.

For all the soxhlets, the highest values measured for each parameter, except pH, occurred after 16 hr of leaching and decreased rapidly to very low concentrations after 64 hr (Table 4). The highest levels of acidity, SO4, Fe and Mn occurred at the highest extraction temperatures and leaching rates and decreased linearly with reduced extraction temperatures. The relationship between the concentration of Fe extracted as a function of leaching time and temperature is illustrated in Fig. 2. Similar relationships exist for manganese, aluminium, and sulphate concentrations.

Soxhlet 1 was maintained at a higher temperature than

Soxhlet	Hours of	pH	EC.	Acidity, mg		Concentration, µg/g		
Number	leaching		mmho/cm $CaCO3/g$	refuse	SO <sub>4</sub>	Fe	Mn	Al
$\mathbf{1}$	16	1.7	17.0	27.8	34,500	8,153	63.0	828
	32	2.6	4.3	1.8	9,000	465	2.9	9.2
	48	3.1	2.7	0.7	5,250	71	0.0	2.7
	64	3.6	2.4	0.4	5,400	56	0.0	1.8
Total				30.7	54,150	9,105	65.9	842
$\overline{2}$	16	1.8	14.2	21.4	23,138	4,838	48.8	1,278
	32	2.5	4.3	2.7	7,013	780	4.7	44.7
	48	2.8	3.4	1.4	4,905	360	0.0	6.1
	64	3.1	2.8	0.7	5,100	161	0.0	2.7
	128	3.2	2.8	0.6	4,778	99	0.0	3.9
	192	3.6	1.6	0.4	2,414	51	0.0	2.6
<b>Total</b>				23.8	54,416	6,289	53.5	1,337
$\mathbf{3}$	16	1.9	10.1	15.3	14,513	3,338	37.5	918
	32	2.4	5.1	4.3	8,235	1,054	11.6	176
	48	2.4	4.7	4.7	7,725	926	10.9	218
	64	3.0	2.6	0.4	4,553	60	0.0	6.5
	128	2.5	4.8	1.0	8,850	81	2.1	13.5
	192	3.1	2.9	0.4	5,085	82	0.0	3.7
<b>Total</b>				25.8	48,961	5,541	62.1	1,335
4	16	2.0	8.7	12.8	15,150	2,700	31.1	873
	32	2.3	5.6	5.4	8,738	1,185	14.4	228
	48	2.5	4.5	2.8	7,245	596	6.9	85.2
	64	2.8	3.1	1.0	4,463	289	1.7	15.4
	128	2.2	6.1	0.9	4,883	329	2.6	19.5
	192	2.7	3.3	0.5	2,701	83	0.0	4.0
Total				22.5	43,180	5,182	56.7	1,124

Table 4 Soxhlet leachate data

Table 5 Humidity cell leachate data

		EC,	Acidity, mg	Concentration, µg/g				
Week	pH	mmho/cm	CaCO <sub>3</sub> /g refuse	SO <sub>4</sub>	Fe	Mn	Al	
	2.50	4.94	3.39	4284	656	11.3	298	
2	2.70	4.87	3.55	4872	722	12.4	331	
	2.64	4.09	2.17	3640	326	8.1	215	
4	2.70	3.60	1.10	2500	161	4.2	109	
ͻ	2.78	3.25	0.917	1825	133	3.3	78.4	
6	2.78	2.73	0.766	1673	82.5	2.9	64.6	
	2.79	2.79	0.680	1519	79.8	2.4	52,4	
8	2.79	3.70	1.05	2179	230	3.3	73.3	
9	2.94	1.92	0.360	659	67.8	1.0	26.4	
10	2.87	2.05	0.467	855	91.3	1.3	29.7	
11	2.88	2.03	0.413	1027	108	1.1	28.2	
<b>Total</b>			14.85	25033	2707	51.3	1306	



Fig. 2 Iron extracted with soxhlet apparatus as a function of leaching time and temperature

soxhlet 2, but soxhlet 2 was run three times as long. As a consequence, the total concentration of sulphate extracted by soxhlet 2 exceeds that extracted by soxhlet 1. These data show that temperature and leaching time are both important to the extraction process.

After 11 weeks of weathering using the humidity cell, concentrations of extracted metal, i.e. Fe, Mn and AI, and SO4 slightly exceed 50% of the concentrations recorded with the soxhlet apparatus. However, Fe levels extracted after 6 weeks have approached those obtained with the soxhlet after 48 to 64 hr. Total acidity for the humidity cell reached only 66% of that extracted with the soxhlets.

It should be noted that at 128 hr in soxhlets 3 and 4, Mn AI, Fe, SO4, EC, and total acidity had increased concentrations over the 64-hr extraction, while pH decreased. These data suggest that a small amount of mineral weathering other than dissolution of soluble species may have occurred. At 192 hr, concentrations were similar to the 64-hr extraction levels. Changes observed in these short extraction periods indicate that longer leaching times will be required to document the different phases of the. geochemical weathering process. These changes were not observed in the humdidity cell data.

Even though the leaching data of the soxhlets and humidity cell differ significantly, SO<sub>4</sub> is strongly correlated with EC, pH and Fe, as is Fe with Mn (Table 6). These results indicate that the same dissolution/oxidation processes are operating on the same mineral fractions. A lower correlation value for pH and SO4 from the soxhlets can be explained by the inclusion of high-temperature data, i.e. higher SO4 concentrations. At lower temperatures, the correlation coefficients for  $pH$  and  $SO<sub>4</sub>$  in both systems are approximately the same as the humidity cell value. All correlations in Table 6 are similar to results reported by Renton and Hidalgo,<sup>10</sup> Hanna and Brant,<sup>13</sup> and Caruccio.<sup>15</sup>

#### *Electrical conductivity and sulphate*

The EC values were 75% higher at the 16-hr extraction than at the 32-hr extraction for soxhlets 1 and 2. Soxhlet 3 had an EC value 50% greater, and soxhlet 4 was 33% greater than the 32-hr extraction. This clearly shows that a major portion of the soluble mineral fraction was removed in the first 16 hr of leaching and was highly dependent upon temperature and rate of leaching. This rapid release of soluble salts is most likely the result of the dissolution of sulphate minerals, i.e. gypsum andjarosite.

In the humidity cell, the EC decreases somewhat

Table 6 Correlation coefficients of common factors for soxhlet and humidity cell \*

	Soxhlet+		<b>Humidity Cell</b>		
	SO <sub>4</sub>	<b>Fe</b>	SO <sub>4</sub>	Fe	
EC	0.97		0.97		
	$-0.72$		$-0.87$		
pH Fe	0.98		0.93		
Mn		0.98		0.96	

\*Statistical significance at the 0.01 level. tAll data points from soxhlets 1, 2, 3, and 4 were included.



Fig. 3 Cumulative iron extracted with soxhlet apparatus as a function of time

uniformly from a high of approximately 5 mmho/cm to a low of 2 mmho/cm. This decreasing trend with time is mirrored by decreasing SO4 concentrations. When EC is plotted as a function of  $SO<sub>4</sub>$  concentration, these two parameters are highly correlated (Table 6).

Concentrations of Fe in the soxhlet extractions were highest at the maximum extraction temperature  $(97^{\circ}C)$ and lowest at the minimum extraction temperature  $(45^{\circ}C)$ . There was no noticeable increase in Fe in the leachate as a function of increased oxidation (Fig. 3). Iron concentrations increased only slightly after 64 hr of leaching. These results demonstrate that Fe is being released as a consequence of a dissolution reaction. Because Fe is correlated linearly (Table 6) with  $SO<sub>4</sub>$ , it can be assumed that iron sulphates, e.g. jarosite, are responsible. All data support the conclusion that chemical oxidation of pyrite and the release of Fe either did not occur or is so minor that it was masked be the release of soluble iron sulphates. These data are also supported by Stumm and Morgan.<sup>26</sup> They show that the rate of pyrite oxidation, under acidic conditions, requires 150 days to reach 5% of the complete reaction. Even at the elevated temperature of soxhlet 4, this rate of oxidation would not be detected.

Manganese concentrations follow the same general trends as Fe; however, after 48 hr of leaching, the Mn levels extracted from the soxhlets 3 and 4 are greater than those from soxhlet 2. This variation may occur because the modified soxhlets have a slightly higher leaching rate than the original soxhlets. This variation aside, almost all of the Mn extracted occurred within 64 hr. Once again, this suggests that Mn is being removed from a soluble mineral fraction. Because manganese is linearly correlated with Fe (Table 6), it is possible that Mn is associated with the soluble iron minerals. Unfortunately, there is not enough Mn in a mineral phase to be identified by x-ray analysis.

Like the soxhlet extractions, Fe and Mn concentrations are highly correlated (Table 6). Once again, this trend may indicate that Mn is associated with the soluble Fe phase. Because Fe and Mn levels decrease by 75% in just three weeks, these metals could be complexed with hydrous sulphates.

### *pH and acidity*

The pH levels of the soxhlet extracts were lower at the higher extraction temperatures; at the end of the leaching cycle, however, the pH was higher at the higher temperatures. This corresponds directly with a maximum extraction of Fe and  $SO_4$  (low pH) within the first 64 hr of leaching and almost complete extraction of Fe and SO4 at the end of leaching (higher pH). Sulphate is negatively correlated (Table 6) with pH, suggesting that leachate acidity may be associated with the dissolution of iron sulphates.

Total acidity data shows that as temperatures increases the level of acidity increased. However, after 64 hr of leaching the amount of acidity generated is extremely small. The pH remains somewhat stable but the amount of Fe and A1 in the teachate decreased dramatically. A possible explanation for this may be the release of



**Fig. 4**  Cumulative aluminium extracted with soxhlet apparatus as a function of time

exchangeable acidity, especially given the high CEC of the refuse.

As leaching progressed in the humidity cell, pH increased only 0.4 units. Given a decrease in concentration of metals in the leachate, the fairly stable pH readings could indicate the addition of exchangeable acidity to the system. Total acidity, however, decreased by 80% during the same period. This decrease will not necessarily be reflected by pH values since the change in total acidity is relatively small. With 4.7% pyritic-sulphur (Table 2), the total acidity curve is very similar to those of Caruccio.'

#### *Aluminium*

The results of the Al analysis in Fig. 4 suggests that, with an oxidizing environment, i.e. adding air to the soxhlet, higher concentrations of AI are extracted in the leachate.<br>
Temperature does not seem to be a significant factor for<br>
increasing solubility of aluminium. Soxhlets 2 and 3<br>
extracted more than 1300 ppm AI, soxhlet 4 extrac Temperature does not seem to be a significant factor for  $\sum_{i=1}^{\infty}$ increasing solubility of aluminium. Soxhlets 2 and 3 extracted more than 1300 ppm Al, soxhlet 4 extracted  $\frac{2}{9}$  3 1124 ppm, but soxhlet 1 (97 $\rm ^{o}C$ ) extracted only 842 ppm. Although soxhlet 4 did not have added air, its leaching  $\frac{2}{5}$  2 rate was less than soxhlet I and thus may have had greater oxygen availability. This behaviour may possibly be the result of the clay mineralogy. The refuse does not contain a significant amount of degraded illite; with the addition  $\frac{1}{2}$ of an extremely oxidizing environment, Fe in the illite could have been oxidized and thus aided the release of AI.

In the humidity cell, the release of AI to the leachate exceeded 300 ppm in the first two weeks of weathering and decreased to less than 30 ppm at the end of the study.

This decrease was fairly uniform; the majority of the AI however, was released in the first three weeks of leaching. This suggests that the release of A1 is the result of acid hydrolysis of the clay minerals.

#### **CONCLUSIONS**

Soxhlets remove the higher concentration of soluble materials within the shortest time when compared to the humidity cell, This is illustrated in Fig. 5, using extractable Fe as an example, and is due primarily to the higher temperatures and higher leaching rates used in the soxhlet. This comparison further demonstrates that



Fig. 5 Comparison of total extracted iron extracted by soxhlet and humidity cell apparatus as a function of time

weathered coal refuse contains a significant portion of soluble Fe, Mn and SO4 that may not be accurately recorded using the humidity cell. If only a humidity cell was used to characterize refuse drainage, the high pH, total acidity and metal content could be construed as resulting from the oxidation of pyritic minerals during laboratory leaching. While the production of acidity from the oxidation or pyrite using the soxhlet may be observed near the end of the extraction period, the presence of acidity from the oxidation of pyrite in the humidity cell may be masked by the release of a soluble phase.

The drainage from weathering coal refuse will contain significant levels of dissolved solids. With the use of the soxhlet extractor, the level of dissolved solids is considerably greater than in the humidity cell and more closely approximates the potential of the refuse to contribute high concentrations of soluble salts to drainage water. The production of high concentrations of dissolved solids is not surprising, considering refuse mineralogy and the mobilities of the cations and anions involved. The refuse is highly weathered, and relatively fresh refuse would contain significantly lower levels of soluble materials.

Weathering of coal refuse will release not only Fe but also a relatively high concentration of AI. This will ultimately increase the total acidity by the hydrolosis of Fe and AI. A high refuse CEC will prolong leachate acidity by releasing acidic cations, i.e. Fe, AI and H.

Simulation of coal refuse weathering can be more easily and quickly done with the soxhlets than with the humidity cells. However, further long-term studies are needed in order to fully characterize all the weathering reactions and water quality changes that may occur. Ultimately, soxhlet and humidity cell methods of weathering should also be tested on other materials, i.e. coal and overburden, to determine the application of simulated weathering methods on these materials in order to predict water quality.

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