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TITLE: CONTROL TECHNOLOGY STRATEGIES FOR COAL PREPARATION WASTE DRAINAGES

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Project Short Title: Control Technology Strategies for Coal Preparation Waste Drainages

The purpose of this project is to investigate in the laboratory the options available for treating high-sulfur coal refuse materials either at the preparation plant or during disposal to prevent or reduce subsequent releases of environmentally harmful trace elements during waste dump weathering and leaching. Such control technology might include (1) chemical or physical processing to remove the undesirable elements from the refuse or treating the refuse materials to immobilize these elements, (2) applying neutralizing agents, adsorbents or sealants at the refuse dump site, and burying, grading and compacting the waste materials to control the flow of water and air through refuse piles or (3) treating the contaminated drainage itself to reduce the contaminant concentration to environmentally acceptable levels. These three basic environmental control categories are discussed in this presentation.

Calcining to immobilize refuse constituents

The possibility that the release of toxic trace elements into the environment can be controlled by pre-treatment of coal preparation wastes has been investigated experimentally. One approach that we are examining is that of heat-treating the wastes to form what appears to be a glass-like structure that apparently locks the trace elements in the matrix and prevents their escape.

Several sets of calcining and leaching experiments were conducted to determine optimum heat treatment conditions necessary to chemically immobilize the potentially toxic trace elements in the refuse matrix. These experiments were performed using high sulfur coal preparation waste from a plant in the Illinois Basin. The waste was ground to -20 mesh and calcined in air at

600°C, 800°C 1000°C, and 1200°C for 2 h. The calcined residues were then statically leached (stirred with distilled H_2O) for a period of 48 h using a ratio of 4 mℓ H_2O to 1 g of calcined refuse, and pH's and total dissolved solids determined.

The effect of the refuse calcining treatments on the mineral composition of the refuse is illustrated by Tables 1 and II. Table I delineates the changes in refuse mineralogy that have occurred as a function of heat treatment Table II presents the mineral conversion reactions that occur at high temperature. It is seen from Table I that the two most environmentally active species, pyrite (marcasite) and calcite, have been transformed to high temperature phases by 600°C. By 1000°C even the clay minerals have been converted to structurally indefinable aluminosilicates. The x-ray diffraction analyses reported in Table I confirm that the mineralogical transformations listed in Table II have been effected by heat treatment in the range of 800 to 1000°C. Physically the samples calcined at 1000°C were fused or sintered at particle surfaces.

The success of the calcining treatment at reducing trace element releases during refuse leaching is illustrated by Table III, which lists trace element data from a comparison leaching test of calcined and uncalcined refuse samples. The refuse samples referred to in the table had been subjected to static leaching for 48 h. It is seen that the concentrations of the group of toxic elements listed are reduced in the leachates from the calcined refuse by as much as two orders of magnitude over the concentration in the raw refuse leachates. Leaching experiments were also conducted using acid solutions (pH < 2) with essentially the same results.

Calcining of acid coal refuse materials prior to disposal could produce several beneficial effects. Foremost among these is the conversion of an active, highly polluting waste material into a chemically and geologically inert mass that can be easily and safely disposed almost anywhere with ordinary landfill practices. In so doing calcining presents a highly effective and permanent solution to a most difficult waste control problem. A corollary to this is that the calcined refuse materials might be

TABLE I

EFFECTS OF CALCINING ON COAL REFUSE MINERALOGY

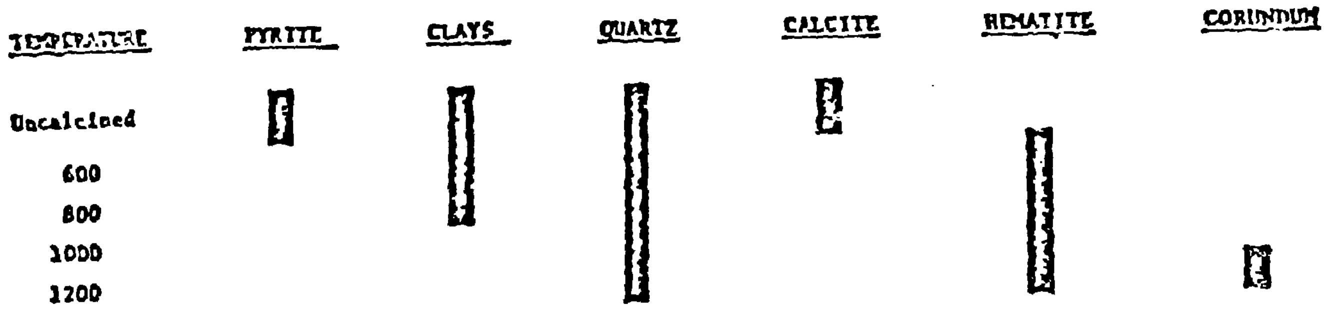

TABLE II

HIGH SULFUR COAL REFUSE

HIGH TEMPERATURE MINERAL TRANSFORMATIONS

Clays	Complex Aluminosilicates
Calcite	$CaO + CO_2 \uparrow$
Pyrite or Marcasite	$Fe_2O_3 + SO_2 \uparrow$
Quartz	Quartz

classified as nonhazardous under the criteria developed in conjunction with the Resource Conservation and Recovery Act (RCRA). This circumvents the need to meet the cumbersome and costly permit and performance requirements that RCRA dictates for the disposal of hazardous wastes, a category into which much, if not all, untreated high sulfur refuse materials will undoubtedly fall. Lastly, there is high potential for the recovery of by-products in connection with refuse calcining that does not exist for many of the other control technology schemes. Potentially recoverable products include sulfur, iron, and aggregated materials.

TABLE III

Trace Element Leachability of a High Sulfur Coal Refuse Sample Calcined at 1000°C

Element	Uncalcined Refuse	Calcined Refuse
Al[a]	100	0.4
Fe	600	< 0.03
Mn	5.8	0.03
Co	2.8	< 0.01
Ni	4.8	0.01
Cu	0.10	0.01
Zn	2.8	0.05
Cd (ppb)	68	0.3
pH	2.9	8.0
TDS (%)	1.4	0.2

[a] Leachate concentration reported as ppm unless otherwise noted (50 g crushed refuse/200 ml H_2O/48 h)

Codisposal of Wastes with Alkaline Materials

One of the major conclusions from our earlier studies of the environmental behavior of coal refuse materials concerned the importance of pH in controlling trace element releases during refuse leaching. In all instances when leachate pH was maintained at or near the neutral point, only minimal amounts of trace elements were solvated by the leachates. Conversely, when oxidative degradation of the pyritic materials in the refuse caused leachate acidities to build up, substantial quantities of such environmentally troublesome elements as Fe, Al, M[illegible] Cu, Co, and Ni were solvated by the acid leachates. This marked dependen[illegible] the degree of trace element contamination of refuse leachates on le[illegible] pH suggested that a fruitful means of preventing trace element releases from discarded refuse materials would be to add neutralizing agents to the refuse prior to disposal to reduce leachate acidity, in situ, as it is formed.

Column leaching experiments that utilized mixtures of powdered lime and high sulfur refuse were conducted to test the effectiveness of this in situ neutralization concept. These experiments proved to be very fruitful. For these experiments, powdered lime in varying amounts (3 to 50 g) was slurried in 150 mℓ of distilled water with -3/8 in. high-sulfur coal refuse (530 g, from Illinois Basin Plant B). The resultant mixture was subsequently dried in air at 50°C and recrushed to -3/8 in. particles. Four different lime concentrations were employed, 0.5, 1.5, 3, and 10% (by weight).

Column leaching experiments were conducted with about 500 g of each of the above samples to determine the effects of the lime additions. The refuse mixtures were packed into pyrex columns 25 cm long by 5 cm diameter and subsequently leached with distilled water at a flow rate of 0.5 mℓ/min until more than 4 ℓ water had been passed through the refuse beds.

A consistent pattern of the effects of the lime additions emerges from these data. The addition of 0.5 and 1.5 wt% lime to the acid refuse had only a small influence on leachate pH and trace element concentration because the acid neutralization provided by these amounts of lime was overwhelmed by the acid generating capability of the refuse. The additions of 3 and 10 wt% of lime, on the other hand, did indeed effectively counteract the acid properties of the refuse; the pH of the leachates for these two systems is higher; TDS values are relatively low, and the trace element concentrations are depressed.

The system containing 3 wt% lime is especially interesting because a leachate pH of 7 was maintained for nearly the entire duration of the continuous part of the leaching experiment (until 4.2 ℓ had been passed through the column). TDS values for this refuse-lime combination were also very respectable (ranging downward from about 0.6 wt%) especially considering that the dissolution of the lime itself adds substantially to the dissolved solids content of the solution. By the end of the leaching experiment, concentrations of troublesome trace elements, especially Fe and Mn, had been reduced to environmentally acceptable levels.

The codisposal of alkaline agents such as lime with acidic coal refuse materials does appear to be an attractive option for controlling trace element contamination of disposal area drainages. The technique is only moderately costly (we estimate \$0.50 to \$1.00 per ton of cleaned coal) and appears to be a highly effective means of preventing the release of a contaminated drainage from coal refuse dumps. The technology for mixing alkaline agents with coal refuse materials should be relatively simple and is immediately effective.

There are also a few uncertain aspects connected with the use of alkaline additives for coal refuse materials. One uncertainty involves the long term effectiveness or permanency of the method. The durability and immobility of the alkaline additives over long geologic periods must be demonstrated. Another potential drawback of codisposing alkaline additivies with high sulfur coal refuse materials concerns the RCRA classification of the resulting refuse-additive mixtures. It is not at all clear whether such a mixture would be classified as hazardous or nonhazardous. As pointed out earlier, a hazardous RCRA designation could be quite costly for the disposal site operator. Another somewhat negative aspect of refuse codisposal with alkaline agents as compared to refuse calcining, for example, is its low potential for by-product recovery. The lack of such potential, of course, negates the possibility of offsetting environmental costs with recovered product value.

Control of trace element contamination of refuse dump drainage

Water treatment is the best known and widely practiced of control technologies. While we have evaluated known and recognized control methods of water treatment for the coal waste drainages, (ion exchange, reverse osmosis, oxidation, chelation), we are most interested in controls that are realistic in terms of cost and ease of implementation; these are most necessary considerations in view of the enormous generation rate (greater than 100 million tons per year) of the coal wastes. One area that we have concentrated on is pH control by means of alkaline neutralization. The effect of pH on drainages from high sulfur coal wastes in the Illinois Basin has been investigated extensively during the progress of our investigations, and in Table IV we show some values of TDS vs pH derived from experiments conducted on laboratory generated leachates. During these studies, coal wastes from three different coal cleaning plants were used and the table represents overall averages.

TABLE IV

EFFECT OF pH ON TOTAL DISSOLVED SOLIDS FOR HIGH SULFUR COAL CLEANING WASTES

pH	TDS (%)
1.7	5.1
2	1.2
3	0.4
5	0.3
7	0.3

Alkaline neutralization is currently used extensively to treat acid drainage from coal mines. While it is well known that alkaline neutralization is very effective in controlling the acid and overall salt compositions of mine waste waters, the degree of control that this method exerts over some of the more highly leachable toxic trace elements remains to be established. Elaboration of this latter point is the basis for the study that we conducted in this area.

The experiments that we conducted were basically titrations in which limestone, lime, or lye (NaOH) were added to one liter of contaminated refuse drainage until a predetermined pH value was reached. The solutions (or slurries) were allowed to sit overnight, were filtered, the pH's were measured, and the compositions of the resulting solutions were analyzed.

As are the other control techniques discussed in this report, alkaline neutralization has been shown to be an effective method for reducing or abating trace element contamination of coal refuse drainage. The projected costs for such a treatment are relatively low ($0.10 to $0.80 per ton of cleaned coal) and the technique is relatively easy to apply, as evidenced by the large number of neutralization plants already in operation to treat acid mine drainage.

However, in spite of the low cost and ease of application, alkaline neutralization has some rather considerable disadvantages. For example, its use never really treats the source of contamination (i.e., the refuse itself) and treatment of drainage from the disposal site may be needed almost indefinitely. Also, although the standard refuse disposal practice at present involves burying the refuse on top of impermeable liners, such as clay, to channel refuse dump drainage into treatment areas, it is very difficult to insure that drainage will not eventually escape through or around these liners and thus negate the effectiveness of this method. Another consideration that may make alkaline neutralization less attractive involves the cost associated with meeting RCRA requirements. Very possibly waste materials disposed of in a way which produces contaminated drainage will be classified as hazardous. Thus the apparent low cost of alkaline neutralization may have to be tempered with additional costs needed to meet RCRA permit and performance requirements. Finally there is little opportunity for by-product recovery during or subsequent to neutralization treatment. Thus the potential for realizing economic gain in this way is quite low.

Summary

We have described our three categories of control strategies and have given examples of the effectiveness of each (see attached material from oral presentation). The conclusions are summarized in Table V.

TABLE V

SUMMARY MATRIX FOR THREE CONTROL TECHNOLOGY OPTIONS

	CALCINING	LIME CODISPOSAL	ALKALINE TREATMENT
COST	HIGH	MODERATE	LOW
EFFECTIVENESS	GOOD	GOOD	GOOD
PROCESS COMPLEXITY	HIGH	MODERATE	MODERATE
TREATMENT DURATION	SHORT	SHORT	VERY LONG
BY-PRODUCTS POTENTIAL	HIGH	NONE	NONE
PERMANENCY	GOOD	?	POOR
RCRA	NO HAZARD	?	HAZARDOUS

ENVIRONMENTAL CONTROL TECHNOLOGY FOR CONTAMINATED DRAINAGE FROM COALS AND COAL WASTES

by

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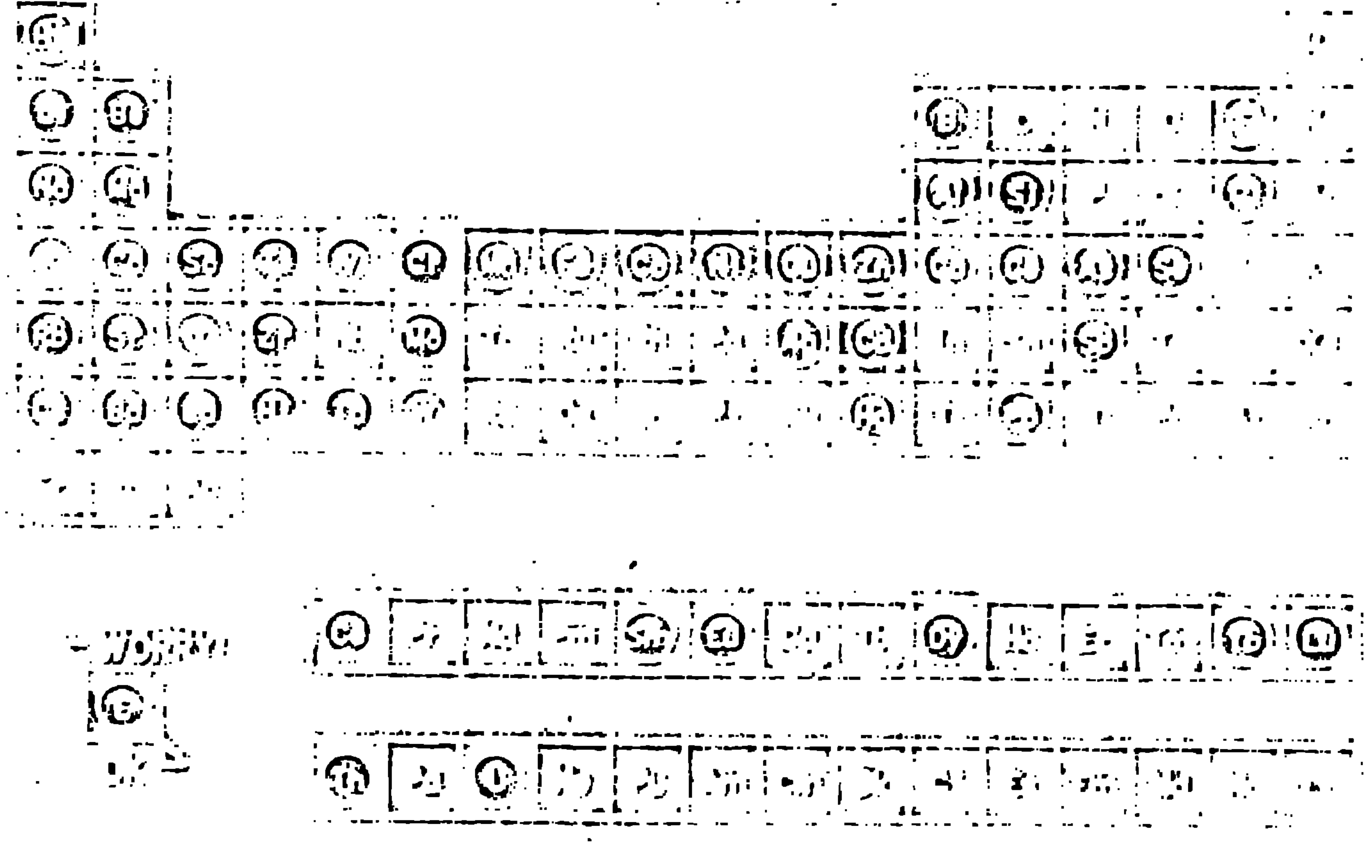
MEG/MATE ANALYSIS OF FLUIDIZED-BED COAL WASTE INCINERATOR



PREFERRED ORDER OF ENVIRONMENTAL CONTROL STRATAGIES FOR ACID COAL REFUSE

- Immobilize or remove toxic elements
- Prevent release of trace elements from refuse dump
- Treat contaminated water from refuse dump

IMMOBILIZATION OR REMOVAL OF TOXIC ELEMENTS

- Calcine
- Preleach acid forming constituents
- Preleach toxic trace elements

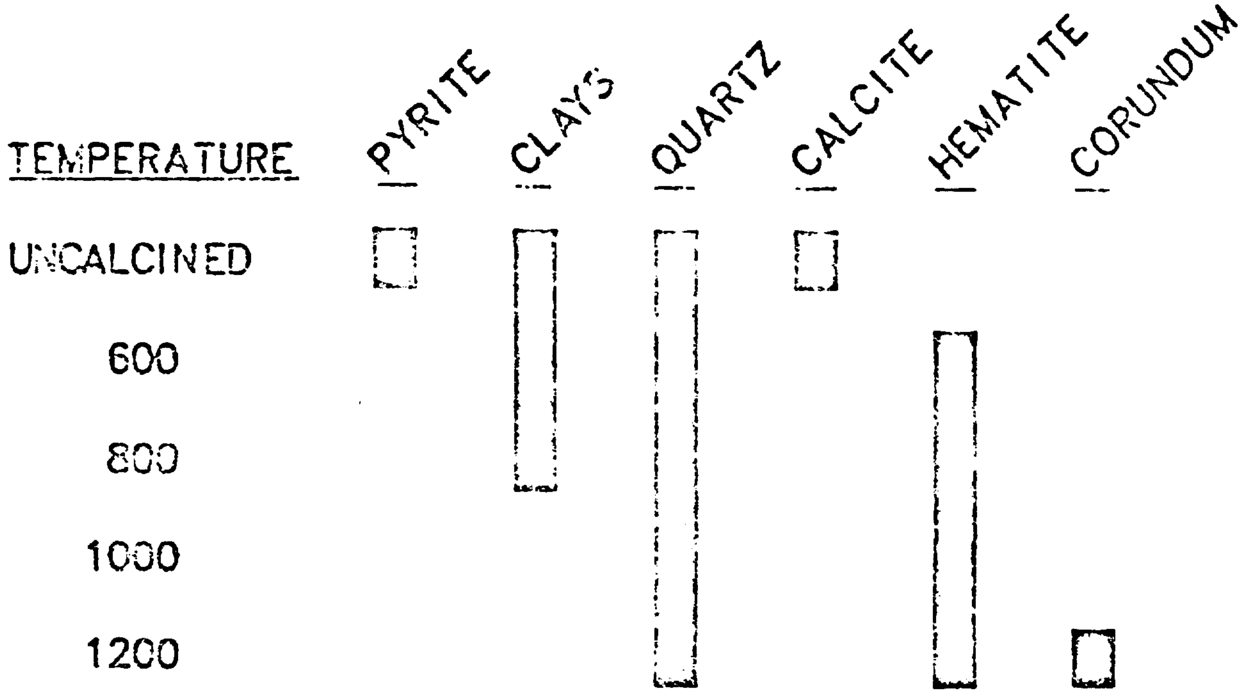
REFUSE CALCINING

To Immobilize the Toxic Contaminants in Acidic Coal Refuse by Converting It to an Inert Mass by Heat Treatment

MINERAL COMPOSITION OF ILLINOIS BASIN COAL REFUSE

MINERAL	AVERAGE (WT %)
Clays	37.3
Pyrite/Marcasite	30.2
Quartz	23.0
Calcite	0.4
Gypsum	0.5

EFFECT OF CALCINING ON COAL REFUSE MINERALOGY



TRACE ELEMENT LEACHABILITY OF A HIGH SULFUR COAL REFUSE SAMPLE CALCINED AT 1000°C

Element	Uncalcined Refuse	Calcined Refuse
Al	40	0.3
Fe	240	<0.02
Mn	2.3	0.02
Co	1.1	<0.01
Ni	1.9	0.01
Cu	0.04	0.01
Zn	1.1	0.04
Cd(ppb)	27	0.2
pH	2.9	8.0
TDS(%)	1.4	0.2

Leachate concentration reported as ppm unless otherwise noted

50g crushed refuse/200 ml H_2O/48h

ADVANTAGES OF REFUSE CALCINING

- HIGHLY EFFECTIVE
- PERMANENT POLLUTANT CONTROL
- SHORT TREATMENT DURATION
- HIGH BY-PRODUCT RECOVERY POTENTIAL
- RCRA NONHAZARDOUS CLASSIFICATION

DISADVANTAGES OF REFUSE CALCINING

- HIGH COST
- POTENTIALLY COMPLEX TECHNOLOGY

REFUSE CALCINING

(COSTS IN DOLLARS PER TON OF CLEANED COAL)

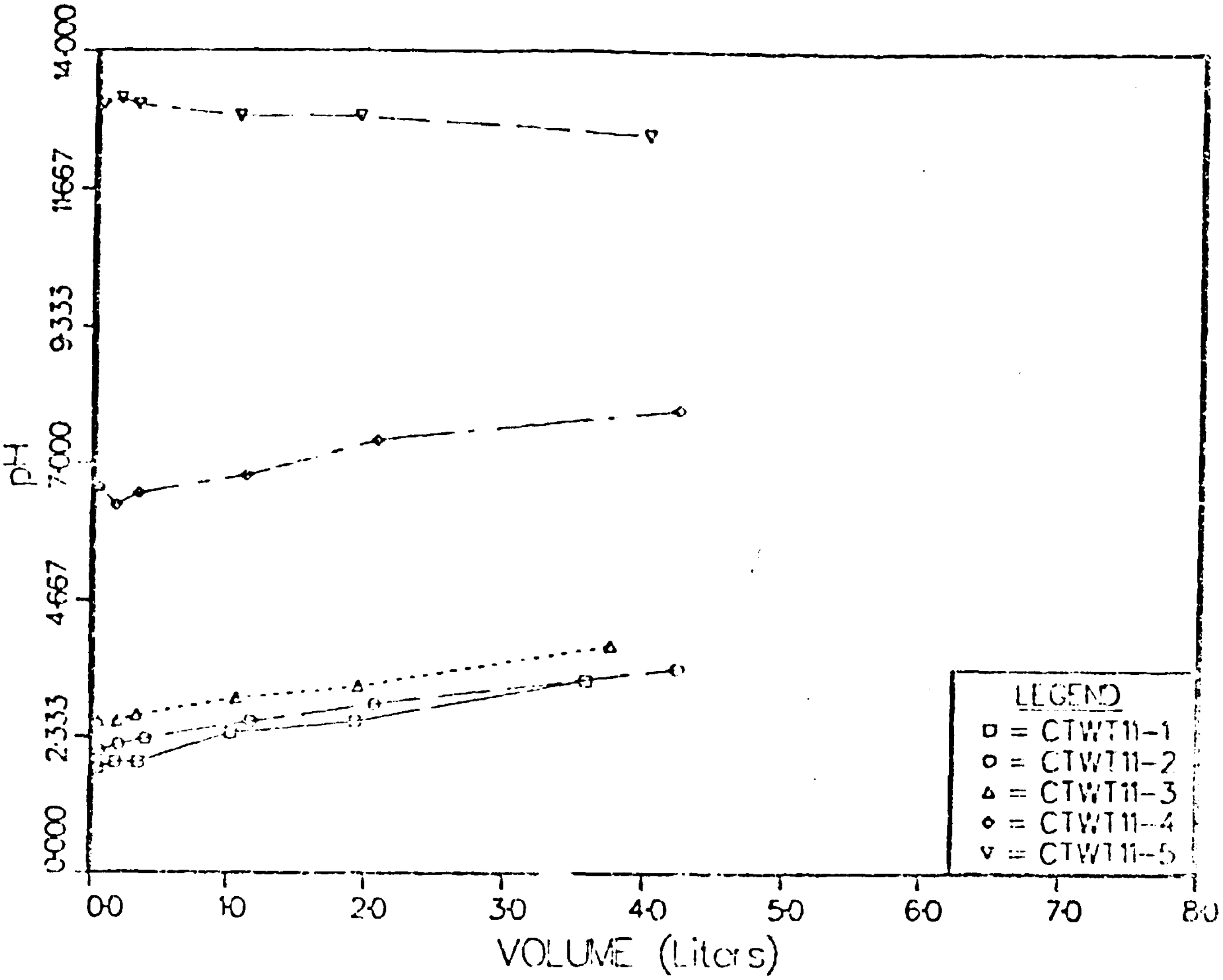
	PLANT A	PLANT B	PLANT C
LIME SCRUBBING	3.29	4.45	4.94
LIME/LIMESTONE SCRUBBING	2.14	2.80	3.08
30% SULFUR RETENTION	1.80		

PREVENT RELEASE OF TOXIC ELEMENTS

- Codispose with neutralizing agents
- Codispose with sorbents
- Apply refuse dump sealants
- Use refuse for something else

LIME CODISPOSAL

The Codisposal of Lime with Coal Refuse
to Control the Acidity of Refuse Leachates
and Prevent the Release of Toxic Contaminants



14.000
11.667
9.333
7.000
4.667
2.333
0.000
pH
0.0
1.0
2.0
3.0
4.0
5.0
6.0
7.0
8.0
VOLUME (Liters)
LEGEND
□ = CTWT11-1
○ = CTWT11-2
△ = CTWT11-3
◇ = CTWT11-4
▽ = CTWT11-5

LEACHABILITY OF REFUSE/LIME MIXTURES

	Control	Refuse+3% Lime	Refuse+10% Lime
pH	2	7	12
TDS(%)	4	0.4	0.5
Al	720	<0.6	<0.5
Fe	7800	40	<0.1
Mn	22	1	<0.02
Co	12	0.3	0.1
Ni	18	0.5	0.1
Zn	29	0.1	0.02

Volume/Refuse ~ 0.7
Values in ppm of leachate

ADVANTAGES OF LIME CODISPOSAL

- MODERATE COST
- HIGHLY EFFECTIVE
- SHORT TREATMENT DURATION
- RELATIVELY SIMPLE TECHNOLOGY

DISADVANTAGES OF LIME CODISPOSAL

- UNKNOWN PERMANENCY
- LOW BY-PRODUCTS RECOVERY POTENTIAL
- RCRA CLASSIFICATION IN QUESTION

LIME CODISPOSAL

COSTS IN DOLLARS PER TON OF CLEANED COAL

	PLANT A	PLANT B	PLANT C
25% OF THEORETICAL LIME	0.58	0.82	0.93

TREATMENT OF CONTAMINATED REFUSE DRAINAGE

- Alkaline Neutralization
- Ion Exchange
- Reverse Osmosis
- Chelating or Complexing Agents
- Freezing or Distilling
- Biological Treatment
- Precipitating Agents
- Selective Sorbents

ALKALINE NEUTRALIZATION

The Use of Alkaline Agents to Reduce the Acidity of Coal Refuse Leachates and Remove Toxic Elements by Precipitation

ALKALINE NEUTRALIZATION OF CONTAMINATED REFUSE DRAINAGE

	Control	Lye	Limestone	Lime
pH	1.1	6	7.1	6.6
TDS(%)	0.5	3.4	3.2	3.2
Al	18	<0.2	<0.2	<0.2
Fe	820	0.06	0.3	0.3
Mn	3.6	0.07	6.4	1.0
Co	2.0	0.05	1.0	0.6
Ni	3.2	0.05	1.0	0.7
Zn	3.9	0.02	0.1	0.1

Values in ppm of effluent

ALKALINE NEUTRALIZATION

(COSTS IN DOLLARS PER TON OF CLEANED COAL)

	PLANT A	PLANT B	PLANT C
LIME PRECIPITATION	0.09	0.83	0.06

MASTER GRID SUMMARY OF ENVIRONMENTAL CONTROL OPTIONS

	CALCINING	LIME CODISPOSAL	ALKALINE TREATMENT
COST	HIGH	MODERATE	LOW
EFFECTIVENESS	GOOD	GOOD	GOOD
PROCESS COMPLEXITY	HIGH	MODERATE	MODERATE
TREATMENT DURATION	SHORT	SHORT	VERY LONG
BY-PRODUCTS POTENTIAL	HIGH	NONE	NONE
PERMANENCY	GOOD	?	POOR
RCRA	NO HAZARD	?	HAZARDOUS