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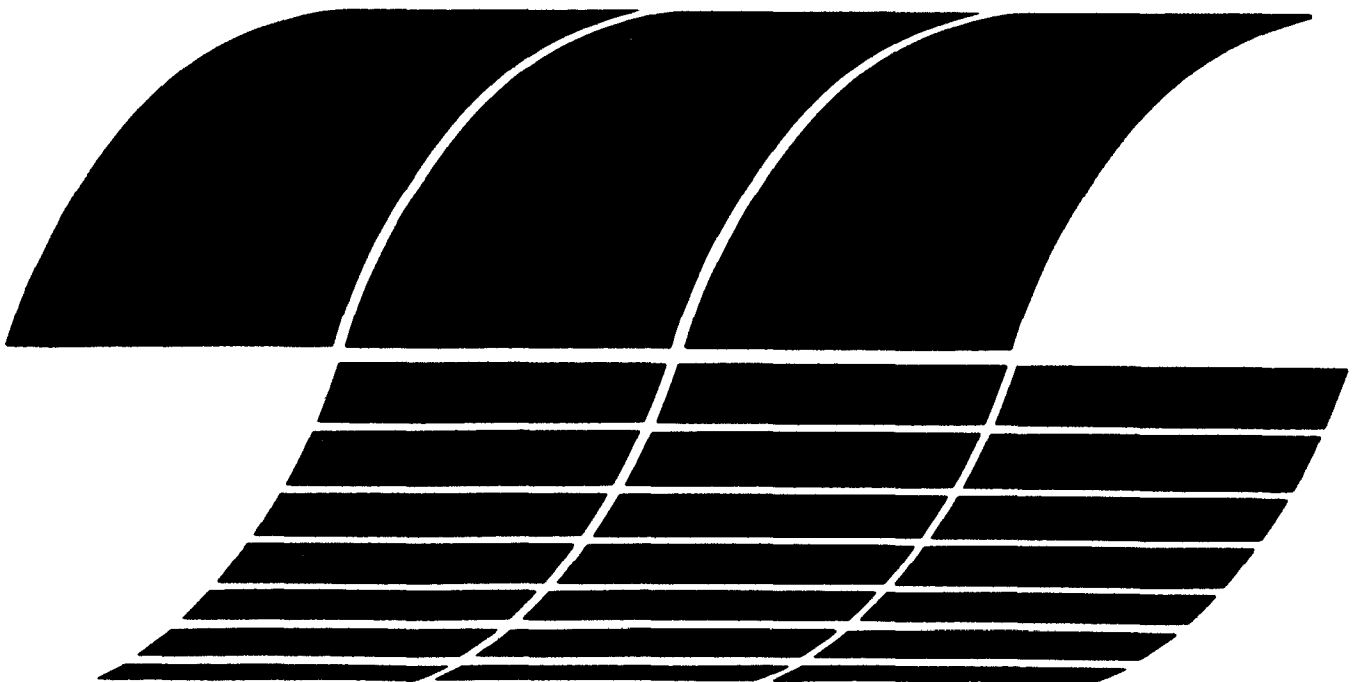
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Leaching Experiments on Coal Preparation Wastes: Comparisons of the EPA Extraction Procedure With Other Methods

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Leaching Experiments on Coal Preparation Wastes: Comparisons of the EPA Extraction Procedure With Other Methods

by

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LEACHING EXPERIMENTS ON COAL PREPARATION WASTES:
COMPARISONS OF THE EPA EXTRACTION PROCEDURE WITH OTHER METHODS

by

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ABSTRACT

Mineral wastes from seven coal preparation plants, located in the Illinois Basin, the Appalachian Region, and the West have been leached in accordance with the EPA extraction procedure published in the Federal Register dated May 19, 1980. This is one of the tests required for the classification of solid wastes under RCRA. When examined according to the procedures set forth in the Federal Register, all of the coal waste leachates had trace element concentrations below the maximum set by EPA. Results of the EPA leaching procedure compare favorably with those of our own leaching experiments for those elements which were analyzed (Ag, As, Ba, Cd, Cr, Hg, Pb, Se). However, we note that coal wastes release substantial quantities of other trace elements not included in the protocols at the present time (Fe, Al, Ni, Mn, Zn, Cu). In addition, the requirement that the test leachate be maintained at $\text{pH} \leq 5$ has the effect of establishing an abnormal environment for those wastes that are neutral or alkaline.

I. INTRODUCTION

The United States Congress, in the fall of 1976, enacted the Resource Conservation and Recovery Act (RCRA), designed to establish a comprehensive program for management of solid industrial and urban wastes. This act requires the Environmental Protection Agency (EPA) to promulgate a series of regulations which classify solid wastes as hazardous or non-hazardous and which set forth various protocols for disposing of these wastes. Among the criteria used to determine whether a solid waste is to be considered hazardous, by virtue of its ability to contaminate aqueous drainages, are the results of a standard

leaching procedure. This test is set forth in detail in the Federal Register (1), but in essence involves leaching the solid material with deionized water under rigidly defined conditions. In past work on coal preparation wastes, we at the Los Alamos Scientific Laboratory (LASL) have used similar procedures for scientific studies of the environmental weathering and leaching of these wastes. Because of this experience we are in a unique position to make comparisons between the RCRA leaching procedure and our own environmental simulation studies on coal cleaning wastes. This report summarizes our recent researches in this area.

In the following discussion we first present the results of RCRA leaching tests carried out on refuse from seven different coal cleaning plants. We then compare these results with those obtained using related procedures which we have developed during the course of our own work and, finally, we offer some comments regarding the RCRA procedures as they relate to coal wastes.

II. RESULTS OBTAINED USING THE EPA EXTRACTION PROCEDURE

Seven mineral wastes from coal preparation plants in the Illinois Basin, the Appalachian Region and the West were leached in accordance with the EPA extraction procedure published in the Federal Register dated May 19, 1980. (1) This, in essence, amounts to using 100g of waste, ground to pass through a 9.3 mm standard sieve ($-3/8"$), adding 1600 ml of deionized water to the waste and agitating for 24 hours in an extractor designed to insure that all sample surfaces are continuously brought into contact with well-mixed extraction fluid.

The pH values of the mixtures must be monitored during the course of the extraction and, in those cases in which the pH is greater than 5, adjustment must be made by addition of 0.5N acetic acid. At the end of the 24 hour extraction period, the solids are removed by filtration, and the concentrations of eight elements (Ag, As, Ba, Cd, Cr, Hg, Pb, Se) in the filtrate are determined. The results of these determinations with seven coal preparation wastes are presented in Tables I and II, while the analytical details may be found in the Experimental Section of this report.

Table I shows the initial and final pH values for each of the samples studied. The pH was well below 5 in all cases except for plant D, which is located in the western U. S. A comparatively small amount (35 ml) of 0.5N acetic acid was sufficient to maintain the required pH of 5 throughout the course of the extraction for this sample. This imposed acidic pH probably represents

TABLE I
INITIAL AND FINAL pH VALUES FOR COAL
WASTE LEACHATES USING THE EPA EXTRACTION PROCEDURE

Plant ^a	A	B	C	D	G	I	K
pH, initial	3.1	2.8	3.3	9.6	4.1	3.1	3.3
Acetic Acid added	-	-	-	35 ml	-	-	-
pH, final	4.2	2.2	3.2	5.0	3.8	2.6	2.7

^aPlants A, B, C: high-sulfur, Illinois Basin waste.

Plant D: low-sulfur, Western Waste.

Plant G: low-sulfur, Appalachian Waste.

Plants I, K: high-sulfur, Appalachian waste.

an abnormal circumstance for the western coal waste sample, but is certainly typical of many coal wastes from the eastern part of the country.

The results of the elemental analyses are presented in Table II. A cursory examination of this table reveals that many of the elements are present at

TABLE II
CONCENTRATIONS (ppm) OF TOXICITY INDICATOR
ELEMENTS IN COAL WASTE LEACHATES FROM SEVEN COAL PREPARATION
PLANTS

Plant	A	B	C	D	G	I	K	HDWS ^a
Ag	<0.006	<0.006	<0.006	<0.006	<0.006	<0.006	<0.006	5.0
As	0.024	0.100	0.007	<0.001	<0.001	0.016	0.096	5.0
Ba	<0.06	0.14	0.08	1.4	0.08	<0.06	<0.06	100
Cd	<0.003	<0.004	<0.003	<0.003	<0.003	<0.003	<0.003	1.0
Cr	<0.005	0.023	0.010	<0.005	<0.005	<0.017	<0.005	5.0
Hg	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	0.2
Pb	<0.012	<0.012	<0.012	<0.012	<0.012	<0.012	<0.012	5.0
Se	0.0015	0.0035	0.0011	0.0016	0.0020	0.0017	0.0038	1.0

^a100 x Primary Drinking Water Standard.

levels below the detection limits of the analytical methods used. In only three instances do any of the values exceed the primary drinking water standards. These cases are the arsenic values for plants B and K and the barium value for plant D. However, the limits specified for these elements in non-hazardous wastes are 100 times the primary drinking water standards (1) and all of the values in Table II are at least an order of magnitude below these.

Statistical analyses were carried out in order to determine the probabilities, based on the analytical data, that the true concentrations of the various elements equal or exceed either the primary drinking water standard or the "Hazardous Waste" limit defined in the Federal Register (1). These were done by calculating the so-called " β errors", using the one-sided t-test with a 95 percent confidence interval. The methodology for doing this has been published elsewhere (6). Some of the results of these calculations are shown in Table III.

TABLE III
PROBABILITIES THAT THE TRUE CONCENTRATIONS
OF TOXICITY INDICATOR ELEMENTS EQUAL OR EXCEED
THE FEDERAL PRIMARY DRINKING WATER STANDARDS

Plant	A	B	C	D	G	K	I
Ag	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
As	<0.01	>0.99	<0.01	<0.01	<0.01	<0.01	>0.99
Ba	<0.01	<0.01	<0.01	>0.99	<0.01	<0.01	<0.01
Cd	<0.5	<0.8	<0.5	<0.5	<0.5	<0.5	<0.5
Cr	<0.01	0.02	<0.01	<0.01	<0.01	<0.01	<0.01
Hg	<0.5	<0.5	<0.5	<0.5	<0.5	<0.7	<0.5
Pb	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4
Se	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01

These results show that the probabilities for exceeding the drinking water standards are significant only for Cd, Hg and Pb generally, and for As and Ba in specific cases. The probabilities for exceeding the "Hazardous Waste" limits, which are 100 times the drinking water standards, are less than 0.01 in all cases.

III. COMPARISONS AMONG DIFFERENT LEACHING PROCEDURES

Let us first consider static leaching experiments. These are experiments in which a fixed amount of liquid phase is maintained in contact with the solid sample throughout the duration of the extraction, as opposed to cases in which the liquid phase is allowed to flow through the solid. There are a number of independent variables in such static experiments. These are the geometric surface area of the solid (mesh size), the liquid to solids ratio, the duration of the extraction, the degree and type of agitation used, the composition of the liquid phase, the temperature, whether the reaction mixture is open to air, and what components are determined in the final leachate. All of our static leaching experiments have been carried out using deionized water as the liquid phase and fairly vigorous agitation (90 strokes/min, 3 inches/stroke). In addition, the vast majority have been done at room temperature with the extractor open to air. With the exception of the presence of air, these conditions are comparable to those called for in the EPA procedure when applied to acidic coal wastes. Consequently we are left free to examine the effects of the liquid to solid ratio, the mesh size and the extraction time. The other variables still deserve comment but that will be reserved for Part IV of this report.

The one-day static leach experiments which we have routinely carried out in the past are directly comparable to the EPA extraction procedure. The only difference is that we have used liquid to solid ratios of either 4 to 1 or 5 to 1, whereas the EPA test calls for a ratio of 16 to 1 during the extraction, and 20 to 1 in the final samples. The concentrations of the toxicity indicator elements in the leachates are presented in Table IVa for the EPA test and in Table Va for our own one-day static leach. (The values in Table IVa are closely related to those in Table II, but have been adjusted to represent the concentrations in the original leachate at a 16 to 1 liquid to solids ratio. This is necessary in order to eliminate the effects of the dilution of the leachate before the filtration and analysis.) If equilibrium were reached during the extraction, then one would expect the concentrations of the elements in the leachate to be independent of the liquid to solid ratio provided that the supply of the original elements in the sample was not exhausted. Comparison of Tables IVa and Va reveals that these extractions are not at equilibrium. Therefore, the concentrations of the elements in the leachates are at least partially kinetically controlled. Under these circumstances it becomes advantageous to

use a low liquid to solids ratio, because this leads to more concentrated leachates which are usually easier to analyze.

A more direct comparison can be made by converting the leachate concentrations to the total amounts of each element leached per unit of solid waste. These results are presented in Tables IVb and Vb. If the release of an element is strictly kinetically controlled, these data should be exactly comparable. The elements Cr and, to a lesser degree, Cd seem to compare fairly well between these two methods. However, much more As was leached using the EPA method, while our own procedure seemed to yield higher Pb values. The As results might be explained by noting that the analytical methods used for As were different, however the Pb results remain unexplained.

Table IVc and Vc show the leachate compositions expressed as the fraction of each element originally present in the solid which is dissolved in the leachate. These results exactly parallel those in Tables IVb and Vb. However it is interesting to note that Cd seems to be highly mobile, with large percentages being extracted, whereas other elements are extracted to much lesser degrees.

Results of extractions done on 20 mesh samples are shown in Tables VIa, VIb, and VIc. In general there is little change between these and the -3/8" samples described in Tables Va, Vb and Vc. Since reduction of the particle size from 3/8" to 20 mesh represents a substantial increase in the geometric surface area, one must conclude that the actual effective surface area is much larger than the geometric surface, or that the surface area does not affect the leaching behaviors of the various elements. The former conclusion seems more likely. Note that cadmium, which does not seem to fit this analysis, is highly mobile and may be much more sensitive to minor changes in accessible surface area than the other, less mobile elements.

Longer term static leaches are summarized in Tables VIIa, VIIb, and VIIc. In most cases, the amounts of leached elements can be seen to remain constant or increase with the duration of the extraction. While this agrees with our conception of how the leaching process works, it is useful to note that the differences between the one-day leaches and the multi-day leaches are not very large. This suggests that most of the action, at least for As, Cd, Cr and Pb, takes place in the early part of the experiment (the first 24 hours).

While our static leaching experiments were designed primarily to determine what might be leached from a coal waste, our dynamic (column) leaching experi-

TABLE IVa
ADJUSTED* LEACHATE COMPOSITIONS OBTAINED USING THE RCRA
LEACHING PROCEDURE FOR COAL WASTE SAMPLES (ppm)

Plant	A	B	C	D	G	I	K
Element							
Ag	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008
As	0.030	0.125	0.009	<0.001	<0.001	0.020	0.120
Ba	<0.075	0.175	0.100	0.075	0.100	<0.075	<0.075
Cd	<0.004	<0.005	<0.004	<0.004	<0.004	<0.004	<0.004
Cr	<0.006	0.029	0.012	<0.006	<0.006	<0.021	<0.006
Hg	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Pb	<0.015	<0.015	<0.015	<0.015	<0.015	<0.015	<0.015
Se	0.0019	0.0044	0.0014	0.0020	0.0025	0.0021	0.0048
pH	4.2	2.2	3.2	5.0	3.8	2.6	2.7

*Adjusted to reflect the original leachate composition at a 16 to 1 liquid to solids ratio, before dilution to the final 20 to 1 ratio.

TABLE IVb
LEACHATE COMPOSITIONS OBTAINED USING THE RCRA LEACHING
PROCEDURE FOR COAL WASTE SAMPLES. RESULTS EXPRESSED AS
mg ELEMENT LEACHED PER Kg SOLID WASTE

Plant	A	B	C	D	G	I	K
Element							
Ag	<0.120	<0.120	<0.120	<0.120	<0.120	<0.120	<0.120
As	0.480	2.00	0.140	<0.020	<0.020	0.320	1.92
Ba	<1.20	2.80	1.60	1.20	1.60	<1.20	<1.20
Cd	<0.060	<0.080	<0.060	<0.060	<0.060	<0.060	<0.060
Cr	<0.100	0.460	0.200	<0.100	<0.100	<0.340	<0.100
Hg	<0.020	<0.020	<0.020	<0.020	<0.020	<0.020	<0.020
Pb	<0.240	<0.240	<0.240	<0.240	<0.240	<0.240	<0.240
Se	0.030	0.070	0.022	0.032	0.040	0.034	0.075
pH	4.2	2.2	3.2	5.0	3.8	2.6	2.7

TABLE IVc
LEACHATE COMPOSITIONS OBTAINED USING THE RCRA LEACHING PROCEDURE
FOR COAL WASTE SAMPLES. RESULTS EXPRESSED AS THE PERCENT OF THE ELEMENT
ORIGINALLY PRESENT THAT APPEARS IN THE LEACHATE

Plant	A	B	C	D	G	I	K
Element							
Ag	-	-	-	<32	-		
As	0.86	2.1	0.64	-	<0.11		
Ba	-	-	-	-	-		
Cd	<25	<20	<5.4	<9.7	<18		
Cr	<0.17	0.74	0.29	<0.07	<0.11		
Hg	-	-	-	-	-		
Pb	<0.49	<0.71	<0.48	<0.86	<1.0		
Se	0.32	1.1	0.26	-	-		
pH	4.2	2.2	3.2	5.0	3.8	2.6	2.7

TABLE Va
LEACHATE COMPOSITIONS OBTAINED FROM ONE-DAY SHAKER LEACHES
OF COAL WASTE SAMPLES (-3/8"). RESULTS EXPRESSED AS ppm

Plant	A	B	C	D	G	I	K
Ag	-	-	-			-	-
As	0.008	-	0.004			-	.054
Ba	-	-	-			-	-
Cd	0.0014	0.024	0.020			.015	.010
Cr	0.001	0.060	0.032			.094	-
Hg	-	-	-			-	-
Pb	0.048	0.300	0.32			-	0.15
Se	-	-	-			-	0.002
pH	7.1	2.2	3.5			2.6	3.0

TABLE Vb
LEACHATE COMPOSITIONS OBTAINED FROM ONE-DAY LEACHES
OF COAL WASTE SAMPLES (-3/8"). RESULTS EXPRESSED AS mg
ELEMENTS LEACHED PER Kg SOLID WASTE

Plant Element	A	B	C	D	G	I	K
Ag	-	-	-			-	-
As	0.04	-	0.02			-	0.270
Ba	-	-	-			-	-
Cd	0.0068	0.12	0.10			0.075	0.050
Cr	0.005	0.30	0.16			0.470	-
Hg	-	-	-			-	-
Pb	0.240	1.5	1.6			-	0.750
Se	-	-	-			-	0.010
pH	7.1	2.2	3.5			2.6	3.0

TABLE Vc
LEACHATE COMPOSITIONS OBTAINED FROM ONE-DAY SHAKER
LEACHES OF COAL WASTE SAMPLES (-3/8"). RESULTS EXPRESSED
AS THE PERCENT OF THE ELEMENT ORIGINALLY PRESENT
THAT APPEARS IN THE LEACHATE

Plant	A	B	C	D	G	I	K
Ag	-	-	-				
As	0.07	-	0.09				
Ba	-	-	-				
Cd	2.8	30	8.9				
Cr	0.008	0.48	0.23				
Hg	-	-	-				
Pb	0.49	4.4	3.2				
Se	-	-	-				
pH	7.1	2.2	3.5				

TABLE VIa
LEACHATE COMPOSITIONS OBTAINED FROM ONE-DAY SHAKER LEACHES
OF COAL WASTE SAMPLES (~20 MESH). RESULTS EXPRESSED AS ppm

Plant	A	B	C	D	G	I	K
Ag	<0.008	-	<0.008		-	-	
As	<0.008	-	<0.004		-	-	
Ba	-	-	-		-	-	
Cd	2.9	48	8.9		9.3		
Cr	<0.001	0.156	0.032		.0018	0.080	
Hg	<0.20	-	<0.20		-	-	
Pb	0.048	0.320	0.320		-	-	
Se	-	-	-		-	-	
pH	7.1	2.2	3.5		4.3	3.2	

TABLE VIb
LEACHATE COMPOSITIONS OBTAINED FROM ONE-DAY SHAKER LEACHES
OF COAL WASTE SAMPLES (~20 MESH). RESULTS EXPRESSED AS mg
ELEMENT LEACHED PER Kg SOLID WASTE

Plant	A	B	C	D	G	I	K
Ag	<0.040	-	<0.040		-	-	
As	<0.040	-	<0.020		-	-	
Ba	-	-	-		-	-	
Cd	0.0070	0.190	0.100		0.031	0.10	
Cr	<0.005	0.780	0.160		0.007	0.40	
Hg	<1.00	-	<1.00		-	-	
Pb	0.24	1.60	1.60		-	-	
Se	-	-	-		-	-	
pH	7.1	2.2	3.5		4.3	3.2	

TABLE VIc
LEACHATE COMPOSITIONS OBTAINED FROM ONE-DAY SHAKER LEACHES
OF COAL WASTE SAMPLES (~20 MESH). RESULTS EXPRESSED AS THE PERCENT OF THE ELEMENT
ORIGINALLY PRESENT THAT APPEARS IN THE LEACHATE

Plant	A	B	C	D	G	I	K
Ag	-	-	-		-		
As	<0.07	-	<0.09		-		
Ba	-	-	-		-		
Cd	2.9	48	8.9		9.3		
Cr	<0.008	1.2	0.23		0.008		
Hg	-	-	-		-		
Pb	0.49	4.7	3.2		-		
Se	-	-	-		-		
pH	7.1	2.2	3.5		4.3		

TABLE VIIa
LEACHATE COMPOSITIONS OBTAINED FROM LONG TERM SHAKER LEACHES
OF COAL WASTE SAMPLES (-3/8"). RESULTS EXPRESSED IN ppm

Plant	A	B	C	D	G	I	K
Days	28	22	28			25	25
Ag	-	-				-	-
As	0.008	-	0.175			-	3.0
Bg	-	-	-			-	-
Cd	0.0003	0.035	0.124			0.01	0.041
Cr	0.0012	0.116	0.240			0.10	-
Hg	-	-	-			-	-
Pb	0.006	0.280	0.360			-	0.004
Se	-	-	-			-	0.036
pH	7.6	1.9	1.9			2.2	2.0

TABLE VIIb
LEACHATE COMPOSITIONS OBTAINED FROM LONG TERM SHAKER LEACHES
OF COAL WASTE SAMPLES (-3/8"). RESULTS EXPRESSED AS mg
ELEMENT LEACHED PER Kg SOLID WASTE

Plant	A	B	C	D	G	I	K
Days	28	28	28			25	25
Ag	-	-	-			-	-
As	0.04	-	0.88			-	15.0
Ba	-	-	-			-	-
Cd	0.0014	0.18	0.62			0.05	0.205
Cr	0.006	0.58	1.20			0.50	-
Hg	-	-	-			-	-
Pb	0.030	1.40	1.80			-	0.020
Se	-	-	-			-	0.180
pH	7.6	1.9	1.9			2.2	2.0

TABLE VIIc
LEACHATE COMPOSITIONS OBTAINED FROM LONG TERM SHAKER LEACHES
OF COAL WASTE SAMPLES (-3/8"). RESULTS EXPRESSED AS THE PERCENT OF THE ELEMENT
ORIGINALLY PRESENT THAT APPEARS IN THE LEACHATE

Plant	A	B	C	D	G	I	K
Days	28	28	28			25	25
Ag	-	-	-				
As	0.071	-	4.0				
Bg	-	-	-				
Cd	0.583	45.0	55.4				
Cr	0.010	0.935	1.74				
Hg	-	-	-				
Pb	0.061	4.12	3.60				
Se	-	-	-				
pH	7.6	1.9	1.9			2.2	2.0

TABLE VIIIa
LEACHABILITIES OF SELECTED ELEMENTS FROM COAL WASTE SAMPLES
(-3/8") OBTAINED FROM CONTINUOUS COLUMN LEACHING EXPERIMENTS.
RESULTS EXPRESSED AS ppm FOR 16 ℓ WATER PER Kg SOLID

Plant	A	B	C	G
Ag	-	-	-	-
As	0.016	0.34	0.50	-
Ba	-	-	-	-
Cd	0.0048	0.016	0.0072	0.0026
Cr	0.031	0.021	0.050	0.0080
Hg	-	-	-	-
Pb	0.014	0.022	0.0075	-
Se	-	-	-	-
pH	2.9-7.7	1.7-3.4	2.4-3.8	2.9-40

TABLE VIIIb
LEACHABILITIES OF SELECTED ELEMENTS FROM COAL WASTE SAMPLES
(-3/8") OBTAINED FROM CONTINUOUS COLUMN LEACHING EXPERIMENTS.
RESULTS EXPRESSED AS mg ELEMENT LEACHED PER Kg SOLID WASTE

Plant	A	B	C	G
Ag	-	-	-	-
As	0.26	5.3	0.80	-
Ba	-	-	-	-
Cd	0.077	0.26	0.12	0.042
Cr	0.49	0.39	0.80	0.13
Hg	-	-	-	-
Pb	0.22	0.35	0.12	-
Se	-	-	-	-
pH	2.9-7.7	1.7-3.4	2.4-3.8	2.9-4.0

TABLE VIIIc
LEACHABILITIES OF SELECTED ELEMENTS FROM COAL WASTE SAMPLES
(-3/8") OBTAINED FROM CONTINUOUS COLUMN LEACHING EXPERIMENTS.
RESULTS EXPRESSED AS THE PERCENT OF THE ELEMENT
ORIGINALLY PRESENT THAT APPEARS IN THE LEACHATE

Plant	A	B	C	G
Ag	-	-	-	-
As	0.46	5.7	3.6	-
Ba	-	-	-	-
Cd	32	66	10	13
Cr	0.82	6.4	1.2	0.14
Hg	-	-	-	-
Pb	0.44	1.0	0.24	-
Se	-	-	-	-
pH	2.9-7.7	1.7-3.4	2.4-3.8	2.9-4.0

ments have been attempts to simulate the weathering of an exposed waste pile. We were primarily interested in studying the leaching of elements as a function of time and the effects of intermittent leaching. Thus these experiments are more difficult to compare to the EPA procedure than the static leaches. However, a comparison can be made by integrating the concentration versus volume curve in each element out to a volume representing a 16 to 1 liquid to solids ratio. This gives the total amount of a given element extracted in that volume. These results were then used to calculate the amounts extracted per unit solids shown in Table VIIIb. The values shown in Tables VIIla and VIIlc were then derived from those in Table VIIIb. One should note that most of the extraction takes place early in the experiment, so that the choice of the upper volume limit to the integration does not drastically affect the results. In general the column leaching experiments show higher extraction efficiencies than the static experiments. This is especially so for As and to lesser degrees for Cd and Cr. Pb shows the reverse trend. This might be due to reprecipitation caused by the increase in pH with time, but this is speculation.

In summary, the EPA leaching procedure compares well with those procedures which we have been using in our work on coal preparation wastes for the last several years, at least to the extent to which these various methods can be compared. The major difference between our procedures and the EPA procedure, in the case of acidic coal preparation wastes, is the higher liquid to solids ratio used in the EPA method. This high ratio has the effect of diluting the leachate and rendering the chemical analyses more difficult. In the case of non-acidic coal wastes, there is the additional difference that acetic acid is added to the extraction mixture in the EPA method. For coal wastes which are not naturally acidic this creates an artificial environment and complicates the scientific interpretation of the results.

IV. THE EPA LEACHING PROCEDURE AS APPLIED TO COAL WASTES

One must remember that the EPA leach test is designed to satisfy a regulatory need to classify solid wastes as hazardous or not. As such it must apply to a wide variety of wastes, including municipal, chemical and industrial by-products, whose properties and chemical behaviors may differ substantially. It seems unlikely that any single test can be entirely appropriate in all these different types of waste, and thus it is important for one to understand what the limitations of the test are for various types of waste. In the following

discussion we shall record our observations on the applicability of the EPA leaching test to coal preparation wastes. The most important question is whether the leaching test is an accurate indicator of the potential of a given waste to harm the environment and our considerations have been carried out with this in mind.

Past studies have revealed that the elements with the highest discharge severities in leachates from coal preparation wastes are Fe, Al, Ni, Mn, Zn, Cu and Cd as well as the acidity (2,3,4). The elements addressed in the EPA leaching test are those included in the Federal Primary Drinking Water Standards, namely Ag, As, Ba, Cd, Cr, Hg, Pb and Se. The only element common to these two groups is Cd. Iron has by far the highest discharge severity, based on the MEG/MATE system (5), followed roughly in order by the other elements listed. Some of the elements included in the EPA procedure, notably Ag, Hg and Ba, are typically present at levels below the detection limits of the methods used for the analysis of the leachates. Furthermore, the parent coal waste materials often contain these elements in such minimal quantities that we have only rarely attempted to determine them in our research on coal waste leaching behavior. Consequently, in the case of typical coal preparation wastes, we conclude that the EPA leaching test in its present form does not address the elements of real concern. If the elements in the secondary drinking water standards were included in the EPA leaching test, then the situation would be markedly improved, since Fe, Mn, Zn and Cu would be covered. This would leave only Al and Ni as elements of potential concern not considered in the leaching test.

When acidic coal wastes are considered, the leachates are sufficiently acid so that no acetic acid additions are called for. Under these circumstances the EPA test is essentially a water leach and reasonably simulates the acid-base conditions one might expect in a more or less stagnant coal waste pile. Since acidic coal wastes are the most abundant type and since they represent the wastes of most concern in the eastern coal fields of the United States, this type of test seems entirely appropriate. However, alkaline coal wastes, typically from the western United States, are treated differently under the EPA test. When acidified to a pH of five with acetic acid, these wastes are subjected to an artificial environment which they are not likely to encounter under normal circumstances. We believe that this test becomes unnecessarily severe for those elements which are mobilized under acidic conditions while ignoring

the possible effects on elements which may be alkaline mobile such as selenium and arsenic.

At first glance, one would expect the results of a leaching experiment to depend on the size of the particles in the solid sample. This is because small particles have a higher geometric surface area per unit mass than large ones. However, our experience with coal wastes has been that the particle size does not strongly affect the results of our leaching experiments. Since the mesh size seems unimportant, one might as well choose one that is convenient. The 9.3 mm (-3/8") size is probably the most convenient for this type of waste.

Agitation of the sample during the leaching procedure is most important. The EPA test procedure calls for vigorous agitation. In our opinion such vigorous agitation is preferred over stagnant leaching because a vigorous agitation is easier to define and to reproduce from one experiment to the next and among different laboratories.

The matter of time is necessarily a compromise. The time needs to be long enough so that whatever chemical reactions are involved can proceed to a reasonable degree and yet short enough to complete the experiment in a timely fashion. With high sulfur coal wastes that do not have any self-neutralizing capacity, the 24 hour extraction time seems reasonable. However, some materials may not become severely acidic for several days or even weeks. Such a delay can be caused by the presence of carbonate minerals acting as in situ neutralizing agents, which must be used up before the pH can become very acidic. Whatever the cause, such a delay in the acid-releasing character of a coal waste could result in a rather toxic material being erroneously classified as non-hazardous. The only straightforward way to avoid this problem is to run leaching experiments for longer periods of time.

One factor which is important in the case of coal wastes, but which may not matter for other types of solid wastes, is the presence of air during the leaching process. The leachates from coal wastes are acidic because the oxidation of pyrite yields sulfuric acid as a by-product. If access of air to the solid is restricted, then less oxidation occurs and the leachates are less acidic. In the case of a 24 hour leaching experiment, most of the acid involved was generated before the actual leach was begun, so that access to air may not be important. However, in longer leaching experiments, the generation of acid during the experiment may be significant and restriction of the air intake may lead to artificially low results.

With reference to coal waste samples, liquid to solid ratios of 20 to 1 for the final analysis tax the detection limits of the analytical procedures. Use of a lower liquid to solids ratio, for example 4 or 5 to 1, would allow greater confidence in the analytical results and their implications concerning pollution potentials

Finally, we would like to offer one comment on the mechanical aspects of the extraction procedure. In order to facilitate the rapid separation of the leachate from the solid residue, thus eliminating long contact times of leachate and residue following the 24-hour agitation period, we have found it advantageous to use a pre-filtering step with a hard, ashless filter paper (Whatman #541) and a Buchner porcelain funnel prior to final filtration through a Millipore 0.45 μ filter. Even a glass fiber pre-filter, as mentioned in the extraction procedure, offers little relief from prolonged separations of materials containing clays, and the pre-filtering with the paper is much more rapid than the centrifuge method described in the EPA test procedure.

V. SUMMARY AND CONCLUSIONS

Mineral wastes from seven coal preparation plants, located in various parts of the country, have been leached in accordance with the EPA extraction procedure published in the Federal Register dated May 19, 1980 (1). When judged according to the toxicity criteria set forth in this procedure, all of the coal wastes are non-hazardous. The probabilities that any of the eight elements examined might actually exceed the levels set forth in the procedure are all less than one percent. The probabilities of the elements exceeding the federal primary drinking water standards are significant only for Cd, Hg and Pb.

When compared to leaching tests which we have used over the past several years in our research on coal wastes, the EPA test gives comparable results for those elements which are examined. The primary differences between our procedures and that prescribed in the Federal Register is the use of a higher liquid to solids ratio in the EPA test and the requirement that alkaline systems be acidified with acetic acid.

With respect to coal preparation wastes we can make the following comments concerning the EPA extraction procedure. First, the elements Fe, Al, Ni and Mn, which have the highest discharge severities in coal waste leachates, are not addressed by the method. Second we believe that the acidification of non-acidic materials is inappropriate in the case of coal wastes. Third, the time

of filtration can be significantly shortened by introducing a pre-filtering step before filtration through the Millipore filter. In addition to these concerns there remains the question of whether longer extraction times should be considered and whether the extraction vessel should be left open to the air.

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EXPERIMENTAL

A. RCRA Leaching Experiments

The procedure published in the Federal Register of May 19, 1980 (1) was carried out with the following modifications. First, we used pre-dried samples, since no fresh (wet) material was available. Second we modified the filtration procedure, as described below. Finally we allowed the pH of the Plant D sample to become a little lower than the specified value when making the initial addition of acetic acid.

Short-term pre-tests were made on all samples by adding 400 ml. deionized water to 25g solid, thereby determining initial pH values. For those samples with a pre-test pH value less than 5 and a history, according to prior LASL leaching procedures, of producing highly acidic leachates, no recording pH meter was used during the 24-hour test period, thus allowing simultaneous leaching of several samples. Refuse from Plants B, C, G, K and I met those criteria. Accordingly, 100g of -3/8" (9 mm) material was put into a 1/2-gallon (2-liter) polyethylene bottle, 1600 ml deionized (Milli-Q) water was added, the bottle capped, and the sample was swirled by hand to assure thorough wetting of the solid. The initial pH was then recorded. The bottle was placed on a platform shaker on its side, and agitation was begun at 90 3-in strokes per minute. Since prior analysis had shown Plant A waste to have some self-neutralizing capacity in the form of calcite, and Plant D waste had an initial pre-test pH value over 5, those wastes were leached separately, and the test was monitored with a recording pH meter. No automatic titrator was used. The pH electrode was fitted through a rubber stopper which was covered with plastic wrap to prevent contamination from the rubber. Thus the system was essentially sealed, as were the samples not monitored with the recording pH meter. The pH of the Plant A refuse leachate remained below 5 for the test period, and no addition of acid was necessary. For Plant D waste, the pH was adjusted manually with 0.5N acetic acid. An initial 10-ml increment of acid lowered the pH from 9.6 to 4.1. Four more additions of acid were made at 2.5 hr, 3.75 hr, 18.5 hr, and 20 hr after agitation was begun to maintain the pH below 5.2. A peak value of 5.75 was reached overnight. A total of 35 ml acetic acid was added.

After the samples were removed from the shaker, final pH values were recorded for those samples not monitored continuously. Vacuum filtration was begun according to the Federal Register procedure on Plant B and C samples and the remaining unfiltered samples were refrigerated. After 4 hours filtering was only partially complete. The vacuum was shut off overnight. After 19 hours (5 hours with vacuum turned on) filtering was still incomplete, though several changes of pre-filters as well as final 0.45 μ filters had been made. At that time a pre-filtering step using a Buchner funnel with Whatman #541 paper was added. Remaining samples were filtered without incident, using the Buchner pre-filter step. 400 ml water was added to bottles as a rinse, and that water added to the filter, except for Plant D, in which 365 ml was added, making the final volume of liquid 2000 ml in all cases. Aliquots of each

sample were poured into polyethylene bottles for analysis and all samples were stored in the refrigerator prior to analysis. (No acid was added for preservation).

Table IX summarizes the methods used to analyze the leachates. The resultant analytical data are shown in Table X. In Table X, \bar{X} is the mean of n independent measurements for the sample. The letter "t" represents the student's t for $\alpha = 0.05$ based upon pooled standard deviations for the sample set. The calculations of the β errors are described in Part II of this report. The β error (DWS) represents the probability, based on the analytical data, that the true concentration of the element equals or exceeds the Interim Drinking Water Standard. The β error (RCRA) represents the same probability relative to 100 times the Interim Drinking Water Standards.

B. LASL Leaching Experiments

1. Static (Shaker) Leaches. Representative samples were obtained by splitting from barrels of pre-dried refuse. All samples leached were no greater than 3/8 in. (9 mm) in particle size. In some cases, the samples were pulverized by alumina shell plates to -20 mesh. Previously split samples were tumbled to mix; portions were weighed into flasks and deionized water was added. The sample size was 50g. The amount of water added was 200 ml for Plants A, B, C, and G (4:1 liquid:solid) and 250 ml for Plants K and I (5:1 liquid:solid). The container used was a 500-ml Erlenmeyer flask with a ground glass neck, fitted with a glass chimney to allow air access without allowing liquid to splash out during agitation (Fig. 1). The refuse/water mixtures were placed on a platform shaker and agitated at 90 3-in strikes per minute. All leaching referred to in this report was done at room temperature, generally around 22° C. After various leaching times, samples were removed from the shaker and filtered by vacuum filtration, using Whatman #541 paper for the first step, followed by either gravity filtration through a fine filter paper (Whatman #42), as with Plant A, B, C, and G samples or through a Millipore 0.45 μ filter (vacuum filtration), as with samples from Plants K and I. Leachates then were diluted by addition of 10% 6N HNO₃ for preservation of sample prior to analysis.

2. Dynamic (Column) Leaches. Coal or refuse material (0.5 kg), crushed to -3/8 in., was packed into a Pyrex column 70 cm long by 4.6 cm diam. in a

TABLE IX
ANALYTICAL METHODS USED FOR THE RCRA LEACHING EXPERIMENTS

<u>ELEMENT</u>	<u>METHOD</u>	1979 Methods Manual <u>EPA EQUIV. METHOD</u>
Arsenic	AA, Hydride (1)	206.3
Barium	AA, N ₂ O flame (2)	208.1
Cadmium	AA, Flame	213.1
Chromium	AA, Flame (3)	218.1
Lead	AA, Flame	239.1
Mercury	AA, Cold vapor (4)	245.1
Selenium	AA, Hydride (1)	270.3
Silver	AA, Flame	272.1

(1) Borohydride Reduction.

(2) 1000 ppm Na instead of K.

(3) Air/C₂H₂ Flame.

(4) Persulfate oxidation not used.

vertical position. The leaching column was equipped with a necked-down inlet at the bottom for introducing the leachates. A side arm located 5 cm below the open top served as an effluent outlet. Both the upper and lower ends of the coal or refuse bed were retained in the column with loosely packed glass-wool plugs. An upward or countercurrent leachate flow was used in most of the experiments to prevent flow blockage from fine sediments that might settle to the bottom of the column.

The leachate, usually deionized water, was fed through the packed column in one of two ways. Early experiments (Plants A, B, C) employ a gravity feed from a reservoir elevated above the column inlet. The flow was regulated by a valve located between the reservoir and the column inlet. Later experiments used a peristaltic pump to feed the effluent through the column. Flow rates used were typically between 0.5 and 1.0 ml/min. Measurements of leachate flow and pH were made at the column outlet. Periodically, samples of leachate were collected for analysis of total solids and trace element composition.

3. Analytical Methods. Cd, Pb and Cr were determined in the acidified leachates by atomic absorption spectrophotometry. In the case of Cr, an air acetylene flame was used. As was determined by neutron activation analysis.

TABLE X
ANALYTICAL RESULTS OF EPA EXTRACTION PROCEDURES
FOR SEVEN COAL WASTE LEACHATES

ELEMENT	ARSENIC ppm				BARIUM ppm				SELENIUM ppm				SILVER ppm			
			β error				β error				β error				β error	
			DWS	RCRA			DWS	RCRA			DWS	RCRA			DWS	RCRA
Sample	$\bar{X} \pm ts/\sqrt{n}$	n			$\bar{X} \pm ts/\sqrt{n}$	n			$\bar{X} \pm ts/\sqrt{n}$	n			$\bar{X} \pm ts/\sqrt{n}$	n		
H ₂ O, Control	<.001	3	<.01	<.01	<.06	4	<.01	<.01	.0014 \pm .0006	3	<.01	<.01	<.006	3	<.01	<.01
Plant A	.024 \pm .001	3	<.01	<.01	<.06	4	<.01	<.01	.0015 \pm .0006	3	<.01	<.01	<.006	3	<.01	<.01
Plant B	.100 \pm .004	3	>.99	<.01	.14 \pm .06	4	<.01	<.01	.0035 \pm .0007	3	<.01	<.01	<.006	3	<.01	<.01
Plant C	.007 \pm .001	3	<.01	<.01	.08 \pm .06	4	<.01	<.01	.0011 \pm .0006	3	<.01	<.01	<.006	3	<.01	<.01
Plant D	<.001	3	<.01	<.01		8	<.99	<.01	.0016 \pm .0006	3	<.01	<.01	<.006	3	<.01	<.01
Plant G	<.001	3	<.01	<.01	.08 \pm .06	4	<.01	<.01	.0020 \pm .0006	3	<.01	<.01	<.006	3	<.01	<.01
Plant I	.016 \pm .001	3	<.01	<.01	<.06	4	<.01	<.01	.0017 \pm .0006	3	<.01	<.01	<.006	3	<.01	<.01
									.0038 \pm .0007	3	<.01	<.01	<.006	3	<.01	<.01
HClAc, Control	<.001	3	<.01	<.01	.08 \pm .06	4	<.01	<.01	.0009 \pm .0006	3	<.01	<.01	<.006	3	<.01	<.01

ELEMENT	CADMIUM ppm				CHROMIUM ppm				LEAD ppm				MERCURY ppm			
			β error				β error				β error				β error	
			DWS	RCRA			DWS	RCRA			DWS	RCRA			DWS	RCRA
Sample	$\bar{X} \pm ts/\sqrt{n}$	n			$\bar{X} \pm ts/\sqrt{n}$	n			$\bar{X} \pm ts/\sqrt{n}$	n			$\bar{X} \pm ts/\sqrt{n}$	n		
H ₂ O, Control	<.003	3	<.50	<.01	<.005	3	<.01	<.01	<.012	5	<.40	<.01	<.001	4	<.4	<.01
Plant A	<.003	3	<.50	<.01	<.005	4	<.01	<.01	<.012	5	<.40	<.01	<.001	3	<.5	<.01
Plant B	<.004	3	<.80	<.01	.023 \pm .006	3	.02	<.01	<.012	5	<.40	<.01	<.001	3	<.5	<.01
Plant C	<.003	3	<.50	<.01	.010 \pm .005	3	<.01	<.01	<.012	5	<.40	<.01	<.001	3	<.5	<.01
Plant D	<.003	3	<.50	<.01	<.005	5	<.01	<.01	<.012	5	<.40	<.01	<.001	3	<.5	<.01
Plant G	<.003	3	<.50	<.01	<.005	5	<.01	<.01	<.012	5	<.40	<.01	<.001	3	<.5	<.01
Plant I	<.003	3	<.50	<.01	.017 \pm .006	3	<.01	<.01	<.012	5	<.40	<.01	<.001	2	<.7	<.01
Plant K	<.003	3	<.50	<.01	<.005	3	<.01	<.01	<.012	5	<.40	<.01	<.001	3	<.5	<.01
HClAc, Control	<.003	3	<.50	<.01	<.005	4	<.01	<.01	<.012	5	<.40	<.01	<.001	2	<.7	<.01

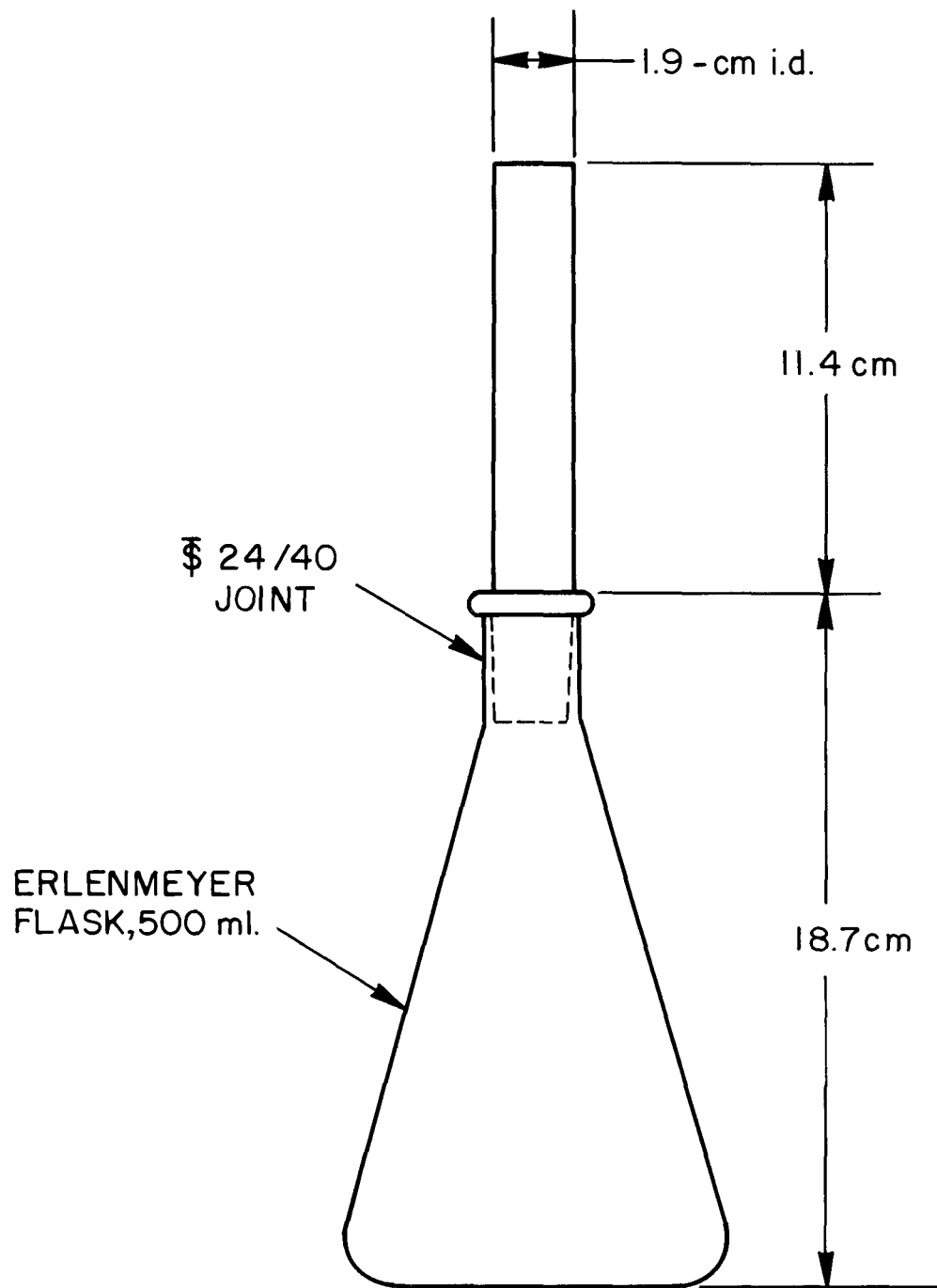


Fig. 1. Extraction Vessel Used for
LASL Shaker Leaching Experiments

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