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RADIOACTIVE WASTE DISPOSAL REPORT ON SEEPAGE PIT LIQUID WASTE - SHALE COLUMN EXPERIMENT

W. J. Lacy

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HEALTH PHYSICS DIVISION

K.Z. Morgan, Director

RADIOACTIVE WASTE DISPOSAL REPORT ON SEEPAGE PIT LIQUID WASTE - SHALE COLUMN EXPERIMENT

William J. Lacy+

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Abstract

The reports covers an investigation of the radioactive liquid waste adsorption capacity of Conasauga shale, the formation in which the ORNL liquid wastes pits have been dug. Included in the study are three other items of general interest (1) the method of predicting when breakthrough of large quantities of radionuclides will occur, (2) the order of occurance for the radionuclides present and (3) if the disposal pits located in the shale are abandoned, will the leaching action of rain and ground water remove dangerous quantities of radioactivity from the saturated shale?

The liquid waste material used was taken from the seepage pits. The study was made on the light colored weathered portion of the Conasauga shale.

INTRODUCTION

Seepage pits have been used as a method for disposing of large volumes of intermediate level radioactive liquid waste at ORNL for over five years.

One question that arises concerns the capacity of the Conasauga shale, the formation in which the pits are excavated for adsorbing the liquid waste released to these pits. Included in this general problem are other questions of interest: (1) is there any way of telling when breakthrough of large quantities of radionuclides will occur; (2) what will be the breakthrough sequence for the radionuclides present in the waste; and (3) when the pits are abandoned, will the leaching action of rain and ground water remove dangerous quantities of activity from the saturated shale?

Various investigators have considered the use of clays and shales to remove radionuclides from waste solutions and contaminated water (Brockett, Goldman, McHenry, Straub, et.al., Amphlett, Hatch, and others) (1,2,3,4,5,6,7,8). Brockett determined the effectiveness of Conasauga shale (the light colored weathered portions) and other shale types for removing radioactive evaporator waste concentrate, called W-6, and various single radionuclides from aqueous solutions employing both jar tests and column tests. His data indicated that the Conasauga shale has a high capacity for removing radioactive materials and compares favorably with the other types of clays which were studied.

However, the three questions mentioned above remained unanswered. Therefore, the following laboratory investigation was undertaken.

Procedure:

An air-dryed sample of the light-color, weathered Conasauga shale, collected from the area of pit No. 4, was graded by standard U. S. sieve sizes and 24 grams of 60-70 mesh placed in an 8 mm.ID. glass column and washed with distilled water to remove air bubbles. The top of the column was connected to a gravity feed system and a five liter reservoir containing the waste solution. This waste solution was a sample of the overflow from waste pit 3 to pit 2 on October 19, 1956. Chemical and a radiochemical analyses of this material were made and are shown in Tables 1 and 2.

The waste solution was allowed to flow downward through the shale, pass a two-way stop-cock, used to feed 1M HCl intermittently through the tygon tubing to decrease the background due to adsorption or plating out of the radionuclides on the tubing wall, and then through a maze (1ml vol) placed under a mica endwindow G-M tube for continuous monitoring of the column effluent, see Figure 1,3.

Composite samples (25ml) were taken from every 100 ml that passed through the column. These composite samples were counted for beta activity with a mica end-window G-M tube and for gamma activity with a 1000 mg/cm^2 Al adsorber and a NaI(Tl) scintillation crystal.

The feed solution was stopped after 9.2 liters of solution had passed through the column because the flow rate dropped from 30 ml/hr to less than 5 ml/hr and the effluent activity increased from 12% to

69% of the influent activity.

The column was allowed to drain for 64 hours, then one liter of tap water was passed through the shale bed at a rate of 10 ml/hr. This leach solution was sampled and counted.

The shale column was then placed inside a 3" aluminum tube previously filled with lead a 10 mm diameter hole through the center, and scanned on a gamma spectrometer. The external radiation from the column was so high that descrimination of discrete energies was impossible.

The column was cut into four almost equal segments and each leached with 1:1 hot H_2SO_4. The acid leach solution was analyzed radiochemically with the procedure developed by Kahn.[9] Chemical and radiochemical analyses were made also of selected samples of the column effluent collected during its operation.

Results:

A plot of the effluent beta activity from a 24 gram column of Conasauga shale versus the volume of ORNL pit No. 3 liquid wastes passed through as the feed solution is shown in Figure 2.

Figure 2 also contains a plot of the gamma activity passing through a 1000 mg/cm^2 Al adsorber and counted with a scintillation counter.

Radiochemical analysis of certain effluent samples were made to ascertain what radionuclides were leaking through the shale column, their order of appearance, and to determine whether the waste solution contained a "built-in" tracer which would indicate when the capacity of

the shale had been exhausted. The results are shown in Table 3.

Table 4 contains the qualitative and quantitative radiochemical analyses of the acid leach solution from the four shale segments. The results shown on this table can be used to determine the capacity of the shale. One can also calculate the fraction of saturation of the three lower segments assuming the first quarter was saturated.

The materials balance sheet is shown in Table 5.

Discussion:

The curves of the effluent activity versus volume of waste passed through the column, shown in Figure 2, indicate (1) the beta to gamma activity ratio remains constant from the initial leak-through until the breaking point, at which time both counting rates began to increase sharply. After the onset of break-through, the ratio of beta to gamma activity increased in favor of the beta activity.

No attempt was made to reproduce field conditions in this laboratory experiment. The tests were set up to obtain information on the soil using liquid waste from the pits. The shale used was 60-70 mesh and the column length 14 inches. The data shown on Table 3 indicate that laboratory studies gave results similar to those obtained in the field. Ruthenium 106, the first radionuclides to appear in test wells and seeps in the pit area, also was first to appear in the column effluent. These laboratory column results also agree with previous work done by the author on removal of ruthenium 106 from W-6 waste by clay adsorption. Using an ion exchange technique about 10% of ruthenium 106 in a basic carbonate solution (pH 12.0) was removed on

a cationic resin, 50% on an anionic resin while 40% was not exchanged. The data using shale indicates that after 3.0 liters have passed through the shale, the ruthenium is about 9% cationic and 91% non-ionic.

Base Exchange:

Clay, shale, and soil materials, when moist, are usually charged electronegatively (zeta potential). What accounts for the exchange capacity in shale are (1) broken bonds at the edges of silica-alumina units, (2) substitution within the lattice structure of a trivalent for quadrivalent ions resulting in unbalanced charges within the shale unit, and (3) replacement of the hydrogen of exposed hydroxyl groups by a cation.

The determination of cation exchange capacity is generally made by saturating the clay with NH_4^+ or Ba^{++} and determining the amount held by the shale or clay at pH 7. The presence of trivalent aluminum and iron tend to block some of the exchange groups and limit the accuracy of the analysis.

The exchange capacities of clay minerals types are: montmorillonite > illite > kaolinite. The exchange affinities for most ionic systems, in aqueous media follow the lyotropic series:

$$Li < Na < K < Rb < Cs$$
$$Mg < Ca < Sr < Ba$$
$$Al < Sc < Y < Eu < Sm < Pr < Ce < La$$

Surface Adsorption:

The outside faces, edges, and corners of the shale crystal have associated with them various cations (hydrogen, calcium, sodium).

When these surface atoms are ionized, the shale particle carries a negative ionic charge: the positive occupy a position in the solution adjacent to this negative charge. This is often the cause of failure of small clay particles in water to coagulate, because when they approach each other mechanically, there is a repulsion due to the similar charges on each.

The surface of a shale or clay particle could adsorb an unimolecular layer of the radioactive material.

Interstitial Precipitation:

When solutions pass through shale columns, if the solubility product constant is exceeded, precipitation of certain ions will take place between the shale particles. Removal of various ions from the liquid phase and their replacement with milliequivalent amounts from the solid phase can cause local precipitation which decreases the flow rate, resulting in more local precipitation and eventual plugging.

From the analysis shown on Table 1, ruthenium was the major radionuclide appearing in the column effluent. The unadsorbed fraction was either anionic or complexed. Ruthenium like other members of the transition series, exhibits a very strong tendency to form complex ions. There is no evidence that the simple or aquo ions of any of the valence states of ruthenium are capable of existence at all. (13)

As the column approached saturation, after 6.5 liters, the concentration of cesium 137 in the column effluent increased rapidly. This resulted in a decreased per cent removal, because cesium 137 contributes about 80% of the total beta activity of the liquid waste

in the pits. Strontium and barium, but not cesium, are precipitated as the carbonates in the waste tank and also in the surface pits. Therefore, the liquid fraction of the waste is mainly radiocesium.

Other fission products are not present in any appreciable amounts because they have been coprecipitated or, at the high pH of this waste, may have formed the metallic hydroxide which decrease in solubility with increasing molecular weight and valence.

Capacity of Soil:

In laboratory experiments, the U. S. Geological Survey determined the shale to have a base exchange capacity of 27.8 milliequivalents per 100 grams.

Considering the actual liquid waste in ORNL pits and the chemically untreated shale, as used in this experiment, about 1 million gallons of ORNL waste can be decontaminated adequately with 10 cubic yards of weathered Conasauga shale. The shale will remove about 85% of the total activity. The remaining 15% will be nine-tenths ruthenium and one-tenth a mixture of unidentified radionuclides. This 10^5 gallon per cubic yard is not the total capacity of the shale but the breakthrough capacity based on the 65 liter value as shown in Figure 2. From the values of Table 4, the capacity is about 7.7×10^8 d/m/gram.

The order of appearance in the effluent based on the rate of increased concentration, Table 3, Part III, was ruthenium, cesium, strontium, and trivalent rare earths. The concentrations of TRE and Sr in the column influent are too dilute to evaluate the significance of the data reported here.

Indicator:

Cesium 137 may be the "built-in" indicator of approaching saturation of the shale. For example, the per cent contributed to the gross influent activity by Cs was 83%; in the column effluent after 3.0 liters, it was .008%, at 6.5 liters, 3.5%, and at 7.5 liters, 19%, while after 3.0 liters Ru^{106} was 87% of the total effluent activity and after 6.5 liters, 59%.

Samples of column influent and effluent were analyzed for radicals and anions. The results are given in Table 2. The non-radioactive analyses included sodium, aluminum, and ammonia for cations and nitrates, carbonates, and sulfate as anionic radicals. A study of this table indicates that a non-radioactive element or radical may exist in the waste that would serve as a good indicator to forewarn when saturation of the soil is being reached. Calcium and sodium concentrations in the effluent before breakthrough was about 6% higher than in the influent. This value is only of borderline significance. However, the nonradioactive radical that may serve as an indication is NH_3. The concentration of ammonia in the influent was approximately 0.002 mg/ml. The concentration in the effluent up until breakthrough was 0.005 mg/ml. After 6.5 liters, the concentration of ammonia decreased as the concentration of radioactivity increased. Before one can say the ratio of the concentration of ammonia in the influent and effluent can be used as an indicator of shale saturation, further investigations are indicated.

However, determinations of the exchangable-cation composition involves the complete displacement of all the exchange cations (not

present in the sample). The solution obtained must be analyzed for all the cations in question, preferably by use of the flame photometer.

Leach With Water:

After the column was allowed to drain dry, the column was leached with approximately 50 column volumes of tap water (1 liter). The total activity in the effluent was about 1% of the total activity on the column. Three-fourths of this 1% activity was obtained in the first 300 mls of water-leach solution. The remaining 700 mls leached only 0.25 per cent of the total adsorbed activity. It is believed much of the first 2 or 3 column volumes (30-45 ml) tap water-leach solution contained radioactive material that was held between the shale particles and was not actually adsorbed, but did not drain when the columns was allowed to drain. The most significant implication of this water-leach test is that once the radionuclides are removed from the liquid phase, only about 1% will be leached, using 50 column volumes. Therefore, the waste adsorbed on the shale in the area of the ORNL waste pits nowwill not be leached to any appreciable extent by rain and ground waters, even when the pits have been abandoned.

Material Balance:

A material balance was made as shown on Table 5. The material balance sheet indicates (1) accounting of the radioactive materials involved, (2) a method of cross-checking values obtained, and (3) the relative degree of accuracy of the radiochemical analysis.

General Discussion:

The column used in this test was very small. Therefore, care was

exercised to minimize the wall effect. Franzini[10] indicates that in general, the ratio of permeameter diameter to mean particle diameter should exceed 40 to eliminate any sizeable effect of the container wall. For ratios less than 40, the Reynolds number of the flow has great influence on wall effect. Calculations indicate the D/d ratio was 32. However, the Reynolds number DV/v when v is kinematic viscosity, was less than five; therefore, the flow through the shale was laminar and the wall effect negligible.

In comparing the clay saturation results reported herein with those of others, it is interesting to note that with a great many differences in mechanics of operation, Blanchard[11] obtained a saturation value of 7.8×10^8 d/m/g. The operation consisted of interupting the flow through the column each evening, using 20 grams of similar shale from the Conasauga formation and treating enough waste to saturate the column (13 liters).

The results reported here (7.7×10^8 d/m/g.) were obtained using a long, thin column, slow flow rate, continuous column operation, 24 grams of the light colored, weathered fraction of Conasauga shale, and treating 9.2 liters of waste before it was stopped (effluent was 68% of the influent activity). In general, both results were similar to those obtained by Goldman and Parker[12] in an earlier column study, i.e., 2.2×10^8 d/m/g of cesium and by the author on a series of slurry type jar test[7] (saturation value of 2×10^9 d/m/gram for a fission product mixture).

Conclusions;

1. Breakthrough may be predicted by the rise in concentration of Cs^{137} from about 12 c/m after 3 liters to 5, 130 c/m at the start of breakthrough to 27,700 after 7.5 liters.

2. The order of breakthrough appears to be: first, ruthenium and other unknown radionuclides; second, trivalent rare earths; third, strontium and soon followed by cesium. However, it should be stated that the TRE and Sr concentrations are very low when compared to the total; therefore, sample contamination is difficult to avoid.

3. Leaching by tap water removes very little activity once it is adsorbed on the shale. (50 column volumes removed less than 1%).

4. Capacity of the shale for cesium = 7.7×10^8 dpm/g.

5. The concentration of ammonia in the effluent from an unsaturated shale column is 2-3 times higher than the influent concentration before breakthrough. This amount starts to decrease and approach the influent value. Ammonia may be a stable constituent that would serve to indicate the approach of breakthrough of the activity.

6. The shale removes about 15% of the total ruthenium in the waste. 50% of the remaining Ru can be removed by a weak basic type anionic ion exchange resin.

7. 68% of the total activity (63% of the Cs activity) appeared after 9.2 liters of waste had passed through the column.

8. Ruthenium and some unidentified gamma emitters appeared in the initial samples of the column effluent. There, concentrations in the effluent remained almost constant over the entire run.

9. Additional work should be done on the following:

   a. Breakthrough sequence.

   b. Identification of the other radionuclides which are present in the waste solution.

   c. Investigation of the change in concentration of ammonia before breakthrough.

   d. More quantitative study on the effects of leaching a saturated column with tap water and with weak acid, salt solutions, etc.

TABLE I

Radiochemical Analysis of Liquid Waste

Radio-element		Activity in 10^3 counts/min/milliliter					
		Influent (Waste Liquid)	Effluent Volume in Liters				
			3.0	6.5	7.5	8.25	9.2
Gross Beta		147	19.	29.	51.5	98.	101.
Ru Beta	14.1% of Gross β	20.7	18.6	19.1	19.2	19.8	20.3
Cs^{137} "	86% of Gross β	127.0	0.012	[illegible].1	27.7	73.7	79.9
TRE Beta	1.3% of Gross β	1.8	0.405	0.707	1.09	0.44	2.04
Sr^{90} "	0.9% of Gross β	1.24	0.017	0.314	0.558	0.601	1.35

TABLE II

Chemical Analysis of Non-Radioactive Constituents

Concentration in mg/ml*

Constituent	Feed Solution	3.0 L.	6.5 L.	7.5 L.	8.25 L.	9.2 L.
Na^+	13.60	14.25	14.00	13.75	14.25	15.50
NH_3^+	<.02	0.05	0.05	0.04	0.03	0.03
Al^{+++}	.25	0.236	0.195	0.265	0.195	0.270
NO_3^-	11.35	14.5	11.0	8.0	9.0	10.0
SO_4^{--}	1.65	1.80	1.80	1.80	1.71	2.00
CO_3^{--}	2.88	2.30	2.36	1.86	2.40	1.97
pH	12.0	10.7	10.3	10.7	10.3	10.2

* except pH values.

TABLE III

Radionuclide Composition of the Activity

I. Total Influent Activity Taken as 100%

Effluent Activity Contains after:	3 liters	6.5 liters	7.5 liters	8.25 liters	9.2 liters
			Per Cent		
Radioelement					
Ruthenium	12.7	12.9	12.9	11.8	15.5
Cesium	.008	3.5	19.0	50.0	51.9
Tri rare earths	0.3	0.5	0.7	0.3	1.4
Strontium	0.12	0.2	0.4	0.4	0.9
Unidentified	6.6	1.3	2.0	3.3	1.1
Adsorbed by soil	87.8	81.5	65.0	34.2	32.2

II. Composition of Effluent Activity in Per Cent

after:	3 liters	6.5 liters	7.5 liters	8.25 liters	9.2 liters
Ruthenium	86.8	59	33	18	19.1
Cesium	.06	18.0	53	75	79
Tri. rare earths	.21	0.24	2.1	4.5	2.0
Strontium	.09	1.1	1.1	0.6	1.3
Unidentified	13.7	21.7	20.8	27	0.0

III. Removal of Each Element in Per Cent*

after:	3 liters	6.5 liters	7.5 liters	8.25 liters	9.2 liters
Ruthenium removal	19	17	17	14	12
in effluent	81	83	83	86	88
Cesium removal	99.991	96.0	78	42	38
in effluent	0.009	4.0	22	58	62
Tri rare earths removal	77	61	39	24	0
in effluent	23	39	61	76	100
Strontium removal	98.6	75	55	52	0
in effluent	1.4	25	45	48	100

* Per cent in effluent values were measured and per cent removal values obtained by subtraction.

TABLE IV

Radiochemical Analysis of the Activity on the Shale

Fraction of Column	Radioelement	Weight of Sample Grams	Amount of Activity Leached d/m	Per Cent Saturation*	Specific Activity Adsorbed d/m/gram
1st Quarter	Cs^{137}	2.0104	1.537×10^9	100%	7.65×10^8
	Ru^{106}		2.11×10^6		
	Sr^{90}		5.05×10^6		
	TRE		7.84×10^6		
2nd Quarter	Cs^{137}	1.6437	1.016×10^9	81	6.18×10^8
	Ru^{106}		1.26×10^6		
	Sr^{90}		4.12×10^6		
	TRE		5.43×10^6		
3rd Quarter	Cs^{137}	2.7749	1.269×10^9	60	4.57×10^8
	Ru^{106}		1.55×10^6		
	Sr^{90}		4.42×10^6		
	TRE		6.22×10^6		
4th Quarter	Cs^{137}	2.6802	8.73×10^8	43	3.25×10^8
	Ru^{106}		9.76×10^5		
	Sr^{90}		3.63×10^6		
	TRE		4.69×10^6		

* Assuming 1st Quarter 100% saturated and 10% counting efficiency for Ru, Sr, and TRE.

TABLE V

Materials Balance Sheet

1.5×10^5 c/m/ml $\times$ 10^3 ml/L $\times$ 9.2 L = 1.38×10^9 c/m feed solution

Effluent (total activity to breakpoint) + (activity breakpoint to end of run)

$$(2.1 \times 10^4 \times 10^3 \times 6.5) + 1/2\,(2.7 \times 10^5 \times 10^3) = \frac{.27 \times 10^9 \text{ c/m leaked}}{1.11 \times 10^9 \text{ on column}}$$

Activity leached using 1 liter tap water

$$(2.5 \times 10^4 \times 300) + (3.6 \times 10^3 \times 700) = \frac{.01 \times 10^9 \text{ water leached}}{1.10 \times 10^9 \text{ on Shale}}$$

Activity as d/m/g leached from each 5-gram sample of shale

$$\frac{(7.65 \times 10^8 \times 5)}{10} + \frac{(6.18 \times 10^8 \times 5)}{10} + \frac{(4.57 \times 10^8 \times 5)}{10} + \frac{(3.26 \times 10^8 \times 5)}{10} = \frac{1.08 \times 10^9 \text{ acid leached}}{0.02 \times 10^9 \text{ remaining on Shale}}$$

or

10^6 c/m/g of Shale unaccounted for

NOTE: Although the balance shows 98% accountibility, the counting, chemical analysis, and experimental procedure, in general, would have limitations of at least $\pm$ 5%.

Also assume $\frac{d/m}{10} = C/M$

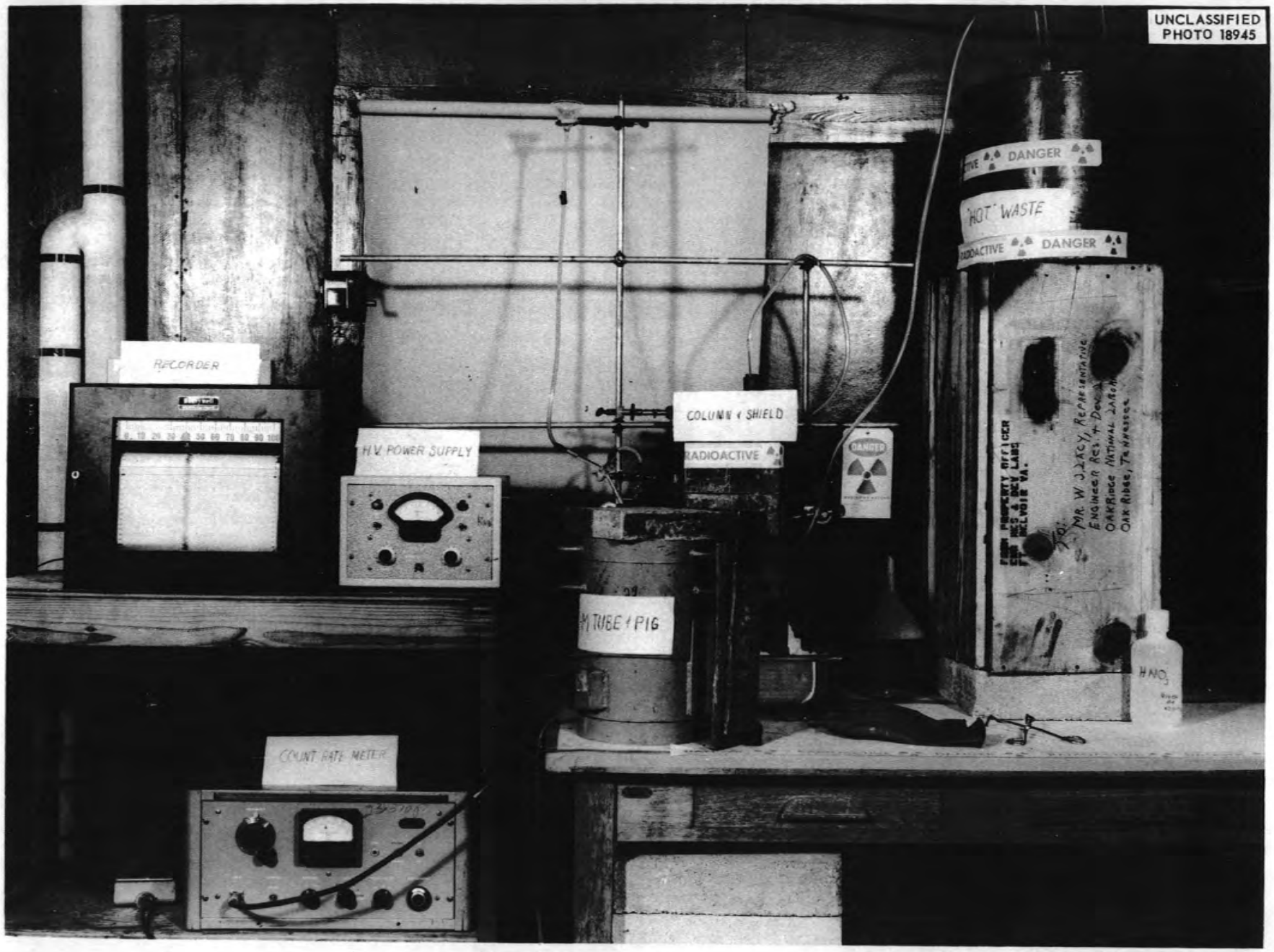

Fig. 1. The Experimental Setup for Evaluation of the Shale Column.

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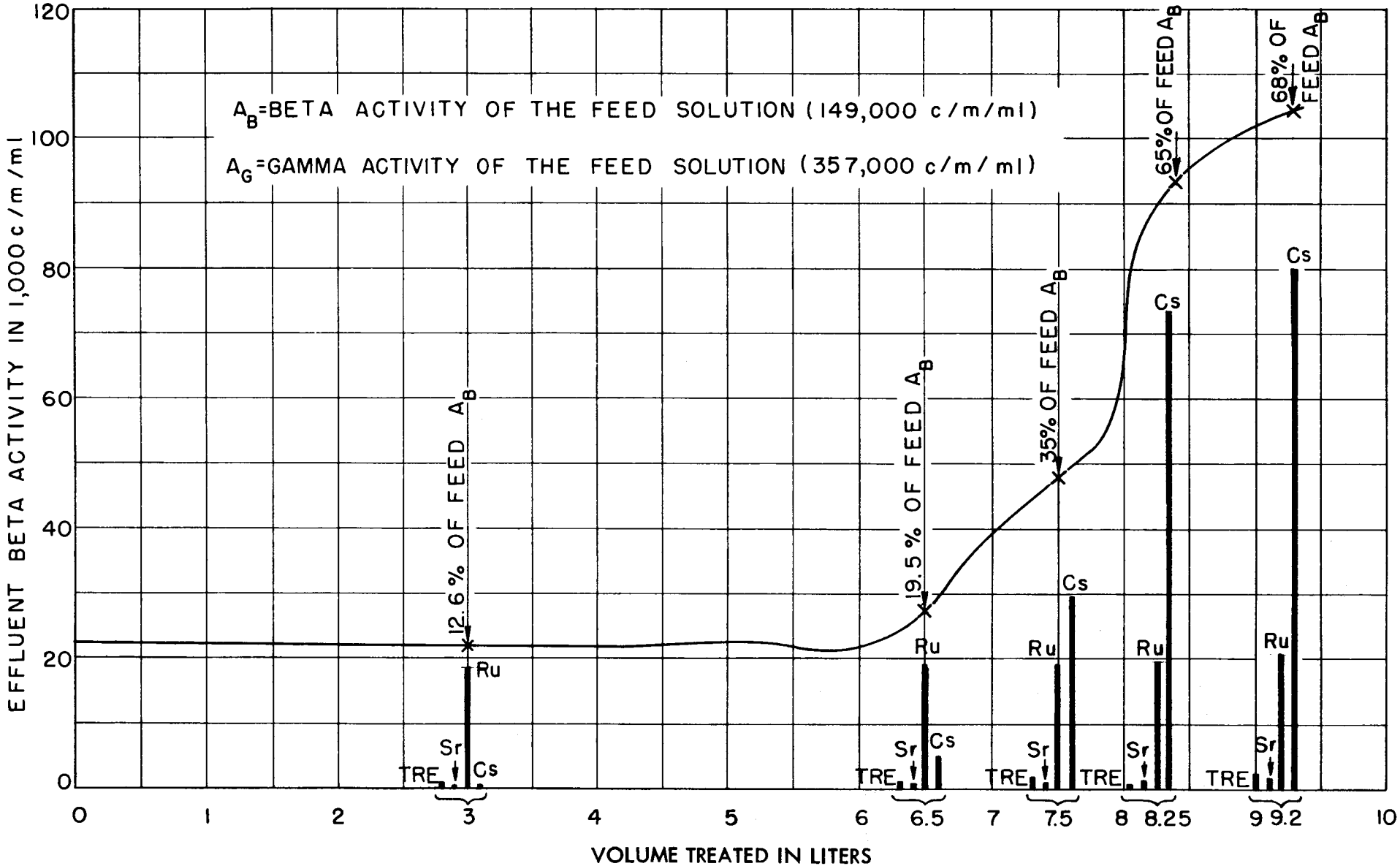

Fig. 2. Curve of the Effluent Beta Activity and Bar Graphs of the Contributors to This Activity.

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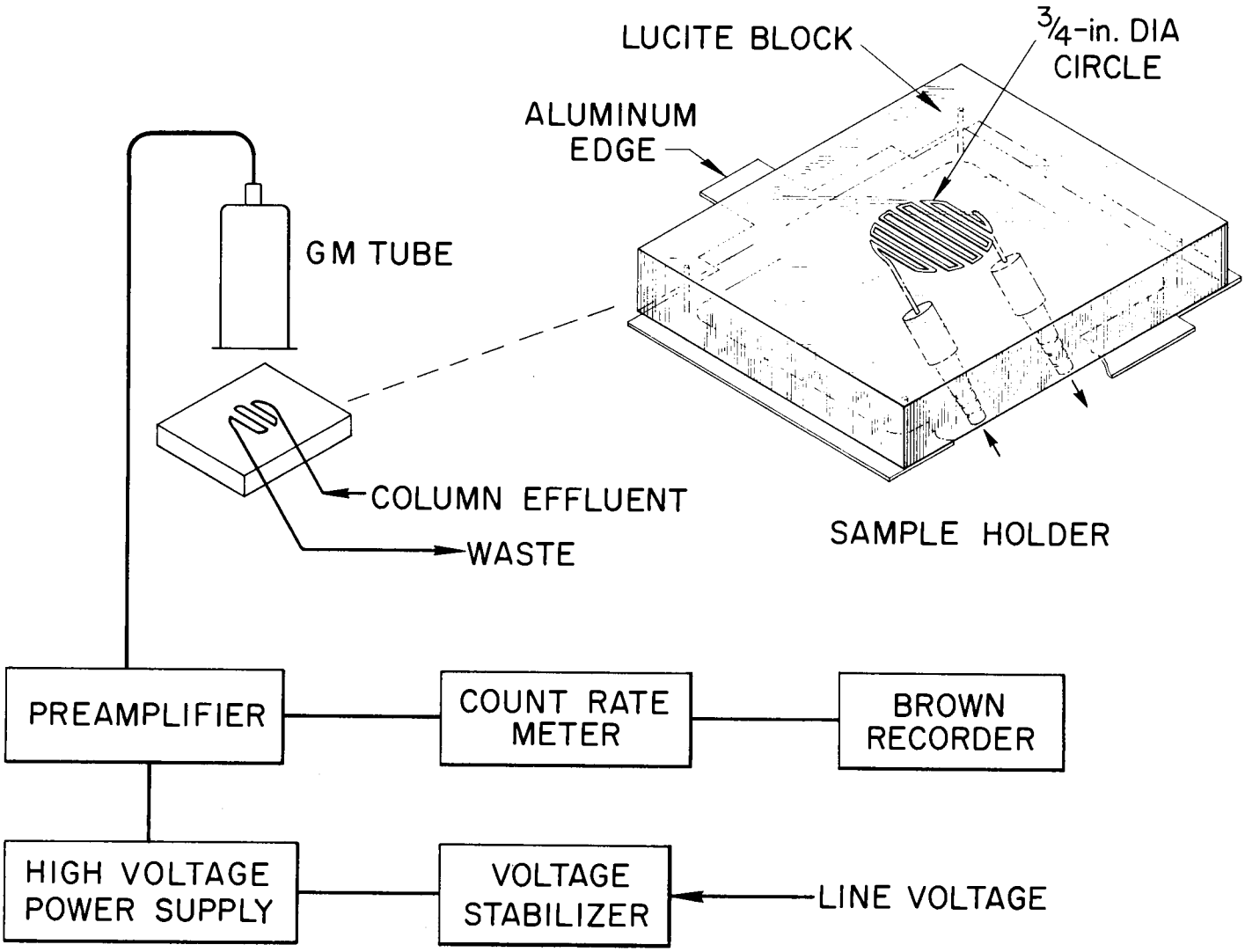

Fig. 3. Schematic Diagram of the Counting Device.

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