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RESEARCH ESTABLISHMENT

LUCAS HEIGHTS

TWO PROCESSES FOR DECONTAMINATION OF RADIOACTIVE AQUEOUS WASTES BY PRECIPITATION AND COAGULATION

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ABSTRACT

In an investigation of the efficiency of the calcium/ferric phosphate process for removal of various radio-isotopes from solution using a sludge blanket clarifier 70 per cent. removal of uranyl and strontium ions and mixed fission products was achieved but cesium removal was poor, in agreement with other workers. Dosing ion concentrations were 80 p.p.m. PO_4^{---}, 50 p.p.m. Ca^{++} and 40 p.p.m. Fe^{+++}, used at an initial pH of 11.5. Use of a lower concentration and lower pH values did not materially affect removals. In batch experiments to find the optimum concentration of ions for precipitation at a pH of 9.5 it was found that concentrations of 90 p.p.m. PO_4^{---}, 50 p.p.m. Ca^{++}, and 40 p.p.m. Fe^{+++} leave very small residual concentrations in the supernate.

Removals of radio-isotopes from solution by precipitation using the aluminium hydroxide process are comparable with those in the calcium/ferric phosphate process, except for that of strontium. It is recommended that 8.0 be used as the maximum pH for the precipitation of aluminium hydroxide in the sludge blanket clarifier.

Thickening experiments with a sludge blanket clarifier using both processes show that the degree of thickening is governed by the size and relative proportions of the clarifier.

CONTENTS

1. INTRODUCTION

Two processes are being considered for use at Lucas Heights to decontaminate low-level radioactive aqueous effluent in a Dorr sludge blanket clarifier which has a rated capacity of 50,000 gallons per day. They are:

(a) the calcium/ferric phosphate process, and

(b) the aluminium hydroxide process.

2. THE CALCIUM/FERRIC PHOSPHATE PROCESS

2.1 Outline of Experiments With the Process

Work at Harwell (Seedhouse et al., 1958) using a sludge blanket clarifier has shown that with concentrations of 80 p.p.m. PO_4^{---}, 50 p.p.m. Ca^{++}, and 40 p.p.m. Fe^{+++} in the effluent at an initial pH of 11.5 and final pH of 10.8 or higher, excellent removal of radio-isotopes was obtained with the residual concentrations of the dosing ions totalling about 5 p.p.m.

The use of such high concentrations of dosing ions makes the cost of chemicals much greater than that for the aluminium hydroxide process using 20 p.p.m. Al^{+++}. In addition, a pH of 11.5 in the sludge blanket clarifier has the disadvantage that the pH of the treated effluent is too high for discharge without pH adjustment. Therefore experiments were carried out to determine whether the use of lower concentrations of the precipitating ions $[PO_4^{---}]$ = 40 p.p.m., $[Ca^{++}]$ = 30 p.p.m., and $[Fe^{+++}]$ = 20 p.p.m. at lower pH values would affect radioactivity removal.

Thickening and precipitation experiments were also carried out.

2.2 Description of Apparatus

2.2.1 Pilot plant

A schematic diagram of the apparatus is shown in Figure 1. Mains water was used as the effluent with a constant head device (A) to give a steady water flow. The water was metered into a rapidly stirred, baffled, 1 litre mixing vessel (B). Solutions containing the coagulating ions (PO_4^{---}, Ca^{++}, and Fe^{+++}), the active solution, and a sodium hydroxide solution also flowed continuously from head tanks by gravity into vessel B. The NaOH solution adjusted the pH of the clarifier influent to the desired value. The residence time in this vessel was short and the various solutions were thoroughly mixed before entering the clarifier(C). Precipitates of calcium phosphate and ferric phosphate were formed in mixing vessel B but, due to rapid agitation, the particles remained colloidal.

2.2.2 Square clarifier

The square clarifier, which was used in the decontamination experiments, is shown in Figure 2. The effluent containing colloidal calcium and ferric phosphate passed through the square distributor into the inner compartment of the clarifier where the floc particles aggregated. The suspension was slowly stirred to prevent floc settling and also to increase the rate of particle collision to facilitate coagulation. Eventually the floc was swept into the outer compartment of the clarifier where the sludge blanket built up. Sludge which settled was withdrawn periodically from the outlets on all sides of the clarifier. In the course of a run the upper part of the blanket was not greatly disturbed. The particles in the blanket were mobile so that there was no channelling of the effluent

at one place. The blanket acted as a filter medium separating solid particles from the liquid phase. The decontaminated water overflowed into the launder and went to drain. The square vessel had the disadvantage that dead spots caused the accumulation of floc at the corners of the inner compartment where it could not be removed.

2.2.3 Circular clarifier

To simulate the operation of the Dorr sludge blanket clarifier in the thickening experiments, it was necessary to build a model circular clarifier of similar design (Figure 3). The model had a perspex tank 21 inches inside diameter and 11½ inches deep, and was used with the apparatus shown in Figure 1. The water containing the colloidal phosphates of calcium and iron was fed into the clarifier from the bottom. It flowed radially along the distributing arms and horizontally through the 4 orifices on each arm into the sludge blanket. The orifices were baffled to reduce the speed of the water as it left the arms. The blanket acted as a filter, separating the particles from the water to maintain a clear clarification zone. The decontaminated water overflowed into the launder and passed to drain. The particles in the blanket gradually settled as they aggregated. The settled sludge was thickened by the slow movement of the distributing arms and was slowly swept towards the annular sludge well which was 3 inches deep and of inside diameter 7 inches. Each distributing arm had a cylindrical rake; these were situated at different distances below the arms to thicken the sludge at different levels. Rakes attached to the foot of the rotating sludge cylinder provided further thickening of the sludge in the well. Sludge was withdrawn by gravity through a spring loaded cock.

2.3 Decontamination Experiments

2.3.1 Experimental procedure using the pilot plant

Seedhouse et al., (1958) found that the flow rate of their 74 litre model clarifier could be raised from 0.5 to 1 litre per minute without affecting operation. In preliminary experiments with the clarifier (C) of 83.5 litres using a flow rate of 1 litre per minute control was achieved with no trouble so this rate was used in all subsequent experiments.

The coagulating ions Fe^{+++}, Ca^{++}, and PO_4^{---} were introduced using ferric chloride, calcium nitrate, and tri–sodium phosphate solutions. The ferric chloride solution was acidified by HCl to give a 0.15 N acid solution to prevent any after–precipitation of ferric hydroxide. The stock solutions were of such strength that a flow rate of 3 ml/min would give the desired ionic concentration in the effluent. Maintenance of feed rates was checked in several runs, and the error was found to be within 6 per cent. The clarifier was emptied after runs with each isotope and a fresh blanket built up using equivalent quantities of tri–sodium phosphate, ferric chloride, and calcium nitrate. A run lasted at least 3 hours so that the supernate was truly representative when sampled. After each run, most of the sludge was drained off to leave only an insubstantial blanket for the next run with the same isotope.

2.3.2 Counting procedure

In each case 5 ml aliquot samples of the supernate were evaporated on 2 in. aluminium trays. After evaporation the trays were weighed. Where necessary the counts were corrected for self–absorption.

The methods used in the various experiments were as follows:

$[U^{233}O_2]^{++}$ Activity. – 2π alpha gas flow proportional counting using pure argon.

Mixed Fission Products. – Total β activity – 2π beta gas flow proportional counting using argon with 10 per cent. methane.

Total γ activity – well crystal scintillation counting using a sodium iodide crystal.

Total strontium activity – 2π beta gas flow proportional counting was used after chemical separation of the strontium from other elements.

Cesium ^{137}. – 2π beta gas flow proportional counting.

Cesium ^{137} + Strontium ^{90} + Yttrium ^{90}. – The total β activity was determined using 2π beta gas flow proportional counting. The cesium γ activity was determined using well crystal scintillation counting. The β activity of the Cs^{137} can be calculated from the γ activity. The strontium β activity was determined by 2π beta gas flow proportional counting after chemical separation. The yttrium β activity was obtained by subtraction.

2.3.3 Results of decontamination experiments

Radioactivity removals from the runs using selected radio–isotopes are shown in Table 1. In these runs the square sludge blanket clarifier was used.

2.3.4 Discussion of results of decontamination experiments

$[U^{233}O_2]^{++}$ Removal

The activity of the $[U^{233}O_2]^{++}$ dosing solution was 7.6×10^3 dis/(min)(ml). The carrier was a 0.1 g/litre uranyl nitrate solution acidified to pH 1.0 by nitric acid. As shown by the results from runs 3, 4, 5, and 6 (Table 1) decontamination by this process is very good at low or high pH values, using high or low concentrations of the ions.

Cs^{137} and Sr^{90}–Y^{90} Removal

The activity of the Cs^{137} solution was 1.456×10^4 dis/(min)(ml); the carrier used was 0.01 g/litre Cs Cl. The activity of the equilibrium $Sr^{90} - Y^{90}$ solution was 1.735×10^4 dis/(min)(ml); the carrier used was 0.01 g/litre $Sr(NO_3)_2$. Equal volumes of the two solutions were mixed to give the active dosing solution for runs 1 and 2. Since differentiation between the three isotopes is subject to ± 25 per cent. error, the total β results have been tabulated as well as those for the two components.

Strontium ^{90}– yttrium ^{90} removal was very good, which is to be expected in view of the chemical similarity of strontium and calcium and the fact that yttrium decontamination by calcium phosphate precipitation is excellent (Straub et al., 1951). Cesium removal was very poor, in agreement with the experience of Seedhouse et al., (1958).

Cesium is very difficult to remove by precipitation because:

(a) the large size of the ion tends to prevent it taking a place in the lattices of smaller cations like calcium and iron, and

(b) its monovalency makes adsorption unlikely.

TABLE 1

Removals of Selected Radio-Isotopes

Run No.	Isotope		Final pH	Initial Activity, dis/(min)(ml), and Counting Precision, %	Overflow Activity, dis/(min)(ml), and Counting Precision, %		Activity Removal %		Concentrations of Dosing Ions
					Unfiltered	Filtered	Unfiltered	Filtered	
1	$Cs^{137}-Sr^{90}-Y^{90}$		10.75	90 ± 5%	31 ± 10%	31 ± 10%	65.0	65.0	High concentrations
2	$Cs^{137}-Sr^{90}-Y^{90}$		10.95	90 ± 5%	43 ± 8%	43 ± 8%	52.0	52.0	Low concentrations
1	$Sr^{90}-Y^{90}$		10.75	48 ± 8%	2 ± 25%	2 ± 25%	96.0	96.0	High concentrations
2	$Sr^{90}-Y^{90}$		10.95	48 ± 8%	1 ± 25%	1 ± 25%	98.0	98.0	Low concentrations
1	Cs^{137}		10.75	42 ± 8%	27 ± 25%	27 ± 25%	35.0	35.0	High concentrations
2	Cs^{137}		10.95	42 ± 8%	41 ± 25%	41 ± 25%	2.0	2.0	Low concentrations
3	U233		9.6	43	1	1	98.0	98.0	Low concentrations
4	U233		10.1	43	0.98	0.98	97.0	97.0	High concentrations
5	U233		11.0	43	1.8	1.8	96.0	96.0	High concentrations
6	U233		11.5	43	0.8	0.8	98.0	98.0	High concentrations
7	Mixed Fission Products	Total Sr	8.6	27 ± 10%	4 ± 100%	2 ± 100%	86.0	93.0	High concentrations
		Total β	8.6	129 ± 5%	30 ± 10%	30 ± 10%	77.0	77.0	
8	Mixed Fission Products	Total Sr	8.7	27 ± 10%	3 ± 100%	2 ± 100%	98.0	97.0	Low concentrations
		Total β	8.7	129 ± 5%	38 ± 10%	38 ± 8%	72.0	72.0	
9	Mixed Fission Products	Total Sr	8.95	27 + 10%	6 ± 100%	3 ± 100%	78.0	89.0	Low concentrations
		Total β	8.95	129 ± 5%	47 ± 8%	47 ± 8%	57.0	57.0	

High concentrations: 50 p.p.m. Ca^{++}
40 p.p.m. Fe^{+++}
80 p.p.m. PO_4^{---}

Low concentrations: 30 p.p.m. Ca^{++}
20 p.p.m. Fe^{+++}
40 p.p.m. PO_4^{---}

Mixed Fission Product Removal

The solution of 6 months old mixed fission products used in runs 7, 8, and 9 was acidified by nitric acid to pH 1.0 to prevent adsorption of the radio-isotopes on apparatus.

The radioactivity of this solution was :-

total γ radioactivity = 1.87×10^4 dis./(min)(ml)

total β radioactivity = 2.22×10^4 dis./(min)(ml)

total Sr β radioactivity = 4.7×10^3 dis./(min)(ml)

Differentiation between strontium ($Sr^{89} + Sr^{90}$) and other β-emitters in the clarifier overflow was subject to an error of ± 100 per cent., but taking the highest filtered overflow strontium activity and adding 100 per cent. to the count the strontium removal is 78 per cent. which agrees fairly well with the removal from a $Cs^{137} - Sr^{90} - Y^{90}$ mixture.

The Effect of Using Reduced Concentrations of Reagents

The effect on decontamination using the lower concentrations of the precipitating ions and pH values lower than 11.5 is small for all radioactive species used. In the case of the Dorr sludge blanket clarifier this could lead to halving the cost of chemical used. Operation with the high concentrations (Table 1) at a pH of 11.5 and the low concentrations (Table 1) at a pH of 9.5 would cost £2400 p.a. and £1200 p.a. respectively for a clarifier throughput of 30,000 gallons per day and five-day working week. (The maximum throughput is 50,000 gallons per day but with the coagulant concentrations used, 30,000 gallons per day is the average throughput).

2.4 Thickening Experiments

A comparison of operating costs of processes must allow for the volume of sludge produced per 10^6 gallons of effluent treated in addition to costs of labour, depreciation, and chemicals.

An investigation was made of the sludge thickening that can be obtained in the circular model sludge blanket clarifier similar in design to the Dorr clarifier (Figure 3). It was found that the sludge increased in concentration along the bottom from the periphery to the sludge well of the clarifier, and reached a maximum at the bottom of the well, at the take-off level. The highest sludge concentration attained in the calcium/ferric phosphate process was 10 per cent. solids. This can be compared with 20 per cent. solids in the sludge produced by the Dorr clarifier. It appears that the size and relative proportions of the clarifier govern the degree of thickening produced.

2.5 Precipitation Experiments

2.5.1 Effect of lowering of pH value

As already mentioned, Seedhouse et al., (1958) have shown that with concentrations of ions of 80 p.p.m. PO_4^{---}, 50 p.p.m. Ca^{++}, and 40 p.p.m. Fe^{+++} and a final pH value of 10.8 or higher virtually complete precipitation is obtained. The use of a lower pH value would:

(a) reduce the cost of alkali for initial adjustment of the pH of the effluent, and

(b) give a treated effluent with a pH value more acceptable for discharge without pH readjustment.

A final pH value of 9.5 was chosen for batch experiments to test whether the above concentrations of ions continued to give complete precipitation.

2.5.2 Experimental procedure

Two preliminary experiments were made; details are summarised in Table 2 and the results are shown in Table 3. In each case 950 ml of water was made sufficiently alkaline to give 1,000 ml of water containing the required concentrations of ions at the desired final pH after the addition of the dosing reagents. Phosphate solution and then the cations in the order described in Table 2 were added to the alkaline water. After precipitation the suspension was boiled for 6 hours and then filtered with filter pulp to remove any colloidal solids from the supernate before analysis. The precipitate was analysed for the weight ratio of the coagulant ions.

These experiments showed that at a final pH value of 9.5 precipitation of the dosing ions was incomplete, and four further experiments 3 (a) to 3 (d) using increased concentrations of PO_4^{---} ions were made. In these experiments the dosing reagents were added over a period of one minute to water whose pH had been adjusted to 11.4 to give a final pH of 9.5. The results of these precipitation experiments are given in Table 4 together with experiment 3 (e) in which dosing reagents were added to give concentrations of 45 p.p.m. PO_4^{---}, 25 p.p.m. Ca^{++}, and 20 p.p.m. Fe^{+++} ions.

2.5.3 Discussion of results of precipitation experiments

Sedimentation. The rates of sedimentation of the smallest particles of calcium/ferric phosphate in experiments 1(a) and 1(b) were comparatively low. When calcium phosphate alone was precipitated, the sedimentation velocity was lower still. This shows that co–precipitation with ferric phosphate improves the settling properties of calcium phosphate.

Precipitation. Bjerrum (1959) defined the degree of neutralisation N= calcium equivalents per molecule of H_3PO_4 in the precipitate. When precipitating calcium phosphate he found that as the pH increased the value of N rose to a value of 3.397 at a pH value of 11.0, and concluded that "a continuous range of calcium phosphate precipitates exists with N values varying from about 3–1/3 to less than 3" (N = 3–1/3 corresponds to hydroxyl apatite).

The increase of N to values greater than that for hydroxyl apatite shown in experiment 2(c) (Table 3) and in Bjerrum's work is due possibly to increasing proportions of the hydroxide being co–precipitated with the hydroxyl apatite as the pH of the environment increases. The co–precipitation of the hydroxide at higher pH values certainly occurs in the case of iron, as ferric phosphate is much less stable and is readily hydrolysed. The results given in Table 3 show that both N and the ratio of cation equivalents per molecule of H_3PO_4 are higher when precipitation occurs at the higher pH value. When co–precipitating calcium, iron, and phosphate ions there was a residual calcium ion concentration at a pH value of 9.5 and a residual phosphate ion concentration at a pH of 11.5. When precipitating calcium phosphate there was a higher residual phosphate concentration at a pH of 11.5.

All these results show that in this process a greater proportion of phosphates is precipitated as the pH of the environment falls. Consequently the concentrations found at Harwell to give complete precipitation of the ions at an initial pH of 11.5 and a final pH of 10.8 or higher do not give complete precipitation when the final pH is 9.5, when an increased concentration of phosphate ions is required. The results in Table 4 show that more complete precipitation is obtained as the concentration of phosphate ions is increased, 90 p.p.m. PO_4^{---} giving virtually complete precipitation of the cations.

Use of half concentrations of the coagulant ions, (45 p.p.m. PO_4^{---}, 25 p.p.m. Ca^{++}, and 20 p.p.m. Fe^{+++}) at a final pH of 9.5 was equally effective in giving complete precipitation.

TABLE 2

Details of Preliminary Precipitation Experiments

Expt. No.	Final pH	Initial Reagent Concn. (p.p.m.)			Method of Reagent Addition	Comment
		Ca^{++}	Fe^{+++}	PO_4^{---}		
1(a)	9.5	50	40	80	Iron and calcium solutions added over a 1-minute period	Immediate coagulation
1(b)	9.5	50	40	80	Iron solution added 5 minutes after the calcium solution	Calcium solution addition produced turbidity. Iron solution rapidly produced coagulation
1(c)	9.5	50	–	80	No iron solution addition	Faint turbidity
2(a)	11.5	50	40	80	Iron and calcium solutions added over a 1-minute period together	Immediate coagulation
2(b)	11.5	50	40	80	Iron solution added 5 minutes after the calcium solution	Calcium solution addition produced large floc. Iron solution addition caused settling
2(c)	11.5	50	–	80	No iron solution addition	Calcium solution addition produced large floc

TABLE 3

Results of Preliminary Precipitation Experiments

Expt. No. (see Table 2)	Final pH	Precipitate									Supernate		
		Wt. Ratio of Ions			Ratio of Ionic Equivalents			N	EQ. Cations / Mol. H_3PO_4	Sedimentation Velocity (mm/min)	Residual Concentration p.p.m.		
		Ca^{++}	Fe^{+++}	PO_4^{---}	Ca^{++}	Fe^{+++}	PO_4^{---}				Ca^{++}	Fe^{+++}	PO_4^{---}
1(a)	9.5	1.0	1.11	1.87	0.85	1.01	1.0	–	1.86	0.33	10.0	<0.05	<2.0
1(b)	9.5	1.0	0.9	1.78	0.94	0.9	1.0	–	1.84	0.38	5.5	<0.05	<2.0
1(c)	9.5	1.0	–	1.55	1.03	–	1.0	3.08		0.24	<0.01	–	7.5
2(a)	11.5	1.0	0.73	1.42	1.11	0.87	1.0	–	1.98	–	<0.3	<0.05	12.0
2(b)	11.5	1.0	0.4	0.9	1.75	0.75	1.0	–	2.5	–	<0.3	<0.05	12.0
2(c)	11.5	1.0	–	1.26	1.26	–	1.0	3.78	–	–	<0.3	<0.05	12.0

TABLE 4

Results of Precipitation Experiments

Expt. No.	Final pH	Initial Concentration of Ions (p.p.m.)			Final Concentration of Ions (p.p.m.)		
		Ca^{++}	Fe^{+++}	PO_4^{---}	Ca^{++}	Fe^{+++}	PO_4^{---}
3 (a)	9.5	50	40	80	8 ±1	0.05	2.0
3 (b)	9.5	50	40	83	7 ± 1	0.05	2.0
3 (c)	9.5	50	40	86	3 ± 1	0.05	2.0
3 (d)	9.5	50	40	90	0.3	0.02	2.0
3 (e)	9.5	25	20	45	3 ± 1	0.05	2.0

3. THE ALUMINIUM HYDROXIDE PROCESS

3.1 Outline of Experiments with the Process

In the aluminium hydroxide process the radioactive effluent is made alkaline by the addition of sodium carbonate; commercial aluminium sulphate is added to it in the sludge blanket clarifier described in Section 2.2 to give an aluminium concentration of 20 p.p.m. It was concluded from a literature survey that the optimum pH for precipitation of aluminium hydroxide would be 8.0 . Soluble iron in the aluminium sulphate used precipitates as ferric hydroxide and improves the settling characteristics of the floc..

The experiments were made at pH values between 6.5 and 9.5 to determine the optimum pH value for clarifier operation and effluent decontamination.

3.2 Decontamination Experiments

3.2.1 Results of decontamination experiments

The removal analyses and activities of samples from the runs are shown in Tables 5, 6, 7 and 8. An attempt was made to differentiate between Sr^{90} and Y^{90} in the strontium runs and between strontium and other β–emitters in the mixed fission product runs. The aluminium concentration of the unfiltered and filtered clarifier overflow samples was determined in most of the runs to indicate:–

(a) the degree of sodium aluminate formation, and

(b) whether colloid stabilisation occurs at high pH values.

The activities of the filtered and unfiltered samples show how the activity removal was affected by these two factors.

TABLE 5

Results from runs with an equilibrium Sr^{90} – Y^{90} mixture

Activity of clarifier influent = 97 dis/(min)(ml)

Run No.	Final pH	Al^{+++} concentration in overflow p.p.m.		Isotope	Final Activity dis/(min)(ml)		Activity Removal %			
		Unfiltered	Filtered		Unfiltered	Filtered	Unfiltered	Filtered	Average	
									Unfiltered	Filtered
1	8.05	1.3	0.8	Sr^{90} + Y^{90}	61	48	37.1	50.5		
				Sr^{90}	41	21	16	56.6		
2	8.84			Sr^{90} + Y^{90}	57	57	41.2	41.2		
				Sr^{90}	32	33	32	32	33.5	47.4
3	9.20	8.4	2.0	Sr^{90} + Y^{90}	54	40	44.3	58.7	43.8	51.25
				Sr^{90}	42	27	44	42		
4	9.47			Sr^{90} + Y^{90}	46	44	52.6	54.6		
				Sr^{90}	59	28	42	59		

TABLE 6

Results from runs with Cs^{137}

Activity of influent = 82 dis/(min)(ml)

Run No.	Final pH	Al^{+++} Concentration in overflow (p.p.m.)		Final Activity dis/(min)(ml)		Activity Removal %			
		Unfiltered	Filtered	Unfiltered	Filtered	Unfiltered	Filtered	Average	
								Unfiltered	Filtered
5	8.27	1.0	<0.5	86	47	0	43		
6	8.3	<0.5	<0.5	76	76	8	8		
7	8.6	3.2	1.6	80	78	3	5	4	17.2
8	9.2	12.0	10.3	78	72	5	13		

TABLE 7

Results from runs with $(U^{233}O_2)(NO_3)_2$

Activity of influent = 43 dis/(min)(ml)

Run No.	Final pH	Al^{+++} Concentration in overflow (p.p.m.)		Final Activity dis/(min)(ml)		Activity Removal %			
		Unfiltered	Filtered	Unfiltered	Filtered	Unfiltered	Filtered	Average	
								Unfiltered	Filtered
9	6.5	0.7	<0.5	4	0	92	100		
10	6.9	1.0	0.6	3	1	93	98	92.3	97.7
11	8.55	0.6	<0.5	4	2	92	95		

TABLE 8

Results from runs with mixed fission products

Activity of influent: Total Sr = 27 dis/(min)(ml)
Total β = 129 dis/(min)(ml)

Run No.	Final pH	Al^{+++} Concentration in overflow (p.p.m.)		Species	Final Activity dis/(min)(ml)		Activity Removal %			
		Unfiltered	Filtered		Unfiltered	Filtered	Unfiltered	Filtered	Average	
									Unfiltered	Filtered
12	6.6	0.45	< 0.1	Total Sr	13	13	52	52		
				Total β	34	34	76	76		
13	7.7	0.68	0.62	Total Sr	9	8	67	70	61	63
				Total β	44	44	66	66	72.7	75
14	9.5	1.3	1.08	Total Sr	10	9	63	67		
				Total β	28	19	78.4	85		

$Sr^{90} - Y^{90}$ Removals. Table 5 shows the results using a clarifier influent containing an equilibrium $Sr^{90} - Y^{90}$ mixture with an activity of 97 dis/(min)(ml). Due to the short counting time used, the Sr^{90} activities given are accurate to ± 25 per cent. Removals of Sr^{90} and Y^{90} are comparable, averaging about 50 per cent. These results may be compared with results by American workers (Straub, 1951) with 25.7 p.p.m. alum where Sr^{89} removal was 5.8 per cent. at a pH of 7.8 and Y^{91} removal was 93.3 per cent. at a pH of 7.1.

Cs^{137} Removal. Table 6 shows that the cesium removals are variable averaging 17.2 per cent. removal for the filtered overflow. Cesium removal can be expected to be poor for the reasons given in Section 2.3.4 and because cesium hydroxide and cesium carbonate are soluble.

Many papers (Morton and Straub, 1956; Seedhouse et al., 1958) refer to the difficulty of removing cesium ions from solution by coagulation.

$[U^{233} O_2]^{++}$ Removal. Table 7 shows that uranyl ions are very effectively removed from solution by this process, over the pH range 6.5 to 8.5.

Mixed Fission Products. For the mixed fission product runs shown in Table 8, an attempt has been made to differentiate between the total strontium (Sr^{89} and Sr^{90}) and the other β-emitters. Although the strontium activities are accurate to ± 50 per cent., removals are of the same order as those achieved in the earlier $Sr^{90} - Y^{90}$ runs.

Removal of all β-emitters averages 75 per cent., agreeing well with the results of Straub (1951), who achieved removals of 73 per cent. by settling and filtration after the coagulation of 15 p.p.m. aluminium sulphate, 14 p.p.m. lime, 7 p.p.m. sodium silicate, with 104 p.p.m. turbidity.

Ra^{226} Removal. At a pH of 7.6 and an influent activity of 23 dis/(min)(ml), the Ra^{226} activity of the filtered and unfiltered overflow after coagulation was 11 dis/(min)(ml), giving an activity removal of 52 per cent. Ra^{226} is chemically similar to strontium so similar removals would be expected for the two elements.

3.2.2 Discussion of results of decontamination experiments

Variation in pH has no significant effect on the removal of any of the isotopes used in this work. However, it is noticeable that as the pH is raised above 9.0, a high proportion of the aluminium hydroxide is stabilised and passes from the clarifier in the overflow. The analyses of the filtered samples of overflow show that the amount of sodium aluminate formed increases with a rise in pH. Since a new blanket was built up for each series of experiments, the activity of the stabilised particles was low, and therefore floc stabilisation did not reduce the decontamination efficiency. From these results and information in the literature (Kaufman et al., 1951) it seems advisable to make a pH value of 8.0 the maximum for this process.

Except for strontium, the activity removals in this process are comparable with those in the calcium/ferric phosphate process.

3.3 Thickening Experiments

Thickening experiments similar to those described in Section 2.4 were carried out with the aluminium hydroxide process. The maximum solids content of the sludge produced was about 4 per cent. This can be compared with the thickening in the Dorr clarifier where sludge of 7.7 per cent. solids was produced. Again this shows that the size and design of the clarifier control the thickening.

4. CONCLUSIONS

The calcium/ferric phosphate process for removal of radio–isotopes from solution can effect greater than 70 per cent. removal of uranyl and strontium ions and mixed fission products but cesium removal is poor. A pH value lower than 11.5 can be used without appreciably reducing radioactivity removal. Concentrations of dosing ions 40 p.p.m. PO_4^{---}, 30 p.p.m. Ca^{++}, and 20 p.p.m. Fe^{+++} in the effluent can be used without effectively reducing radioactivity removals. Complete precipitation of the dosing ions can be achieved at a final pH of 9.5 using concentrations 90 p.p.m. PO_4^{---}, 50 p.p.m. Ca^{++}, and 40 p.p.m. Fe^{+++} or proportional values.

Activity removals using the aluminium hydroxide process are comparable with those in the calcium/ferric phosphate process except for strontium. It is recommended that 8.0 be used as the maximum pH for the aluminium hydroxide process in the sludge blanket clarifier.

A series of trials with the Dorr clarifier should be made to verify the removals obtained with the model clarifier and to determine the volume of sludge produced per 10^6 gallons.

5. ACKNOWLEDGMENTS

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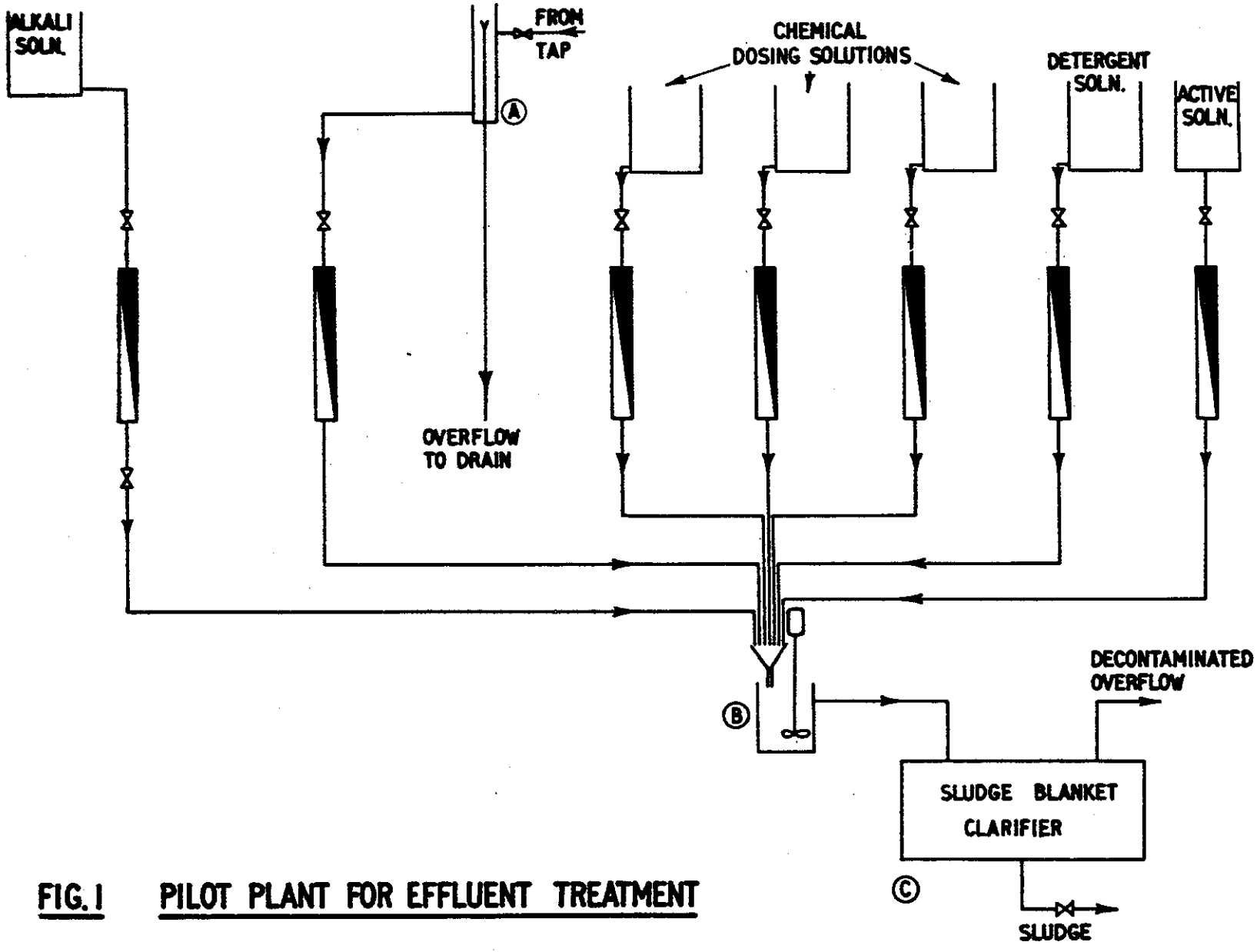

FIG. 1 PILOT PLANT FOR EFFLUENT TREATMENT

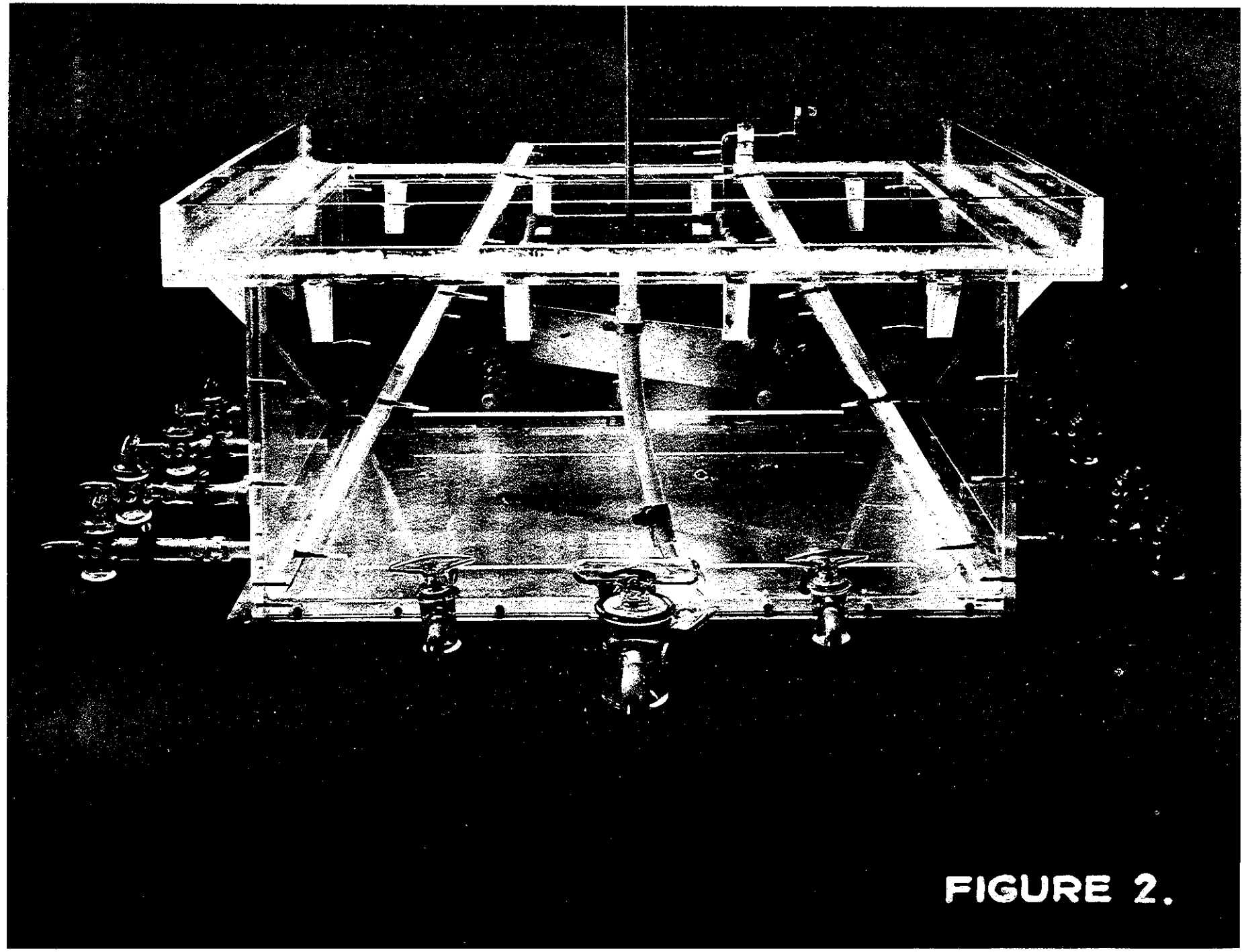

FIGURE 2. THE SQUARE SLUDGE BLANKET CLARIFIER

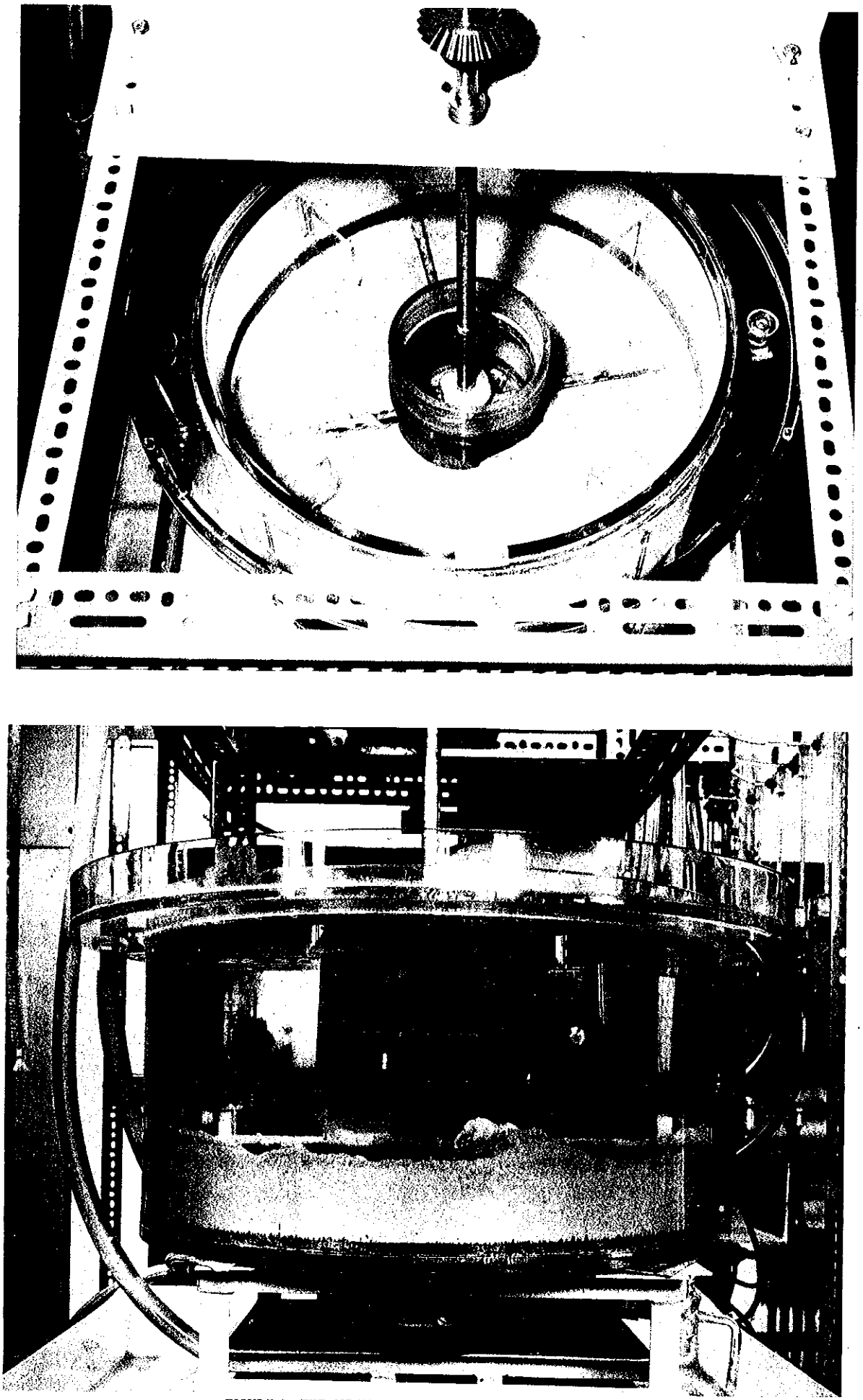

FIGURE 3. THE CIRCULAR SLUDGE BLANKET CLARIFIER