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The Effect of Radioactive Substances on Sludge Digestion

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CONTENTS

FIGURES

TABLES

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I. INTRODUCTION

1. Statement of the Problem

Two main sources of radioactive wastes have, in the past, been the production installations of the national atomic energy program and laboratories and hospitals using radioactive materials as research tools or for medical treatment. Radioactive wastes from national atomic energy installations have, in general, been treated and retained within the sites in a highly satisfactory manner.

Successful use of atomic energy is dependent upon the proper handling of radioactive wastes so that no harm results to man or other life. The possibilities of danger to sewage treatment plant personnel and to persons using contaminated sludge, or of harm to sewage treatment processes have become the concern of sanitary engineers and others involved in the problems of sewage treatment and in the prevention of stream pollution. The rapidly expanding use of radioactive materials, the growing list of users of isotopes, and the developing interest of industry in nuclear power contribute further concern in that the desire to achieve higher goals may lead to the improper or unintentional disposal of hazardous quantities of radioactive materials into sewage systems.

Although tentative standards of treatment and disposal have been established for radioactive wastes from hospitals and laboratories and specific recommendations have been made authoritatively in regard to long-term research and development programs, authoritative information is almost completely unavailable on the effects of radioactive materials on sewage treatment processes or on the degree of reconcentration of radioactive materials during such processes.

Information is needed on the questions of the effect of radioactive materials on sewage treatment processes and the possible reconcentration of radioactive materials during treatment processes in order to:

(1) Protect sewage treatment works personnel and the general public from radiation hazards.

(2) Develop, insofar as possible, means for the concentration and disposal of wastes, such means being consistent with the general principles recommended by the Atomic Energy Commission Advisory Committee which was appointed in June, 1948.

(3) Develop, through experimental evidence, suitable tolerances for various radioactive wastes.

(4) Determine the effects of radioactive wastes on standard industrial waste treatment processes and equipment, and the possibilities of utilizing such processes and equipment in the treatment of radioactive wastes.

(5) Obtain fundamental information on the chemistry and bacteriology of waste treatment and disposal, and the effects of radioactive materials on these aspects.

2. Sponsoring Agency

The research done in the Sanitary Engineering Laboratory of the University of Illinois was made possible through a contract between the United States Atomic Energy Commission and the University, which was entered into on April 1, 1952, and was terminated on May 31, 1954. Under the terms of Contract AEC-AT-(11-1)-218, the University was authorized to carry out the following work:

(1) Undertake the study of the effect of various concentrations and various combinations of radioactive materials on the anaerobic digestion of sewage sludges at various temperatures. Particular emphasis

was to be placed on studies of P^{32} and I^{131} in various concentrations and combinations at room temperature. The effects of these materials on digestion to be measured by quantitative observations of the recognized indicia of digestion such as: (1) rate, volume, and quality of gas production, (2) pH, (3) acidity, (4) volatile acids, (5) odor, (6) drying characteristics of sludge, and (7) characteristics of gas, digested sludge, supernatant liquor, dried sludge, and moisture obtained from drying a sludge.

(2) Include such other investigations in this general field as may be mutually agreed upon.

3. Related Projects

Studies relating to the various aspects of digestion of radioactive sludges furnished background materials for these tests. Studies such as those conducted by Babbitt, Leland, Whitley, and Schlenz [(1), (2)], provided reference on the mechanism of sludge digestion and properties of sludges to be tested.

An exchange of progress reports was made with other universities engaged in similar projects. Information and techniques obtained from the exchange were used in these studies on anaerobic digestion.

One of the primary objectives of the investigations at the University of Illinois was to determine the possibility of assimilation of radioactive materials by anaerobic organisms in a manner similar to that which has been demonstrated to be possible with aerobic processes by investigators at the University of California, the University of Texas, and New York University.

4. Methods of Research or General Procedures

In order to determine the effects of radioactive materials on the anaerobic digestion of sewage sludges and the degree of reconcentration attained during the digestion process, the following general methods of research were used.

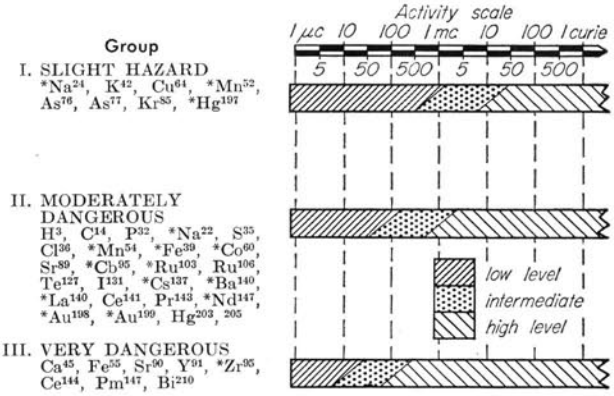
The effects of radioactive materials on digestion were first tested at varying concentrations by batch digestion experiments. These batch digestion experiments were utilized to establish the concentration of radioactivity as the controlled variable and to permit the use of high concentrations of radioactivity in relatively small volumes of sludge. The batch digestion method has the advantages of minimizing radiation hazard to research personnel and of reducing the cost of radioactive materials for high-level experiments.

Batch digestion experiments were effective only in determining the effects of radioactive materials on the anaerobic digestion of sludge since no effort was made to maintain or continue the digestion process by the addition of fresh sewage solids. There were no withdrawals of sludge or supernatant, nor were there any additions of fresh sewage solids during any particular batch digestion experiment.

The effects of radioactive materials on digestion and the degree of reconcentration of radioactivity during the digestion process were tested in specially constructed digestion apparatus which was built to approximate, as nearly as possible, conditions existing in standard digestion equipment.

Use of the specially constructed digesters provided the opportunity for control of such variables as digestion period, sludge withdrawal, solids content of feed, level of concentration of radioactivity, and temperature of digestion.

Throughout this report references will be made to levels of activity. In order to establish a basis for comparison of high and low levels of concentration, the following material is presented from Handbook 52 entitled "Maximum Permissible



Effective radiotoxicity is obtained from a weighting of the following factors:

Half-life
Energy and character of radiations
Degree of selective localization in the body
Rates of elimination
Quantities involved and modes of handling in typical experiments.

The slant boundaries between levels indicate border-line zones, and emphasize that there is no sharp transition between the levels and the associated protection techniques. The principal gamma-emitters are indicated by asterisk (e.g., $*Na^{24}$). The above level system does not apply to the hazards of external radiation.

Fig. 1. Selected Radioisotopes Grouped According to Relative Radiotoxicity

Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water," and from Handbook 42 entitled "Safe Handling of Radioisotopes," which were published by the National Bureau of Standards in 1953 and 1949, respectively.

Medium in Which Contained	Beta or Gamma Emitter (microcuries per ml.)	Alpha Emitter (microcuries per ml.)
Air	10^{-9} (a)	5×10^{-12} (b)
Water	10^{-7}	10^{-7} (c)

The values in the table are considered safe for any of the radioisotopes if (a) is reduced to 0.2×10^{-9} for Sr^{90}, if (b) is reduced to 2×10^{-12} for Pu^{239}, and (c) is reduced to 0.4×10^{-7} for Ra^{226}.

On the basis of the definition of levels presented in Figure 1, it will be apparent that the work done in the experiments in this project were mostly high level with some of the early experiments with P^{32} falling in the intermediate level.

The following laboratory tests were used in these investigations to aid in the control of digestion and some were intended as parameters thereof. In the case of those intended to be used as parameters the optimum values are listed.

(1) Gas Production — quality and quantity — volumes of gas generated were measured daily in all cases and in some cases twice daily. Quality of the gas produced was determined periodically.

(2) Solids, volatile and fixed — Analyses were made on samples from the digesters and on the sewage fed. Attempts were made to maintain the total solids in the continuously-stirred and -fed digesters at 50,000 to 70,000 ppm., and the volatile solids at 30,000 to 40,000 ppm.

(3) pH.

(4) Volatile Acids — Attempts were made to maintain volatile acids at values less than 2000 ppm. as acetic acid (as determined by the procedure outlined in the Ninth Edition of Standard Methods of Water and Sewage Analysis).

(5) Sludge characteristics.

(6) Odor

II. LABORATORY APPARATUS

5. General

The apparatus used in the entire study consisted of equipment essential for (1) storage and collection of sludge, (2) controlled digestion of sewage sludge by either the batch or the continuously-fed process, and (3) mechanical, chemical, radiological, and bacteriological analysis of sewage and sewage treatment processes.

6. Sources of Sewage and Sludge

Sewage for the digestion tests was obtained from various sources and at various times. In the majority of instances, seed sludge was obtained from the digester at the Urbana-Champaign Sanitary District Sewage Treatment Works. On two occasions when gas production was very low at the Urbana-Champaign treatment works, sludge was obtained from the Danville, Illinois sewage treatment plant. For the sake of convenience, stock digesters seeded with Urbana-Champaign sludge were maintained in the laboratory as a readily accessible source of digesting sludge for some of the experiments.

7. Equipment

(1) The Sanitary Engineering Laboratory at the University of Illinois has facilities for pumping sewage from a 30-inch outfall sewer, located within 200 feet of the laboratory, into two 10-foot diameter tanks within the building. These tanks have a capacity of approximately 7,000 gallons each and are arranged for continuous addition and removal of sewage from the outfall sewer. Raw sludge from settled, fresh sewage from these tanks provided the feed for all digestion tanks.

(2) All batch digestion experiments were conducted with essentially the same type of equipment. The digesters consisted of Erlenmeyer flasks, the size of which was dependent upon the volume of sludge being digested. A rubber tube led from the stopper in the digester flask to an eudiometer. In all cases, the eudiometer was constructed by inverting a brine-filled graduated cylinder in a brine-filled beaker. A diagram of the assembled digester and eudiometer is shown in Figure 2.

The temperature of the digesters was maintained at 95 F by installing them in a constant-95 F temperature water bath or by carrying out the tests in a constant-temperature room.

In some batch digestion experiments the rate of gas production was low, making it necessary to give consideration to the loss of carbon dioxide by diffusion through the saturated sodium chloride solution in the eudiometers. Consequently, tests were made using pure carbon dioxide generated by treating commercial sodium carbonate with hydrochloric acid to determine the loss of carbon dioxide by absorption and diffusion through saturated sodium chloride solutions. The results of these tests demonstrated that careful control of the apparatus and gas collection methods restricted the absorption and diffusion losses to an insignificant minimum.

(3) Initial digestion equipment consisted of two 5-gallon bottle digesters operated at room temperature. These two digesters did not function properly because of poor quality seed sludge and temperatures too low for proper digestion. Because

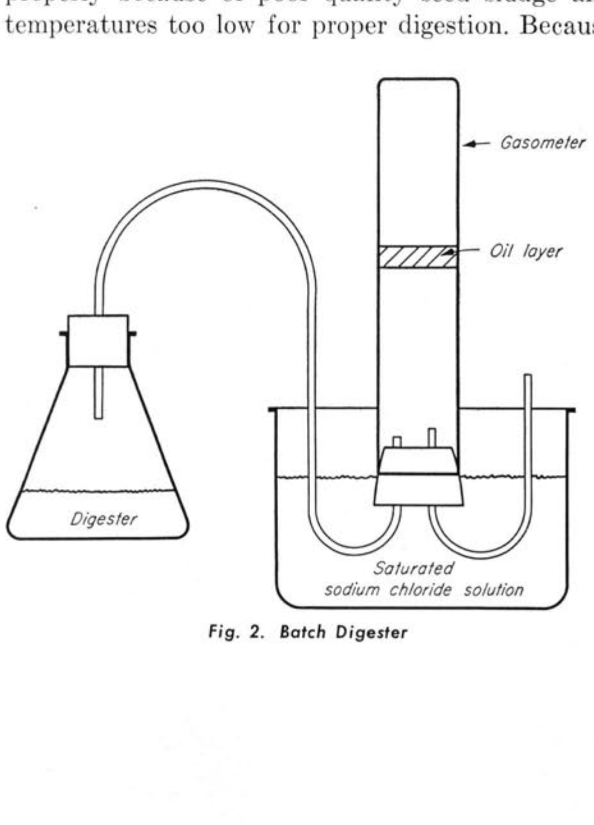


Fig. 2. Batch Digester

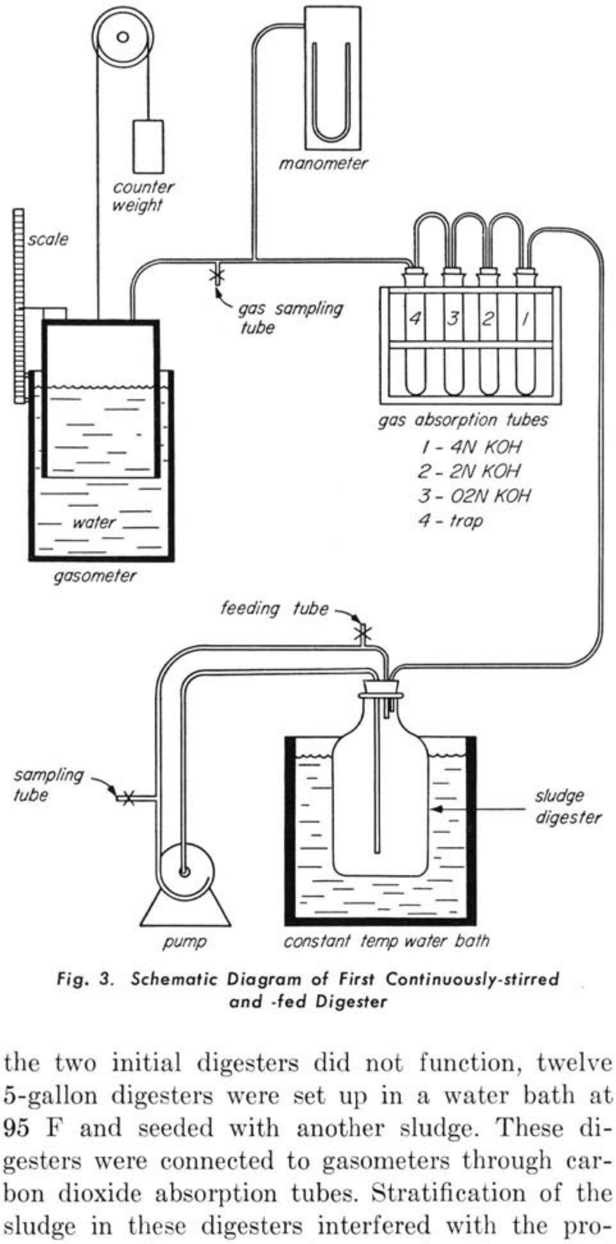


Fig. 3. Schematic Diagram of First Continuously-stirred and -fed Digester

the two initial digesters did not function, twelve 5-gallon digesters were set up in a water bath at 95 F and seeded with another sludge. These digesters were connected to gasometers through carbon dioxide absorption tubes. Stratification of the sludge in these digesters interfered with the procurement of representative samples for control analyses.

In order to attempt to determine the ability of the sludge digestion process to concentrate radioactivity, it was necessary to construct digestion equipment capable of maintaining a normal digestion rate and permitting a continuous feeding and sampling basis. It was desirable that samples taken from the digesters be as completely homogeneous as possible. The first digesters constructed to achieve these aims consisted of 5-gallon glass bottles mounted in a constant-temperature water bath as shown in Figure 3. Gas was collected in a brine-filled gasometer after passing through four carbon dioxide gas absorption tubes. The weight of the gas dome was counterbalanced by means of a counterweight and pulley arrangement. A small centrifugal pump, powered by a 1/30 hp electric motor, with a capacity of 4.5 gpm was installed on each digester as shown in the figure. An electrically controlled timer operated the circulating pumps for 1½ minutes out of each half-hour. The purpose of the circulating pumps was to provide a convenient means of sampling and feeding and to mix thoroughly the digester contents.

The method of mixing the digester contents by means of the centrifugal pumps proved unsatisfactory because of the rapid settling out and imperfect mixing of the heavier particles in the digesting sludge mixture. This imperfect mixing of the digester contents was found to be responsible for analytical errors greater than can be tolerated in work with radioactive tracers. The carbon dioxide gas absorbers proved ineffective in removing carbon dioxide from the gas produced and were therefore abandoned. Subsequent gas samples were analyzed on the Orsat apparatus for carbon dioxide and other constituents.

Difficulties with the type of digester shown in Figure 3 prompted the design of one in which constant temperature could be maintained without using a water bath, radiation hazard from exposed tubing would be minimized, and maintenance and shielding could be easily provided.

Six new digesters were constructed to conform to the requirements listed in the preceding paragraph. These digesters were circular, silicon-bronze containers 8 inches in diameter and 27 inches long, each having a capacity of approximately 5.8 gallons. A lucite draft tube and top assembly was bolted into the metal shell over a rubber gasket, making a tight seal. A stainless steel stirring shaft, having three propellers mounted inside the draft tube, protruded through a mercury and oil seal in the center at the top of the draft tube and top assembly. The stirring shafts of all six digesters were connected by bevel gears to a common horizontal drive shaft which was belt driven by a 1 hp electric motor at 400 rpm. A 1¼-inch sampling port extended through the top assembly and below the surface of the digester contents. A ¼-inch gas port entered the opposite side of the top assembly. Gas was collected through the gas port in a counter-balanced gasometer containing a saturated sodium chloride solution. Gasometer corrosion was inhibited by adding 1 ounce of potassium chromate

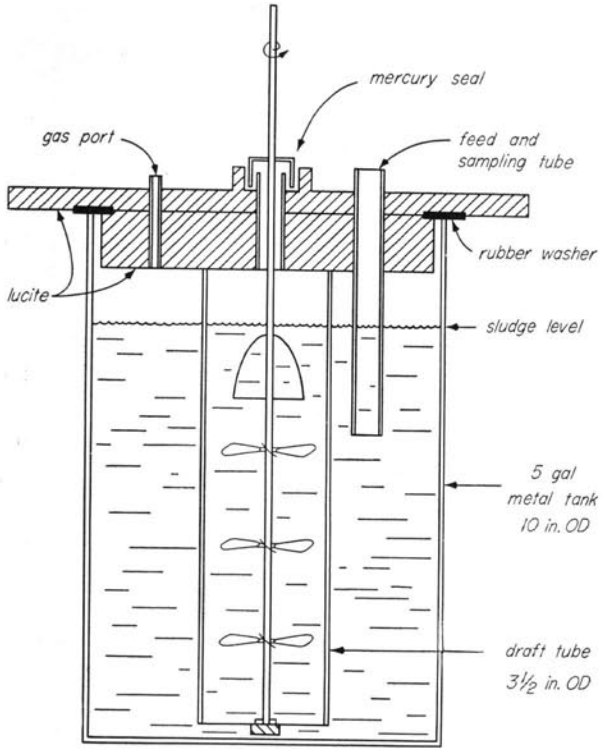


Fig. 4. Sectioned View of Ultimate Continuously-stirred and -fed Digester

per gallon of saturated sodium chloride solution. This digestion equipment was installed in a room on the second floor of the laboratory, in which the temperature was controlled to ± 1 F by a forced circulation steam heater with thermostatic control. The constant-temperature room was sufficiently isolated from other parts of the laboratory to decrease materially radiation hazard to laboratory personnel. Details of the digester assembly and arrangement are shown in Figures 4 and 5.

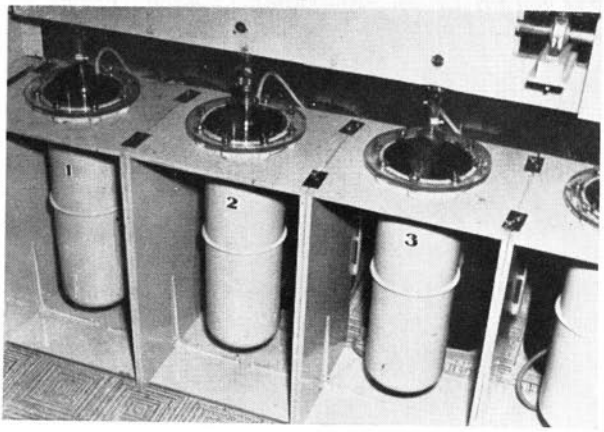


Fig. 5. Assembled Digester Equipment

III. INVESTIGATIONS CONDUCTED

8. Radioactive Materials and Their Characteristics

A list of the radioactive materials tested in these investigations and of their characteristics is shown in Table 1. Materials having intermediate half-lives were considered preferable for these tests. Intermediate half-lives permitted a reasonable test period without undue expense for radioactive materials and at the same time avoided the difficulties of disposal of radioactive sludge after testing. The choice of isotopes with intermediate half-lives was possible in all tests except those using fission products.

9. Preliminary Sludge Digestion Tests

Preliminary sludge digestion experiments were aimed at developing suitable equipment, adaptation of chemical control tests and radiological techniques to this investigation, and the study of effects of stable elements on anaerobic digestion.

(1) Development of Synthetic Sludges — Lack of uniformity of samples due to stratification and imperfect mixing caused analytical difficulties with the sewage sludge from the early 5-gallon bottle digester experiments. It was felt that the use of a synthetic sludge, the composition of which could be controlled sufficiently to regulate the digestion process, would avoid such analytical difficulties. Simplification of preparation of samples for counting radioactivity levels provided another reason for development of a synthetic sludge.

Numerous synthetic sludges were made, several of which had high rates of gas production. The synthetic sludges were tested in batch digesters and the gas production, at 95 F ±1 F, compared with that from sewage sludge and various mixtures of synthetic and sewage sludges. Comparison of the gas produced was made both quantitatively and qualitatively. On the basis of these batch experiments, gas production from the synthetic sludges was found to be from four to six times as great as from sewage sludge. The composition of the gas from synthetic sludge and from sewage sludge was about the same. Microscopic examination of the synthetic sludge revealed an abundance of large cocci and long slender curved rods. Because it was difficult to control the drop in pH caused by lactose fermentation in the synthetic sludges, the use of synthetic sludges was abandoned and new techniques were developed to prevent the stratification of sewage sludges.

(2) Effects of Stable Elements on Digestion — Prior to determining the effects of radioactive isotopes on digesting sludge it was necessary to determine the effects of the stable isotopes on sludge

Table 1
Characteristics of Radioactive Materials Used in Anaerobic Sewage Sludge Digestion Tests

Isotope	Concentration (mc./ml.)	Specific Activity	Chemical Form	Half Life	Type Radiation and Energy (mev)	Total Solids	Acidity or Basicity
P^{32}	>0.5 mc./ml.	Approx. 0.025 mg. P/mc. P^{32}	Phosphate in weak HCl.	14.3 days	β-1.712 γ-0	<5 mg./mc.	<0.5N HCl
I^{131}	5-20 mc./ml.	Carrier-free	NaI in basic Na_2SO_3 solut.	8 days	β-0.33 0.60 0.15 0.81 γ-0.720 0.364 0.284 0.080 0.16	<2 mg./mc.	0.005N to 0.05N NaOH
Ca^{45}	>0.01 mc./ml.	0.2 to 0.4 mc./gm. Ca	$CaCl_2$ in HCl solut.	152 days	β-0.254 γ-0		1N HCl
S^{35}	>1 mc./ml.	Carrier-free	Sulphate in weak HCl	87.1 days	β-0.166 γ-0	<10 mg./m ml.	
Fission Products (unseparated)	1-25 mc./ml. (estimated gross beta activity only)		Nitrates in HNO_3 solution			<10 mg./ml.	1-6N

digestion. Batch digestion experiments were used in these determinations of the effects of stable isotopes on anaerobic digestion. Stable iodine was added as sodium iodide and as potassium iodide. Sodium sulfite was also included with some of the potassium iodide experiments since I^{131} contains some

Table 2
Tabulation of Batch Digestion Tests for Effects of Stable Elements on the Anaerobic Sludge Digestion Process

Experiment Number	Digester Number	Element Tested	Concentration Used	Gas Production ml. hrs.	Remarks
1 (sewage sludge used)	1	KI	None	Not Measured	Microscopic examination revealed large numbers of long, slender curved rods, cocci in pairs or small groups, cocci in short chains, short thick rods, decreasing in numbers as concentration of KI increased.
	2	"	1% as I_2	" "	
	3	"	2% " "	" "	
	4	"	3% " "	" "	
	5	"	4% " "	" "	
	6	"	5% " "	" "	
2 (sewage sludge used)	1	KI	None	See Fig. 6	Microscopic examination indicated decreasing numbers of organisms with increasing concentrations of KI.
	2	"	0.5% as I_2		
	3	"	1.0% " "		
	4	"	1.5% " "		
	5	"	2.0% " "		
	6	"	2.5% " "		
3 (sewage sludge used)	1	KI	None	See Fig. 6	Na_2SO_3 added to digester No. 10.
	2	"	0.1% as I_2		
	3	"	0.2% " "		
	4	"	0.3% " "		
	5	"	0.5% " "		
	6	"	0.75% " "		
	7	"	1.0% " "		
	8	"	1.25% " "		
	9	"	1.5% " "		
	10	"	None		
4 (sewage sludge used)	1	NaI	None	See Fig. 6	
	2	"	0.2% as I_2		
	3	"	0.4% " "		
	4	"	0.6% " "		
	5	"	1.0% " "		
	6	"	2.5% " "		
	7	"	5.0% " "		
5 (sewage sludge used)	1	K_2HPO_4	None	See Fig. 7	See Table 4 for Results of Soluble and Total Phosphorus Determinations made at end of experiment.
	2	"	0.05%		
	3	"	0.1%		
	4	"	0.5%		
	5	"	1.0%		
	6	KH_2PO_4	1.5%		
	7	"	2.5%		
6 (sewage sludge used)	1	K_2HPO_4	None	See Fig. 7	See Table 5 for Results of Soluble and Total Phosphorus Determinations made at end of experiment.
	2	"	0.01%		
	3	"	0.03%		
	4	"	0.05%		
	5	"	0.07%		
7 (synthetic sludge used)	1	K_2HPO_4	None	See Fig. 7	
	2	"	0.01%		
	3	"	0.02%		
	4	"	0.03%		
	5	"	0.05%		
	6	"	0.07%		
8 (synthetic sludge used)	1	KI	None	See Fig. 8	
	2	"	1.0%		
	3	"	2.0%		
	4	"	3.0%		
	5	"	4.0%		
	6	"	5.0%		
9 (synthetic sludge used)	1	KI	None	See Fig. 8	Microscopic examination indicated predominance of large cocci in all concentrations with decreasing numbers of long, slender rods as concentration of KI increased.
	2	"	1.0%		
	3	"	2.0%		
	4	"	3.0%		
	5	"	4.0%		
	6	"	5.0%		
10 (sewage sludge used)	1	$CaCl_2$	None	See Fig. 9	
	2	"	"		
	3	"	"		
	4	"	0.05%		
	5	"	0.1%		
	6	"	0.15%		
	7	"	0.2%		
	8	"	0.4%		
	9	"	0.6%		
	10	"	1.0%		
11 (sewage sludge used)	1	$Na_2(SO_4)_3$	None	See Fig. 9	
	2	"	"		
	3	"	"		
	4	"	0.1%		
	5	"	0.21%		
	6	"	0.31%		
	7	"	0.42%		
	8	"	0.83%		
	9	"	1.25%		
	10	"	2.1%		
12 (sewage sludge used)	1	$Na_2(SO_4)_3$	None	See Fig. 9	
	2	"	"		
	3	"	"		
	4	"	0.005%		
	5	"	0.01%		
	6	"	0.05%		
	7	"	0.1%		
	8	"	0.5%		

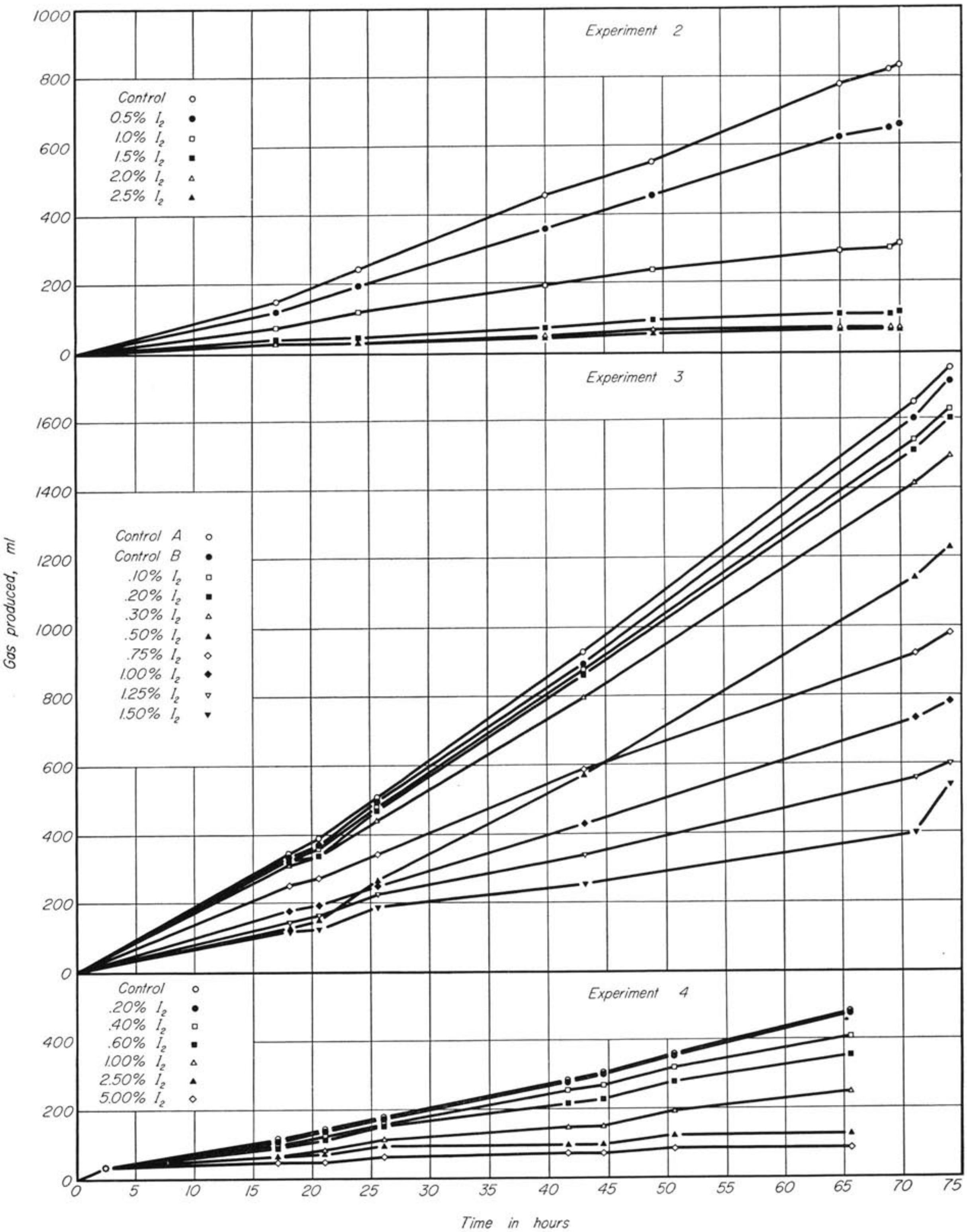


Fig. 6. Gas Production Curves for Batch Digestion Tests 2, 3, and 4

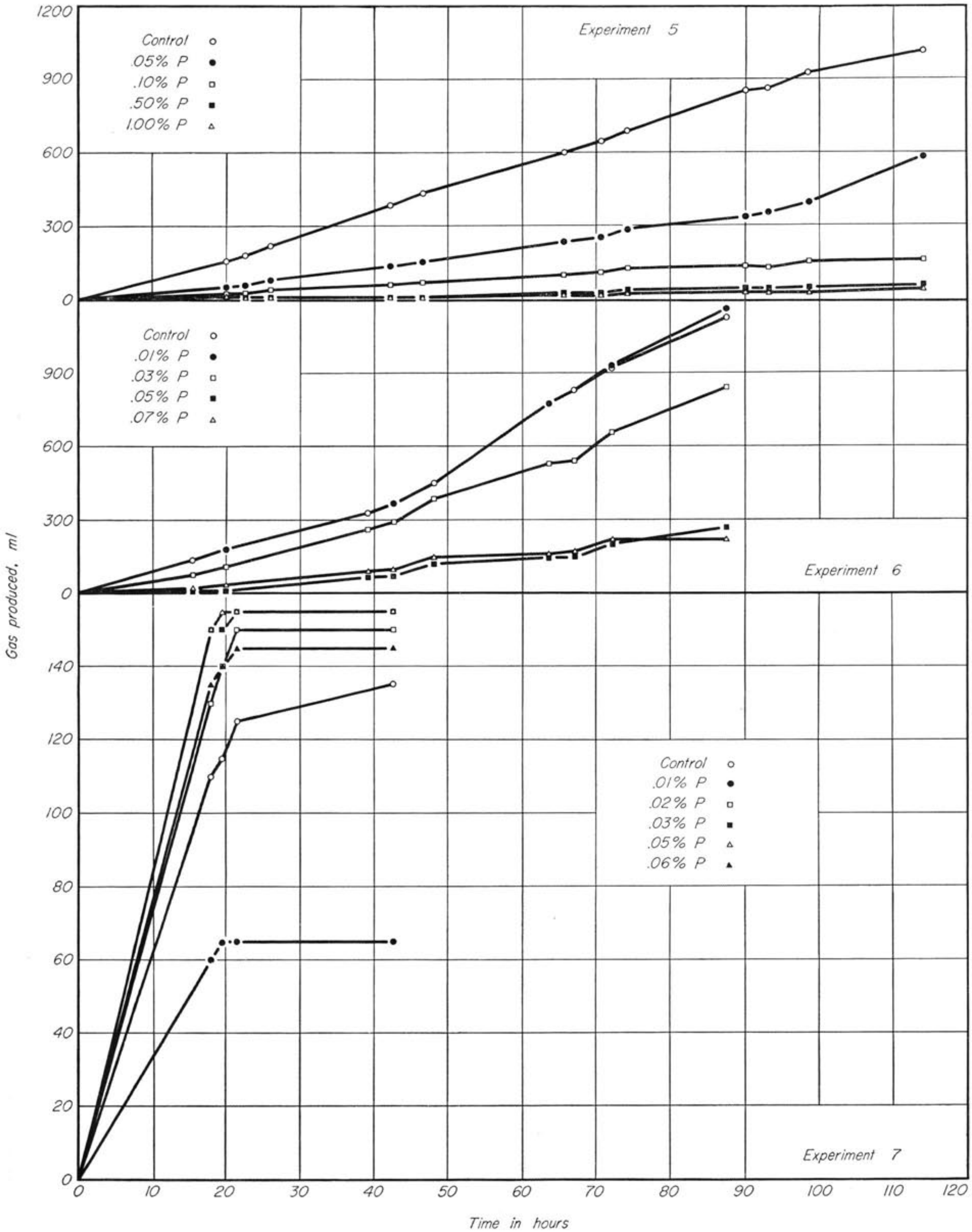


Fig. 7. Gas Production Curves for Batch Digestion Tests 5, 6, and 7

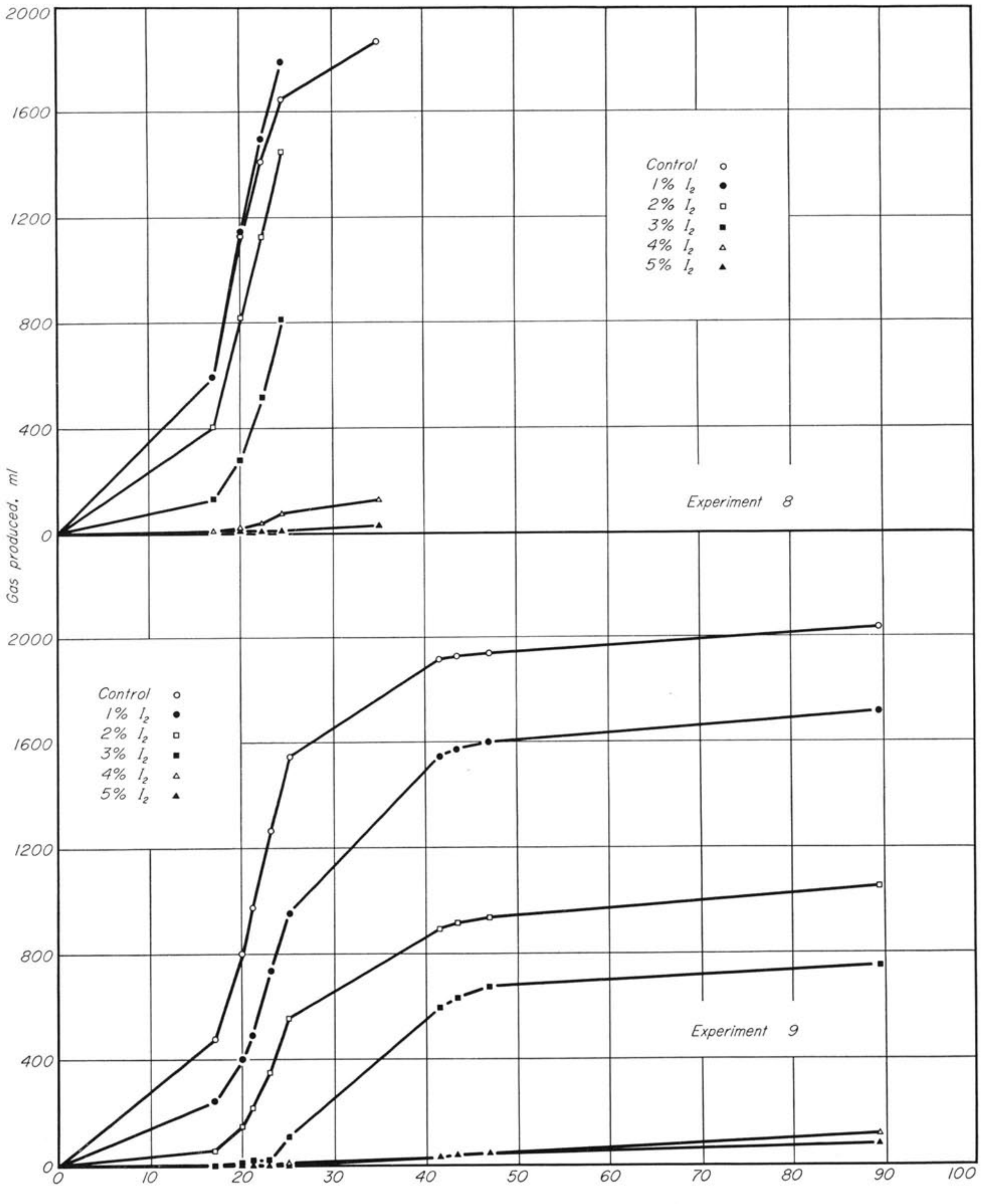


Fig. 8. Gas Production Curves for Batch Digestion Tests 8 and 9

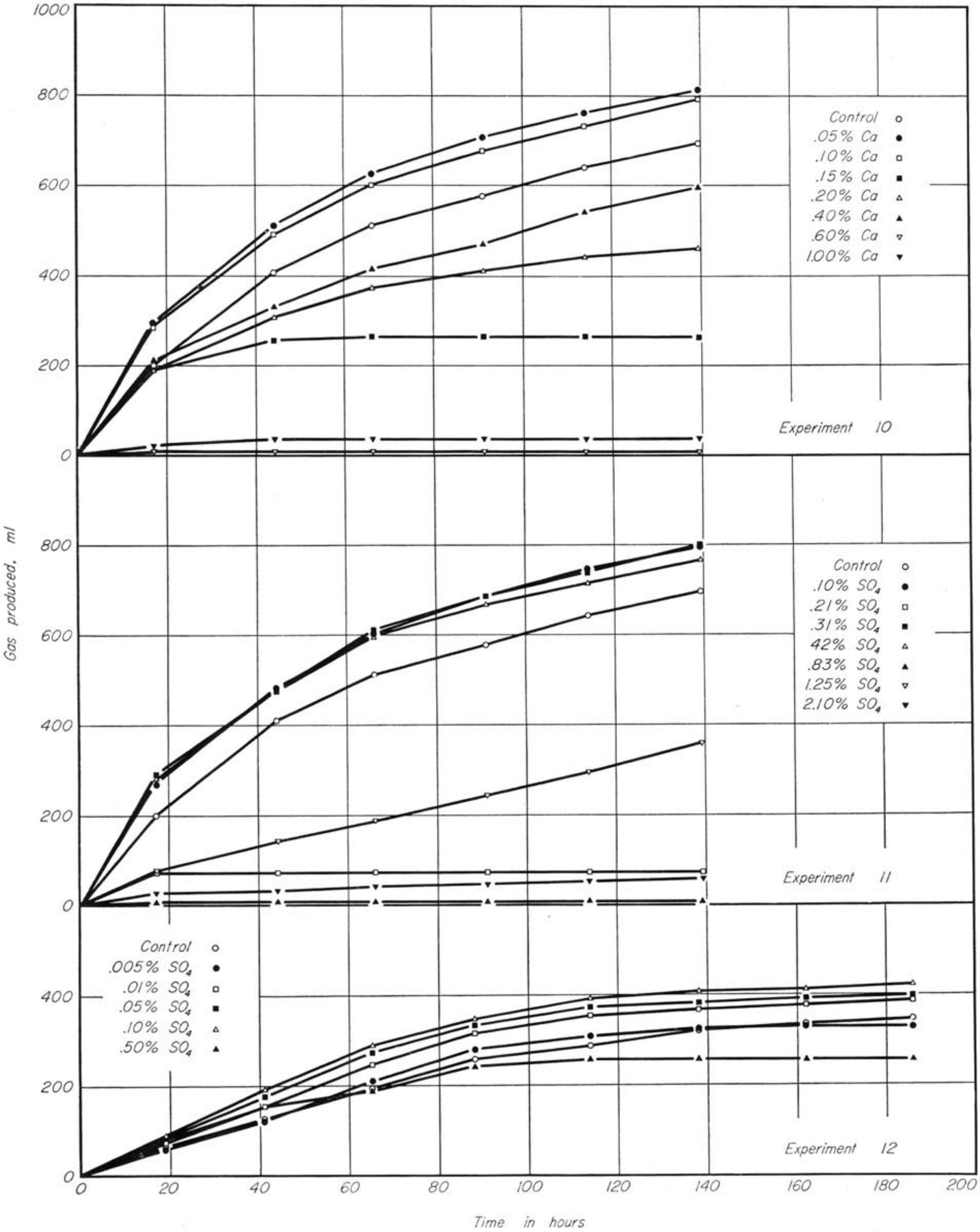


Fig. 9. Gas Production Curves for Batch Digestion Tests 10, 11, and 12

sodium sulfite as a carrier. Stable phosphorus was added as hydrogen potassium phosphate and as a mixture of hydrogen potassium phosphate and dihydrogen potassium phosphate. Stable calcium was added as calcium chloride and stable sulphur as sodium sulphate. The stable elements tested and the concentrations in which they were used are shown in Table 2.

10. Batch Digestion Tests of the Effects of Radioactive Materials on Digestion

The batch digestion experiment method which was used to determine the effects of stable elements was also chosen as the means of determining the effects of radioactive materials on digestion, particularly for high levels of radioactivity. The various batch digestion tests which were run with radioactive materials are shown in Table 3.

Table 3

Tabulation of Batch Digestion Tests for Effects of Radioactive Materials on the Anaerobic Sludge Digestion Process

Experiment Number	Digester Number	Radioactive Material	Concentration Used	Gas Production ml. hrs.	Remarks
13	1	P^{32}	None	See	
8/30/52	2	"	4 μc./l.	Fig.	
	3	"	20 μc./l.	10	
	4	"	40 μc./l.		
	5	"	210 μc./l.		
	6	"	420 μc./l.		
	7	"	None		
	8	"	None		
14	1	P^{32}	None	5850-384	1-liter digesters, each containing 800 ml. of digesting sludge.
11/13/52	2	"	None	5210- "	
	3	"	0.25 mc./l.	4857- "	
	4	"	0.25 mc./l.	5105- "	
	5	"	0.5 mc./l.	4885- "	
	6	"	0.5 mc./l.	3890- "	
	7	"	1.0 mc./l.	3425- "	
	8	"	1.0 mc./l.	4740- "	
15	1	P^{32}	None	150-500	No gas production in any of the digesters during the first 192 hours of the test. 1-liter digesters, each containing 200 ml. of digesting sludge.
2/2/53	2	"	None	600- "	
	3	"	4.0 mc./l.	475- "	
	4	"	4.0 mc./l.	625- "	
	5	"	7.0 mc./l.	165- "	
	6	"	7.0 mc./l.	650- "	
	7	"	12.0 mc./l.	530- "	
	8	"	16.0 mc./l.	650- "	
16	1	P^{32}	None	Avg. 950 -404	1-liter digesters, each containing 800 ml. of digesting sludge.
3/18/53	2	"	"	"	
	3	"	"	"	
	4	"	"	"	
	5	"	"	"	
	6	"	0.7 mc./ml.	"	
	7	"	"	"	
	8	"	"	"	
	9	"	"	"	
	10	"	"	"	
	11	"	"	Avg. 940 "	
	12	"	"	"	
	13	"	"	"	
	14	"	"	"	
	15	"	"	"	
	16	"	"	"	
	17	"	"	"	
	18	"	"	"	
	19	"	"	"	
	20	"	"	"	
17	1	P^{32}	None	See Fig. 10	125-ml. digesters, each containing 50 ml. of digesting sludge.
7/2/53	2	"	"		
	3	"	"		
	4	"	"		
	5	"	"		
	6	"	30 mc./l.		30 mc./l. level of activity maintained for 240 hours by daily additions of P^{32}.
	7	"	"		
	8	"	"		
	9	"	"		
	10	"	"		
18	1	P^{32}	None	365-183	125-ml. digesters, each containing 50 ml. of digesting sludge. Initial levels of activity maintained for 144 hours. See Note for explanation of Controls.*
7/14/53	2	"	60 mc./l.	430- "	
	3	"	"	440- "	
	4	"	100 mc./l.	420- "	
	5	"	"	420- "	
	6	"	100 Control*	410- "	
	7	"	" *	385- "	
	8	"	60 Control*	400- "	
	9	"	" *	375- "	
	10	"	None	400- "	
19	1	P^{32}	60 mc./l.	See Fig. 11	125-ml. digesters, each containing 50 ml. of digesting sludge. Initial levels of activity maintained for 120 hours. See Note for explanation of Control.*
7/28/53	2	"	"		
	3	"	100 mc./l.		
	4	"	"		
	5	"	60 Control*		
	6	"	" *		
	7	"	100 Control*		
	8	"	"		
	9	"	Nore		
	10	"	"		
	11	"	"		
20	1	P^{32}	200 mc./l.		125-ml. digesters, each containing 40 ml. of digesting sludge. Initial levels of activity maintained for 100 hours.
8/15/53	2	"	150 mc./l.		
	3	"	"		
	4	"	100 mc./l.		
	5	"	"		
	6	"	50 mc./l.	See Fig. 12	
	7	"	"		
	8	"	200 Control*		
	9	"	150 Control*		
	10	"	"		
	11	"	100 Control*		
	12	"	"		
	13	"	50 Control*		
	14	"	"		
	15	"	None		
	16	"	"		
	17	"	"		
21	1	I^{131}	200 mc./l.		
8/26/53	2	"	100 mc./l.		
	3	"	"		
	4	"	50 mc./l.		
	5	"	"		
	6	"	200 Control*	See Fig. 13	
	7	"	100 Control*		
	8	"	" *		
	9	"	50 Control*		
	10	"	"		
	11	"	None		
	12	"	"		
	13	"	"		
	14	"	"		
	15	"	"		
	16	"	"		
22	1	S^{35}	200 mc./l.		
2/16/54	2	"	100 mc./l.		
	3	"	"		
	4	"	50 mc./l.		
	5	"	"		
	6	"	200 Control*	See Fig. 14	
	7	"	100 Control*		
	8	"	" *		
	9	"	50 Control*		
	10	"	" *		
	11	"	None		
	12	"	"		
	13	"	"		
	14	"	"		
23	1	Fission	200 mc./l.		
4/24/54	2	Products	100 mc./l.		
	3	"	"		
	4	"	50 mc./l.		
	5	"	"		
	6	"	200 Control*	See Fig. 15	
	7	"	100 Control*		
	8	"	" *		
	9	"	50 Control*		
	10	"	" *		
	11	"	None		
	12	"	"		
	13	"	"		
	14	"	"		
	15	"	"		
24	1	Fission	200 mc./l.		* Nitric acid added to adjust pH to same value as that in spiked counterpart.
5/22/54	2	Products	100 mc./l.		
	3	"	"		
	4	"	50 mc./l.	See Fig. 16	
	5	"	"		
	6	"	200 Control*		
	7	"	100 Control*		
	8	"	" *		
	9	"	50 Control*		
	10	"	"		
	11	"	None		
	12	"	"		

* Controls contained no radioactive material but had distilled water added to them in amounts equivalent to the volume of radioactive material added to their counterparts.

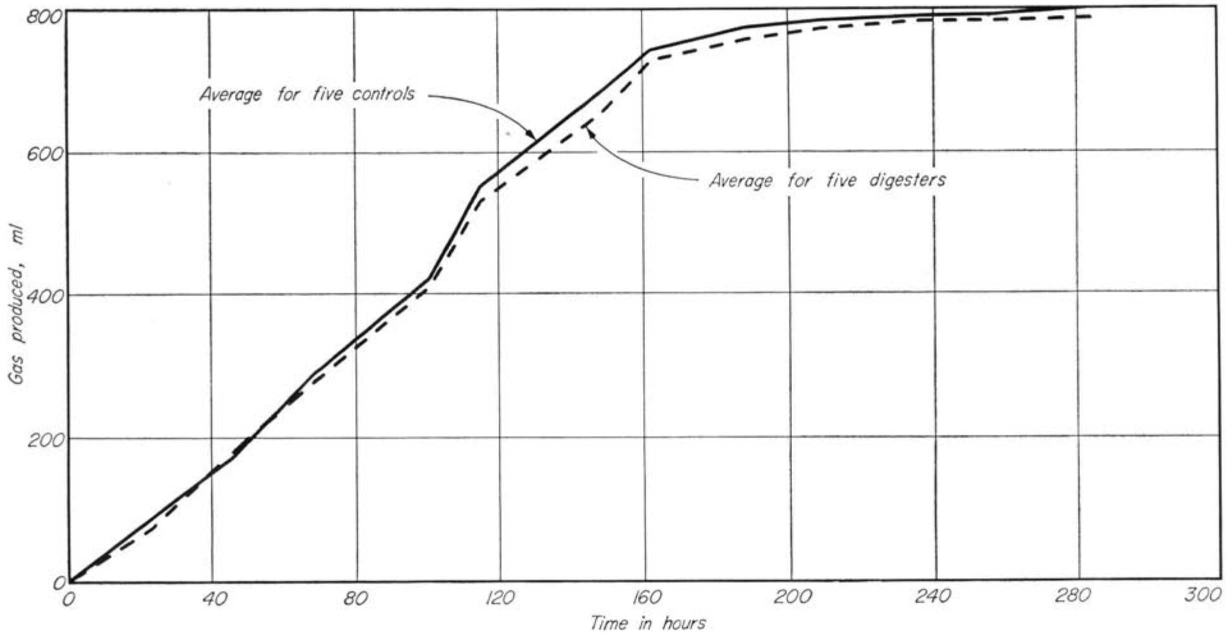


Fig. 10. Gas Production of Digesting Sludge With 30 Millicuries of Radioactive Phosphorus per Liter

11. Continuously-stirred and -fed Digestion Tests

At the same time that the batch digestion tests were being carried out, other tests on the effects of radioactive materials on digestion and on the degree of concentration of radioactivity during the digestion process were run in the six specially constructed stirred digesters. A description of the stirred digestion equipment is given in section II, 7, (3) of this report.

The stirred digesters were normally charged by adding weighed amounts, between 36 and 38 pounds, of a blended mixture of raw and digesting sludge. At the beginning of the tests with the stirred digesters, levels of activity as low as 10 microcuries per liter of wet sludge were used. Digesters 1, 2, and 3 were spiked with sufficient radioactive material to produce an activity level of approximately 110 microcuries per liter of wet sludge. The level of 110 microcuries per liter was standardized as being sufficiently high to produce significant results of the effects of radioactivity on sludge digestion and as a sufficiently high tracer level for chemical and radiological assay. This level was considered to be approximately 5500 times greater than that which a sewage treatment plant operator might encounter if the radioactive level in the plant digester

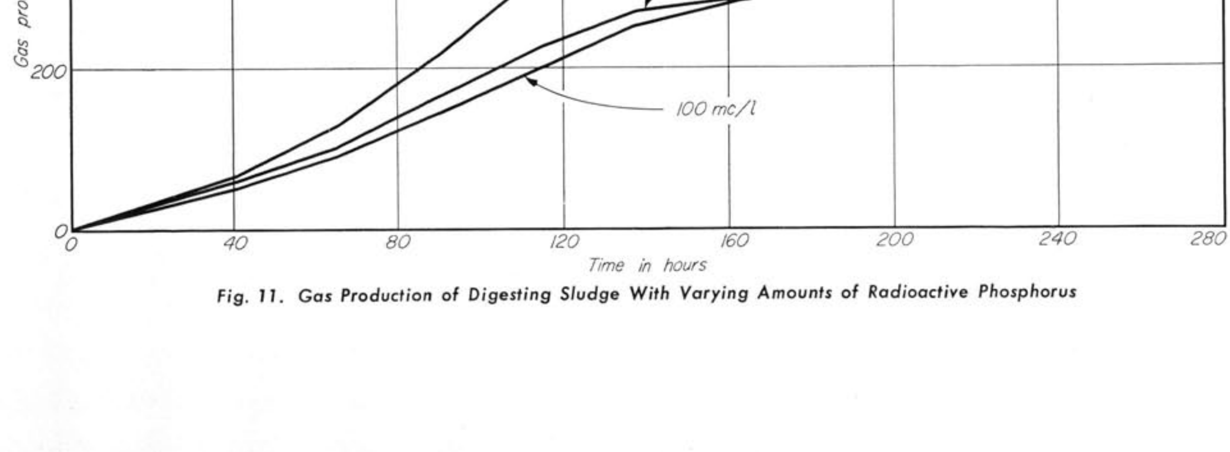


Fig. 11. Gas Production of Digesting Sludge With Varying Amounts of Radioactive Phosphorus

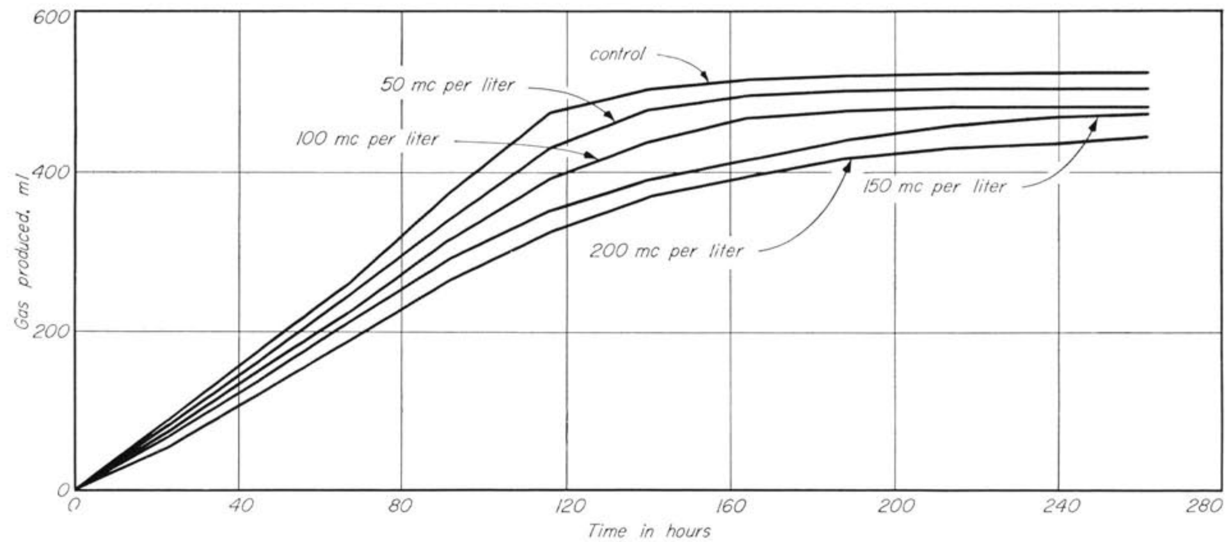


Fig. 12. Gas Production of Digesting Sludge With Varying Amounts of Radioactive Phosphorus

did not exceed the tolerance established in Handbook No. 52 of the National Bureau of Standards. Digesters 4, 5, and 6 received no radioactive materials and served as controls throughout each of the tests. The six stirred digesters were located in a constant-temperature room where the contents of the digesters could be maintained at temperatures near 95 F. During the summer months it was impossible to maintain the temperature of the digesters at 95 F and the control temperature was maintained at 102 F. In most cases the digesters were stirred continuously during any test period to avoid the possibility of foaming and spilling of the digester contents on the floor of the constant-temperature room. The daily amounts of sewage fed to and of samples taken from the digesters were arranged to provide a digestion period of approximately 30 days. During each test period regular observations were made of:

(1) volume of gas produced
(2) quality of gas produced
(3) weight of samples taken from the digesters each day
(4) weight of feed added to the digesters each day
(5) total solids in digester samples and in feed samples
(6) volatile solids in digester samples and in feed samples
(7) volatile acids in digester samples and in feed samples

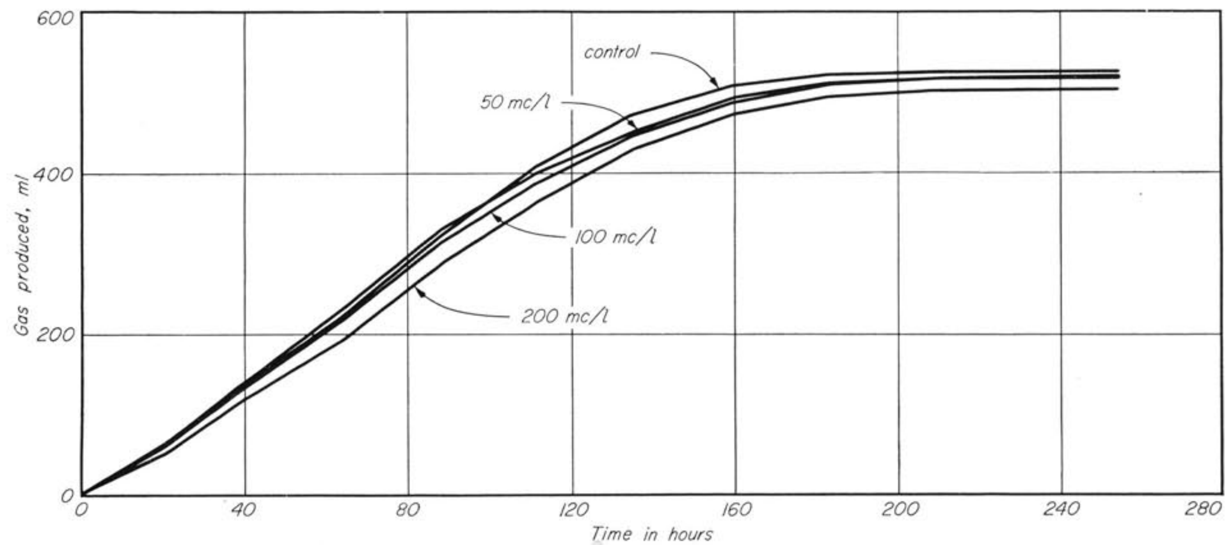


Fig. 13. Gas Production of Digesting Sludge With Varying Amounts of Radioactive Iodine

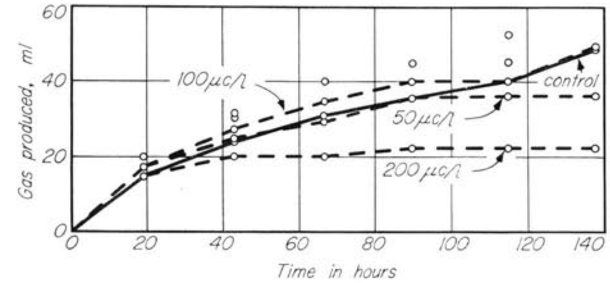


Fig. 14. Gas Production of Digesting Sludge With Varying Amounts of Radioactive Sulphur

(8) pH of digester samples

(9) temperatures in digesters

The level of radioactivity in the spiked digesters was maintained by periodic additions of radioactive materials to compensate for losses due to decay and sampling, and for the addition of fresh solids.

In addition to the observations enumerated above, routine measurements of radioactivity and chemical determinations were made for the specific element undergoing test.

Use was made of the various daily observations in maintaining proper digestion throughout each test. The feeding schedule was regulated by the observations for volatile acids, total solids, and volatile solids. In order to avoid a gradual decrease of the solids in the digesters it was necessary to feed a concentrated raw sludge. A 30-gallon metal drum was filled with fresh sludge every 24 hours. At the end of each 24-hour concentration period the floating solids were skimmed off. The solids concentrated in this manner were screened and mixed in a blender in preparation for feeding in the digesters.

12. Additional Methods of Treatment of Radioactive Sludges

(1) Filtration — Tests of the disposal of radioactive sludge on sand beds were conducted

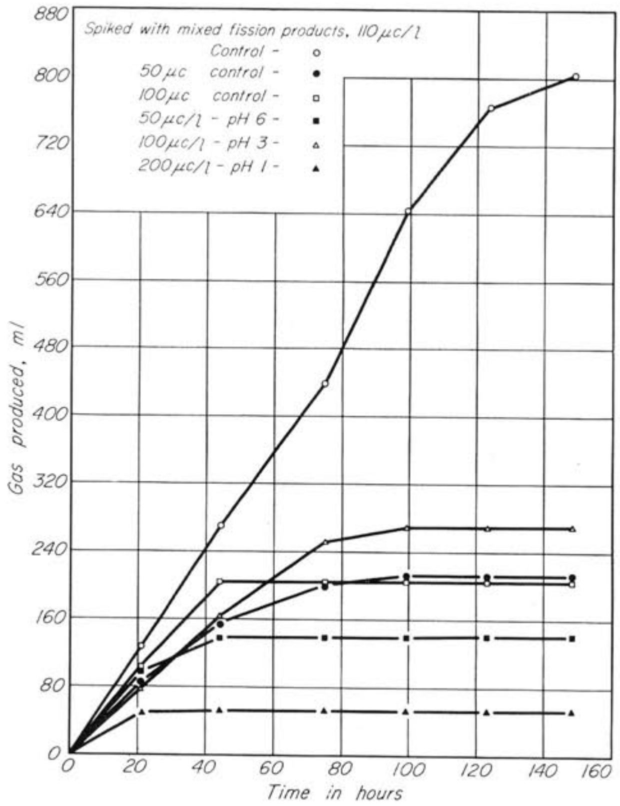


Fig. 16. Gas Production of Digesting Sludge With Varying Amounts of Mixed Fission Products

to determine the levels of radioactivity in the dried cake and in the filtrate.

Sand drying beds were simulated by six 2-inch diameter lucite cylinders each 6 inches in length, filled with standard Ottawa sand. Each lucite cylinder was filled with 300 grams of sand. This amount provided a sand depth of approximately 3 inches with enough freeboard above the sand to permit placing 100 ml. of sludge on top of the sand. Filtrate was collected in graduated cylinders placed underneath the filters.

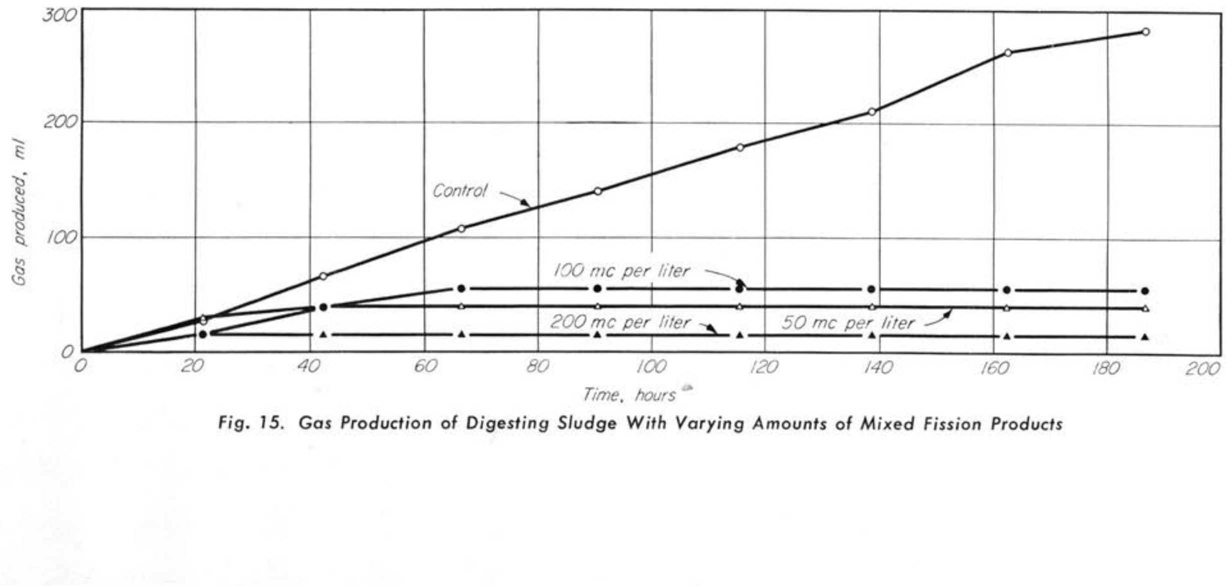


Fig. 15. Gas Production of Digesting Sludge With Varying Amounts of Mixed Fission Products

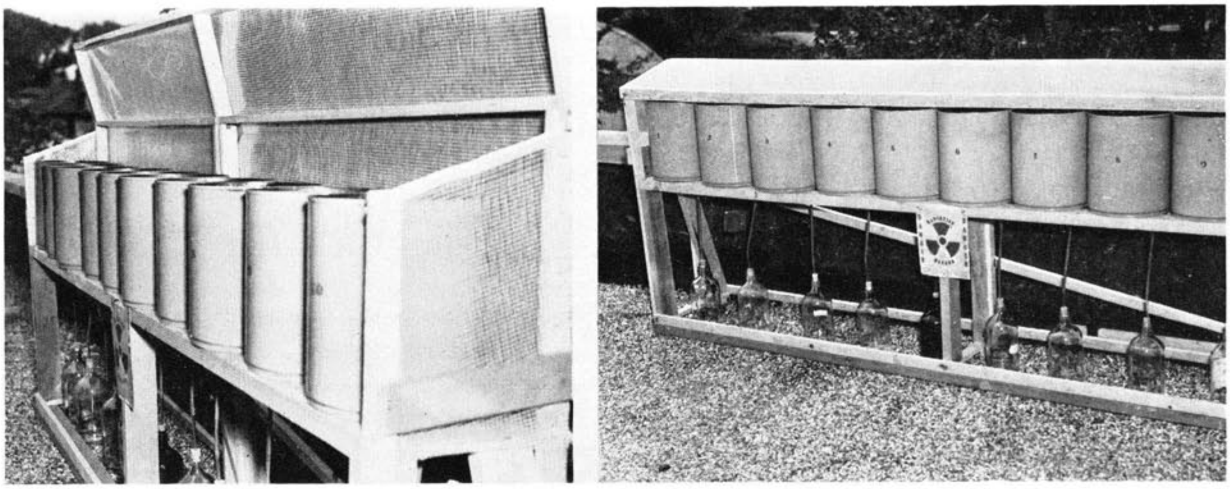


Fig. 17. Photographs of Filtration and Leaching Equipment

(2) Filtration and Leaching — Additional filtration tests were made to determine the effects of weathering and leaching on radioactive sludges placed on sand drying beds. Filters for these tests were constructed from 5-gallon metal drums as shown in Figures 17 and 18. The 10 filters were each loaded with 10 pounds of digested sludge, which was sufficient to cover each filter with 3 inches of sludge. The sludge on filter No. 1 contained no radioactivity and served as the control for the experiment. Filters No. 2 and No. 3 contained digesting sludge which was spiked with 110 microcuries of fission products per liter of wet sludge. Filters No. 4 and No. 5 contained digesting sludge spiked with 110 microcuries of P^{32} per liter of wet sludge. Filters No. 6 and No. 7 contained digesting sludge spiked with 110 microcuries of I^{131} per liter of wet sludge. Filters No. 8 and No. 9 contained sludge from the stirred digestion experiment with Ca^{45}, which was a month old and in which the level of activity had decreased by decay to approximately 96 microcuries per liter. Filter No. 10 contained sludge from the stirred digestion experiment with S^{35}, which was 2 months old and in which the level of activity had dropped to approximately 66 microcuries per liter.

Samples of the sludge placed on the 10 filters were counted for radioactivity at the beginning of the test and the total amount of radioactivity on each filter, expressed in counts per minute, was determined. Filtrates were collected periodically, measured volumetrically, and the total volume of filtrate from each filter evaporated to a volume less than 100 ml. Each concentrated filtrate was placed in a 100-ml. volumetric flask and brought to 100 ml. with distilled water. A 2-ml. portion of each concentrated solution was pipetted into a planchet, dried by evaporation, and counted in the scaler. From the counts obtained in this manner it was possible to determine the total activity, expressed as counts per minute, in each sample of filtrate. All counts were corrected for decay and expressed as a percentage of the original activity in the sludge on the filter.

The 10 sand filters were mounted on the first floor rooftop of the Sanitary Engineering Laboratory. A transparent plastic cover protected them from rainfall but allowed the drying effects of sunlight and wind to take place.

(3) Chemical Coagulation and Filtration — Raw and digesting radioactive sludges were treated

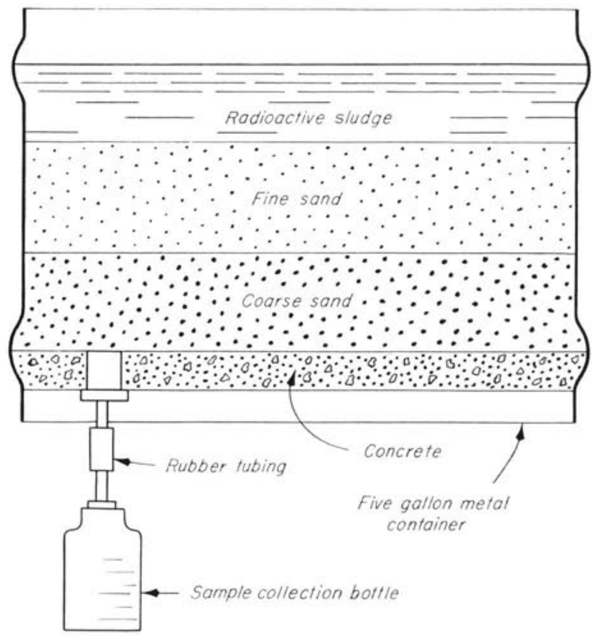


Fig. 18. Schematic Diagram of Filtration and Leaching Equipment

to determine the effects of chemical treatment on the partition of radioactive elements between the solid and liquid components of the sludge. The general procedure followed was to weigh out 200 grams of sludge into each of three 1-liter beakers. To beaker No. 1 were added 20 ml. of distilled water; to beaker No. 2, 20 ml. of 15% aluminum sulphate solution were added; and to beaker No. 3, 20 ml. of 1.5% ferric chloride solution were added. Immediately after the addition of chemicals, each beaker was stirred for 15 minutes at 50 rpm. The samples were removed from the stirrer and a 50-ml. portion of each filtered through semi-crepe filter paper into graduated cylinders. The volume of filtrate obtained from each 50-ml. portion was recorded after 5, 10, 15, 30, and 60 minutes. Simultaneously with the gravity filtrations, a 100-ml. portion of each sludge sample was poured onto a 4-inch Buchner funnel and suction filtered under a vacuum of 8 inches of mercury through No. 41 Whatman filter paper for 1 hour. Total and volatile solids determinations were made on all sludge filter cakes and on the untreated sludge samples to ascertain the degree of dewatering. A 2-ml. portion of each filtrate (gravity and vacuum) was pipetted onto a planchet, dried by evaporation, and counted for radioactivity in the scaler. Approximately 15 grams of each unfiltered sludge were diluted to 100-ml. with distilled water in a volumetric flask and thoroughly dispersed by shaking. A 2-ml. portion of this dispersed sludge was pipetted into a planchet, dried by evaporation, and counted in the scaler. The pH of chemically treated sludges was determined using a glass electrode pH meter.

From the data collected in these tests the counts per minute per 100 grams of sludge were determined. The activity of the filtrate was expressed as a percentage of the activity in the unfiltered sludge.

(4) Elutriation — Two experiments were made to determine whether radioactivity could be washed out of sludges. In the first test, 50 grams of radioactive raw sludge were weighed out into a beaker. This sludge was washed four times, each wash consisting of 100 ml. of distilled water. After each addition of distilled water the mixture was stirred vigorously and then allowed to settle for 10 minutes. After settling, the supernatant was decanted carefully and its volume measured. After the final wash the residue was diluted to 250 ml. with distilled water. Two-ml. portions of each of the four washes and of the diluted residue were pipetted into planchets, evaporated to dryness, and counted for radioactivity in the scaler. The counts obtained were corrected for background and calculated back to the total volumes of washwater and residue. The sum of the decanted washwater counts plus the residue count was considered as 100%. The percentage activity present in each wash was calculated on this basis.

In the second test, 50 grams of radioactive sludge were weighed out into a beaker as before. This sludge was washed seven times, with 20 minutes settling time between each wash. Radioactivity was measured and calculated in the same manner as in the first test. In addition, a 15-ml. portion of each wash was centrifuged for 30 minutes at 2500 rpm. in the 6-inch centrifuge. Counts on the subnatant of the centrifuged washwater samples gave an estimate of the amount of radioactivity carried over into the washwater by colloidal and suspended matter.

(5) Flotation — Two flotation experiments were made using raw sludge containing fission products and one was made with digesting sludge containing fission products. The experiment with digesting sludge was not satisfactory because the supernatant contained large amounts of suspended matter, making it extremely difficult to distinguish between the sludge and the subnatant. In these tests, 100 grams of radioactive sludge were weighed out into a beaker. The pH of the sludge was adjusted to 3.0 with 10% sulphuric acid. The beaker was covered with a watchglass and allowed to stand on the steam bath for 3 hours. The subnatant was pipetted out and its volume measured. Two-ml. portions of the subnatant and of the total sludge sample were pipetted into planchets, dried by evaporation, and counted in the scaler. The radioactivity in the subnatant was expressed as a percentage of the activity in the total sludge sample.

13. Chemical and Radiological Analytical Procedures

In attempting to find the effects of digestion on the concentration of radioactivity it was necessary to determine chemically and radiologically the distribution of the element being tested. During early tests with radioactive materials, sludge samples which were to be analyzed chemically and radiologically were separated into filtrate and precipitate by gravity filtration through filter paper. This method of obtaining the so-called solid and liquid components of sludge samples was unsatisfactory because consistent, reproducible results could not be obtained due to variations in such factors as the character of the sludge and humidity of the atmosphere. More consistent, reproducible results were obtained by separating samples into solid and liquid components by centrifuging 15-ml. portions for 1 hour at 2500 rpm. in the 6-inch diameter centrifuge. The liquid component, or supernatant found in the centrifuge tube, was removed by decantation. The solid component was removed by quantitative transfer after decantation of the liquid from the centrifuge tube. Chemical and radiological analyses of these two components were then compared with analyses of the unseparated or total sludge sample as a means of determining the distribution of chemical and radioactive constituents between the solid and liquid components.

In general, the analytical methods used were those outlined in the Ninth Edition of Standard Methods for Examination of Water and Sewage. Of necessity there were some variations and adaptations of other methods.

Results of determinations of stable and radioactive iodine concentrations in the various components of sludge were erratic using the Spector and Hamilton procedure. The loss of iodine due to sublimation during the drying process was subsequently confirmed. The loss of radioactive iodine was confirmed by comparing counts of samples which were run through the entire extraction procedure with counts of identical samples which were merely diluted to a known volume and dried for counting.

Various chemicals were employed in attempts to fix the iodine in the sample prior to initial drying and also prior to drying in the counting planchets. However, the major loss appeared to occur during the initial drying. Potassium carbonate was used to create an alkaline condition and sodium sulfite to attempt to reduce any free iodine to iodide. These two chemicals were used both separately and together. The maximum concentration used was 1 ml. of 1 molar solutions of each of the two chemicals. This maximum concentration was successful in achieving partial fixation of the iodine, however it was necessary to use lower concentrations since the maximum concentration interfered with the complete combustion of samples in the Parr apparatus.

Because of the erratic results obtained using the Spector and Hamilton iodine determination procedure, a mechanical separation technique was adopted for the determination of the distribution of radioactive iodine between the solid and liquid components of sludge samples. The mechanical separation technique was also used in studies with other radioactive materials.

IV. TEST RESULTS

14. Batch Digestion Tests Using Stable Isotopes

(1) Iodine — A total of six tests were made to determine the effects of iodine on the anaerobic digestion of sewage and synthetic sludges. These tests are shown in Table 2 as experiments No. 1, No. 2, No. 3, No. 4, No. 8, and No. 9. Experiments No. 1, No. 2, and No. 9 were primarily concerned with the effects of various concentrations of iodine, as potassium iodide, on the microscopic organisms found in digesting sludge. Microscopic examinations made in these tests indicated that the number of organisms in the sludge decreased as the concentration of potassium iodide increased. In general, the sludges with no potassium iodide contained large numbers of long slender curved rods, cocci in pairs or small groups, some cocci in short chains, and short thick rods. On the other hand, the sludges having high concentrations of potassium iodide usually contained some large and some small cocci, many short beaded rods, and a marked absence of long slender rods. Measurement of gas production in experiments No. 2 and No. 9 indicated a decided reduction in the amount of gas produced with an increase in the concentration of potassium iodide. Experiment No. 3 served to confirm the results of preceding experiments regarding the effects of the concentration of potassium iodide on digestion and also indicated that sodium sulfite, which is found in I^{131} solutions, exerted a slight inhibitory effect on gas production. In experiment No. 4 sodium iodide was used to determine whether the sodium salt had any greater effect than the potassium salt. The amounts of gas produced in these tests seem to indicate that iodine as potassium iodide or sodium iodide can be added in concentrations as high as 0.2%, expressed as I_2, without much inhibitory effect on sludge digestion.

(2) Phosphorus — Experiments No. 5, No. 6, and No. 7 were used to determine the effects of mono- and di-hydrogen potassium orthophosphate on anaerobic digestion. The results of these tests indicate that phosphorus, in concentrations greater than 0.03%, inhibits sludge digestion by 13% as measured by gas production. Microscopic examinations made of sludge samples from experiment No. 5 indicated that the number of organisms present decreased with a corresponding increase in concentration of phosphorus.

(3) Calcium — Experiment No. 10 was used to determine the effects of calcium chloride on anaerobic digestion. The results of this test indicated that more than 0.1% calcium had to be added to the sludge before the effects became apparent. Lower concentrations appear to have a stimulating effect, as measured by gas production, thereby verifying the findings of Hotchkiss [3] regarding the stimulative action of this salt in the growth of E. coli.

(4) Sulphur — Experiments No. 11 and No. 12 were made to determine the effects of sodium sulphate on anaerobic sludge digestion. The results of these experiments indicated that a concentration of 0.1% sulphate was the maximum which could be added to the sludge without inhibiting digestion, as measured by gas production.

15. Batch Digestion Experiments to Determine Effects of Radioactive Materials on Sludge Digestion

(1) Radioactive Phosphorus — The details of experiment No. 14 are given in Table 3. During the first 180 hours of the experiment the gas production of the radioactive sludge was nearly the same as that of the controls. The total volume of gas produced during the experiment varied about 10% between the radioactive and the control sludges, with no distinctive trend established between the two. About 99% of the radioactivity was found in the solid component of the radioactive sludge samples at the conclusion of this test.

The sludges used in experiment No. 15 produced very low amounts of gas during the first 190 hours. Increasing amounts of gas were produced during the period from 190 to 500 hours, but there was no conclusive correlation between the amounts of gas produced and the concentrations of radioactive phosphorus added to the sludges. It is believed that the erratic results were due to the selection of

Table 4

Tabulation of Bacterial Counts on Samples from I^{131} Batch Test

	1/14/54	1/18/54					1/22/54				
	Unstirred Digester	Control			I^{131}		Control			I^{131}	
		#1	#2	#3	#4	#5	#1	#2	#3	#4	#5
Gas Volume		105	115	100	165	0	125	140	120	215	40
Total Count	158	108	91	117	114	129	123	91	104	78	57
Per Cent Cocci											
Small	43.7	52.8	56.0	67.5	63.1	65.1	67.5	48.4	54.8	51.3	61.4
Large	5.1	(P-4)	1.1	0.8	2.6	1.6	....	1.1	1.9	1.3	....
Per Cent Micrococci											
Small	10.1	7.4	7.7	6.0	6.1	4.7	11.4	11.0	5.8	12.8	10.5
Large	0.6	....	....	(2)	1.7	(1)	....	....	....	....	....
Per Cent Streptococci	....	....	....	....	....	....	....	....	....	....	....
Per Cent Sarcina and Staphylococcus	(P-8)	....	(1)	....	1.7	(4)	1.6	1.1	1.0	(1)	(1)
Per Cent Rods											
Long	....	....	....	....	....	....	....	....	....	....	....
Heavy Long	....	....	....	....	....	....	....	....	....	....	....
Short	25.9	35.2	26.4	12.8	22.8	18.6	13.0	31.9	28.8	24.4	26.3
Heavy Short	2.5	0.9	1.1	0.8	(18)	0.8	0.8	(1)	1.0	....	(1)
Per Cent Diplobacilli	3.8	0.9	3.3	0.8	(2)	....	0.8	1.1*	1.0	2.6	....
Heavy	(P-3)	0.9	1.1	(3)	0.9	....	0.8	....	....	....	....
Per Cent Streptobacilli	3.2	....	....	....	....	....	....	....	....	....	....
Per Cent Curved Rods											
Long	0.6	....	....	....	....	....	....	(P)	....	....	....
Heavy Long	....	....	....	....	....	....	....	....	....	....	....
Short	2.5	1.8	4.4	2.6	0.9	7.6	1.6	3.3	3.8	6.4	1.8
Heavy Short	(P-6)	....	....	....	....	....	0.8	....	1.0	....	....
Per Cent Vibrio	1.9	....	....	....	....	2.3	0.8	....	1.0	1.3	(1)
Per Cent Neisseria	....	....	....	....	....	....	0.8	2.2	(2)	(1)	1.0

* Curved Diplobacilli
(No.) — Not counted in total; present in field
(P) — Present on slide but not in fields in which count was made

a poorly digesting sludge with which to start this experiment. No conclusions were drawn from this experiment. Experiment No. 15 was not repeated since it was felt that subsequent experiments served to cover the scope intended. A larger number of digesters was used in experiment No. 16 in the hope of obtaining conclusive results. Results in this test indicated that effects, as measured by gas production, were insignificant at a level of activity of 0.7 millicurie per liter.

Experiment No. 17 contained digesters which were spiked to a level of 30 millicuries per liter and maintained at that level for a period of 240 hours by daily additions of P^{32}. Distilled water was added daily to the control flasks to simulate the dilution of the radioactive flasks by addition of radioactive phosphorus. The radioactive flasks produced 1% less gas than did the control flasks.

Levels of activity of 60 millicuries per liter and 100 millicuries per liter were reached in experiment No. 18. Gas production during the early part of the experiment was extremely low and was interpreted as indicating that a poorly digesting sludge had been used.

Experiment No. 19 was started because of the difficulties with digestion experienced in the preceding experiment. Results of this experiment indicated that digestion, as measured by gas production was inhibited by about 33% for both the 60 and 100 millicurie per liter levels of activity. The slope of the gas production curve, shown in Figure 11 during the first 100 hours of the experiment indicates that the sludge used in this test was digesting poorly when it was chosen for the experiment. The results seem to indicate that the inhibiting effects of radioactive materials are more pronounced in poorly digesting sludges.

In experiment No. 20 the maximum level of activity was increased to 200 millicuries per liter. This maximum level of activity appeared to inhibit gas production by about 13%.

(2) Radioactive Iodine — Experiment No. 21 tested the effects of radioactive iodine on sludge digestion at levels of activity of 50 millicuries per liter, 100 millicuries per liter, and 200 millicuries per liter. From the curves shown in Figure 13 it is apparent that the digesters containing I^{131} produced less gas than did the controls, although inhibition was not so marked as in the case of the P^{32} batch digestion experiments.

Microscopic examinations were made of samples from a batch digestion test with radioactive iodine which is not listed in Table 4. In this test, two digesters were spiked to produce a level of 190 millicuries of I^{131} per liter of wet sludge in each. Three additional digesters which were not spiked served as controls. The five digesters were maintained at 95 ±1 F for 185 hours. Samples for microscopic examination were taken at the beginning of the test, after 88 hours, and after 185 hours. The results of these microscopic observations are shown in Table 4.

The over-all appearance of the slides examined indicated a lack of bacteria in the samples collected January 18 and January 22. Great difficulty was experienced in finding areas with enough cells to count in all samples. The greatest difficulty was with the spiked sample taken January 22.

In general, the same organisms found in the control samples were also found in the spiked samples. In addition, the organisms observed were similar to those found in samples taken from the stirred digestion apparatus. Small cocci and short rods were most prevalent.

Heavy short rods were present in profusion in the spiked sample No. 4 of January 18, although they were not found in the portion of the fields counted. The same organisms were lacking in the sample from the same source on January 22. Incidentally, this digester produced the largest volume of gas during the test.

Streptobacilli and long curved rods, present initially in the sludge, disappeared. Heavy short rods and Vibrio, present in the original sludge, but absent in the first test sample, reappeared at the end of the test. Neisseria made its first appearance in the last samples collected. The only time at which curved diplobacilli were observed in this test was in the observations on samples taken January 22.

A reversal in counts was noted in the spiked samples No. 4, and No. 5 for January 18 and January 22, especially in both thin and heavy short rods, short and thin curved rods, and in vibrio. The gas production from these samples showed a remarkable difference, although the types of organisms present differed only in the small cocci and thin diplobacilli.

Long rods, of the type resembling Barker's[4] methane producers, were entirely lacking in these batch digestion tests. Long curved rods were found only in the sludge with which this batch digestion test was started and in the No. 2 control sample of January 22. These long curved rods had previously been found in only two samples from batch digestion tests. Sarcina was generally present in small numbers which increased with time.

(3) Radioactive Sulphur — The effects of radioactive sulphur were tested in experiment No. 22. The results of this test indicated that the presence of S^{35} in concentrations up to 200 millicuries per liter inhibit gas production by as much as 55%.

(4) Mixed Fission Products — Experiments No. 23 and No. 24 were made to test the effects of fission products on digestion. The results of experiment No. 23 were considered to be inconclusive due to the possibilities of low pH in the digesters caused by the high acidity of the fission product solution added. Experiment No. 24 was made as a check on the possibility of interference by the acid solution. Fission products were added in the concentrations shown in Table 3. In addition, nitric acid was added to some of the controls, as indicated in Table 3, to adjust the pH of the controls to the same values as those of the spiked digesters. The results of this test indicated that inhibition of digestion was caused in part by the radioactive material and in part by the nitric acid present in the radioactive solution added to the digesters.

16. Experiments in Stirred Digestion Apparatus to Determine the Effects of Radioactive Materials on Digestion and to Determine the Degree of Concentration of Radioactivity During the Digestion Process

(1) Radioactive Phosphorus —

(a) 1 microcurie per liter level of activity — Experiments with radioactive materials using the six stirred digesters were started on March 18, 1953, at which time two of the six digesters were spiked with radioactive phosphorus to a level of approximately 1 microcurie per liter. This experiment was abandoned at the end of one week when operational difficulties caused the overflow of sludge from the digesters.

(b) 11 microcuries per liter of activity — On April 29, 1953, the stirred digesters were started again. Three digesters were spiked with sufficient radioactive phosphorus to give a level of activity of approximately 11 microcuries per liter in each, and maintained at that level by daily additions of P^{32} until May 31, 1953. This experiment gave no indications of inhibition of digestion due to the presence of radioactive phosphorus, and appeared to indicate concentration of the activity in the solid component of samples from the digesters.

(c) 22 microcuries per liter level of activity — An experiment started on June 15, 1953 at a level of activity of 22 microcuries per liter was inconclusive. It was not repeated since previous and subsequent experiments provided data on the effects of higher and lower levels of radioactivity.

(d) 110 microcuries per liter level of activity — For the test period of July 16, to September 30, 1953, the digesters were charged with digesting sludge and a level of activity of 110 microcuries per liter of wet sludge was maintained in digesters No. 1, No. 2, and No. 3 from July 16, until September 4. The level then decreased by decay until September 23, at which time it was again raised to 110 microcuries per liter and maintained until the end of the test.

The results of determinations for radioactivity and for phosphate shown in Figure 19 are representative of the results obtained in the three spiked digesters during this test period. The figure shows that the amount of radioactivity in the solid component of sludge samples analyzed was greater than the amount of radioactivity in the liquid component. This was despite the fact that the intensity of radioactivity in the solid component, expressed as counts per minute per milligram of phosphate, was less than the intensity of radioactivity in the liquid component. Determinations made on samples from these tests indicated the amount of radioactivity in the solid component to be approximately three to four times greater than that in the liquid component.

(2) Radioactive Iodine — Radioactive iodine was tested in the stirred digesters from November 25, 1953 until January 14, 1954. The level of activity in digesters 1, 2, and 3 was started at 110 microcuries of I^{131} per liter of wet sludge on November 25 and maintained at that level until December 14. From December 14 until December 24 the level decreased by decay. On December 24 it was raised to 110 microcuries per liter and held at that level until January 14. Periodic observations were made for:

a. total and volatile solids of sludge samples
b. volatile acids
c. volume and quality of gas produced
d. distribution of iodine between the solid and liquid components
e. radioactive counts

Attempts to determine the distribution of iodine were unproductive, due to losses of iodine by sublimation, and were finally discontinued in the interests of economy. Distribution of radioactivity between the solid and liquid components of sludge samples analyzed was determined by a mechanical separation method. These determinations of distribution indicated that 65 to 85% of the radioactivity in samples from this experiment was found in the liquid component of the samples. These percentage distributions were found in samples containing levels of activity from 33 to 110 microcuries per liter.

(3) Radioactive Sulphur — The six stirred digesters were charged with digesting sludge on January 19, 1954, in preparation for tests with S^{35}. Sufficient volumes of S^{35} were added to digesters 1, 2, and 3 on February 14 to produce a level of activity of 110 microcuries per liter in each. The 110-microcurie-per-liter level was maintained until March 4 by periodic additions of S^{35}. The usual periodic observations for solids, volatile acids, etc.

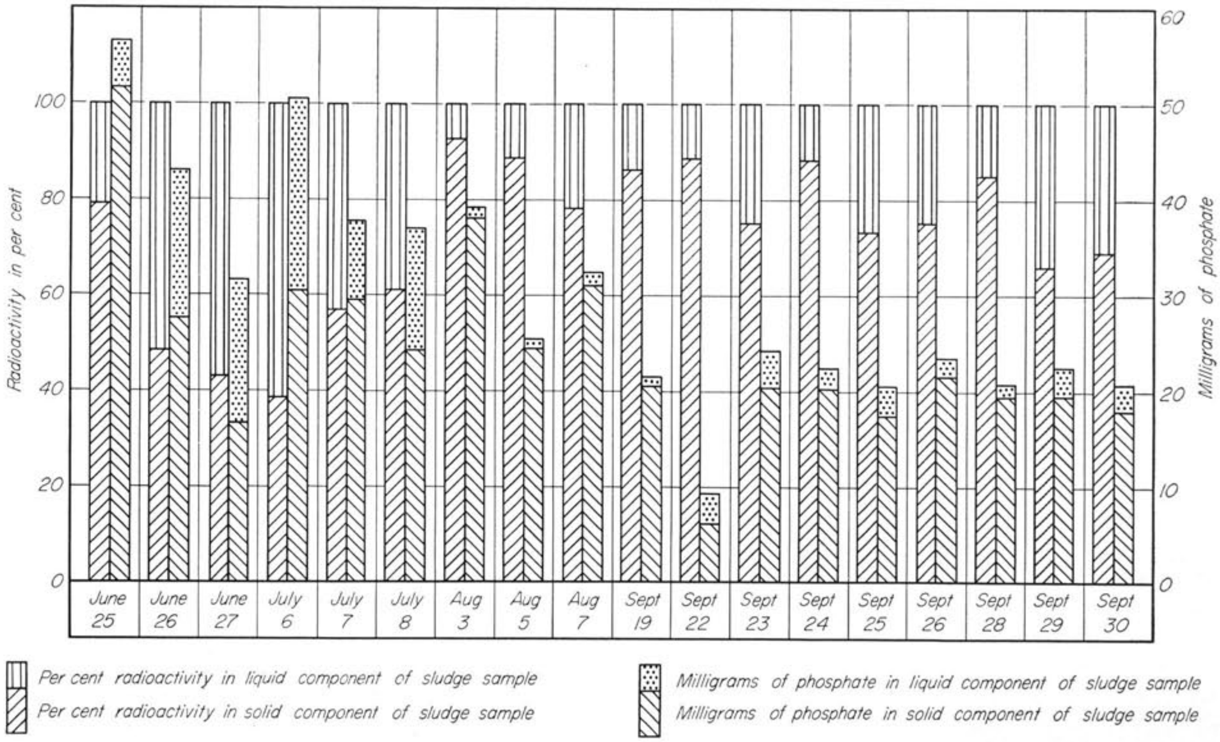


Fig. 19. Distribution of Radioactivity and of Phosphate in Digested Sludge

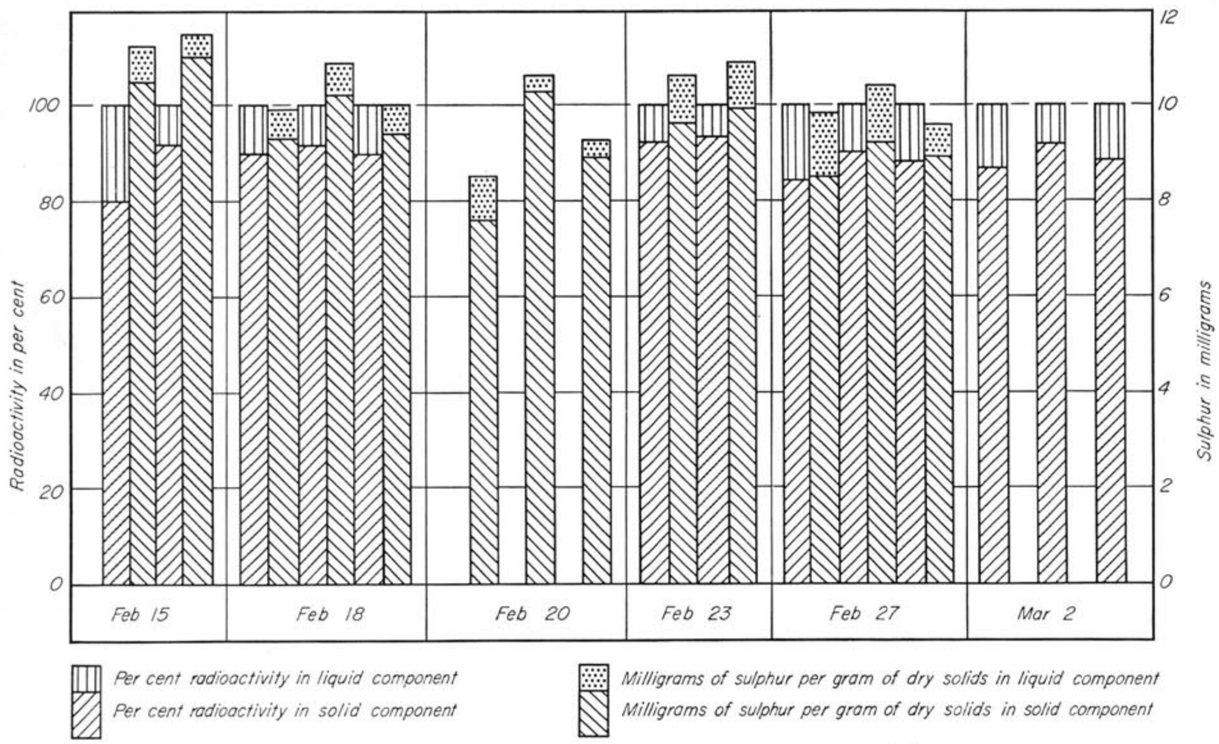


Fig. 20. Distribution of Radioactivity and of Sulphur in Digested Sludge

were made during this test. The distribution of radioactivity and of sulphur was determined on samples taken from digesters 1, 2, and 3. The results of these determinations are shown in Figure 20. Approximately 89% of the radioactive sulphur was found in the solid component of the samples analyzed during this experiment. Radioactive sulphur, like the radioisotopes used in previous tests, did not follow the laws of isotopic dilution in this experiment. In the samples analyzed it was found that the solid component contained from 6 to 14 times as much radioactivity as did the liquid component, but that in some cases the solid component contained as much as 34 times the sulphur as did the liquid component.

(4) Fission Products — On April 16, 1954, the stirred digesters were started with a new charge of digesting sludge. Digesters No. 1, No. 2, and No. 3 were spiked with mixed fission products to a level of activity of approximately 110 microcuries per liter in each, on April 19, 1954. The level of activity was allowed to decrease by decay, however the rate of decay was very slow. While no chemical determinations were made, a series of determinations of the distribution of radioactivity were made from April 20, to May 27. The results of these determinations, shown in Figure 21, indicated that approximately 86% of the fission product was found in the solid component of samples taken from the digesters.

17. Results of Tests of Additional Methods of Treatment of Radioactive Sludge

(1) Chemical Coagulation and Filtration — Chemical coagulation and filtration experiments were carried out on both raw and digesting sludges, using P^{32}, I^{131}, Ca^{45}, and fission products. Eight sets of data were collected for each experiment. Using the data collected, the activity in the filter was expressed in terms of percentage of the activity in the unfiltered sludge samples.

(a) Radioactive Calcium — The experiments made with digesting sludge containing Ca^{45} at the 110-microcuries-per-liter level fell into two categories. The first three experiments were made within a period of one week on samples of digesting sludge which were spiked at the beginning of the week. The first experiment was conducted the day after spiking, the other two at two-day intervals. There appeared to be no change in the concentration of activity in the filtrate during the period of one week. Gas production indicated that active digestion was taking place throughout the week. In subsequent Ca^{45} experiments, the digesting sludge

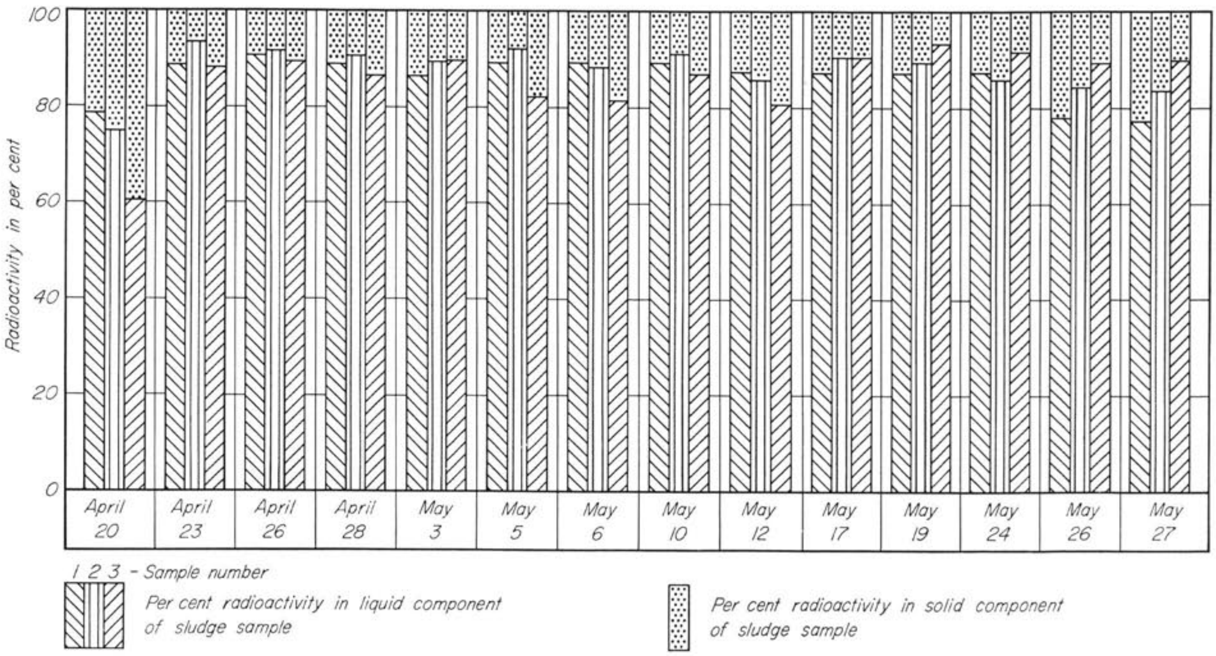


Fig. 21. Distribution of Fission Product Radioactivity Between Solid and Liquid Components of Digested Sludge Samples

samples were spiked several hours prior to the experiments.

In all experiments run with Ca^{45} on raw and digesting sludges, both aluminum sulphate and ferric chloride increased significantly the percentage of activity found in the filtrate. The results indicating percentages of activity found in the filtrates are shown in Tables 5 and 6. Calcium determinations, which are shown in Table 7, showed a release of calcium into the filtrate.

(b) Radioactive Iodine — The results obtained with I^{131} showed a marked difference in behavior between raw and digesting sludges subjected to ferric chloride treatment. Whereas the aluminum sulphate appeared to have no effect, ferric chloride reduced significantly the apparent I^{131} content of the filtrates from digesting sludge, and drastically reduced it in the filtrates from raw sludge, as shown in Tables 8 and 9. However, it is important to consider here the possible loss of iodine during the drying of the counting planchets. An attempt was made to avoid such loss by the addition of 2 drops of 0.2M sodium sulfite in 2M potassium carbonate solution to each planchet prior to drying. The ferric ion readily oxidizes iodine to free iodine. It was suspected, particularly in the

Table 5
Results of Chemical Coagulation and Filtration Tests on Raw Sludge Containing Radioactive Calcium

Set	Activity in Filtrate % of total		pH	Total Solids	Volatile Solids	Total Solids	Volatile Solids	Chemical Used
	Gravity Filtrate	Vacuum Filtrate						
1	39	39	6.9	15.8	10.9	9.7	6.7	None
	63	66	6.7	31.5	21.8			1.5% Alum
	63	59	6.6	30.5	21.0			1.5% $FeCl_3$
2	39	41	6.8	15.3	10.7			None
	59	58	6.7	33.2	23.0			1.5% Alum
	51	63	6.6	30.3	20.9			1.5% $FeCl_3$
3	42	44		13.8	9.7			None
	68	75		31.0	21.3			1.5% Alum
	84	74		29.1	20.6			1.5% $FeCl_3$
4	41	42	6.9	17.5	12.2	11.1	7.6	None
	56	62	6.75	33.4	23.1			1.5% Alum
	51	58	6.7	28.7	20.0			1.5% $FeCl_3$
5	39	40		19.2	13.3			None
	56	69	6.7	33.1	22.9			1.5% Alum
	48	68	6.65	24.3	16.9			1.5% $FeCl_3$
6	38	41		22.0	15.6			None
	59	60		32.6	22.6			1.5% Alum
	64	66		29.1	20.4			1.5% $FeCl_3$
7	57	68		33.3	9.2	9.9	7.2	None
	55	64		20.2	12.7			1.5% Alum
	54	59		51.3	15.4			1.5% $FeCl_3$
8	34	38		16.2	11.8			None
	55	53		36.3	25.3			1.5% Alum
	60	74		29.1	20.0			1.5% $FeCl_3$

Table 6
Results of Chemical Coagulation and Filtration Tests
On Digesting Sludge Containing Ca^{45}

Set	Activity in Filtrate % of total: Gravity Filtrate	Vacuum Filtrate	pH	Filter Cake: Total Solids	Volatile Solids	Sludge: Total Solids	Volatile Solids	Chemical Used
1	16							None
	36							1.5% Alum
	46							1.5% $FeCl_3$
2	13							None
	30							1.5% Alum
	43							1.5% $FeCl_3$
3	12							None
	31							1.5% Alum
	42							1.5% $FeCl_3$
4	18	19	6.9	11.1	6.8	4.7	2.8	None
	48	52	6.7	30.5	17.7			1.5% Alum
	49	47	6.6	25.9	15.1			1.5% $FeCl_3$
5	15	14	6.8	10.1	6.0			None
	51	58	6.7	31.3	18.3			1.5% Alum
	54	61	6.6	23.9	14.1			1.5% $FeCl_3$
6	18	20		7.1	4.4			None
	49	48		23.6	18.1			1.5% Alum
	55	55		24.4	14.3			1.5% $FeCl_3$
7	17	21		6.8	4.1	4.9	2.9	None
	52	53		30.5	17.7			1.5% Alum
	51	51		23.6	13.6			1.5% $FeCl_3$
8	21	14		10.1	5.9			None
	47	47		33.0	19.3			1.5% Alum
	50	52		72.1	15.3			1.5% $FeCl_3$

case of raw sludge, that loss of iodine occurred, since appreciable amounts of iron salts were present in the filtrates as indicated by the color and oxide precipitation. Sulfite is also oxidized by the ferric ion; and with sufficient ferric ion present, both sulfite and iodide will be oxidized.

(c) Radioactive Phosphorus — The results with P^{32} indicated that more of the phosphorus present in digesting sludge was available for equilibration with the added $P^{32}O_4^{=}$ since less activity was found in the filtrates of digesting sludge than in the filtrates of raw sludge, as shown in Tables **10** and **11**. This difference appeared to be reasonable since the digestion process probably liberates organic and bound phosphates as inorganic phosphate. The amount of activity remaining in the filtrates after chemical treatment seemed high, since the activity was added as phosphate and should have precipitated out as aluminum phosphate and ferric phosphate, both of which are very insoluble. The P^{32} was added to the sludges a relatively short time prior to the experiments, and therefore the effects due to microbiological action would probably be small. Phosphate determinations, shown in Table **12**, showed precipitation of the phosphate into the sludge cake remaining on the filters.

Table 7
Calcium Determinations on Chemically Treated Raw
Sludge Containing Radioactive Calcium

Chemical Used	Component Analyzed	Milligrams of Calcium	Counts per Minute
None	Liquid	2.59	10,112
	Solid	20.32	24,454
	Total	22.59	30,054
Alum	Liquid	4.51	16,644
	Solid	13.56	11,607
	Total	20.12	30,249
$FeCl_3$	Liquid	8.07	19,970
	Solid	12.32	9,060
	Total	19.21	27,735

(d) Fission Products — The results of the experiments with fission products are shown in Tables **13** and **14**. The addition of either aluminum sulphate or ferric chloride resulted in a greater

Table 8
Results of Chemical Coagulation and Filtration Tests on
Raw Sludge Containing Radioactive Iodine

Set	Activity in Filtrate % of total: Gravity Filtrate	Vacuum Filtrate	pH	Total Solids	Volatile Solids	Total Solids	Volatile Solids	Chemical Used
1	56	60	5.5	9.0	6.9			None
	55	56	3.7	26.1	19.9			1.5% Alum
	3.2	3.2	2.3	34.9	26.7			1.5% $FeCl_3$
2	61	61		10.2	7.9			None
	62	70		19.7	14.7			1.5% Alum
	3.7	3.3		28.4	21.8	6.1	4.6	1.5% $FeCl_3$
3	45	53	5.6	12.4	9.8			None
	49	59	3.9	31.2	24.0			1.5% Alum
	4.0	4.0	2.5	34.2	25.8			1.5% $FeCl_3$
4	56	49		10.3	8.2			None
	56	49		34.5	27.0			1.5% Alum
	4.0	4.0		42.8	21.2	4.1	3.2	1.5% $FeCl_3$
5	70	60		8.5	6.7			None
	68	60		37.0	29.3			1.5% Alum
	4.0	4.0		33.8	26.7			1.5% $FeCl_3$
6	56	44		8.3	6.6			None
	82	69		36.8	28.9			1.5% Alum
	4.0	4.0		30.9	24.7	4.1	3.2	1.5% $FeCl_3$

Table 9
Results of Chemical Coagulation and Filtration Tests on Digesting Sludge Containing Radioactive Iodine

Set	Activity in Filtrate % of total		pH	Total Solids	Volatile Solids	Total Solids	Volatile Solids	Chemical Used
	Gravity Filtrate	Vacuum Filtrate						
1	57	61	8.0	9.3	5.4			None
	54	58	6.3	37.0	15.6			1.5% Alum
	38	44	5.2	29.3	16.6			1.5% $FeCl_3$
2	55	57		9.5	5.8			None
	62	68		26.9	16.7			1.5% Alum
	41	49		27.5	17.8			1.5% $FeCl_3$
3	55	55		11.3	7.0			None
	55	60		36.3	22.3			1.5% Alum
	34	40		30.2	18.3			1.5% $FeCl_3$
4	51	62		11.3	6.6			None
	57	62		34.0	20.4			1.5% Alum
	33	46		24.6	14.6	6.9	4.0	1.5% $FeCl_3$
5	47	53		9.3	5.7			None
	53	55		26.9	16.4			1.5% Alum
	33	39		27.3	16.4			1.5% $FeCl_3$
6	51	50		9.1	5.1	7.8	4.3	None
	64	52		17.6	9.6			1.5% Alum
	41	52		30.1	16.8			1.5% $FeCl_3$
7	52	52		9.7	5.6			None
	51	55		28.0	16.3			1.5% Alum
	30	24		24.4	14.3			1.5% $FeCl_3$
8	55	42		9.3				None
	55	46		25.7	14.4			1.5% Alum
	41	42		24.1	13.2	6.8	3.7	1.5% $FeCl_3$

Table 10
Results of Chemical Coagulation and Filtration Tests on Raw Sludge Containing Radioactive Phosphorus

Set	Activity in Filtrate % of total		pH	Total Solids	Volatile Solids	Total Solids	Volatile Solids	Chemical Used
	Gravity Filtrate	Vacuum Filtrate						
1	48	50	5.9	16.4	12.7			None
	24	27	4.3	27.0	20.7			1.5% Alum
	18	18	3.0	30.7	23.9			1.5% $FeCl_3$
2	47	58		13.6	10.5			None
	22	10		31.5	24.1			1.5% Alum
	19	15		30.7	23.7	5.9	4.5	1.5% $FeCl_3$
3	59	61		9.7	7.6			None
	24	24		35.3	27.4			1.5% Alum
	21	21		34.8	27.4			1.5% $FeCl_3$
4	51	54		7.2	7.2			None
	25	21		29.2	25.4			1.5% Alum
	34	23		28.9	22.5			1.5% $FeCl_3$
5	13	28		14.2	10.6			None
	19	20		20.5	15.7			1.5% Alum
	15	16		34.4	27.0			1.5% $FeCl_3$
6	25	32		12.1	9.8			None
	17	14		26.8	20.5			1.5% Alum
	11	9		28.9	22.9	8.3	6.4	1.5% $FeCl_3$
7	34	40		25.9	20.1			None
	16	16		25.2	19.2			1.5% Alum
	14	13		30.1	23.0			1.5% $FeCl_3$
8	35	37		21.4	16.7			None
	17	10		26.0	19.4			1.5% Alum
	8	5		27.8	19.5			1.5% $FeCl_3$

Table 11
Results of Chemical Coagulation and Filtration Tests on Digesting Sludge Containing Radioactive Phosphorus

Set	Activity in Filtrate % of total		pH	Total Solids	Volatile Solids	Total Solids	Volatile Solids	Chemical Used
	Gravity Filtrate	Vacuum Filtrate						
1	21	....		24.1	14.8			None
	1.9	....		37.1	22.7			1.5% Alum
	0.3	....		28.3	16.8			1.5% $FeCl_3$
2	22	....		34.4	20.5			None
	1.4	....		38.5	22.9			1.5% Alum
	0.2	....		33.0	19.6			1.5% $FeCl_3$
3	21	....		17.3	10.6			None
	1.8	....		33.4	20.3			1.5% Alum
	0.2	....		28.7	16.9			1.5% $FeCl_3$
4	12	....		25.3	15.7			None
	1.6	....		38.5	23.9			1.5% Alum
	0.3	....		56.2	41.8	6.8	4.2	1.5% $FeCl_3$
5	40	29	8.1	20.9	12.5			None
	1.2	1.1	6.7	31.2	18.6			1.5% Alum
	0.2	0.1	5.6	30.7	18.1			1.5% $FeCl_3$
6	26	20		11.6	7.3			None
	1.1	1.3		28.8	17.8			1.5% Alum
	0.1	0.1		27.1	16.6			1.5% $FeCl_3$
7	20	18		6.6	6.3			None
	1.7	3.4		28.6	17.5			1.5% Alum
	0.3	0.2		28.2	17.7			1.5% $FeCl_3$
8	21	14		10.8	6.7			None
	1.4	0.8		26.0	15.9			1.5% Alum
	0.3	0.2		27.8	16.9			1.5% $FeCl_3$

Table 12

Phosphorus Determinations on Chemically Treated Digesting Sludge Containing Radioactive Phosphorus

Chemical Used	Component Analyzed	Total Milligrams of Phosphorus	Total Counts per Minute
None	Liquid	0.197	156,320
	Solid	23.638	178,039
	Total	26.812	318,890
Alum	Liquid	0.018	9,585
	Solid	24.316	364,164
	Total	23.280	261,033
$FeCl_3$	Liquid	0	240
	Solid	24.368	260,134
	Total	22.390	288,522

Table 13

Results of Chemical Coagulation and Filtration Tests on Raw Sludge Containing Mixed Fission Products

Set	Activity in Filtrate % of total		Chemical Used	Set	Activity in Filtrate % of total		Chemical Used
	Gravity Filtrate	Vacuum Filtrate			Gravity Filtrate	Vacuum Filtrate	
1	10	12	None	5	15	12	None
	28	31	1.5% Alum		32	32	1.5% Alum
	39	42	1.5% $FeCl_3$		32	35	1.5% $FeCl_3$
2	23	27	None	6	12	13	None
	18	24	1.5% Alum		26	29	1.5% Alum
	35	33	1.5% $FeCl_3$		14	27	1.5% $FeCl_3$
3	9	10	None	7	12	12	None
	22	26	1.5% Alum		31	33	1.5% Alum
	30	30	1.5% $FeCl_3$		40	43	1.5% $FeCl_3$
4	12	13	None	8	12	11	None
	25	29	1.5% Alum		25	27	1.5% Alum
	29	28	1.5% $FeCl_3$		41	44	1.5% $FeCl_3$

amount of activity being released into the filtrate than when untreated.

(2) Elutriation —

(a) Fission Products — In the elutriation experiments there was fairly good agreement between the results obtained for different samples of the same sludge. The results of these experiments, shown in Table 15, indicated in the first experiment that 70% of the activity caused by fission products could be washed out with four washings. However, the washwater was very cloudy and contained much finely divided suspended matter. In the second experiment it was decided to centrifuge some of the decanted washwater and count the centrifuged supernatant the decanted washwater.

Although the centrifuged samples were still faintly cloudy, the more readily settleable matter which amounted to about 0.1 to 0.3 c.c. from a 15 ml. portion of the washwater, had been centrifuged out. Whereas counts on the decanted washwater indicated that 80% of the activity was removed by seven washings, these same washwater samples after centrifuging showed the removal of only 32% of the activity. In other words, 48% of the activity present in the decanted washwater was present on the solids which had not settled out. The amount of activity removed in solution was undoubtedly even lower than the centrifuged sample data show since such samples still contained appreciable amounts of finely divided suspended matter. The data indicated that the amount of activity removed after the second wash decreased very rapidly.

(b) Radioactive Phosphorus — An elutriation experiment with P^{32} gave the results shown in Table 16 indicating that 70% of the activity could be washed out in seven washings. Counts on centrifuged samples indicated that 46% of the activity washed out in the seven washings was present on the solids in the washwater.

(3) Flotation — The results of two experiments with raw sludge containing 110 microcuries of fission per liter of sludge are shown in Table 17. A third experiment was attempted with digesting sludge but flotation was not satisfactory and the subnatant contained large amounts of suspended matter. In the two experiments with raw sludge,

Table 14

Results of Chemical Coagulation and Filtration Tests on Digesting Sludge Containing Mixed Fission Products

Set	Activity in Filtrate % of total		pH	Total Solids	Volatile Solids	Total Solids	Volatile Solids	Chemical Used
	Gravity Filtrate	Vacuum Filtrate						
1	5.0	6.0		6.6	3.6			None
	18	16		33.9	18.8			1.5% Alum
	13	14		22.9	12.2			1.5% $FeCl_3$
2	6	6		7.9	4.1			None
	20	14		29.3	15.0			1.5% Alum
	14	17		25.2	13.2			1.5% $FeCl_3$
3	6	7		6.0	3.3			None
	15	12		24.5	12.9			1.5% Alum
	15	16		25.0	13.3	5.5	2.9	1.5% $FeCl_3$
4	6	5		7.7	4.2			None
	16	15		23.4	16.1			1.5% Alum
	14	14		24.2	12.9			1.5% $FeCl_3$
5	5	6		8.3	4.6			None
	20	15		29.0	15.4			1.5% Alum
	14	14		20.8	11.1			1.5% $FeCl_3$
6	5	6		5.6	5.1			None
	15	16		24.6	13.4			1.5% Alum
	12	12		27.9	15.1	5.4	2.9	1.5% $FeCl_3$
7	4	3		11.4	6.3			None
	15	14		29.3	15.8			1.5% Alum
	13	14		24.0	12.9			1.5% $FeCl_3$
8	4	5		8.7	4.7			None
	17	14		26.3	14.1			1.5% Alum
	13	14		21.3	11.4	5.6	3.0	1.5% $FeCl_3$

Table 15
Results of Elutriation Tests on Raw Sludge Containing Mixed Fission Products

Experiment No.	Wash No.	Per Cent of Total Activity Found in Washwater	Per Cent of Total Found in Supernatant of Centrifuged Washwater
I	1	23.5	....
	2	21.2	....
	3	14.9	....
	4	10.4	....
II	1	13.9	6.5
	2	18.6	15.9
	3	15.1	3.9
	4	12.9	2.1
	5	8.6	1.7
	6	6.6	1.2
	7	4.6	1.0

Table 16
Results of Elutriation Tests on Raw Sludge Containing Radioactive Phosphorus

Experiment No.	Wash No.	Per Cent of Total Activity Found in Washwater	Per Cent of Total Activity Found in Supernatant of Centrifuged Washwater
I	1	9.0	5.5
	2	15.7	7.7
	3	11.4	4.1
	4	10.3	2.6
	5	8.8	1.8
	6	8.2	1.2
	7	6.6	1.1
	Total	70.0	24.0

Table 17
Results of Flotation Tests on Raw Sludge Containing Mixed Fission Products

Experiment No.	Sample No.	Total Activity in Sample counts per minute	Activity in Subnatant % of total
I	Subnatant 1	446,800	
	" 2	409,150	
	" 3	379,750	
	" 4	419,225	
	Total Sludge	2,043,250	20.2
II	Subnatant 1	246,876	
	Subnatant 2	285,752	
	Total Sludge	1,337,350	19.9

approximately 20% of the original activity was found in the subnatant after flotation.

(4) Filtration and Weathering — The experiment to test the effects of filtration and weathering on radioactive sludges ran for a period of twenty days. During this period the filters were periodically dosed with distilled water and filtrates collected for analysis. On the thirteenth day of the experiment, distilled water was applied to alternate filters to determine whether the radioactivity could be leached out or washed through. The results of determinations of the amounts of radioactivity found

Table 18
Results of Filtration and Leaching Tests on Radioactive Sludges

Date	Filter No.	Radioactive Material	Volume of Filtrate Collected ml.	Activity in Filtrate % of total on filter	Volume of Distilled Water Applied to Filter ml.
5/8/54	1	None	2095	0.5	1000
	2	MFP	1810	0.59	"
	3	MFP	2115	0.37	"
	4	P^{32}	1940	0.59	"
	5	P^{32}	2100	0.53	"
	6	I^{131}	2000	5.71	"
	7	I^{131}	2070	4.21	"
	8	Ca^{45}	1465	0.06	"
	9	Ca^{45}	1475	0.29	"
	10	S^{35}	875	0.12	"
5/9/54	1	None	1105	0.08	None
	2	MFP	970	0.63	"
	3	MFP	1125	0.69	"
	4	P^{32}	890	0.89	"
	5	P^{32}	1115	1.23	"
	6	I^{131}	1055	6.14	"
	7	I^{131}	1105	6.08	"
	8	I^{45}	500	0.19	"
	9	Ca^{45}	630	0.65	"
	10	S^{35}	400	0.27	"
5/12/54	1	None	415	0.01	1000
	2	MFP	570	0.46	"
	3	MFP	505	0.39	"
	4	P^{32}	560	1.16	"
	5	P^{32}	420	1.11	"
	6	I^{131}	380	3.84	"
	7	I^{131}	400	4.45	"
	8	Ca^{45}	815	0.59	"
	9	Ca^{45}	900	0.96	"
	10	S^{35}	630	0.63	"
5/14/54	1	None	845	0.09	1000
	2	MFP	640	0.38	"
	3	MFP	620	0.25	"
	4	P^{32}	690	0.96	"
	5	P^{32}	775	1.68	"
	6	I^{131}	755	3.84	"
	7	I^{131}	725	5.42	"
	8	Ca^{45}	795	1.35	"
	9	Ca^{45}	585	1.00	"
	10	S^{35}	270	0.46	"
5/17/54	1	None	495	0.07	1000
	2	MFP	650	0.28	"
	3	MFP	790	0.27	"
	4	P^{32}	840	0.31	"
	5	P^{32}	830	0.54	"
	6	I^{131}	910	1.41	"
	7	I^{131}	790	1.47	"
	8	Ca^{45}	830	0.83	"
	9	Ca^{45}	690	1.02	"
	10	S^{35}	450	0.58	"
5/19/54	1	None	665	11.35	1000
	2	MFP	710	0.16	"
	3	MFP	800	0.16	"
	4	P^{32}	810	0.26	"
	5	P^{32}	750	0.30	"
	6	I^{131}	725	0.83	"
	7	I^{131}	800	1.08	"
	8	Ca^{45}	365	0.51	"
	9	Ca^{45}	435	0.69	"
	10	S^{35}	380	0.40	"
5/20/54	1	None	670	0.09	None
	2	MFP	760	0.12	1000
	3	MFP	780	0.09	None
	4	P^{32}	760	0.22	1000
	5	P^{32}	780	0.26	None
	6	I^{131}	780	0.47	1000
	7	I^{131}	740	0.43	None
	8	Ca^{45}	600	1.48	1000
	9	Ca^{45}	280	0.79	None
	10	S^{35}	220	0.14	1000
5/21/54	1	None	20	0	None
	2	MFP	680	0.12	"
	3	MFP	10	0.003	"
	4	P^{32}	830	0.12	"
	5	P^{32}	10	0.003	"
	6	I^{131}	775	0.29	"
	7	I^{131}	10	0.004	"
	8	Ca^{45}	260	0.60	"
	9	Ca^{45}	260	0.007	"
	10	S^{35}	250	0.07	"
5/24/54	1	None	0	....	1000
	2	MFP	0	....	"
	3	MFP	0	....	"
	4	P^{32}	0	....	"
	5	P^{32}	0	....	"
	6	I^{131}	0	....	"
	7	I^{131}	0	....	"
	8	Ca^{45}	400	0.85	"
	9	Ca^{45}	270	0.69	"
	10	S^{35}	600	0.24	"
5/26/54	1	None	725	0.06	
	2	MFP	760	0.13	
	3	MFP	805	0.10	
	4	P^{32}	850	0.42	
	5	P^{32}	755	0.32	
	6	I^{131}	780	0.54	
	7	I^{131}	760	0.69	
	8	Ca^{45}	835	0.73	
	9	Ca^{45}	860	0.79	
	10	S^{35}	290	0.15	

in the filtrates are shown in Table 18. From these experiments it appears that the amount of radioactivity released in the filtrate is proportionate to the volume of filtrate. This was indicated in the instance where alternate filters were dosed with distilled water and the filtrates from all filters analyzed for radioactivity.

(5) Filtration — A series of filtration experiments using sludge containing P^{32} were made in the lucite cylinder filters. The purpose of these tests was to determine the amount of radioactivity and phosphate which could be expected to appear in the filtrate from radioactive sludge applied to sand drying beds. The results of these experiments, shown in Table 19, indicated a higher percentage of the activity in the filtrate than was found in the filtrates of the filtration and leaching experiment. The results of this experiment were comparable to results obtained on samples from the stirred digesters which were separated into solid and liquid components by filtration through filter paper and by centrifuging.

Table 19

Results of Sand Filtration Tests Using Digested Sludge Containing Radioactive Phosphorus

Date	Sample	Activity in Filtrate % of total in sludge	Distribution of Phosphate mg. in filtrate	mg. in sludge cake
10/23/53	1	4.3	0.36	28.53
	1	8.0	0.47	28.32
	2	10.0	0.42	20.17
	2	13.1	0.57	19.79
	3	10.9	0.37	18.76
	3	4.0	0.17	25.03
11/6/53	1	12.0	0.33	10.73
	2	7.4	0.21	8.84
	3	9.2	0.29	8.21
11/13/53	1	5.0	0.38	8.64
	3	5.0	0.29	7.72

V. SUMMARY AND CONCLUSIONS

18. Summary of Test Results

Tests made during this study, and their results, were:

(1) To determine the effects of stable elements on sludge digestion prior to testing radioisotopes. These tests, which are described on pages 13 through 19, indicated that concentrations of the elements listed below have little effect on anaerobic digestion as measured by gas production and bacteriological examination.

a. phosphorus — less than 0.03% (as P)
b. iodine — less than 0.2 % (as I)
c. calcium — less than 0.01% (as Ca^2)
d. sulphur — less than 0.1 % (as SO_4)

(2) To determine the effects of radioactive materials on sludge digestion in batch type experiments using gas production as the primary index to digestion and to the limiting amounts of radioactivity producing inhibition of the digestion process. A description of these tests is found on pages 19 through 22. The effects of concentrations as high as 200 millicuries per liter were tested and indicated that gas production was reduced by about 15% at the highest concentration of P^{32}, presumably from the effects of radioactivity. Maximum concentrations of 200 millicuries per liter were tested using I^{131}, S^{35}, and fission products. At the 200 millicurie per liter level, these tests indicated that I^{131} inhibited gas production by as much as 4%, S^{35} by as much as 55%, and fission products by as much as 93%. In the case of the fission products, the tests indicated that about three-fourths of the inhibition was due to nitric acid present in the radioactive solution.

(3) To determine the effects of radioactive materials on sludge digestion under conditions approximating normal sewage treatment sludge digestion processes. The experiments made on this phase of the study were carried out in the stirred digestion apparatus, and the majority were run at levels of activity of 110 microcuries per liter. These tests, described on pages 20 through 22, and 28, indicated that the 110 microcurie per liter level of activity had no significant effect on digestion when P^{32}, I^{131}, and S^{35} were used as indicated by the cumulative gas production curves shown in Figures 22, 23, and 24. However, the tests with fission products indicated that digestion over a 30-day period, as measured by gas production, was inhibited by approximately 17% as shown in Figure 25. The quality of gas produced in digesters spiked with radioactive materials and in the control digesters is shown in Table 20. A comparison of these qualitative analyses indicates practically no difference between the quality of gas from spiked digesters and gas from control digesters.

(4) To determine the degree of concentration of radioactivity in either the solid or liquid component of sludge during the digestion process. Samples from the stirred digesters were separated into solid and liquid components which were analyzed for radioactivity and for the element being tested. Results of these tests, shown in Figures 19, 20, and 21 indicated that radioactive phosphorus was distributed about four to one in the solid component of samples tested; radioactive sulphur distributed about nine to one in the solid component; and fission products about five or six to one in the solid component. The results of determinations of distribution of radioactive iodine are not shown graphically. However, they indicated that 65 to 85% of the radioactive iodine was to be found in the liquid component of samples tested. In addition to determining the distribution of radioactivity, the distribution of the element tested was determined in an attempt to find whether isotopic dilution of the radioisotope occurred. Neither S^{35} nor P^{32} was found to follow this law. In either case, varying concentrations of radioactivity per milligram of stable element were found in a series of tests made on the liquid and solid components.

(5) To determine the potentialities of utilizing chemical coagulation and filtration, elutriation, or flotation as a means of preparation of radioactive wastes for ultimate disposal. Descriptions of those

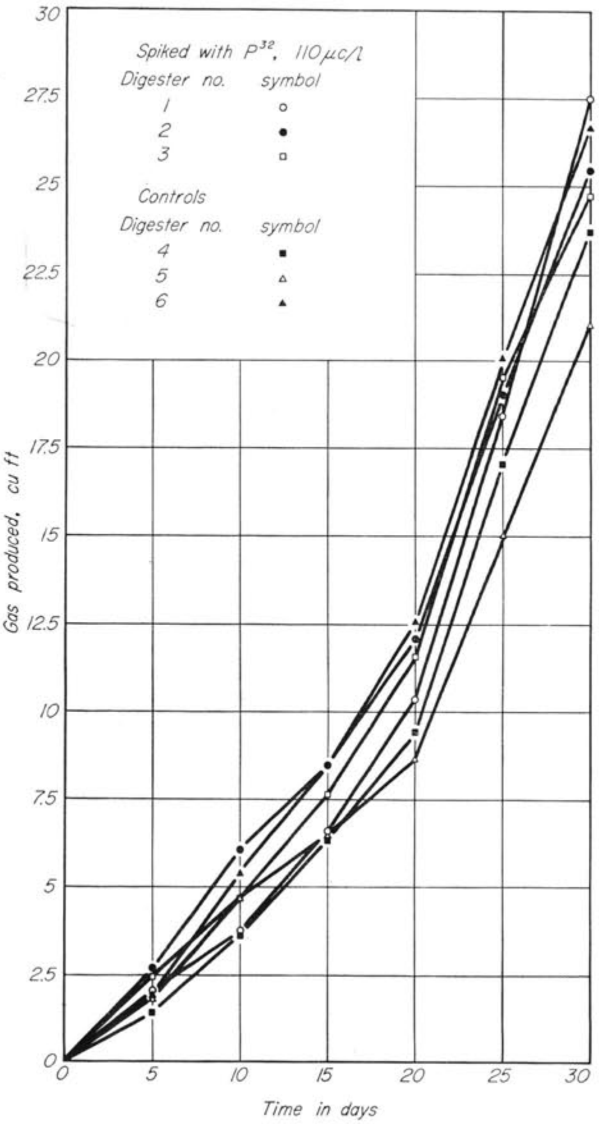


Fig. 22. Cumulative Gas Production Curve for Stirred Digestion Tests Using Radioactive Phosphorus

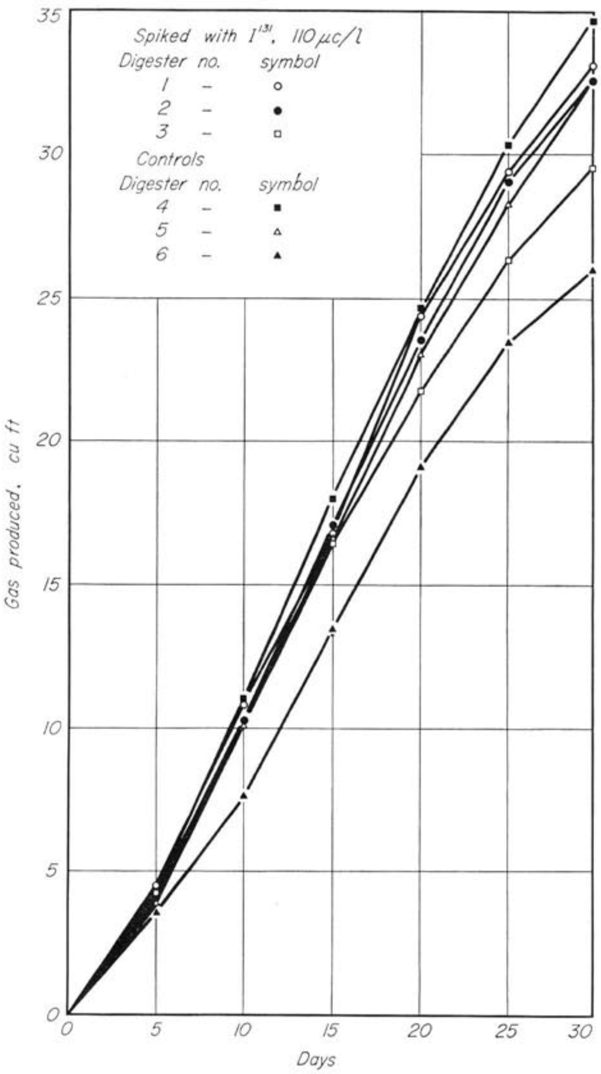


Fig. 23. Cumulative Gas Production Curve for Stirred Digestion Tests Using Radioactive Iodine

tests are given on pages 22 through 24 and 30 through 36. The chemical coagulation and filtration tests on raw and digesting sludges containing Ca^{45} indicated that aluminum sulphate and ferric chloride increased significantly the amount of activity found in the filtrate from the sludge samples. Calcium determinations likewise showed a release of calcium into the filtrate due to treatment with the two coagulants.

These results can be attributed in part to the insolubility of ferric and aluminum phosphates, both of which are less soluble than the corresponding calcium salt. Previous experiments indicated that a portion of the calcium of the solid component is held as calcium phosphate, since precipitation of the calcium as calcium oxalate prior to the addition of $P^{32}O_4$ caused most of the labeled phosphate to remain in the liquid component, instead of being held as calcium phosphate. The filtrates from coagulated samples contained from 40 to 60% of the Ca^{45} activity in the original samples, whereas the filtrate from untreated samples contained from 16 to 40% of the original Ca^{45} activity.

Tests with raw and digesting sludges containing fission products indicated that coagulation increased the amount of activity passed in the filtrates of raw sludge by as much as 22%, and by as much as 12% in digesting sludge filtrates. Tests with I^{131} indicated that ferric chloride coagulation reduced the apparent activity in the filtrates quite drastically

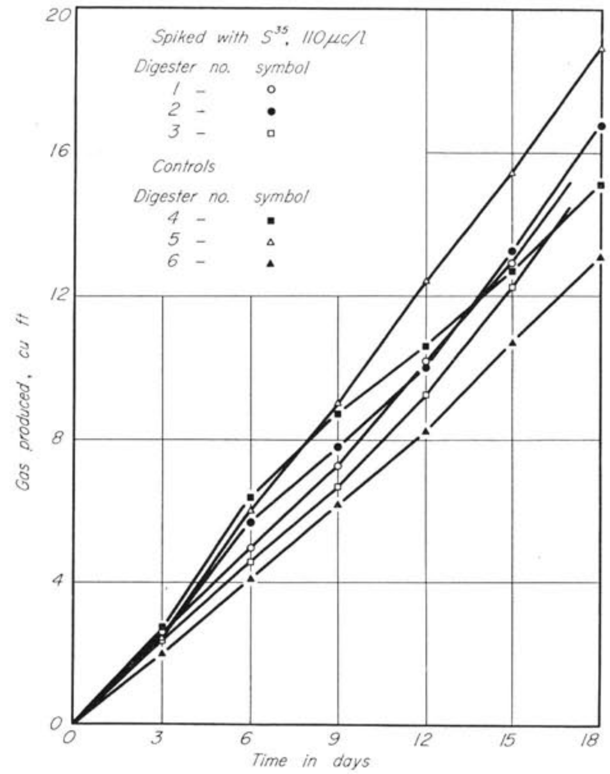


Fig. 24. Cumulative Gas Production Curve for Stirred Digestion Tests Using Radioactive Sulphur

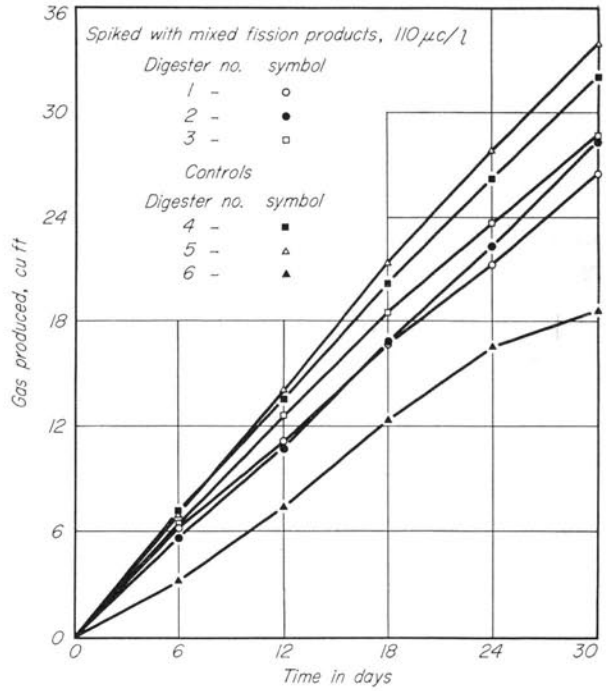


Fig. 25. Cumulative Gas Production Curve for Stirred Digestion Tests Using Mixed Fission Products

Table 20

Representative Analyses of Gas Produced During Tests in Stirred Digestion Apparatus

Date of Analysis	Gas From Radioactive Digesters		Gas From Control Digesters	
	% CH_4	% CO_2	% CH_4	% CO_2
11/10/53	55.8	42.1	55.5	40.5
11/23/53	65.8	33.8	60.4	39.1
11/27/53	65.0	34.4	66.0	33.4
11/30/53	64.4	34.8	65.1	34.3
12/2/53	65.4	34.2	64.8	34.7
12/7/53	65.1	34.1	64.1	35.4
12/10/53	65.1	34.2	65.3	34.0
12/14/53	61.2	37.9	60.9	38.5
12/15/53	63.8	34.9	62.4	36.7
12/21/53	64.9	34.9	64.4	35.1
12/28/53	68.2	30.6	66.5	32.8
1/4/54	69.0	30.3	67.7	31.7
1/7/54	66.9	31.7	63.7	35.5
1/11/54	63.4	35.5	65.1	34.1
1/25/54	65.5	33.9	66.0	33.4
2/3/54	65.2	34.7	64.1	35.0
2/10/54	66.2	32.5	64.7	34.7
2/16/54	66.5	32.8	67.5	31.8
2/23/54	63.7	35.6	65.4	34.1
3/2/54	64.9	34.1	66.9	32.6
3/30/54	65.9	33.4	66.7	32.5
4/21/54	65.6	33.9	67.6	32.1
4/26/54	66.4	33.1	67.0	32.3
4/28/54	64.1	35.4	64.7	34.9
5/5/54	65.1	34.6	64.0	35.6
5/11/54*	51.8	32.5	48.2	32.1
5/13/54*	42.1	32.3	54.9	34.4
5/17/54	67.5	32.1	63.5	36.0
5/19/54*	54.6	33.7	49.7	36.3
5/24/54*	54.6	33.1	53.8	41.1
Median	65.1	34.1	64.7	34.4

Note: All methane values except those on dates marked with an asterisk (*) actually include all combustible gases and inerts present in the gas. Those values for the dates marked with an asterisk represent methane values only. The median values for the four dates on which complete analyses were possible, are about ten percentage points lower than those indicated above for methane.

in the case of raw sludge and significantly in the case of digesting sludge. The results with P^{32} indicated that more of the phosphorus present in the digesting sludge used in these tests was available for equilibration with the added $P^{32}O_4$, since less activity was found in the filtrates from digesting sludge than in the filtrates from raw sludge.

Elutriation tests with fission products and with P^{32} indicated that from 70 to 80% of the activity in raw sludge spiked with fission products could be washed out with from four to seven washings. Seventy per cent of the activity in raw sludge spiked with P^{32} was washed out in seven washings. In general, the maximum amount of activity washed out in any single elutriation occurred during the second wash.

Flotation experiments, which are described on pages 24, 34, and 35, indicated that about 20% of the total activity in radioactive raw sludge was found in the subnatant after flotation.

Sand filtration tests in the lucite cylinder filters with digested sludge containing P^{32} indicated that approximately 10% of the activity in the sludge passed through the filter in the filtrate.

In subsequent filtration and leaching experiments with several radioactive materials, including P^{32}, the percentage of activity passing through the filter from the P^{32} sludge reached only about 5%.

However, the radioactive sludge used in the subsequent tests had not digested as long as that used in the lucite cylinder tests.

The results of the filtration and leaching experiments with various radioactive materials, shown in Table 18, indicated 2 to 3% of fission products in the filtrate, 5 to 6% of P^{32} in the filtrate, 7% of Ca^{45} in the filtrate, and 3% of S^{35} in the filtrate. These tests indicated that the amount of activity leaving the sludge at any given time was dependent, to a degree, upon the amount of moisture placed on the filter. A comparison of the percentage of activity in the filtrates from one collection period to another indicated that the amount of activity was dependent upon the amount of filtrate collected. Dosing alternate filters with distilled water demonstrated that it was possible to wash activity out of the filters. This observation verified results obtained in the elutriation tests.

19. Conclusions

From the results of the tests in this study it is concluded that:

(1) Radioactive materials such as P^{32}, I^{131}, and S^{35} in concentrations of 110 microcuries per liter have no apparent effect on normal sludge digestion. Concentrations up to 200 millicuries per liter can be reached before the inhibitory effect on anaerobic digestion becomes significant. The concentration of 200 millicuries per liter in sludge exceeds by 17×10^6 to 40×10^6 times the concentration of activity in sludge from sewage containing activity in the range 0.25×10^{-4} to 0.60×10^{-4} microcuries per liter estimated by Ruchhoft and Feitelberg(5) to be expected in sewage of six large American cities.

(2) Fission products in the 110 microcuries per liter concentration showed a definite inhibitory effect on digestion during the 30-day digestion period. The gas production from the stirred digestion apparatus indicated that digestion capacity would have to be increased by approximately 17% for sludge containing 110 microcuries of fission products per liter.

(3) Those radioactive materials tested, which Porter lists as satisfying some of the mineral requirements of bacteria, such as phosphorus, calcium, and sulphur, appeared to concentrate in the solid component of the sludge during the digestion process. On the other hand, iodine, the function of which is unknown in regard to bacterial requirements, appeared to concentrate in the liquid component. Whether these distributions were due to bacterial action alone was not determined in this study.

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