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COO-196

PROGRESS REPORT 5

UTILIZATION OF THE GROSS FISSION PRODUCTS
(Unclassified)

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Project M943

U. S. ATOMIC ENERGY COMMISSION
CONTRACT NO. AT (11-1)-162
CHICAGO 80, ILLINOIS

September, 1953

PREFACE

This report presents the results of research performed during the period January 1, 1953, to June 30, 1953, on Project M943 of the Engineering Research Institute, University of Michigan, under AEC Contract No. AT(11-1)-162.

Results of other research studies in which the irradiation facilities of the Fission Products Laboratory have been used are also reported because it is believed that these studies may provide additional data on the use of waste fission products. For this reason some of the studies supported by Michigan Memorial-Phoenix projects 41, 43, 54, 73, and 83 and some of the academic studies of graduate students are included in this report. It is to be noted that there is no relationship between these studies and Project M943 except that the personnel have a common interest in the effects of radiation and the possible uses of radioactive materials, and they use the same source of gamma radiation. The research of the Michigan Memorial-Phoenix Project is supported by funds of the University of Michigan obtained through contributions by individuals and corporations and is in no way supported by, or connected with, the Atomic Energy Commission. The results of the studies reported by the Michigan Memorial-Phoenix Project in most instances will appear in the scientific literature at some future date. To protect the authorship of research personnel of the Michigan Memorial-Phoenix Project, no part of their results is to be reproduced without permission of the respective authors.

ABSTRACT

This report describes the work accomplished since Progress Report 4 (COO-124), dated March, 1953, was prepared. The experimental studies detailed in this report may be summarized as follows:

The fundamental study of the effect of radiation on combustion (subproject M943-2) has continued and the experimental equipment has been used successfully; however, as yet experimental difficulties have not permitted the comparison of explosion-limit curves with and without the influence of beta-particle radiation. This study will be completed as a portion of the studies on the effect of radiation on chemical reactions (subproject M943-4).

The study of performance of combustion engines under the influence of radiation (subproject M943-3) has been continued with tests on drag-stabilized hydrocarbon-air flames under the influence of radiation from 2050 curies of gold-198. The results of these tests indicate that more efficient combustion is obtained when radiation intensity is high. These tests were concluded with support from Michigan Memorial-Phoenix project 83.

The study of the effect of radiation on the promotion of chemical reactions has been continued with studies on the polymerization of ethylene and the chlorination of benzene and toluene. Initial studies indicate that the chlorination of toluene gives the hexachloro addition product. A discussion of the thermodynamics and kinetics of reactions is included.

Dosimetry studies on both the 1-kc and 10-kc sources are reported, and the nominal activities and those determined from dosimetry measurements are compared. The actual activity of both the 1-kc and 10-kc sources is found to be considerably less than the reported activity. Personnel of Brookhaven National Laboratory have reached the same conclusion regarding the true activity of the 1-kc source, based on reactor computations.

The Michigan Memorial-Phoenix projects have used gamma radiation in a number of studies which are reported. Phoenix project 41 is supporting an extensive animal-feeding experiment using gamma-irradiated food. The original plan of the feeding experiment reported in Progress Report 4 has been modified to include one diet in which all the food is irradiated. Personnel of Phoenix project 54 have treated large sections of trichinous pork with gamma radiation using both the 10-kc cobalt source and radiation from spent reactor fuel slugs. A preliminary design for a pork processing plant to handle 2000 hogs per day indicates that the irradiation of pork using waste fission products would require 1-1/2 megacuries of fission products and would increase the cost of the pork approximately 2.3 mills per pound. Various designs of irradiation chambers are considered.

Michigan Memorial-Phoenix project 73 is supporting an investigation of the nutritive value of irradiated media using the protozoan Tetrahymena

pyriformis as the test organism. The requirements of this protozoan for the essential amino acids and the B vitamins are very similar to those of man. The short life cycle of this organism compared to experimental animals such as rats should accelerate the accumulation of data on this problem.

Preliminary studies of the effect of radiation on rubber and on polymerization have been initiated with the support of Phoenix project 43. The nature of the polymer, the action of compounding agents, and possibly the degree and nature of cure all influence the resistance of a given rubber formulation to gamma radiation.

The gamma-catalyzed polymerization of a mixture of styrene and methyl methoxylate appears to be a free-radical process; however, the gamma-catalyzed polymerization of styrene is mildly promoted by several rubber antioxidants and vulcanization accelerators and is retarded by sulfur, dinitrobenzene, and a few other materials.

Academic studies in the Department of Bacteriology by N. J. Williams on the irradiation of human blood culture media indicate that radiation sterilization of media is not feasible because changes are produced which alter the growth of certain organisms on the irradiated media.

The new 10-kc gamma source has been installed in the Fission Products Laboratory and has been in satisfactory operation for several months. It was found necessary to add concrete shielding to the ceiling of the radiation "cave" in order to reduce radiation effects in nearby buildings to levels approved by the Radiation Policy Committee of the University of Michigan. Some difficulty has been experienced in controlling the pH and clarity of the water in the well of the radiation cave; however, to date no corrosion of the aluminum jackets of the cobalt rods is evident.

Academic studies on the use of polyvinyl chloride films in dosimetry measurements has been conducted by L. M. Welshans. Procedures for preparing and using these films and modifications of published techniques are reported and discussed.

TABLE OF CONTENTS

TABLE OF CONTENTS (cont.)

TABLE OF CONTENTS (cont.)

TABLE OF CONTENTS (cont.)

LIST OF FIGURES

LIST OF FIGURES (cont.)

LIST OF FIGURES (cont.)

LIST OF FIGURES (cont.)

LIST OF TABLES

LIST OF TABLES (cont.)

PROGRESS REPORT 5

UTILIZATION OF THE GROSS FISSION PRODUCTS

PART I. THE EFFECT OF IONIZING RADIATION ON COMBUSTION ENGINE PERFORMANCE

A. SUBPROJECT M943-2, FUNDAMENTAL STUDY OF EFFECT OF RADIATION ON COMBUSTION

Personnel:

Subproject Supervisor: R. A. Wolfe, Professor of Physics; F. L. Tobey, Research Assistant.

1. Review

This subproject is investigating the effects of intense beta radiation on gaseous combustion. In particular, a study of the explosion limits of gaseous mixtures has been undertaken. In these experiments a "bomb" is charged with a mixture of combustible gas, oxygen, and nitrogen at pressures up to 20 atmospheres. The mixture is then exploded by a spark if possible. The pressure of nitrogen is varied until the mixture just fails to explode. This condition defines the explosion limit for a given pressure of $(2H_2 + O_2)$. The process is then repeated with a different pressure of $(2H_2 + O_2)$. By plotting pressure of $(2H_2 + O_2)$ against pressure of nitrogen, an explosion-limit curve can be obtained. The experiments are then repeated in the presence of an intense beta-source. If such a source is capable of affecting the combustion process, a shift in the explosion-limit curve should be observed.

2. Equipment

Some modifications have been made in the equipment described in Progress Report 3, as follows:

(a) The blowout assembly on the bomb has been replaced by an Aminco superpressure needle valve. It was found that an explosion in the bomb could be heard readily even when completely contained. The use of the needle valve eliminates the potentially difficult problem of filtering a high-pressure gas-exhaust stream which might contain radioactive material. The bomb may now be exhausted safely into the regular laboratory hood system, through suitable prefilters.

(b) A manometer and McLeod gauge have been added to the system to insure complete evacuation of the bomb between runs.

(c) An additional exhaust line has been introduced to improve the evacuation rate of the bomb. This was necessary because of the highly restricted openings required in an explosion system.

Figure 1a is a schematic diagram of the present system. Figure 1b is a photograph of the head of the pressure vessel, and Fig. 1c is a photograph of the control panel for the explosion system

3. Experimental Work

The equipment is now operating satisfactorily, and an attempt is being made to determine the normal explosion-limit curve. Two difficulties have been encountered which have prevented reproducible results being obtained as yet. These are:

(a) a marked dependence of the spark plug on its past history insofar as its ability to produce an explosion is concerned, and

(b) the unexpectedly long time which is required for proper mixing of gases to take place in the bomb.

Once an explosion-limit curve is established, the effects of intense beta radiation will be tested. The use of phosphorus-32 as a source was found

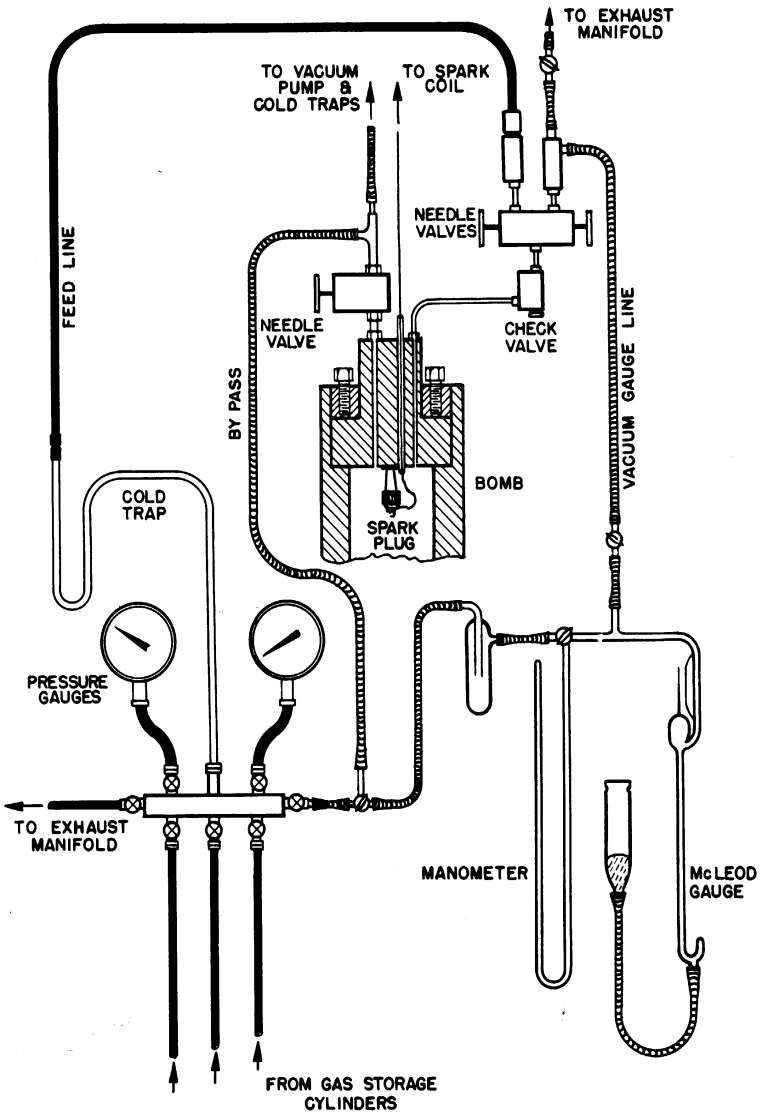

Fig. 1a. Schematic Diagram of Explosion Limit Equipment.

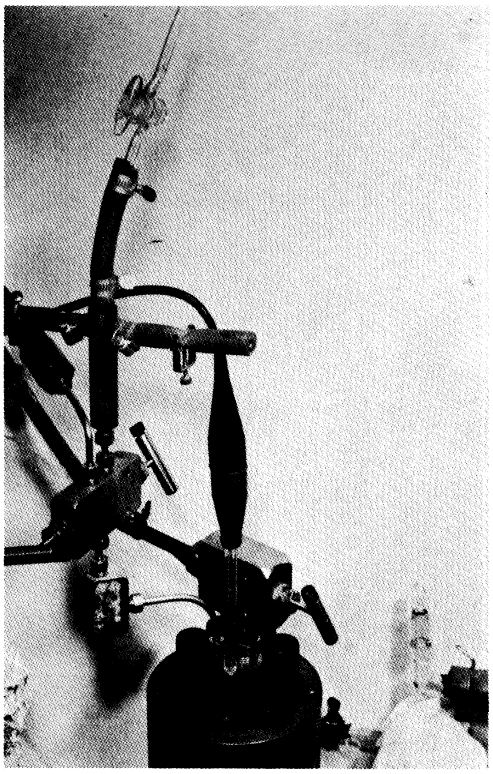

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Fig. 1b. Head of Pressure Vessel.

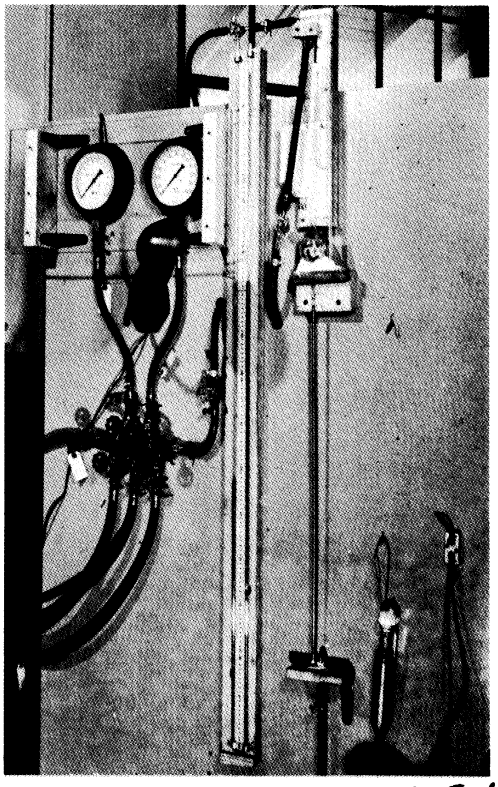

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Fig. 1c. Control Panel for Explosion System.

to be impractical, as the bakelite placques in which the phosphorus is suspended are badly melted and burned by the explosion.

Gold-198 has been tentatively selected as the source material. A 5-mil gold foil will be irradiated in a high neutron flux to produce a source strength of about 1000 curies.

B. SUBPROJECT M943-3, PERFORMANCE OF COMBUSTION ENGINES UNDER THE INFLUENCE OF RADIATION

1. Internal-Combustion Engines

Personnel:

Subproject Supervisors: E. T. Vincent, Professor of Mechanical Engineering and Chairman of Department; G. J. Van Wylen, Assistant Professor of Mechanical Engineering.

a. Introduction. A study of the effects of beta radiation on the performance of internal-combustion engines is underway. Of two previous tests on diesel engines, one using a Pd disc source indicated a slight improvement of performance when the combustion process was subjected to beta radiation, and the other using a Pd rod source showed no change. The tests outlined in this present progress report are an attempt to utilize the experience and information gained from previous tests and establish definitely whether beta radiation has an effect on the combustion process in internal-combustion engines.

The equipment for the tests outlined in this report has been set up and the tests will be performed as soon as the irradiated palladium is received. There has been a delay in getting these samples irradiated; the tests are now scheduled to be conducted about the end of August. The support of this study by Project M943 ceased on June 30, 1953. The experiments described will be completed with support from Michigan Memorial-Phoenix project 83.

b. Description of Diesel Engine Test. The same engine will be used as has been used in previous tests, namely a single-cylinder CFR diesel test engine. On the basis of the second test which was outlined in Progress Report 4, two changes in experimental technique will be made. First, two identical pieces of palladium will be used, one irradiated and the other not. The two

tests will be run successively thus maintaining identical operating conditions for the two runs. The second change is to surround the combustion chamber with a radioactive material, thus keeping the center of the combustion chamber free and thereby promoting good swirl and mixing.

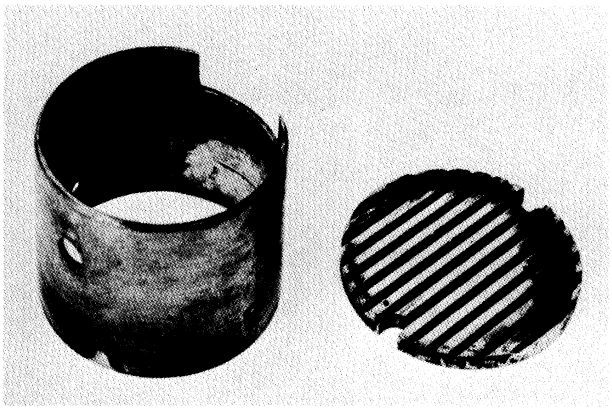

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Fig. 2. Palladium Disk and Cylinder for Diesel Engine Experiments.

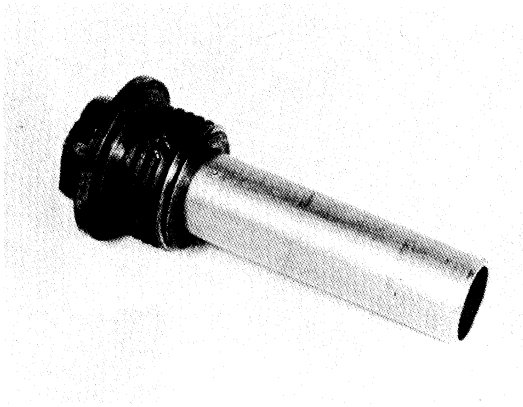

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Fig. 3. Palladium Tube for Tests on Spark Ignition Engine.

These changes have been incorporated in this test setup. Figure 2 shows the palladium sources to be used. The cylinder and the disc will both be radioactive; thus the combustion chamber is almost entirely surrounded by beta emitting surfaces. This design does not permit variation in the compression ratio, but the compression ratio selected (which determined the length of the palladium cylinder) was 16.5 to 1, a typical value for diesel engines.

The data to be taken during this test are the specific fuel consumption and pressure-time measurements. The latter will be obtained by photographing the trace from a pressure pick-up on the oscilloscope. A more suitable dynamometer and improved fuel-rate measuring technique will be used, which it is believed will increase the accuracy of the results.

c. Description of Spark-Ignition Engine Test Procedure. This will be the first test conducted on a spark-ignition engine. The engine used is a single-cylinder CFR spark-ignition enigne, which has a variable compression ratio. The source of radiation is a palladium tube, 7/16 inch in diameter, 2 inches long, and 1/32 inch thick. Of this length, 1.6 inches are exposed in the cylinder. Figure 3 shows this palladium tube in its holder. The cylinder head for this engine has four spark-plug holes; the palladium tube will be inserted in one of these and held in place by a fixture which fits into the spark-plug hole.

This engine is fitted with a fuel pump and mixing evaporator tank. The purpose of this arrangement is to insure that a dry air-fuel mixture is

being supplied to the engine. This leads to greater uniformity and reliability of results.

The data to be obtained in this test are the specific fuel consumption, pressure-time measurements, and knock intensity, all as functions of compression ratio.

As stated previously the equipment for these experiments has been assembled and the pieces of palladium to be irradiated are at the pile site awaiting irradiation. The results of this test will be presented in the next progress report.

2. Jet Engines

Personnel:

Subproject Supervisors: E. T. Vincent, Professor of Mechanical Engineering and Chairman of Department; R. B. Morrison, Assistant Professor of Aeronautical Engineering; R. E. Cullen, Associate Research Engineer; M. E. Gluckstein, Research Associate.

a. Introduction. Drag-stabilized hydrocarbon-air flames have been subjected to the radiation from a multicurie source of gold-198. The equipment used in this work was described in Progress Report 4.

b. Experimental Techniques. (1) Handling the gold source: The source used in this work was a tube of gold and copper which could be set in the burner wall. The inner surface, which was 999.5-fine gold, was bonded to the copper. The copper served only as a structural matrix for the gold.

This source was activated in the Materials Testing Reactor at Idaho Falls. The radioisotopes Au^{198} and Cu^{64} were formed by the reactions:

$$Au^{197}\ (n,\gamma)\ Au^{197}$$

$$Cu^{63}\ (n,\gamma)\ Cu^{64}$$

Decay of these isotopes takes place as follows:

$$Au^{198} \rightarrow Hg^{198} + \beta^- + \gamma$$

$$Cu^{64} \quad Zn^{64} + (\beta^-) \text{ or } (\beta^+) + \gamma$$

The mercury and zinc daughters produced are stable and the energies involved in the decay schemes are given in Table I.

TABLE I[1]

ENERGY FROM DECAY OF GOLD-198 AND COPPER 64

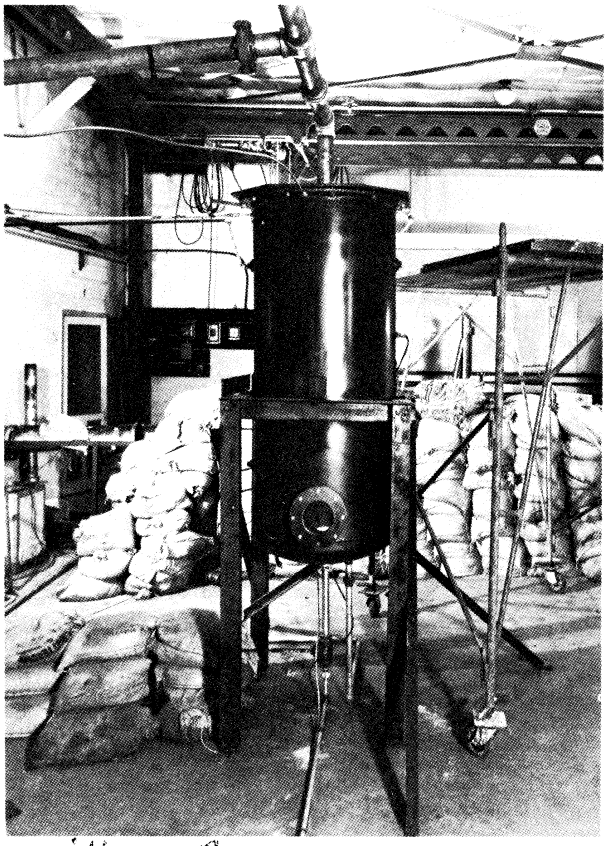
Isotope	Half Life, Hrs	Energy, mev			Initial Activity,* Curies
		γ	β^+	β^-	
Au^{198}	65	.41	0	.96	2050
Cu^{64}	12.9	1.35	.66	.56	1300

* At time of removal from reactor.

Since the copper gives rise to a high level of gamma radiation, it was necessary to design a shipping container and handling system that would allow safe handling of the source at its maximum level. The shipping container, equivalent to 5.5 inches of lead shielding in all directions, was adequate for this source. The experimental system is discussed in Progress Report 4. Figure 4 shows a view of the experimental equipment with the test tank prior to the installation of the shielding; Fig. 5 is a similar view showing the sandbag shield in place.

The source was shipped from Idaho Falls to the Aircraft Propulsion Laboratory at Willow Run Airport by a special airlift arranged with the United States Air Force. Figure 7 shows the unloading of the shipping container at Willow Run Airport. At the Laboratory, the lead cask was removed from the pallette, hoisted into place, and lowered into the experimental tank. The tank had previously been filled with 5 feet of water, which was the shield for the assembly operation. Figure 6 shows the cask being lowered into place just prior to removing the top.

After the cask was in place the health-physics personnel set up monitoring equipment above the tank and the top of the cask was removed. The radiation level through the water from the open cask remained below 5 mr/hr. The source was then removed from the cask by means of 8-foot-long "crow's foot" tongs. The cask was then removed and surveyed for contamination. At no time was the source raised above the water level, and this operation was constantly monitored. A surface dose-rate measurement showed the cask to be clean, except for a 15-mr/hr reading above the opening. The cask was wrapped in absorbent paper and lowered to a covered area on the floor. The dose rate was below 5 mr/hr at the surface of the water.



Willow Run

Fig. 4. Tank Prior to Installation of Sandbag Barriers.

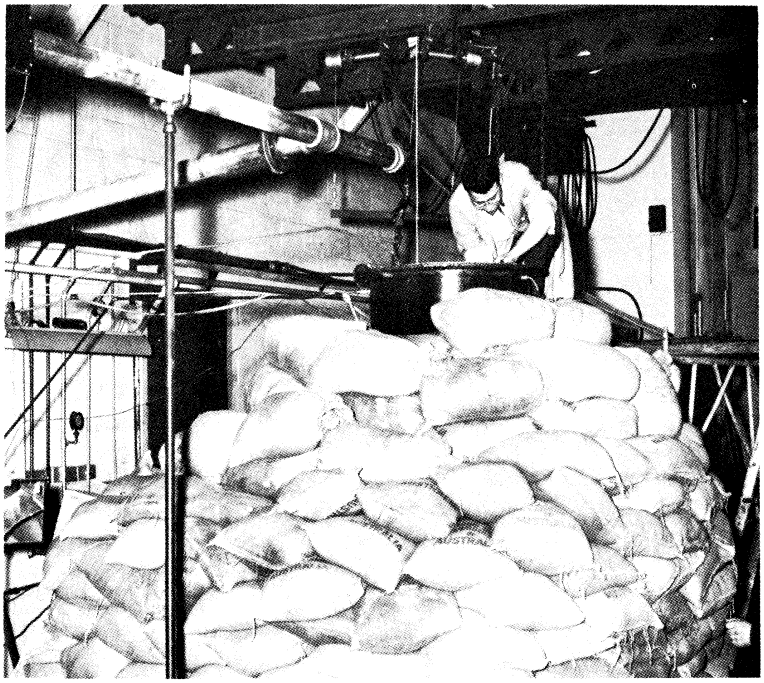


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Fig. 5. Completed Tank Assembly Showing Shielding and Periscope (Lower Left).

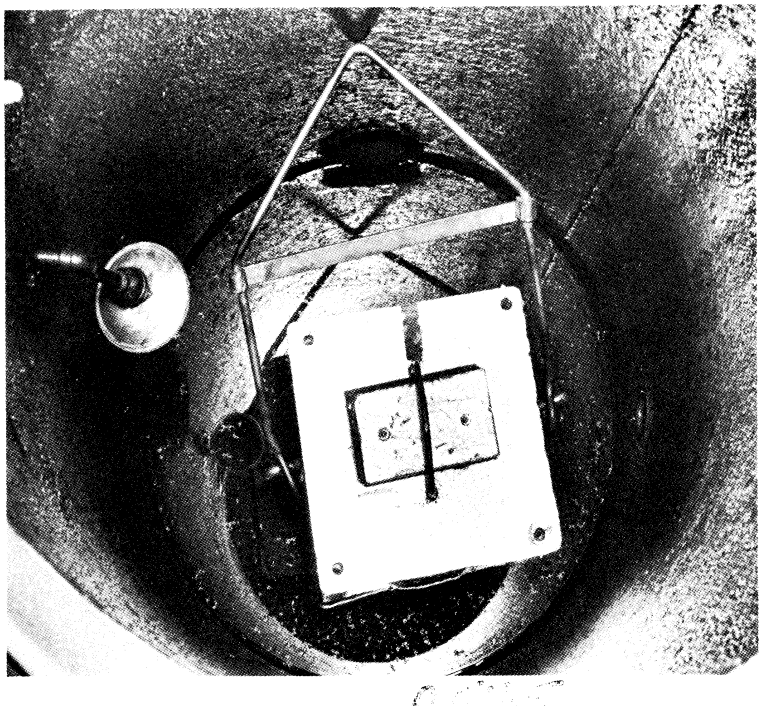


Fig. 6. Shipping Container Under Water Preparatory to Opening.

9517

Fig. 7. Receiving Shipment of Au^{198} at Aircraft Propulsion Lab., Willow Run Airport.

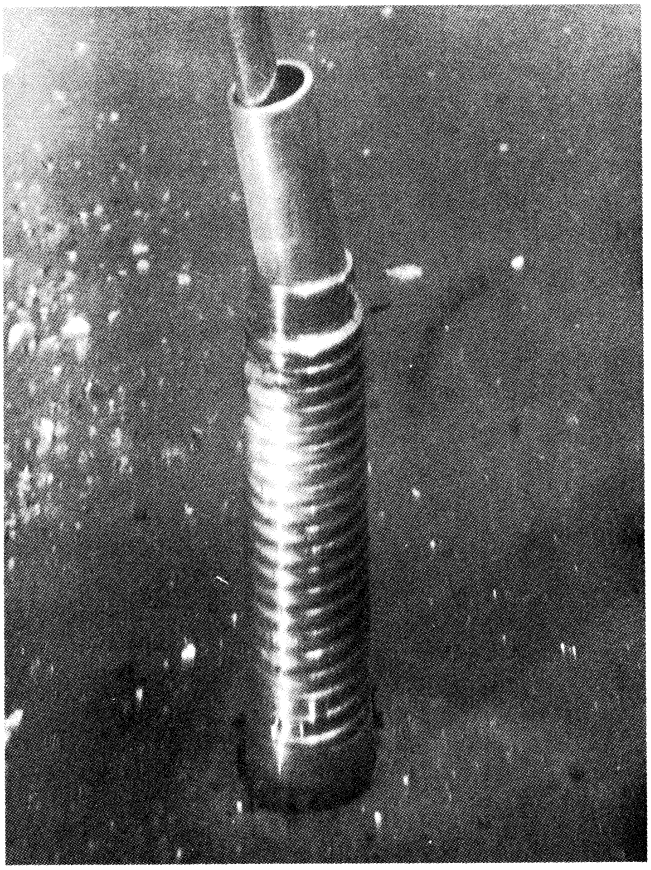


Fig. 8. Source Being Placed in Burner Wall. 45168

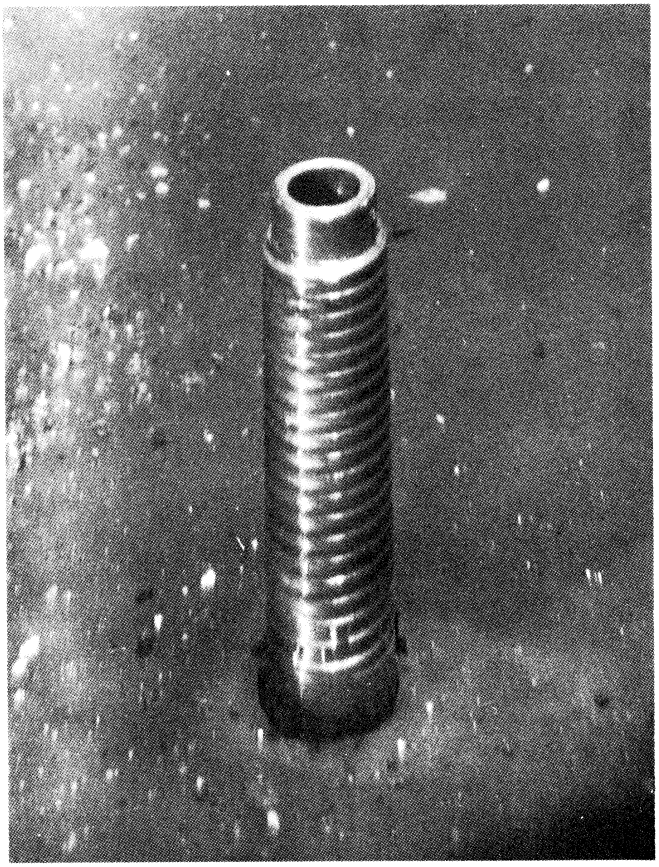


Fig. 9. Source in Place. 45167

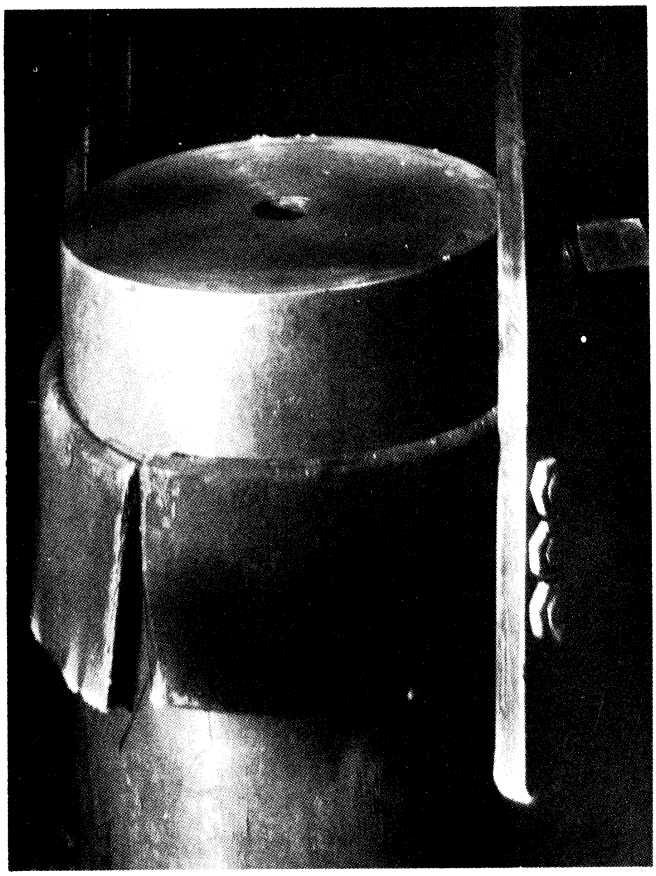


Fig. 10. Tightening Head in Place. 45170

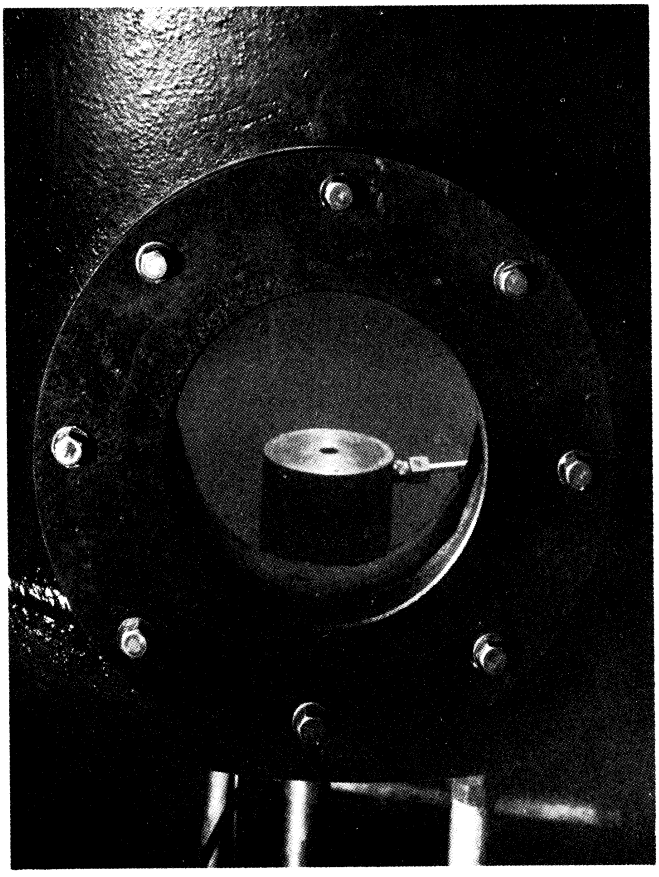


Fig. 11. Completed Burner Assembly. 45171

Fig. 12. Special Tongs for Fastening and Removing Head of Burner.

Assembly of the burner was accomplished without any difficulty. Figures 8, 9, 10, and 11 illustrate the assembly operation. These photographs of a practice run were taken through the observation window. It was not convenient to take pictures of the "hot" assembly, since the radiation field at the exit of the periscope was 150 mr/hr. Figure 8 shows the gold source being inserted into the burner tube, and Fig. 9 shows the source in place. The long-handled tongs (Fig. 12) were used to tighten the head in place (Fig. 10). Figure 11 is a view of the completed assembly. The top head of the tank was bolted in place and the piping connected. The water from the tank was drained into a storage vessel and surveyed for contamination. No evidence of contamination was found and the water was discharged to the sewer.

The entire assembly was surveyed for radiation leakage through the sandbag barrier and except for the 150 mr/hr at the periscope exit, no area exceeded 5 mr/hr. Barriers were erected to prevent access to the periscope exit and an auxiliary mirror was set in place.

Assembly required about 2 hours and proceeded according to plan.

(2) Operation of burner equipment: Controls and instrumentation for the burner were located at some distance from the sandbagged area. Figure 13 shows the relative location of the various areas. Figure 14 is a schematic diagram of this system which was designed to permit operation either with a premixed fuel-air mixture or with fuel and air mixed upstream of the burner in a mixing tee. The flame was ignited by means of a spark plug located upstream of the flameholder. Measurements of pressures and gas flow rates were made by means of instruments located at the control table.

Care was exercised in the planning and installation of the controls and instrumentation to facilitate collection of data. No unusual or difficult techniques in operation were required except when fuel and air were metered separately and mixed at the tee. This case necessitated periodic evacuation and bleeding of the plenum chamber to remove the combustibles which occasionally accumulated. The observation window and periscope assembly permitted visual observation of the flame during ignition and burning. Figure 15 is a photograph of a typical flame.

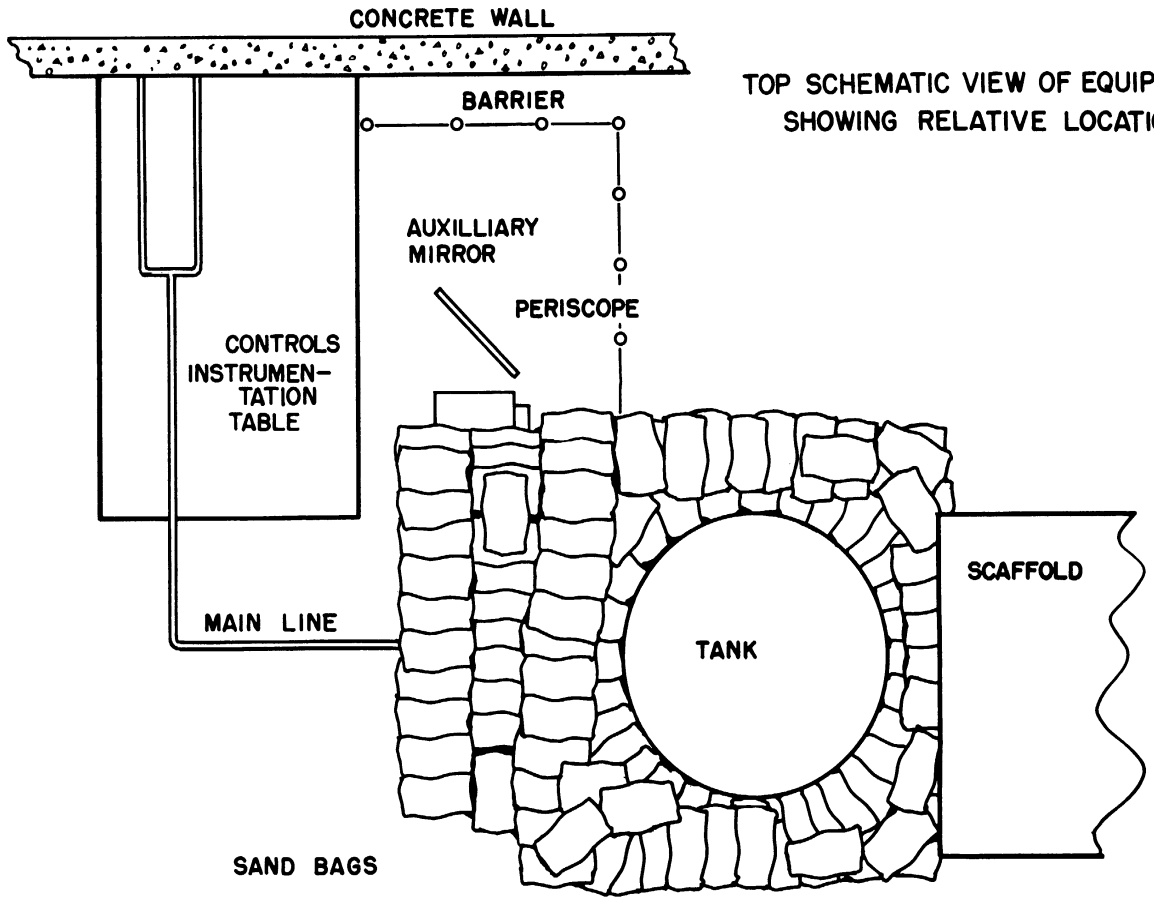


Fig. 13. Top Schematic View of Equipment Showing Relative Locations.

c. Discussion. Since the air specific impulse, S_a, is used to evaluate the performance of this burner, it would be helpful to discuss the significance of this parameter and the method of evaluation.

For a one-dimensional combustion chamber the following equation for conservation of momentum may be written:[2]

$$P_1A + \rho_1AV_1^2 - D_f = P_2A + \rho_2AV_2^2, \qquad (1)$$

where A is area, ρ is density, V is velocity, and D_f is drag, all in absolute units. The subscripts 1 and 2 refer to the positions immediately upstream and downstream of the flame zone (Fig. 16). The units in Equation (1) represent force, and the equation may therefore be rewritten as:

$$F_1 - D_f = F_2 \quad . \qquad (2)$$

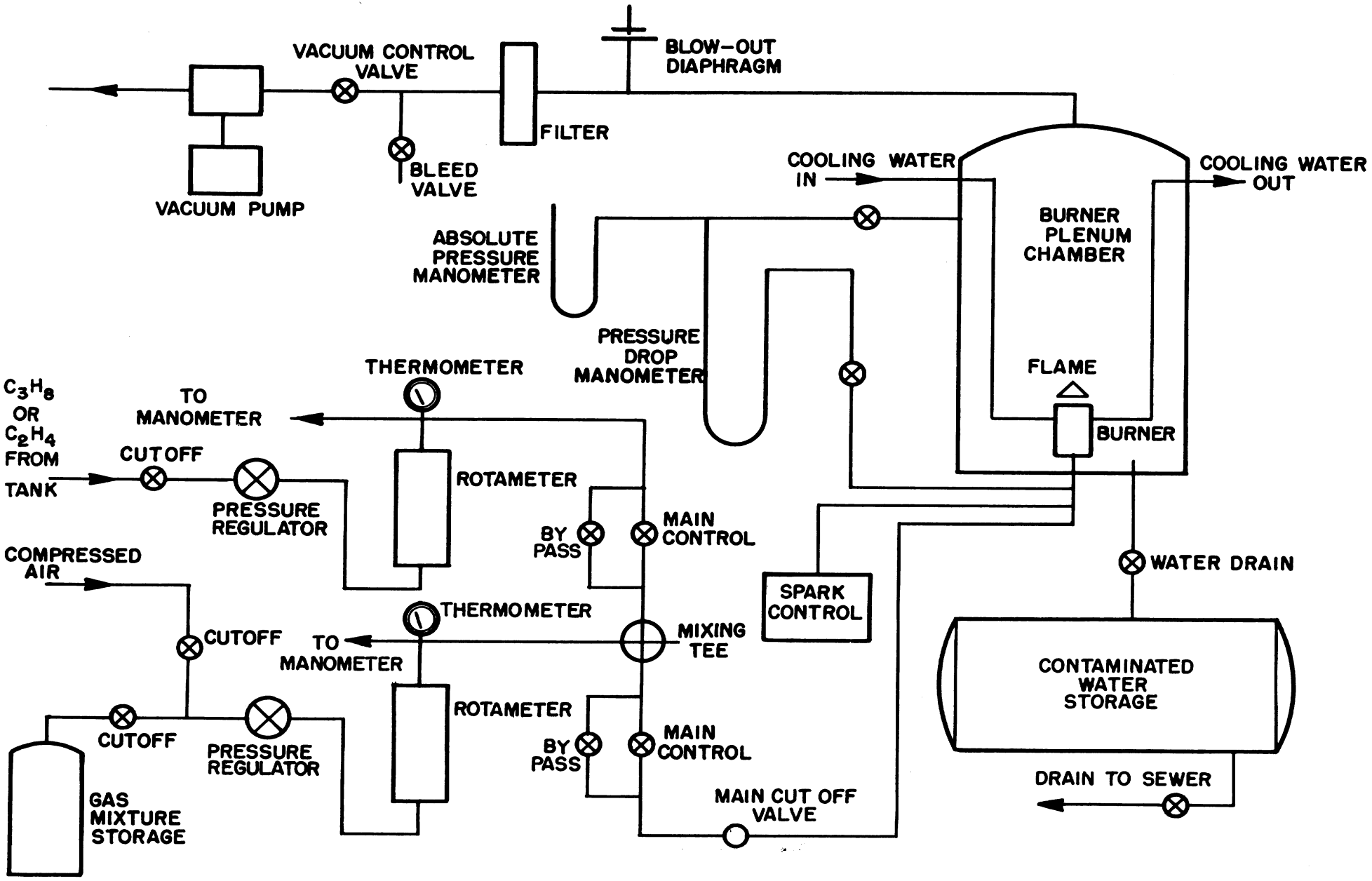


Fig. 14. Schematic Diagram of Burner Equipment and Instrumentation.

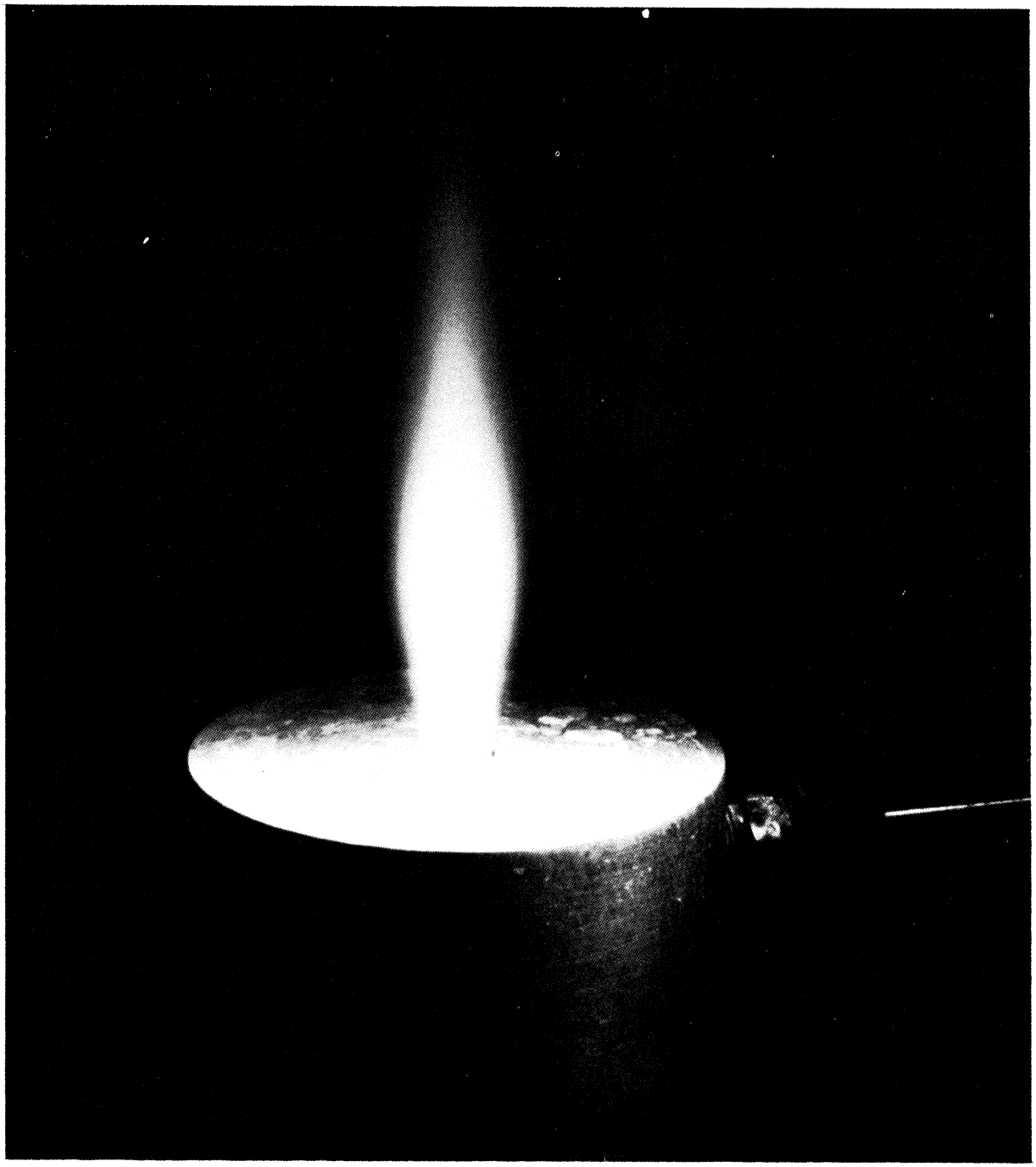


Fig. 15. Typical Hydrocarbon Air Flame. 9512

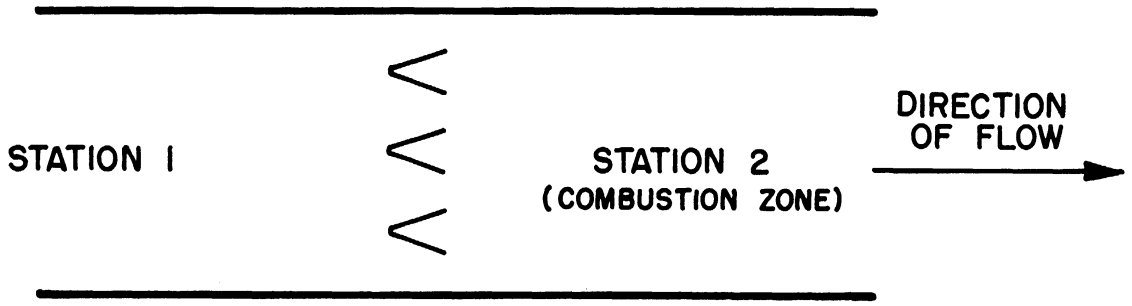


Fig. 16. Flameholder, Heat Addition in One-dimensional Duct.

Introducing the equation of state for an ideal gas, $p = \rho RT$, and the definition of the Mach number

$$M = \frac{V}{C}, \text{ where } C = \sqrt{\gamma g_c RT} \quad , \qquad (3a)$$

and rearranging,

$$\frac{F_2}{W_a} = \left[\frac{C_{s2}(1+f)\sqrt{2(\gamma+1)}}{\gamma g_c}\right]\left[\frac{1 + M_2^2}{M_2\sqrt{2(\gamma+1)}\left(1 + \frac{\gamma-1}{2}M_2^2\right)^{1/2}}\right] \quad , \qquad (3)$$

Where C_{s2} is the speed of sound corresponding to the stagnation temperature at station 2, g_c is gravitational acceleration, f is the fuel-air ratio, and W_a is the air flow rate in lbs/sec. Denoting the first term in the bracket as S_a and the second as $\phi(M)$, Equation (3) may be written as:

$$S_a = \frac{F_2}{W_a\,\phi(M)} \quad . \qquad (4)$$

Equation (4) is the basic equation used in these calculations. The various terms may be determined from the experimental data by the following techniques.

F_2, the stream thrust at the exit of the combustion chamber, is calculated directly from inlet conditions by means of Equation (2). Since ΔP_c, the pressure drop due to cold drag, is proportional to the dynamic head, ρV_1^2, a plot of ΔP_c vs ρV^2 at various conditions was prepared. The design of the flameholder and burner was such that the pressure drop due to drag was given by the equation:

$$\Delta P_c = .006 \ (\rho V^2), \tag{5}$$

which fits the plot of ΔP_c vs ρV^2 to within 1 per cent. The unit of ΔP_c is inches of H_2O and ρV^2 is in lbs/ft^2.

The mass velocity G is obtained from the volumetric flow rate, as measured by rotameters, and W_a is then given by equation

$$W_a = \frac{G_{total} A}{1+f} \tag{6}$$

if premixed fuel-air is used, or by

$$W_a = G_a A$$

if the fuel and air are metered separately.

The final term, $\phi(M)$, is the most difficult to evaluate, since it requires complete description of the state variables at station 2. The pressure at 2 was measured and the other terms, i.e., ρ_2, T_2, and m_2 the molecular weight, are calculated from equilibrium considerations in addition to the perfect-gas and momentum equations. The speed of sound can then be calculated by means of Equation (3a). The function $\phi(M)$ is a measure of the completeness of heat addition to the moving stream.

Since the evaluation of S_a requires a knowledge of the equilibrium data for the combustion products, it was not possible to calculate S_a for C_2H_4-air mixtures. For C_3H_8-air, however, these data were available and S_a was calculated.

The variation of S_a with fuel-air ratio and activity using propane-air mixtures is shown in Figs. 17-21. Examination of these figures shows that the air specific impulse is increased at high levels of activity. The data shown in Figs. 17 and 19 differ from those of Figs. 18 and 20 in both the level of activity and the source location. For the data presented in Figs. 17 and 19 the source was located immediately upstream of the flameholder and for the data of Figs. 18 and 20 it was immediately downstream of the flameholder. This corresponds essentially to irradiation of the precombustion zone in the former case and to the irradiation of the reaction zone in the latter case. Since the radiation is emitted radially, there is no sharp line of demarcation and in both cases there is overlap of the radiation field in all zones of the burner.

It is apparent from examination of Figs. 17-21 that the change in S_a due to radiation depends on several factors. It is apparent that as the mass velocity, G, increases, the magnitude of the change in S_a decreases, with no

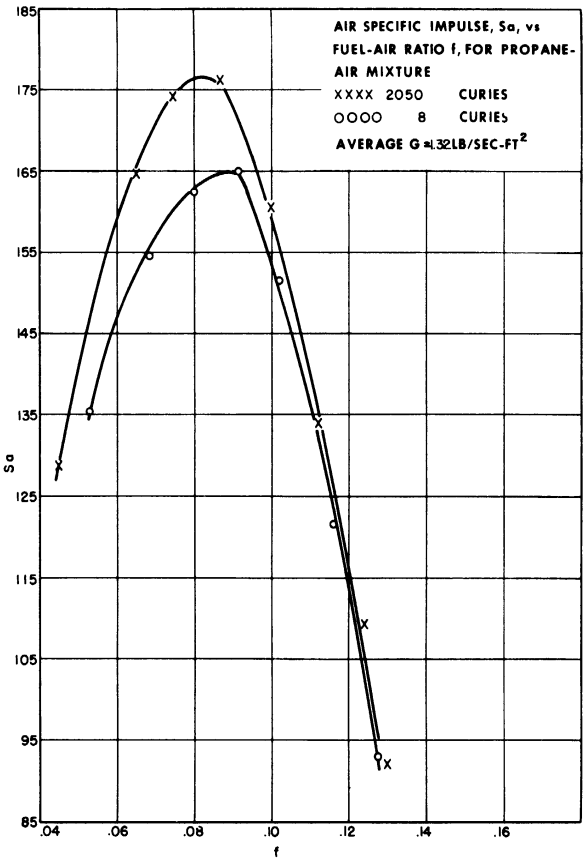


Fig. 17. Air Specific Impulse, Sa, vs Fuel-Air Ratio f, for Propane-Air Mixture.

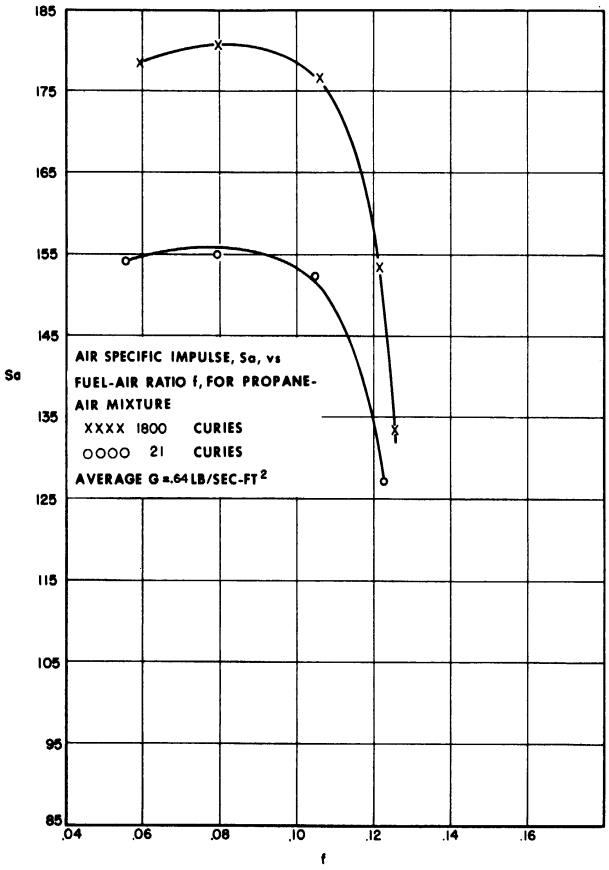


Fig. 18. Air Specific Impulse, Sa, vs Fuel-Air Ratio f, for Propane-Air Mixture.

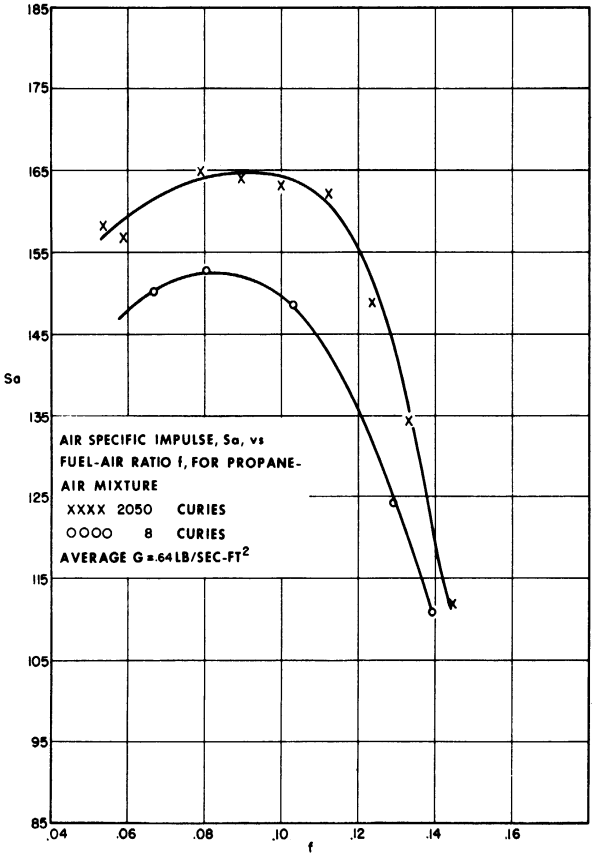


Fig. 19. Air Specific Impulse, Sa, vs Fuel-Air Ratio f, for Propane-Air Mixture.

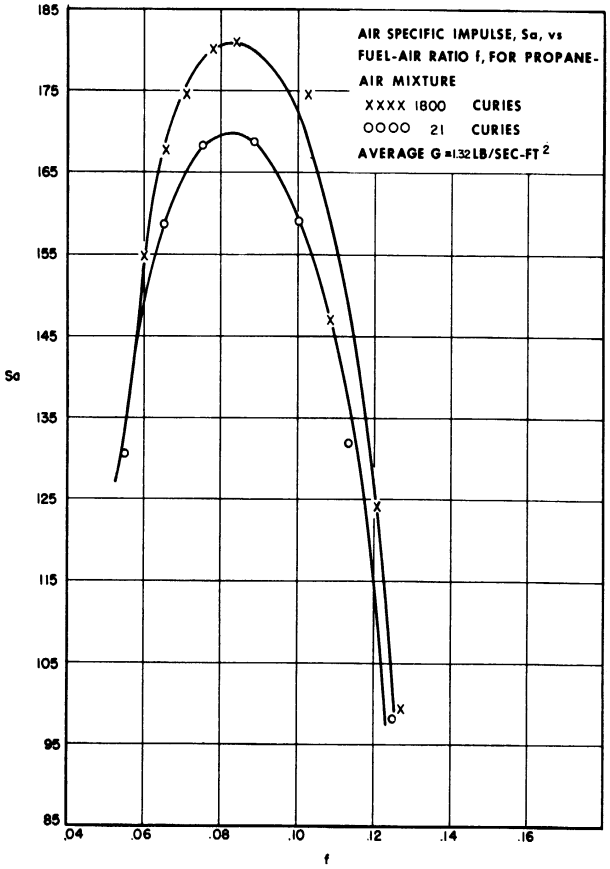


Fig. 20. Air Specific Impulse, Sa, vs Fuel-Air Ratio f, for Propane-Air Mixture.

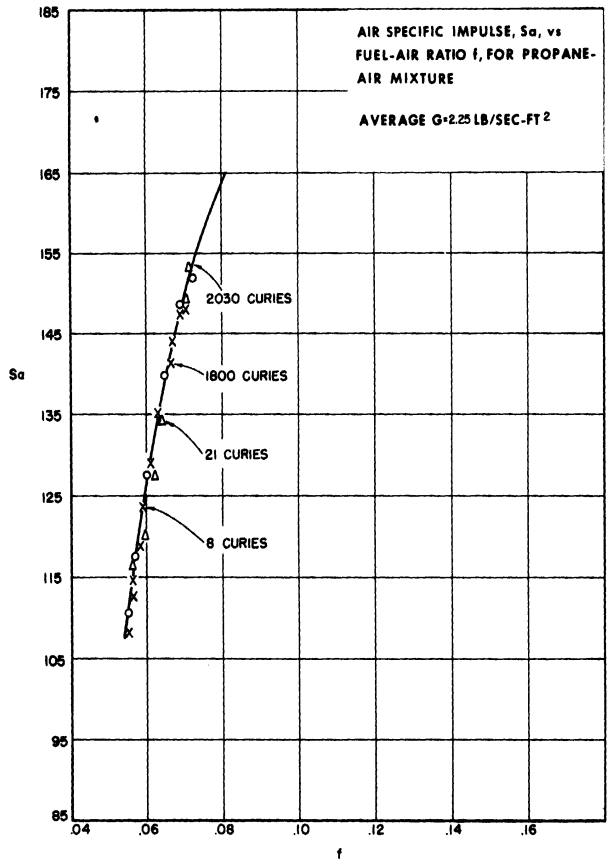


Fig. 21. Air Specific Impulse, Sa, vs Fuel-Air Ratio f, for Propane-Air Mixture.

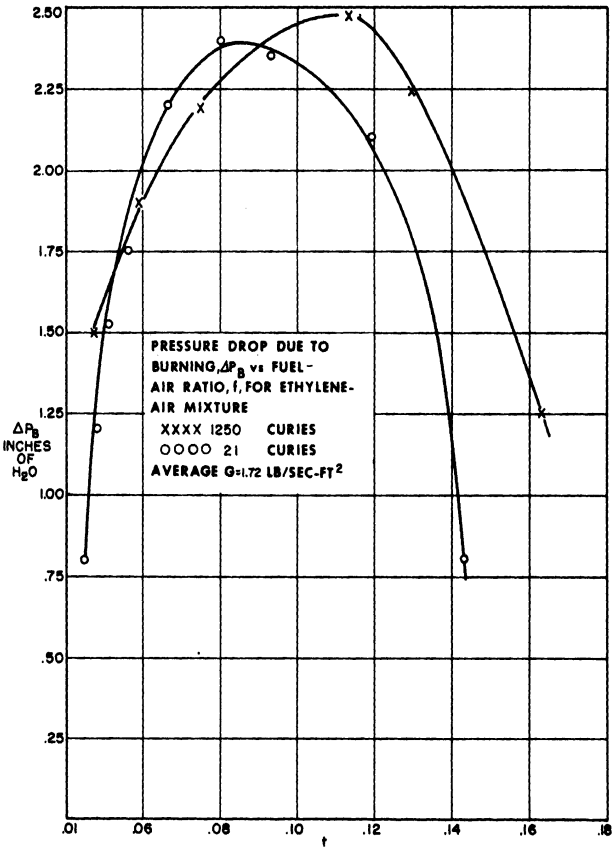


Fig. 22. Pressure Drop Due to Burning, ΔP_B vs Fuel-Air Ratio f, for Ethylene-Air Mixture.

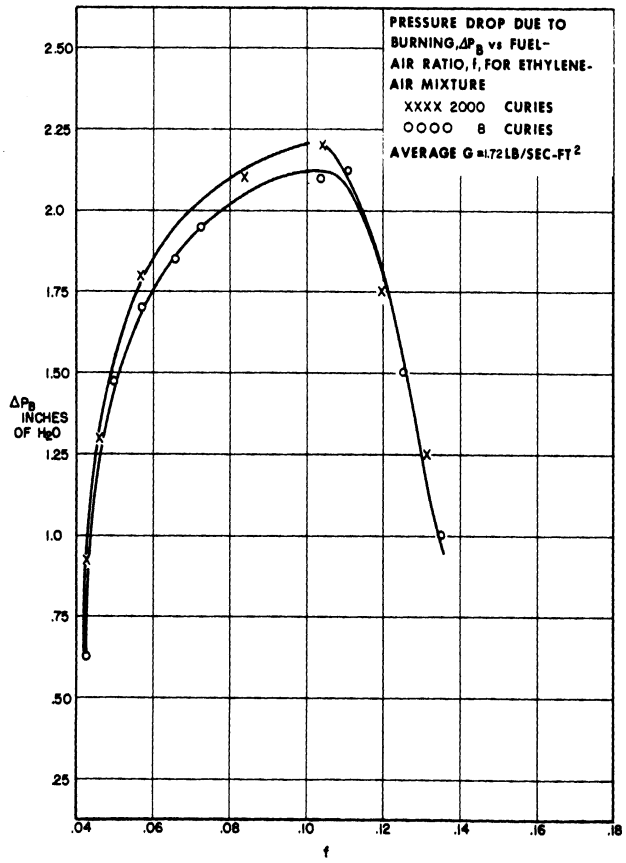


Fig. 23. Pressure Drop Due to Burning, ΔP_B vs Fuel-Air Ratio f, for Ethylene-Air Mixture.

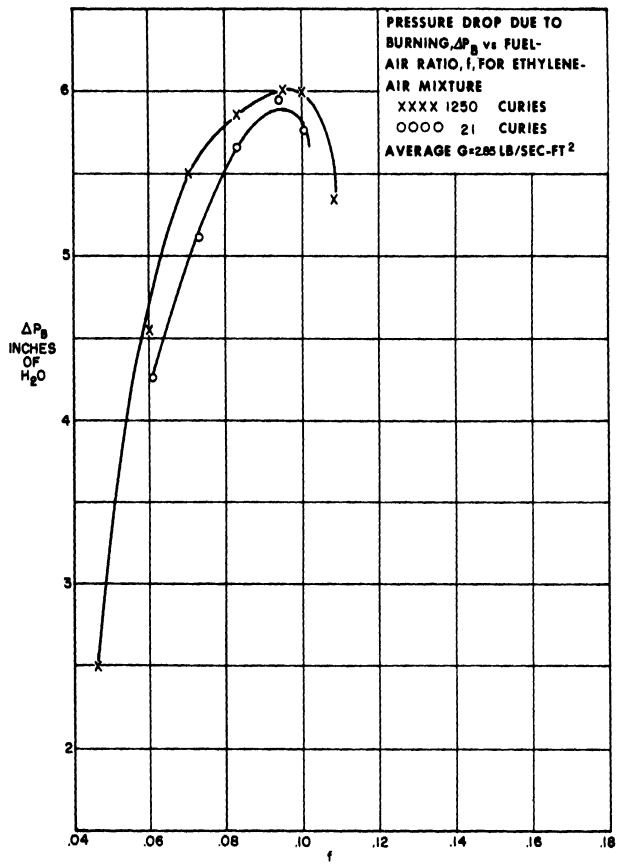


Fig. 24. Pressure Drop Due to Burning, ΔP_B vs Fuel-Air Ratio f, for Ethylene-Air Mixture.

apparent change occurring at the highest mass velocity (Fig. 21). The fact that the data in Fig. 21 does not peak is due to blow off by an acoustic effect, which is a function of the system.

Position of the source evidently has little or no effect, as may be determined by comparing Figs. 17 and 18 with Figs. 19 and 20.

In view of the data presented in Figs. 17-21, it can be stated that for a given fuel, high levels of radiation increase the air specific impulse. The magnitude of the effect depends on:

(1) mass velocity,

(2) fuel-air ratio, and

(3) source activity.

These data are for 1 atmosphere ambient pressure.

The limited data collected show that the rich and lean limits of blow-off are slightly increased under the influence of radiation.

In addition to the data collected at variable fuel-air ratios, considerable data were obtained at the fuel-air ratio for maximum flame speed (0.067). The mass velocities for these runs varied from G = 0.75 lbs/sec-ft^2 to G = 5 lbs/sec-ft^2. No appreciable change was found in the pressure drop due to burning at this fuel-air ratio. This is primarily due to an unfortunate choice of fuel-air ratio and mass velocity for operation, since above G = 2 lbs/sec-ft^2, at f = 0.067, any effect of the radiation is slight (cf. Fig. 21).

The data shown in Figs. 22-27 were obtained from runs using ethylene-air as the combustible mixture.

For the runs made at a source activity of 2000 curies, the source was located upstream of the flameholder; for the runs at 1250 curies, the source was downstream of the flameholder. The values of S_a were not determined for an ethylene-air mixture for the reasons given above. However, since the pressure drop due to burning, ΔP_B, is also a measure of the degree of completeness of reaction, these data are presented as ΔP_B vs fuel-air ratio for different source activities. For C_2H_4-air, the position of the source appears to have little effect.

The experimental data obtained with ethylene-air flames appear to be quite similar to the data obtained with propane-air flames.

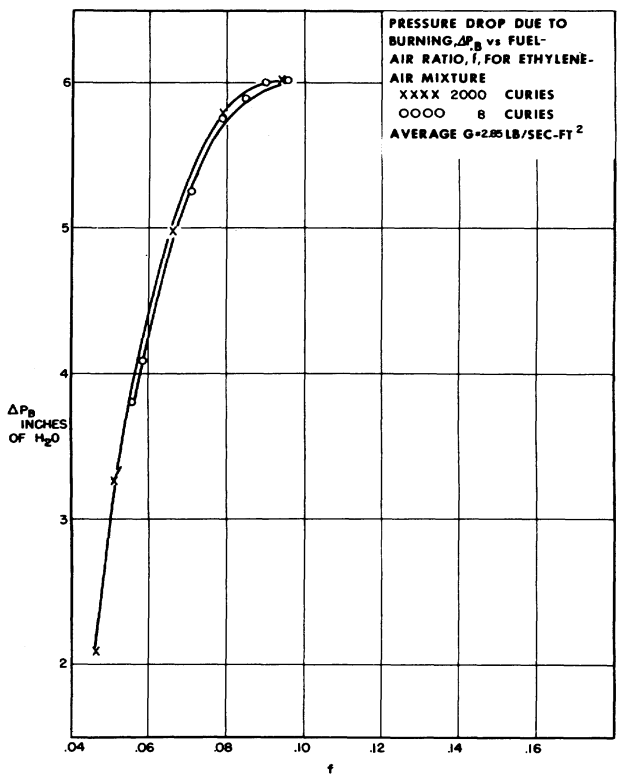


Fig. 25. Pressure Drop Due to Burning, ΔP_B vs Fuel-Air Ratio f, for Ethylene-Air Mixture.

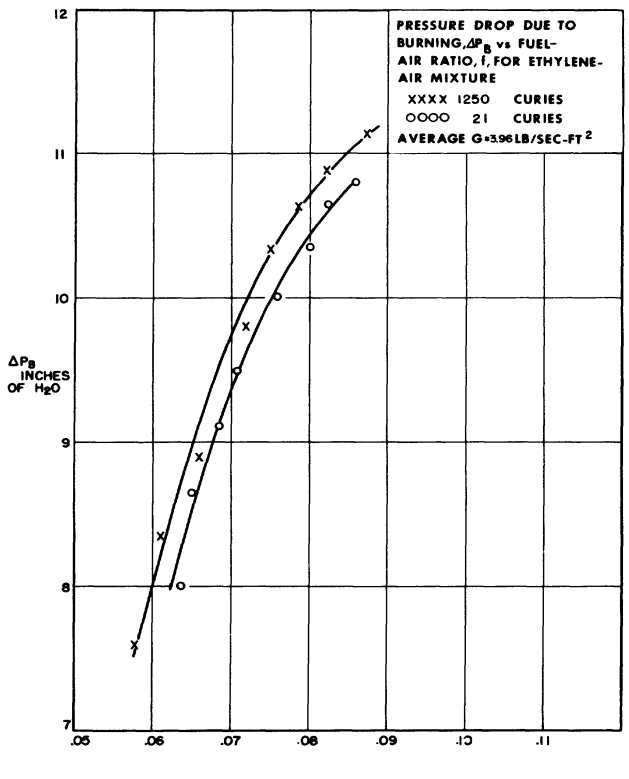


Fig. 26. Pressure Drop Due to Burning, ΔP_B vs Fuel-Air Ratio f, for Ethylene-Air Mixture.

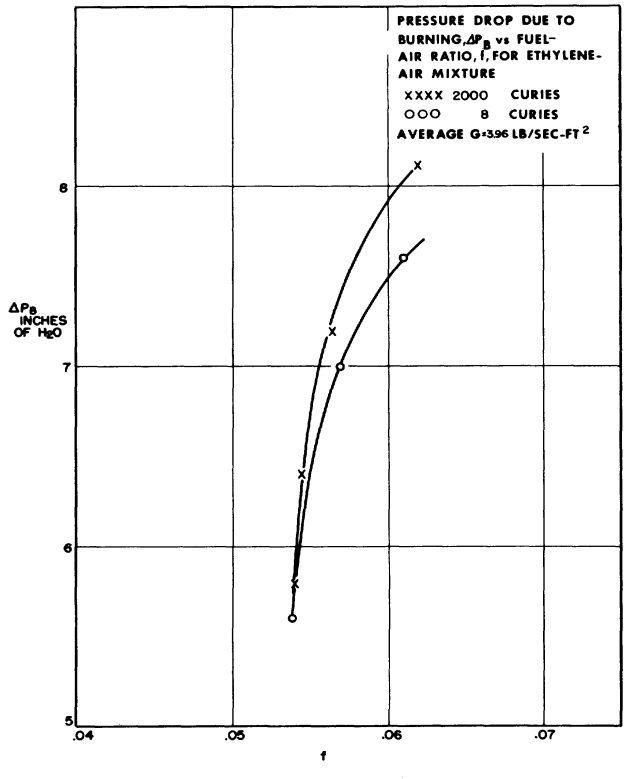


Fig. 27. Pressure Drop Due to Burning, ΔP_B vs Fuel-Air Ratio f, for Ethylene-Air Mixture.

One notable difference, however, is the greater mass velocity at which the effect is still noticed with the C_2H_4-air as compared to the mixtures of C_3H_8-air. This is not entirely unexpected, as the double bond is generally more susceptible to excitation than the single bond.

The properties of flames are functions of the geometry of the combustion chamber, and any results obtained are applicable only to the system investigated.

d. Conclusions. The efficiency of combustion in drag-stabilized flames may be increased by the presence of high levels of beta radiation. Such changes are influenced by the type of fuel, mass velocity, source activity and fuel-air ratios. Rich and lean blow-off limits are also slightly improved.

This work is exploratory; a great deal of additional basic data must necessarily be collected before a clearer understanding of the behavior of combustion in the presence of radioactive sources may be developed.

e. Acknowledgements. The authors wish to acknowledge the financial assistance of the Michigan Memorial-Phoenix Project in providing funds to complete this work. The United States Air Force, 10th Air Force, 2242nd Reserve Combat Training Center, provided the airlift to transport the source from Idaho Falls to Willow Run.

f. References.

1. General Electric Corp., "Chart of the Nuclides", Schenectady, N. Y., 1950.

2. Progress Report 4 (COO-124), Eng. Res. Inst. Proj. M943, Univ. of Mich., Ann Arbor, Mich., March, 1953.

PART II. SUBPROJECT M943-4, THE EFFECT OF RADIATION ON CHEMICAL REACTIONS

Personnel:

Subproject Supervisors: Joseph J. Martin, Associate Professor of Chemical and Metallurgical Engineering; and Leigh C. Anderson, Professor of Chemistry and Chairman of Department.

Research Assistants: David E. Harmer and John G. Lewis Assistants in Research: C. Eckfield; J. Holmes; R. Kinney; and E. Rosen.

A. INTRODUCTION

Reference to previous progress reports will reveal the general nature of the studies being carried on at this laboratory. During the period covered by this report, the activities of the personnel of this subproject have not been confined strictly to determining experimentally, the effect of ionizing radiation on the promotion of chemical reactions. Consequently, the work is being reported under four major topic headings as follows: Chemical Reactions, Thermodynamics and Kinetics of Reactions, Dosimetry Problems, and Equipment Changes. These topics will now be considered in that order.

B. CHEMICAL REACTIONS

1. Polymerization

The polymerization of ethylene at pressures as high as 1600 psi and temperatures up to 400°F has received considerable attention. As shown in Table II, the early runs showed fairly high yields of a fluffy white powder. This was true for the runs in which the ethylene and a small amount of acetone were irradiated and for the runs in which ethylene alone was irradiated. The acetone was added with the idea that it would release free radicals on radiolysis and initiate chain polymerization of the ethylene. Later runs have

TABLE II

IRRADIATION OF ETHYLENE WITH COBALT-60 GAMMA RAYS

Page	Starting Date	Pressure psig	Ave. Pres., psia	Ave. Temp. °F	Grams Polymer	Hours Irrad.	Grams Polymer Per Hr.	Other Reactants	Source	Position of Base of Reactor
132113	19 Dec 52	785-830	822	69	1.6+	69.7	0.02+	1 ml acetone	1-kc	-
132118	12 Jan 53	795-820	822	70	0.666	61	0.011	-	1-kc	-
132147	3 Mar 53	1330-1430	1395	230	0.292	17.2	0.017	-	10-kc	8" NE 9" UP
132149-II	7 Mar 53	700-820 424-435	775 445	50	0.004	0.9 15. 15.9	0.0003	-	10-kc	8" NE 9" UP
132159	26 Nov 52	780-790	800	68	0.394	42	0.009	1 ml acetone	1-kc	-
132250	10 Mar 53	715-810	777	50	0.167	16.2	0.010	-	10-kc	GL on GL at Base
132252	11 Mar 53	1330-1430	1395	200	1.04	16.1	0.06	-	10-kc	Do.
132253	12 Mar 53	692-698	710	45	1.48	87.7	0.017	-	10-kc	8" NE 9" UP
132254	17 Mar 53	655	670	45	-	23.3	-	650 psi NA 15 psi Air Vent, Chg. C_2H_4	10-kc	8" NE 9" UP
132255	18 Mar 53	710-800	770	45	0.088	21.5	0.004	-	10-kc	8" NE 9" UP
132256	19 Mar 53	760-840	815	420	0.381	15.3	0.025	-	10-kc	8" NE 9" UP

132258	23 Mar 53	70-810	455	45	0.010	39.	0.0003	10 ml acetone	10-kc	-
132259	26 Mar 53	183-788	485	61	-	15.3	-	-	10-kc	GL on GL at Base
132262-I	4 May 53	1425-1600	1528	72	-	21.9	-	-	1-kc	-
132263-II	5 May 53	595-620	623	72	0.022	23.	0.001	-	1-kc	-
132264	7 May 53	600-615	622	69	0.0001	20.7	0.000,005	100 ml O_2	1-kc	-
132265	8 May 53	608-620	631	67	0.0001	21.4	0.000,005	25 ml O_2	1-kc	-
132266	9 May 53	577-594	601	72	0.034	25.6	0.0013	5.0 ml O_2	1-kc	-
132267	10 May 53	595-600	603	75	0.001	17.0	0.000,05	2.0 ml O_2	1-kc	-
132268	11 May 53	597-617	622	70	0.017	22.3	0.0008	10.0 ml O_2	1-kc	-
132269	12 May 53	590-605	612	71	0.030	21.1	0.0014	10.0 ml O_2	1-kc	-
132270	13 May 53	629-637	638	66	0.016	14.8	0.0011	5.0 ml O_2	1-kc	-
132275	4 Jun 53	525-550	553	70	0.031	16.8	0.002	-	10-kc	8" sw 13-7/8" UP

produced very erratic results, and as noted in Table II, the yields have fallen considerably. At first it was thought this might be caused by the presence of oxygen in the system, and therefore a number of runs were made in which a known amount of oxygen was introduced. Although the results of these runs showed that the extent of reaction is dependent on the amount of oxygen, as shown in Fig. 28, the effect was far smaller than the decrease in reactivity from the early runs to the later runs. See Figs. 29 and 30. Therefore, it seemed unlikely that oxygen alone could be responsible for inhibiting the reaction. It should be noted that some uncertainty exists as to the slope of the curve of Fig. 28 for oxygen content near zero.

Other ideas which have been suggested are that the cylinder of ethylene contained impurities which acted either as promoters for the early runs or as inhibitors for the later runs. Alternative possibilities are that there might have been some polyethylene powder in the original cylinder of ethylene, that impurities might have been introduced by leakage during evacuation, that impurities might have been adsorbed on the packing or sealing washers, that acetone might have been present in the early runs even though it was not intentionally added except for three runs, and that some kind of separation reaction had taken place in the feed cylinder which resulted in different products over a period of time. These various ideas are being investigated and it is hoped to have a more definite information soon on just what causes ethylene to polymerize and what inhibits that polymerization in the system under investigation.

It should also be mentioned that the analysis for polymer after irradiation has been very crude. The procedure involves bleeding off all the gas, opening the reactor, and scraping out the fluffy powder. Analyses of the bleed-off gas are contemplated for future runs, as well as molecular-weight determinations on the polymer that is formed.

Liquid propylene was irradiated for 67.6 hours at a dose rate of about 25,000 rep/hr; the results are reported in Table III. A small quantity of a

TABLE III

IRRADIATION OF PROPYLENE IN 1-KC COBALT-60 GAMMA SOURCE

Date	Starting Date	Time	Hours Irrad.	Dose Rate, Rep/hr	Dose, Rep	Temp., °F	Pres., psia	Remarks
132140	6 Feb 53	1735	0.	65,000 ± 10%	0.	71.	147	2.8-in.-depth liq. propylene in bomb; 3000-lb gauge.
132140	9 Feb 53	1313	67.6	65,000 ± 10%	4,400,000.	65.5	145	Out of vault.
132140	18 Feb 53	1000	--	--	--	Room	25-30	0.2-0.3-in.-depth liquid in bottom, quickly evaporated.

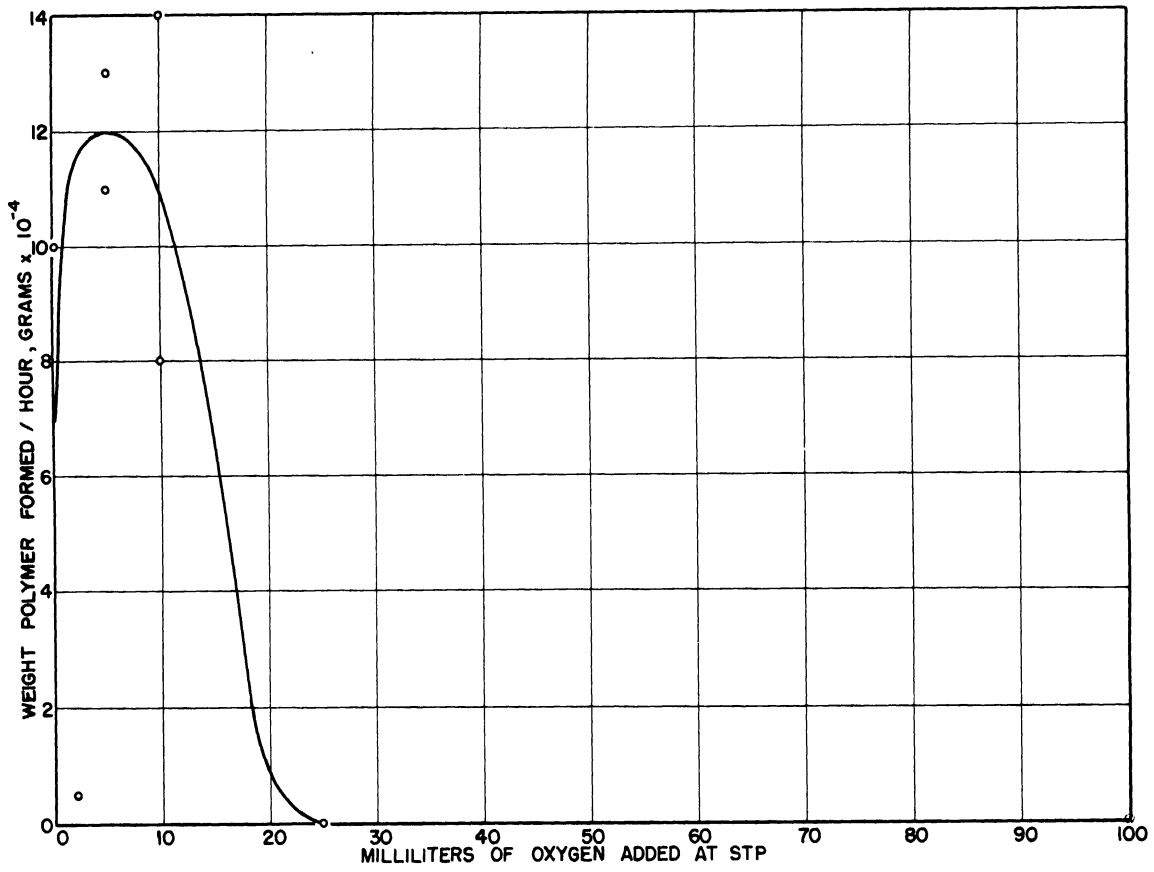


Fig. 28. Effect of Oxygen on Rate of Ethylene Polymerization.

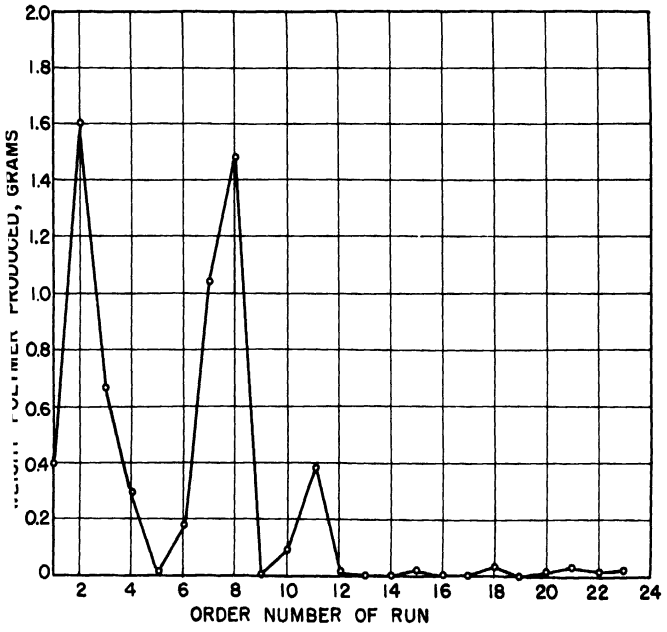


Fig. 29. Ethylene Yields for Successive Runs.

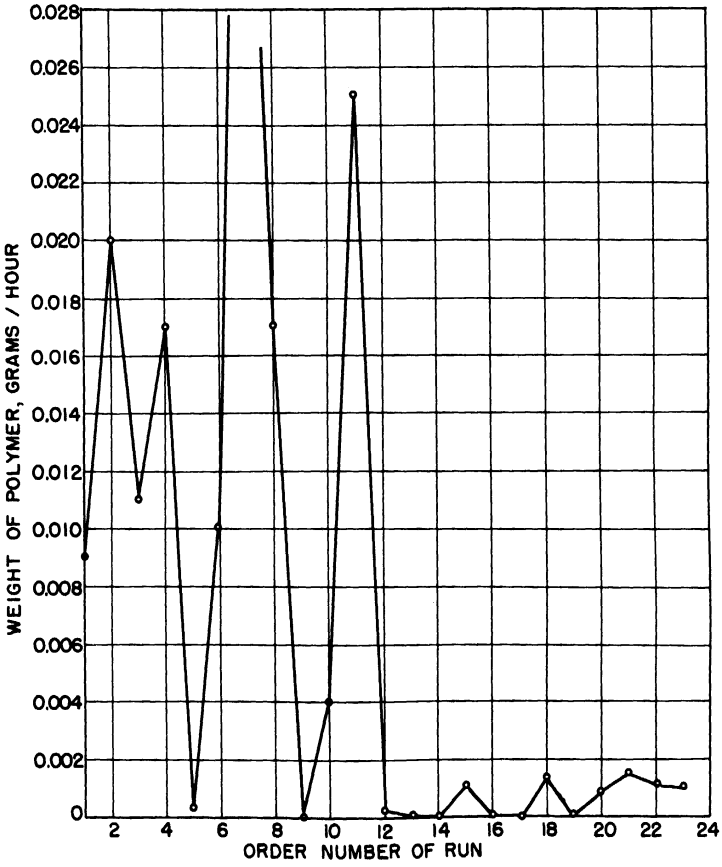


Fig. 30. Rate of Ethylene Production for Successive Runs.

volatile liquid was found in the reactor when it was vented and opened as in the ethylene runs. Undoubtedly some polymerization took place, although the quantitative amount has not yet been determined.

2. Chlorination

The addition of chlorine to benzene under gamma radiation to form the addition compound, benzenehexachloride, has been reported by this laboratory previously. The speed and extent of this reaction have led to investigations of the chlorination of other hydrocarbons such as toluene. Also, further work is anticipated on the kinetics of the benzene chlorination under gamma radiation to compare it with the reaction induced by ultraviolet radiation.

The reaction of chlorine with toluene has been carried out at 20°C. The experimental runs consisted of a 10-minute nitrogen purge through the toluene and the entire system, followed by passing an excess of chlorine gas through the toluene for 30 minutes during irradiation. After this, nitrogen was again passed into the system for 10 minutes, in order to allow the apparatus to be disconnected without escape of chlorine gas into the room. Equipment for the runs was identical to that used for benzene runs described in Progress Report 4[1].

An attempt was made to separate the products of this reaction by distillation at atmospheric pressure. Fractions were obtained up to 225°C, but a vigorous decomposition began at 170°C with evolution of hydrogen chloride. About 30 per cent of the total chloride content of the sample was given off in this decomposition. When the original sample was extracted with water, it was found that less than 1 per cent of the total chloride passed into the aqueous layer, indicating that the chloride was not present in an active or ionic form.

A distillation of the products of another toluene chlorination run was carried out in a column packed with glass helices and operated at a pressure of 4.55 cm Hg. Table IV summarizes the fractions and some of their properties. The organic chloride analyses were carried out by the sodium diphenyl reagent method. Table IV shows that the largest single fraction (excepting the recovered starting material) is that boiling between 206 and 216°C (fraction X). This fraction has a chlorine content which is remarkably close to the theoretical percentage for a hexachloro addition product of toluene. Because of the possibilities which might be realized from the production of a new type of toluene derivative, further investigations are being made.

A control run was made on the reaction of chlorine and toluene in the absence of gamma radiation. All conditions were the same as for the runs previously described. Table V summarizes the fractions obtained from the distillation of the reaction product mixture at 4.50 cm Hg pressure. This control run

TABLE IV

DISTILLATION AT 4.55 CM HG PRESSURE OF THE PRODUCTS OF A TOLUENE CHLORINATION RUN WITH GAMMA IRRADIATION

Fraction	Volume, ml	Temperature Range, °C	Per Cent Chlorine	Comment
I	28.5	35-36°		Unreacted toluene
II and III	8.9	62-104°		
IV	5.6	95-96°	26.5	Mostly benzyl chloride (pure benzyl chloride is 28% Cl)
V to VII	6.2	96-127°		
VIII	2.8	142-157°	56.2	Mostly benzo trichloride (pure is 54.4% Cl)
IX	4.6	157-180°		
X	15.0	206-216°	68.8	Probably the hexachloro addition product. Theoretical chlorine content is 69.8%.
XI	1.0	216-222°		Slight decomposition
Residue	12.0		70	

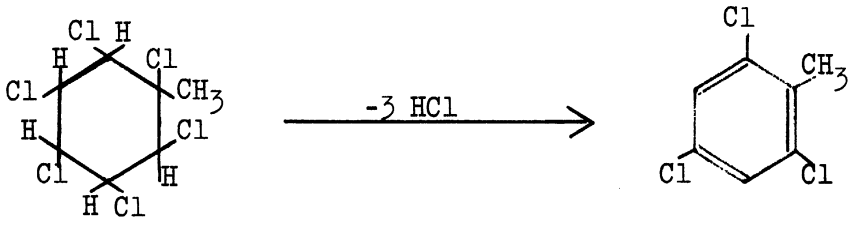
shows that very little reaction takes place in the absence of gamma radiation. Experiments which will allow comparison of the effectiveness and nature of products obtained under gamma irradiation and ultraviolet irradiation are contemplated.

TABLE V

DISTILLATION AT 4.50 CM HG PRESSURE OF THE
PRODUCTS OF A TOLUENE CHLORINATION RUN WITHOUT IRRADIATION

Fraction	Volume	Temperature Range, °C	Per Cent Chlorine	Comment
I	85.5	34°		Toluene (identified by odor)
II	1.9	80-87°	25	Mostly benzyl chloride or chlorotoluene (theoretically 28% Cl)
Residue	4.1		68	High chlorine content, but dissimilar appearance to fraction X of Table IV.

Since fraction X of Table IV appeared to be somewhat unique, it was investigated further. If the structure is assumed to be the hexachloro addition compound of toluene, it should be possible to remove three molecules of hydrogen chloride and obtain 2,4,6-trichlorotoluene, or some corresponding isomer, by procedures similar to those which yield trichlorobenzene from hexachlorocyclohexane.



The thermal decompositions observed during distillation might lead to this type of compound in addition to other products. A sample of fraction X from Table IV was refluxed with pyridine for 63 hours. Aliquot portions of the pyridine mixture were placed in a mixture of equal volumes of benzene and ether and extracted with 0.1 N sodium hydroxide. The alkaline extracts were analyzed for ionic chloride. Of the total chlorine content of the original fraction, 52.8 per cent was found in this aqueous layer. This fact indicates that the hexachloro addition product of toluene was probably present in the original

material and suffered elimination of three molecules of hydrogen chloride while being teated with pyridine. The proof of structure is being given further consideration.

In an effort to ascertain whether the addition reaction could be applied to other substituted aromatic systems, benzyl chloride, benzal chloride, and benzotrichloride were each treated with chlorine under gamma irradiation at 20°C for 30 minutes. In each case, practically no increase in chlorine content was found and the starting material remained virtually unchanged.

Because of the nearly complete nonreactivity of the side-chain-substituted chlorine compounds of toluene, it seemed valuable to ascertain the effect their presence would have on the rate of reaction of chlorine with benzene under gamma irradiation. A series of runs was made in which mixtures of benzene and benzyl chloride were treated with chlorine under gamma irradiation for a 15-minute interval, at the end of which time the solid benzene hexachloride was separated and weighed. Table VI gives the results of these runs.

TABLE VI

EFFECT OF THE PRESENCE OF BENZYL CHLORIDE ON THE REACTION BETWEEN BENZENE AND CHLORINE

Mole Per Cent of Benzyl Chloride	Per Cent Yield of Benzene Hexachloride in 15 Minutes	Temperature Rise over Control Point
5	0.72	None observed
4	0.01	None observed
2	3.6	None observed
1	5.7	10°F after 13 minutes
0.5	5.6	22°F after 9 minutes
0.5		20°F after 10 minutes

The above table shows that the presence of benzyl chloride has a pronounced effect on the rate of addition of chlorine to benzene. The exact mathematical character of the function is not satisfactorily indicated by the data because of the experimental difficulties which arose at the minimum and maximum concentrations. The time for running the experiments was set at 15-minutes because previous work in this laboratory has shown that solutions of benzene

TABLE VII

IRRADIATION OF OXYGEN-BENZENE
MIXTURES IN 1-KC COBALT-60 GAMMA SOURCE AT 1 ATM ABS AND ROOM TEMPERATURE

Page	Starting Date	Time	Hours Irrad.	Dose Rate Rep/hr	Dose, Rep	Procedure	Observations
132121	17 Jan 53	1615	0	65,000±10%	0	100 ml Benzene into vault	
132121	17 Jan 53	1626	0.18		12,000.	Oxygen on	
132121	17 Jan 53	1705	0.83		54,000.	Oxygen off	
132121	17 Jan 53	1707	0.87		57,000.	Out of vault	No changes in temperature or tail gas rate.
132122	20 Jan 53	-	-	-	-	5 ml of 132121 Product with sat'd. Bromine water, Evaporate on watch glass	No visible Ppt. in liquid. Cloudiness in benzene layer.
132122	21 Jan 53	-	-	-	-		Small oily smear on water glass, smelled of halo-genated hydro-carbons.

diluted with carbon tetrachloride reacted almost completely in this length of time. Although great care was taken to control the temperature of the reaction by adequate cooling, some of the reactions displayed a tendency to "run away" after an initial period of good temperature control; this effect was more marked at lower concentrations of benzyl chloride. In the last run reported in Table VI, the mixture heated up so rapidly that chlorine escaped and no quantitative yield could be ascertained.

3. Oxidations

Several attempts have been made to oxidize partially some aromatic compounds by procedures analagous to those used in chlorinations runs.

Pure oxygen was bubbled through benzene for 30 minutes while the mixture was being subjected to gamma irradiation at the rate of 50,000 rep/hr. The results are reported in Table VII. No changes in temperature or tail-gas flow rate were noted during irradiation, no changes were noted in the color or odor of the product, and no visible reaction occurred between a sample of the product and saturated bromine water.

An attempt to oxidize toluene as reported in Table VIII, was carried out in a manner similar to that just described for benzene. The same equipment and dose rate were used and oxygen was bubbled through the toluene at 68°F for 1/2 hour. No changes were noted in temperature, tail-gas flow rate, color, or odor of the reaction product.

TABLE VIII

IRRADIATION OF OXYGEN-TOLUENE
MIXTURES IN 1-KC COBALT-60 GAMMA SOURCE AT 1 ATM ABS and 68°F

Page	Starting Date	Time	Hours Irrad.	Dose Rate Rep/hr	Dose, Rep	Procedure	Observations
132126	31 Jan 53	1708	0.	65,000±10%	0.	100 ml Toluene into source.	
132126	31 Jan 53	1712	0.07	65,000±10%	4,500.	Oxygen on	
132126	31 Jan 53	1732	0.40	65,000±10%	25,000.	Oxygen off	
132126	31 Jan 53	1735	0.45	65,000±10%	30,000.	Toluene out of vault	No noticeable change in toluene

TABLE IX

IRRADIATION OF POTASSIUM PERMANGANATE - TOLUENE
MIXTURES IN 1-kc COBALT-60 GAMMA SOURCE AT 1 ATM ABS and ROOM TEMPERATURE

Page	Starting Date	Time	Hours Irrad.	Dose Rate, Rep/hr	Dose, Rep	Procedure	Observations
132125	29 Jan 53	1056	0	65,000±10%	0.	0.5g permanganate 25 ml acetone, 25 ml toluene	
132125	29 Jan 53	1400	3.1	65,000±10%	200,000.	inspect sample	Supernatant liquid clear pink when shaken.
132125	29 Jan 53	1409	3.1	65,000±10%	-	resume irradiation	-
132125	29 Jan 53	1530	4.5	65,000±10%	300,000.	inspect sample	still faint pink
132125	29 Jan 53	1531	4.5	65,000±10%	-	resume irradiation	-
132125	29 Jan 53	1616	5.1	65,000±10%	330,000.	inspect sample	Supernatant liquid clear. Faint pink when shaken. Pungent odor.
132125	29 Jan 53	1627	5.1	65,000±10%	-	resume irradiation	-
132125	30 Jan 53	1519	28.0	65,000±10%	1,800,000.	inspect sample	Supernatant liquid clear and remained clear on shaking; decolorized bromine water; slowly decolorized a solution of permanganate in acetone; did not react visibly with aqueous bromide-bromate. The central sample had also decolorized.

In a different type of oxidation a sample consisting of 25 ml of acetone, 0.5 gram of potassium permanganate, and 25 ml of toluene was irradiated 28 hours at a dose rate of about 50,000 rep/hr, as reported in Table IX. Observations were made at intervals, and it was noted that there was a gradual bleaching of the permanganate color. The unirradiated control sample also bleached, but not as rapidly as the irradiated sample.

A solution of 0.25 gram of iodine in 50 ml of toluene was irradiated 44.5 hours at a dose rate of about 50,000 rep/hr, as reported in Table X. No difference was discernible between the irradiated sample and the unirradiated control.

TABLE X

IRRADIATION OF IODINE-TOLUENE MIXTURE IN 1-KC COBALT-60 GAMMA SOURCE AT 1 ATM ABS AND ROOM TEMPERATURE

Page	Starting Date	Time	Hours Irrad.	Dose Rate, Rep/hr	Dose, Rep	Procedure	Observations
132130	4 Feb 53	2108	44.5	65,000±10%	2,900,000.	0.25 g. iodine dissolved in 50 ml toluene	No discernible difference between run and control kept in dark.

4. A Dosimetry Reaction

Preliminary tests were made of the use of starch-iodide solutions for dose measurements. A solution of 1 gram of potassium iodide and 1 drop of glacial acetic acid in 100 ml of water was prepared together with a 1-per cent water solution of soluble starch. Samples consisting of 1 part of the starch solution to 19 parts of the iodide solution were subjected to doses of 5,000 to 300,000 rep, and the results are reported in Table XI. Visual observations indicated correspondence between dose and depth of color.

Since the above results indicate that an iodide system might be used for dose measurements, application of the method is considered: The possibility of measuring the concentrations of iodine produced in the oxidation of an iodide by means of a concentration potential for the solution of the mixture was discussed in Progress Report 4. Consider the usual voltage equation for an electrode dipping into a solution of iodine and iodide:

$$E = E^\circ - \frac{2.303\,RT}{N\,F}\log_{10}\frac{(\gamma_B C_B)^b}{(\gamma_A C_A)^a}$$

$$= E^\circ - \frac{0.05915}{N}\log_{10}\frac{(\gamma_B C_B)^b}{(\gamma_A C_A)^a}$$

TABLE XI

IRRADIATION OF SOLUTIONS OF POTASSIUM IODIDE IN 1-KC COBALT-60 GAMMA SOURCE AT 1 ATM ABS AND ROOM TEMPERATURE: POSSIBLE DOSIMETRIC METHOD

Stock Solution was: 100 ml water, 1 g potassium iodine, 1 drop glacial acetic acid.
Starch Solution was: 100 ml water, 1 g soluble starch.

Page	Starting Date	Time	Hours Irrad.	Dose Rate, Rep/hr	Dose, Rep	Observation
132127	3 Feb 53	1430	0	65,000±10%	0.	Add 1 ml starch soln. to 100 ml stock soln. 5 samples, 25 drops each, then put into vault Remove one at stated time.
132127	3 Feb 53	1435	0.083	65,000±10%	5,500.	Faint blue tint
132127	3 Feb 53	1500	0.50	65,000±10%	33,000.	Moderate blue
132127	3 Feb 53	1630	2.0	65,000±10%	130,000.	Purplish blue
132127	3 Feb 53	2030	6.0	65,000±10%	390,000.	2 out-both blue-black; put one sample into clean tube, added starch, color darkened.
132128	4 Feb 53	0930	0.	65,000±10%	0.	Add 75 drops starch soln. to 90 ml stock soln. and starch from 132127. Made 5 samples, 25 drops each, put all into vault. Remove one at stated time.
132128	4 Feb 53	0935	0.083	65,000±10%	5,500.	No change - clear.
132128	4 Feb 53	0940	0.17	65,000±10%	11,000.	No change - clear.
132128	4 Feb 53	1010	0.7	65,000±10%	45,000.	Moderate blue.
132128	4 Feb 53	1530	6.0	65,000±10%	390,000.	Deep purple; control clear.
132129	4 Feb 53	1030	0.	65,000±10%	0.	10 g potassium iodide, 100 ml water, 1 drop glacial acetic acid, 5 ml starch soln. Procedure as above.
132129	4 Feb 53	1035	0.083	65,000±10%	5,500.	Faint blue.
132129	4 Feb 53	1040	0.17	65,000±10%	11,000.	Light blue.
132129	4 Feb 53	1110	0.7	65,000±10%	45,000.	Deep blue; control clear.

where

E = the single-electrode potential at 25°C of the electrode,

E° = the single-electrode potential of the electrode when the concentration of all reactants and products involved is one molar (unit activity),

F = coulombs per Faraday of charge,

N = number of electrons involved in the electrode reaction,

C_A = concentration of the reactant, iodide, for which the activity coefficient is γ_A,

C_B = concentration of the product, iodine, for which the activity coefficient is γ_B, and

a and b are the stoichiometric coefficients of the reactants and products respectively, in the electrode reaction.

Now let two identical aqueous solutions of acidic iodide be prepared, such that a reaction will occur in either of them when it is subjected to gamma irradiation. Then concentration of iodine in the irradiated solution will be increased by a small amount. A concentration cell employing platinum electrodes and these two solutions, one irradiated and one not, will then have a potential as follows:

$$E = E_1 - E_2$$

$$= \left(E^\circ - \frac{2.303\,RT}{NF} \log_{10} \frac{(C_B)^b \text{ control}}{(C_A)^a \text{ control}} \frac{\gamma_B{}^b}{\gamma_A{}^a} \right)$$

$$- \left(E^\circ - \frac{2.303\,RT}{NF} \log_{10} \frac{(C_B)^b \text{ test}}{(C_A)^a \text{ test}} \frac{\gamma_B{}^b}{\gamma_A{}^a} \right) ,$$

where

C_A = iodide concentration, and

C_B = iodine concentration.

Since the iodide concentration of the control solution is approximately equal to the iodide concentration of the test solution for small degrees

of reaction, the above equation simplifies to:

$$E = 0.05915 \frac{b}{N} \log_{10} \frac{C_B \text{ test}}{C_B \text{ control}} \quad . \qquad (7)$$

Equation (7) shows that a small change in the amount of product may be detected by measuring accurately the oxidation potential corresponding to the ratio of the test value to the control value.

C. THERMODYNAMICS AND KINETICS OF REACTIONS

In the study of chemical equilibria, the standard free energy of reaction is a criterion of the degree of reaction to be anticipated at equilibrium in a given system. As usually defined, a negative standard free energy for a given reaction indicates the possibility of obtaining a preponderance of the product of the reaction in the equilibrium mixture. In addition, a reaction with a negative standard free energy of reaction may proceed spontaneously, but a positive free-energy change precludes the possibility of a spontaneous reaction. Only reactions with negative standard free energies of reaction will be considered in the following discussion. It is often assumed that the standard free energy of reaction is merely the net change in the energy of the system, and that the rate of the reaction may be explained on the assumption that a certain energy of activation must be possessed by a molecule or group of molecules before a reaction can occur. (See Fig. 31).

In a given system the fraction of molecules possessing a given energy is a statistical function of the energy possessed. Energies greater than or equal to the energy of activation needed for chemical reaction will be possessed by a certain fraction of molecules at a temperature above absolute zero. The rate of a chemical reaction proceeding as a result of thermal activation will vary exponentially with temperature if other conditions are fixed, since the number of molecules possessing energies equal to or greater than the energy of activation will vary exponentially with temperature.

As an alternative to thermal activation, energy might be introduced into a system in the form of electromagnetic radiation. For photon energies from 500 kev to 5 mev the predominant process would be ionization as a result of Compton scatter. The resulting ions or excited molecules, radicals or other excited species would have large energies compared with those ordinarily derived from statistical distribution of thermal energy. Each excited species activated by the encounter with radiation would probably possess sufficient

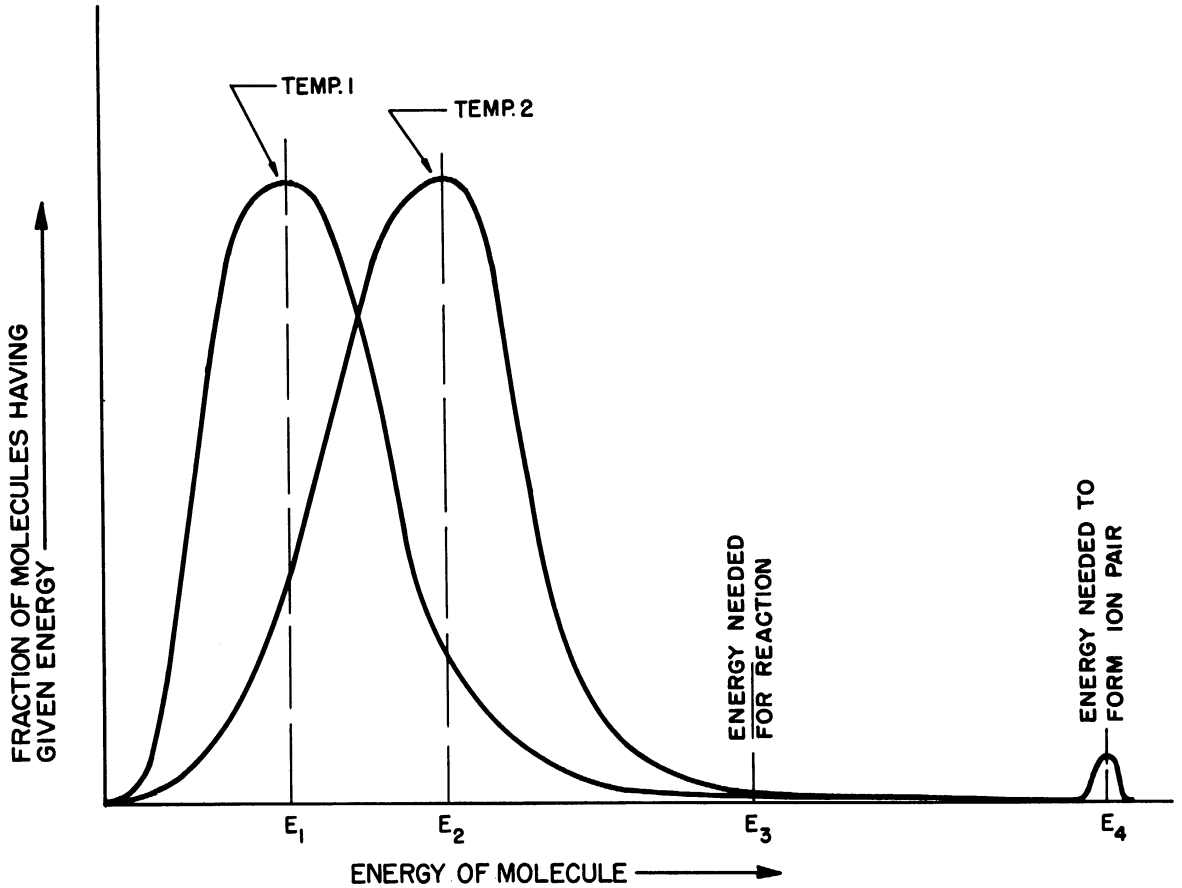


Fig. 31. Energy Distribution Among Molecules.

energy to react. Therefore, radiation might be used to induce chemical reactions in a system at a temperature too low for these reactions to occur measurably by thermal activation.

If the probability of attaining a sufficiently high energy to overcome the activation barrier were left to the Maxwell-Boltzmann distribution of energies among the molecules, the fraction reacting in unit time would be a function of the temperature only. In addition, for an exothermic reaction the net energy of the reaction is released and increases the random energy of the system as a whole. Thus in a sense the passage of each molecule over the barrier of activation helps to raise other molecules over this barrier.

However, if ions are produced by the absorption of highly energetic electromagnetic radiation and these ions react directly or indirectly to produce chemical products, then such an occurrence would not be a function of the temperature of the system. Rather the probability of such an occurrence would be governed by the intensity and energy of incident radiation. The energy of reaction and probably also the energy of ionization (barring fluorescence) would be released to the system and cause an increase in the random energy of the system as a whole, similar to that noted by thermal activation. However, there are cases in which the temperature is so low that this additional energy is not

sufficient to increase thermal activation by a measurable amount. Therefore, each molecule reacting as a result of radiation activation does not "lift" another molecule over the barrier of activation. Under these conditions, molecular interactions induced by radiation would be random events when considered as a function of the reacting system only, but the number of such interactions would be proportional to the dose rate of radiation. Furthermore, since the rate of reaction due only to thermal activation increases exponentially with the temperature, it should be possible to attain a temperature above which the contribution of radiation to the rate of reaction would become negligible compared with that of thermal activation.

In a chain reaction the energy of activation is transmitted directly from reacted molecules to unreacted molecules. It then becomes unnecessary for the energy of activation to be distributed at random among all molecules and then concentrated by random motion to activate the unreacted molecule. In effect the rate of a single-hit reaction is maintained in the chain reaction, but multiplied by the chain length.

The influences of radiation and temperature on the rate of a given reaction may be described quantitatively as follows:

The Arrhenius equation may be assumed to hold for the specific reaction velocity constant, k_t.

$$k_t = Ae^{-E_t/RT} \quad , \tag{8}$$

where R is the gas constant, T is the absolute temperature, E_t is the energy of activation, and A is a frequency factor for the reaction. It can be shown[5] that the factor $e^{-E_t/RT}$ represents the fraction of molecules having energies equal to or greater than E_t, the energy of activation. By postulate, these molecules are the only ones with sufficient energy to initiate reaction.

The specific reaction velocity constant, k_{tr}, of a reaction occurring in a field of radiation may be examined and equation (8) may be modified to:

$$k_{tr} = A(e^{-E_t/RT} + g) \quad , \tag{9}$$

where g is the fraction of molecules activated by irradiation.

Equations (8) and (9) may be rearranged and combined as follows:

$$\begin{aligned} \ln (k_{tr} - Ag) &= \ln Ae^{-E_t/RT} \\ &= \ln k_t \quad . \end{aligned} \tag{10}$$

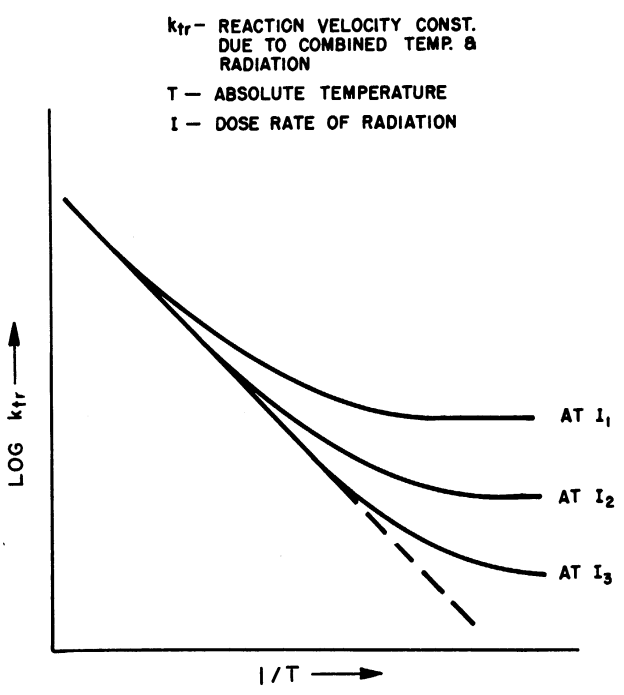


Fig. 32. Log K_{tr} Versus Reciprocal Temperature Plot.

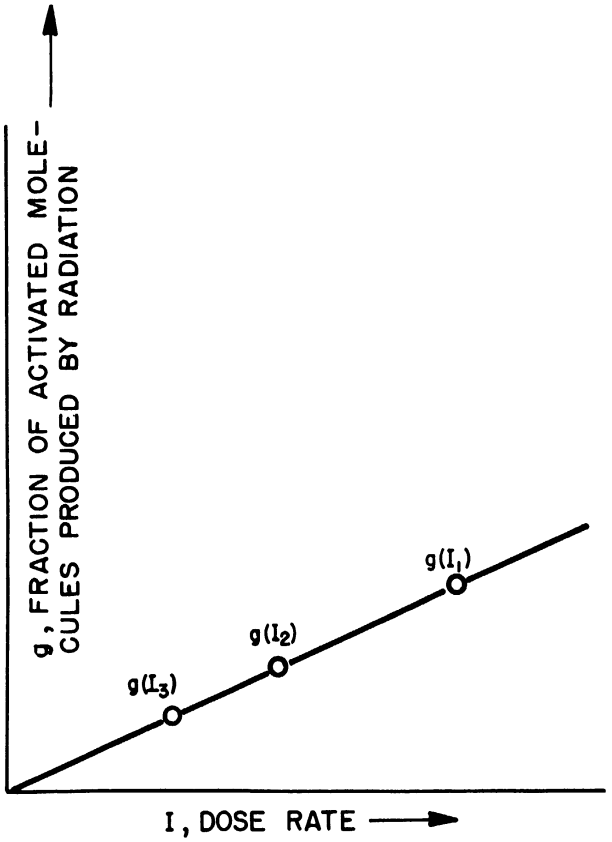


Fig. 33. Fraction of Activated Molecules as a Function of Dose Rate.

The validity of equation (10) may be tested as described below. Note that a plot of ln (k_{tr} - Ag) versus 1/T should be linear. Consequently a plot of ln k_{tr} versus 1/T would be expected to be curved (see Fig. 32). If such a plot were obtained experimentally (as by Bretton et al.[2]), it could be straightened by the subtraction of an empirically determined parameter from k_{tr}. The parameter determined by this method would equal Ag, the part of k_{tr} due to radiation alone.

The values of E_t and A could be closely approximated from equation (8) at temperatures sufficiently high that k_{tr} would be nearly indistinguishable from k_t. Then the value of g could be computed from equation (9). Then g could be plotted as a function of the dose rate, as shown in Fig. 33. Thus the separate effects of temperature and gamma radiation on the kinetics of a reaction with no temperature-dependent chain could be studied.

Figure 31 indicates that the fraction of molecules with energy sufficient to react is almost solely a function of radiation effects at temperature 1. However, at temperature 2 the area under the curve due to thermal excitation

in the region lying above E_3 is a considerable proportion of the total area lying above E_3. These observations illustrate the conclusion drawn from equation (10) above. That is, the rate of a reaction induced by gamma radiation is dependent on the dose rate and may be nearly independent of temperature over a wide range of temperatures less than temperature 2. However, at temperatures above the neighborhood of temperature 2 the rate of reaction becomes a function of both the temperature and dose rate due to radiation. At still higher temperatures the reaction due to thermal activation is sufficiently great that the rate due to radiation becomes small by comparison.

D. DOSIMETRY PROBLEMS

For any quantitative study of the effect of radiation on chemical reactions, it is necessary to know the dose rate of the radiation. Considerable work has been done since the last progress report on measurement of dose rates. The following discussion is taken from an article on that subject which was written by four of the authors of this report (Harmer, Lewis, Martin and Nehemias) for the 1953 Gordon Research Conference on Radiation Chemistry.

1. Experimental Procedure

Dose rates were measured chemically by the method employing the oxidation of ferrous sulfate solutions. Dilute solutions of ferrous sulfate (5×10^{-4} M) in aerated 0.8 N sulfuric acid solution were exposed to gamma radiation for doses of between 5 and 20 kilorep. The ferric ion produced by the gamma radiation depends on the presence of a small amount of oxygen, which is readily furnished by first passing air through the solution. For quantitative determinations of the ferric ion produced by irradiation, the spectrophotometric method described by J. Weiss[9] of Brookhaven National Laboratory was employed. This method makes use of a spectral absorption peak of ferric ion at about 304 millimicrons in the ultraviolet region. Optical densities of the irradiated solutions are measured at 305 millimicrons and compared with those of known ferric solutions made up by dilution of standardized ferric stock solution.

In converting the chemical yield to radiation dosage, a value of 15.4 micromoles/liter-kilorep was used. This value is based on the absorption of 93 ergs/gm of water for each equivalent roentgen of radiation. The solutions were irradiated in glass bottles about 3 cm in inside diameter, which were filled to a depth of about 4 cm.

The bottles of solution were placed inside and outside the 10-kilocurie source, as shown in Figs. 34 and 35, and inside the 1-kilocurie source. (See

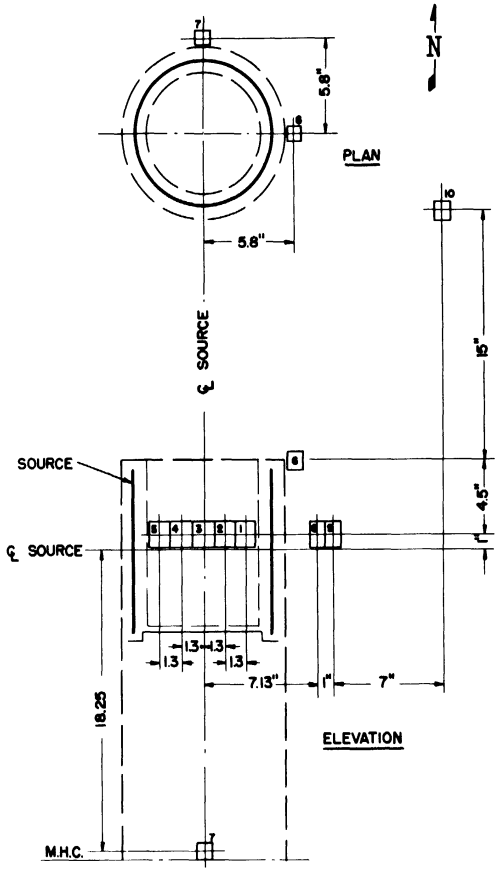


Fig. 34. Location of Samples--
10-kc Source Dosimetry.

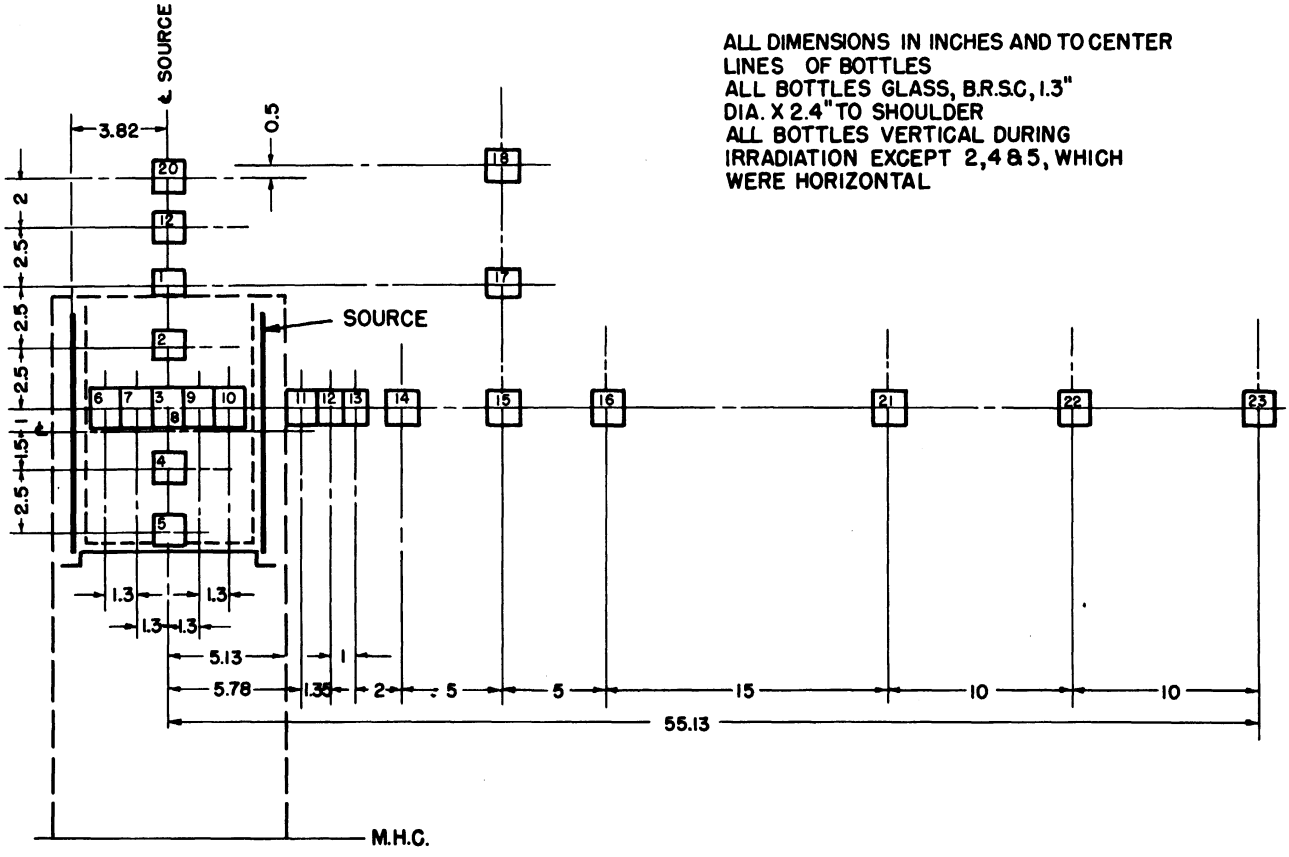


Fig. 35. Location of Samples--10-kc Source Dosimetry.

Table XII). Proper exposure times were calculated to fall within the range of the method of ferric ion determination. Measurement of dose rate in the 1-kilocurie source was carried out at times separated by an interval of 1 year, and values were found to be consistent after corrections for radioactive decay were applied. Measurements using a ceric sulfate system were also made and found to agree within experimental error with the ferrous sulfate results.

TABLE XII

IRRADIATION OF FERROUS SULFATE
SOLUTIONS IN COBALT-60 SOURCE - DOSIMETRY BY METHOD OF WEISS[9]
DATA FROM 10-KILOCURIE SOURCE UNLESS NOTED OTHERWISE

Page 132308
Date 16 Mar 53
See Fig. 34 For location of Samples

Sample Number	Dose Rate, Kilorep/Hr
1	280
2	250
3	242
4	244
5	292
6	342
7	38
8	102
9	80
10	8
1 kilocurie	57
1 kilocurie	60

Page 132306
Date 13 Mar 53
See Fig. 35 For Location of Samples

Sample Number	Dose Rate, Kilorep/Hr
1	144
2	194
3	249
4	261
5	266
6	281
7	243
8	234
9	248
10	274
11	168
12	86
13	115
14	52
15	24
16	13
17	22.
18	17
19	74.
20	42.
21	4.4
22	2.6
23	2.0
1 kilocurie	55.

Physical determinations of dose rate have also been carried out on both sources. Two instruments have been employed in these determinations: a Victoreen roentgen rate meter, which measures the current flow between electrodes in an ionization chamber which is placed in the radiation field; and a Victoreen r-meter, which measures the drop in potential of a charged condenser due to ionization current caused by the radiation. The rate meter was calibrated against radium standards by the manufacturer, while the r-meter was calibrated against a cobalt standard at the University of Michigan.

Within 50 cm of the center of the 10-kilocurie source the rate-meter readings were 15 to 20 per cent lower than the ferrous sulfate determinations. The r-meter readings were 15 to 20 per cent higher than the ferrous sulfate measurements in the 10-kilocurie source, and were 25 to 30 per cent higher in the 1-kilocurie source. The detailed significance of these differences is not clear.

2. Calculation Procedure

Since gamma radiation from a point source may be assumed to follow the usual inverse-square relation, it is possible to calculate the dose rate at any position in the neighborhood of a source of known shape and total activity by an integration technique similar to that employed in radiant heat transfer. If the geometry of the source shape is complicated, the resulting integration may be difficult. As a simple shape somewhat similar to the two cobalt-60 sources, consider first a hollow cylinder of negligible wall thickness and assign to it the power or curie rating of the actual source. The dimensions of the cylinder are taken to correspond as nearly as possible to those of the actual source, and the assigned curies are assumed to be distributed uniformly over the surface of the cylinder. Then the contribution to the radiation intensity at any given point due to an element of source area, dA, at a distance ρ is given by equation (11); see Fig. 36.

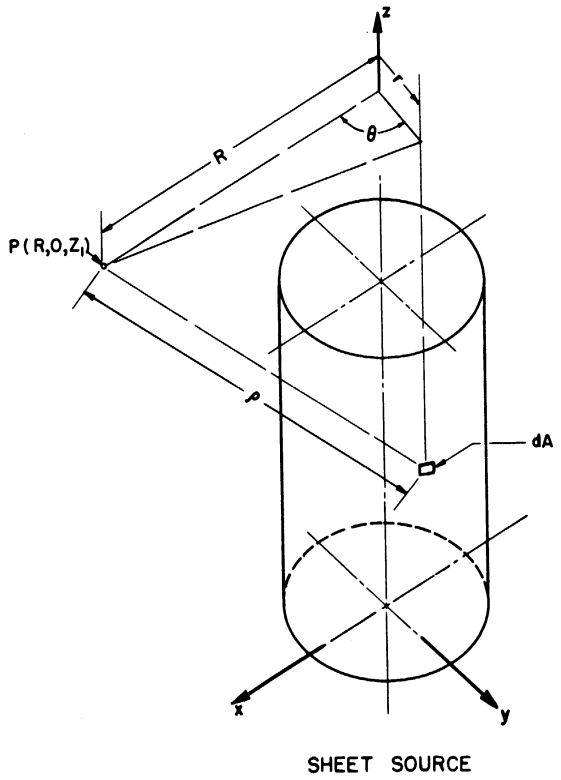


Fig. 36. Source With Negligible Wall Thickness.

$$dI = \alpha \frac{dA}{\rho^2} \quad . \qquad (11)$$

The total intensity at the given point is obtained by summing the contributions from all elemental areas as

$$I = \int_{Z=0}^{Z=L} 2 \int_{\theta=0}^{\theta=\pi} \frac{\alpha r d\theta dZ}{R^2+r^2-2Rr\cos\theta + (Z_1-Z)^2} \quad . \qquad (12)$$

Integrating equation (12) gives

$$I = \frac{2\alpha\pi r}{R+r}\left[F(\tan^{-1}\frac{R+r}{Z_1-L},k)-F(\tan^{-1}\frac{R+r}{Z_1},\ k) \right] \qquad (13)$$

for

$$Z_1 > L > 0,\ R \geq 0,\ r > 0,\ 0 \leq \tan^{-1}\frac{R+r}{Z_1-L},\ \tan^{-1}\frac{R+r}{Z_1} < \frac{\pi}{2}$$

and,

$$I = \frac{2\alpha\pi r}{R+r}\left\{ 2K(k)-\left[F(\tan^{-1}\frac{R+r}{L-Z_1},k)+F(\tan^{-1}\frac{R+r}{Z_1},k) \right] \right\} \qquad (14)$$

for

$$L \geq Z_1 > 0,\ R \geq 0,\ r > 0,\ R \neq r,\ 0 \leq \tan^{-1}\frac{R+r}{L-Z_1}\ ,\ \tan^{-1}\frac{R+r}{Z_1} < \frac{\pi}{2} \quad .$$

An alternative form may be obtained as shown by equation (15). Dewes and Goodale[3] have indicated the preliminary steps in this development.

$$I = \frac{2\alpha\pi r}{R+r}\left[F(\tan^{-1}\frac{Z_1}{|r-R|},\ k)-F(\tan^{-1}\frac{Z_1-L}{|r-R|},\ k) \right] \qquad (15)$$

for $Z_1 \geq 0,\ R \geq 0,\ r > 0,\ R \neq r,\ -\frac{\pi}{2} < \tan^{-1}\frac{Z_1-L}{|r-R|} < \frac{\pi}{2},\ 0 \leq \tan^{-1}\frac{Z_1}{|r-R|} < \frac{\pi}{2}$.

A relation given by Hancock[4] permits the transformation of equations (13) and (14) into equation (15), and vice versa. Equation (15) is considered more convenient in most computations, except for $R=r$, $Z_1 > L$, where equation (13) may be used to advantage.

The symbols used above are defined as follows:

I = dose rate, equivalent roentgens/hour;

A = area of source;

ρ = distance from elemental area dA to the point at which I is taken;

$\alpha = \left(\frac{\text{total activity, curies}}{\text{area of source, cm}^2}\right)\left(1000\frac{\text{millicuries}}{\text{curie}}\right)\left(\frac{\text{equivalent roentgens at 1 cm}}{\text{hour x millicurie point source}}\right)$;

r = radius of source
or constant radius vector of cylinder;

R = radial distance of point at which I is taken from axis of source;

θ = central angle from R to r;

Z = distance parallel to axis of source from base of source to element dA;

Z_1 = Z coordinate of point at which I is taken;

$k = \frac{2\sqrt{Rr}}{R+r}$;

$F(\phi,k)$ = elliptic integral of first kind of modulus k and amplitude ϕ; and

$K(k)$ = complete elliptic integral of first kind of modulus k.

Self-absorption of a hollow cylindrical source of finite thickness may be approximated along the axis of the source by the following procedure (see Fig. 37). It will be assumed (1) that the source is of uniform unit-volume activity and density, (2) that absorption occurs only within the source, (3) that scattered radiation due to the absorber will not affect the dose rate, (4) that radiation intensity and dose rate vary inversely with the square of the distance from a point source and inversely with an exponential function of absorber thickness, and (5) that the part of the source lying outside the cone, $\phi = \tan^{-1}\left[r_1/(Z_1-L)\right]$, also fulfills the foregoing assumptions. The resulting differential equation and its approximate integration are as shown in Fig. 38, where

P = distance between point and element of volume;

ρ = density, grams/cm^3

μ = mass absorption coefficient, cm^2/gram;

$V = \left(\frac{\text{total activity, curies}}{\text{volume of source, cm}^3}\right)\left(1000\frac{\text{millicuries}}{\text{curie}}\right)\left(\frac{\text{equivalent roentgens at 1 cm}}{\text{hour x millicurie point source}}\right)$;

dv = element of volume of source;
and all other terms are defined as above or in Fig. 37.

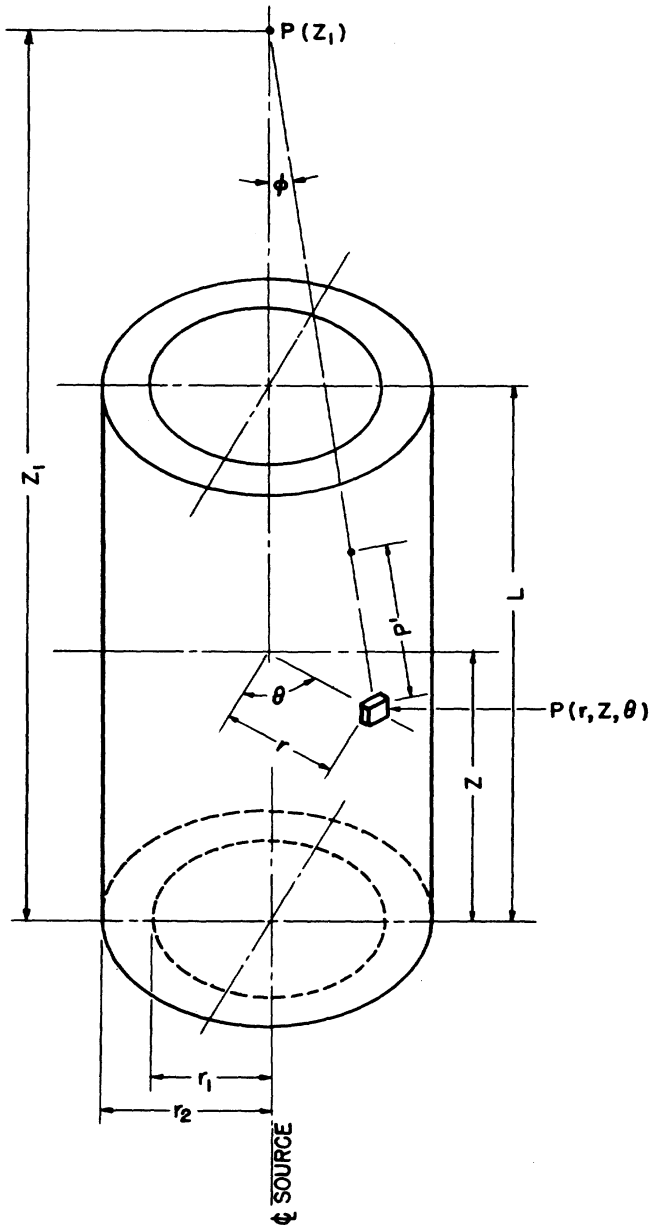


Fig. 37. Source with Finite Wall Thickness.

$$dI = V\frac{dv}{P^2}\, e^{-\mu P'\rho} \qquad \text{EQ. (16)}$$

$$I = \int_{r=r_1}^{r=r_2}\int_{Z=0}^{Z=L}\int_{\theta=0}^{\theta=2\pi} V\frac{r\,d\theta\,dZ\,dr}{r^2+(Z_1-Z)^2}\, e^{-\mu\rho(r-r_1)\csc\phi} \qquad \text{EQ. (17)}$$

if a three-term approximation to the exponential is employed:

$$\begin{aligned} I = {} & 2\pi V\Bigg[r_2\left(\tan^{-1}\frac{Z_1}{r_2}-\tan^{-1}\frac{Z_1-L}{r_2}\right)-r_1\left(\tan^{-1}\frac{Z_1}{r_1}-\tan^{-1}\frac{Z_1-L}{r_1}\right) \\ & +\frac{Z_1-L}{2}\ln\frac{1+\left(\frac{r_1}{Z_1-L}\right)^2}{1+\left(\frac{r_2}{Z_1-L}\right)^2}+\frac{Z_1}{2}\ln\frac{1+\left(\frac{r_2}{Z_1}\right)^2}{1+\left(\frac{r_1}{Z_1}\right)^2}\Bigg] \\ & +2\pi V\mu\rho\Bigg[\left(\frac{r_2^2}{2}-r_2r_1\right)\left(\sinh^{-1}\frac{Z_1-L}{r_2}-\sinh^{-1}\frac{Z_1}{r_2}\right) \\ & +\frac{r_1^2}{2}\left(\sinh^{-1}\frac{Z_1-L}{r_1}-\sinh^{-1}\frac{Z_1}{r_1}\right) \\ & +\left(\frac{Z_1-L}{2}\right)\left(r_2\sqrt{\left(\frac{Z_1-L}{r_2}\right)^2+1}-r_1\sqrt{\left(\frac{Z_1-L}{r_1}\right)^2+1}\right) \\ & -\frac{Z_1}{2}\left(r_2\sqrt{\left(\frac{Z_1}{r_2}\right)^2+1}-r_1\sqrt{\left(\frac{Z_1}{r_1}\right)^2+1}\right) \\ & +(Z_1-L)r_1\ln\frac{r_1}{r_2}\left(\frac{\sqrt{\left(\frac{Z_1-L}{r_1}\right)^2+1}+1}{\sqrt{\left(\frac{Z_1-L}{r_2}\right)^2+1}+1}\right) \\ & -Z_1r_1\ln\frac{r_1}{r_2}\left(\frac{\sqrt{\left(\frac{Z_1}{r_1}\right)^2+1}+1}{\sqrt{\left(\frac{Z_1}{r_2}\right)^2+1}+1}\right)\Bigg] \\ & +\pi V\mu^2\rho^2L\left[\frac{r_2^2-r_1^2}{2}-2r_1(r_2-r_1)+r_1^2\ln\frac{r_2}{r_1}\right] \end{aligned} \qquad \text{EQ. (18)}$$

for $Z_1 \geq 0$, $-\frac{\pi}{2} < \tan^{-1}\frac{Z_1-L}{r_1}$, $\tan^{-1}\frac{Z_1-L}{r_2} < \frac{\pi}{2}$;

$0 \leq \tan^{-1}\frac{Z_1}{r_1}$, $\tan^{-1}\frac{Z_1}{r_2} < \frac{\pi}{2}$; $r_1 > 0$, $r_2 > 0$, $r_1 \neq r_2$.

Fig. 38. Differential Equation and Its Integration for Source with Finite Wall Thickness. (Dosimetry Calculations)

Equations (15) and (18) were applied to both the 1000- and 10,000-curie sources. In the case of the 1000-curie source, it was simple to assume a cylinder with dimensions corresponding to the actual cobalt cylinder. In the case of the 10,000-curie source, the nest or bundle of 100 rods was assumed equivalent to a cylinder whose inside and outside diameters were the shortest and longest diametrical distances across the rod bundle. The 10,000 curies was assumed to be uniformly divided throughout this volume and the density of the assumed cylinder was taken so that its mass equalled that of the rods themselves.

Calculated and observed values of dose rate for the 10-kilocurie source of cobalt-60 are compared in Tables XIII and XIV and are plotted in Figs. 39 and 40. The calculated values were based on an assumed activity of 10,000 curies. The observed values are considerably less than the calculated values.

TABLE XIII

DOSE RATES ON AXIS OF 10-KC SOURCE

$Z_1 - \frac{L}{2}$ = Distance above midplane, cm; R = 0

	Calculated Rep/hr for 10,000 curies				Measured Rep/hr	
$Z_1 - \frac{L}{2}$	Annular Source		Sheet Source	$Z_1 - \frac{L}{2}$	Ferrous Oxidation*	Victoreen Rate Meter
	No Absorption	With Absorption	No Absorption			
0	1,020,000	830,000	1,010,000	0	242,000 (16 Mar 53)	
6.35	910,000	747,000	928,000	2.5	249,000	
12.7	665,000	527,000	662,000	2.5	234,000	
25.4	222,000	154,000	218,000	8.9	194,000	
38.1	97,000	61,000	96,000	15.2	144,000	
63.5	-	-	32,000	20.3	-------	61,000
				21.6	74,000	------
				22.8	-------	48,000
				25.4	-------	38,000
				26.7	42,000	------
				38.1		16,000
				50.8		8,100
				60.8		5,000
				76.2		3,300

* 13 Mar 53 unless noted.

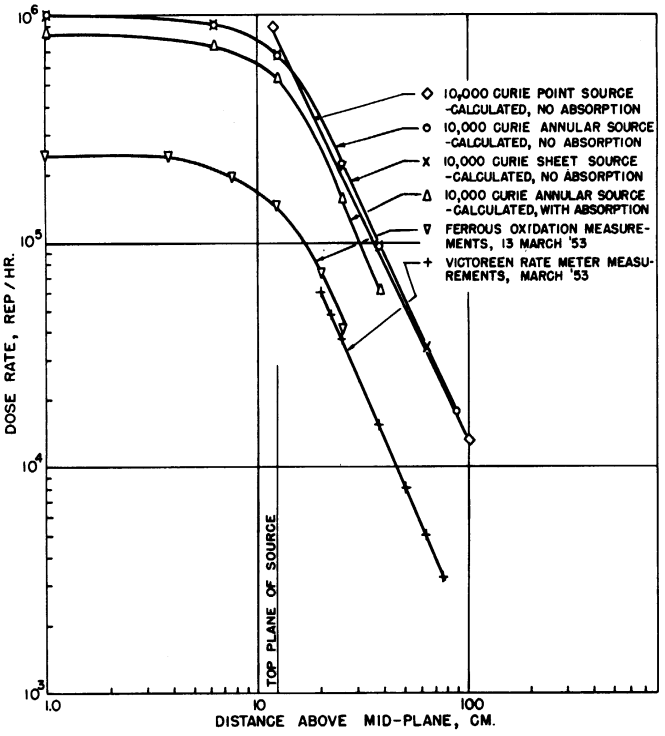


Fig. 39. Dose Rate on Axis of 10-kc Cobalt-60 Source.

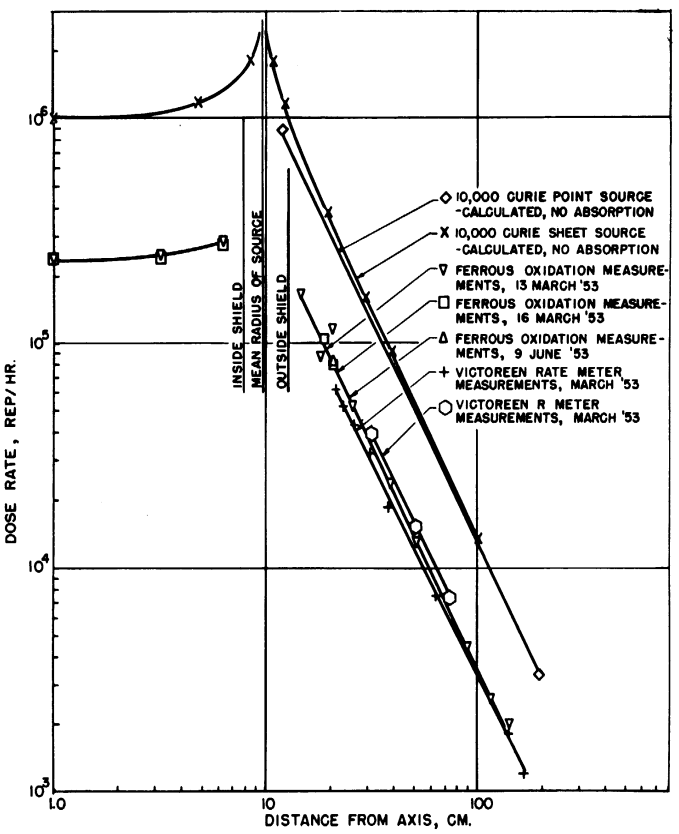


Fig. 40. Dose Rate on Mid-Plane of 10-kc Cobalt-60 Source.

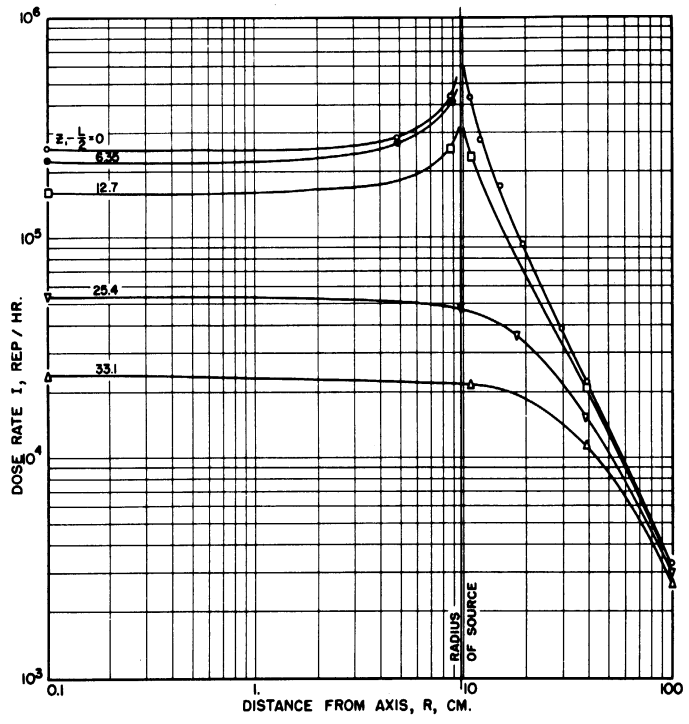


Fig. 41. Dose Rates Parallel to Mid-Plane Interpolated from Measurements--10-kc Source.

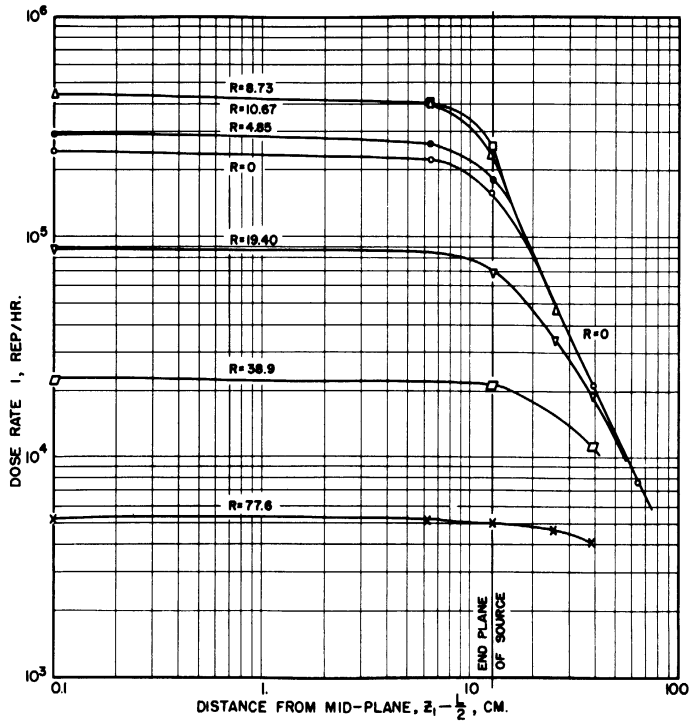


Fig. 42. Dose Rates Parallel to Axis Interpolated from Measurements--10-kc Source.

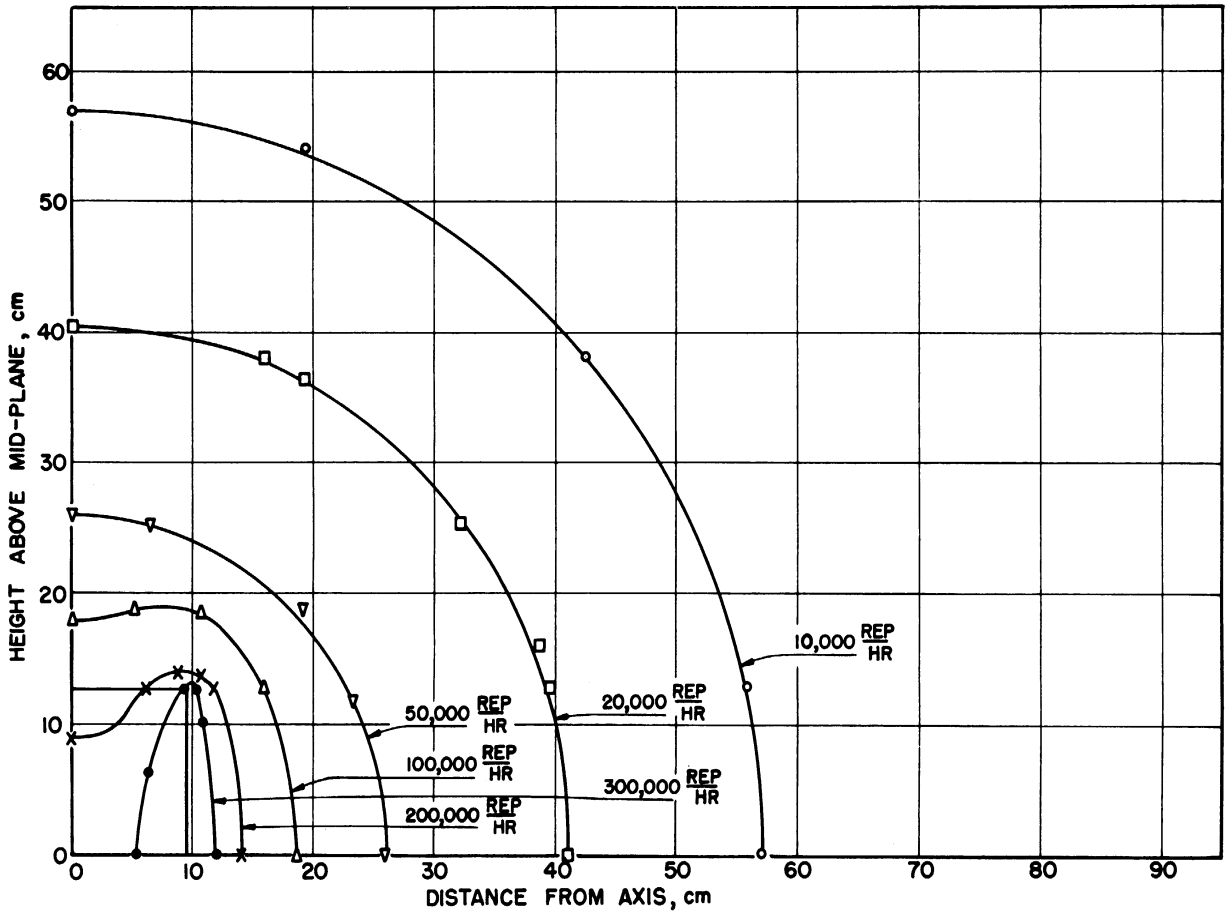


Fig. 43. Isodose Surfaces Interpolated From Measurement--10-KC Source.

For any given method of measurement the observed values are a nearly constant fraction of the calculated values. In Figs. 41, 42, and 43 appear cross plots of equation (15) when the latter is made to agree with data from the oxidation of ferrous ion. The data were taken on the midplane and on the axis in March, 1953. The source was irradiated at the Chalk River NRX reactor and was rated at 9250 curies on shipment from the Chalk River site in January, 1953. The activity computed from each means of measurement appears in Table XV. In the extreme right column of Table XV the ratio of the curies estimated from observed values of dose rate to the 9250-curie nominal value after correction of the latter value for decay appears. If self-absorption is not considered, the activity is estimated to be from 17 to 33 per cent of the nominal value. If self-absorption is considered, the activity is estimated to be from 28 to 33 per cent of the nominal value. These figures are computed from data taken both on the midplane and on the axis. No estimate of self-absorption was made on the midplane however.

Calculated and observed values of dose rate and curies for the 1-kilocurie source are compared in Table XVI and in Fig. 44. The 1-kilocurie source was irradiated at Brookhaven National Laboratory and was assumed to have a nominal activity of 1000 curies in July, 1951. The activity computed from each means of

TABLE XIV

DOSE RATES ON MIDPLANE OF 10-KC SOURCE

$Z_1 = \frac{L}{2} = 12.7$ cm; R = Distance from Axis, cm.

R	Rep/hr for Sheet Source; No Absorption Calculation for 10,000 curies	R	Rep/hr for Ferrous Oxidation		R	Rep/hr for Victoreen Meters	
			13 Mar 53	16 Mar 53		Rate Meter	R-Meter
0	1,000,000.	0	249,000.	242,000.	21.3	61,000.	
		0	234,000.	-			
4.85	1,120,000.	3.30	243,000.	250,000.	23.1	52,000.	
8.70	1,800,000.	3.30	248,000.	244,000.	26.2	43,000.	
9.70	∞	6.30	281,000.	280,000.	30.8	32,000.	
10.7	1,800,000.	6.30	274,000.	292,000.	31.8	------	40,000.
12.0	1,150,000.	14.7	168,000.	-	38.4	18,500.	------
19.4	379,000.	18.1	86,000.	102,000.	51.0	------	15,500.
29.1	160,000.	20.6	115,000.	79,500.	64.0	7,500.	------
38.8	92,000.	25.7	52,000.		73.9	------	7,500.
100.	14,000.	38.4	24,000.		140.	1,800.	
		51.0	13,000.		165	1,200.	
		89.1	4,400.				
		114.8	2,600.				
		140.0	2,000.				
		20.6		82,000			
				9 June 53			

TABLE XV

ESTIMATES OF ACTIVITIES FROM MEASUREMENTS OF DOSE RATES

Source	Measurements: Where Taken	Measurements: Method	Date	Estimate of Activity, Curies*: Maximum	Estimate of Activity, Curies*: Minimum	Estimate of Activity, Curies*: Arithmetic Mean	Mean Value Divided By Decayed Nominal Value
10-KC	Axis	Ferrous Oxidation	Mar 53	2500 3100A	2100 2800A	2300 2950A	0.26 0.33A
		Victoreen Rate Meter	Mar 53	1700 2600A	1400 2500A	1550 2550A	0.17 0.28A
	Mid-plane	Ferrous Oxidation	Mar 53	2600	2200	2400	0.27
		Victoreen Rate Meter	Mar 53	2500	2000	2250	0.25
		Victoreen R Meter	Mar 53	3000	2900	2950	0.33
1-KC	Axis	Ferrous Oxidation	May 52			140 180A	0.16 0.20A
			May 53			130 170A	0.16 0.21A
		Victoreen Rate Meter	Feb 53	150 190A	110 150A	130 170A	0.16 0.20A
		Victoreen R Meter	May 52			170 230A	0.19 0.26A
			May 53			160 210A	0.20 0.26A

* A after value indicates self-absorption was considered.

TABLE XVI

DOSE RATES ON AXIS OF 1-KC SOURCE

$Z_1 - \frac{L}{2}$ = Distance above midplane, cm

R = 0

r = 2.493 cm

	Calculated Rep/hr for 1,000 curies				Measured Rep/hr		
$Z_1 - \frac{L}{2}$	Annular Source		Sheet Source	$Z_1 - \frac{L}{2}$	Ferrous Oxidation	Victoreen Rate Meter	Victoreen R-Meter
	No Absorption	With Absorption	No Absorption		Feb 53		
0	442,000.	341,000.	460,000.	0	62,300. May, 52		79,000. May, 52
8.75	429,000.	336,000.	450,000.	0	57,200. May, 53		72,000. May, 53
17.5	232,000.	184,000.	240,000.	1.3		51,600.	
35.0	-------	-------	15,200.	3. 8		52,800.	
87.5	--------	-------	1,900.	6.3		54,000.	
				8.9		55,800.	
				11.4		57,500.	
				14.0		46,800.	
				16.5		19,200.	
				19.0		9,300.	
				21.6		5,150.	
				24.2		3,240.	
				26.7		1,860.	
				29.2		1,320.	
				34.2		490.	

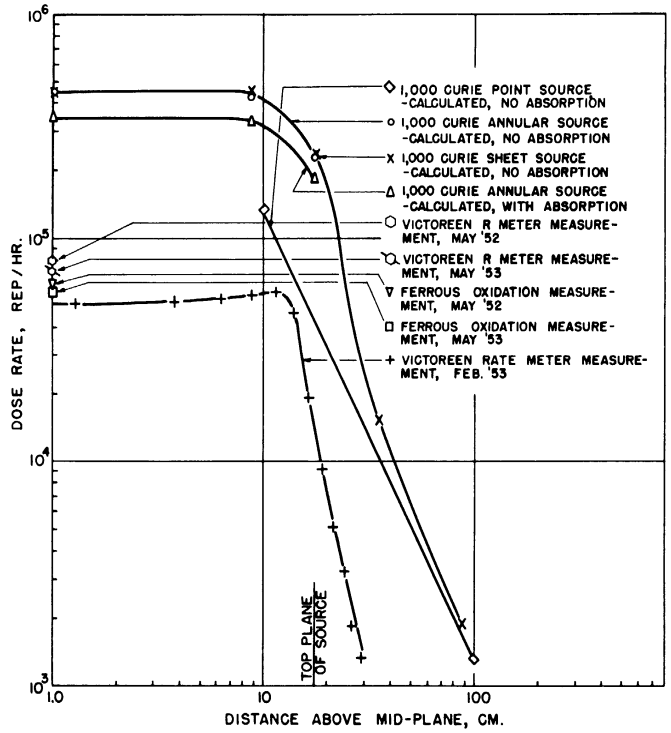


Fig. 44. Dose Rate on Axis of 1-kc Cobalt-60 Source.

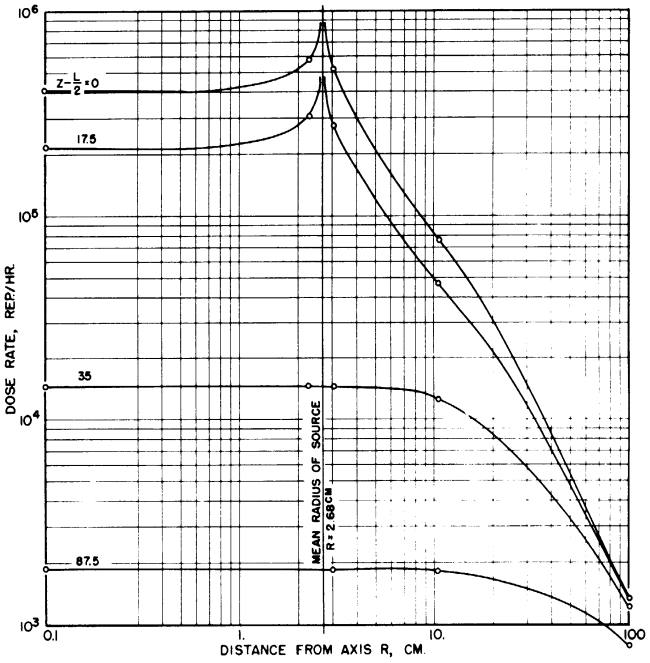


Fig. 45. Calculated Dose Rates Parallel to Mid-Plane--1-kc Source.

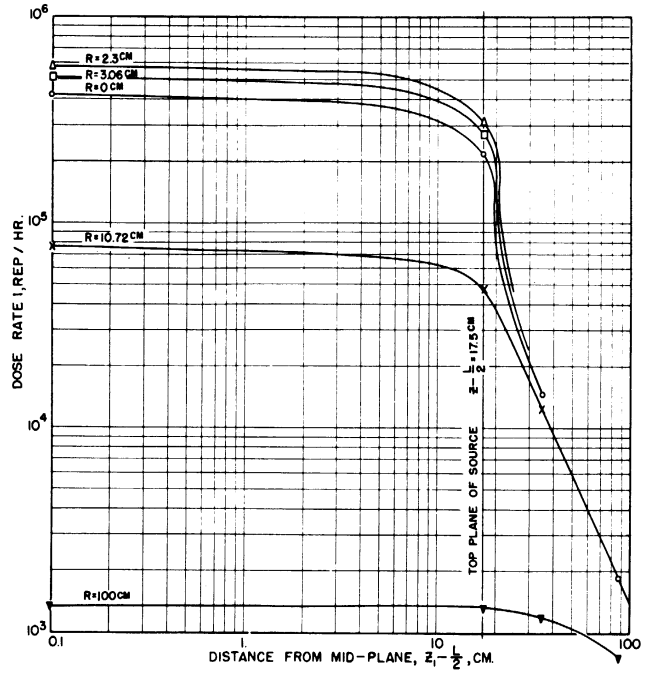


Fig. 46. Calculated Dose Rates Parallel to Axis--1-kc Source.

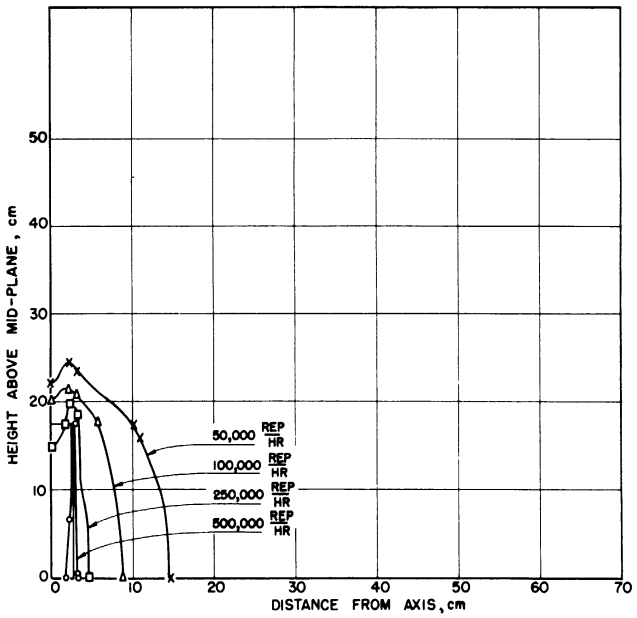


Fig. 47. Calculated Isodose Surfaces--1-kc Source.

measurement is given in Table XV. The ratios of observed to decayed nominal curies appear in the right column. If self-absorption is not considered, the activity is estimated to be from 16 to 20 per cent of the nominal value. If self-absorption is considered, the activity is estimated to be from 20 to 26 per cent of the nominal value. These figures are computed from data taken on the axis only, since it was not possible to make measurements external to the source. However, it was desired to compare the dose rates predicted by equation (15) for the 1-kilocurie source with those predicted for the 10-kilocurie source in order to observe differences caused by the different geometrical proportions of the two sources. Consequently Figs. 45, 46, and 47 are presented to portray the dependence of dose rate on position in the neighborhood of the 1-kilocurie source. The data for these figures were computed on the assumption that the source actually contained 1000 curies.

Judging from the above results, there appears to be about a threefold discrepancy between the curies in the 10-kilocurie source as estimated from ionization measurements and as calculated from neutron absorption. The comparisons for the 1-kilocurie source are not so meaningful, since no firm estimate of the activity of the source was supplied by Brookhaven National Laboratory.

The errors in the methods of calculation summarized in equations (13), (15), and (18) probably arise chiefly from the simplifying assumptions made. The assumption that the source has no thickness is evidently justified by the agreement of values calculated on this assumption with those in which the thickness of the source is considered (see Figs. 39 and 44). The results in Tables XII and XVI show that absorption is not negligible. However, in Figs. 39 and 40 it can be seen that the plots from data and from equations (13), (15), and (18) differ by an approximately constant factor between any pair of curves. This result is interpreted to mean that equations (13) and (15) may be used within limits to predict the distribution of dose rates without consideration of self-absorption, but that accurate prediction of dose rates requires consideration of self-absorption.

The 1-kilocurie source is evidently not of uniform activity throughout its whole volume. This conclusion was reached from a study of Fig. 44. Note that the measured dose rates in the 1-kilocurie source do not vary with distance along the axis in the manner predicted by the calculated curves. The depression near the midplane is probably caused by lower unit activity inside the source in this region, which in turn is probably caused by failure of neutrons to penetrate to the interior of the source near the midplane as abundantly as near the ends.[8] The other assumptions introduced are thought to be reasonably acceptable, although equation (18) converges much more slowly as Z_1 is increased. The value of 13.5 equivalent roentgens/hour at 1 cm per millicurie point source of cobalt-60 was taken from the work of Marinelli, Quimby, and Hine[7], and was assumed to be correct within our experimental error.

Although there were some differences between the chemical and physical dosimetry measurements, they were not sufficiently large to account for the factor of three or four between the nominal activities of the sources and those which result from the dosimetry measurements themselves. In an attempt to understand this discrepancy, attention was turned to the calculations of activity of a cobalt sample which is subjected to neutron radiation. Neutron absorption calculations are generally used to predict activities of irradiated samples.

3. Consideration of Neutron Activation Equations

Exact computation of the absolute amount of an isotope produced during neutron bombardment is dependent on precise knowledge of several factors. The relationship commonly used for such computations is:

$$C = \frac{0.6\phi\sigma Mr}{3.7\times10^{10}A}\left(1-e^{-0.693t/T}\right) , \qquad (19)$$

where

C = total activity of isotope produced, curies;

ϕ = thermal neutron flux, particles/cm^2-sec;

σ = activation cross section, barns;

A = atomic weight of irradiated isotope, grams;

t = length of irradiation, arbitrary time units;

T = half-life of isotope produced, same time units;

M = mass of element present, grams; and

r = fraction of irradiated isotope present.

The factors which might contribute to a serious error in the computed value of C will be considered. Of these factors, A is known accurately in most cases and t, the length of irradiation, may be measured as accurately as desired, the limiting consideration being the length of time required for insertion and removal of the samples.

Half-life may in general be measured as accurately as desired. Exceptions to this statement must be made for extremely long- and extremely short-lived isotopes. The buildup of activity under bombardment is exponential (Fig. 48). The plateau or saturation value of activity is simply the coefficient of the

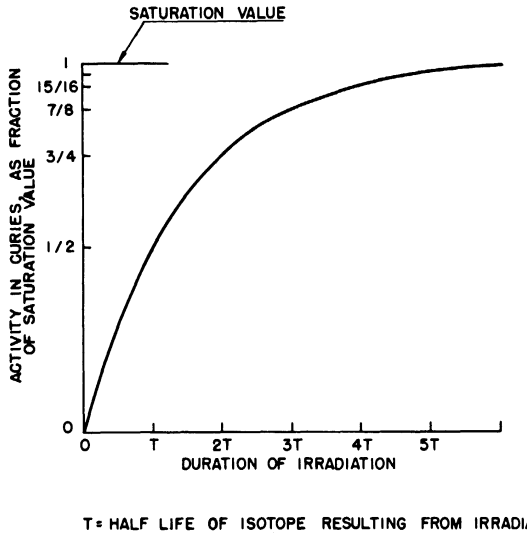


Fig. 48. Increase in Activity During Neutron Irradiation.

bracketed term in equation (19). It can readily be seen that in the case of short-lived isotopes, i.e., for $T \ll t$, the exponential term is of negligible importance, and that errors in the determination of T have little effect on the computed value of C at the time of removal. For long-lived isotopes, however, the exponential term becomes:

$$1-e^{-0.693t/T} \cong 1-(1-+0.693t/T) = 0.693t/T \qquad (20)$$

and errors in the determination of T have a direct effect on the computed value of C.

The case of cobalt-60, with a half-life of the order of 5 years and irradiations of the order of months, is still on the nearly linear portion of the activation curve. However, recent determinations of the half-life of cobalt-60 are within 5 per cent of one another. Differences of this magnitude, although serious in many cases, cannot be considered as significant contributions to the discrepancy under discussion.

The accurate determination of the remaining factors (neutron flux density, ϕ, and activation cross section, σ) is not as straightforward as those previously discussed. There are several reasons for this difficulty.

First, the determinations as commonly made are interdependent. To measure ϕ using an isotope of known σ , or to measure σ using a neutron source of known ϕ is relatively simple. However, the calibration of the quantity to be used as a reference is more complex. A closer examination of these measurements might uncover a factor partially responsible for the observed discrepancy, but it seems unlikely that such a factor could explain a threefold error.

Second, in any practical case the observed neutron flux is not completely thermalized but contains a finite range of energies higher than thermal. The activation cross section is a function of energy. Therefore it is necessary in principle to know the neutron spectrum and the cross section as a function of energy. In general, however, the cross section at nonthermal energies is vanishingly small, with the exception of occasional resonance energies. Such activations could contribute a source of error to activity computations, but the computed value would then be too low rather than the observed high value.

Third, local neutron depression caused by the insertion of irradiation samples of high cross section must be considered. It is this consideration which seems the most probable source of the observed discrepancy. Recent work of Levin and Hughes[6] indicates that this correction is of the order of magnitude of the observed discrepancy. Therefore, at this writing it is believed that the activity of a source calculated from neutron absorption, such as the 9250 curies mentioned above for our source, may be several times too high because of the depression of the neutron flux in the vicinity of the cobalt.

E. DOSE RATE WITHIN A CYLINDRICAL PRESSURE REACTOR

All the foregoing calculations and dosimetry measurements are for points in air lying at different distances from the sources. In the experiments reported on the polymerization of ethylene, however, the reaction took place inside a stainless-steel pressure vessel. A series of calculations was therefore made on the intensity of radiation inside the pressure vessel, taking into account the absorption of gamma radiation by the walls of the vessel (see Fig. 49).

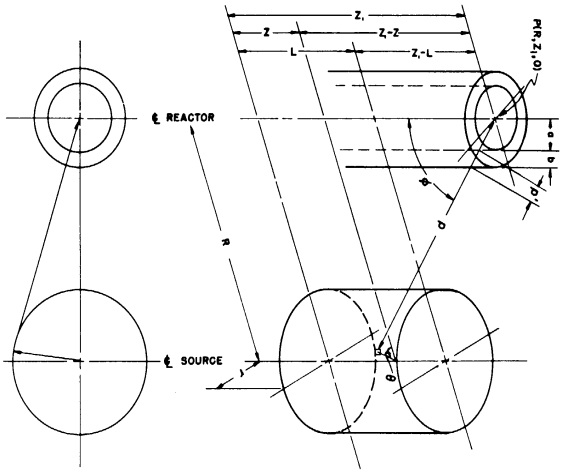


Fig. 49. Gamma Source and Pressure Reactor Shown Diagrammatically.

It is assumed (1) that the source of gamma radiation is a cylindrical sheet of no thickness and of uniform activity per unit area, (2) that the source is transparent to its own radiation, (3) that the dose rate varies inversely with the square of the distance and inversely with an exponential function of absorber thickness, and (4) that no absorption occurs inside the reactor.

Let the terminology be defined as in Fig. 49 and as follows:

I = dose rate at $P(R,Z)$, rep/hr;

$\alpha = \left(\frac{\text{activity of source, curies}}{\text{area of source, cm}^2}\right)\left(1000\frac{\text{millicuries}}{\text{curie}}\right)\left(\frac{\text{equivalent roentgens at 1 cm}}{\text{hr x millicurie point source}}\right)$;

μ = absorption coefficient, cm^2/gram;

A = area of source, cm^2;

P = distance from dA at $P(r,Z,\theta)$ to $P(R,Z)$;

P' = distance through bomb wall, cm;

$K(k)$ = complete elliptic integral of first kind of modulus k;

$k_1 = \frac{2\sqrt{Rr}}{R+r}$;

ρ = density of bomb wall, grams/cm^3; and

X = distance in Fig. 50 from source to point at which I is measured.

See Fig. 50.

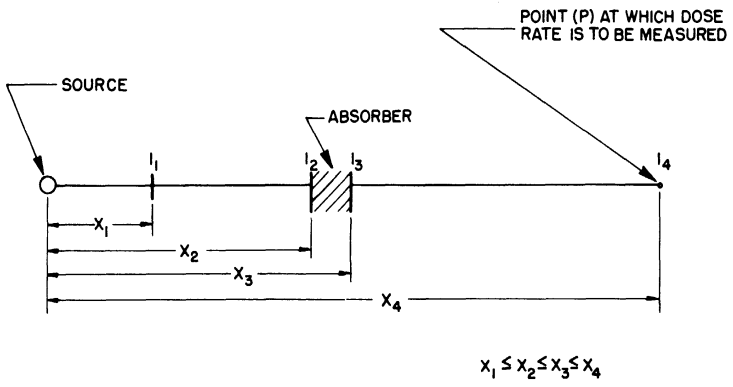


Fig. 50. Attenuation of Gamma Radiation by Absorption and Distance.

From assumption (3) above the following equation may be written:

$$dI = -\mu I dX - \frac{2}{X} I dX \tag{21}$$

Integration and substitution of limits yields the expressions:

$$I_2 = \left(\frac{X_1}{X_2}\right)^2 I_1 \tag{22}$$

$$I_4 = \left(\frac{X_3}{X_4}\right)^2 I_3 \tag{23}$$

$$\frac{I_3}{I_2} = \left(\frac{X_2}{X_3}\right)^2 e^{-\mu(X_3 - X_2)} \tag{24}$$

Combining equations (22), (23), and (24) results in the expression:

$$I_4 = \left(\frac{X_1}{X_4}\right)^2 I_1 \, e^{-\mu(X_3 - X_2)} \tag{25}$$

From equation (25) we may deduce that the location of an absorber is immaterial as long as it is between the source and point P. Only the thickness of the absorber need be considered.

Now the equations may be written for dose rate on the axis of the bomb (see Fig. 49):

$$P' = b \csc \phi \quad , \tag{26}$$

$$\csc \phi = \sqrt{\frac{R^2+r^2- 2Rr \cos \theta + (Z_1-Z)^2}{R^2+r^2- 2Rr \cos \theta}} \quad , \tag{27}$$

and

$$dI = \alpha_\rho \frac{dA}{2} \, e^{-\mu\rho P'} \tag{28}$$

Equation (28) must be integrated over the entire source, as shown by equation (29). A three-term approximation to the exponential is employed.

$$I = \int_{Z=0}^{Z=L} 2 \int_{\theta=0}^{\theta=\pi} \alpha r d\theta dZ \left\{ \frac{1}{R^2+r^2-2Rr\cos\theta + (Z_1-Z)^2} \right.$$

$$- \frac{\mu b\rho}{\sqrt{R^2+r^2-2Rr\cos\theta + (Z_1-Z)^2}\,\sqrt{R^2+r^2-2Rr\cos\theta}}$$

$$\left. + \frac{\mu^2 b^2 \rho^2}{2!} \frac{1}{(R^2+r^2-2Rr\cos\theta)} - \ldots \right\} \qquad (29)$$

The first term within braces has been integrated above, and presented as equation (15). Integration of equation (29) yields equation (30).

$$I = \frac{2\alpha\pi r}{R+r}\left[F\left(\tan^{-1}\frac{Z_1}{|r-R|}, k_1\right) - F\left(\tan^{-1}\frac{Z_1-L}{|r-R|}, k_1\right)\right]$$

$$-2\alpha\mu b\rho\sqrt{\frac{r}{R}}\int_{Z_1-Z=Z_1-L}^{Z_1-Z=Z_1}\frac{K(k)dk}{1-(k/k_1)^2} + \frac{\alpha\pi r L\mu^2 b^2\rho^2}{R^2-r^2} \quad , \qquad (30)$$

where

$$k = k_1\frac{(Z_1-Z)}{\sqrt{(R-r)^2+(Z_1-Z)^2}} \quad .$$

Equation (30) holds for

$$Z_1 \geq L > 0, \quad 0 < \tan^{-1}\frac{Z_1-L}{|r-R|}, \ \tan^{-1}\frac{Z_1}{|r-R|} < \frac{\pi}{2} \quad , \quad R \neq r \neq 0 \quad ,$$

If, however,

$$L \geq Z_1 > 0, \quad R \neq r \neq 0 \quad ,$$

then

$$I = 2\frac{\alpha\pi r}{R+r}\left[F\left(\tan^{-1}\frac{Z_1}{|r-R|}, k_1\right) - F\left(\tan^{-1}\frac{Z_1-L}{|r-R|}, k_1\right)\right]$$

$$- 2\alpha\mu b\rho\sqrt{\frac{r}{R}}\left[\int_{Z_1-Z=Z_1-L}^{Z_1-Z=0}\frac{K(k)dk}{1-(k/k_1)^2} + \int_{Z_1-Z=0}^{Z_1-Z=Z_1}\frac{K(k)dk}{1-(k/k_1)^2}\right] \qquad (31)$$

$$+ \frac{\alpha\pi r L\mu^2 b^2\rho^2}{R^2-r^2} - \ldots \quad .$$

Since k, defined above, is the modulus of an elliptic integral of the first kind,

$$1 \geq k \geq 0 \quad .$$

Consequently, for equation (31) the following definitions are employed.

$$k = k_1 \frac{(Z_1-Z)}{\sqrt{(R-r)^2 + (Z_1-Z)^2}} \quad \text{for} \quad Z_1 \geq Z$$

$$k = k_1 \frac{(Z-Z_1)}{\sqrt{(R-r)^2 + (Z_1-Z)^2}} \quad . \quad \text{for} \quad Z > Z_1$$

If

$$Z_1 \geq L \geq 0, \; R=0$$

then

$$I = 2\alpha\pi \left(\tan^{-1} \frac{Z_1}{r} - \tan^{-1} \frac{(Z_1-L)}{r}\right) - 2\alpha\mu b\rho\pi \ln\left(\frac{Z_1 + \sqrt{r^2 + Z_1^2}}{(Z_1-L) + \sqrt{r^2 + (Z_1-L)^2}} \right)$$

$$+ \frac{\alpha\pi L\mu^2 b^2 \rho^2}{r} - \cdots \quad , \qquad (32)$$

where

$$0 \leq \tan^{-1}\left(\frac{Z_1}{r} \right), \; \tan^{-1} \left(\frac{Z_1-L}{r}\right) < \pi/2 \quad .$$

If, however,

$$L \geq Z_1 \geq 0, \; R=0,$$

then

$$I = 2\alpha\pi \left[\tan^{-1}\left(\frac{Z_1}{r} \right) + \tan^{-1} \left(\frac{L-Z_1}{r}\right)\right]$$

$$- 2\alpha\mu b\rho\pi \ln \left\{ \frac{\left[(L-Z_1)+\sqrt{r^2+(L-Z_1)^2}\right]\left[Z_1+\sqrt{r^2+Z_1}\right]}{r^2} \right\}$$

$$+ \frac{\alpha\pi L\mu^2 b^2 \rho^2}{r} - \cdots \quad , \qquad (33)$$

where

$$0 < \tan^{-1}\left(\frac{Z_1}{r}\right),\ \tan^{-1}\left(\frac{L-Z_1}{r}\right) < \pi/2 \quad .$$

In equations (30) and (31) terms of the form:

$$\int \frac{K(k)dk}{1-(k/k_1)^2}$$

may be integrated graphically.

F. EQUIPMENT CHANGES

A number of additions to and modifications of the equipment in the laboratory have been made during the past several months. These are as follows:

Some special heaters were designed and built to maintain the stainless-steel reactor at elevated temperatures. The heaters were designed to be added to or removed from the reactor readily and to absorb a minimum of gamma radiation. Chromel-A resistance wire previously wound around a 1/8-inch arbor was wound around porcelain tubes. The turns were insulated from each other by winding asbestos rope soaked in water glass between turns. An insulating jacket of asbestos paper and a protective jacket of steel shim stock were applied over each element. A photograph of the heaters appears in Fig. 51, and details of construction appear in Figs. 52 and 53.

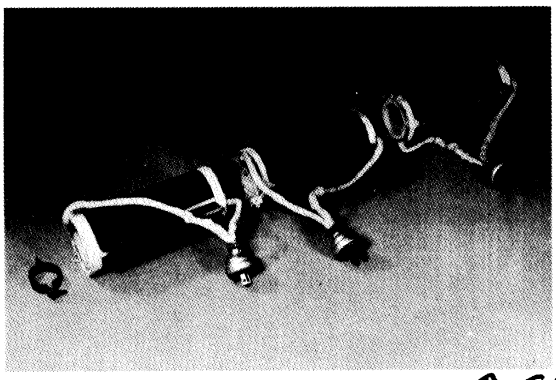


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Fig. 51. Heaters for Pressure Reactor.

A steel rack was designed and built for the purpose of positioning the stainless-steel reactor approximately symmetrically with respect to the 10-kilocurie source; see Figs. 54 and 55 for details. Extension legs (Fig. 56) permit use of the rack to hold the reactor on the axis of the source. The new steel

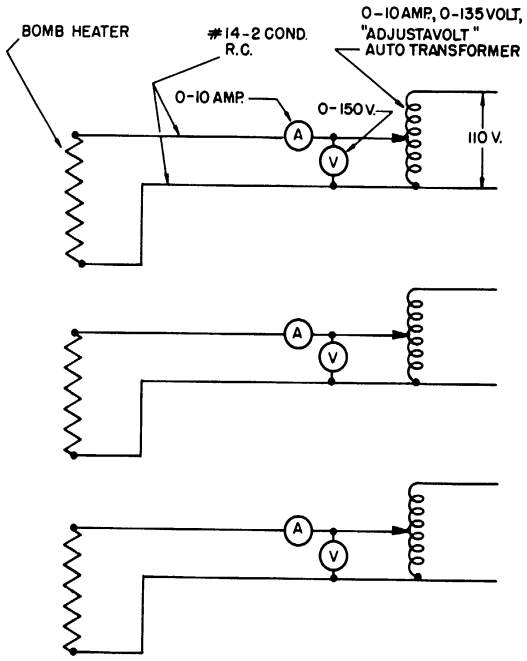

Fig. 52. Wiring Diagram for Heaters for Pressure Reactor.

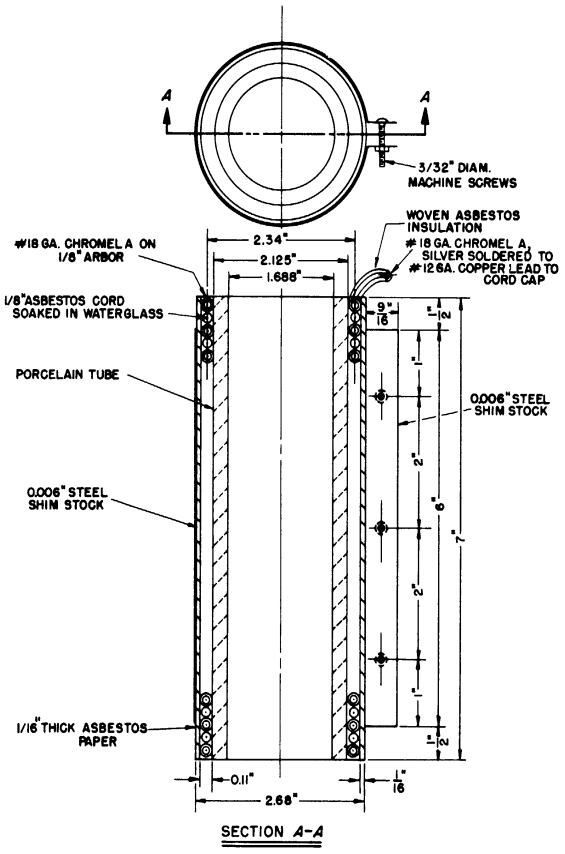

Fig. 53. Details of Heaters for Pressure Reactor.

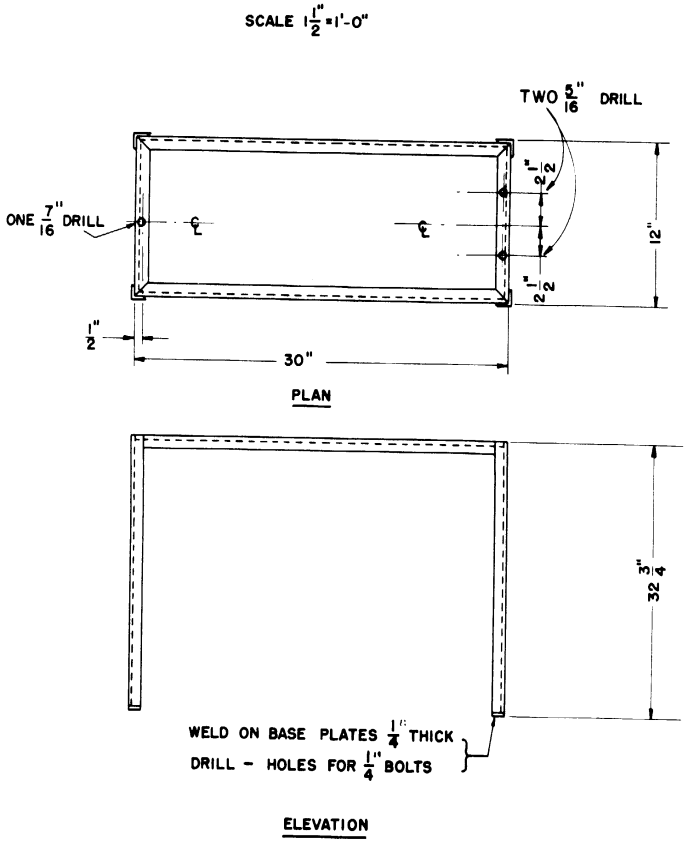

Fig. 54. Rack for Pressure Reactors.

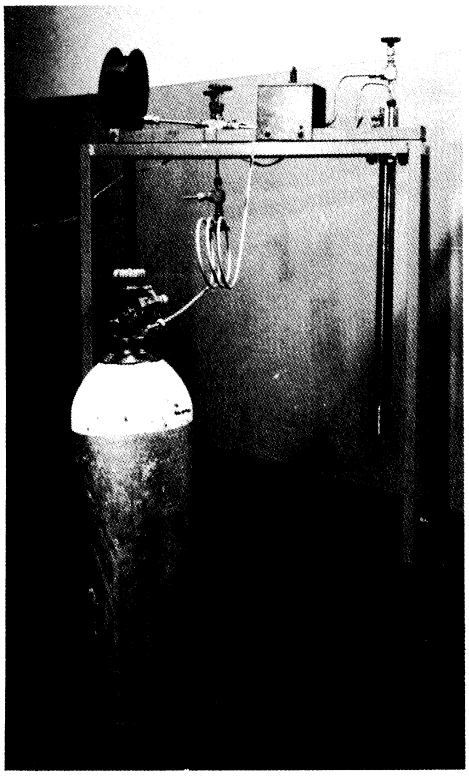

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Fig. 55. Pressure Reactor in Rack and Connected to High Pressure Gas Cylinder.

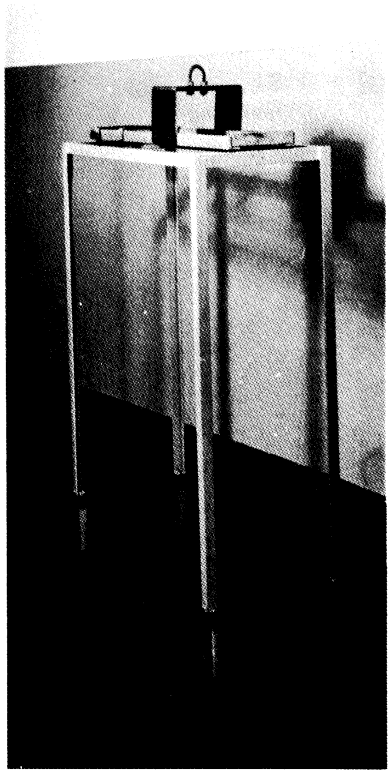

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Fig. 56. Rack for Pressure Reactor.

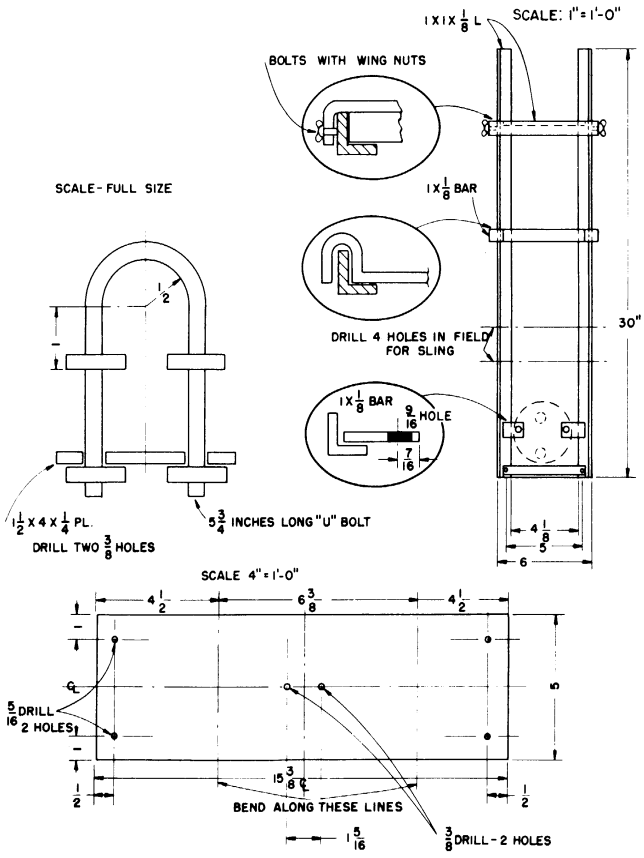


Fig. 57. Sling for Pressure Reactor.

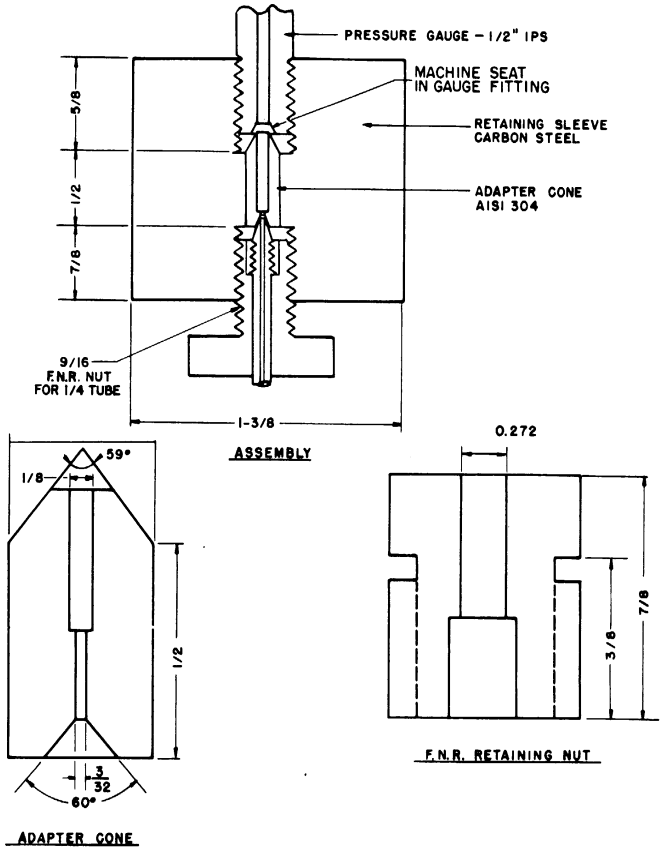


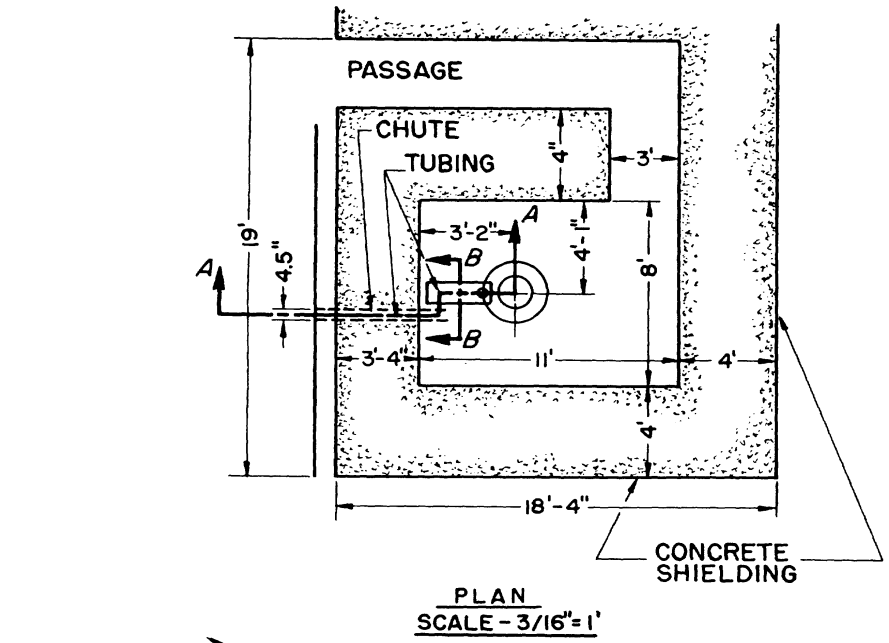
Fig. 58. Cone Joint to Iron Pipe Thread Adapter.

rack was designed to accommodate the sling (Figs. 56 and 57) previously constructed for the purpose of supporting the reactor with its attached tubing in the 1-kilocurie source.

A special connection was constructed to permit attaching a compression tubing fitting to a pressure gauge having an iron-pipe-size thread. The details are given in Fig. 58. The pressure gauge fitting was machined out to provide a cone seat to match the standard cone on the end of the tubing. This kind of joint was used to provide a better seal than was thought to be possible by the use of tapered pipe threads.

An access opening was provided during the construction of the shielding for the 10-kilocurie source; see Figs. 59 and 60. Electrical leads and aluminum tubing lines were placed in this opening to permit the operation of the pressure reactor remotely from the second floor of the laboratory. Two copper leads were also carried through this opening to connect a thermocouple cold junction in the vault with a potentiometer on the second floor. Assembly of the screwed compression joints for the aluminum tubing was accomplished by means of specially constructed socket wrenches (Fig. 61). Two wrenches were required, one to be inserted from each leg of the right-angled access opening and over the length of tubing extending out from the tubing ell. The wrenches were constructed by brazing spark-plug wrenches to the ends of pieces of electrical conduit of appropriate diameter. The socket was then turned by means of a pipe wrench. A special wooden holder was devised in order to protect and support glass tubing introduced into the same opening with the electrical leads and aluminum tubing.

A modified form of the original glass chlorination reactor has been designed and constructed for use with the 1-kilocurie cobalt-60 source. This equipment is illustrated in Figs. 62 and 63. The use of concentric tubes joined at the top by means of ground joints facilitates cleaning of the reactor and at the same time assures a tighter seal for the gaseous contents than the earlier design afforded. In runs where a large heat of reaction is not anticipated, cooling may be done with water. The water is in turn cooled by recycling through a chamber containing ice frozen around a can containing a mixture of chloroform, carbon tetrachloride, and dry ice. If a large heat of reaction is expected, the water may be replaced with methyl alcohol cooled well below the freezing point of water. A knife-type heater has also been installed, actuated by means of a double-throw switch on the temperature controller, in order to provide better temperature control.



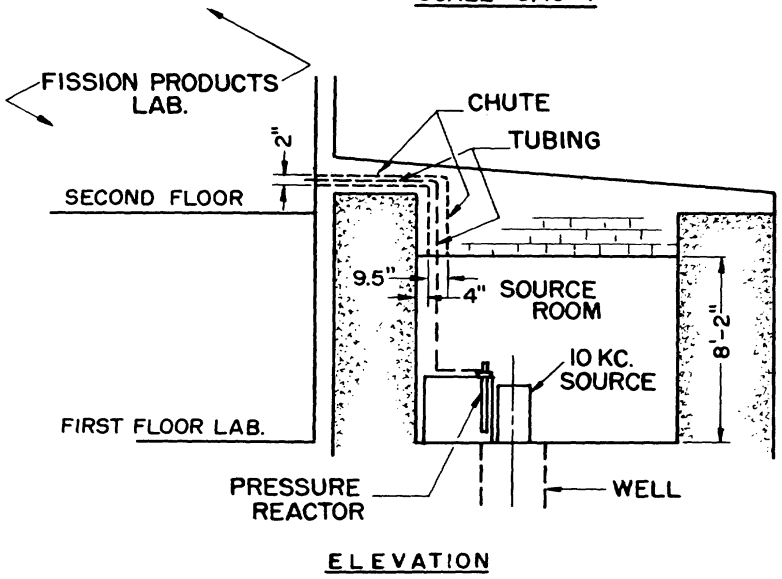
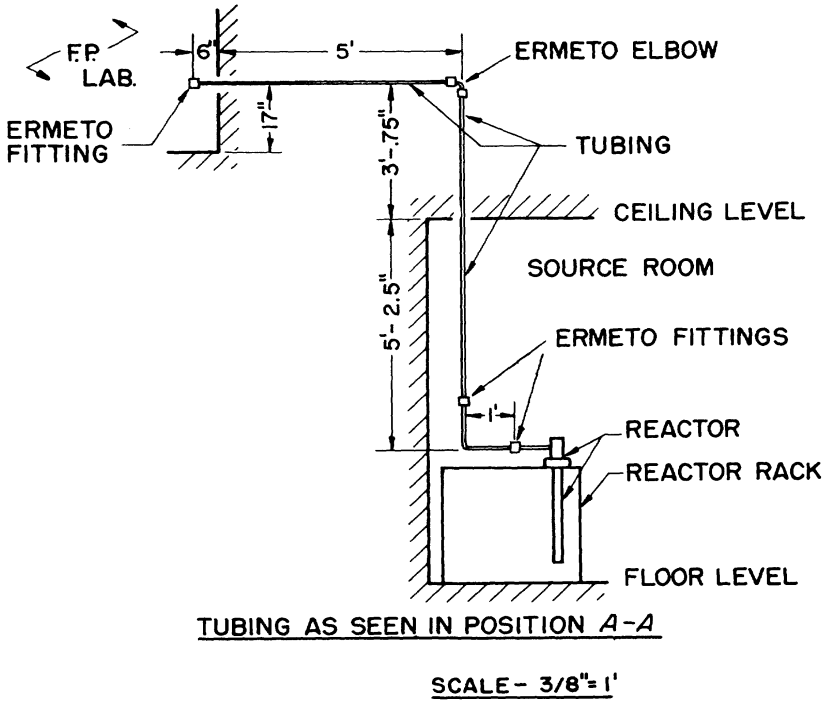


Fig. 59. Location of Tubing Between Fission Products Laboratory and 10-kc Source.



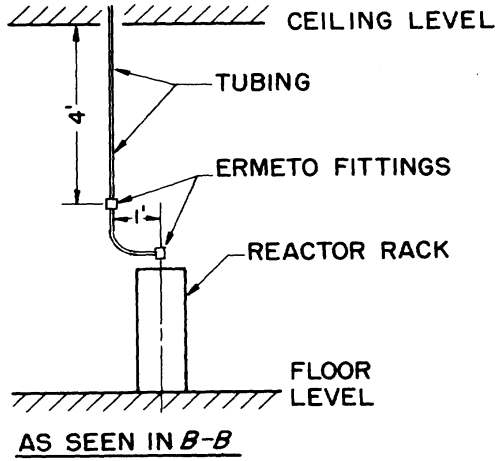


Fig. 60. Details of Tubing Shown in Figure 59.

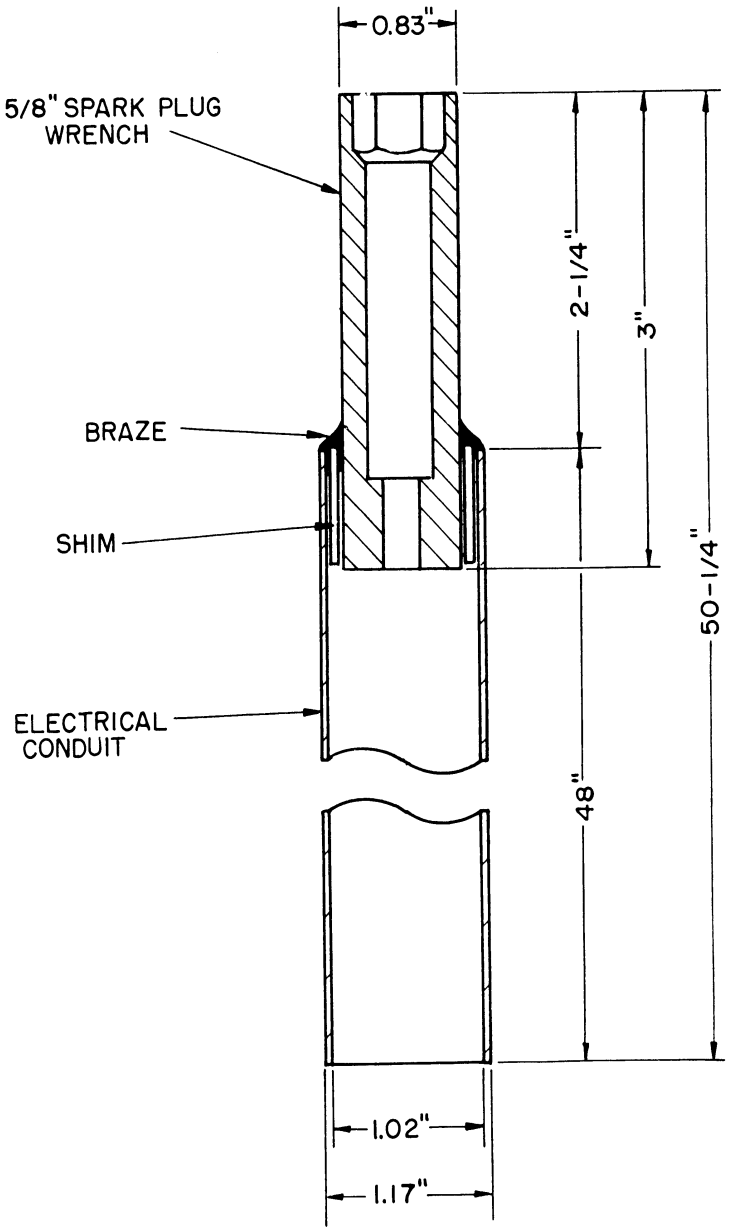


Fig. 61. Socket Wrench Brazed to End of Electrical Conduit Used for Installing Aluminum Tubing for 10-kc Source.

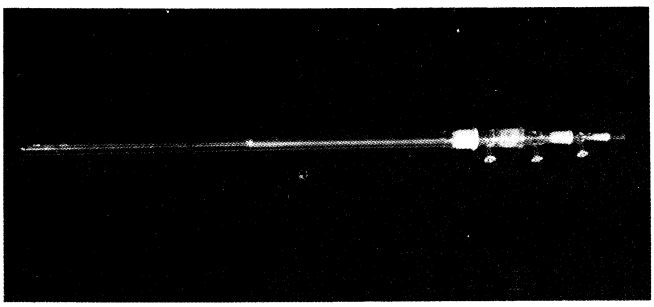


Fig. 62. Glass Apparatus Used in Chlorination Runs.

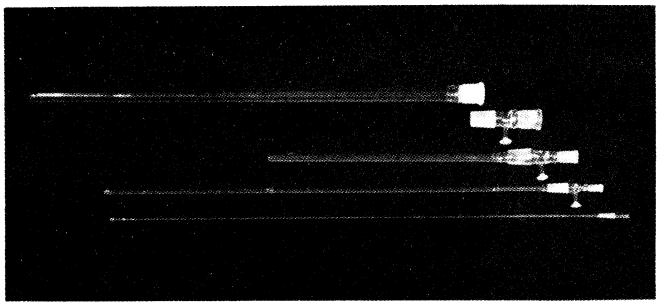


Fig. 63. Glass Apparatus Used in Chlorination Runs.

G. FUTURE WORK

It is expected that the work in the near future will concentrate on the following objectives: (1) Determine the conditions that cause ethylene to polymerize (and also not to polymerize) under the influence of gamma radiation. (2) Analyze more completely the products produced by irradiating ethylene. (3) Continue studies on the analysis of products from the chlorination of toluene. (4) Chlorinate other hydrocarbons. (5) Study the kinetics in the chlorination of benzene under gamma radiation. (6) Perhaps try some partial oxidations of olefins and aromatics and also some nitrations.

H. REFERENCES

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(2) Bretton, R. H., et al., Effect of Gamma Radiation on Hydrocarbon Gases, Progress Report IV (NYO-3311) Dept. of Chemical Engineering, Yale University, New Haven, Conn., October 30, 1952.

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III. COOPERATIVE RESEARCH WITH MICHIGAN MEMORIAL-PHOENIX PROJECTS

A. INTRODUCTION

As explained in the preface, this report includes a description of various research projects supported by The Michigan Memorial-Phoenix Project in which the irradiation facilities in the Fission Products Laboratory have been used. This material has been included because it is believed to have considerable bearing on the possible utilization of the waste fission products. Most of the material presented in this manner in this report will appear at some later date in the technical literature. No portion of this report dealing with work supported by the Michigan Memorial-Phoenix Project is to be reproduced without permission of the authors.

B. BREAKING THE CYCLE OF TRICHINOSIS BY GAMMA IRRADIATION

Personnel:

H. J. Gomberg, Assistant Director of the Michigan Memorial-Phoenix Project, Associate Professor of Electrical Engineering, and Research Associate with the U.S. A.E.C. Project on Biological Effects of Irradiation at the University of Michigan; S. E. Gould, Clinical Professor of Pathology at Wayne University, Pathologist at Wayne County General Hospital, Eloise, Michigan, and Research Associate with the U.S. A.E.C. Project on Biological Effects of Irradiation at the University of Michigan; and J. V. Nehemias, Research Associate and Health Physicist, Fission Products Laboratory, University of Michigan.

1. Errata in Progress Report 4

In Progress Report 4, an introductory explanation of these experiments was made as follows:

> "These experiments were first undertaken using x-radiation in the U.S. Atomic Energy Commission Laboratory on Biological Effects of Irradiation, University of Michigan. The results of the study using x-radiation have been described in an article, 'Effect of X-Rays on Trichina Larvae', American Journal of Pathology, 29:323-337, 1953.

This research has been continued with support of Phoenix project 54 on a cooperative basis with the Fission Products Laboratory."

It has since been brought to the attention of the Fission Products Laboratory that this explanation was only partially correct, in that the U.S. Atomic Energy Commission Laboratory on Biological Effects of Irradiation under the supervision of Dr. F. H. Bethell had continued the support of this study, including the work reported in Progress Report 4, on samples of rat tissue infected with trichinae and treated with gamma radiation in the Fission Products Laboratory. Michigan Memorial-Phoenix project 54 supported the experiments on large sections of pork carcass exposed to gamma radiation from the 10,000-curie cobalt-60 source in the Fission Products Laboratory, and on pork-carcass sections exposed to gamma radiation from fission products in spent reactor fuel rods at Arco, Idaho.

2. Introduction

A brief review of the problem of trichinosis in man was given in Progress Report 4. Vernon B. Link of the Communicable Disease Center, U.S. Public Health Service, Atlanta, Georgia, in the opening paper of the First National Conference on Trichinosis held in Chicago in December, 1952, expressed the opinion of many in the field of public health: "In any case, let us not continue with our customary laissez-faire policy toward this preventable disease; let us rather proceed to correct these conditions in our country which we and the whole world know are probably worse than in any other civilized area."

The following section of this report has been reproduced from a manuscript which was presented at the Conference on Nuclear Engineering at the University of California in September, 1953. No parts of the following excerpt from this manuscript are to be reproduced without permission of the authors, H.J. Gomberg and S. E. Gould.

3. Irradiation Study Methods

a. "Introduction to Irradiation Techniques

"To kill the trichinae in situ, about 750,000 to 1,000,000 rep are needed.[1,2] This dose is prohibitively high because of both the cost of the irradiation and because of the deleterious flavor changes which are induced in the pork, turning it rancid and sour.[3]

"However, 1/50 of the above dose, 20,000 rep of Co^{60} gammas, is sufficient to prevent maturation of the larvae to adults and about 1/80, 12,000 rep, will sterilize the females, preventing reproduction;[2] 200-kilovolt x-rays

produce similar results with somewhat lower doses.[4,1] Either of the above effects will prevent the invasion of muscle tissue (myositis) by trichina larvae.

"The results cited, however, have all been obtained by irradiation of relatively small sections of infected material and do not reflect an approach to practical conditions.

"To break the trichinosis cycle most effectively, it is desirable to irradiate the whole carcass so that any cuttings which are returned to feed will be treated. During processing, the hog carcass is gutted, split, and then cooled for 24 hours before cutting. This delay period was selected as a good irradiation time and tests were made to simulate this condition.

b. "Techniques Employed in Present Tests

"A series of tests on sections of hog carcass using the University of Michigan 10-kilocurie radiation source was made. In addition two trichinous hog sections were irradiated with fission products, using old fuel slugs from the Hanford Reactors which were in storage at the American Cyanamid Chemical Processing Plant at Idaho Falls.

"(1) Cobalt-60 Irradiation. (a) Description of equipment:[5] A curve of dose rate in air as a function of distance for the 10-kilocurie source is shown in Fig. 100. Examination of this curve reveals that beyond 10 inches from the source surface, it may be treated as a point source. All the meat irradiated was located at least 13 inches from the surface of the source. Knowing the dosage pattern in air and using an 8-inch effective half-value layer in meat for Co^{60} radiation, the dose at any point in the section could be calculated.

"(b) Procedure and dosimetry: The most significant test of the cobalt series was made with the hog section oriented so that the length along a radius was 14 inches. The whole section, a hind quarter of a 450-pound hog, weighed about 50 pounds and measured about 14 by 8 by 16 inches. The section was oriented so as to obtain maximum variation in the dose delivered to various depths of the section. In addition to dose determination by calculation, ferrous sulfate dosimeters were inserted in the meat at 2-inch intervals so that an independent dose check could be made. The meat was irradiated with the center of the section 17 inches from the source surface for 1/2 hour, then rotated through 180 degrees and irradiated for a second half-hour. Three sets of dosimeters were used, one set during the first half-hour, a second set during the second half-hour (following rotation), and a third set remaining in place for the full hour. A photograph and diagram of the experimental setup are shown in Figs. 64a and 64b. The wire cage surrounds the source when it is in operating position. A set of four calibration (in air) dosimeters was also used.

Fig. 64a. Photograph of Section of Pork Showing Dosimeters in Position with Wire Cage of Source in Background.

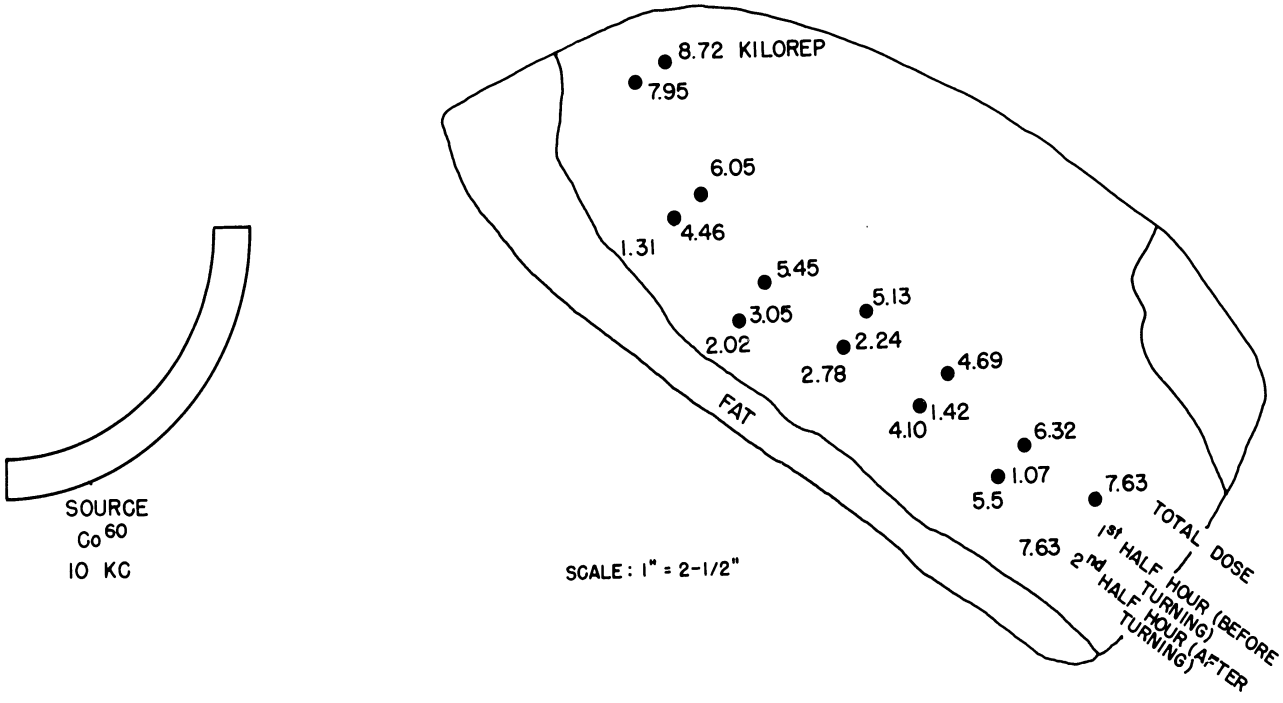

Fig. 64b. Diagram Showing Relation of Pork and Dosimeters to 10-kc Cobalt Source.

"(2) Waste Fission Product Irradiation. (a) Description of equipment: In an attempt to simulate possible large-scale operations, arrangements were made to irradiate 50-pound hog sections, using old fuel slugs from the Hanford reactors in storage at the American Cyanamid Company Chemical Processing Plant, Idaho Falls, Idaho. While it was desired to use principally cesium-137 radiation, the oldest slugs available were about 2 years old, and according to Hunter[5] the gamma radiation for 2-year-old fissioned uranium is 86 per cent 1.2 mev from Pr^{144} and 14 per cent 0.6 mev, approximately, from Ba^{137} and Rh^{106}. Although the spectrum is, therefore, close to that of the cobalt already used, the experiment was carried out to study the effect of differences in operating conditions."

The fuel rods were assembled into two plane rectangular arrays at the bottom of the 20-foot-depth water canal in the Storage Building. The resultant plaques were about 30 inches long and 27 inches wide, and the rods were packed as closely as possible with 20-foot manipulation tongs. All work was done standing over the plaques on an iron grill covering the canal.

"Before irradiation tests were made, the radiation field was mapped using a Victoreen roentgen rate meter. The probe was enclosed in polyethylene tubing, weighted so that no scattered radiation from the weight would reach the measuring thimble, and the probe was then lowered through the water to known positions above the source. Isodose curves derived from these tests are shown in Fig. 65 for the north source. The isodose surfaces closely approximate surfaces generated by rotating these curves in a horizontal plane about the central axis of the source. The variation of dose with distance from the plaque surface has been investigated theoretically and is commensurate with radiation having a 12-inch half-value layer in water. Taking Compton secondaries into account, this half-value layer applies to either 0.5-mev or 1.2 mev gamma radiation in water[6] and so applies to radiation from the fuel slugs.

"(b) Procedures and dosimetry: Two sections, a hind quarter and a fore quarter, of a hog carcass containing encysted trichinae were used for the test irradiation. Before irradiation, lusteroid vials containing ferrous sulfate were inserted into the pork so that the dose distribution within the meat could be determined. The sections were then wrapped in thin plastic sheets to minimize water-logging of the meat, then weighted so that they would sink and lowered into the canal at the end of a rope. The rope was tied when, as well as could be estimated, the bottom surface of the meat was 5 inches from the irradiator surface. The average dimensions of both sections were 15 by 20 by 8 inches. A sketch of the experimental layout is shown in Fig. 66.

"Due to shrinkage of the rope under water, and water currents in the canal which shifted the sections with respect to the irradiators, conditions could not be maintained constant throughout the exposures. The exposure times for the sections were selected so that the bottom layer of the second section,

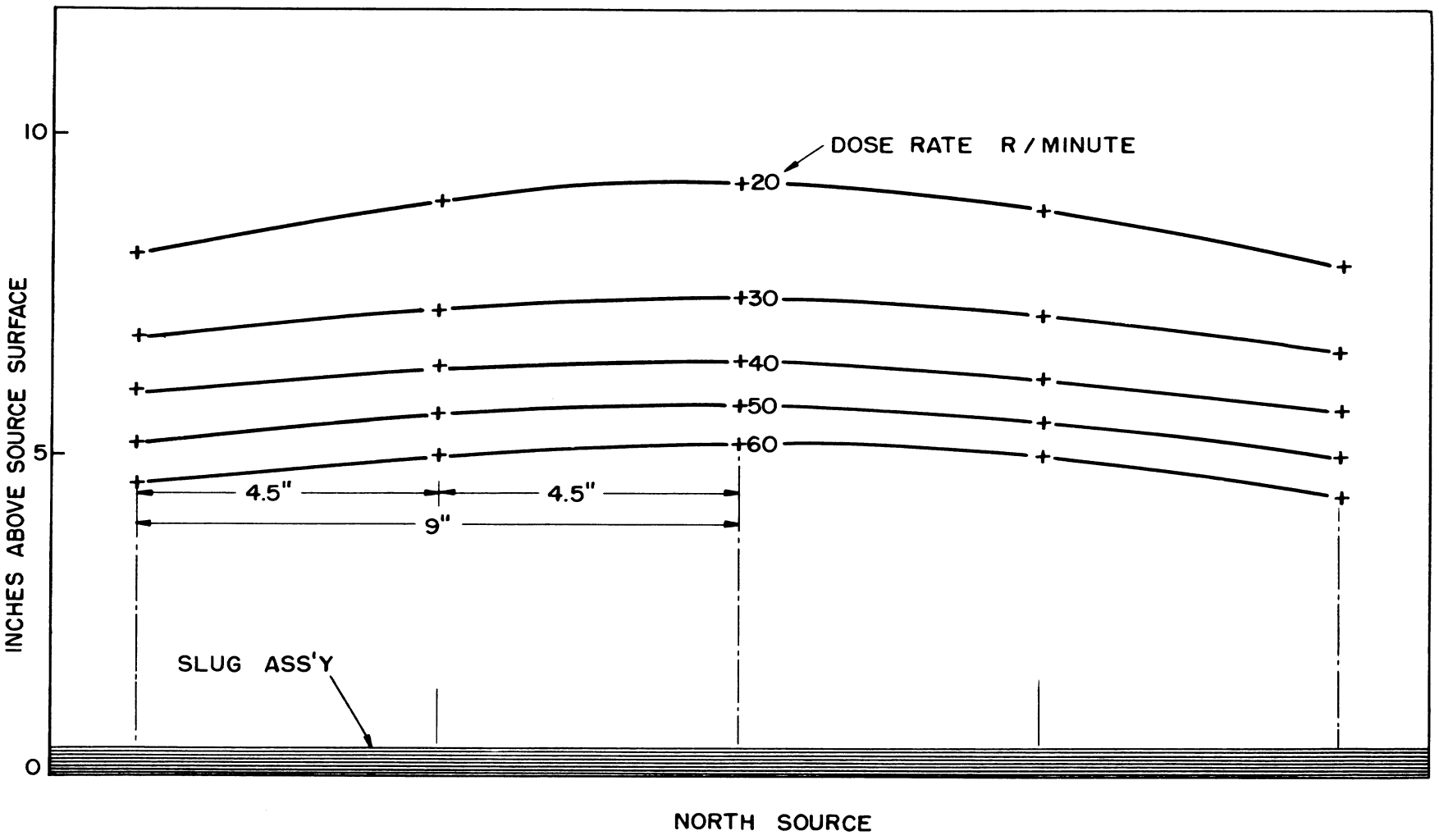

Fig. 65. Dose Rates vs Position over Array of Old Fuel Slugs in Canal at CPP.

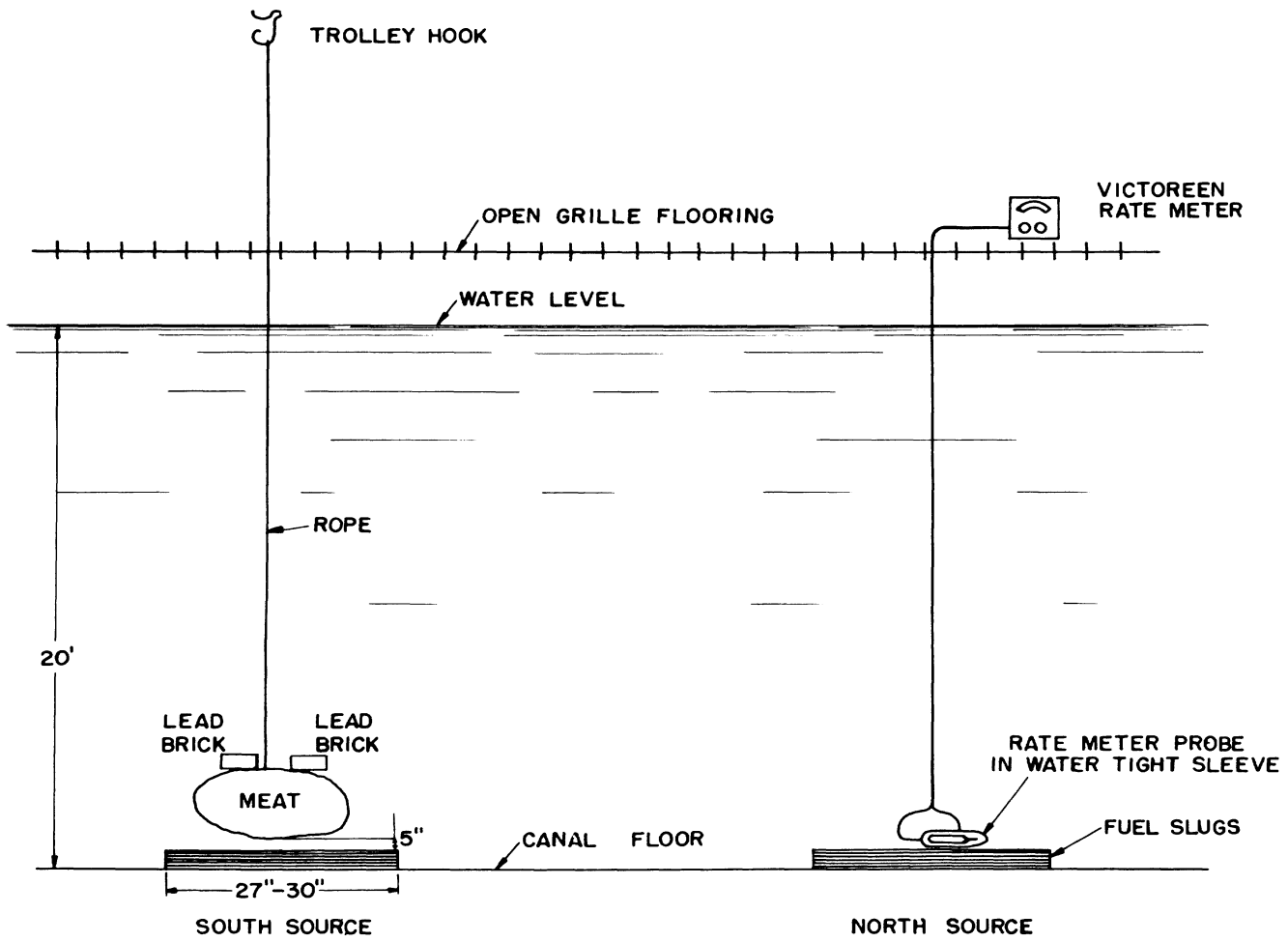

Fig. 66. Irradiation Arrangement Used in Canal at CPP.

which received the largest dose in that section, would be irradiated slightly more than the top layer of the first section. In this way the two sections would show a wide range of dose distribution, including a small overlap. The hind quarter was irradiated for 320 minutes and the fore quarter for 1185 minutes.

"In addition to checking dose rate as a function of position with respect to the source, several ferrous sulfate dosimeters, lusteroid and glass, were attached to the Victoreen probe and lowered into the water to fixed positions either before or after the meat irradiation. From these tests it was found that the glass - ferrous sulfate dosimeters gave results 7/6 as high as the Victoreen meter reading. The same factor was observed in the cobalt-source calibration. It was also found that the ratio of glass dosimeter to lusteroid dosimeter readings was 0.55, due apparently to an oxidizing agent produced by irradiation of lusteroid. In our dosimeters, which contained aerated solutions, the maximum dose measurable before the dissolved oxygen was exhausted was about 28,000 r for glass or 13,950 r for lusteroid.

"As discussed in the section on results, after correcting for shifting of the meat the agreement between predicted dose, from known dose distribution in water, and the dose measured in the meat by dosimeter was quite satisfactory."

4. Results

a. "Cobalt-60 Tests

(1) Radiation Measurements in Meat. A comparison between the calculated dose in the meat and the measured dose delivered by the cobalt source for the two separate half-hour runs is presented in Fig. 67. The agreement is considered satisfactory for the experimental conditions.

"The doses measured in the meat are shown in Fig. 68. The most significant result is the final dose distribution in the meat after rotation. The total variation is about 2 to 1 from the surface to the interior for a 14-inch-thick section. Of this change, less than 20 per cent is due to attenuation by absorption in the meat. These results could be checked quite closely by calculation, using an 8-inch half-value layer for the cobalt gammas in meat. As an example, the calculated dose at the center of the meat was 5225 r and the measured value was 5130 r. A second calculated dose distribution in a 14-inch-thick slab, based on an 8-inch half-value layer but with no geometric attenuation, indicates a dose variation of 12.5 per cent from surface to the interior. This is, of course, a limiting value, never quite attainable in practice.

"(2) Biological Effects. The biological effectiveness of the radiation is demonstrated in Figs. 69a and 69b. Sterilization of the trichina larvae is

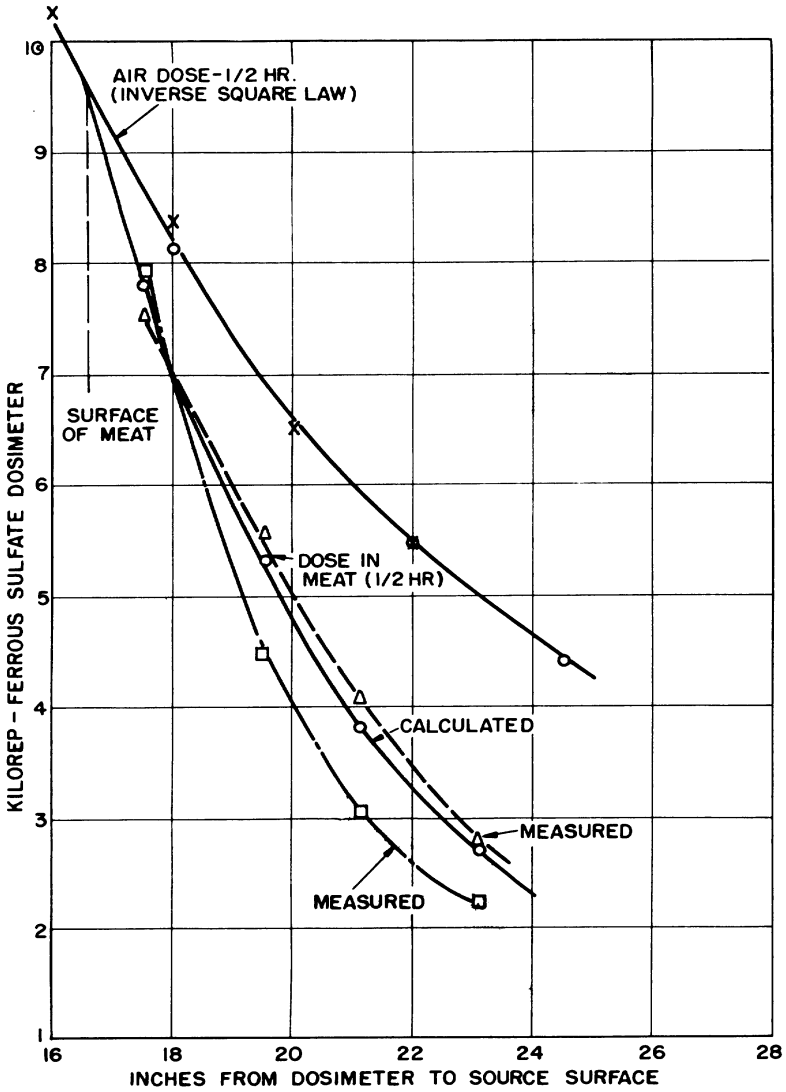

Fig. 67. Roentgen Dose in Air and at Corresponding Points in Meat, Calculated and Measured.

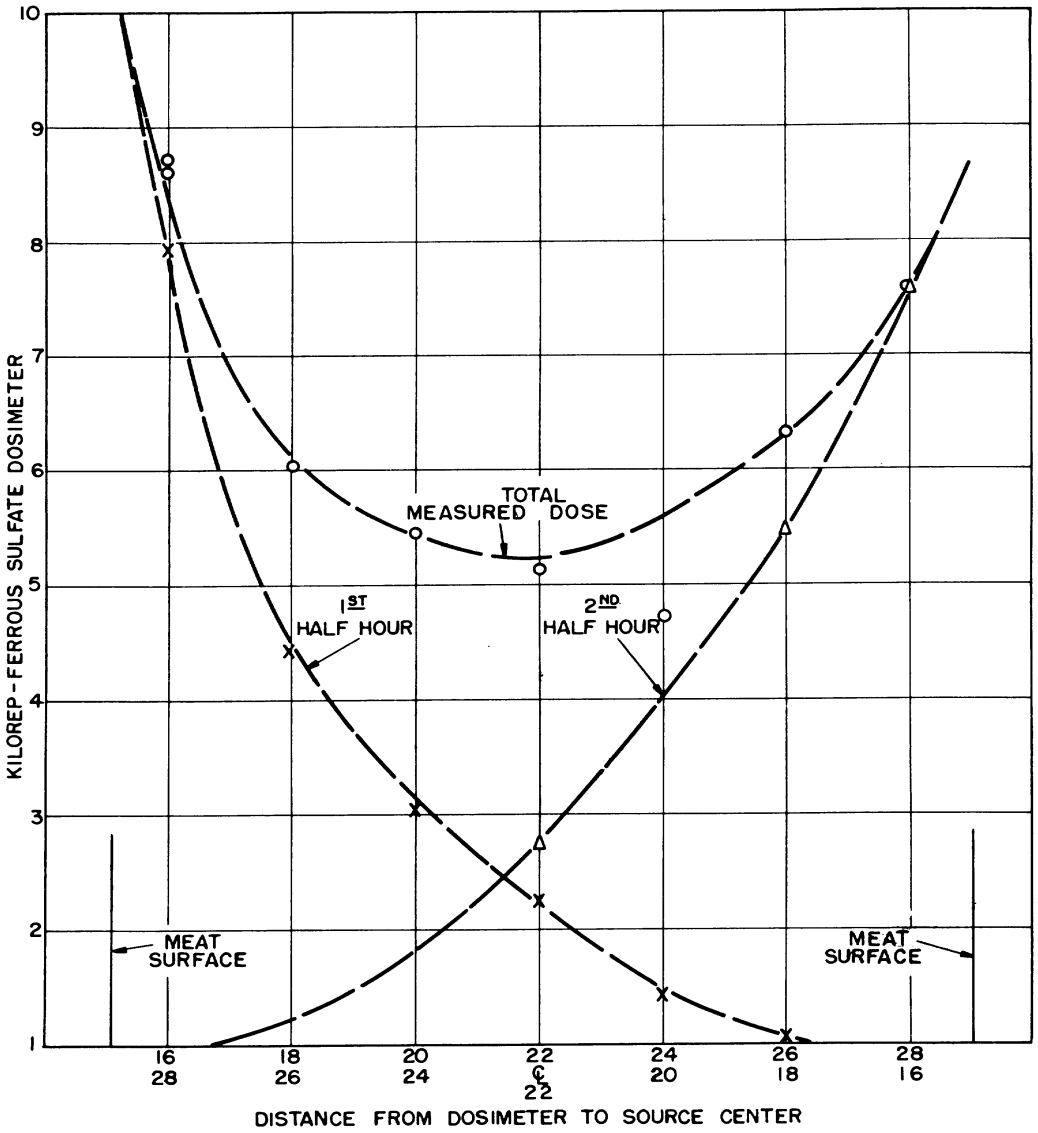

Fig. 68. Measured Doses Delivered to Pork 14 inches Thick.

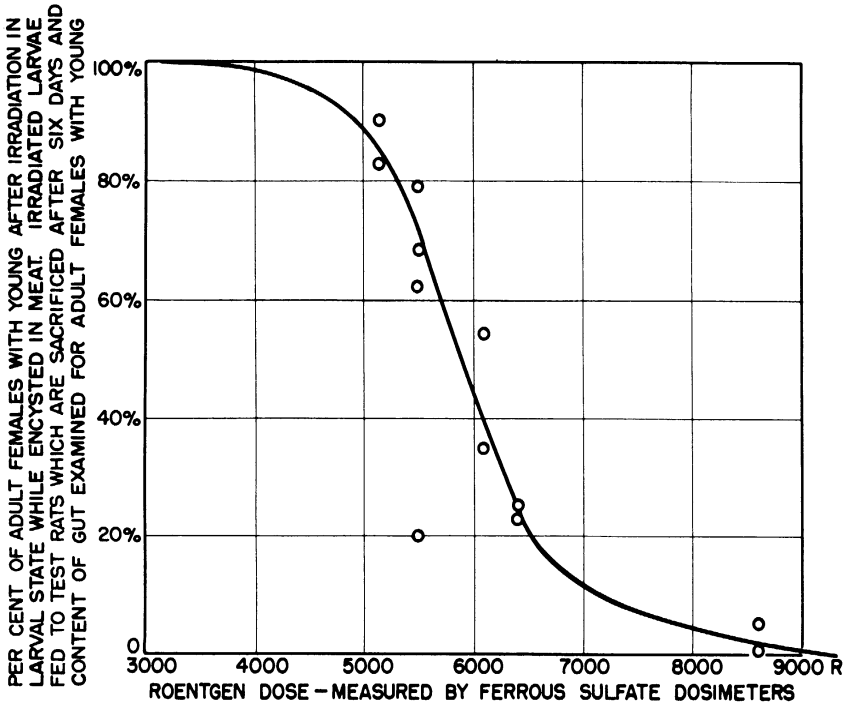

Fig. 69a. Irradiation of Trichinous Pork Using 10-kc Cobalt-60 Source.

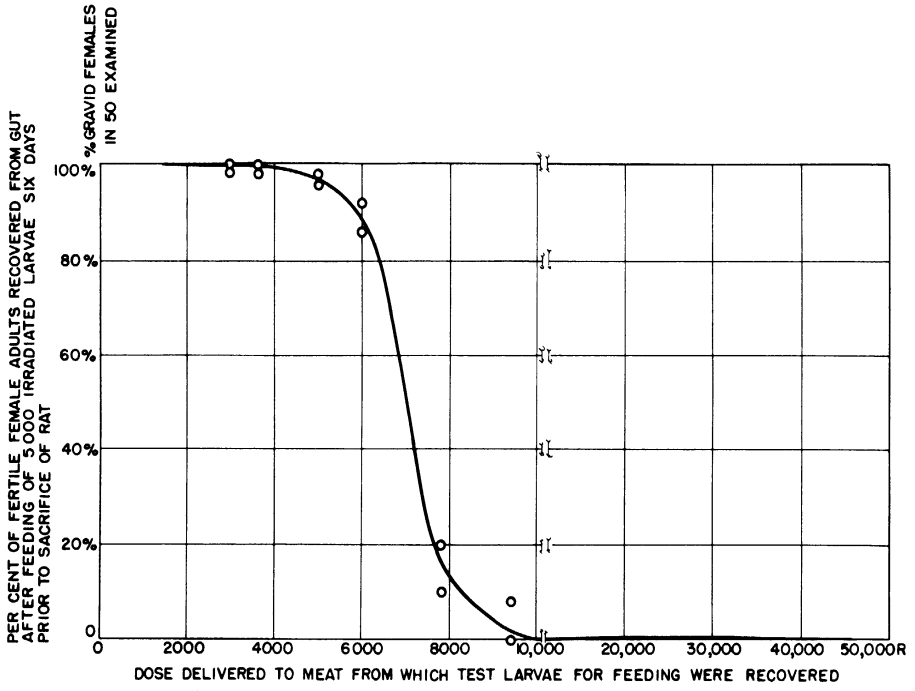

Fig. 69b. Irradiation of Trichinous Pork Using Gammas from Spent Fuel Slugs.

complete at somewhat more than 10,000 r. This is in keeping with determinations made by irradiation of small sections of pork and rat carcasses.[2]

"The number of encysted trichina larvae recovered from the muscle tissue of the 30-day-test rats is shown in Fig. 70. The very rapid depression in the number of encysted trichinae is significant. The best fit to the data appears to be a straight line, implying that inhibition of encystment by trichina larvae is a 'one-hit' effect. Figure 70 also includes the data obtained in the Fission Products Laboratory irradiation. The nature of the results is the same in both cases. Further tests are needed to determine whether the effect noted is due to damage of the germ cells in the female larvae so that a smaller number of young are born, or if the young developing from these germ cells are defective and cannot follow the normal course of invasion and encystment. In this connection, the excellent and very complete work of Alicata[4] is of interest, particularly in the photomicrographs showing a reduced number of fully developed egg cells in female adults which received less than the sterilizing dose.

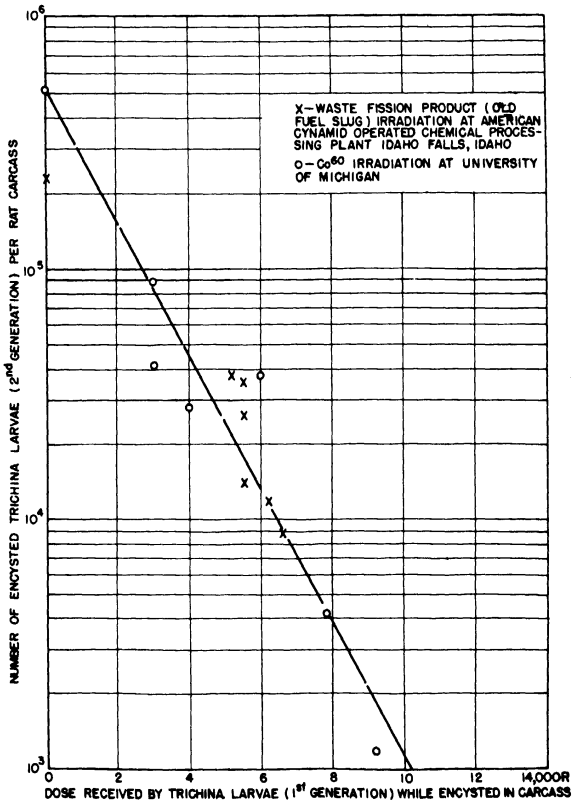

Fig. 70. Larvae in Rat Muscle 30 Days after Feeding 5000 Irradiated Larvae Using Cobalt-60 Gammas.

"It will be noted from Figs. 69a, 69b, and 70 that, at the highest dose received in this experiment, the sterilization process had not been carried to completion. In a similar experiment in which higher doses were used but all doses were calculated instead of measured, a 12,000-r dose resulted in complete sterilization of the females and complete absence of encysted trichina in the test rats.

b. "Waste Fission Product Tests

"(1) Radiation Measurements in Meat. The hind quarter of the carcass was irradiated for 320 minutes, providing measured doses up to about 9000 r at the dosimeter closest to the source. This dose was somewhat lower than had been estimated for the selected position of the meat. Analysis of the data obtained from the dosimeters showed that, due to unequal buoyancy, the section had rolled slightly, so that the isodose surfaces in the meat were not quite parallel to the bottom surface of the meat slab. Also, by checking the dose distribution in water as determined by the Victoreen rate meter against that given by the dosimeters in the meat, it is estimated that the bottom of the meat was 8 inches rather than 5 inches from the source. This may have been due to a minor inaccuracy in positioning of the meat or to shrinkage of the rope. Since the dose rate changes fairly rapidly in this region (Fig. 65) the dose received would be affected noticeably. Fortunately, the dosimeters provided dose measurements, so that the biological effectiveness could be determined accurately.

"The fore quarter was irradiated for 1185 minutes to provide doses overlapping slightly those in the hind quarter and increasing from there. Actually, due to the unexpectedly low dose in the hind quarter, there is a discontinuity from 9000 r to 13,000 r, the dose indicated by the dosimeter in the fore quarter farthest from the irradiator. At the time of the experiment, it was not known that the lusteroid dosimeters would saturate at 13,750 r. This information, cited earlier, was learned from this test. As a result, most information on the dose rate in the meat was obtained by calculation. However, the check between the dosimeters which did not saturate and the predicted dose for their position fell within experimental error.

"(2) Biological Effects. Complete sterilization of the female larvae and the prevention of infection as determined by 6-day and 30-day tests described previously occurred at 10,000 r. Results of the sterilization tests are shown in Fig. 70 and the effect on the number of larvae found in the rat muscle tissue is shown in Figs. 69a and 69b. Data for these curves came entirely from the first or hind-quarter irradiation test. The fore quarter, which received a dose from 13,000 r and higher, yielded trichina larvae which were all completely sterile if they developed at all. None of the rats which received these larvae showed any infection.

5. Discussion

"It has been shown that techniques suitable for sterilizing trichinae in small samples of infected meat can also be used for large full-weight sections. Further, by proper selection of the source geometry and by irradiating the meat from both sides, variation in dose through full-weight sections as much as 14 inches thick can be kept to much less than 50 per cent. If we are to consider this process of pork treatment seriously, some estimate of energy demand must be made. According to Dr. J. G. Trump,[7] 12 kilowatts of ionizing radiation will deliver 1 x 10^6 r to 5 tons of food per hour. On this basis, 12 kw will sterilize trichinae in 2 x 10^6 tons of pork per year, based on a delivered dose of 20,000 r.

"The total production of hogs in the United States in 1952 was 64 x 10^6. At about 200-250 pounds per hog and about 150 pounds of material to be irradiated per hog, the total irradiation load would be 5 x 10^6 tons, or 30 kw of gamma power for the whole country. This much power is obtained from 10^5 curies,[8] a modest amount by present production standards. Since 12,000 r will prevent infection, and insignificant flavor changes occur in pork irradiated with doses up to about 40,000 r, the available operating range for treating pork is more than adequate. From source design studies not discussed in this paper, it is known that variation of much less than 50 per cent over a 12-inch section is feasible."

(The following discussion is taken from a paper by S. E. Gould to be presented November 10th before the Laboratory Section of the American Public Health Association at the 81st Annual Meeting in New York, N.Y.)

"Because of the lack of adequate control of trichinosis in the United States, it is no wonder that the great frequency of trichinosis in our country has long been a source of 'astonishment and horror by many other people in many other countries.' Let us examine briefly the reasons for our failure to institute an adequate program. We find that microscopic inspection of pork was deemed not suited to our high-speed processing methods, even though it has been practiced in many other countries for many years apparently with a large measure of success. The examination of several specimens of meat from every carcass would require a formidable army of inspectors and would impose a substantial additional cost per pound of pork produced. In addition, as previously stated, there is frequently the danger of failure to detect trichina larvae that are present in the specimens examined.

"A second possible method of control that could be instituted is that of low-temperature treatment of all pork. While the method is applied to certain products, the costs of producing the necessary low temperatures make this method too expensive to be applied to all pork produced.

"A third method, that of boiling all garbage that is fed to hogs, has during 1953 been instituted in most states of the United States. It may be noted that this requirement was adopted by the legislature of 41 states not as a means of controlling trichinosis in man, but as an economic measure to protect the pork industry against the ravages of vesicular exanthema, a disease of hogs that is also transmitted by feeding of raw garbage. As a control measure for trichinosis, this method is desirable and beneficial in that it tends to reduce the incidence of the infection. And yet, among so-called farm-fed and grain-fed hogs, and in Canada where garbage fed to hogs must be cooked, the incidence of infection is still appreciable, ranging from 0.3 to 0.7 per cent. In all likelihood, this degree of infection is the result of permitting hogs to eat kitchen scraps containing infected pork. Furthermore, the states of California, New Jersey, and Rhode Island, in which feeding of raw garbage to hogs has been most extensively practiced, still allow this practice to continue.

"The method of irradiation provides direct protection to the consumer and has many advantages over other methods:

1. It is sanitary. In the dairy industry two approaches to the control of disease are in use. One is sanitation throughout all phases of production and processing, the other is pasteurization of the final product. In the processing of pork the irradiation process is analogous to pasteurization. Irradiation of pork is not a substitute for sanitation, but it guarantees protection to the consumer should sanitation be defective. The practice of feeding garbage to hogs is insanitary and should not be permitted. Regardless of whether garbage feeding is or is not permitted, the irradiation of pork would protect the consumer.

2. It is simple and rapid. The apparatus required is compact and long-lasting. The time of irradiation required is about 1 minute.

3. It is effective. The meat is rendered free from the danger of trichinosis, thus removing this worry from all who eat pork. The method should eliminate this fear and thus tend to increase the number of potential consumers of pork.

4. The wholesomeness of the meat is preserved. No radioacitivity is induced in the meat, no flavor changes are produced, and no harm results to the person who eats the meat.

5. The method is relatively inexpensive. It requires only a single room, a source, and a technical staff or only one

or two persons for a large packing plant. The estimated cost for such a plant with an estimated daily capacity of 2000 hogs is approximately 2.3 mills per pound of pork produced, if the total investment (interest included) is amortized over a period of 5 years, and approximately 1.5 mills per pound if investment and interest are recovered over a period of 10 years. The cost in subsequent years is even less. The cost of processing pork for a plant having only one-sixth this capacity (about 80,000 hogs/year) would be about 0.74 cents (7.4 mills) per pound. Costwise, the larger and more centralized plant, therefore, would be desirable.

6. The method should largely obviate the need for special processing (heating, freezing or curing) of certain ready-to-eat pork products such as sausages and smoked ham, in order to destroy trichinae larvae. In U. S.-inspected plants these products comprise approximately 30 per cent of the pork produced.

7. The irradiation method could well become exceedingly valuable in advertising, in educating the public to eat pork as an article of diet that is safe besides being nutritious and rich in thiamine (Vitamin B_1). Those producers who adopt the method early should have a tremendous economic advantage. Here again the situation is comparable to that in the dairy industry, in which pasteurization of milk has become one of the principal factors in promoting the sale of milk and its products and in making greater production possible and profitable.

"As yet there is no commercial plant that is equipped to apply gamma irradiation to pork. The use of x-ray and cobalt-60 could theoretically be applied, but no study has been made of the design of construction necessary or the cost of these agents. The use of atomic waste fission material is still under security control by the Atomic Energy Commission and its release for commercial purposes would no doubt be controlled under license. It is assumed that its release for industrial use will be forthcoming at some time in the near future. When such material does become available, it is expected that forward-looking public health workers will cooperate with forward-looking pork producers in introducing this method for making pork safe for human consumption. Such a boon in the interest of the public health would redound to the credit and advantage of everyone concerned."

6. References

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3. Brownell, L. E., et al., "Utilization of Gross Fission Products" Progress Report 3, U.S.A.E.C. Report COO-91.

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C. THE DESIGN OF AN IRRADIATION CHAMBER FOR A PORK PROCESSING PLANT

Personnel:

H. J. Gomberg, Assistant Director of Michigan Memorial-Phoenix Project, Associate Professor of Electrical Engineering, and Research Associate in AEC Biological Effects of Irradiation Laboratory; L. E. Brownell, Director of Fission Products Laboratory and Associate Professor of Chemical Engineering; J. V. Nehemias, Research Associate in Fission Products Laboratory; and E. Coleman, Research Assistant in Fission Products Laboratory.

1. Introduction

This portion of the report is the result of a study supported by Michigan Memorial-Phoenix project 54, supervised by H. J. Gomberg, with some assistance from personnel of the Fission Products Laboratory. It follows the earlier work on the problem of trichinosis supported by the AEC Biological Effects of Irradiation Laboratory and Phoenix project 54 described in Progress Report 4 and Part III B of this report. The calculations and designs were prepared primarily by E. Coleman.

The occurrence of trichinosis and subclinical infections of trichinosis has been well established as a health problem in the United States.[1,2,3] The source of infection to humans is pork infected by the parasite Trichinella spiralis. If infected pork containing viable trichinae is ingested, either by human or hog, the living larvae are released in the digestive tract of the host, mature to adult forms, and copulate. The male soon dies, but the female, lying partially embedded within the mucosa, gives birth to living larvae of the second generation. These young larvae are carried by the circulatory system of the host to fibers of voluntary muscles, where they become encysted. If the host is a hog, the cycle can be repeated if the hog is killed and scraps of the infected pork ingested by a new host. The cycle of the parasite must be broken at some point if this disease is to be controlled effectively.

Although steps have been taken to reduce the incidence of infection of both man and hog, the situation is far from ideal. Attempts to break the cycle have been concentrated mainly on preventing live trichinae from being ingested. The application of heat in curing or thorough cooking of pork is one method used to prevent infection of humans. Cooking of garbage fed to hogs is required by law in more than 50 per cent of the states at the present time. The main disadvantage of such treatment is that it is not nationwide and the remedies are applied largely by individuals as consumers. No treatment is given at the processing plant to fresh pork products.

Work conducted by S. E. Gould, and H. J. Gomberg has shown that x-rays or gamma rays can be used to break the cycle at either of two points.[4,5] A radiation dose of about 750,000 roentgens can be used to kill the trichinae larvae. More important, a radiation dose of less than 20,000 roentgens is sufficient to sterilize the adult female worms and prevent motile larvae from attaining maturity.

The studies on the use of radiation to break the cycle of trichinosis have explored rather completely the effects of radiation on trichinae. The conclusion from these studies was that radiation in rather small dosages of about 20,000 rep is sufficient to "pasteurize" pork so as to prevent reproduction of any trichinae which may infect the meat. Taste studies by panels in the Fission Products Laboratory have indicated that such low dosages of radiation have no undesirable effects on the flavor of fresh pork.

An animal-feeding program is under way using irradiated meat with radiation dosages 100 times or more greater than that required to "pasteurize" pork. There is as yet no evidence contradicting the hypothesis that pork processed with 20,000 rep is wholesome.

This suggests a new approach to the control of trichinosis, since large amounts of gamma-emitting isotopes are being produced as waste fission products in the atomic energy program. Utilization of these waste products for mass gamma irradiation of pork could conceivably eliminate this disease or certainly reduce it to one of minor concern.

For economic reasons, such treatment would probably be applied in large packing houses using modern production-line methods, where the cost would be distributed over a large product volume. This is therefore a report on the economics of design, construction, and operation of large gamma radiation sources for mass-production plants of the pork processing industry.

2. Size of Pork Processing Plant Selected

A visit was made in June, 1953, to the Hygrade Food Products Corporation plant in Detroit, Michigan, to inspect the processing of pork from the live hog to the finished products and to determine the logical point in the production process at which gamma radiation might most effectively be utilized. A schematic diagram of the Hygrade plant is shown in Fig. 71. This plant uses a gravity system to move hogs through the plant. The hogs are slaughtered on one of the upper floors of the building and proceed, largely by gravity, to the ground floor for shipment as fresh meat and by-products.

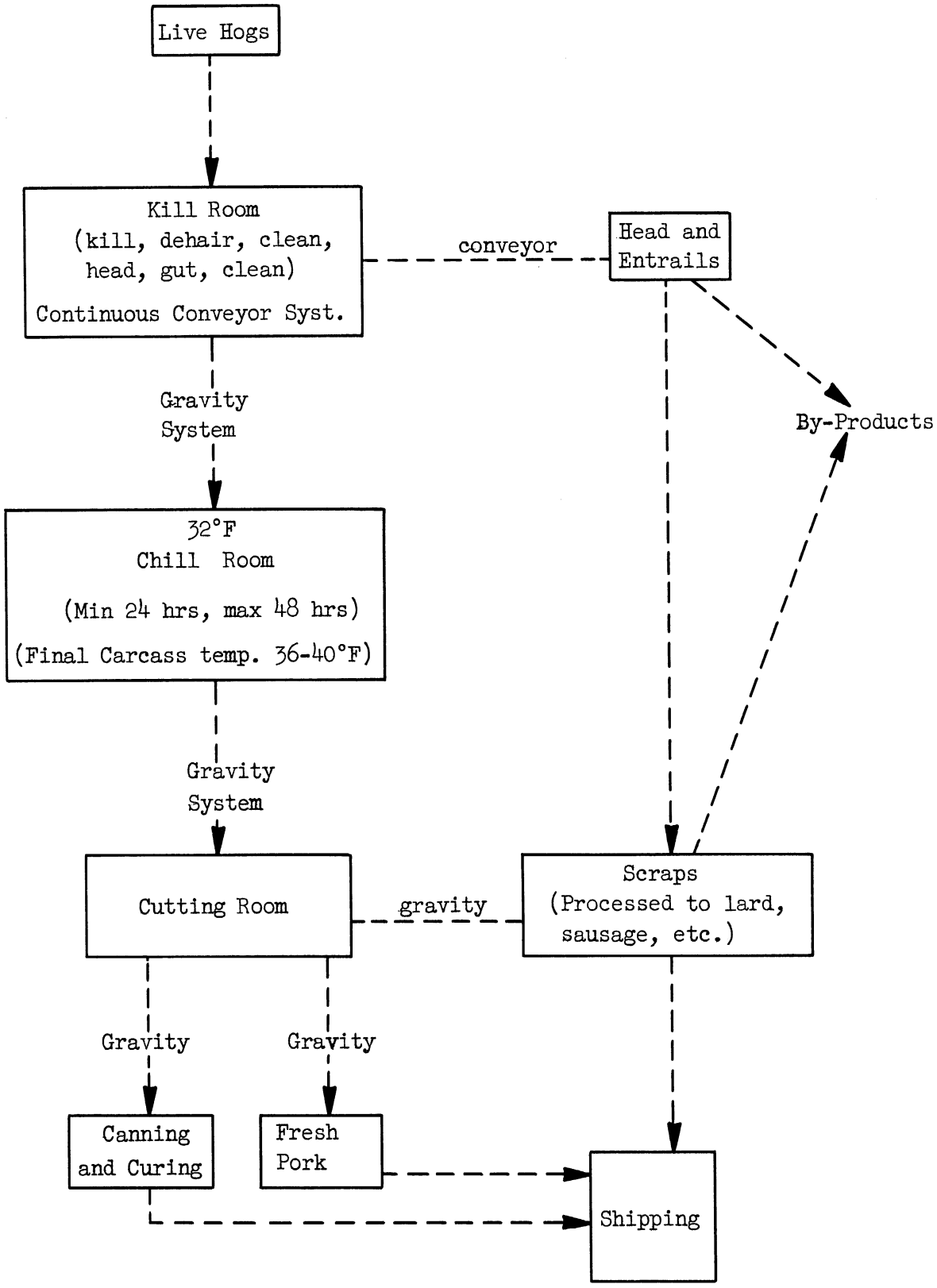

Fig. 71. Schematic Diagram of Hygrade Plant.

The live hog is driven up a ramp to the fourth floor and into the kill room. There the hog is hung head down from an overhead conveyor. The animal is then stuck, dehaired, gutted, headed, cleaned, split, and spread. It leaves the kill room on a gambrel suspended from a pulley wheel riding the conveyor rail. The carcass proceeds by gravity into the chill room on the same floor, remaining there for 24 to 48 hours. The removal of body heat facilitates cutting. Prolonged storage is not desirable, however, since loss of body moisture during the chilling process results in a real economic loss. Shrinkage rates due to moisture loss are about 3 per cent during the first 24 hours, 1 per cent during the next 24 hours, and 1/2 per cent for the third 24-hour period.

On entering the cutting room at Hygrade, the carcass is removed from the rail system and placed on a table-height horizontal conveyor. In the cutting room the feet, hams, shoulders, butts, side meat and ribs are separated, trimmed, and dropped through chutes to lower floors. The loins are trimmed, weighed, and crated in the cutting room. Scraps removed during cutting and trimming pass through chutes to the lower floors for further processing.

Products that require smoking, such as hams and bacon, are then placed in the smoke house. Some of the products are canned and autoclaved. Other products, that are to be sold ready-to-eat, are heated sufficiently to produce a temperature of 137°F at the center of the meat. Finished products are assembled on the ground floor for shipment by truck or rail.

The plant operates on a 5-day week and a 7-3/4-hour day. Union regulations require that all employees in the processing department be given a 15-minute break every 2-1/2 hours. This necessitates a 15-minute shutdown of the plant once in the morning and again after lunch.

The statistical data acquired at the Hygrade plant are presented in Table XVII. These data are believed to be representative of large-scale pork processing plants. Correspondence with the American Meat Institute Foundation and a survey of the literature on modern pork processing plants has led to the conclusion that it is customary for pork plants to use the gravity flow process in which fresh carcasses are taken to the upper floors, cleaned and prepared, and thence moved to floors below for chilling and cutting, eventually arriving at the first floor for shipping. There are differences of design at various installations and many plants have only two floors. In the following discussion a plant of two floors has been selected as typical.

After careful study of various designs, it appeared that irradiation would be possible at any one of five points in the processing system, using a single radiation unit. These are:

1. Irradiation of the live hog prior to killing. This is the only point at which the entire animal, including the head, can be irradiated

without affecting the continuity of the plant process other than by installing a radiation room. Irradiation of the live animal may be undesirable for several reasons, however. The effect of radiation on glands is not known, and loss of potency in those used for pharmaceutical purposes might result. There may also be some objection from

TABLE XVII

PORK PROCESSING DATA SHEET

Plant Hygrade Food Products Corporation

Location Detroit, Michigan

I. Overhead Conveyor System

Type

Kill Room Chain-driven continuous, traveling 8-18 ft/min
Transporting Gravity, roller wheel
Minimum height of running surface above floor, 7 feet 10 inches

Operation of continuous conveyor: Frequency and duration of shutdown

Starting up, shutdown approx. 30 seconds, Variable frequency
Full operation, shutdown 15 minutes every 2-1/2 hours

II. Plant Data

Location of Kill Room, Fourth Floor
Location of Chill Room, Fourth Floor
Location of Cutting Room, Fourth Floor
Approximate design capacity, 3500 hogs/day
Kill days per year, 260-312
Hours per kill day, 7-3/4
Approximate operating capacity 850,000 hogs/yr

III. Treatment of pork to destroy trichinae as described by Department of Agriculture for processed pork products.

Approximate amount processed, 30%
Methods used

Heating yes
Freezing no
Curing yes

the humane viewpoint since the required dose would certainly cause the animal discomfort. In addition, this treatment of the live animal might alter the flavor of the meat as a result of changes in the flowing blood. Before designing a unit for irradiation of live hogs, preliminary investigation of the effects of total body irradiation on the living animal and its tissues would be required.

2. Irradiation of the carcass, less head and entrails, in the interval between leaving the kill room and entering the chill room. At this point, the objective of this work would not be completely accomplished, since portions of the hog muscle in the head and diaphragms would not be irradiated.

3. Irradiation of the carcass, less head and entrails, in the chill room during the chilling period. Here a much longer exposure time would be possible, lower-specific-activity source material could be used, and heat-transfer problems would be lessened by the lower ambient air temperature.

4. Irradiation of the carcass, less head and entrails, after leaving the chill room and before entering the cutting room.

5. Irradiation of the finished packaged products containing raw pork prior to shipment.

By utilizing a second radiation source in conjunction with method (2), (3), (4), or (5), the head and any other muscle-containing scraps could be irradiated after removal of the portions used for pharmaceutical purposes.

Of the five possibilities, (1), (2), and (3) could be carried out as continuous processes, (4) would be a batch process, and (5) might conceivably be either batch or continuous.

With methods (1) through (4), a source could be installed either on the upper floor of the plant, or on the ground floor with a conveyor to the upper floor, assuming the gravity system is used in the plant. The physical dimensions of the shielding material required for a large source might easily be greater than the available floor space. Ceiling clearance might also be a problem, since shielding would be required above and below the source room. In addition, the mass of the shielding is quite concentrated, and it is not likely that a packing house would be designed for such large, highly concentrated loads (upwards of 1 ton/ft^2). It is possible to distribute such a load but it is doubtful that such an installation could be made without extensive reinforcement of the building.

From the standpoint of cost of shielding material, the ideal solution would be an irradiation room below ground level, thus taking advantage of the natural shielding of the earth. However a significant objection to such an installation is the necessity of constructing a conveyor system to carry the hogs up to the processing floor of the plant.

On the basis of these considerations, it was decided to select a plant design in which the radiation would be performed on pork conveyed at the second-floor level. A design was selected for a plant operating 260 days/year with a capacity of 2,000 hogs/day, about 60 per cent of the capacity of the Detroit Hygrade plant. However it is still a plant of considerable size, and is probably more typical of the capacity of many hog processing plants operated by the larger packing companies. For example, the Hygrade Company operates a modern pork processing plant of this size at Storm Lake, Iowa, and the Wilson Company operates a packing house at Albert-Lea, Minnesota, which has about the same capacity for processing pork.

3. Design of Source

Gross fission products would appear to be the most economical source of radiation from the view point of material costs. However, reliable information on cost and availability is lacking on the gross fission products. Cesium 137 has been separated from the fission products at ORNL, and the Radioisotopes Division has offered (by private communication) to supply 1000-curie cesium-137 sources at $5.00 per curie. As this is the only quotation on high-activity fission product sources available to this laboratory the design was based on the use of cesium 137.

There are many shapes and types of containers which might prove suitable for holding concentrated radioactive fission products. Some preliminary calculations were made using cylindrical rods and rectangular strips, each of which has certain inherent advantages.

The chief advantages of using cylindrical containers are the simplicity of filling and the structural strength, while the chief advantage of the strips is the reduced number of source pieces required. A rectangular strip can be used to irradiate samples on either side and presents a continuous, uniform plane from which rays are emitted, which is advantageous in irradiating food conveyed by mechanical means through an irradiation chamber. In such an application it is desirable to introduce a minimum of direction changes, and preferably no change in conveyor mechanism, so as to minimize maintenance problems in the irradiation chamber. Preliminary calculations indicate that it is not feasible to use cylindrical source rods at 2-foot intervals, because rods so spaced must be unduly large if a high radiation flux is desired. Source

rods of large diameter are undesirable because self-absorption of the radiation causes generation of heat in the rods. The alternative is to place the rods at shorter intervals. This process can be continued until the limit is reached, at which the rods are adjacent to one another. Then the rods form a plaque for purposes of flux calculations, and a very large number of rods would be required. For these reasons the calculations have been limited to designs involving the use of a plaque made up of plane strips.

A Disk-Shaped Source. Calculating the radiation field emitted from a finite rectangular plaque is not a simple problem. The calculation is simpler for a disk-shaped plaque. In the accompanying calculations the radiation from a plaque 6 feet by 5 feet by 1 cm is compared with that from a disk 6 feet in diameter, and also with a disk 6.1 feet in diameter. The latter disk has the same area as the 6-foot by 5-foot plaque. A calculation was also made for an infinite strip 6 feet wide. These calculations are for radioactive fission wastes having an activity of 10,000 curies/lb. The calculations which may be of interest follow.

Required: A plane source 6 feet high and as long as necessary to provide a dose of approximately 30,000 rep to the surface of carcasses moving approximately 20 ft/min, 21 inches from the source, assuming 10,000 curies/lb for the source. To estimate the order of magnitude of the length required, compute the dose at a point P, 21 inches from a 6-foot disk, from Fig. 72:

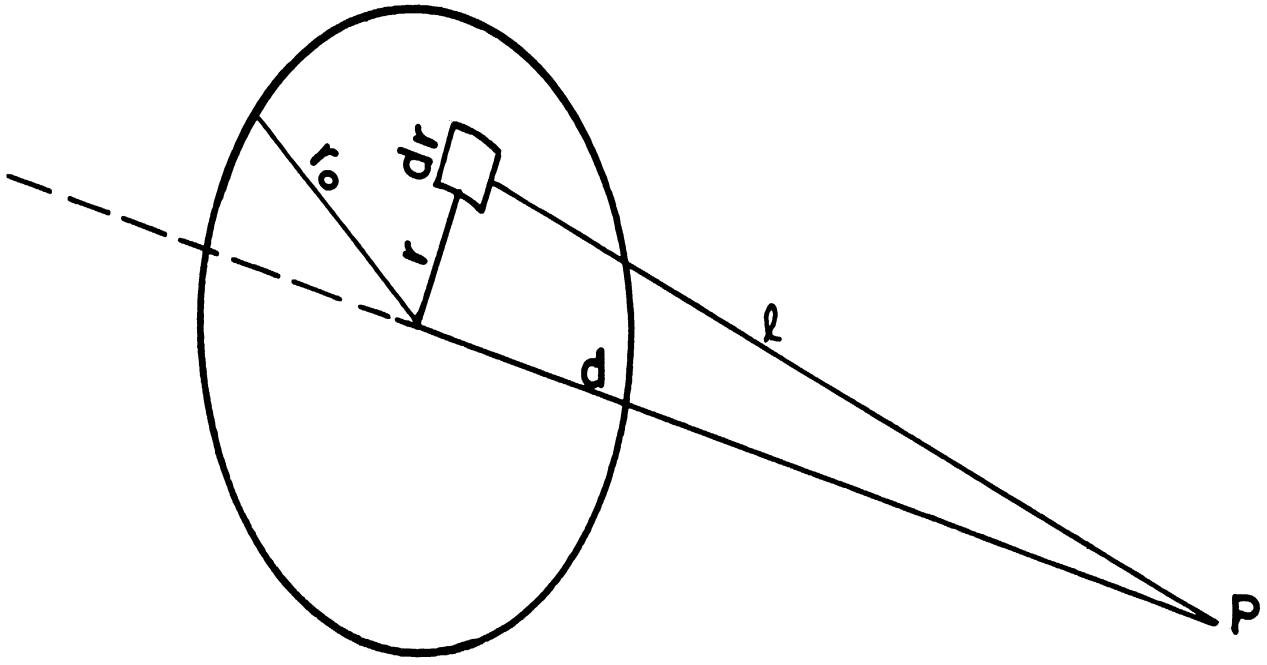

Fig. 72. Geometry of a Disk-Shaped Plaque.

$$I\ (r/hr) = \alpha \iint \frac{dA}{l^2} = 2\alpha \int_0^{r_o} \int_0^{\pi} \frac{r\ dr}{r^2 + d^2}$$

$$= 2\pi\alpha \int_0^{r_o} \frac{r\ dr}{r^2 + d^2}$$

$$= \alpha\pi \ln \left(1 + \frac{r_o^2}{d^2}\right) \quad ,$$

where α, the radiation coefficient, is

$$\alpha = R_o \rho s$$

and

R_o = 3.8 x 10^3 r/hr-curie at 1 cm for Cs^{137} *

ρ = 4 g/cm^3, typical salt density

ρ = 4 g/cm^2 for 1-cm-thick plaque

s = 10,000 curies/lb, specific activity;

thus

α = (3.8 x 10^3) (10,000) (4) (2.2 x 10^{-3} lb/g)

= 340,000 r/hr

Thus, at 21 inches from a 6-foot disk:

$I = (340{,}000) \ln \left[1 + (36/21)^2\right]$

= 1,470,000 r/hr

b. Infinite Strip Source. This result may be compared with the calculated dose rate for a plaque 6 feet high and infinitely long, from Fig.73a.

*Marinelli, L. D., et al,, American Journal Roentgenology, Radium Therapy, 59, 260 (1948).

$$I = \alpha \iint \frac{dA}{l^2} = 4\alpha \int_0^y \int_0^\infty \frac{dx\,dy}{x^2 + y^2 + d^2}$$

$$= 4\alpha \int_0^y \frac{dy}{\sqrt{y^2 + d^2}} \tan^{-1} \left[\frac{x}{\sqrt{y^2 + d^2}} \right]_0^\infty$$

$$= 2\pi\alpha \int_0^y \frac{dy}{\sqrt{y^2 + d^2}} = 2\pi\alpha \left[\ln \left(y + \sqrt{y^2 + d^2} \right) \right]_0^y$$

$$= 2\pi\alpha \ln \frac{y + \sqrt{y^2 + d^2}}{\ln d}$$

$$= 2\pi\ 340{,}000 \ln \frac{36 + 42}{21}$$

$$= 2{,}800{,}000 \text{ r/hr.}$$

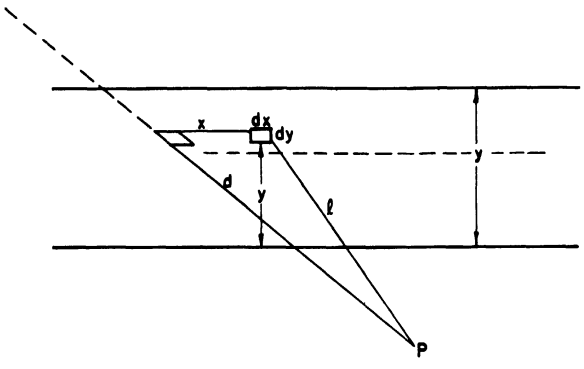

Fig. 73a. Geometry of an Infinite Strip Plaque.

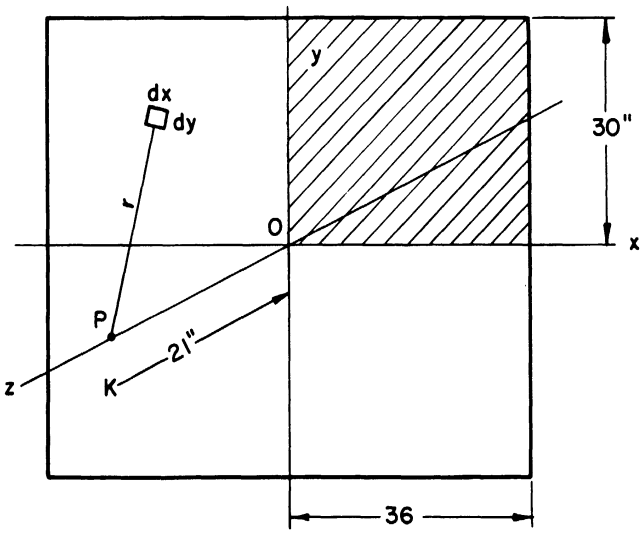

Fig. 73b. Geometry of a Finite Rectangular Source.

The dosage rate at 21 inches from a plaque of finite length and 6 feet high should lie between these two values. These are feasible radiation levels which would require approximately 1 minute of radiation time to provide the required dose of 30,000 rep.

Therefore, a plaque 6 feet high and 5 feet long is selected. As a first approximation, the dosage rate at 21 inches from a circular disk of equal area (3.08-foot radius) would be:

$$I = \pi\,(340{,}000)\,\ln\left[1 + (37/21)^2\right]$$

$$= 1{,}510{,}000 \text{ r/hr at 21 inches.}$$

C. Finite Rectangular Source. The objective was to determine the theoretical dose rate at a position P, 21 inches from the center of a plane rectangular 60- by 72-inch source (see Fig. 73b).

Procedure: Consider the contribution of an infinitesmal rectangular element of the source, dx by dy, whose position in space is given by the coordinates (x,y,0). The distance, r, of this element from the point P is given by

$$r = \sqrt{(x - 0)^2 + (y - 0)^2 + (0 - 21)^2}\ ,$$

$$= \sqrt{x^2 + y^2 + 441}\ .$$

By the inverse-square law, the contribution of this infinitesimal element is

$$\alpha \left(\frac{dy\ dx}{x^2 + y^2 + 441} \right)\ ,$$

where α is a coefficient determined by the characteristics of the source material.

Integrating this expression with respect to x from 0 to 36, and then with respect to y from 0 to 30, the contribution of the cross-hatched portion of the source shown in Fig. 73b is obtained; i.e.,

$$\alpha \int_0^{30} \int_0^{36} \frac{dy\ dx}{x^2 + y^2 + 441}\ .$$

Since the source is symmetric about the x and y axes, multiplication of the above expression by 4 yields the total dose rate I, at position P; i.e.,

$$I = 4\alpha \int_0^{30} \int_0^{36} \frac{dy\,dx}{x^2 + y^2 + 441} \quad , \qquad (34)$$
$$= 4\alpha H$$

where, for convenience, the double integral is denoted by H.

Integration with respect to x yields

$$H = \int_0^{30} \frac{1}{\sqrt{y^2 + 441}} \tan^{-1} \frac{36}{\sqrt{y^2 + 441}}\, dy \quad .$$

This integral cannot be formally integrated. Approximating H by means of Euler's quadrature formula[6], using 31 equally spaced ordinates, yields the value

$$H = 1.097 \pm 0.001.$$

Substituting this value and the proper value of α (see page 93) into equation (34) yields the dose rate:

$$I = 4\alpha H$$
$$= 4\,(340{,}000)\quad (1.097 \pm 0.001),$$
$$= 1{,}491{,}920 \pm 1360 \text{ r/hr.}$$

In a graphical check solution of the integral:

$$I = 4\alpha \int_0^{30} \frac{1}{\sqrt{y^2 + 441}} \tan^{-1} \frac{36}{\sqrt{y^2 + 441}}\, dy,$$

the values of the integrand (Y) as a function of y are plotted in Fig. 74a.

where

$$y = \frac{1}{\sqrt{y^2 + 441}} \tan^{-1} \frac{36}{\sqrt{y^2 + 441}}$$

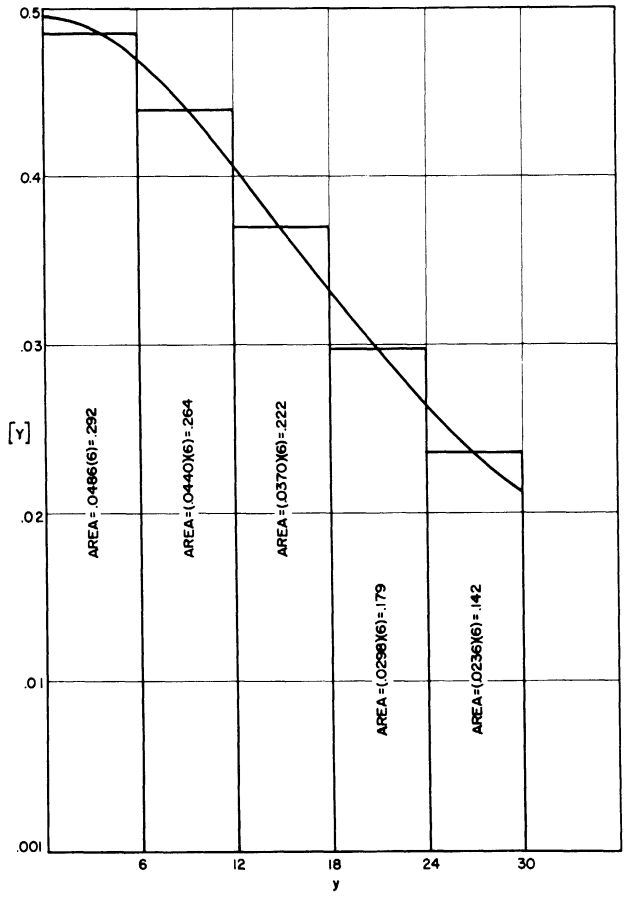

Fig. 74a. Graphical Integration of Equation 34a.

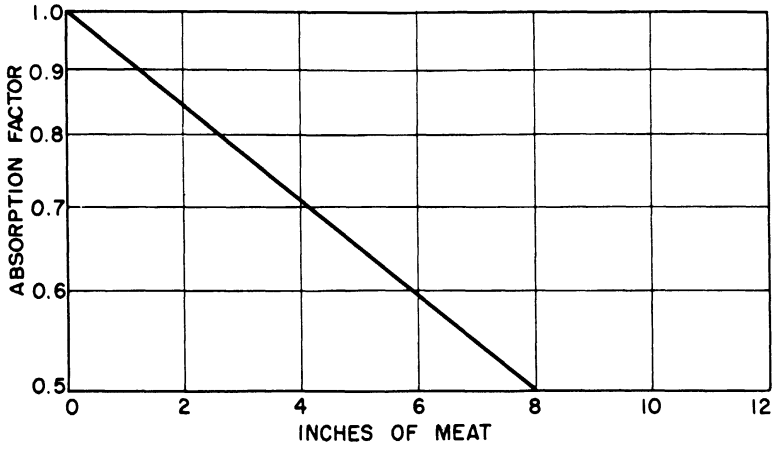

Fig. 74b. Absorption of Gamma Radiation in Pork.

GRAPHICAL INTEGRATION

y	Y	Δy	Area (YΔy)
0	0.0496	0-6	.292
5	.0477	6-12	.264
10	.0430	12-18	.222
15	.0368	18-24	.179
20	.0308	24-30	.142
25	.0256		1.099
30	.0212		

The area beneath the curve from 0 to 30 is thus found to be 1.099 area units.

Thus

$$I = 4\ (340{,}000)\ 1.099$$

$$= 1{,}495{,}000 \text{ rep/hr.}$$

A similar solution at the edge of the plaque and 1 foot past the edge of the plaque yields dosage rates of 1,010,000 rep/hr and 696,000 rep/hr respectively. The average dosage rate over the traverse of the plaque plus an additional foot on either end is approximately 1,140,000 rep/hr or 19,000 rep/min. Similar calculations for the case in which the hogs are spaced 17 inches and 25 inches from the plaque give radiation fluxes of 23,500 and 15,700 rep/min. respectively.

4. Capacity Calculations

a. Absorption Correction. The hogs to be irradiated will traverse 14 feet of radiation field having an average flux of 19,000 rep/min. This flux must be corrected for absorption in determining the dosage rate. Tests reported in the previous section describing irradiation of pork using cooling reactor fuel slugs indicated that the thickness of pork required to reduce the field of 0.7-mev gamma radiation to one-half value is approximately 8 inches as shown in Fig. 74b.

Reading from Fig. 74b for 4 inches of pork absorption, the radiation field will be reduced by 0.71. Therefore the center of the hog at 21 inches from the plaque will receive an average dose of

$$\text{Average Center Flux} = I = 19{,}000\ (0.71) = 13{,}500 \text{ rep/min.}$$

The surface of a hog 8 inches in thickness will be exposed to one traverse at 25 inches with 8 inches of absorption and one traverse at 17 inches with no absorption. For 17 inches,

$$I = 23{,}500 \text{ rep/min;}$$

for 25 inches,

$$I = 15{,}700\ (.5) = 7{,}850 \text{ rep/min.}$$

$$\text{Average surface flux} = \frac{23{,}500 + 7{,}850}{2} = 15{,}675 \text{ rep/min.}$$

For a spacing of 4 feet between hogs and a plant capacity of 2,000 hogs/day:

$$\text{Conveyor Speed} = \frac{2000 \text{ hogs/day } (4 \text{ ft/hog})}{20 \text{ hrs/day } (60 \text{ min/hr})} = 6.67 \text{ ft/min}$$

$$\text{Irradiation time} = \frac{14 \text{ ft}}{6.67 \text{ ft/min}} = 2.1 \text{ min}$$

$$\text{Center Dose} = 2.1 \text{ min } (13{,}500 \text{ rep/min}) = 28{,}400$$

$$\text{Surface Dose} = 2.1 \text{ min. } (15{,}600 \text{ rep/min}) = 32{,}800.$$

b. Thermal Considerations. For a plaque 0.4 inch thick of specific activity 10,000 curies/lb;

$$\sigma = \rho t s,$$

where

σ = curies/sq ft,

ρ = typical density = 4.0 gm/cc or 250 lbs/cu ft,

t = plaque thickness = 0.4 inch, and

s = specific activity.

$$\sigma = 250 \text{ lbs/cu ft } \left(\frac{0.4}{12} \text{ ft}\right)(10{,}000 \text{ curies/lb}) = 83{,}200 \text{ curies/sq ft.}$$

To compute the energy absorbed per square foot of plaque:

$$\text{particles/second(sq ft)} = 83{,}200\ (3.7 \times 10^{10})$$

$$= 3.08 \times 10^{15} \text{ particles/sec (sq ft).}$$

Ignoring the small energy absorption due to gamma photons, the average energy per disintegration will be derived from the absorption of beta particles and may be calculated as follows:

$$\frac{\text{Average energy}}{\text{disintegration}} = 0.4\ [0.05\ (1.2) - 0.95\ (0.51)]$$

$$= 0.218 \text{ mev/disintegration}$$

(Cesium-137 decays 5 per cent of the time emitting beta particles of 1.2 mev maximum energy, 95 per cent of the time with 0.51 mev maximum energy. In this region, average energy is approximately equal to 0.4 of the maximum energy.)*

$$\frac{\text{Energy absorbed}}{\text{sq ft}} = \left(0.218 \frac{\text{mev}}{\text{dis}}\right)\left(3.08 \times 10^{15}\ \frac{\text{dis}}{\text{ft}^2}\right)\left(1.6 \times 10^{6}\ \frac{\text{erg}}{\text{mev}}\right)$$

$$(10^{-7} \text{ joules/erg.})\ (3600 \text{ sec/hr})\ (1/1054.8 \text{ Btu/joules})$$

$$= 3.66 \times 10^{2} \text{ Btu/ft}^2 \quad .$$

If all this energy is transferred by convection to the air

$$Q = hA\Delta T;$$

assuming h = 4.0 Btu/(hr) (ft^2) (°F), and

ΔT = temperature difference between air and plaque surface,

Then

$$\Delta T = Q/hA$$

$$= (3.66 \times 10^{2})\ (1/4) = 92.5°F.$$

Or at a mean room temperature of 60°F the temperature of the surface of the plaque becomes

$$t_s = 60°F + 92.5°F = 152.5°F \quad .$$

5. Design of Radiation Chamber

Using a single plaque 6 feet long by 5 feet by 0.4 inch, 2,000 hogs can be irradiated each day by bringing the hogs on a conveyor past both sides

*Novey, T. B., et al., "Theoretical Study of β Absorption Curves" CC-579.

of the plaque. The flux at 21 inches is calculated to be approximately 13,500 rep/min. Therefore, the necessary time for the pork to accumulate 30,000 rep will be 2.22 minutes in the radiation chamber. Using a radioactive plaque 6 feet by 5 feet the pork would be conveyed at a speed of 6.67 ft/min. The amount of radioactivity required is calculated to be 1.5 megacuries. This amount of cesium activity would require 3 feet 4 inches of concrete shielding if placed above grade as shown by the following calculation:

a. Shielding Calculations for Concrete. At 8 feet from the plaque the dosage rate (no shielding) would be:

$$I = (340{,}000) \ln \left[1 + \left(\frac{37 \text{ in.}}{96 \text{ in.}} \right)^2 \right]$$

$$= 50{,}000 \text{ r/hr.}$$

The tenth-thickness of concrete for gamma radiation of 0.7 mev is 4.5 inches.[8]

Nine tenth-thicknesses (39.5 inches) reduce this to 0.05 mr/hr, which is considered safe.

To check this value: solve $I = I_0 e^{-\mu x}$, where $\mu = 0.2 \text{ cm}^{-1}$:[9]

$$0.05 = I = 50{,}000\, e^{-\mu x} = 50{,}000\, e^{-0.2(40)2.54}$$

$$0.05 = 50{,}000{,}000\, e^{-\mu x}$$

$$10^{-9} = e^{-\mu x} \quad .$$

Substituting for μ and taking the logarithms

$$9(2.303) = (0.2 \text{ cm}^{-1}) (x \text{ cm})$$

$$x = 103.6 \text{ cm, or } 41 \text{ inches.}$$

b. Shielding Calculation for Water. The tenth-thickness of water for this radiation is 11 inches.[8] In this case seven tenth-thicknesses (6 feet 5 inches) would reduce the dosage rate at 8 feet to 5 mr/hr, probably an acceptable level inside the cave. To be conservative, however, the design allows 6 feet 6 inches of water above the vertical source, whereas the calculated dose is on a normal to the plane.

The walls of such a one-story radiation chamber, with a total elevation of about 12 feet above grade, can be made uniform in thickness with no extra width for footings providing suitable bearing soil is available. The walls should continue far enough below grade to be below the frost line. Such a design for a one-story structure will result in a unit bearing pressure of about 2,000 lbs/sq ft, which is reasonable for most types of good bearing soils. The design also includes a safety well into which the radioactive plaque can be lowered to permit access to the radiation room.

A plan view of the radiation chamber one floor above grade is shown in Fig. 75. With the radiation chamber in this location the dimensions on the outside of the shielding are 28 feet 10 inches by 20 feet 6 inches. Figure 76 shows a cutaway isometric view of the design shown in Fig. 75. If the pork is to be taken from the conveyor on the second floor and returned, an inclined conveyor might be used such as shown in Fig. 77.

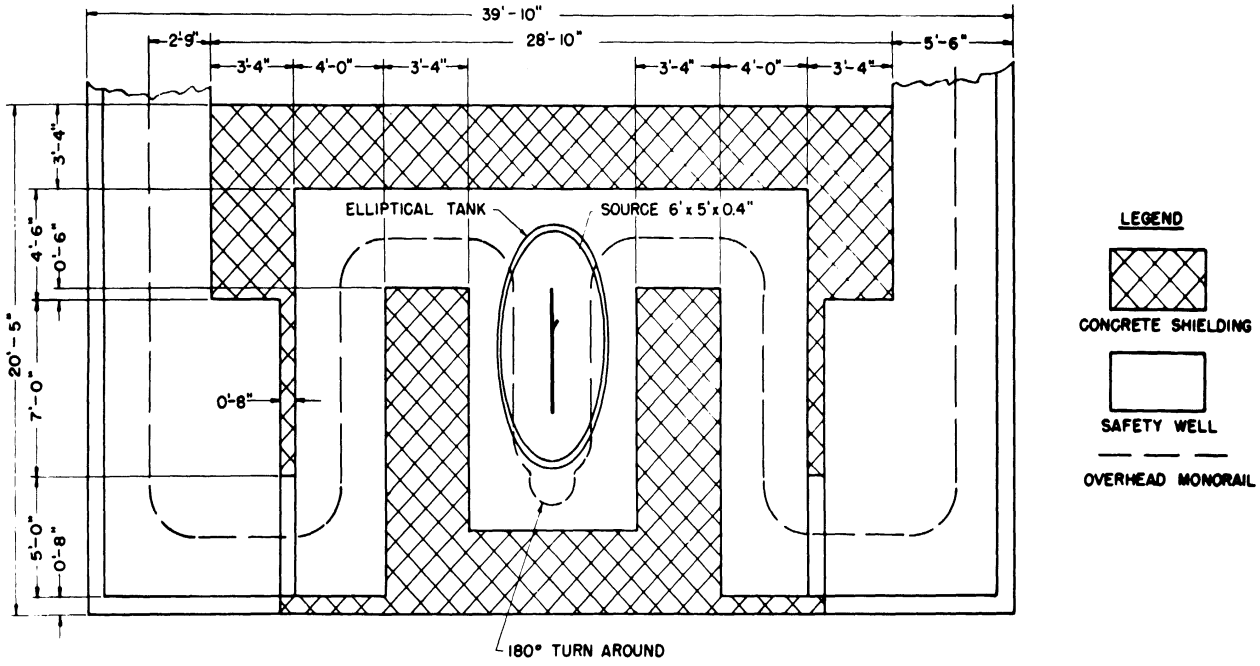

Fig. 75. Plane View of Radiation Chamber One Floor Above Grade.

Alternative designs are feasible in which the radiation chamber is placed one story below grade, one story above grade, or two stories above grade. There are some advantages and disadvantages for each location. Placing the radiation chamber below grade would reduce the amount of concrete shielding required. However, this advantage is partially offset by the additional cost of excavation and the requirement of steel reinforcing for retaining walls, water proofing or installation of water sump pumps, etc. Placing the radiation chamber on either the first floor above grade or below grade has the disadvantage of performing the irradiation below the floor at which the pork is being handled

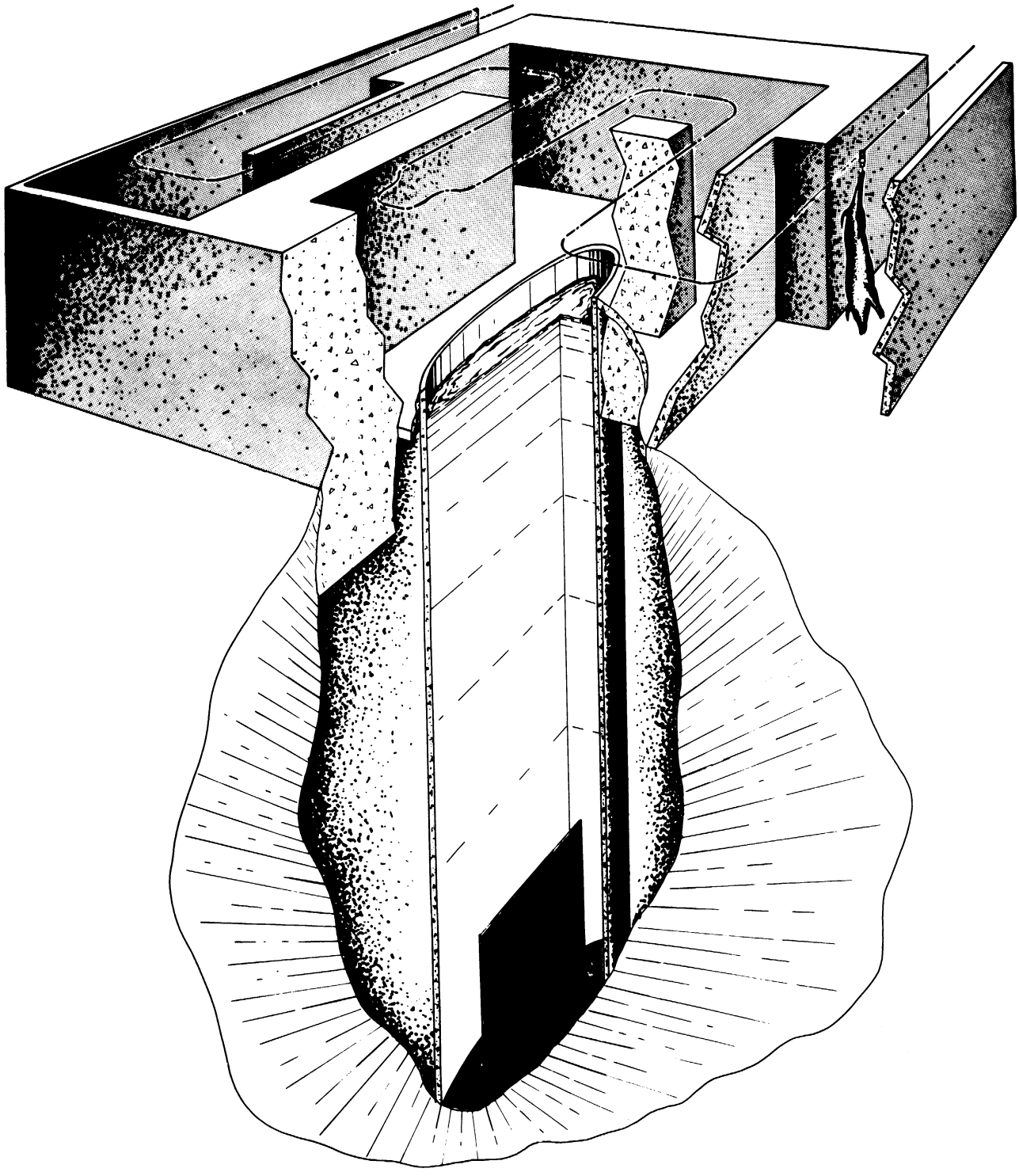

Fig. 76. Perspective View of Radiation Chamber One Floor Above Grade.

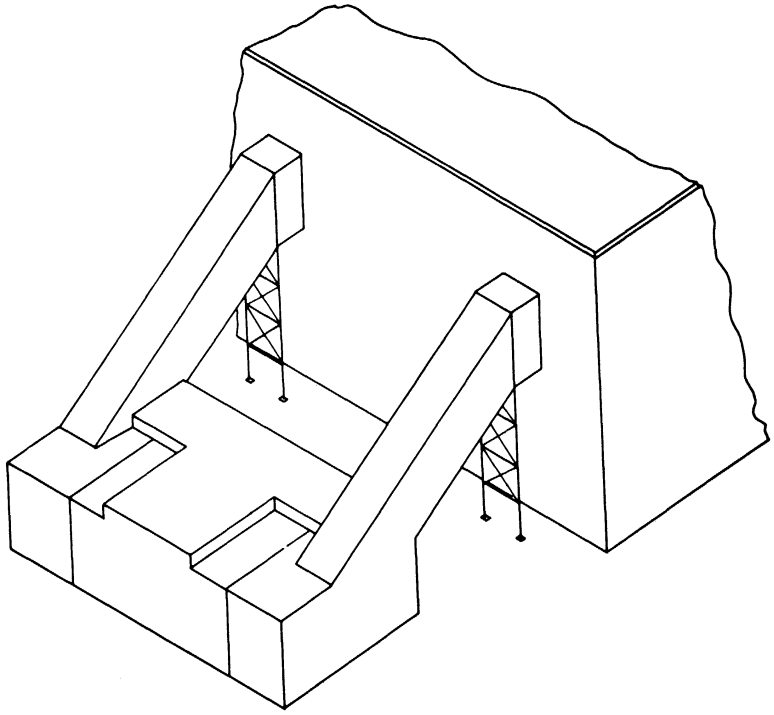

Fig. 77. Incline Conveyor to Bring Pork From 2nd Floor to First Floor for Irradiation and Then Return.

and therefore, requiring the use of a conveyor traveling up an incline or an elevator.

An alternate design is shown in Fig. 78 in which the radiation well and chamber are both placed above grade, with the radiation chamber at the level of the second floor so as to make it possible to irradiate the pork on the second floor. This would require footings having about twice the area of the footings of the previous design to provide adequate foundation, which would thus more than double the volume of concrete required. Such a radiation chamber might be placed adjacent to an existing building. It would then be necessary to bring the conveyor in and out of the new structure.

Another alternate design is shown in Fig. 79 in which the radiation chamber is located one floor below grade. A pit for a radiation chamber might be excavated in the basement of existing plants and a suitable elevator included to the conveyor on the second floor.

6. Estimation of Cost of Radiation Chamber

Although a radiation chamber is essentially a room with thick walls of concrete, the cost of the concrete is not a large part of the total cost. It is estimated that the cost of concrete for shielding will be in the range

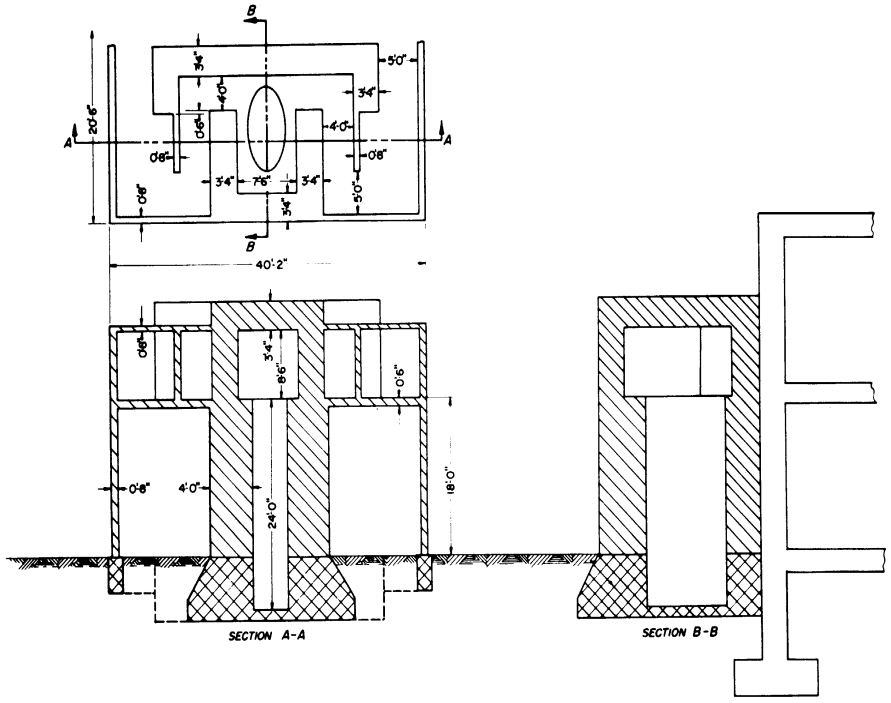

Fig. 78. Radiation Chamber Two Floors Above Grade.

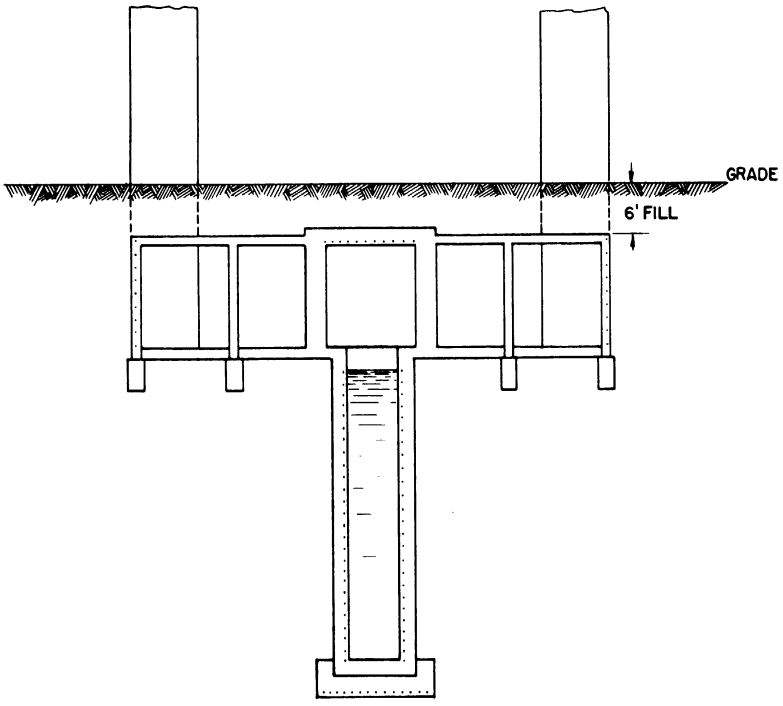

Fig. 79. Elevation of Radiation Chamber Located One Floor Below Grade.

of $30 to $50 per cubic yard for poured concrete, including the cost of forms. The cost will be slightly higher if the structure is formed out of solid concrete blocks, with mortar joints, because of the additional cost for masons' time.

It is estimated that a suitable radiation chamber of the type shown in Figs. 75 and 76 might be constructed for a total cost of $41,800, not including the cost of the radiation source itself or shipping and installation charges. Cost estimates are given in Table XVIII.

TABLE XVIII

COST ESTIMATES FOR RADIATION CHAMBERS FOR PORK

1. Estimate for radiation chamber one floor above grade

Excavation for footings and excavation and shoring for well (24 feet deep)	800
Stainless-steel well (elliptical-16 ga x 9 ft 0 in. x 4 ft 6 in. x 24 ft 0 in.)	2,500
Concrete for well (20 yds. at $20.00/yd)	400
Forms for concrete walls of radiation chamber (5,000 bd ft at $100.00/M)	500
Concrete for walls (150 yds at $20.00/yd)	3,000
Labor for forming and pouring walls	1,500
Forms and reinforcing for roof	150
Concrete for roof (50 yds at $20.00/yd)	1,000
Labor for forming and pouring roof	400
Concrete for floor (5 yds at $20.00/yd)	100
Reinforcing and labor for floor	100
Elevator mechanism	2,500
Ion exchange for well water	3,000
Monitoring equipment	4,000
Wiring	200
Water lines and labor for pipe fitting	800
Access doors (with safety interlock)	1,200
Ventilation system	2,000
Backgrading	200
Painting	250
Conveyor mechanism in radiation chamber	3,200
Conveyor mechanism to and from 2nd floor	4,200
Sub total for labor and materials	32,000
Miscellaneous contingencies (10% sub total)	3,200
Engineering costs (9% labor and materials)	2,800
Contractors fee (10% of costs)	3,800
Total	41,800

2. Estimate for radiation chamber one floor below grade

Note: The estimate is the same as for (1) with the following exceptions:

TABLE XVIII (cont.)

a. Additional costs	
Additional excavation, shoring, and trucking costs (1,000 yds at $1.00/yd)	$ 1,000
Waterproofing walls	300
Sump well and pump	300
Stair well to lower floor	1,000
Additional conveyor to and from 2nd floor	2,400
Steel reinforcing for walls	400
	$ 5,400
b. Costs saved	
Concrete saved in wall (110 yds at $20.00/yd)	$ 2,200
Concrete saved in roof (35 yds at $20.00/yd)	700
	$ 2,900
Total cost of chamber one floor below grade $41,800 + $5,400 - $2,900 =	$44,300

3. Estimate for radiation chamber two floors above grade

Note: The estimate is the same as for (1) with the following exceptions:

a. Additional costs	
Additional excavation for footings	$ 200
Additional concrete for footings (30 yds at $20.00/yd)	600
Additional concrete for walls (120 yds at $20.00/yd)	2,400
Additional forms (4,000 bd ft at $100.00/M)	400
Additional labor for forming and for pouring from second floor	1,600
Use of crane to raise concrete to second floor	600
	$ 5,800
b. Costs saved	
Conveyor mechanism	$ 1,000
Excavation and shoring for well	400
	$ 1,400
Total cost of chamber on second floor $41,800 + $5,800 - $1,400 =	$46,200

To date there is not sufficient information to make a reliable estimate of the cost of packaged fission products. However, an estimate may be made of the magnitude of the cost of the fission products, using the 0.6-power scaling factor frequently used in estimating the costs of chemical plants.[7] Kilocurie gamma sources of cesium-137 processed from fission products can be supplied by the Radioisotopes Division of AEC, Oak Ridge, Tennessee, for $5,000. The experience of the Fission Products Laboratory with a 1,000-curie cobalt-60 source indicated shipping (700 miles) and installation costs (not including cost of shipping container) total about $1,000. Therefore it is assumed that $6,000 is a reasonable installed cost for 1,000 curies of cesium 137. Using the 0.6-power factor for scaling up to a 1.5-megacurie source, the installed cost is estimated to be:

$$x = \$6{,}000 \left(\frac{1{,}500{,}000}{1{,}000}\right)^{0.6}$$

$$= \$483{,}000$$

A shipping container for such sources might be supplied by AEC or an industrial handler of fission product sources. However, if it was not, a steel-reinforced lead container approximately 2 feet by 7 feet by 8 feet would be required at a cost of about $25,000. This container would weigh about 50 tons and therefore could be shipped on a flat car. A much cheaper but more unwieldy container could be constructed of concrete.

Assuming the maximum cost conditions, for a radiation chamber on the second floor, and including the cost of a lead shipping container, the initial investment is estimated to be:

Cost of radiation chamber (from Table XVIII, part 3)	$ 46,200
Shipping container	25,000
1.5-megacurie source, installed	483,000
	$554,200

If the plant is written off in a five-year period and 6% of the investment is included for interest, the annual costs become:

$$\frac{1.18}{5} (554{,}000) = \$130{,}500/\text{yr.}$$

An estimate of $30,000 per year is made for operation with two experienced health physicists and for maintenance, making the total annual cost:

$$\$130{,}500 + \$30{,}000 = \$160{,}500$$

Based on the production figures for 1950, 68,504,000 head of hogs yielded 9.265 million pounds of pork; thus the average hog yields 135 pounds of pork, excluding lard. Basing the design on a rate of 2,000 hogs/day and operation 260 days/year, the production for the plant would be:

(2,000 hogs/day) (260 days/year) (135 lbs pork/hog) = 70,200,000 lbs pork/year.

The cost of operating the irradiation chamber to be added to the price of pork would thus be:

$$\frac{\$160,500/\text{year}}{70,200,000\ \text{lbs/year}} = \$0.0023/\text{lb} = 2.3\ \text{mills/lb}.$$

$$(\$0.0023/\text{lb})\ (135\ \text{lbs/hog}) = \$0.31/\text{hog}.$$

Cesium 137 has a half-life of 33 years. Therefore, the plant might readily be written off over a period of ten years rather than five years. This would reduce the computed costs to about one-half. However, a charge of 2.3 mills/lb or $0.31/hog would be small price to pay to aid in wiping out the problem of trichinosis.

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D. ANIMAL FEEDING EXPERIMENTS

Personnel:

Supervisors and Advisors: H. B. Lewis,* Professor of Biological Chemistry and Chariman of Department of Biologial Chemistry; H. C. Eckstein, Professor of Biological Chemistry; L. E. Brownell, Director of Fission Products Laboratory and Associate Professor of Chemical Engineering; L. L. Kempe, Assistant Professor of Bacteriology and Assistant Professor of Chemical Engineering. Laboratory Personnel: B. W. Uhlendorf, Project leader and Research Assistant; J. T. Graikoski, Research Assistant; R. Dennis, Statistican; E. Ambo, Laboratory Assistant; R. De Pestel, Laboratory Assistant.

1. Introduction

At a meeting held recently at the Oak Ridge Institute of Nuclear Studies, Dr. W. B. Rankin, Assistant Director of the Food and Drug Administration, Division of Field Operations, discussed the major issues involved in the use of radiation for food sterilization under the federal food and drug laws.

It was stressed that before irradiation-sterilization of foods and drugs can be used commercially, much information is needed regarding the effects of radiation on the nutritional properties of food, the therapeutic efficiency of drugs, and the possibility of irradiation by-products being toxic. It was pointed out that the FDA does not wish to bar progress in the use of radioactive materials but that the absolute safety of the process must be demonstrated.

Drs. H. B. Lewis, H. C. Eckstein, L. E. Brownell, and L. L. Kempe, and Mr. B. W. Uhlendorf have planned an animal-feeding experiment to evaluate the wholesomeness of irradiated food. This experiment will be referred to as Experiment No. 1 and is presently underway with support from Phoenix project 41.

*On leave of absence because of illness.

In this experiment a synthetic diet containing 50-per cent heat-sterilized meat is being used to minimize the problems of storage and to provide more adequate controls than would be possible using raw meat. No attempt is being made to study the effect of storage on irradiated food.

A second experiment to study the effects of storage (Feeding Experiment No. 2) should be performed in which irradiated foods are stored for various lengths of time prior to feeding. It has been shown that peroxides and other compounds are formed as a result of irradiation of fats and their effects may not appear until after an appreciable storage period.

A third experiment (Feeding Experiment No. 3) which should be conducted, is the sterilization of raw perishable food for subsequent animal feeding. This experiment is necessary because there is an indication that food containing active enzymes behaves differently when irradiated than foods in which the enzymes have been destroyed and the proteins denatured prior to irradiation.

In the present animal-feeding experiment rats are used as the experimental animals, as they would be in the proposed experiments also. The Food and Drug Administration has indicated that it is necessary to extend such tests to species other than rodents, such as dogs or monkeys.[2] Feeding Experiment No. 4 using nonrodents should therefore be performed. The Food and Drug Administration and representatives of the National Cancer Institute have also indicated that in order to determine whether irradiation could induce the formation of carcinogens from naturally occurring steroids, it would be necessary to run tests using strains of cancer-sensitive mice (Feeding Experiment No. 5).

2. Revision of Feeding Experiment No. 1

The original plan for Feeding Experiment No. 1, described in Progress Report 4, was based on the knowledge that irradiation does cause some loss in vitamin content and the formation of peroxides from fats. Therefore, it was planned to irradiate all the proteins and carbohydrates but not the supplements of vitamins and fats. Later it was realized that the irradiation capacity of the 10,000-curie source is more than ample to permit an increase in the scope of the experiment. Other minor modifications have also been made in the diet and procedure.

A small-scale pilot experiment is now in progress. This experiment was designed primarily to determine whether high-energy radiation induces the formation of toxic substances from normal food constituents. However, if some essential component of food is largely destroyed, the effect produced would be

a deficiency effect. It is known, for example, that organic peroxides derived from fat destroy vitamin E. Also, it was decided to add to the original experiment another group of animals in which the entire diet received is irradiated. Consequently, in order to distinguish between toxicity effects and deficiency effects, if any, there will be a control diet and two experimental diets.

The control diet (A) will consist of nonirradiated food. For experimental diet (B) all but the vitamins, fats, and salts will be irradiated, while for the other experimental diet (C) all components will be irradiated. The radiation dose is two million rep.

3. Animal Room

The air-conditioned animal room shown in Fig. 80 is kept at a temperature of 76° to 78°F. The relative humidity will be maintained at 40 to 50 per cent by injecting live steam into the room in the winter. The rapid exchange of air and condensation on the cooling coils of the refrigeration unit of the conditioning system will keep the humidity from exceeding 50 per cent in the summer. This temperature and humidity will prevent the condition known as ring-tail in the newborn rats (this is a condition in which newborn rats develop reddened, swollen tails with corrugated constrictions).[3] A sensitive recording thermometer and hygrometer will provide a continuous record of the temperature and relative humidity. In addition to the air-conditioning unit, which recirculates the air in the room, an exhaust fan removes stale air from the room and pulls fresh air into it providing a complete exchange of air approximately every 15 minutes. Light and dark periods are alternated every 12 hours by an automatic switch. According to Farris,[4] rats with regulated light and dark display more regular estrous cycles. The floor of the room is swept and mopped with Lysol solution daily.

In Feeding Experiment No. 1 the rats will be housed individually in cages measuring 7 inches by 9.5 inches by 7 inches, with No. 2 gauge wire cloth on the front and bottom. During mating and before the litters are weaned, a similar cage measuring 16.25 inches by 9.5 inches by 7 inches will be used.* The cages are sterilized weekly using boiling water in the sterilizer shown in Fig. 80.

4. Diet

The diet must be a complete ration and yet such that quantitative experiments, such as vitamin assays, can be performed. From the point of view

*The cages and racks were obtained from Hoeltge Bros., Cincinnati, Ohio.

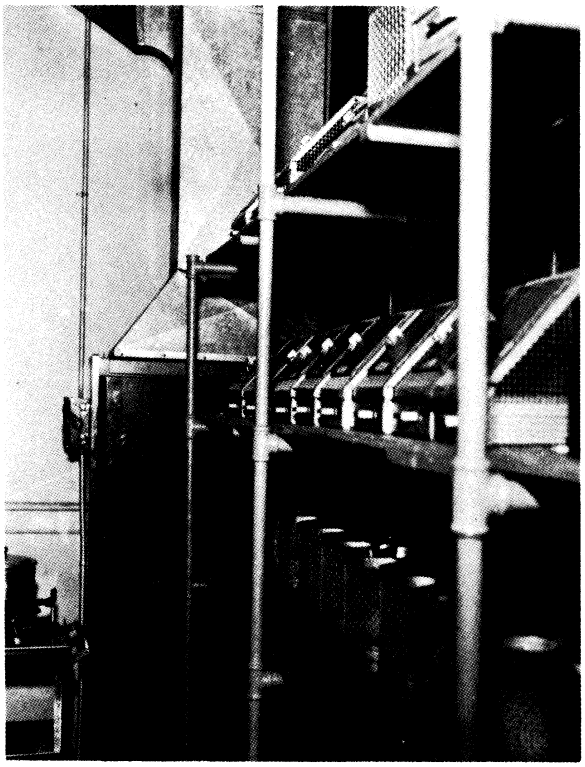

Fig. 80. One Side of the Rat Room Showing the Air Conditioning Unit in the Background and the Original Cages on the Right.

of applying these results to human consumption, a diet of natural foods would be best. The food mixture selected is a semisynthetic diet in which a large proportion of the protein is in the form of a canned meat product, Swift's Beef for Babies. Calculated on a dry basis, this diet contains approximately 29% protein, 45% carbohydrate, 17% fat, 3.5% salts, and 3% nonnutritive bulk. The composition of the diet is given in Table XIX, and an analysis of Swift's Beef for Babies in Table XX.

TABLE XIX

COMPOSITION OF RAT DIET

Swift's Beef for Babies	50.00%
Casein	7.50%
Corn starch	27.17%
Cellulose	1.85%
Salad oil*	6.00%
Cod-liver oil, U.S.P.	3.00%
Salts	1.84%
Vitamin mixture	2.46%
Choline chloride	0.18%

* Durkee's soybean oil.

TABLE XX

TYPICAL ANALYSIS OF SWIFT'S BEEF FOR BABIES

Protein	18.7%	Calcium	8 mg/100 g
Fat	3.0%	Phosphorus	1.30 mg/100 g
Moisture	77.1%	Iron	2 mg/100 g
Total Ash	1.2%	Thiamin	0.01 mg/100 g
Salt	0.6%	Riboflavine	0.21 mg/100 g
		Niacin	3.39 mg/100 g

According to Tables XIX and XX, the canned meat contains 18.7% protein and 77.1% moisture. If beef were used as the sole source of protein, it would provide the diet with about 15% protein, calculated on a dry basis. McCoy[5] states that 25 to 30% protein yields optimum growth and excellent reproduction. For this reason, casein* is used to supplement the meat protein. The cellulose** provides the diet with nonnutritive bulk. The cod-liver oil, U.S.P., which contains at least 850 units of vitamin A, and 85 units of vitamin D per gram, is also a vehicle for added α-tocopherol. This mixture is stored in a dark glass bottle to prevent photodestruction of vitamin A.

TABLE XXI

SALT MIXTURE, H.M.W.

	%
Calcium carbonate	54.300
Magnesium carbonate	2.500
Magnesium sulfate	1.600
Sodium chloride	6.900
Potassium chloride	11.200
Potassium phosphate (monobasic)	21.200
Ferric phosphate	2.050
Potassium iodide	0.008
Manganese sulfate	0.035
Sodium fluoride	0.100
Aluminum potassium sulfate	0.017
Copper sulfate	0.090

This salt mixture is used as 2-1/2% of the diet on a dry basis.

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* Vitamin-free casein obtained from the Borden Company.

** Alphacel, obtained from Nutritional Biochemicals Corporation, Cleveland, Ohio.

The salt mixture* was formulated by Hubbell, Mendel, and Wakeman.[6] According to the recommendations of McCoy[5] this salt mixture is insufficient in manganese and potassium; therefore, it is supplemented with manganese sulfate, 92 mg/kgm of diet, and dibasic potassium phosphate, 2.77 gms/kgm of diet. The composition of the salt mixture is given in Table XXI. The total salt content of the diet is calculated on the basis of the elements the salts provide to the diet on a dry basis. The recommended minimum quantities are given in Table XXII.

The composition of the vitamin mixture is given in Table XXIII. Pure crystalline B vitamins are used with the exception of vitamin B_{12}, which is supplied in the form of liver powder. The liver powder** and Brewer's yeast*** also supply possible growth and lactation factors not yet identified. The choline chloride is not included in the vitamin mixture because it is extremely deliquescent. This vitamin is dried, weighed, and added as a separate item

TABLE XXII

TOTAL SALT CONTENT OF DIET

	mg/100 g diet	mg/rat/day	Recommended[5]
Calcium	550	110	500/100 g
Phosphorus	391	78	500/100 g
Magnesium	26	5	5/100 g
Sodium	262	50	500/100 g
Potassium	499	100	503/100 g
Chlorine	636	130	5 mg/day
Iron	14.6	2.9	5 mg/day
Iodine	0.15	0.03	1-2 mg/day
Manganese	4.82	0.96	0.8 mg/day
Fluorine	1.13	0.23	----------
Aluminum	0.025	0.005	1 mg/day
Copper	0.895	0.18	.05 mg/day

* Obtained from Nutritional Biochemicals Corporation, Cleveland, Ohio.

** Liver concentrate, N.F., obtained from the Wilson Laboratories, Chicago, Ill.

*** Brewer's yeast, Anheuser and Busch, obtained from Fisher Scientific Company.

when the diet is mixed. Table XXIV shows the total vitamin content of the complete diet including the vitamins from the meat, liver powder, yeast powder, cod-liver oil, and those added in pure form, the amounts recommended by various authors for optimum growth, reproduction, etc. are also included. It will be noted that in many cases the vitamin content of this diet is considerably greater than the recommended minimum. However, as stated in the introduction, this experiment was designed primarily to determine whether any toxic products were formed in the food and not to test the destruction of vitamins. The composition of the diet with respect to vitamins was set up before it was decided to feed a group of animals a diet in which the vitamins are also irradiated. Since it is possible that toxic substances could be produced by irradiation of vitamins, it is important that all vitamins required for human consumption be included in the diet, whether they are essential for the rat or not.

TABLE XXIII

COMPOSITION OF VITAMIN MIXTURE

	Gm	Mgm
Ascorbic acid	0.25	-
Thiamine hydrochloride	0.05	-
Pyridoxine hydrochloride	0.1	-
Riboflavine	0.1	-
Calcium pantothenate	0.4	-
Niacin	0.5	-
p-Aminobenzoic acid	5.0	-
Inositol	10.0	-
Biotin	-	2.0
Folic acid	-	7.0
Menadione	-	25.0
Brewer's yeast, U.S.F.	150.0	-
Liver concentrate, N.F.	150.0	-
Corn starch	83.6	-
	400.034	

This mixture is used as 4% of the diet on a dry basis.

TABLE XXIV

VITAMIN CONTENT OF COMPLETE DIET ON DRY BASIS

	Mg/100 g diet	Amount/rat/day	Recommended Amount/rat/day	Reference
Thiamine	0.756	150 mcg	75 mcg	7
Pyridoxine HCl	1.290	260 mcg	50 mcg	8
Riboflavine	1.487	300 mcg	60 mcg	7
Calcium pantothenate	4.975	1 mg	600 mcg	9
Niacin	10.850	2.1 mg	1 mg*	10
p-Aminobenzoic acid	50.02	10 mg	****	-
Inositol	106.00	21 mg	****	-
Biotin	0.0238	4.8 mcg	3 mcg	9
Folic acid	0.088	.8 mcg	****	-
Menadione	0.25	50 mcg	**	-
Vitamin B_{12}	0.008	1.6 mcg	1 mcg	11
Choline chloride	328.5	66 mg	30-35 mg	5
α-tocopherol	32.5	6.4 mg	3 mg	5
Ascorbic aicd	2.5	500 mcg	***	12
Vitamin A	2550 units	510 units	50 units	5
Vitamin D	255 units	51 units	60 units	5

5. Mixing of the Diet

The crystalline vitamins, liver powder, yeast powder, and some corn starch are mixed in a ball mill. This mixture is made up in batches of 2 gm,

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* There is no evidence that niacin is required for growth; however, 22 investigators added it to the rat diet at a median value of 5 mg/100 g.

** An alteration of the intestinal flora must take place before a K-avitaminosis can be elicited. Nearly all rats grow and reproduce normally in its absence.[5]

*** Ascorbic acid (Vitamin C) is included even though it is not required preformed in the diet of the rat, in order to check the possible formation of toxic compounds from the irradiation of ascorbic acid. This amount of vitamin C is based on the human requirement of 75 mg/70 kg/man/day.[12]

**** The amounts of these 3 vitamins represent averages of the amounts used by 112 other workers in experimental rat diets.

which is enough for about 80 gm of diet. The vitamin mixture and the salt mixture are then mixed with the other dry ingredients, with the exception of the choline chloride, in a MacLellan Dry-Batch Mixer. The meat, choline chloride, dry mix, and oils are then thoroughly mixed in a Hobart Mixer (shown in Fig. 81).

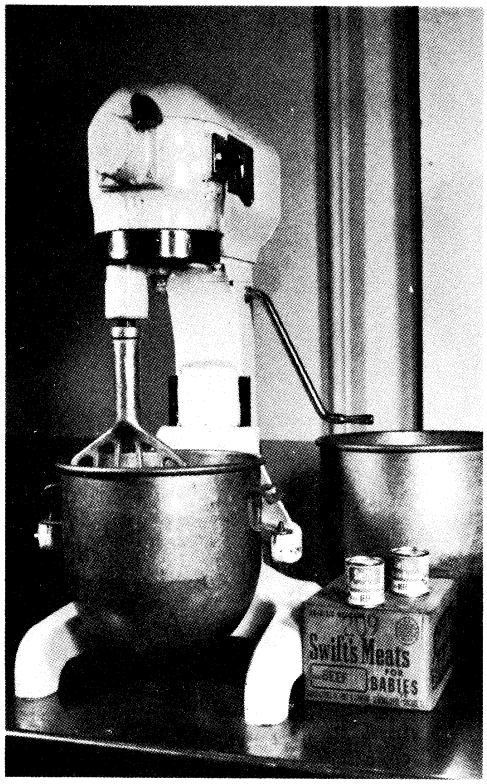

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Fig. 81. The Hobart Mixer Used to Mix the Diet.

For the B diet, the meat is irradiated in the manufacturer's cans and the dry mix irradiated separately. These are then combined with the vitamins, salts, and oils in the Hobart Mixer. In preparing the A and C diets the ingredients are mixed together; half is then used as the control diet (A) and the other is irradiated for diet (C). The two diets will be made up daily or possibly every other day. A one-week supply will be kept of each diet to allow for shutdown and changes in the irradiation facilities, and will be stored at 4°C.

6. Plan of Experiments

The plan of the experiments is essentially that described by Lehman, et al.,[2] for the appraisal of toxicity of chemicals in food.

a. Long-Term Study for Sub-Acute or Chronic Toxicity. Weanling litter mates (Holtzman) rats will be distributed evenly with respect to numbers,

sex, and weight into the three groups to supply 16 males and 16 females in each group. One group will receive the control diet (A), another group the diet in which the vitamins and oils are not irradiated (B), and the third group the diet in which all components are irradiated (C). The rats will receive food ad libitum, but a record of the daily food consumption will be kept. Fresh water will be supplied daily. The growth of each animal will be recorded by weighing them once a week. Observations will be made on general appearance, behavior, and mortality of the animals, and hemoglobin determinations, red, white, and differential blood counts, will be made monthly.

Animals showing any gross abnormalities, such as tumor, severe loss of weight, etc., will be posted and tissues preserved for histopathological examination. The long-term experiment will be terminated when the rats are 2 years old. At that time the surviving animals will be sacrificed and autopsied, the principal organs weighed, and the tissues preserved for histopathological study. Any animals that die before the termination of the experiment will be treated likewise. Biochemical studies will be conducted if necessary; for example, if one group of animals is utilizing protein more efficiently, it would be advisable to compute nitrogen balance.

b. Reproduction Studies. The rats used for the long-term studies will also be used as the parent generation for the reproduction studies. When these animals are 100 days old, 16 females and 8 males will be selected and mated (1 male to 2 females). The litters (F_{1a}) will be destroyed when they are weaned. In mating, the males will be rotated among the females weekly within each group. Before the 21st day after the original mating, the males will be removed and the females put in separate cages. Since the estrous cycle of the albino rat is four or five days, this will give four to five opportunities for conception. Then 10 days after the last litter is weaned, the rats will be remated in a like manner. From these litters (F_{1b}) 16 females and 8 males will be selected to continue the reproduction study. These will be mated when at least 100 days old using the procedure described above. The (F_{2a}) litters will be destroyed and the (F_{2b}) litters continued in the study. The reproduction studies will be terminated when the (F_{3b}) litters are weaned.

The selection of 16 females and 8 males from the (1b) and (2b) litters in such a way as to give a fair sample of the generation is complicated by two factors; first, there may be a spread of a month or so over which the litters are born, and second, some females may not produce a litter. The method selected is a compromise, since complete random selection is not practical. When each of the 16 (or less) litters are 21 days old, the pups will be tagged and 2 females and 1 male will be selected from each litter by the use of a table of random numbers. This will yield an excess over the number of rats needed to continue the experiment.

The number of animals will be reduced to 16 females and 8 males by taking 1 of the 2 females from each litter and by randomly selecting 8 males from the 16. In the event that some parent females do not have litters, or do not raise the litter to weaning, or the litter is all of one sex, animals will be selected from the excess available.

The litters exceeding 10 offspring will be reduced to this number of animals on the day the litter is born. A record will be kept of the number of litters, the size of the litter at 1, 5, and 21 days, and the weight of the litter at 21 days. The parent generation will be sacrificed and autopsied when their second litters are weaned (except for the original rats which will be continued in the long-term study), and the tissues will be preserved for histopathological studies. The procedure is shown diagrammatically in Fig. 82.

7. The Pilot Experiment

The pilot experiment preliminary to this long-term study has three main functions: (1) to determine whether the experimental diet is acutely toxic; (2) to give the personnel experience in the tasks and experimental techniques associated with this type of experiment and to iron out any unforeseen difficulties; and (3) to yield preliminary data which can be used as a guide for planning the main experiment, such as how often to weigh the animals, what food consumption, can be expected, etc.

The pilot experiment in progress is successfully serving these functions. The data show that the irradiated food definitely is not acutely toxic under the conditions of the experiment.

In the pilot experiment a total of 27 Holtzman rats were divided into three groups and fed diets A, B, and C. Unfortunately, due to various unanticipated delays, these animals were not fed the experimental diets during the period of most rapid growth. The growth data for a period of over two months are summarized in Table XXV. The females are not included here because of the weight variation during the pregnancy and nursing periods. The growth curves for the animals during this period are shown in Figs. 83, 84 and 85 where the average weight of each group is plotted as a function of time. It should be noted that the feeding of diets A, B, and C commenced on May 22, and that the animals were first mated on June 1. The average growth curves for the males shows no significant variation between the groups fed diets A, B, and C.

a. Discussion of Growth Data. It is obvious from these data that for the period these animals were fed, the partly irradiated and the completely irradiated diets did not have a deleterious effect as far as can be ascertained from the rate of growth. Statistical analysis shows that at the 1 per cent or

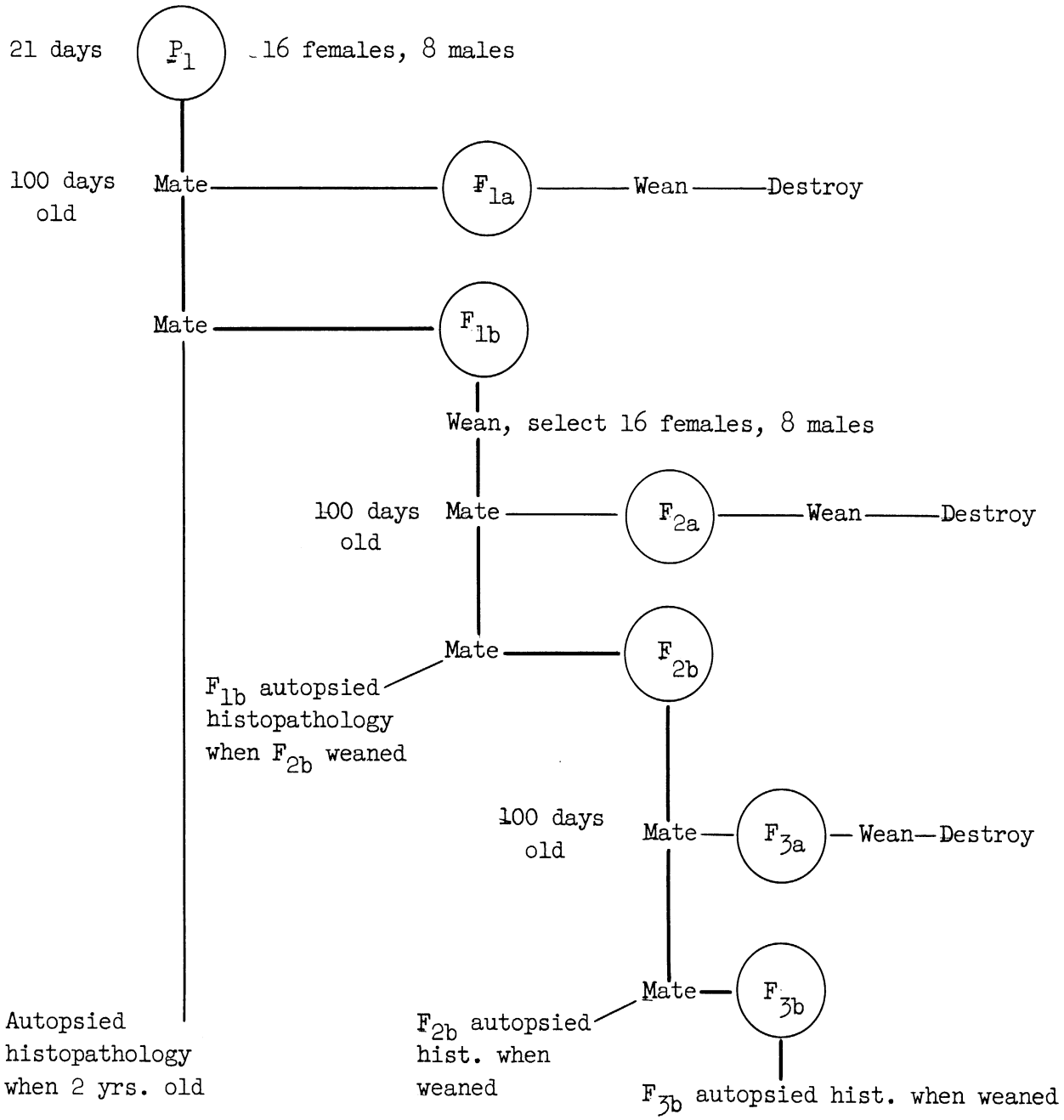

Fig. 82. Schematic Diagram for a Given Diet for Reproduction Studies Using Irradiated Food.

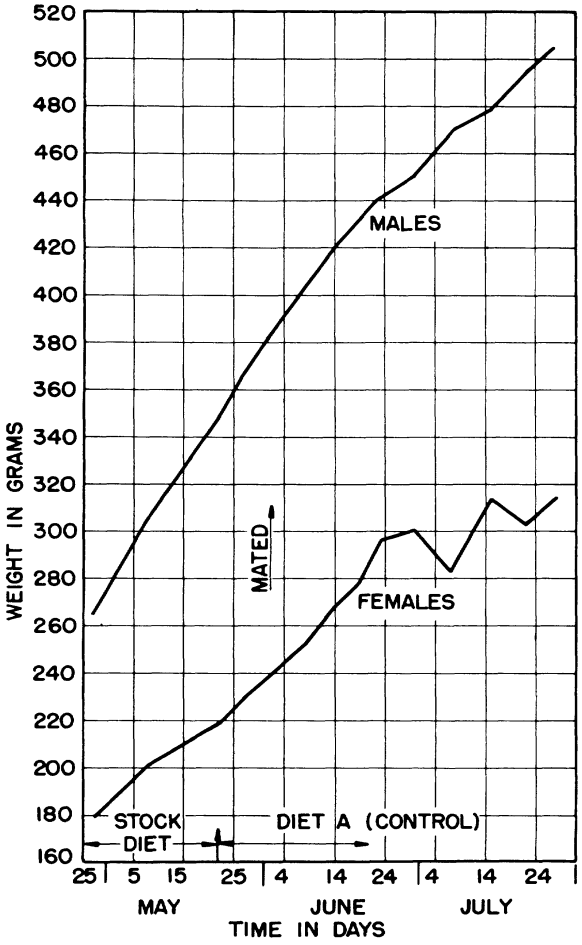

Fig. 83. Growth Curve for Rats Fed Diet A (control) in Pilot Experiment No. 1.

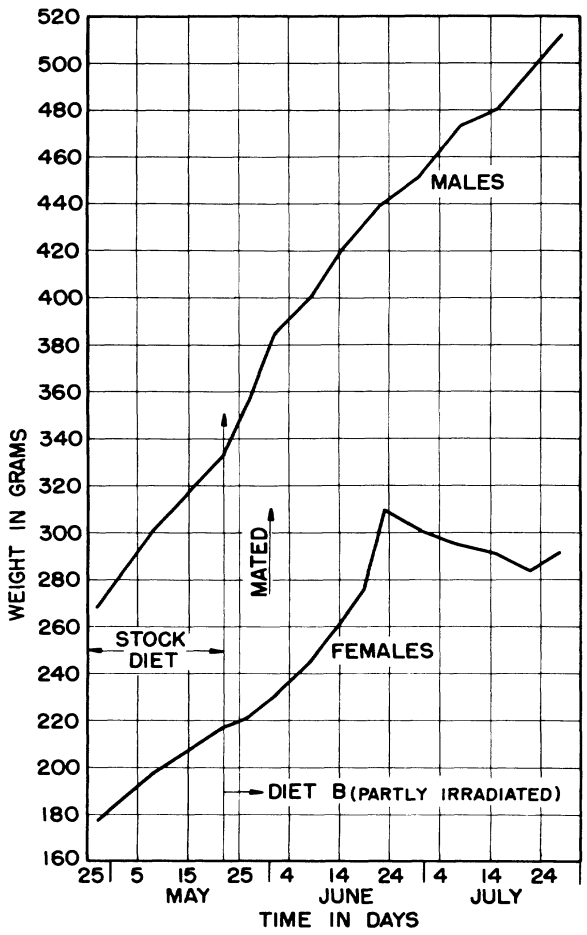

Fig. 84. Growth Curves for Rats Fed Diet B (partly irradiated) in Pilot Experiment No. 1.

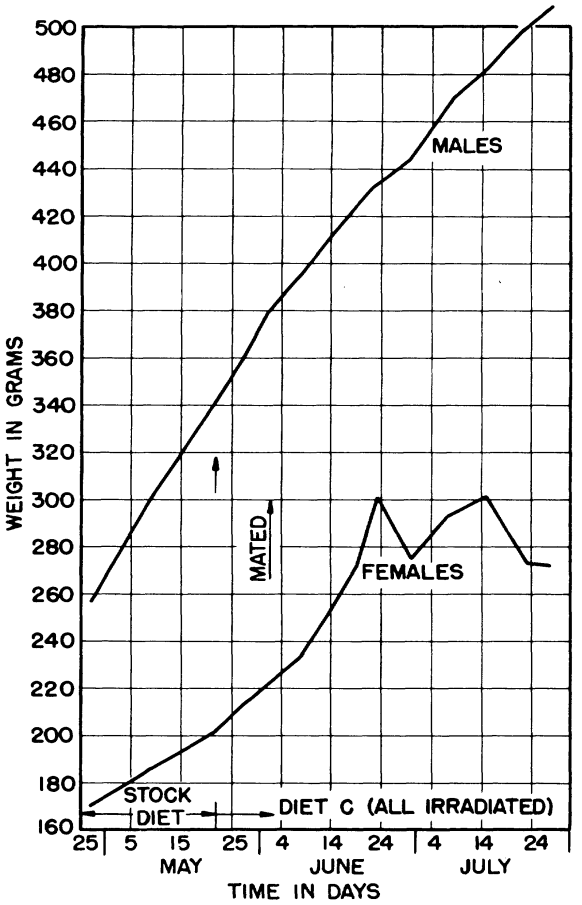

Fig. 85. Growth Curve for Rats Fed Diet C (all irradiated) in Pilot Experiment No. 1.

TABLE XXV

GROWTH DATA FOR MALES IN PILOT EXPERIMENT

	Number of Male	Weight on 5/22/53	Weight on 7/28/53	Gain in Weight	Standard Deviation of Mean
Control (A)	A_1	355	527	172	
	A_3	347	483	136	
	A_5	340	505	165	
	A_7	330	477	147	
	A_9	368	535	167	
	Mean	348	505	157	15.2
Partly Irradiated Diet (B)	B_{11}	363	558	195	
	B_{13}	318	496	178	
	B_{15}	309	485	176	
	B_{17}	340	507	167	
	Mean	332	512	180	11.7
All-Irradiated Diet (C)	C_{19}	328	471	143	
	C_{22}	352	491	139	
	C_{24}	348	583	235	
	C_{26}	336	492	156	
	Mean	341	509	168	44.8

5 per cent level of confidence, the differences between the mean increase in weight of the experimental groups and the control groups are not significant. This confidence test, however, is not strictly applicable to such small samples. It may well be that the difference is significant, i.e., that the rats fed

irradiated food grow faster. The main experiment using a greater number of animals will show whether or not this is true. If it is true, it might be explained on the basis that radiation partially depolymerizes the protein or in some other way makes it more readily available for enzymatic digestion.

b. Reproduction Studies. The reproduction was 100 per cent in each group, group A containing 4 females, and groups B and C, 5 females each. The data are given in Table XXVI.

TABLE XXVI

REPRODUCTION DATA FOR FEMALES IN PILOT EXPERIMENT

Group	No. of Females	Total No. Born	Average No. per Litter	Litter Range	No. Found Dead	Per Cent Weaned*	Average Weight at Weaning
A	4	47	11.75	10-13	5	97.4	55.8
B	4**	42	10.5	7-10	14***	71.1	45.2
C	5	60	12.0	10-13	2	100	55.4

*Since the litters are reduced to 10 on the fifth day, the percentage weaned is calculated on the basis of the reduced litter. This may give a false impression of the ability of the mother to rear the litter to weaning age. In the future the litters will be decreased to 10 on the day they are born.

**One female in this group fell approximately 4 feet to the floor. The next day, she gave birth to 7 pups; 4 were born dead and the other 3 died a few hours later. Her performance as a foster mother was good. This rat is not included in the table.

***One litter of 10 pups died before the fifth day. This was apparently due to a failure in lactation. Of the pups in the other 3 litters, 96.4 per cent were weaned. The rats have since been remated and this rat was again unsuccessful in lactating. This is probably a characteristic of the individual animal and not caused by the diet.

The success of the mothers in rearing their litters to weaning time was at least as good in the C group as in the control (A). It appears, however, that the performance in the B group was considerably poorer than that of the control group. Whether this was due to the partly irradiated diet will be established by future experiments.

After this experiment was well under way, it was learned that if the thiamine content of a rat ration is increased without a corresponding increase in the manganese content, the females will exhibit a loss of maternal instinct[5] and even cannabalism toward the young. Since the thiamine-manganese content was in a range where this effect may have been beginning to manifest itself, the thiamine content was reduced. The tables showing the vitamin content of the diet have been corrected to the new value.

c. Physical Characteristics of the Diet. The complete diet has the consistency of peanut butter but is not as sticky.

At first the food was rolled into a ball and put into the cage as such; however, the rats' hair became discolored and greasy from contact with the ball of food. The food is now put into 2- or 4-ounce ointment jars.

The B diet is somewhat more mushy and the C diet is more crumbly. There appears to be no difference in the rats' liking for the (A) diet and the B and C diets, the latter two which have received 2×10^6 rep.

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E. MICROBIOLOGICAL ASSAY OF IRRADIATED NUTRILITES

Personnel:

A. M. Elliott, Associate Professor of Zoology; L. E. Brownell, Associate Professor of Chemical Engineering; J. A. Gross, Research Assistant.

1. Introduction

A microbiological assay for essential amino acids and growth factors in an irradiated medium is being conducted by the personnel of Michigan Memorial-Phoenix project 73.

The effect of irradiation on the nutrient value of food is very complex because of the great variety of high-molecular-weight organic compounds which may be oxidized, reduced, or otherwise modified as a result of irradiation. It is possible to study the effects of irradiation on individual amino acids, vitamins, etc.; however, the results of such studies are complicated by the protective effect that one substance may have on the other, so that it is necessary to examine the complete spectrum of nutrients en masse to determine the overall effect.

Animal feeding experiments are being supported by Phoenix project 41 to investigate the effects of irradiation on the complete diet and selected portions of the diet using rats as experimental animals. It is expected that these tests will require two or more years before significant results are available. It is believed, however, that valuable information may be obtained at a much earlier date using protozoa to determine nutritive values of irradiated food constituents.

Professor A. M. Elliott and his associates in the Department of Zoology at the University of Michigan have made an extensive study of the nutritional requirements of the ciliated protozoan, Tetrahymena pyriformis (T. geleii). L. E. Hamilton, S. H. Hutner, and L. Provasoli[1] recognize the usefulness of protozoa in analysis and state that protozoa offer unique advantages in assaying metabolites that are difficult to analyze by chemical or enzymatic methods. These authors discuss the use of Tetrahymena pyriformis and point out that this organism has at last reached the stage where it can be grown in a completely defined medium, as a result of the recent availability of synthetic protogen. According to these authors, this fact has made Tetrahymena available for assaying wide varieties of metabolites.

J. O. Corliss[2] has made comparative studies on ciliates and mentions that pure culture strains such as Tetrahymena, Colpidium, Glaucoma, and Leucophrys have been used to advantage in studies on growth, nutrition, respiration, serology, genetics, morphogenesis, cannibalism, and even facultative parasitism. He feels that these small ciliates are now showing much promise as microbiological assay organisms in the determination of certain amino acids and vitamins.

C. Wu and J. F. Hogg[3] compared the amino acid composition of T. geleii with that of the average for various other microorganisms, except algae, and with that of vertebrates.

2. Description of Tetrahymena pyriformis

Tetrahymena pyriformis, strain E, was isolated in 1932 by A. M. Elliott from a lake in the Bronx, New York. Elliott established the organism in pure, bacteria-free culture and it has been maintained in that manner in suitable media since that time. Figure 86 is a photomicrograph of the organism, T. pyriformis, and Fig. 87 is a diagrammatic sketch of the organism prepared by Elliott.

The organism is classified as a holotrichous ciliate and is a member of the Colpidium-Glaucoma-Leucophrys-Tetrahymena group. Its natural habitat is fresh water (rivers, lakes, ponds, and streams). It is holozoic in nature; i.e., it feeds on particulate food. It may be cultured in an infusion of hay or lettuce, in peptone broth, or in a chemically defined medium. The nutritional requirements

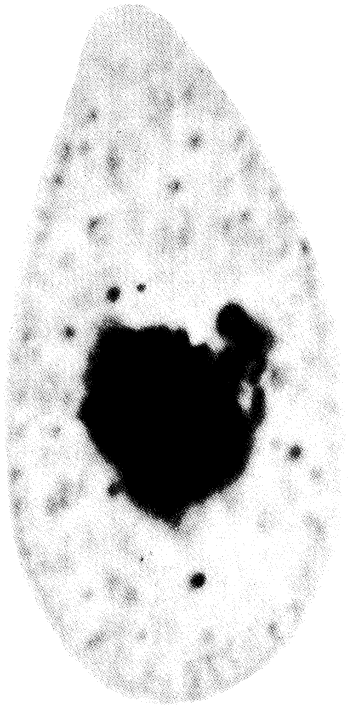

Fig. 86. Photograph of Tetrahymena pyriformis 2000X

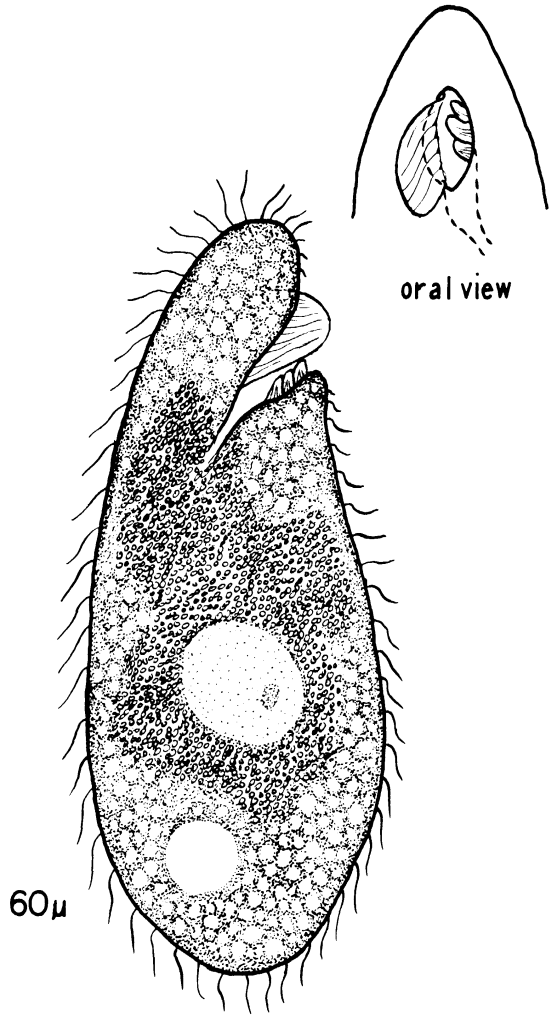

Fig. 87. Sketch of Tetrahymena pyriformis. (according to Elliott)

of the organism are quite similar to those of higher animals in regard to amino acids, vitamins, and physiologic salts.

3. Nutritional Requirements of T. pyriformis E

In 1949, A. M. Elliott[4] reported the amino acid requirements of T. pyriformis E. Earlier attempts to determine the specific amino acid demands of this organism were unsuccessful because at that time the essential amino acids, methionine and threonine, were unknown. Elliott's investigation was undertaken to determine the amino acid needs of strain E for the support of optimal growth. The protozoan was cultured free from other organisms in 15- by 125-mm pyrex test tubes.

All estimates of growth were carried out turbidimetrically with the use of a Klett-Summerson photoelectric colorimeter which has been adapted to this type of work. Readings were taken in Klett units from the inoculated tubes at the beginning of the experiment and again at the end, or at regular intervals throughout the course of the experiment. The difference between these readings was taken as indicative of the amount of growth.

The composition of the basic culture medium used in all these experiments, exclusive of the amino acids, is shown in Table XXVII.

TABLE XXVII

BASAL CULTURE MEDIUM FOR TETRAHYMENA PYRIFORMIS E FOR DETERMINING ESSENTIAL AMINO ACIDS

	mg/l
Dextrose	1,000
Sodium acetate	1,000
$MgSO_4 \cdot 7H_2O$	100
K_2HPO_4	100
$CaCl_2 \cdot 2H_2O$	50
$FeCl_3 \cdot 6H_2O$	1
$Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$	25
$CuCl_2 \cdot 2H_2O$	5
$MnCl_2 \cdot 4H_2O$	0.05
$ZnCl_2$	0.05
Ca pantothenate	0.10
Nicotinamide	0.10
Pyridoxine•HCl	2.00
Riboflavin	0.10
Pteroyltriglutamic acid	0.01
Biotin	0.0005
Thiamine•HCl	1.0
Choline•Cl	1.0
Yeast nucleic acid	100
Protogen	1.0 unit

The basic medium was prepared in large quantities, to which amino acids were added in graded amounts, depending on the experiment. The various concentrations were made in triplicate tubes. With the exception of the essential amino acid series, inoculations were made with pipettes, using a 0.2-ml inoculum from a heavy suspension of washed ciliates in their logarithmic growth phase maintained in purified medium. The organisms were washed several times with

sterile distilled water before serving as the inoculum; this procedure allowed a carry-over of very little, if any, of the original medium. Washing did not injure the protozoa in any way that could be detected with the microscope or in their subsequent growth. By using relatively high concentrations of organisms at the beginning of the experiments, the results could be read much sooner and were more consistent. It was difficult to obtain uniform initial inoculations with a loop. The results of the first two tubes cultured simultaneously were checked against each other; the third tube was a loop transfer which was used only to confirm the results of the first two tubes. All experiments were repeated several times, and the most consistent series were selected for reporting here.

All media were prepared in double-distilled water, and the pH was set at 7.4 at the start of the experiments. A test series demonstrated that growth is good over a pH range of 6.5-7.5 but that optimal growth in purified media occurs either at 6.5 or at 7.5; hence 7.4 was arbitrarily selected. All cultures were incubated in the dark at a constant temperature of 25°C.[4]

In these studies the ten amino acids essential for growth of rats plus serine were investigated. It was found that arginine, histidine, isoleucine, leucine, lysine, methionine, phenylalanine, serine, threonine, tryptophane, and valine are essential for the growth of T. pyriformis E.

A comparison of the amino acid metabolism of T. pyriformis with vertebrates was made by J. F. Hogg and A. M. Elliott.[5] These authors pointed out that T. pyriformis is similar to the higher animals in its nutritional requirements. Thus it may utilize glycogen, starch, and galactose as well as glucose and sucrose for its carbohydrate supply. In addition, it requires for optimal growth nearly all the B vitamins known at present and all the amino acids essential to mammalian growth and maintenance. However, T. pyriformis differs in requiring certain purines and pyrimidines, the amino acid, serine, and the growth factor, protogen, which has recently been identified as thioctic acid. The protozoan also differs in that it does not absolutely require an unsaturated fatty acid or any of the fat-soluble vitamins.[5]

Metabolic studies with T. pyriformis are of value from the standpoint of comparative biochemistry to allow a more critical comparison with higher animals. Such studies offer the possibility of recommending T. pyriformis as an experimental animal. In the study by Hogg and Elliott the compounds tested were related either structurally or metabolically to the amino acids and were used to replace or supplement essential amino acids used in the medium. The results indicated similarities of T. pyriformis to higher animals and in some cases showed that its requirements differ from those of other microorganisms. Twenty-four compounds were tested for their ability to replace 8 of the 11 amino acids

essential for T. pyriformis. It was found that there was a marked similarity between the amino acid metabolism of this ciliated protozoan and higher animals.

Using the technique described previously, A. M. Elliott[6] reported growth-factor requirements of Tetrahymena. In this study calcium pantothenate, nicotinamide, thiamine, riboflavin, pteroyltriglutamic acid (PGA), biotin, choline, and protogen were tested for their essentiallity in the normal nutrition of T. pyriformis E, since each of these was suspected of being necessary for growth. In the experimental procedure each factor was omitted singly from the complete medium and growth was carried through successive transfers until the organism failed to grow altogether, or until at least eight transfers were made in which growth continued normally (in such a case, the growth factor was considered nonessential). The basal medium used with these growth factors is given in Table XXVIII.

TABLE XXVIII

BASAL MEDIUM FOR DETERMINING ESSENTIAL GROWTH FACTORS

	mg/l
l-Arginine	600
l-Histidine	150
dl-Isoleucine	75
l-Leucine	75
l-Lysine	150
dl-Methionine	150
dl-Phenylalanine	150
dl-Serine	300
dl-Threonine	300
l-Tryptophane	150
dl-Valine	150
Dextrose	1,000
Sodium acetate	1,000
$MgSO_4 \cdot 7H_2O$	100
K_2HPO_4	100
$CaCl_2 \cdot 2H_2O$	50
$FeCl_3 \cdot 6H_2O$	1.0
$Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$	5.0
$CuCl_2 \cdot 2H_2O$	5.0
$ZnCl_2$	0.05
$MnCl_2 \cdot 4H_2O$	0.05
Yeast nucleic acid	100

In these studies it was found that tubes which did not contain pantothenate, nicotinamide, pyridoxine, PGA, or protogen would not support the growth of the ciliates after three transfers, and that some dying-out occurred in the second transfer. In tubes in which biotin and choline were omitted no reduction in growth was detected.

The conclusions were that Tetrahymena requires pantothenic acid, nicotinamide, pyridoxine, thiamine, PGA, riboflavin and protogen, for normal nutrition. Biotin, choline, and B_{12} are neither stimulatory nor essential.

4. Proposed Study with Irradiated Medium

Recently the culture conditions for T. pyriformis E have been improved in that a quantitatively minimal medium has been developed by Elliott and his co-workers.[7] The composition of this medium is listed in Table XXIX.

TABLE XXIX

IMPROVED MINIMAL BASAL MEDIUM FOR TETRAHYMENA PYRIFORMIS E*

Amino Acids	L	or	DL
Arginine	150 mg/l		---
Histidine	110		---
Isoleucine	50		100 mg/l
Leucine	70		---
Lysine	35		---
Methionine	35		35
Phenylalanine	50		100
Serine	90		180
Threonine	90		180
Tryptophane	20		40
Valine	30		60

Growth Factors	
Thiamine·HCl	1.00 mg/l
Riboflavin	0.10
Ca pantothenate	0.10
Niacin	0.10
Pyridoxine·HCl	2.00
Folic Acid (PGA)	0.01
Thioctic acid	0.001
	(1000 units/l)

Carbon Source	
Glucose	1000 mg/l
Na acetate	1000

Nucleic Acid	
Guanylic acid	25 mg/l
Adenylic Acid	25
Cytidylic acid	25
Uracil	25

Inorganic Salts	
K_2HPO_4	100.0 mg/l
$MgSO_4 \cdot 7H_2O$	10.0
$Zn(NO_3)_2 \cdot 6H_2O$	5.0
$FeSO_4 \cdot 7H_2O$	0.5
$CuCl_2 \cdot 2H_2O$	0.5

*Modified from Elliott (1949, 1950) by Elliott, Hogg, Slater, and Wu (1952).

Knowing the minimum growth requirements of this organism for amino acids makes it applicable as an assay method for possible radiation damage to these compounds.

The proposed experiment is to irradiate the medium in the 10-kilocurie cobalt-60 source, subsequently inoculate with Tetrahymena, and observe growth of the organism in the irradiated medium as compared to growth in control medium. Preliminary studies have already shown that an irradiation dosage of 4×10^6 rep damages the medium so that it will not support growth of the organism. In order to determine the cause of growth failure in the irradiated medium the following experiments will be performed:

(1) Complete nonirradiated medium will be added back to the irradiated, innoculated broth to determine recovery of growth.

(2) To ascertain the type of nutrilite that has been affected, groups of components (amino acids, vitamins, etc.) will be added back.

(3) Individual amino acids, vitamins, etc. will be added to the experimental tubes of medium to determine exactly which component has been destroyed.

Furthermore, it is thought advisable to irradiate separately each individual component, both in solution and in the dry state, to detect possible differences from effects on mixtures of components, since it is suspected that protection of some compounds by others occurs in mixtures.

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F. GAMMA IRRADIATION OF THE PROTOZOAN, TETRAHYMENA

Personnel:

A. M. Elliott, Associate Professor of Zoology, and J. V Slater, Research Assistant.

1. Introduction

A preliminary study of the effect of gamma radiation on the protozoan, Tetrahymena pyriformis, strain E, was supported by grant no. R-196 from the Horace H. Rackham School of Graduate Studies.

Studies were made of the effects of x- and ultraviolet rays on Tetrahymena (Elliott and Slater, 1951). It was found in both cases that older cultures were more sensitive than younger cultures. The same investigators tested the effect of exposing cultures to a graded dosage of gamma radiation from the 1-kilocurie cobalt-60 source at the Fission Products Laboratory of the University of Michigan.

2. Experimental Procedures

Tetrahymena pyriformis E was grown in a medium of the following composition:

TABLE XXX

STOCK CULTURE MEDIUM FOR TETRAHYMENA PYRIFORMIS E

	g/l
Tryptone	5
Proteose-peptone	5
KH_2PO_4	1
Dextrose	1
Na acetate	1
Yeast extract	0.1
Thiamine•HCl	0.002
Adjust to pH 7.4	

Cultures were grown for 48 hours (young cultures), and for 7 to 10 days (old cultures), before being harvested for irradiation. At the designated age of culture, the ciliates were centrifuged, washed several times with sterile glass-redistilled water, suspended in distilled water, and irradiated for periods of from 1 to 8 hours. Old and young irradiated ciliates were reinoculated respectively into tubes of medium, their subsequent growth was measured turbidimetrically in a photoelectric colorimeter, and results were compared.

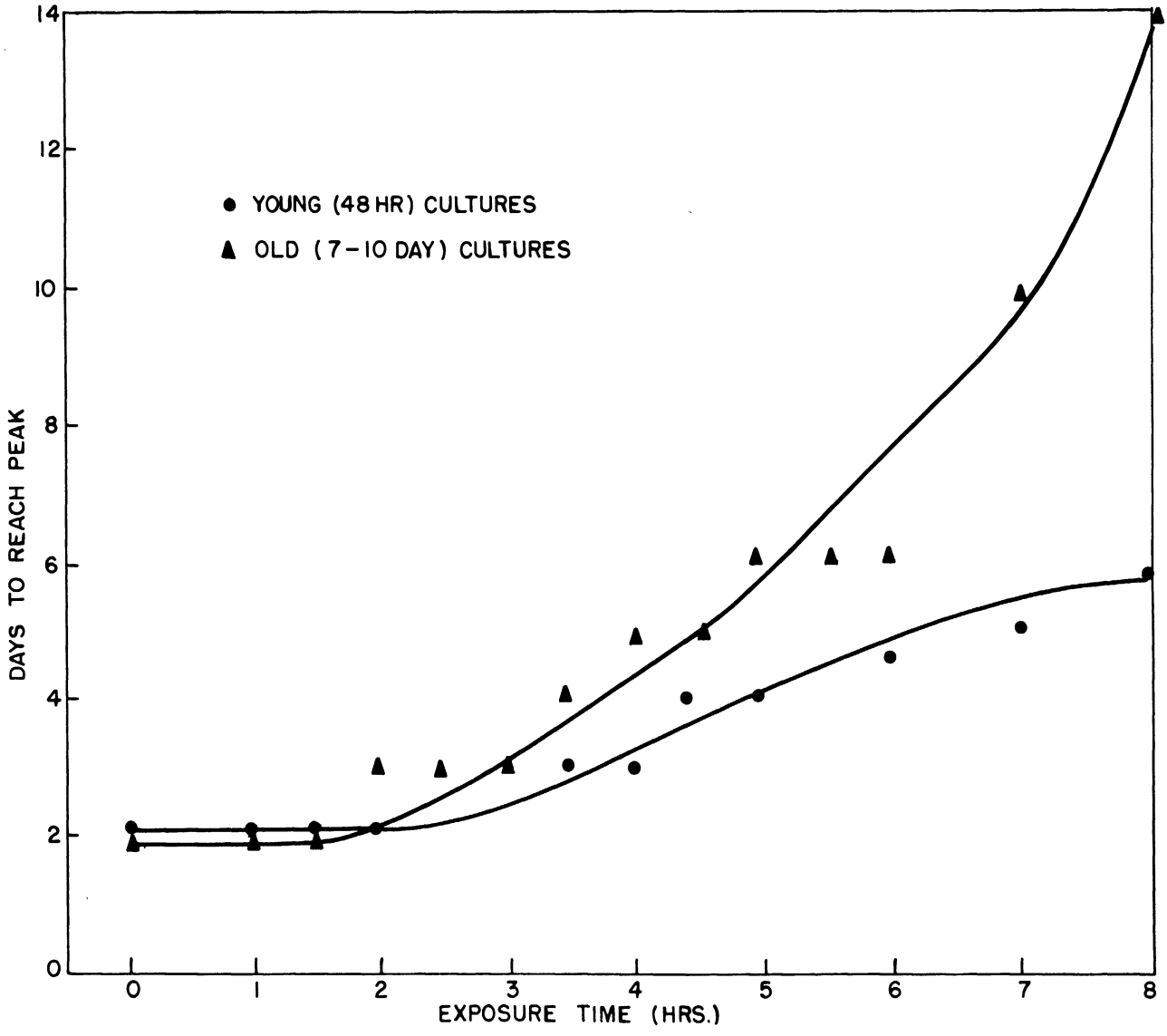

Fig. 88. Response of Tetrahymena to Cobalt-60 Irradiation.

3. Results

In neither case was an effect noted after 1-1/2 hours of exposure. However, after 2 hours of exposure a definite delay in reaching maximal growth (approximately 150,000 protozoa/ml of medium) became obvious. After 8 hours of irradiation it took young cultures 6 days to reach a maximum, while old cultures reached peak growth only after 14 days of incubation (Fig. 88). Experiments in which the basal synthetic medium recommended by Elliott (1949, 1950) was used yielded similar results. The results are similar to those found with x- and ultraviolet irradiation: old cultures are more susceptible to irradiation damage than are young cultures.

G. IRRADIATION OF FOOD

Personnel:

L. E. Brownell, Associate Professor of Chemical Engineering and Director of Fission Products Laboratory: L. L. Kempe, Assistant Professor of Bacteriology and Assistant Professor of Chemical Engineering; J. T. Graikoski, Bacteriologist, Phoenix project 41; and J. V. Nehemias, Research Associate and Health Physicist for Fission Products Laboratory.

1. Introduction

Since the preparation of Progress Report No. 4, the 10-kilocurie Co^{60} source has been received and put into operation in this laboratory as described in Part IV of this report. With the new source it is possible to irradiate much larger volumes of food than could be handled with the 1000-curie Co^{60} source. Another advantage of the new radiation source is that conventional-type food containers can be utilized for the packaging of foods. It is now possible to use commercial types of containers and food packaging procedures in the experiments on radiation sterilization and pasteurization.

The research on irradiation of foods performed during the past year in the Fission Products Laboratory has been supported by Michigan Memorial-Phoenix project 41. Budget limitations necessitated a careful selection of problems. As the Fission Products Laboratory was the only known laboratory that had an adequate experimental gamma source capable of irradiating appreciable quantities of food, the decision was made to allocate the major portion of the funds of Phoenix project 41 to an extensive animal-feeding experiment, which is described in Part IIID of this report. Studies of the effect of gamma radiation on microorganisms have been in progress with support by Phoenix project 41 and the help of a bacteriologist, Mr. J. T. Graikoski.

The feeding experiments and studies on microorganisms require the major portion of the man-hours available from support by Phoenix project 41. As a result, the studies on packaging, irradiating, and organoleptic testing of food sterilized or pasteurized by irradiation have been very limited and have taken second place to the other studies underway. However, there is ample irradiation capacity for studies on food and it is hoped that additional support will make it possible to direct more attention to the organoleptic and packaging problems.

2. Procedures for Preparation and Irradiation of Foods

Food or other biological material for irradiation studies are placed in No. 2 (3-7/16 by 4-9/16-inch) or No. 10 (6-3/16 by 7-inch) tin containers. Sealing of these containers is accomplished by means of a Western Type Closing Machine* (Fig. 89). Since tin containers do not permit visual observation of irradiated samples, quart- and pint-size mason jars are also used. Although considerable glass discolorization occurs a certain degree of visual observation is possible, and use of the 2-piece Kerr-type lid permits a very effective method of packaging samples in a vacuum. The caps are fitted loosely on jars containing samples to be irradiated. The jars are placed in a typical bell jar and evacuation is accomplished by means of a vacuum pump. After the desired evacuation is obtained the pump is turned off and the vacuum broken quickly on the bell jar. The Kerr lid, acting as a one-way valve is sealed snugly against the mouth of the mason jar. Vacuums of approximately 28 inches (Hg) as measured with a canner's test gauge are obtained by this method.

Samples for irradiation are placed next to the wire cage which encloses the source when it is in the raised position. In order to give them a uniform irradiation, the samples are rotated on turntables powered by small synchronous gearhead motors rotating at 1 rpm. Since the bottom of the radiation source extends nearly 10 inches above the floor level of the cave, the turntables are placed on an elevated platform. In this way samples being irradiated are kept near the central plane of the source. Outside the source at a minimum distance from the cage it is possible to irradiate eight No. 10 cans simultaneously. By stacking two No. 2 tin containers on each turntable a total of 20-No. 2 cans can be irradiated simultaneously in this position. Since slight variation in dose rate occurs along the axis of the radioactive rods, it is necessary to turn the cans end-over-end at intervals in order to insure as uniform a dose as possible. By placing samples at a greater distance from the source, a greater number can be irradiated, but the radiation time is increased.

*The closing machine and cans were obtained through the courtesy of the American Can Company.

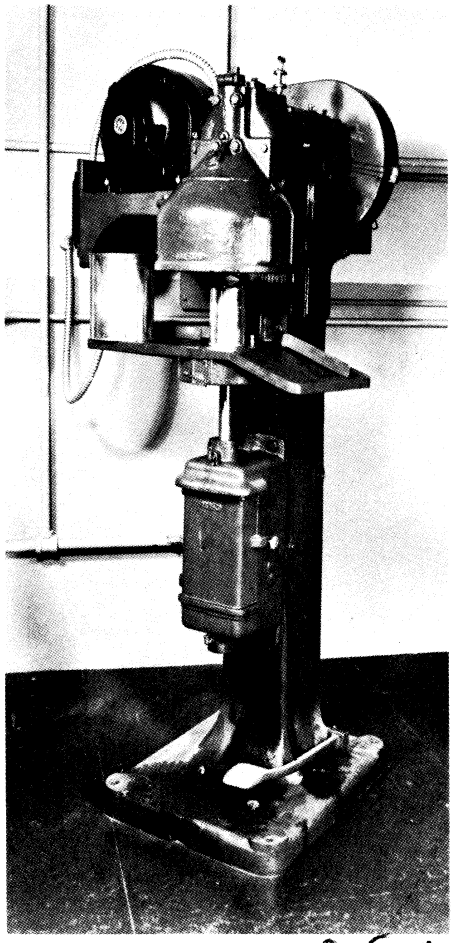

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Fig. 89. Western Type Can Closing Machine.

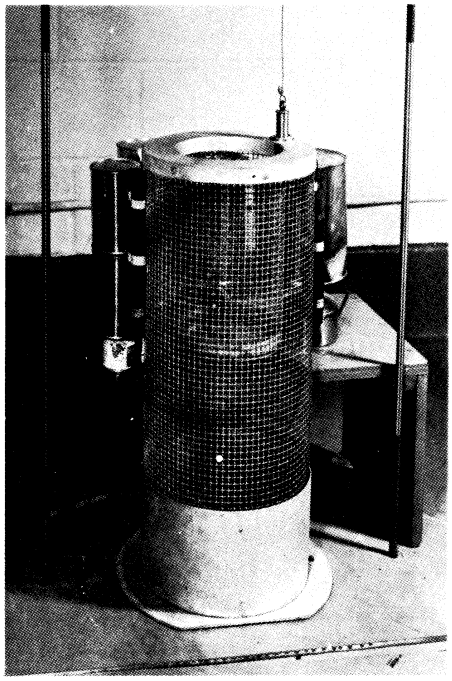

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Fig. 90. Wire Cage for 10,000-Curie Source with Food Samples in Background.

In the present experiments only about 50 lbs of food per day is being sterilized, but more than 200 lbs per day probably could be sterilized and 1/2 ton of food per day probably could be pasteurized. This production is based on an average daily rate. In the center position, which is the point of high flux (about 300,000 rep/hr) food samples can be sterilized in about 8 hours. A No. 10 can fits very nicely in this position, making the actual capacity about 10 lbs. A larger number of smaller containers can be irradiated in this position. On the outside of the source about 50 lbs of food can be irradiated, but as the flux is only about 80,000 rep/hr, the time required for sterilization is about 30 hours. In some of the feeding experiments in which a radiation dose of 2-1/2 million rep is to be accumulated, the food can be stored for several days and positions of still lesser flux can be used.

Figure 90 shows some samples and their relation to the source, during irradiation. For low-temperature irradiation the samples can be placed in an

insulated box surrounding the source. The box can be cooled either by frozen CO_2 or ice. Samples can also be irradiated at elevated temperature by use of heating coils if this is desired.

Since the preparation of Progress Report No. 4, a considerable number of man-hours have been spent in installation and development of new techniques with the new source.

3. Organoleptic Tests

Several exploratory experiments have been performed irradiating foods in No. 10 tin containers and in mason-type jars. The purpose was to study intact foods in larger volumes than could be irradiated in the small Co^{60} source. Foods were packed in No. 10 containers for irradiation without evacuation. Several samples were precooked before irradiation in order to compare the effects of radiation on raw and cooked foods. Gross changes due to irradiation were observed and some of the typical results are reported in Table XXXI.

TABLE XXXI

EFFECTS OF GAMMA RADIATION ON FOODS

Sample	Dose Megarep	Texture	Color	Taste
Whole Potatoes				
Raw	2.5	Slight Softening	Browning	Off Slight
Pre-Cooked	2.5	No Change	No Change	Good
Whole Carrots				
Raw	2.5	Soft	No Change	Good
Pre-Cooked	2.5	No Change	No Change	Good
Cabbage				
Raw	2.5	Softening	Slight Bleaching	Off
Pre-Cooked	2.5	No Change	No Change	Off
Bread	2.5	No Change	Light Straw	Good
Grapefruit	0.5	Slight Softening	No Change	Good
Oranges	0.5	Slight Softening	No Change	Off
Bing Cherries	0.5	Slight Softening	No Change	Good
Bing Cherries	1.0	Softening	No Change	Good

Some preliminary experiments to investigate the effect of storage at elevated temperatures were also initiated. In one experiment approximately 1-1/2 pounds of fresh side of pork containing both lean and fat were irradiated in mason jars evacuated as described previously. Samples received approximately 3.5×10^6 rep, a dose assumed to give sterility. The unirradiated controls were stored at 37.5°C. The samples were removed 11 weeks later, but the controls had to be disposed of earlier because of spoilage. Visual observation showed that samples developed a certain amount of fluid on storage. The fluid solidified at ice-box temperature and appeared to consist of lower-melting-point fats and other cell juices. Some enzymatic degradation undoubtedly occurred; however, there was no evidence of microbial growth on opening. At the time of opening the sample jars still held a vacuum. The lean portion of the meat was red but of a lighter color than fresh meat. Fatty portions were solid and white. The sample was tasted after autoclaving at 15 lbs for 15 minutes. The flavor of the sample was good but bland, as if it needed seasoning. The fat did not seem to taste rancid.

Another experiment is in progress at present using top round beef. Two different samples of meat, each weighing approximately 1 pound and consisting of 3 small steaks, were packed in mason jars. Each series consisted of 7 jars with 2 controls. The samples were evacuated at 14 mm Hg for 10 minutes as described previously, and then received a dose of 3.5×10^6 rep. One sample of each was tested immediately after the last radiation, while the other samples were placed in a hot room at 37.5°C. Samples tested after irradiation did not have the distinctive red color of fresh meat, but still had a very good red color. A slight amount of fluid was present. The odor of the raw samples was characteristic of irradiated meats, although the odor of one sample was more pronounced than that of the others. These samples were pan-fried over slow heat and tasted. Both samples had an off-taste, but again one was more pronounced than the other. Vacuum packing seemed to improve the odor and taste of irradiated samples. The texture of the meat was softer and more tender than that of the control.

A sample of each series was removed 1 week later. One observation was that the characteristic irradiated odor was absent. The cooked samples had a better flavor than those tested immediately after irradiation. The amount of fluid in the jar had increased. Another sample of each series was tested after 1-month storage at 37.5°C. These samples were similar in appearance to the earlier samples. The samples tested were edible but had a slight bitter taste which appears to develop on storage.

Other samples will be stored and tested up to a year's incubation, and information from these preliminary experiments will be used in planning additional tests. A definite change occurs subsequent to irradiation and incubation at an elevated temperature which will be investigated further.

4. Further Experiments

A statistician and a food chemist trained in food and nutrition and experienced in the organoleptic evaluation of foods are being added to the laboratory personnel. The test kitchen of the Food Service Department of the University of Michigan will be made available for the preparation of food for evaluation by a taste panel. The results of these organized taste-panel experiments will be described in the next progress report.

H. STERILIZATION OF FOOD-PACKAGING MATERIALS

Personnel:

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1. Introduction

Gamma-ray sterilization of food containers made from paper, plastic, metal laminate, cloth, etc. is of current interest because large sources of gamma radiation from waste fission products are potentially available. The problem of sterilizing packaging materials becomes increasingly important as the need for sterile food containers receives wider recognition by industrial and public health authorities. Owen[5] has recently presented data to show that food packages are usually contaminated with microorganisms when ready for filling, although containers for dairy and bakery products have recently been improved in this respect. Unlabeled tin and glass containers can be sterilized with steam or hot air. However, such treatment of paper, cardboard, plastics, printed cloth, etc. would usually cause irreparable damage.

The possible large-scale availability of gamma radiation from fission by-products suggests an alternative sterilizing technique. Gamma radiation will instantaneously penetrate considerable depths of material and kill microorganisms throughout the mass. Fortunately, this radiation is reported to be essentially harmless to common organic packaging materials at the levels required for sterilization [1,5,7].

Commercial adaptation of this process would probably involve establishment of a centrally located sterilizing plant. Here packaging materials, inclosed in hermetically sealed paper boxes or wrappings, could be sterilized and then shipped to food-processing plants for storage in this sterile condition until needed. It should be pointed out that the cost of irradiation with waste fission products has not yet been determined accurately, so the economic feasibility of

any process based on these materials cannot be properly evaluated at the present time.[8] On the other hand, there may be specialized instances where the unusual advantages of gamma-ray sterilization will make its use mandatory. For these reasons, it was considered desirable to test the feasibility of using gamma radiation for sterilizing food-packaging materials. This research was supported by Michigan Memorial-Phoenix project 41.

2. Experimental Tests

Two bacteria, E. coli and B. stearothermophilus NCA 1518, were used in this study. These organisms were selected because they represent approximate minimum and maximum degrees of radiosensitivity. Also, since it has been shown[3] that yeast and mold spores are killed by radiation doses of approximately the same intensity that are effective against E. coli, it was not considered necessary to include yeasts and molds in this work.

The spore suspensions were prepared by growing B. Stearothermophilus on the surface of nutrient agar in Roux flasks at 55°C for 48 hours. The cells were next harvested by washing from the agar surface with a small amount of sterile distilled water, strained through glass wool to remove agar clumps, centrifuged to recover the cells, suspended in sterile distilled water, and then boiled for 5 minutes to kill the vegetative cells. This spore suspension was stored at 4°C for future use.

E. coli cultures were grown for 24 hours at 37°C in nutrient broth, and then used in this condition.

Specimens for irradiation experiments were inoculated by placing either 0.2 ml of an E. coli broth culture or 0.2 ml of the B. stearothermophilus spore suspension on a 1-square-inch section of the test material. The impregnated specimens were next dried at 37°C for 24 to 48 hours, after which they were placed inside individual sterile envelopes made from filter paper.

For irradiation, the envelopes containing inoculated specimens were placed near the center of a corrugated-paper box which was tightly packed with paper to simulate a box full of packages. The box was cubical and measured 14 inches on each edge. During irradiation it rested on a stand adjacent to the 10,000-curie cobalt-60 gamma-ray source and was positioned so that a 1-inch air space existed between the box face and the source. The distance between the cobalt-60 rods and the paper envelopes was thus established at 7 inches.

Figure 91 is a photograph showing an interior view of the radiation "cave". The source is in the lowered position. The experimental box is shown next to the cage which covers the source when it is raised for use. The operator is placing an envelope containing a contaminated plastic sample in position.

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Fig. 91. View of Radiation Chamber with 10,000-Curie Source Down, Showing Placing of Envelope Containing a Contaminated Plastic in Position.

The No. 10 tin cans of food in the background are being irradiated in connection with another experiment.

After irradiation, the specimens were transferred aseptically to a sterile flask containing 100 ml of sterile distilled water. The flasks were shaken at 200 revolutions per minute on a New Brunswick shaker which develops a 1-inch-diameter circular agitation pattern. Control samples were treated identically except that they were not irradiated. Decimal dilutions were next made from the sterile flasks into culture media suitable for counting the resulting colonies.

Beef extract-peptone media was used for plating E. coli. Incubation was carried out for approximately 30 hours at 37.5°C before counts were made. For B. stearothermophilus, Bacto-Dextrose Tryptone agar (Difco) was used and the plates were incubated for 48 hours at 55°C before counting. A conventional-type Quebec Colony Counter was employed in all counting procedures.

In order to arrive at dosages to be used for sterilization, samples were irradiated at various time intervals. Tables XXXII to XXXV give the results of a series of experiments.

3. Results

The experimental data from representative experiments shown in Tables XXXII to XXXV demonstrate that packaging materials can be sterilized with gamma

TABLE XXXII

EFFECT OF GAMMA RADIATION FROM COBALT-60
ON E. COLI PRESENT ON MANILA PAPER

Paper Sample Number	Total Dosage-Rep (in air)	Number of Organisms on the 1 square inch of paper
Control	0	3.8×10^7
No. 1 Irradiated	50,000	7.0×10^5
No. 2 Irradiated	99,000	2.5×10^5
No. 3 Irradiated	149,000	7.5×10^3
No. 4 Irradiated	199,000	0
Control	0	1.4×10^7
No. 1 Irradiated	284,000	0
No. 2 Irradiated	409,000	0
Control	0	1.5×10^8
No. 1 Irradiated	522,000	0
No. 2 Irradiated	684,000	0
No. 3 Irradiated	994,000	0
No. 4 Irradiated	1,086,000	0

TABLE XXXIII

EFFECT OF GAMMA RADIATION ON E. COLI
PRESENT ON VARIOUS PACKAGING MATERIALS

Material	Total Dosage Rep(in air)	Number of Organisms on 1 square inch of material	
		Control	Irradiated
Paraffin Paper	199,000	1.65×10^4	0
Cryovac	199,000	6.0×10^3	0
Polyethyl-ene	199,000	1.65×10^4	0
Cloth	199,000	7.0×10^4	0
Saran	199,000	8.0×10^3	0

TABLE XXXIV

EFFECT OF GAMMA RADIATION ON
B. STEAROTHERMOPHILUS PRESENT ON MANILA PAPER

Paper Sample Number	Total Dosage Rep (in air)	Number of Organisms on 1 Square Inch of Paper
Control	0	3.0×10^6
No. 1 Irradiated	143,000	6.5×10^6
No. 2 Irradiated	252,000	3.0×10^6
No. 3 Irradiated	521,000	7.5×10^5
No. 4 Irradiated	1,950,000	3.0×10^3
Control	0	3.2×10^5
No. 1 Irradiated	521,000	2.2×10^5
No. 2 Irradiated	1,200,000	0
No. 3 Irradiated	1,790,000	0
No. 4 Irradiated	2,330,000	0

TABLE XXXV

EFFECT OF GAMMA RADIATION ON
B. STEAROTHERMOPHILUS PRESENT ON VARIOUS PACKAGING MATERIALS

Material	Total Dosage Rep (in air)	Number of Organisms on 1 Square Inch of Material	
		Control	Irradiated
Paraffin Paper	1,310,000	7.0×10^6	1.7×10^5
Cryovac	1,310,000	9.5×10^6	1.5×10^5
Polyethylene	1,310,000	16.5×10^6	4.75×10^4
Cloth	1,310,000	2.3×10^6	0
Saran	1,310,000	7.5×10^6	0
Paraffin Paper	1,900,000	2.7×10^7	0
Cryovac	1,900,000	3.25×10^6	0
Polyethylene	1,900,000	3.0×10^7	0
Cloth	1,900,000	50,000	0
Paper	1,900,000	20,000	0

radiation from cobalt-60. The dosages required for sterilization are approximately ten thousand times smaller than those reported to cause damage to plastics,[5,2] cellulose, and cotton fibres[7]. No damage, such as color change, embrittlement, clouding, etc., was observed in any of the samples of plastic, cloth, or paper irradiated in the course of the work reported in this paper.

These results indicate that the heat- and moisture-sensitive materials tested, and possibly a variety of others which are used in the fabrication of food packages, can be sterilized in quantity by gamma irradiation without significant damage to the materials.

During the course of the work, it was also found practical to sterilize "Lusteroid" plastic centrifuge tubes with gamma radiation for routine use in our laboratory. Several successive sterilizations have caused no apparent damage to these tubes.

The results of this study further suggest that large unit quantities of a rather wide variety of packaged organic materials can be sterilized by gamma radiation. For example, it might be desirable to irradiate cotton fibres, leather, etc. in bulk to prevent mildew damage or to prepare them for use in special situations where sterile materials are required.

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I. EFFECT OF RADIATION ON MICROORGANISMS

Personnel:

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In the study of preservation of food from spoilage by microorganisms it is necessary to know the dosage required to inactivate the organisms most commonly incriminated in food spoilage. Studies are underway on the effect of radiation on various bacteria. These studies have not been completed and therefore will not be reported in detail until the next progress report.

Emphasis is being placed on spore-forming bacteria, since these are recognized to be the most resistant to radiation. Strains being used are B. stearothermophilus NCA1518, Putrifactive anaerobe NCA3679, Thermophilic anaerobe NCA3814, Cl. botulinum 62-A, and Cl. botulinum 213-B. The technique calls for inoculating known concentrations in food substrates [being irradiated in tin containers (No. 2 cans)] and post-irradiation recovery of organism. Since many factors affect radip-sensitivity of organisms (such as oxygen tension, presence of anti-oxidants, and temperature) it will be necessary to determine the effect of radiation on microorganisms for each set of conditions.

J. IRRADIATION OF HUMAN BLOOD CULTURE MEDIA

Personnel:

Professor L. L. Kempe, Assistant Professor of Bacteriology and Assistant Professor of Chemical Engineering; and Nancy J. Williams, Graduate Student, Department of Bacteriology.

1. Introduction

This portion of the report is based on research done by Nancy J. Williams for Graduate School credit.

The original problem was to investigate gamma radiation as a method for the sterilization of the bacteriological media which cannot be sterilized by known present-day methods. The sterilization must leave the media unchanged and permit normal growth of the organisms cultured on it. With this purpose in mind gamma irradiation of blood media was attempted, but the results obtained with the first experiment indicated that the media had been changed by this treatment. Alpha- and beta-hemolytic streptococci would no longer undergo their

normal reactions when grown on it. Since the irradiated media could not be used as originally intended, the research continued along the lines of investigating the possible mechanisms of this change.

2. Materials and Methods

Human blood collected aseptically by the Blood Bank of the University Hospital was used for these experiments. Blood to be irradiated was taken from the original container using sterile technique and placed in a glass tube 1-1/4 inches in diameter and 12 inches long, which was placed in an aluminum cylinder 1-1/2 inches in diameter and 13 inches long. This cylinder was in turn placed in the 1-kilocurie gamma source in the Fission Products Laboratory.

A Wright's stain was performed on normal and irradiated red blood cells in one of the experiments, according to the following procedure:

1. A slide with the dried blood film on it was placed in a perfectly horizontal position and Wright's stain added to cover the surface completely. The stain was allowed to stand for 1 to 2 minutes.

2. Then an equal number of drops of buffered water were added to bring the meniscus to the margin.

3. The mixture was allowed to stand for 4 to 6 minutes. (A green metallic sheen is indicative of proper staining.)

4. The stain was washed off with distilled water. (Do not pour the stain off before washing, because this will leave a precipitate.)

5. The bottom of the slide was wiped to remove dried stain and the slides were placed endways on filter paper to allow excess fluid to drain off.

The organisms used for the experimentations were: (1) Beta-hemolytic streptococcus, Strain A-3; (2) Alpha-hemolytic streptococcus; and (3) Hemophilus pertussis. The media necessary for the cultivation of these organisms was of two kinds. Bordet-Gengou agar media was used for the cultivation of the Hemophilus pertussis, and blood agar media for the cultivation of the alpha- and beta-hemolytic streptococci. The Bordet-Gengou media was prepared as follows:

1. The base agar was obtained from the Department of Bacteriology Media Room in 200-ml bottled quantities.

2. The base was heated in the autoclave until completely liquefied. It was then cooled to between 45° and 50°C by placing the flask in a water bath and the blood was added to make a concentration of from 20 to 25 per cent.

3. Pour plates were made and these plates used for cultures which had been prepared within 72 hours.

The blood agar media for the alpha- and beta-hemolytic streptococci was prepared in a manner similar to that used for the Bordet-Gengou media except blood was added to make a concentration of 5 per cent in BHI base agar obtained from the Department of Bacteriology Media Room in 200-ml bottled quantities.

The chocolate agar used in one of the experiments was prepared as follows:

1. BHI base agar was obtained from the Department of Bacteriology Media Room in 200-ml bottled quantities.

2. The base was heated in the autoclave until completely liquefied. It was then cooled to between 45° and 50°C by placing the flask in a water bath and the blood was added to make a concentration of 5 per cent.

3. The blood media was then heated to 85°C and held there for 10 minutes. It was then allowed to cool to 45°C again.

4. Pour plates were made and these plates used for cultures which had been prepared within 72 hours.

For the work with blood fractions, the citrated whole blood was placed aseptically in large centrifuge bottles and centrifuged at a great enough speed to separate the plasma and red blood cells. The plasma was then siphoned off into separate bottles and the red blood cells placed in containers suitable for irradiation purposes.

The blood used in this work was irradiated with cobalt-60 gamma radiation for approximately 29 hours in each instance (equivalent to a dose of 2,234,000 rep in air), except the plasma in the experiment on blood fractions, which was irradiated for 44 hours (3,203,000 rep in air).

3. Experiments and Results

a. Experiment 1. Fifteen blood agar plates were inoculated with alpha-hemolytic streptococci. Five of these plates had been prepared with normal, non-irradiated blood and were to serve as controls. Ten of the plates had been prepared with irradiated blood and were to show the effect of the irradiated blood on alpha streptococci. The plates were incubated at 37°C for 48 hours. The results obtained are tabulated in the following table.

TABLE XXXVI

ALPHA-HEMOLYTIC STREPTOCOCCI GROWN ON IRRADIATED BLOOD

Control Plate	Growth	Hemolysis	Irradiated Plate	Growth	Hemolysis
1	++++	++++	1	++++	±
2	++++	++++	2	++++	±
3	++++	++++	3	++++	±
4	++++	++++	4	++++	±
5	++++	++++	5	++++	±
			6	++++	±
			7	++++	±
			8	++++	±
			9	++++	±
			10	++++	±

This would indicate that although the organisms produce essentially the same amount of growth on irradiated media as on normal media, there is a marked decrease in the amount of hemolysis produced by the organisms after 48 hours.

The plates were examined again at the end of one week; definite alpha-hemolysis had appeared at this time, indicating that it was delayed rather than completely inhibited on irradiated media.

b. Experiment 2. Fifteen blood agar plates were inoculated with beta-hemolytic streptococci. Five of these plates were prepared with normal, nonirradiated blood to serve as controls. Ten of the plates were prepared with nonirradiated blood and were to show the effect of the irradiated blood on beta streptococci. The plates were incubated at 37°C for 48 hours. The results obtained are tabulated in the following table.

TABLE XXXVII

BETA-HEMOLYTIC STREPTOCOCCI GROWN ON IRRADIATED BLOOD

Control Plate	Growth	Hemolysis	Irradiated Plate	Growth	Hemolysis
1	++++	++++	1	++++	-
2	++++	++++	2	++++	-
3	++++	++++	3	++++	-
4	++++	++++	4	++++	-
5	++++	++++	5	++++	-
			6	++++	-
			7	++++	-
			8	++++	-
			9	++++	-
			10	++++	-

As was the case with the alpha streptococci, the organisms produced essentially the same amount of growth on irradiated media as on normal media. Here, however, there was no hemolysis produced by the organisms on the former media after 48 hours.

The plates were examined again at the end of one week and there was still no hemolysis, so a final examination was made at the end of two weeks. No hemolysis had appeared at this time, either, so the plates were discarded and it was concluded that irradiation of the media permanently inhibited beta hemolysis.

c. Experiment 3. Forty plates of Bordet-Gengou media were inoculated with Hemophilus pertussis. Twenty plates were prepared with normal, nonirradiated blood to serve as controls. Twenty plates were prepared with irradiated blood to determine the effect of the irradiated blood on the growth of the pertussis organisms. The plates were incubated at 37°C for 48 hours and examined for differences in the amount or kind of growth. It was found, however, that H. pertussis grows equally well on media prepared with either irradiated or nonirradiated blood (i.e., ++++ growth on all plates) and the type of growth is no different for the two types of media. Thus, results were negative, indicating that irradiation does not change the blood in a way which will affect the H. pertussis organisms, so they were dropped from further experimentation in this research.

d. Experiment 4. Since the blood had obviously been changed in a way which affected the hemolytic abilities of streptococci when it was irradiated, the next step was to examine the red blood cells of both irradiated and normal blood to observe any difference between the two, since this is the level at which hemolysis of blood operates. Blood films were made and stained by Wright's stain according to the method described in Part 2 of this section.

The slides were examined in oil immersion under the microscope and it was observed that the cell wall of both the irradiated and normal blood cells was intact, the cell wall not being destroyed by irradiation. However, irradiation apparently had changed the permeability of the cell wall, because cells of irradiated blood were ghost cells; i.e., they did not take the Wright's stain, indicating a loss of their hemoglobin into the surrounding medium, while the normal cells took a Wright's stain, indicating that the hemoglobin was still intact within the cell. This would account for the almost black color of irradiated blood and is, apparently, the major difference between normal and irradiated cells.

e. Experiment 5. Thirty plates of media prepared with freshly irradiated blood (ten plates were Bordet-Gengou media, twenty plates were blood agar media) were inoculated with the following three organisms: (1) Beta-hemolytic

streptococci, (2) alpha-hemolytic streptococci, and (3) H. pertussis. The purpose of the experiment was to determine whether or not toxic products are formed in irradiated blood which would prevent the growth of organisms cultured on such blood within 24 hours after its removal from the irradiation source. The work of previous investigators had shown that if such products are formed, they disappear on standing. The blood was therefore removed from the Co^{60} source at 1:00 p.m. on March 25, 1953, and was used in the media at 4:00 p.m. on the same day. The plates were inoculated with the organisms indicated above at that time and incubated at 37°C for 48 hours. The following results were obtained:

TABLE XXXVIII

GROWTH OF ORGANISMS ON FRESHLY IRRADIATED BLOOD

Alpha-hemolytic Streptococci

Plate	Growth	Hemolysis
1	++++	+++
2	++++	+++
3	++++	+++
4	++++	+++
5	++++	+++
6	++++	+++
7	++++	+++
8	++++	++
9	++++	++
10	++++	++

Beta-hemolytic Streptococci

Plate	Growth	Hemolysis
1	++++	+
2	++++	+
3	++++	+
4	++++	+
5	++++	+
6	++++	+
7	++++	+
8	++++	+
9	++++	+
10	++++	+

Hemophilus pertussis

Plate	Growth
1	++++
2	++++
3	++++
4	++++
5	++++
6	++++
7	++++
8	++++
9	++++
10	++++

As is seen from these results, no toxic products were formed in the blood due to irradiation which would prevent the growth of the organisms inoculated on it within 24 hours after radiation. In fact, both alpha- and beta-hemolysis seemed somewhat stronger on this media than on older irradiated media, although still not equal to that on normal blood media.

f. Experiment 6. In this experiment, the plasma of citrated whole blood was separated from the red blood cells by centrifugation to determine in which fraction of the blood the factor is located which prevents hemolysis of beta-hemolytic streptococci and inhibits that of alpha-hemolytic streptococci temporarily. These fractions (plasma and red blood cells) were irradiated separately and then an irradiated fraction was recombined with its corresponding unirradiated fraction. That is, irradiated red blood cells were combined with normal plasma and, using another sample of blood, irradiated plasma was combined with normal red blood cells. Media for growing the streptococcal organisms was then prepared using this blood to see which fraction contained the hemolysis-inhibiting factor.

Fifteen plates were prepared using irradiated red blood cells plus normal plasma. Five of these were inoculated with alpha-hemolytic streptococci and ten with beta-hemolytic streptococci. Fifteen other plates were prepared using normal red blood cells plus irradiated plasma. Five of these were also inoculated with beta-hemolytic streptococci. The plates were incubated at 37°C for 48 hours and the results read. They are tabulated in the following table.

TABLE XXXIX

NORMAL RBC PLUS IRRADIATED PLASMA

Plate No.		-1	2	3	4	5	6	7	8	9	10
Alpha Strepto-cocci	Growth	++++	++++	++++	++++	++++					
	Hemoly-sis	++++	++++	++++	++++	++++					
Beta Strepto-cocci	Growth	++++	++++	++++	++++	++++	++++	++++	++++	++++	++++
	Hemoly-sis	++++	++++	++++	++++	++++	++++	++++	++++	++++	++++

TABLE XL

IRRADIATED RBC PLUS NORMAL PLASMA

Plate No.		1	2	3	4	5	6	7	8	9	10
Alpha Strepto-cocci	Growth	++++	++++	++++	++++	++++					
	Hemoly-sis	++++	++++	++++	++++	++++					
Beta Strepto-cocci	Growth	++++	++++	++++	++++	++++	++++	++++	++++	++++	++++
	Hemoly-sis	++++	++++	++++	++++	++++	++++	++++	++++	++++	++++

This experiment was repeated twice to be sure the results were reproducible; the same results were obtained each time. As can be seen from the tables, no evidence was obtained concerning which fraction of the blood contains the anti-hemolysis factor. When only a fraction of the whole blood used in the media had been irradiated, hemolysis occurred just as though the media was completely normal, whichever fraction had been irradiated. It might be concluded from this that hemolysis depends on factors present in both plasma and red blood cells, not on the latter alone, but since the mechanism of hemolysis (beta-hemolysis, especially) is not yet fully understood definite conclusions cannot be drawn from only one experiment. Therefore, the results of this experiment were considered essentially negative for the purpose intended.

g. Experiment 7. Blood media was prepared for the growth of beta-hemolytic streptococci in which 50 per cent of the blood added to the agar base was irradiated whole blood and 50 per cent was normal whole blood. The purpose of the experiment was to determine whether or not the loss of hemolyzing power of the organism is a function of the amount of irradiated blood present or whether it is complete when any irradiated blood is present. Ten plates were inoculated with the organism and incubated at 37°C for 48 hours.

When the plates were read, it was found that hemolysis was present and was about 50 per cent as great as that obtained on normal media. This would indicate that the effect of the irradiated blood is quantitative, the more irradiated blood being present the greater the loss of hemolyzing power, and that the presence of any normal blood will cause hemolysis to occur, its extent being dependent on the percentage of the normal blood present.

h. Experiment 8. The BHI agar base used for the preparation of the blood media was irradiated with Co^{60} (exposed to radiation for 23 hours; near the center of the 10-kilocurie source) to be sure that the agar base itself played no part in the loss of hemolyzing power of the organisms. This irradiated agar was then melted in the autoclave and cooled to 45°C, nonirradiated blood added to it, and twenty plates poured. Ten plates were also prepared using irradiated blood to serve as controls. Fifteen plates each (ten test plates, five control plates) were then inoculated with alpha- and beta-hemolytic streptococci, respectively. The plates were incubated at 37°C for 48 hours and results recorded. They are tabulated in Tables XLI and XLII.

The results of this experiment indicate that the agar base is not a significant factor in the loss of hemolyzing power by the streptococci.

An interesting sidelight on this experiment is that irradiation of the media affected the dextrans in the agar so that complete solidification never took place. This made inoculation of the media particularly hard, since just the slightest touch of a needle would cause a break in the surface.

TABLE XLI

ALPHA-HEMOLYTIC STREPTOCOCCI GROWN ON IRRADIATED BHI AGAR

Plate No.		1	2	3	4	5	6	7	8	9	10
Irradiated media plus normal blood	Growth	++++	++++	++++	++++	++++	++++	++++	++++	++++	++++
	Hemolysis	++++	++++	++++	++++	++++	++++	++++	++++	++++	++++
Irradiated media plus normal blood	Growth	++++	++++	++++	++++	++++					
	Hemolysis	++++*	++++	++++	++++	++++					

*Hemolysis was good but atypical. It produced a dark clouding of the surrounding media with a deep greenish cast to it rather than the usual bright green hemolysis with lightening of the media.

TABLE XXLII

BETA-HEMOLYTIC STREPTOCOCCI GROWN ON IRRADIATED BHI AGAR

Plate No.		1	2	3	4	5	6	7	8	9	10
Irradiated media plus normal blood	Growth	++++	++++	++++	++++	++++	++++	++++	++++	++++	++++
	Hemolysis	++++	++++	++++	++++	++++	++++	++++	++++	++++	++++
Irradiated media plus normal blood	Growth	++++	++++	++++	++++	++++					
	Hemolysis	-	-	-	-	-					

i. Experiment 9. An experiment was run doing serial passage of beta-hemolytic streptococci on irradiated blood media to determine whether or not the loss of hemolyzing ability would become a permanent thing. However, after twenty passages of the organism on this media (a passage made every 48 hours), it was found that the streptococci had not lost their ability to cause hemolysis on normal blood when transferred back to this media. On the contrary, the beta streptococci apparently increased their hemolyzing power on the irradiated blood because, by the end of the twenty passages, the streptococci showed slight hemolysis on the irradiated blood.

j. Experiment 10. A final experiment was run using chocolate agar as the media on which to grow beta-hemolytic streptococci to determine whether the loss of hemolyzing ability was purely a function of a physical change in the media or if it was due to some peculiar effects of radiation alone. Ten chocolate agar plates were inoculated with the organism and these plates incubated for 48 hours at 37°C. The results were read at that time and no hemolysis was found on the plates, indicating that the loss of hemolyzing power is not an effect peculiar to irradiated media alone but that any physical change in blood media may cause this to occur.

4. Conclusions

a. Radiation of media would not be suitable for sterilizing blood to be used in culture media, since changes are produced which alter the growth of certain organisms upon it:
(1) alpha hemolysis is delayed in its appearance, and
(2) beta hemolysis is completely inhibited (the growth of H. pertussis is unaffected by inoculation on irradiated media, however).

b. Radiation apparently changes the permeability of the cell wall of red blood cells, for after radiation they have lost their hemoglobin to the surrounding fluid, leaving an intact "ghost cell".

c. No toxic products are formed in blood by radiation which will prevent the growth of organisms cultured on it within 24 hours after its removal from the radiation source.

d. Loss of hemolyzing ability be beta streptococci is a function of the amount of irradiated blood present in the media, and this appears to be a quantitative relationship.

e. Loss of hemolysis by the beta streptococci is not a permanent change. The ability is immediately restored on thier return to normal blood agar media.

f. There appears to be some relationship between the changes produced by radiation and by heating of media to form chocolate agar media. This might suggest a possible use of radiation for sterilization purposes in the future.

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K. EFFECTS OF GAMMA RADIATION ON RUBBER AND ON POLYMERIZATION

Personnel:

L. M. Hobbs, Director of Phoenix project 43 and Associate Professor of Chemical Engineering; D. W. Pletcher, Research Assistant; and D. E. Brown, Graduate Fellow.

1. Introduction

Early this year a program of fundamental research on rubber and elastomers was initiated. The program is identified as Phoenix project 43. Emphasis in the work is directed toward gaining a better understanding of the molecular, structural, and physical properties of elastomeric materials. Funds for fellowships and equipment were provided by a gift from The Goodyear Tire and Rubber Company to the Michigan Memorial-Phoenix Project.

A portion of the research which has been started under Phoenix project 43 has comprised the examination of the effects on Co^{60} gamma radiation on:

(a) several rubber compositions,
(b) the polymerization of equimolar mixtures of styrene and methyl methacrylate, and
(c) the polymerization of styrene in the presence of various added agents.

The purpose of this section is to summarize the preliminary results of these three lines of experimentation.

2. Effect of Gamma Radiation on Rubber Compositions

Dr. Gehman of the Goodyear Company has kindly prepared the rubber compositions and has made the tensile measurements described in Table XLIII. The materials listed in Table XLIII were exposed to 10^8 rep of Co^{60} gamma radiation. The test pieces were packaged in polyethylene bags in an atmosphere of nitrogen, and the bags sealed with petroleum wax. During the course of the exposure the polyethylene developed cracks and holes, allowing the rubber samples to be exposed to an unknown amount of ozone. On completion of the exposure the measurements in Table XLIIIa were made and at the same time the exposure of a duplicate group of samples packaged under nitrogen in a vacuum desiccator was started. The exposure of the second group to 10^8 rep has been completed and the physical measurements are essentially complete. While the results of the second group will be given in detail in the next report, it can be stated that they confirm those recorded in Table XLIIIb, indicating that the effects of ozone on the first group were negligible.

TABLE XLIII

EFFECT OF GAMMA RADIATION ON RUBBER COMPOSITIONS
10^8 rep Co^{60} Gamma Radiation - Room Temperature

a. COMPOUND FORMULAS

	1	2	3	4	5	6	7
GR-1 18, Butyl	100						
GR-S 100, cold rubber			100	100		100	
Neoprene GN		100					
Hevea, smoked sheet					100		100
Sulfur	3			2	3	2	2.75
Zinc Oxide	5	5	3.5	3.5	5	3.5	5
Stearic Acid	3	1	1.5	1.5	1.5	1.5	3
Pine Tar							5
Magnesia		4					
Captax					1		
MBT Disulfide				1		1.5	.75
Tuads	1		4				2
HAF carbon black					50	50	

b. STRESS-STRAIN PROPERTIES

Compound No.	Cure	100% Modulus Original	100% Modulus Exposed	100% Modulus % Change	Tensile Strength Original	Tensile Strength Exposed	Tensile Strength % Change	Ultimate Elongation Original	Ultimate Elongation Exposed	Ultimate Elongation % Change
1	30/290		Good							
	50/290									
2	30/290	141	-		4300	384	-91	1016	60	-94
	50/290	156	-		4520	364	-92	1000	70	-93
3	30/290	123	171	+ 39	278	216	-22	320	170	-47
	60/290	125	175	+ 40	246	238	- 3	285	170	-40
4	30/290	44	139	+216	515	215	-58	1350	225	-83
	60/290	89	189	+112	190	352	+86	465	245	-47
5	20/290	356	686	+ 93	2480	2060	-17	370	235	-37
6	40/290	304	975	+221	3290	3420	+ 4	520	250	-52
7	10/311	67	135	+101	1750	2080	+19	660	590	-11
	20/311	67	130	+ 94	1650	1820	+10	700	570	-19
	30/311	95	131	+ 38	1700	1500	-12	740	540	-27
a 8 Milled smoked sheet		21	86	+310	23	152	+560	120	240	+100
a 9 GR-S 100, milled		18	-		14	87	+520	735	67	-91
10 Polyethylene		2200	-		2300	1080	-53	115	6	-95

Examination of the data in Table XLIIIb, indicates the following:
(a) serious deterioration of various rubber compositions takes place on exposure to 10^8 rep of Co^{60} gamma radiation at room temperature;
(b) natural and cold GR-S compositions have about the same resistance, with the natural rubber possibly being a little more resistant;

(c) the nature of the compounding agents and possibly the degree of cure influence the resistance on rubber compositions to gamma radiation;
(d) the resistance of Neoprene compositions to gamma radiation is less than that of natural and cold GR-S compounds;
(e) butyl rubber has very low resistance to gamma radiation;
(f) gamma radiation causes strengthening and a large increase in the gel content of smoked sheet and raw cold GR-S; and
(g) 10^8 rep of gamma radiation causes serious deterioration of polyethylene sheet.

3. Polymerization of Equimolar Mixtures of Styrene and Methyl Methacrylate

Equimolar mixtures of styrene and methyl methacrylate were prepared from freshly fractionated monomers. The mixtures were placed in pyrex tubes and the catalyst added under an atmosphere of oxygen-free nitrogen. The tubes were evacuated, degassed, and sealed at a pressure of about 0.1 mm Hg. After the polymerizations, indicated in Table XLIV, each mixture was diluted with benzene and the solid polymer precipitated by the addition of methyl alcohol.

TABLE XLIV

POLYMERIZATION OF EQUIMOLAR MIXTURES OF STYRENE AND METHYL METHACRYLATE, 25-30°C

	Catalyst mol %	Wt. % Yield Solid Polymer	Mol % Styrene in Solid Polymer[a]
Na	5.6	30	3
$SnCl_4$	2.0	41	99
Benzoyl Peroxide	0.11	3	51
Gamma (10^7 rep)		8	51

a Determined by C-analysis

Solution in benzene and precipitation with methanol was repeated and finally the solid polymer was dissolved in benzene. The solution was frozen and the benzene removed by sublimation at reduced pressure. The polymer was dried to constant weight in a vacuum oven at about 1 mm Hg and 50°C. The styrene contents of the polymers were determined by carbon analysis; the latter were made by the Clark Microanalytical Laboratory of Urbana, Illinois.

The scheme represented by the data in Table XLIV was developed by Walling, Mayo and co-workers (J.A.C.S. 72, 48, 1950) and can be used to throw some light on the possible mechanism of a given polymerization. The compositions of the products of initial polymerization are quite different for the three types of reactions involved:

Catalyst	Composition of Initial Copolymer	Type of Reaction
Na	mainly methyl methacrylate	carbanionic
$SnCl_4$	mainly styrene	carbonium
Benzoyl peroxide	equimolar mixture	free radical

It is apparent that the composition of the gamma-catalyzed product is identical with that formed in the presence of benzoyl peroxide and quite different from that produced in the presence of sodium or stannic chloride.

At this point it must be concluded that the gamma-catalyzed process bears a resemblence to free-radical-catalyzed reactions. This view is strengthened, as pointed out recently by Dr. Gehman, by the appearance of butyl rubber after gamma radiation, which is similar to that of the product formed by milling butyl rubber with benzoyl peroxide. If a free-radical process is involved, then some of the common antioxidants would be expected to retard the gamma-catalyzed polymerization.

4. Effect of Agents on Gamma Catalyzed Polymerization of Styrene

Various agents in catalytic amounts were added to samples of freshly fractionated styrene in pyrex tubes under atmospheres of oxygen-free nitrogen. The tubes were evacuated, degassed, and sealed as described above and exposed to about 10^7 rep of gamma radiation in two groups; those of Table XLVa were exposed at about 85°F and those of Table XLVb were exposed at about 95°F. The solid polymers were isolated and dried to constant weight by the methods described above.

The fact that the blanks in Tables XLVa and XLVb did not polymerize to the same degree on being exposed to equal amounts of radiation is of concern. In fact, the degree of polymerization of all the samples of Table XLVb seems to be uniformly higher than those in Table XLVa. This point will be examined in further experiments. Nevertheless, it appears that some of the common inhibitors (which are also rubber antioxidants) and some vulcanization accelerators exhibit a moderate promoting action, and that sulfur, dinitrobenzene, a secondary disulfide, and a few other materials show a retarding effect on the gamma-catalyzed polymerization.

5. Conclusions

Serious deterioration of several rubber compositions takes place on exposure to some 10^8 rep of Co^{60} gamma radiation at room temperature. The

TABLE XLV

EFFECT OF AGENTS ON GAMMA CATALYZED POLYMERIZATION OF STYRENE

a. 1.16 x 10^7 rep; Average temperature, 20-25°C

Added Agent	Wt.%	Mol %	% Yield of Solid Polymer
Hydroquinone	0.12	0.08	28
2,5-di t Bu"	0.11	0.05	28
C_6H_5-NH-$C_{10}H_7$	4.9	1.7	25
Copper Powd.	0.14	0.09	24
NONE			22
Sulfur	0.07	0.08	12
Dinitrobenzene	0.19	0.08	3

b. 1.16 x 10^7 rep; Average temperature, 25-30°C

Added Agent	Wt.%	Mol %	% Yield of Solid Polymer
Mercaptobenzotiazole (Captax)	0.17	0.10	46
Benzothiazyl disulfide (Altax)	0.17	0.10	43
Titramethylthiuram disulfide (Methyl Tuads)	0.22	0.11	39
NONE			36
p-t Bu catechol	0.17	0.10	34
Organic sulfonate (Daxad 11)	0.19		31
t-Dodceyl mercaptan (Sulfole)	0.24	0.12	31
Zinc stearate	0.63	0.10	27
1-Methyl heptyl disulfide	0.32	0.11	26

degradation usually comprises hardening, stiffening, and embrittlement; however, butyl rubber decomposes to a sticky mass long before the exposure level reaches 10^8 rep. Natural rubber and cold GR-S formulations appear to have about the same resistance to gamma radiation and both are more resistant than Neoprene compounds.

It appears that the nature of the polymer, the action of compounding agents, and possibly the degree and nature of the cure effect the resistance of a given rubber formulation to gamma radiation.

The gamma-catalyzed polymerization of a mixture of styrene and methyl methacrylate gives a copolymer of composition identical to that formed in the presence of benzoyl peroxide.

Several compounds belonging to the classes of rubber antioxidants and vulcanization accelerators exhibit a moderate promoting effect on the gamma-catalyzed polymerization of styrene, whereas dinitrobenzene, sulfur, and a few other materials show a retarding effect on that polymerization.

It must be emphasized that these results are of a preliminary nature. In extending these experiments, special attention will be given to the effect of compounding agents on the resistance of rubber compositions and to the possibility that there may be more than one basic type of reaction involved in what is now referred to as a "free-radical reaction in solution".

IV. SUBPROJECT M943-7, OPERATION OF THE FISSION PRODUCTS LABORATORY

Personnel:

Advisors: L. E. Brownell, Associate Professor of Chemical and Metallurgical Engineering; H. J. Gomberg, Associate Professor of Electrical Engineering; W. W. Meinke, Assistant Professor of Chemistry; L. Thomassen, Professor of Chemical and Metallurgical Engineering; J. V. Nehemias, Health Physicist; D. E. Harmer, Chemist; E. W. Coleman, Research Assistant; and E. Ambo Research Assistant.

A. INSTALLATION OF 10-KILOCURIE COBALT-60 SOURCE

1. Shipment of Source and Transfer to Well

The source material arrived at the laboratory by University truck after an uneventful trip from Chalk River. The 3-ton shipping container was then lifted from the truck, dollied into the radiation cave, and set at position A, between the storage well and the barrier wall as shown in the plan view of the cave room (Fig. 92). The unloading operation was performed remotely, with the operator standing at position B, behind the barrier wall. Visibility was maintained through a double-mirror system.

The original plan involved placing the source rods in an aluminum bucket inside the shipping container with a wire loop from the aluminum bucket extending outside. In the transfer at the University it was planned to fasten the chain hoist to the 300-pound cover of the shipping container, and to fasten the rope pulley hoist to the bucket of source rods, without removing the shipping container cover. The cover could then have been removed and the rod bucket lifted out and lowered into the well, all by remote control from behind the barrier wall, in a matter of a few minutes. Close machining of the shipping container, however, precluded the use of the external wire loop. Instead a steel chain was attached to the bucket and after loading was allowed to fall among the source rods. Hence, when the actual transfer took place, the heavy shipping container cover was lifted off remotely as scheduled but it was then necessary to "fish" for the chain attached to the bucket.

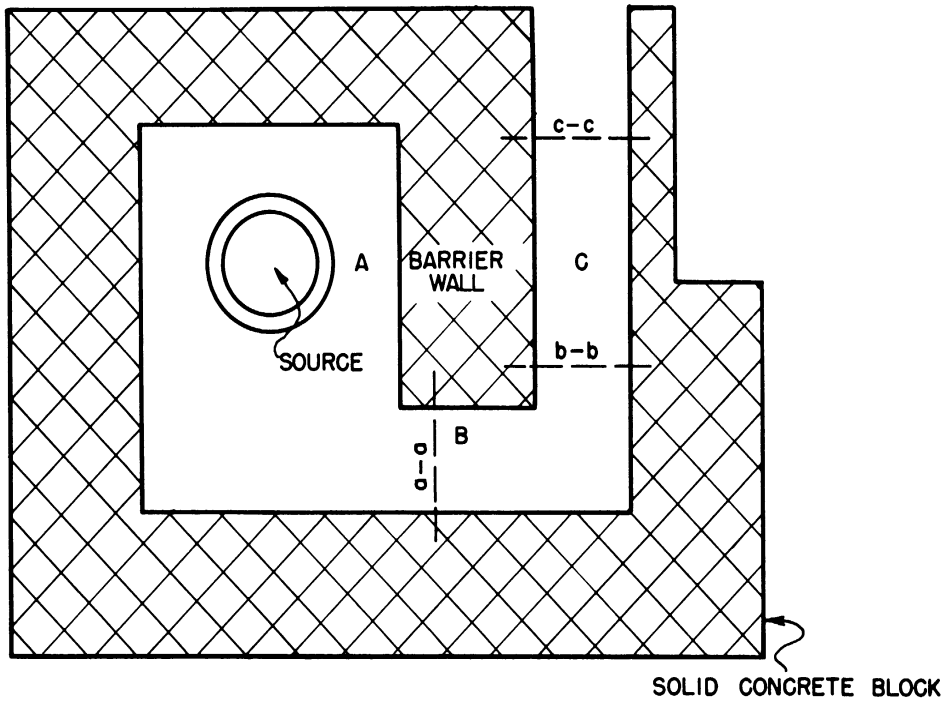

Fig. 92. Plan View of Radiation Cave.

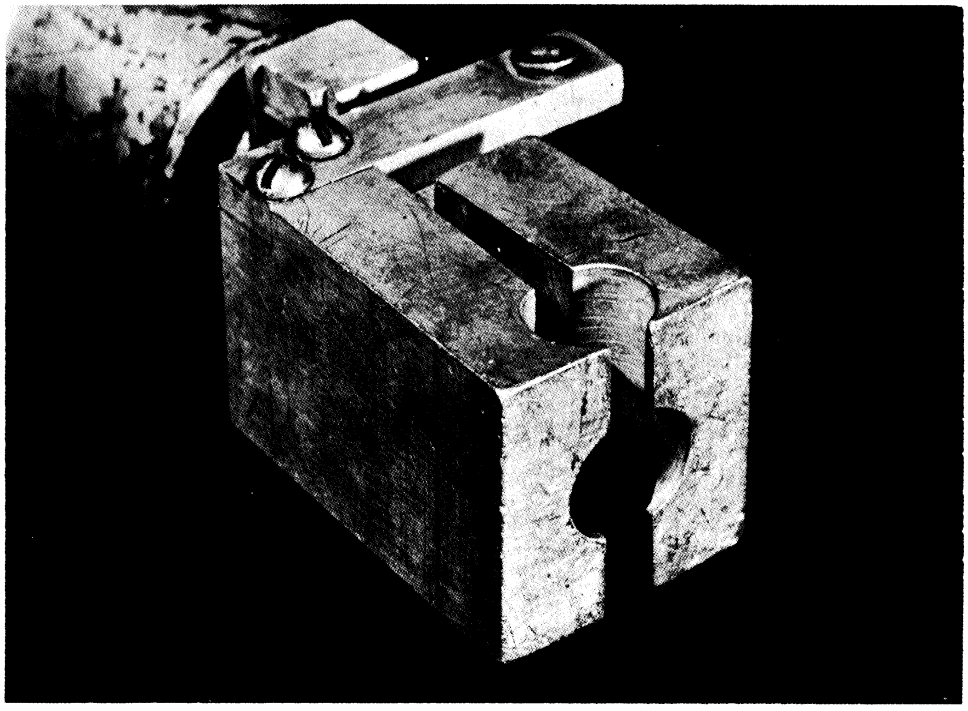

Fig. 93. Jaws of 18-Foot Tongs for Handling Cobalt Rods. A 5183

When the shipping container cover had been removed, the dosage rate at position C rose to 20 mr/hr. At position B, protected by some concrete, the dosage rate was 400 mr/hr. It was therefore feasible for a man at B to operate offset tools and "fish" for the steel chain in the bucket using a mirror system to see inside the shipping container. After trying several methods the chain was finally raised with a three-pronged gang fishhook and placed over the hook of the rope pulley hoist. Once engaged by the hoist, the source bucket was lifted from the shipping container, rolled into position over the well, and lowered into the water without incident in a matter of seconds.

The entire transfer operation was completed in about 3 hours with no personal whole-body exposures in excess of 180 mr and no hand exposures in excess of 240 mr. These exposures were determined both with film badges and rings, and with pocket and finger ionization chambers.

2. Loading of Source Rods into Holder

When the bucket of source rods had been lowered into the well, it was safe for personnel to enter the cave room. The rods were poured from the bucket onto the bottom of the well and scattered loosely over the floor of the well. From there the rods were picked up individually and set in place in the rack; 100 rods, 10 inches long and less than 1/2 inch in diameter had to be picked up from a horizontal position under 16 feet of water, turned to a vertical position, and inserted into closely machined holes in the rack. This tedious operation was completed in two days, using the specially designed 18-foot tongs, the jaws of which are illustrated in Fig. 93. The rods were first picked up by the tongs in the horizontal grip position and allowed to fall into a metal beaker on the bottom of the well, to bring them into a position near the vertical. They were then taken from the beaker by the tongs in the vertical grip position and inserted into place in the rack. This operation was also completed according to plan without incident. Figure 94a shows the 18-foot tongs in use during the loading procedure. The stream of water shown in the figure is coming from a small filter through which the water in the well was circulated to maintain clarity. Figure 94b shows the source in the light of the Cerenkov glow under water.

B. SHIELDING PROBLEMS AND RADIATION LEVELS

The source is lowered into the water, which makes it possible for workers to enter the cave without hazard. Calculations reported in Progress Report 4 were made to determine the well depth required. Calculated and experimental values of dosage rate at the surface of the water are plotted as a function of shielding water thickness in Fig. 95. On the basis of these

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Fig. 94a. Loading Cobalt Rods Into Holder in 14 Feet of Water Using Submarine Light and 18 Foot Tongs.

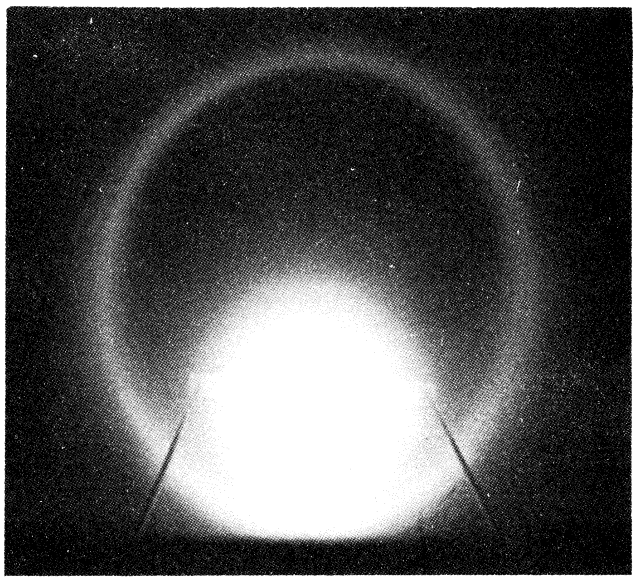

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Fig. 94b. Cerenkov Glow of 10-kc Source Under Water.

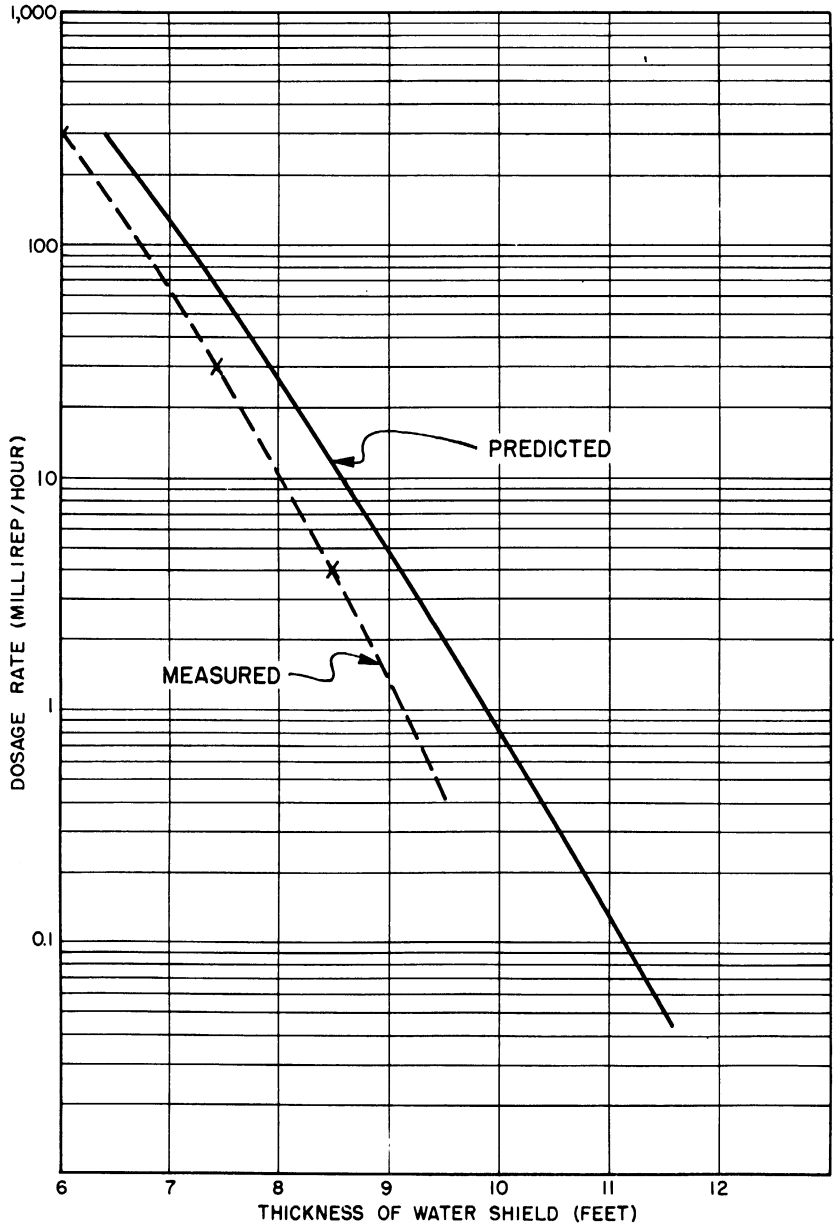

Fig. 95. Predicted and Measured Radiation Dosage as a Function of Shielding Water Thickness.

calculations, a shielding thickness of 11 feet was considered sufficient. In present practice the source is lowered approximately 8-1/2 feet; dosage rate at the surface of the water is 4 mr/hr.

The predicted values are appreciably higher than the measured values. The explanation for this discrepancy appears to lie in the reactor computations, as discussed in Part II of this report.

C. OVERHEAD SHIELDING REQUIREMENT

To facilitate the transfer and loading operations, no overhead shielding was incorporated in the original cave structure. When ten source rods had been loaded into the rack, the source assembly was raised into its operating position in the cave and the dosage rates in the surrounding areas determined. Shielding was then added over the cave until the dosage rates were satisfactorily reduced. Points of particular interest in this radiation survey are shown in Fig. 96, while the dosage rates at these points for several shielding configurations are enumerated in Table XLVI.

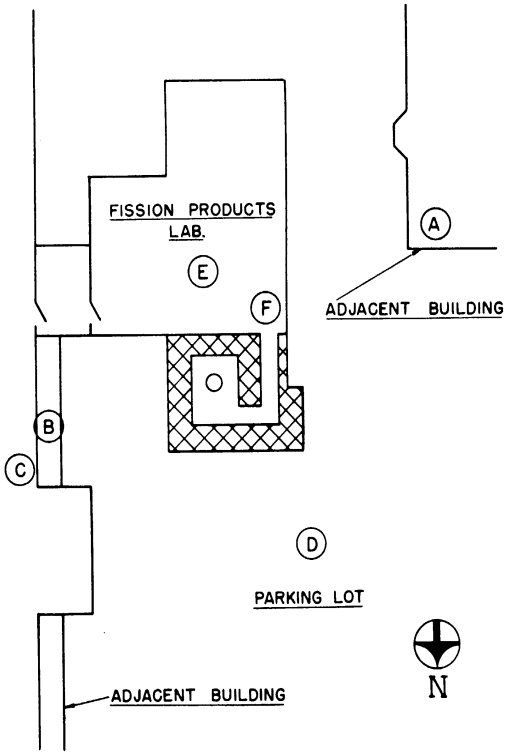

Fig. 96. Plan View Showing Points of Interest in Radiation Survey.

Plan location A indicates the corner rooms of an adjacent building; A-1 refers to readings at the ground floor; A-2, second floor; A-3, third floor; A-4, fourth floor; and A-5, the roof. Location B indicates the position of highest dosage rate on a third-floor fire escape on the opposite adjoining building. Location C indicates the room in this building in which the highest dosage rate was found. Location D indicates the position of maximum dosage rate immediately outside the concrete cave. Location E indicates the window area on the second floor of the Fission Products Laboratory and location F indicates the entrance to the cave on the first floor of the Fission Products Laboratory. In each case, the dosage rates given are the highest that could be found in the area in question.

As the successful prediction of the magnitude of scattered radiation levels is somewhat less than an exact science, it is felt that these data may be of general interest. The reduction of radiation levels at all points outside the laboratory to tolerable levels, permitting early routine operation of the radiation facility, was the prime consideration during the installation of overhead shielding. No time, therefore, was taken for detailed studies of the spectrum and distribution of the scattered radiation field for configuration I. Readings for Configuration I through VII were taken with a radium-calibrated Juno portable ionization-chamber instrument and for configuration VIII with a radium-calibrated portable geiger-tube instrument. An analysis of these data is in progress and will be reported later.

TABLE XLVI

EFFECT OF OVERHEAD SHIELDING ON DOSAGE RATE IN SURROUNDING AREA (All figures in mr/hr)

Position	I	II	III	IV	V	VI	VII	VIII
A-1	2	1	-	-	Bkgd	Bkgd	Bkgd	0.18
A-2	6	2	-	-	1	1	1	0.60
A-3	8	4	-	-	1	5	2	1.0
A-4	13	6	-	-	2	10	4	1.6
A-5	22	-	-	-	3	16	5	0.3
B	70	70	45	28	3	7	-	3.0
C	50	-	-	-	3	5	-	2.0
D	9	6	-	-	1	1	1	1.0
E	400	320	300	160	60	160	60	30
F	20	20	20	20	20	100	100	100

I. No overhead shielding, only plywood and paper roof; ten rods up.
II. Two courses (approximately 8 inches) of concrete over cave up to line a-a, with exception of 1 by 3 foot hole over source for source lift mechanism; ten rods up.
III. Same as II with hole closed with 4 inches of concrete.
IV. 12 inches of concrete over cave out to line b-b, 8 inches over remainder to a-a; ten rods up.
V. 20 inches of concrete over cave out to line a-a; ten rods up.
VI. 24 inches of concrete over cave out to line a-a; one hundred rods up.
VII. Same as VI plus 1/4-inch lead sheet over passage to line c-c; one hundred rods up.
VIII. Same as VI plus 8 inches of concrete over passage to line c-c; one hundred rods up. (Refer to Fig. 92 for a-a, b-b, and c-c lines.)

The addition of further shielding has since reduced radiation levels in nearby occupied buildings to the point that 30 mr/week is the maximum obtainable dose.

D. CONTROL OF CORROSION OF THE RODS

Corrosion of the source rods in water has been an important consideration at this installation. This problem is somewhat similar to that of aluminum-jacketed uranium fuel rods in a water-moderated reactor.[4] A lower gamma-radiation flux is encountered in the case of the cobalt source, but the rods must be protected for several years, while fuel rods are in use for a much shorter time.

To control the spread of contamination, cobalt sources are generally encased in aluminum or gold. The cobalt rods of this source were encased in 3S-H18 aluminum pipe with an analysis of 98.8% aluminum and 1.2% manganese. The ends of the pipe were welded shut with Alcan 2S welding rod (commercially pure Al), machined clean, and tested for leaks by immersion in hot water. Absence of detectable cobalt-60 contamination in the water of the well after 3 months of operation indicates that the welds were satisfactory.

The walls of the well are lined with an hydraulic cement which presents a problem in pH control. This lining contains, among other constitutents, certain alkaline earth silicates which have been subject to leaching by the well water, which brings hydrolysis products into the well water. These products are of such alkaline nature that the pH of the unbuffered water rises very rapidly above the neutral point. The use of phosphate buffers in any appreciable concentration to control pH results in a precipitate with the alkaline earth ions, and visibility is then reduced to a point where underwater inspection of the source is difficult. Attempts to maintain a neutral solution through continuous circulation of the water through a deionizer have not been practical with the small deionizer available in this laboratory (see Fig. 97).

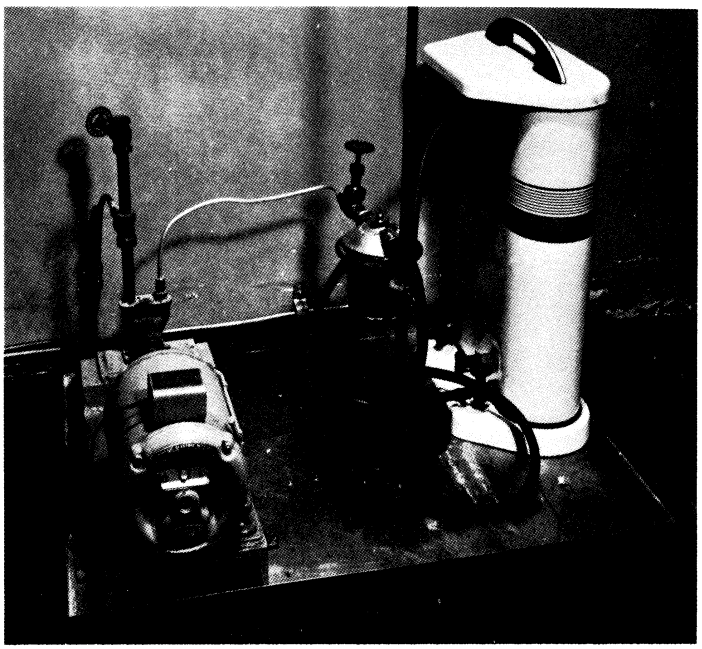

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Fig. 97. Ion Exchanger Used to Treat Water in Well for 10,000-Curie Source.

Successful control of pH with excellent visibility has been attained through the use of extremely dilute phosphate buffers (of the order of 15 ppm) and a small cation exchange unit operated intermittently. As the alkaline materials leach from the walls, the pH gradually increases. When a pH of about

7.5 is reached, the cation exchanger is connected to a circulating pump system, and the excess cations removed until a pH of about 6.3 is reached. It should be noted that the ion exchange resin employed removes all metallic ions and exchanges them for hydrogen ions.

Corrosion studies on aluminum conducted at the Oak Ridge National Laboratories in connection with the aluminum-encased uranium fuel rods show that sodium chromate added to the water to the extent of 60 ppm gives effective inhibition of corrosion to these aluminum casings.[4] To take advantage of this inhibiting action, a sodium chromate - sodium dichromate buffer system is now used in a manner exactly analogous to the previously described phosphate system using the cation exchanger. With this system the required water visibility, pH, and inhibitor action are attained.

A check for corrosion of the aluminum jackets was made by a qualitative analysis of the salts leached out of the cation exchanger by the acid regeneration. The ions of calcium, magnesium, sodium, and chromium were identified. It is interesting to note that aluminum ions could not be found. Thus it appears that the aluminum jackets of the cobalt rods and the aluminum rack are not suffering corrosion to any great extent. It is probable that the calcium, magnesium, and sodium ions are those leached from the porcelain lining of the well, while the trivalent chromium arises from reduction of the chromate inhibitor by foreign matter in the well water or by radiation effects.

E. FAILURE OF PLASTIC PARTS

The original design of the source installation included a plastic cap into which the source was raised for irradiation experiments, and a plastic rack to hold the 100 component rods of the source. The cap was constructed of Plexiglas plastic. Within a few weeks after the source was put into use, the cap developed innumerable fine cracks. It became very brittle and failed by crumbling into small pieces under its own weight. Figure 98 is a photograph of one of the fragments showing the numerous cracks which caused failure. The cap was then replaced with one constructed of galvanized wire mesh, shown previously in Fig. 90, which has proven satisfactory.

The plastic rack which originally supported the cobalt rods was constructed of cloth-laminated phenolic-type resin, with tie-rod spacers of Lucite. After about 4 months of usage, the tie-rod spacers failed as a result of severe decomposition and the holder was replaced with a rack of aluminum. At the time of replacement the parts of cloth-laminated phenolic resin had experienced an appreciable loss in strength. This new rack is fabricated from 2-S alloy, which is similar to the 3-S alloy, and little difficulty from electrolytic corrosion is anticipated. The rack is separated from the steel elevator platform by a replaceable plastic disk to avoid electrical coupling with the steel. Provision

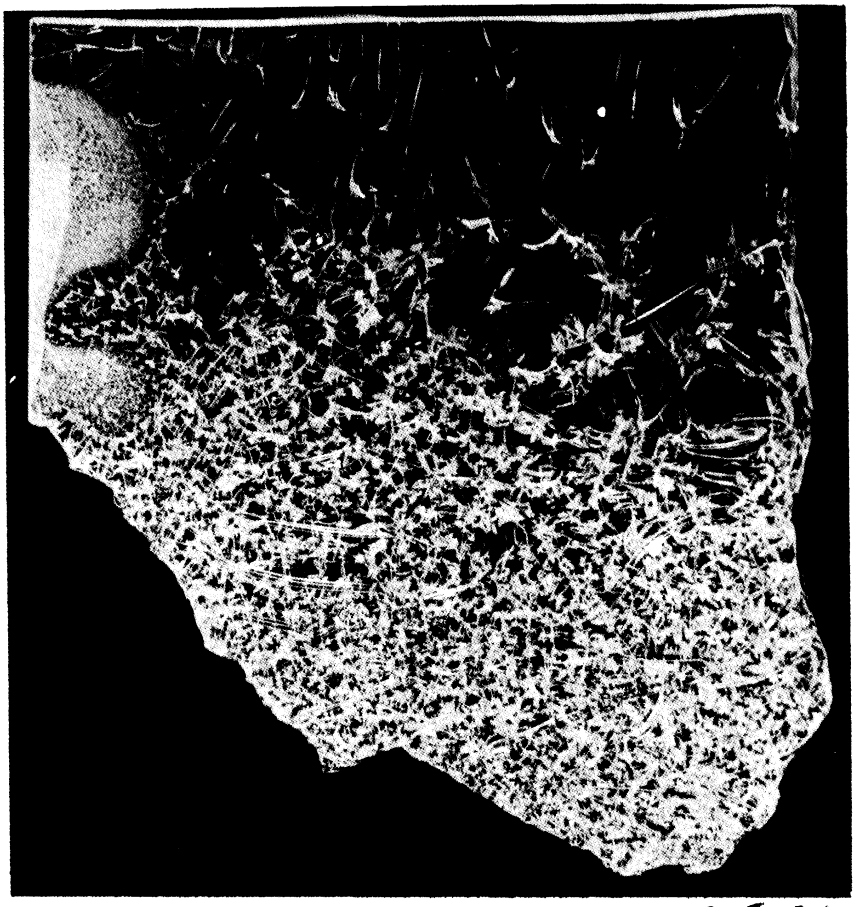


Fig. 98. Fragment of Lucite Cap which Failed After Exposure to Gamma Radiation.

has been made to inspect this plastic disc periodically to insure that it has not disintegrated.

F. DOSIMETRY MEASUREMENTS

After the source rods were properly placed in the support rack and were ready for routine operation, a dosimetry program was undertaken. Dosage rates were determined as a function of position throughout the cave by several methods.

1. Measurements with Ferrous Sulfate

Dosimetry measurements with ferrous sulfate solutions were used as the principal method of calibration. Most of the analytical work was performed by the personnel of the group studying chemical reactions (M943-4). The description of the ferrous sulfate tests has been included in Part II of this report. Figures 99 and 100 show the results of ferrous sulfate dosimetry measurements inside and outside the source, respectively.

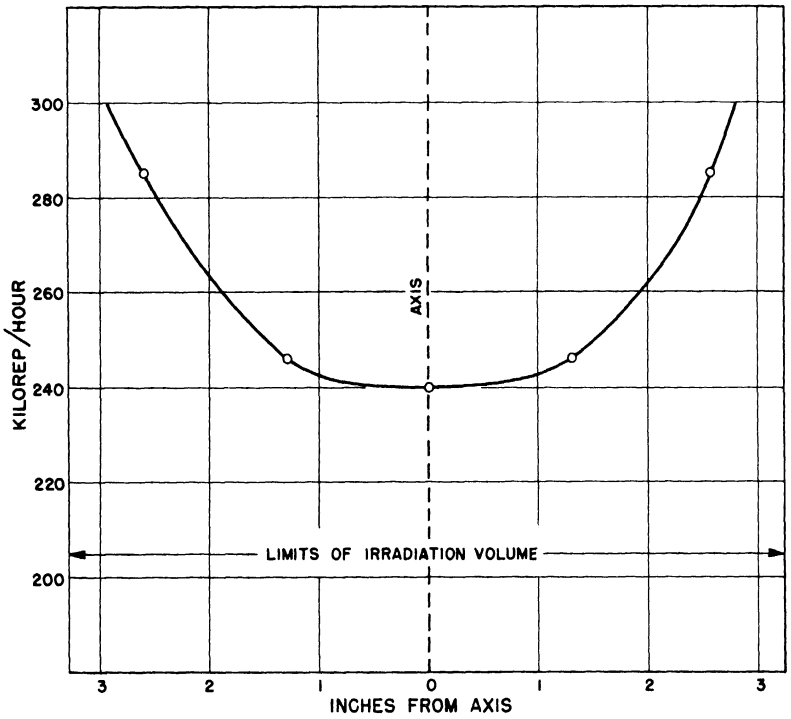


Fig. 99. Gamma Flux Inside Cylinder of 10-Kilocurie Cobalt-60 Source.

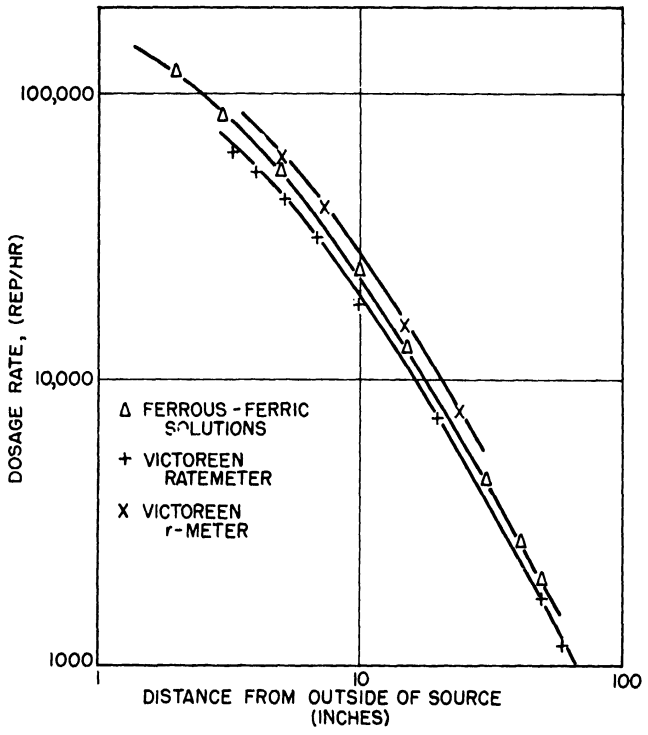


Fig. 100. Gamma Flux Outside Cylinder of 10-Kilocurie Cobalt-60 Source.

2. Measurements with Polyvinyl-Chloride Film

a. Introduction. The study of polyvinyl chloride films which follows is from a report by Mr. L. M. Welshans which was submitted in partial fulfillment of the requirements for the degree of M.S. in Nuclear Engineering.

The purpose of this investigation was to check the use of polyvinyl chloride films as a gamma radiation dosimeter and, with their use, provide an estimate of the strength of the 1- and 10-kilocurie cobalt-60 sources. The possibility of increasing the sensitivity of the films and their use in x-ray dosimetry measurements was also investigated in a limited way.

The application of polyvinyl chloride films to gamma radiation dosimetry measurements was suggested by Steigman[1] and investigated in greater detail by Henley and Miller.[2] The reaction is thought to take place by the action of gamma radiation on the plastic to liberate HCl within the film. If an acid-alkali indicator has been incorporated into the film, a color change will occur under the influence of the HCl.

Henley and Miller give the following four requirements that a system must meet for acceptance as a dosimeter method. They are:

1. The system must vary in an exact manner and be independent of radiation intensity, wavelength, temperature, pH, etc.
2. The reaction must be irreversible.
3. The results must be reproducible within a few per cent.
4. The chemical system must be stable under ordinary storage conditions.

In general, the main investigation will be to determine if polyvinyl chloride films meet these requirements or, if they don't, what limitations must be placed on their use as a dosimetry method.

b. Experimental Procedure. (1) Preparation of Polyvinyl Chloride Films: the first method used to prepare the films was that described by Henley and Miller.[2] Briefly, their procedure involved the addition of powdered resin to a filtered solution of dye in chlorobenzene and heating the mixture for 1 hour at 110°C. The solution is then poured on level glass plates and set aside until dry.

The films prepared in this manner were specked with dye and undissolved plastic. These difficulties were overcome by diluting the dye solution with solvent and by filtering the solution before the films were cast. However, the surface of the films then had a mottled appearance, resembling fish scales, which is characteristic of a film which dries on the surface faster than the solvent diffuses through the main body of the plastic. The appearance could

be improved by varying the drying time, solvent concentration, and casting temperature, but none of these methods would eliminate the honeycombed appearance.

It was found that films cast from cyclohexanone were clear and free from surface imperfections, with the additional advantage that stock solutions of uniform-quality plastic can be prepared and cast at room temperature. However, the drying time was increased to 9-10 days instead of 1-3 days from chlorobenzene. Hence, in practice about 40% chlorobenzene was added to the solvent to decrease the drying period without affecting the appearance of the film.

A typical stock solution has the composition:

200 cc chlorobenzene (tech)
300 cc cyclohexanone (Eastman)
0.11 g methyl violet 6B (General Aniline)
23.7 g polyvinyl chloride resin (Geon 101)

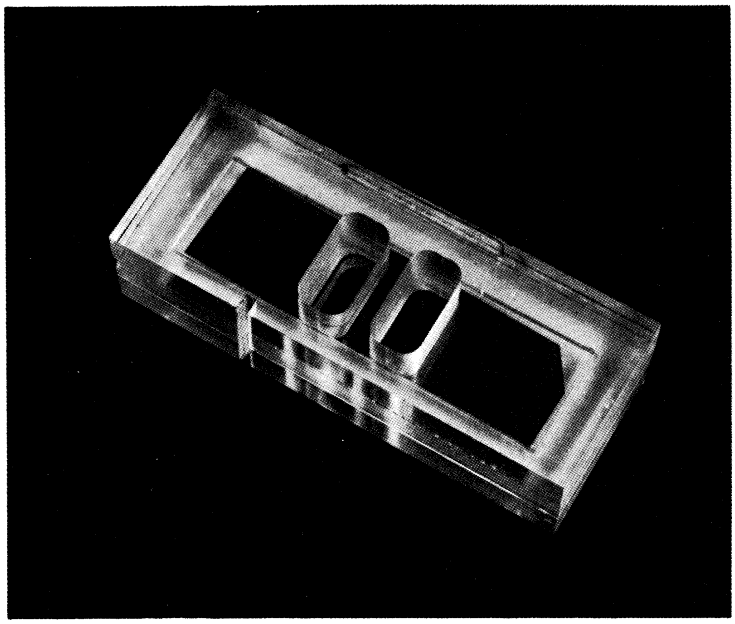
The dye is dissolved in the mixed solvents and resin added slowly to avoid lumps. The mixture is heated to 115°C for 1 hour, cooled, and filtered through cotton into a dark-brown bottle. If 76.5 cc of this solution are poured on a 19- by 29-cm glass plate, the dried film is from 0.0016 to 0.0018 inch thick.

These films were cast on an 8- by 12-inch glass plate which had a wall of sauereisen cement around the edges to act as a dam for the solution. The plate was floated in a pool of mercury contained in a large pyrex dish, as shown in Fig. 101. In this way the surface of the plate could be kept perfectly level.

The plate containing the dried film is placed under water and an edge of the film lifted with a sharp knife. Then the film can be peeled off in one piece without tearing. It is finally dried with blotting paper and cut into the proper shape with a razor blade.

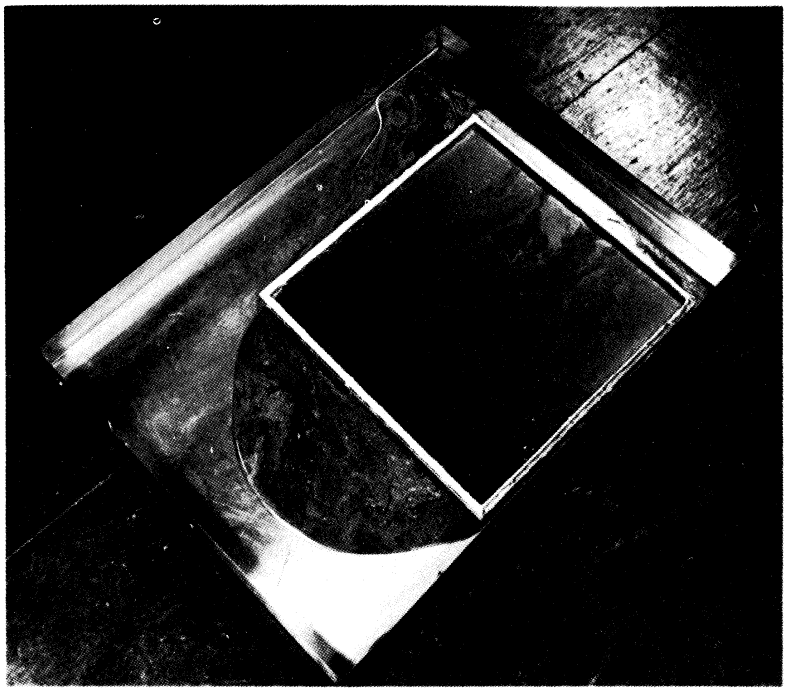
The stability of the film was demonstrated by boiling it with water for several minutes. The appearance and shape of the film remained the same after the test and the water remained colorless, although the dye is normally water-soluble.

(2) Film Mounts: It is necessary to provide some type of support to hold the films during exposure or reading in the spectrophotometer. Two methods were used in these experiments. The first method consisted of the insertion of the film in a 35-mm Kodak ready-mount. These were fastened with scotch tape



A 5187

Fig. 101. Leveling Glass Plate for Film Solution by Use of Mercury.



A 5188

Fig. 102. Lucite Holder and Polyvinyl Chloride Film Used in Obtaining Transmission Data.

to a container or cardboard support during exposure. It was necessary to insert the Kodak ready-mounts from the side. The thin cross section of these mounts resulted in some difficulty in keeping the films flat and free from buckles and bends. Another type of mount was tried in which the films were laid flatly inside a recess in the 2- by 2-inch frame, and held in place with a retainer frame which snaps into the recess, tightly holding the film on all four sides. With this frame wrinkles and folds were eliminated. This method results in easy handling and filling of the films, but the size and shape of the mount somewhat limits its use. For example, the mount will not fit inside the aluminum sample holder used with the 1-kc cobalt-60 source. Furthermore, it is rather difficult to insert the thin films in the ready-mount. The films were placed in a Lucite holder for reading the transmission data. A view of the film and holder is shown in Fig. 102.

The second mounting method consisted of cutting the films into 1- by 3-inch strips. These strips were inserted into a 1-inch-diameter test tube and exposed in this manner. The method has several advantages: first, the films could be suspended in either cobalt-60 source, and second, many of the experiments involving chemical reactions were performed in glass equipment of similar dimensions. A special holder was devised to permit the reading of the per cent transmission in the spectrophotometer.

(3) Dosimeter Studies with the Film: The films were placed in the proper holder and transmission readings taken before and after exposure to gamma radiation with a Beckman Model DU spectrophotometer kept at a wavelength of 600 mu and a slit width of 0.80 mm. The tungsten lamp was used in all measurements with the Beckman filter in the "in" position and the phototube in the "out" position. The sensitivity of the instrument was adjusted to give 100% transmission in air without any film in the holder. It was possible to make readings on the same sample that agreed within 1%.

(4) Exposure Procedure: It was decided that all measurements would be taken with the 1- by 3-inch film strips placed inside the test tube. The films were centered vertically in the tube so that they would lie in a plane through the center of the source. The test tube was then placed in the aluminum holder, for exposures in the 1-kilocurie source, or suspended in the center of the 10-kilocurie source from a ring stand. The exposure time varied from 4 to 48 hours, depending on the source used and the desired range of transmission readings.

c. Experimental Results. (1) Tests with Cobalt-60 Source: The results of the dosimeter experiments in the cobalt-60 sources are tabulated in Tables XLVII and XLVIII. Sample calculations of the data are given on page (184). If the reciprocal of the exposure time is plotted versus the per cent transmission, a value of the transmission, T_{inf}, is obtained when the curve is

extrapolated to infinite time. This value is necessary for quantitative interpretation of the data. This plot was constructed, Fig. 103, for the data from both the 1- and the 10-kilocurie cobalt-60 source. The two curves have different shapes because the abscissa should be plotted in terms of dosage instead of exposure time. If the correct source strengths were known and plotted properly, the two curves would be identical.

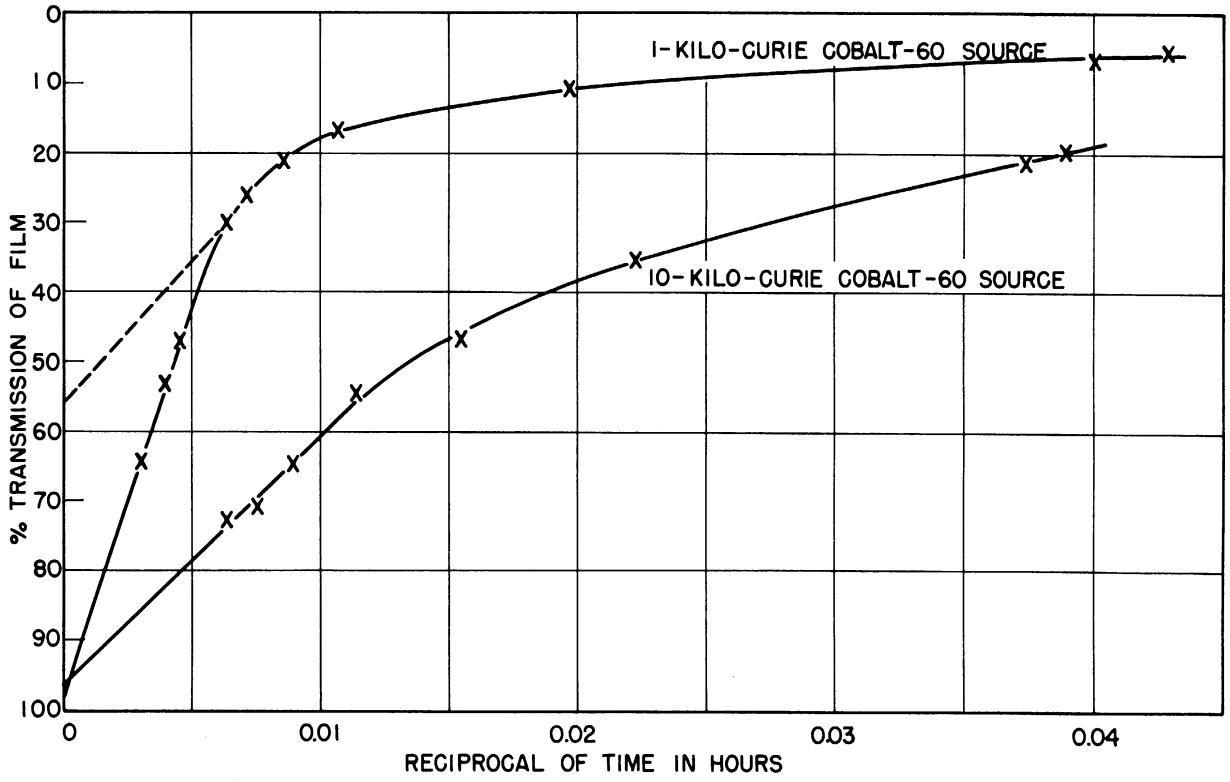


Fig. 103. Percentage Transmission of Film vs Reciprocal of Exposure Time, Hours.

The values of T_{inf} obtained check very closely and within the error of the extrapolation. This would be expected, since the film strips used in all the experiments were cut from the same sheet of plastic. In general, T_{inf} is a function of the dye concentration of the film and would be determined for each batch of film.

The values of T_{inf} do not agree with those obtained by Henley and Miller[2] because they chose to extrapolate the curve at a higher value, as indicated by the dotted line. This procedure is followed because the film base deteriorates at high dosages and the first-order relation between the percentage of dye bleached and the exposure is no longer maintained. Calculations indicate that better agreement is obtained with a value of T_{inf} extrapolated according to the solid line rather than using the procedure followed by Henley and Miller.

TABLE XLVII

EXPOSURE DATA ON POLYVINYL CHLORIDE FILM
RADIATION SOURCE: 1-KILOCURIE COBALT-60

Film No.	Transmission, %	Exposure Time, hr	Reciprocal of Exposure	Dye Retained %
5	4.12	0	infinity	100.00
5	6.78	24.95	0.040	84.22
5	11.00	50.78	0.0197	69.00
5	16.7	93.38	0.0107	55.80
6	4.26	0	infinity	100.00
6	6.57	23.35	0.0428	86.14
6	21.05	116.77	0.00856	49.00
6	25.8	139.24	0.00717	42.50
6	30.0	157.36	0.00635	37.60
6	47.2	225.59	0.00444	23.1
6	53.1	251.62	0.00398	19.5
6	56.9	272.45	0.00367	17.3
6	62.9	293.08	0.00342	14.1
6	64.6	319.83	0.00313	
6	67.0	345.67	0.00290	
6	72.2	388.27	0.00257	

Unfortunately, accurate dosimetry data on the sources is needed in order to check this difference and determine the correct method.

Figure 104 is a plot of the percentage of dye unbleached by the radiation versus the exposure time. For moderate exposures, the film appears to follow the simple relation that the rate of bleaching is proportional to the amount of dye present. At higher dosages the relation becomes more complex and the data tends to scatter considerably.

From the slopes of these curves it is possible to estimate the relative strengths of the two cobalt-60 sources (see page 184). The results indicate that the 10-kilocurie source has 4.0 times the intensity of the 1-kilocurie source. It is understood, of course, that these relative intensities apply only in the center of each source. With this information it is possible to replot Fig. 103 with the exposure time of the 10-kilocurie source increased by a factor of four to place both sources on an equal exposure basis. This replot is shown as Fig. 105; in which the solid line represents the 1-kilocurie source data and the small circles are the 10-kilocurie-source experimental points.

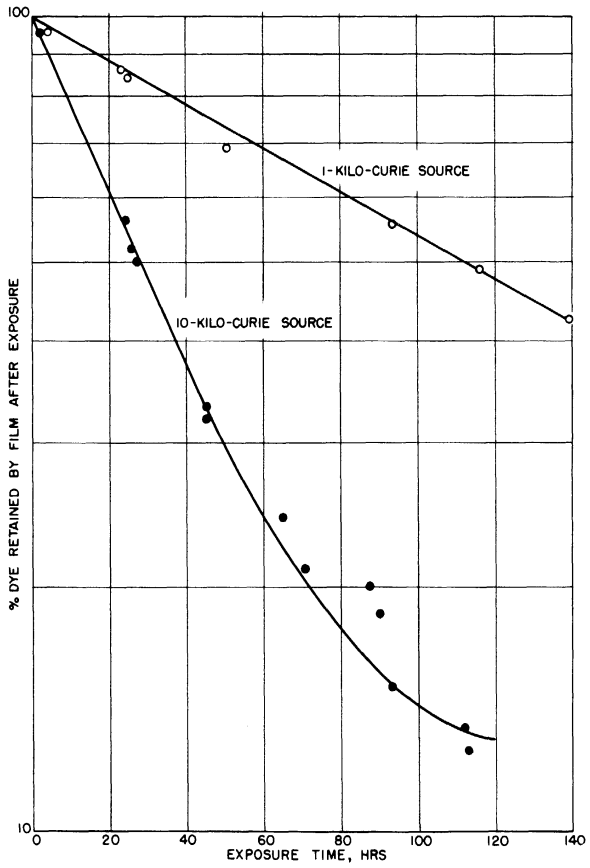


Fig. 104. Percentage Dye Retained by Film After Exposure.

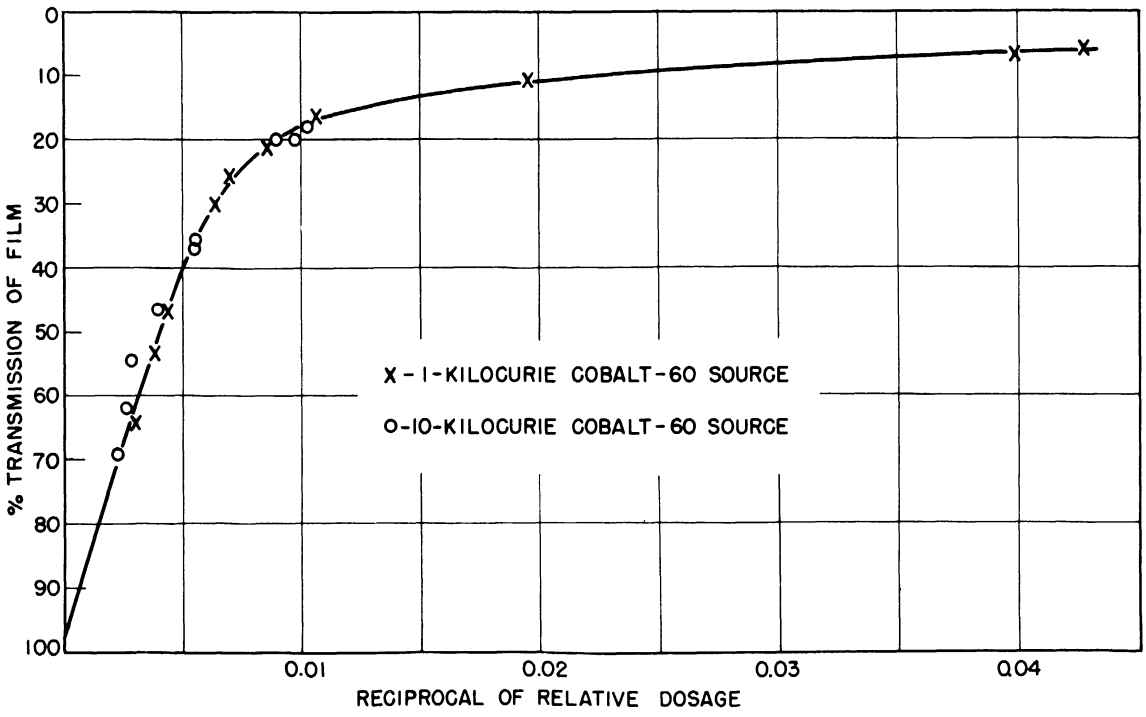


Fig. 105. Percentage Transmission of Film vs Reciprocal of Relative Dosage.

TABLE XLVIII

EXPOSURE DATA ON POLYVINYL CHLORIDE FILMS
RADIATION SOURCE: 10-KILOCURIE COBALT-60

Film No.	Transmission, %	Exposure Time, hr	Reciprocal of Exposure	Dye Retained, %
14	5.10	0	infinity	100.00
14	54.50	87.9	0.0114	20.00
14	68.7	113.7	0.00880	12.2
14	70.9	133.43	0.00750	10.09
14	73.1	158.32	0.00634	10.0
14	71.0	181.22	0.00551	10.08
15	4.68	0	infinity	100.00
15	17.7	24.38	0.0411	56.2
15	35.6	45.21	0.0222	33.2
15	46.8	64.94	0.0154	24.4
15	56.0	89.83	0.0111	18.5
15	64.9	112.73	0.00887	13.4
16	4.50	0	infinity	100.00
16	20.00	25.80	0.0388	51.9
16	37.10	45.53	0.0220	32.0
16	52.0	70.42	0.0142	21.0
16	62.5	93.32	0.0107	15.0
3	4.68	0	infinity	100.00
3	5.36	4	0.25	95.55
3	21.4	26.9	0.0372	50.0

From the fact that these points fall almost exactly on the graph, the following conclusions can be drawn:

1. The system is independent of intensity of radiation over a wide range of exposures.
2. The estimated relative intensity of the two sources (4:1) is close to the actual value.
3. It appears that results may be reproducible within 5-10% in most cases. However, there are some points that differ by more than 10% and more data are needed in the low-exposure region before any conclusions can be reached.

(2) Tests with X-Ray Machines: It has been reported[2] that the bleaching rate of the polyvinyl chloride films is a function of the wavelength of the

incident gamma radiation. This factor was checked by exposing the films to unfiltered radiation from a 250-kv x-ray machine. The output of the tube, as measured with a condenser-type roentgen meter, had the value of 1000 r/min in air. The film was exposed at this dosage rate for 2 hours and the transmission readings then compared to similar readings from the 1-kilocurie cobalt-60 source. It appears that the unfiltered x-ray radiation is about 2-1/2 times as effective in bleaching as the cobalt-60 radiation for equal dosages. This is in agreement with previous findings and indicates that it will not be possible to use the films to compare various sources of gamma radiation. Each wavelength will have its own calibration curve, unless some analytical method is devised to take this factor into account.

If a 1-inch test tube is placed in the beam path and the dose rate measured, the indicated reading is 2150 r/min inside the tube. A piece of film exposed inside the tube showed a transmission change twice that of a film suspended in air and exposed for the same length of time. This effect of the container on dose rates is a further complicating factor in attempting to determine the strength of various sources.

3. Studies with Leaded Films

One of the chief objections to the use of polyvinyl chloride films is the relatively long exposures necessary for calibration and use of the films. It takes from 75 to 100 hours of exposure to establish the calibration curve, and poor results are obtained from exposures of less than 8 hours.

One method of decreasing the exposure time is by increasing the gamma radiation absorption of the film. It was thought that this could be accomplished with the use of a backing material, or by incorporating a heavy metal within the film itself. A brief survey of the readily available compounds indicated that lead oleate and tetraethyl lead dissolve in the plastic solution. The former compound was found to be impure and contained free acid; hence, it was not used. Instead, 10 grams of tetraethyl lead were dissolved in 10 cc of cyclohexanone and the solution mixed well with an amount of stock plastic solution normally used to prepare a film. The film was rather long in drying (2 weeks), but the final film was clear and uniform in dye content. It had an odor of tetraethyl lead and was lighter in color than the other cast films.

Exposure in the 10-kilocurie cobalt-60 source showed that the film was about 50 times more effective in bleaching the dye than an unleaded film. Unfortunately, the films proved to be unstable and bleached out under normal storage conditions. A strip exposed to the sunlight would be rendered colorless within an hour. Samples stored under light-excluded conditions have shown a continuing bleaching-out of the dye. Apparently there is some type of chemical

action taking place within the film that liberates a dye-decolorizing agent, because the films turn from blue to a colorless condition. The unleaded films turn from blue to green and have a permanent brown color when bleached.

Although no further work was done along this line, it is believed that stable metal-containing films can be made. One source of metal-organic compounds is the large number of mercury derivatives that have been prepared.

4. Conclusions

Four requirements were given in the introduction to this section as a basis of judging whether a particular chemical or physical method would be suitable as a dosimeter device. If the polyvinyl chloride film results are compared with these requirements, the following conclusions can be reached.

1. The system does not vary in an exact manner. It has been shown to be a function of wavelength, pH, and possibly intensity at high levels of radiation. The effect of temperature was not studied.
2. The reaction is irreversible as carried out in dosimetry methods.
3. The results indicate that they are reproducible, over a limited range, within 5%.
4. The system is stable if stored out of sunlight or bright artificial light. Sunlight will almost completely bleach the film in two days. A No. 2 photoflood, 2 feet from the film, will cause a 10% increase in transmission in 20 minutes. However, it appears that exposure in the 10-kc source cave with the light on has no effect.

While the films do not meet all the requirements, neither do any of the other methods currently used in dosimeter measurements. Therefore, if the films are used in a fixed gamma radiation source, such as cobalt-60, and the total exposure is kept below 10 million roentgens, they will provide a convenient and reproducible method of measuring dose-rates.

5. Sample Calculations

a. Calculation of % Dye Retained after Exposure.

Data:

Initial Transmission	4.26
Transmission after exposure	25.80
Infinite Transmission	97.50

$$X = \frac{\log T - \log T_o}{\log T_i - \log T_o} ,$$

where

X = % dye converted,
T = final transmission,
T_o = initial transmission, and
T_i = infinite transmission.

Thus,

$$X = \frac{\log 25.8 - \log 4.26}{\log 97.5 - \log 4.26} = \frac{1.41162 - 0.62941}{1.98900 - 0.62941} = 0.575 ;$$

then,

$$\% \text{ dye retained} = (1-X)100 = (1.000-0.575)100 = 42.5\%.$$

b. Relative Strengths of the Two Cobalt-60 Sources.

Data: Slope of 1-kc Co-60 source (Fig. 103) 0.002685
Slope of 10-kc Co-60 source (Fig. 103) 0.01072

Assume each curve (straight-line portion) of Fig. 103 has the equation

$$\log X = mZ ,$$

where

X = % dye retained by film,
m = slope of curve, and
Z = exposure in hrs.

For equal dosages:

$$X_1 = X_2 \text{ or } \log X_1 = \log X_2 ,$$

where subscripts 1 and 2 refer to 1- and 10-kilocurie source, respectively.

Then,

$$m_1 Z_1 = m_2 Z_2$$

or

$$\frac{Z_1}{Z_2} = \frac{m_2}{m_1} = \frac{0.01072}{0.002685} = 4.0 .$$

Hence, for the same dosage, a sample must be exposed four times longer in the 1-kilocurie source than in the 10-kilocurie source.

6. Properties of the Materials Used in Preparation of Films.

Chlorobenzene - Eastman technical grade. Used without purification
Density - 1.39 g/cc

Cyclohexanone - Eastman technical grade. Distilled before use.
Store in dark bottle away from light.
Density - 0.948 g/cc

Polyvinyl chloride - Geon 101 Goodrich Rubber Co.
Density (finished film) - 1.39 g/cc

Methyl Violet 6B - General Aniline
Solubility - approx. 0.05 g/100 cc chlorobenzene

Concentration of dye in film	Initial transmission, %
0.40	5-6
0.25	10
0.08	51
0.0125	83

Tetraethyl Lead - Eastman Technical grade

7. References

1. Miller A., B.S. Thesis, Department of Chemistry, Brooklyn Polytechnic Institute.

2. Henely, E. J., and Miller, A., Nucleonics 9 No. 6, 62-66 (Dec., 1951).

3. Progress Report 3 (C00-91) Eng. Res. Inst., Univ. of Mich.

4. Breazeale, W. M. A Low-Cost Experimental Chain Reactor, ORNL-1105, (August, 1952).