ENGINEERING RESEARCH INSTITUTE
UNIVERSITY OF MICHIGAN
ANN ARBOR

PROGRESS REPORT NO. 2

UTILIZATION OF THE GROSS FISSION PRODUCTS

By

L. E. BROWNELL, Supervisor
L. C. ANDERSON
H. H. GOMBERG
J. J. MARTIN
W. W. MEINKE
R. B. MORRISON
L. THOMASSEN
E. T. VINCENT
R. A. WOLFE
G. J. VAN WYLEN

Assisted by

H. S. DOMBROWSKI
R. E. CULLEN
M. E. GLUCKSTEIN
D. E. HARMER
W. KERR
J. G. LEWIS
J. NEHEMIAS
R. D. PIERCE
F. L. TOBY
ALEXANDER WIER

Project M943

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INTRODUCTORY ABSTRACT

This report describes the progress on this project since Progress Report I, dated August 31, 1951. The modification of the allocated building space to adapt it to a laboratory for handling radioactive materials has been essentially completed. The high-level laboratory has been in full operation since September, 1951 and the low-level laboratory was put into operation in January, 1952.

Some experimental results have been obtained on all subprojects. These results are described in the report and are abstracted as follows: On subproject M943-A (Fundamental Study of Effects of Radiation on Combustion Engine Performance), combustion has been conducted with gaseous mixtures under nonexplosive conditions and reaction rates have been measured for various gaseous fuels without the influence of radioactive material. On subproject M943-B (Performance of Combustion Engines under the Influence of Radiation), a small-sized experimental burner has been operated with and without ionizing beta radiation. Palladium-109 has been used as a source of radiation. Tests have also been made on an experimental diesel engine (subproject M943-B), which will be operated with radioactive palladium in the combustion zone. A pressure reaction chamber or bomb has been fabricated for subproject M943-C (Effects of Radiation on Chemical Reactions), and some preliminary tests have been made in the 1000-curie cobalt-60 source and with palladium-109. On subproject M943-D (Preservation of Foods and Drugs by Irradiation), a variety of foods, drugs and microorganisms have been irradiated in the cobalt-60 source. The sterilization of heat-labile drugs by gamma irradiation appears to be one of the most promising commercial uses of the radioactive waste fission products. On subproject M943-E (Exploratory Research), alloys of strontium and aluminum have been prepared, which may have possibilities in the industrial use of the fission products.

Cooperative research has been undertaken with other groups interested in the use of the fission products. These other groups include (1) Project 20 of the Michigan Memorial-Phoenix Project; (2) the Atomic Energy Commission, Biological Effects of Irradiation Laboratory; and (3) Research and Development Division of Parke, Davis and Company. Results of this cooperative research are reported under subproject M943-D.

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PROGRESS REPORT II

UTILIZATION OF THE GROSS FISSION PRODUCTS

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

The effect of radiation on combustion engine performance is under vigorous investigation by three research groups of project M943. Originally this research was divided into two subprojects: M943-A (Reciprocating Engines) and M943-B (Jet Engines). However, as a result of the laboratory equipment available for research and the different approaches taken by the research investigators, the original plan has been modified.

Subproject M943-A, supervised by Dr. R. A. Wolfe, Associate Professor of Physics, is at present a basic investigation of the effects of radiation on combustion. Combustion reactions are carried out under nonexplosive conditions in glass equipment, previously evacuated, and reaction rates are determined. This is a fundamental approach to the problem, and therefore it would seem advisable to modify the title of this subproject to "M943-A, Fundamental Study of Effects of Radiation in Combustion Engine Performance".

Subproject M943-B is supervised by Professor E. T. Vincent, Chairman of the Department of Mechanical Engineering. This subproject is making use of the experimental engines available in different research laboratories of the University. An experimental diesel engine is available in the automotive laboratory, and tests will be performed by Professor G. J. Van Wylen, Assistant Professor of Mechanical Engineering. Equipment is available for the study of flame phenomena applicable to jet engines at the Combustion Research Laboratory, supervised by R. B. Morrison. Tests will be performed by R. E. Cullen and Alexander Weir, Research Associates, and R. B. Morrison, Research Engineer. A considerable amount of research has previously been performed on these engines for other projects, and, as a result, a quantity of reference data is available. In the initial tests, radioactive palladium will be used in the combustion zone, and the effects of ionizing radiation on engine performance will be noted. These direct performance tests tackle the problem from the opposite direction of the fundamental study of M943-A. In this manner, subprojects M943-A and M943-B complement each other and take advantage of the personnel and laboratory facilities available. It seems advisable to modify the title of subproject M943-B to "Performance of Combustion Engines under the Influence of Radiation".

A. SUBPROJECT M943-A FUNDAMENTAL STUDY OF EFFECT OF RADIATION ON COMBUSTION ENGINE PERFORMANCE

Personnel

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

1. Introduction

For the initial investigation of the effects of radiation on internal combustion, it was decided to study mixtures of combustible gases at pressures low enough to preclude explosions. For this purpose a vacuum system and high voltage d-c source have been built so that the changes in pressure in a fixed volume of gas can be followed, while maintaining a d-c glow discharge in the gas. It has been found that the glow discharge can be adjusted to give a reaction rate slow enough to be followed easily.

Before the effects of radioactive sources on this reaction rate can be measured, it is necessary to determine the normal behavior without radioactive sources present. This report deals entirely with the first experiments, designed to establish such a normal behavior.

2. Experimental Procedure

Mixtures of methane and oxygen and of hydrogen and oxygen were subjected to a d-c glow discharge in a spherical glass bulb of approximately 15 cm inside diameter. The electrodes consisted of aluminum discs 20 mm in diameter and 4 mm thick, separated a distance of 15 mm between front surfaces. Potentials of 400 to 600 volts were applied across the electrodes. The discharge current varied between 15 and 20 milliamps, depending on the gas pressure and composition.

Pressure measurements were made with a McLeod gauge separated from the bulb by a cold trap which was cooled to a temperature of about -78°C by means of a mixture of dry-ice and acetone. Starting pressures used in the reaction bulb varied between 5 and 15 mm. During most runs a pressure reading was taken one minute after the initiation of the discharge, and at two-minute intervals thereafter. When the rate of change of pressure became sufficiently small, the time intervals between readings were gradually increased to 5, 10, and 20 minutes. A stopwatch was used to measure these intervals.

No attempt was made to regulate voltage or current during these runs. The power applied to the electrodes was the maximum available from the current rectifier used. The current was limited by a ballast resistor of 50,000 ohms. The powerstat control was set at maximum output at all times. Between runs the system was pumped down to less than one micron pressure, with the cold trap in operation, before a new charge was placed in the bulb.

Fig. 1 is a schematic diagram of the vacuum system. Figs. 2 and 3 show the completed system. Fig. 5 shows the reaction bulb in operation.

3. Results

a) Behavior of methane-oxygen mixtures. The runs on methane-oxygen mixtures are of two distinct types depending on the relative proportions of oxygen and methane. For high proportions of oxygen, the pressure increased sharply to a maximum in a few minutes, and then decreased. The shape and location of the maxima appear to depend on both the composition and the total pressure of the charge.

Table I gives data from runs of this type and Fig. 6 is a plot of pressure vs time for some of these runs. Similar runs have been grouped together to provide an indication of reproducibility. Note that the curves in each set show considerable similarity, including even the small but definite minima observed in sets B and C.

Sets B and C differ from each other essentially only in their total pressure. Set C shows the same characteristics as set B, but the minima occurs somewhat earlier. On the other hand, sets A and B, which differ considerably as to composition, show a noticeable difference in shape. Note the break occurring in set A at a value of between 11 and 15 minutes after the start of the discharge. Run 9 differed from the others in set B only in that a cold-trap was not used. The resulting curve shows considerable broadening of the second maximum and a much more erratic tail. In Table I, note particularly the stability of the ratio P_{max}/P_T.

For oxygen-to-methane ratios of one or less, the pressure increased assympotically and then became essentially constant. Runs made on pure methane showed a similar behavior. Table II gives data on some runs of this type, and Fig. 7 shows curves for runs at three different compositions.

Fig. 8 shows the effect of stopping the discharge part way through the run. The circles indicate the points at which the discharge was stopped. Note the relatively sharp drop immediately after cut-off. This is followed by a gradual decrease in slope which eventually becomes less than that for the curve corresponding to a continued application of the discharge.

SCHEMATIC DIAGRAM OF VACUUM SYSTEM AND REACTION BULB

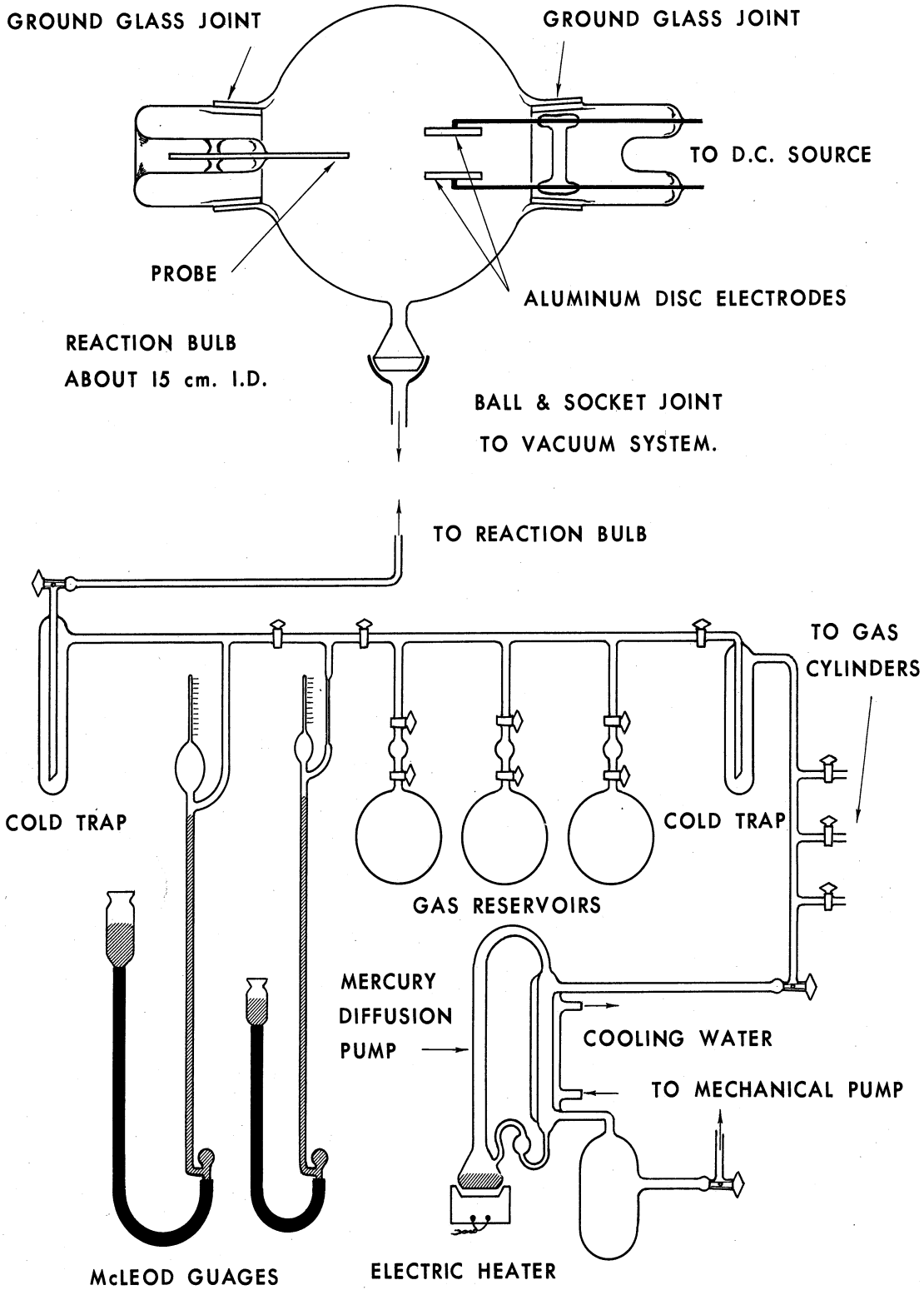

Fig. 1

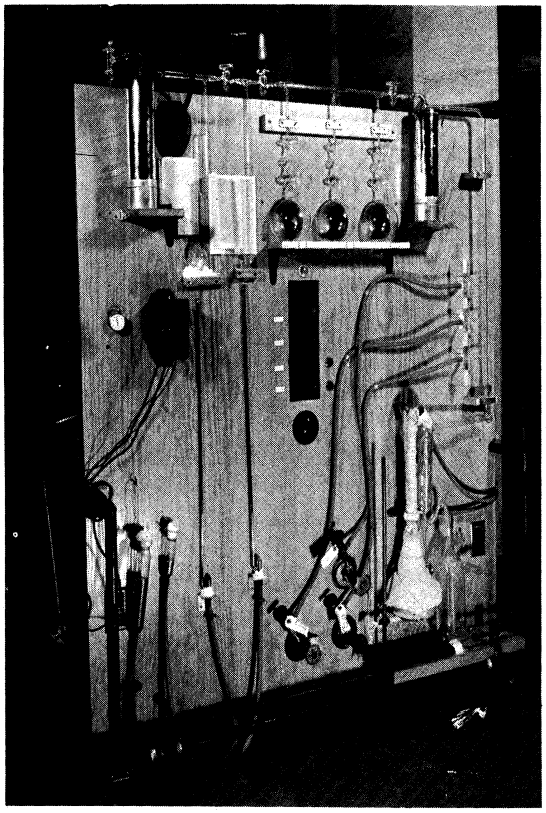

Fig. 2. Front View of Completed System

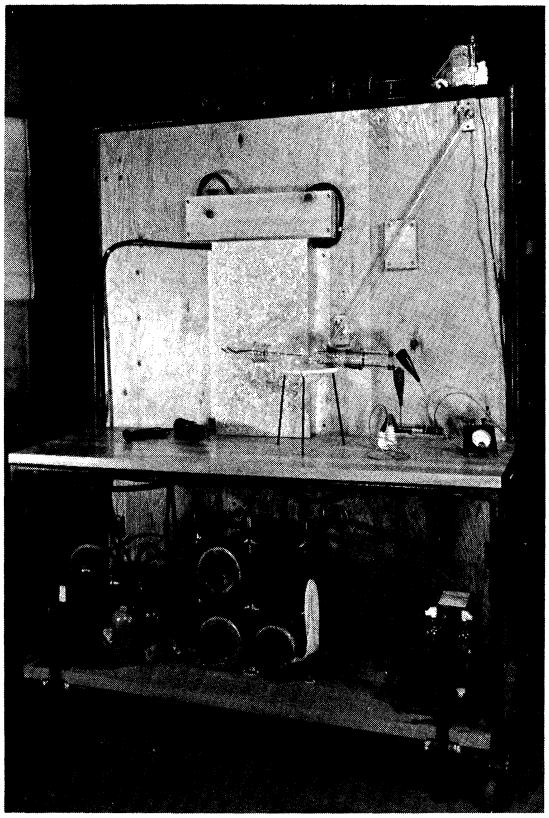

Fig. 3. Rear View of Completed System

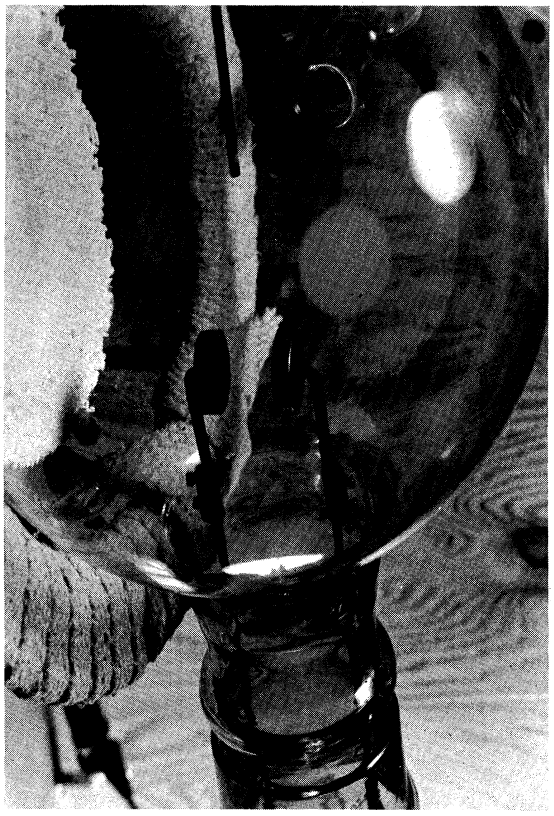

Fig. 4. Close-Up of Reaction Bulb Showing Al Disc Electrodes

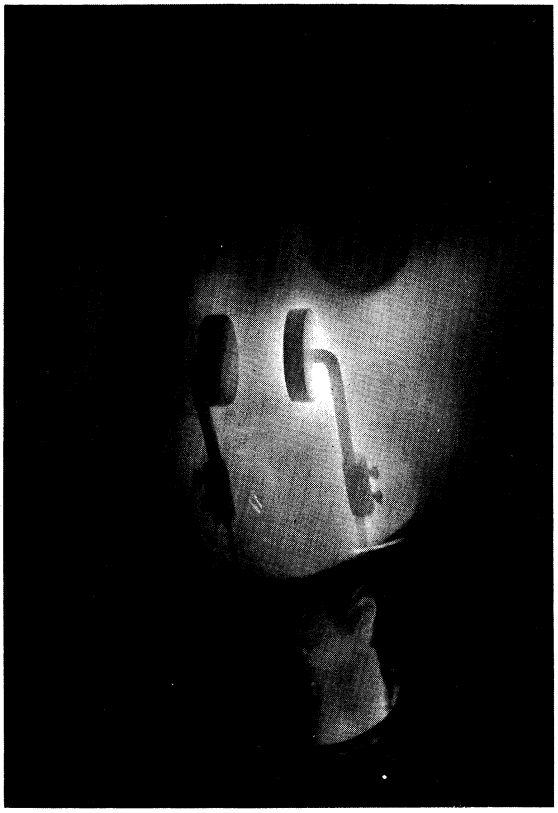

Fig. 5. Reaction Bulb in Operation

BEHAVIOUR OF METHANE–OXYGEN MIXTURES IN D. C. DISCHARGE–HIGH OXYGEN RATIO

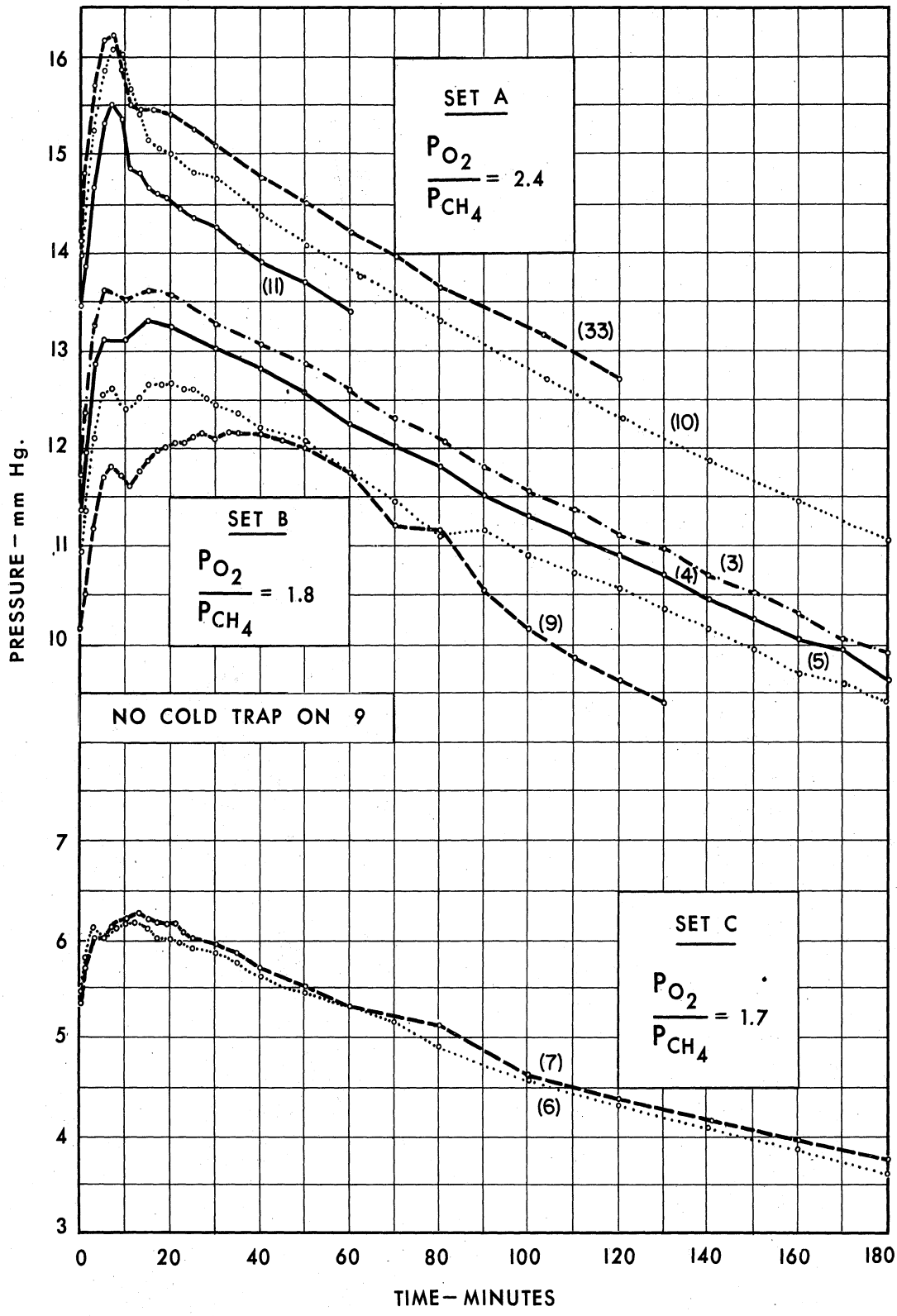

Fig. 6

TABLE I

DATA OF COMBUSTION OF METHANE AND OXYGEN
USING MIXTURES RICH IN OXYGEN

Run No.	P_T	P_{O_2}/P_{CH_4}	P_{max}	$\Delta P/P_{O_2}$	$\Delta P/P_{CH_4}$	P_{max}/P_T	t_{max}	t_{min}
3	11.70	1.85	13.60	0.250	0.463	1.16	15	10
4	11.35	1.84	13.30	0.265	0.487	1.17	15	10
5	10.90	1.82	12.65	0.249	0.452	1.16	18	10
*9	10.15	1.69	12.15	0.314	0.530	1.20	33	11
6	5.45	1.74	6.15	0.202	0.352	1.13	11	5
7	5.35	1.73	6.25	0.266	0.460	1.17	13	5
10	13.95	2.14	16.10	0.226	0.483	1.16	7	-
11	13.40	2.12	15.50	0.231	0.488	1.16	7	-
33	14.10	2.17	16.20	0.218	0.472	1.15	7	-

P_T = Total starting pressure

P_{O_2} = Initial pressure of O_2

P_{CH_4} = Initial pressure of CH_4

P_{max} = Maximum pressure

ΔP = $P_{max} - P_T$

t_{max} = Time of maximum

t_{min} = Time of minimum

All pressures in mm Hg

All times in minutes

*This run made without cold trap.

BEHAVIOUR OF METHANE-OXYGEN MIXTURES

IN D. C. DISCHARGE-LOW OXYGEN RATIO

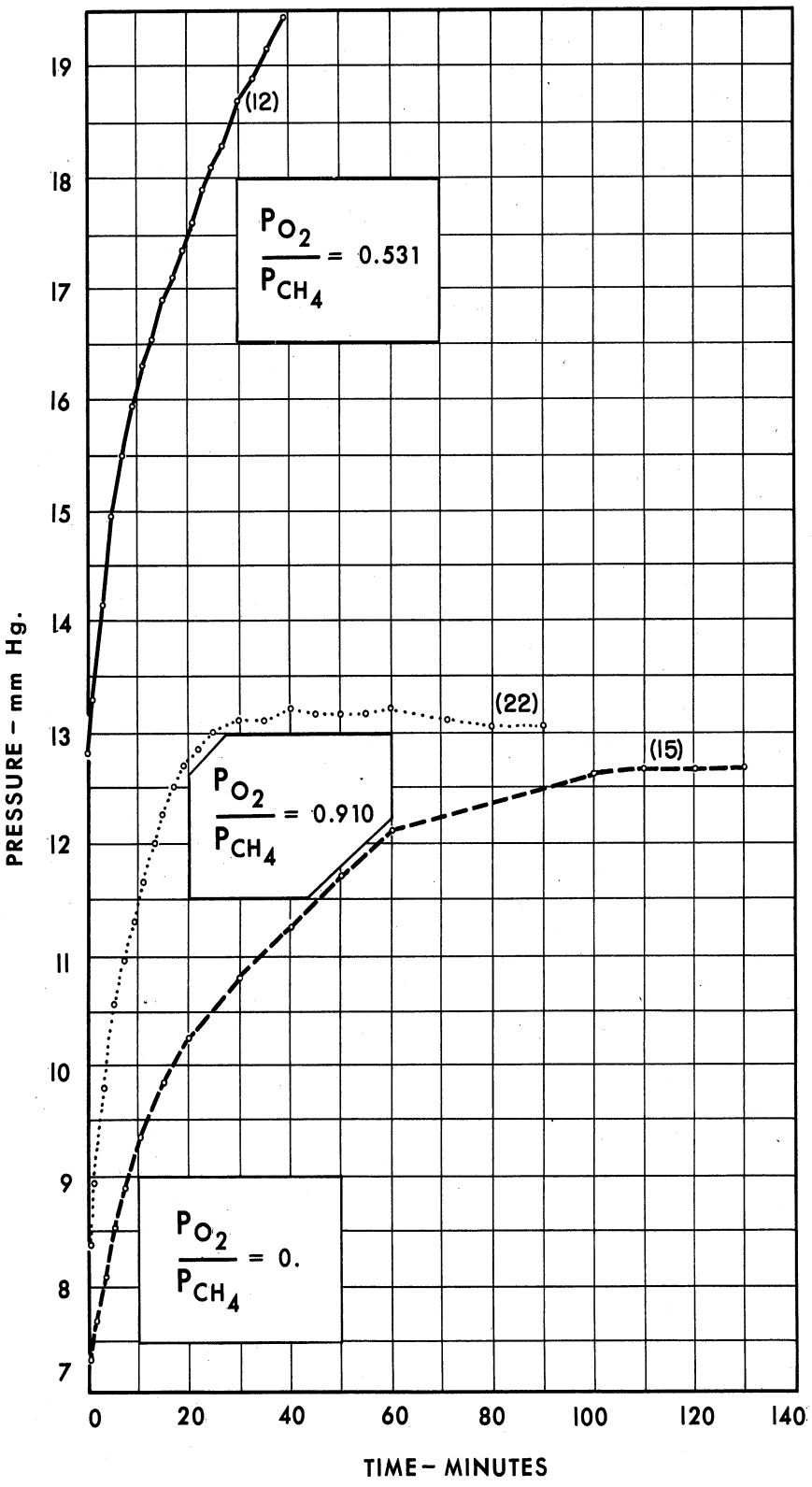

Fig. 7

TABLE II

DATA OF COMBUSTION OF METHANE AND OXYGEN
USING MIXTURES LEAN IN OXYGEN

Run No.	P_T	P_{O_2}/P_{CH_4}	P_{max}	$\Delta P/P_{O_2}$	$\Delta P/P_{CH_4}$	P_{max}/P_T	t_{max}
12	12.83	0.531	23.00	2.28	1.21	1.79	>210
22	8.40	0.910	13.20	1.20	1.09	1.57	40
23	8.20	0.907	12.95	1.22	1.10	1.58	40
13	7.85	0	13.40	----	0.707	1.71	102
14	7.65	0	12.15	----	0.588	1.59	60
15	7.35	0	12.65	----	0.721	1.72	110

For explanation of symbols, see Table I.

b) Behavior of hydrogen-oxygen mixtures. The pressure during these runs dropped immediately and sharply once the discharge was initiated. After 7 to 10 minutes, a break appears in the pressure-time curve. The pressure continues to drop, but more gradually. Thereafter, the slope of the curve decreases gradually. Table III gives data for a set of runs on hydrogen-oxygen mixtures.

Fig. 9 compares two such runs with runs in which the discharge was cut off during the course of the run. These latter runs show a definite similarity in behavior to similar runs made on methane. Thus, in runs 28 and 30 the steeper slope immediately following the cut-off occurs again. This is followed by a leveling off of the curve to a slope less than that of the normal curve at the same point. In runs 27 and 29 there was again observed a very rapid leveling off to a relatively gentle slope immediately after cut-off at three minutes. These curves may be compared with those for run 31 on methane (Fig. 8).

4. Conclusion

An examination of the pressure-time curves for hydrogen-oxygen mixtures in which the discharge was cut off at 35 minutes shows a continued drop in pressure for a long period thereafter. It seems unlikely that a gaseous

BEHAVIOUR OF METHANE–OXYGEN MIXTURES
IN D. C. DISCHARGE–HIGH OXYGEN RATIO

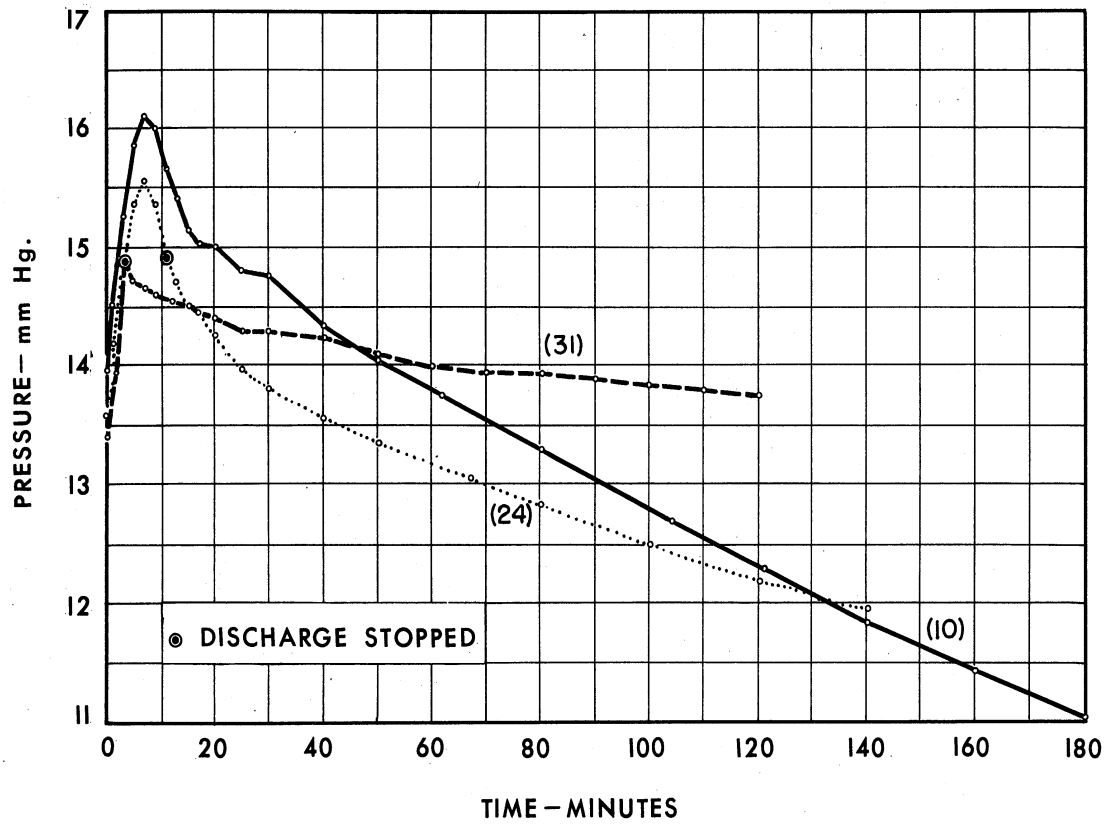

Fig. 8

reaction could have continued for any length of time after the discharge stopped. One possible explanation for this drop in pressure is that water vapor was gradually diffusing into the cold trap, where its contribution to the total pressure was reduced essentially to zero. The cold trap is separated from the reaction bulb by a long tube of comparatively small diameter, so that it would be expected that the water vapor would require a long time to diffuse into the trap.

A McLeod gauge will not measure the true pressure of a gas below its critical temperature. If such a gas has penetrated to the McLeod gauge, it will merely condense in the bulb of the gauge on being compressed by the mercury column. In the present case, however, no condensable vapor could reach

the McLeod gauge due to the action of the cold trap. Therefore, the gauge could measure the pressure due to water vapor in the reaction bulb, since the pressure throughout a connected system must be constant.

A comparison of runs 28 and 30 with 20 and 21 (Fig. 9) indicates that the reaction is nearly completed in a few minutes, probably at the time the break occurs in the pressure-time curve. It might be possible to explain the difference in the tails of the curves by a temperature effect, since the reaction bulb became noticeably warm when the discharge was operating. Thus, the initially steeper slope following cut-off might be the result of the drop in pressure of the gas while cooling. The slopes of these curves at higher values on the time scale are less than those of runs in which the discharge was not stopped. This may be the result of a decreased diffusion rate at lower temperatures.

The same considerations would apply to the runs on methane-oxygen mixtures, where the proportion of oxygen was high (runs 24 and 10, Fig. 8). However, other condensable vapors besides water might enter here.

For those runs in which the oxygen-to-methane ratio was one or less, no significant drop was observed from the maximum pressures attained. This indicates that all the products formed in any quantity must show a measurable vapor pressure at dry-ice temperatures. In particular, the reaction which would produce water must occur to a much smaller extent than some others.

Consider the more probable reactions in pure methane:

	Reaction	P_{max}/P_T
(a)	$CH_4 \longrightarrow C + 2H_2$	2.0
(b)	$2CH_4 \longrightarrow C_2H_6 + H_2$	1.0
(c)	$2CH_4 \longrightarrow C_2H_4 + 2H_2$	1.5
(d)	$2CH_4 \longrightarrow C_2H_2 + 3H_2$	2.0

Consideration of the experimentally observed values of P_{max}/P_T (Table II) and the fact that a black powdery substance formed on both electrodes during these runs indicates that reaction (a) is probably one of the principal reactions. Reactions (b) and (c), which also occur to some extent, are also indicated. Just what reactions occur in the presence of oxygen will require considerably more investigation.

5. Projected Experiments

Further tests will be made to establish more definitely the nature of the principal reactions between methane and oxygen. Runs will be made

BEHAVIOUR OF HYDROGEN–OXYGEN MIXTURES IN D. C. DISCHARGE

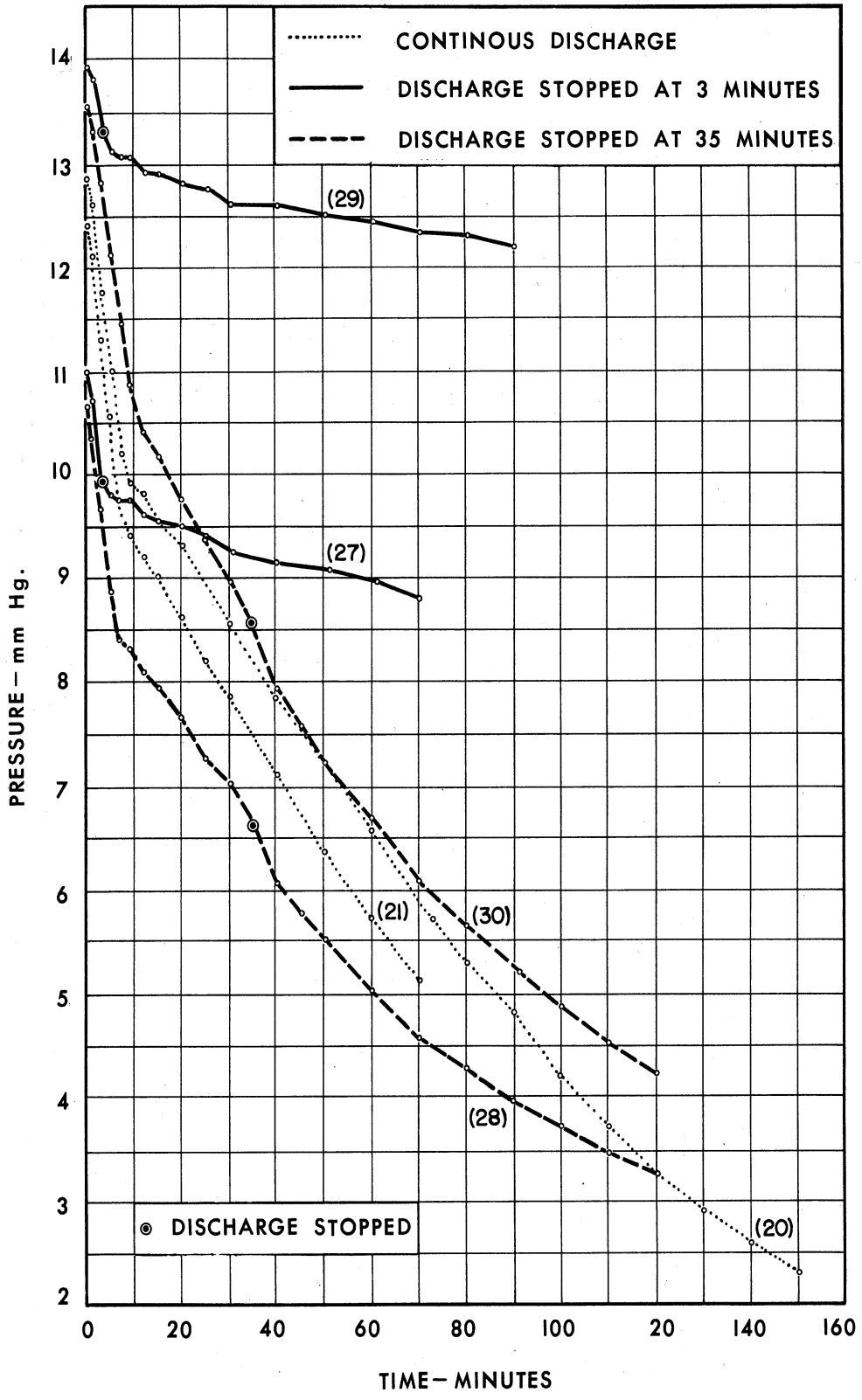

Fig. 9

TABLE III

DATA OF COMBUSTION OF HYDROGEN AND OXYGEN

Run No.	P_T	P_{H_2}/P_{O_2}	t_B
19	6.60	2.07	5
20	12.85	2.06	8
21	12.40	1.99	8

P_T = Total starting pressure

P_{O_2} = Initial pressure of O_2

P_{H_2} = Initial pressure of H_2

t_B = Time of break in curve.

with drying agents and carbon dioxide absorbents in the reaction bulb. It is hoped such runs will show the duration of significant gaseous reactions.

If, as is suspected, the reaction time turns out to be of the order of ten minutes, it will be necessary to slow down the rate so that more readings can be taken during the course of the reaction. To this end the discharge current will probably be reduced to a fraction of its present value. It will also be necessary to regulate the current more closely than was previously done.

If a consistently reproducible pressure-time curve can be obtained for the duration of the reaction, it should be possible to test the effects of radioactive sources on the gaseous reactions. For this purpose, a bakelite plaque impregnated with phosphorous-32 has been ordered from Oak Ridge. This plaque is one inch in diameter and 0.19 inch thick and has a reported radiation intensity of 2000 rep per hour.

It may be desirable later to examine the effects of the radioactive sources on rates of reaction maintained by a short-wave-length light source, such as a mercury lamp in place of the present glow discharge. This should provide additional valuable information about the relative effectiveness of the electric current and light radiation in producing and maintaining reactions.

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

Personnel

Subproject Supervisors: E. T. Vincent, Professor of Mechanical Engineering and Chairman of Department; G. J. Van Wylen, Assistant Professor of Mechanical Engineering; R. B. Morrison, Research Engineer. R. E. Cullen, Research Associate; A. Weir, Jr., Research Associate.

1. Internal-Combustion Engines

Introduction. The effect of radiation on the combustion process of reciprocating engines is being considered in both the compression ignition (diesel) engine and the spark-ignition engine. The necessary combustion qualities of the fuel are quite different for the two engines. The compression-ignition engine requires a fuel of low self-ignition temperature and short ignition delay, whereas the fuel for a spark-ignition engine must have a high self-ignition temperature and a long ignition delay. A preliminary survey of the literature indicates that radiation tends to increase the rate at which a chemical reaction takes place. Since ease of ignition accompanied with an increasing rate of the combustion process would be especially desirable in a compression-ignition engine, it was decided to conduct the first tests on a compression-ignition engine. The effects in the diesel-engine cylinder accompanying easier ignition and faster combustion can be shown on a pressure-time diagram as follows:

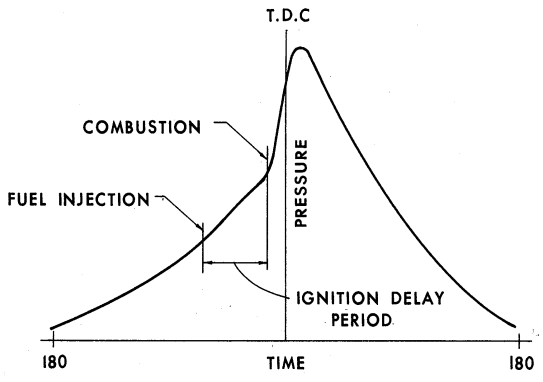

Fig. 10. Typical Pressure-Time Curve for Combustion in Diesel Engine

Fuel is injected some degrees before top dead center (T.D.C.) near the end of the compression stroke. On injection into the cylinder the fuel does not ignite instantaneously, and as a result there is an accumulation of fuel in the cylinder. The period between injection and ignition is called the ignition-delay period. When ignition does take place there is an extremely rapid combustion of the accumulated fuel, producing a very rapid increase in the cylinder pressure. If the ignition-delay period is too long, the ignition of the larger amount of fuel accumulated in the cylinder produces excessive peak pressures and knocking. The cetane number of a diesel fuel is a measure of its anti-knock combustion characteristics in the same manner as the octane number is used to rate spark-ignition fuels. For a diesel engine, therefore, the sooner the fuel ignites after being injected into the cylinder, the better will be the combustion characteristics and anti-knock qualities of the fuel.

Activation of the combustion process by radiation may reduce the delay period with the result that it may be necessary to inject the fuel closer to T.D.C. for the same combustion point. The result will be a reduction in the fuel accumulation in the engine cylinder at the ignition point thereby reducing the tendency to knock by reducing the rapid pressure rise, and producing a lower peak pressure. It follows that for any given peak pressure (for which the engine has been designed) earlier injection can be used, which improves fuel economy and power output. A greater quantity of fuel will be burned under controlled (in place of detonating) conditions, producing greater cycle efficiency.

It is also possible that radiation may cause an increase in the combustion rate, resulting in earlier completion of the process which will again be reflected in increased power and economy. This effect, and the short ignition delay period, may result in considerable gains in compression-ignition engine performance as a result of radiation.

The engine on which the tests are being conducted is a Waukesha CFR diesel test engine shown in Fig. 11. It is the property of the Department of Mechanical Engineering and is located in the Automotive Laboratory. This is a standard test engine on which the cetane number of diesel fuels is determined. Combustion in this CFR engine takes place in a combustion chamber located above the piston, as shown in Fig. 11. The hot gases pass from the combustion chamber through a port into the cylinder, where they expand as work is done on the piston. The compression ratio may be varied by adjusting the position of the compression plug in the combustion chamber.

The source of radiation is a piece of palladium which is mounted on the compression plug as shown in Figs. 12 and 13. This palladium is 1-1/2 inches in diameter, 1/8 inch thick and has a mass of about 37.5 grams. The compression and combustion processes are thus subjected to irradiation from

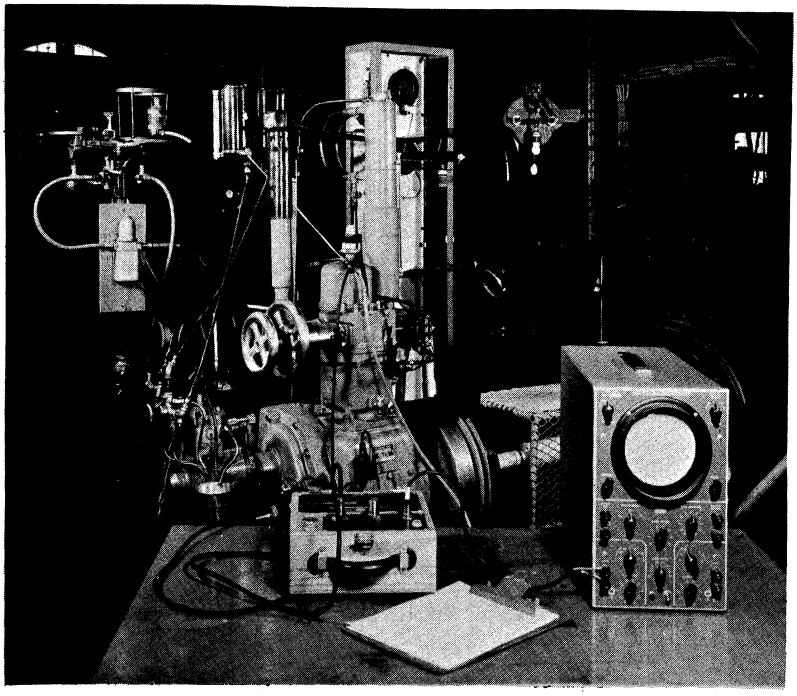

Fig. 11. View of Waukesha CFR Diesel Test Engine

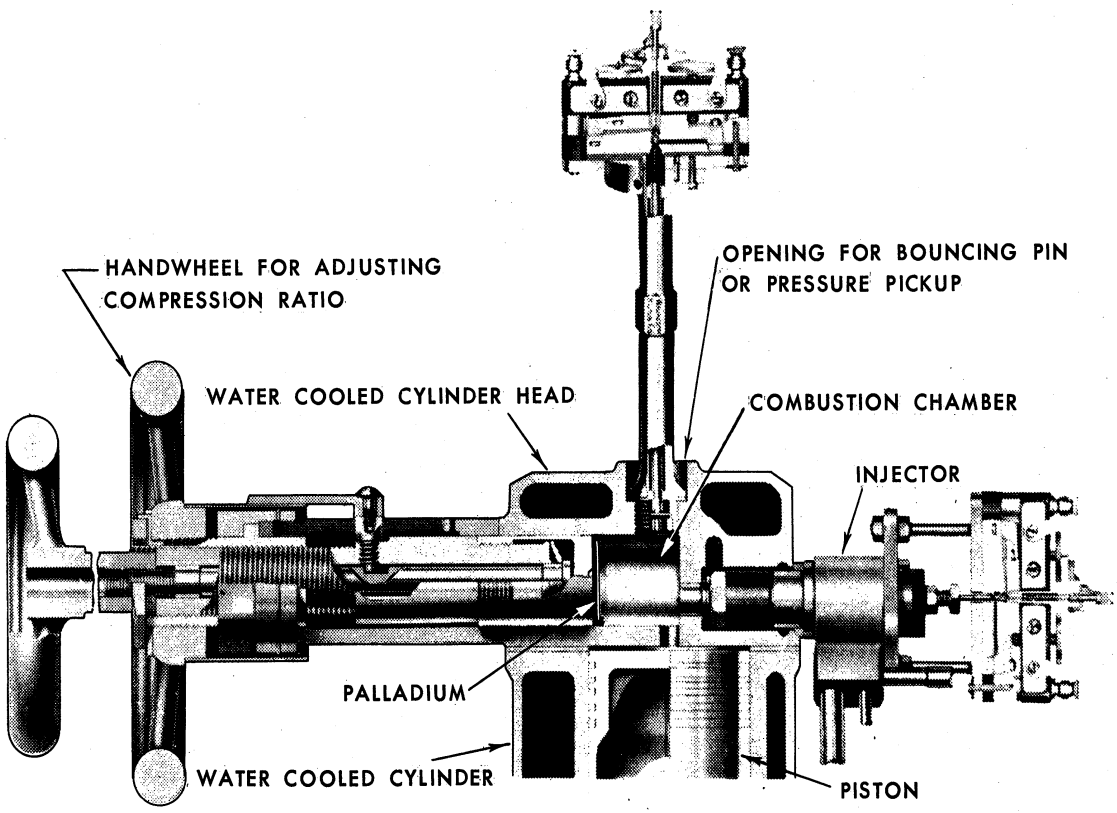

Fig. 12. Sectional View of Combustion Chamber in CFR Engine

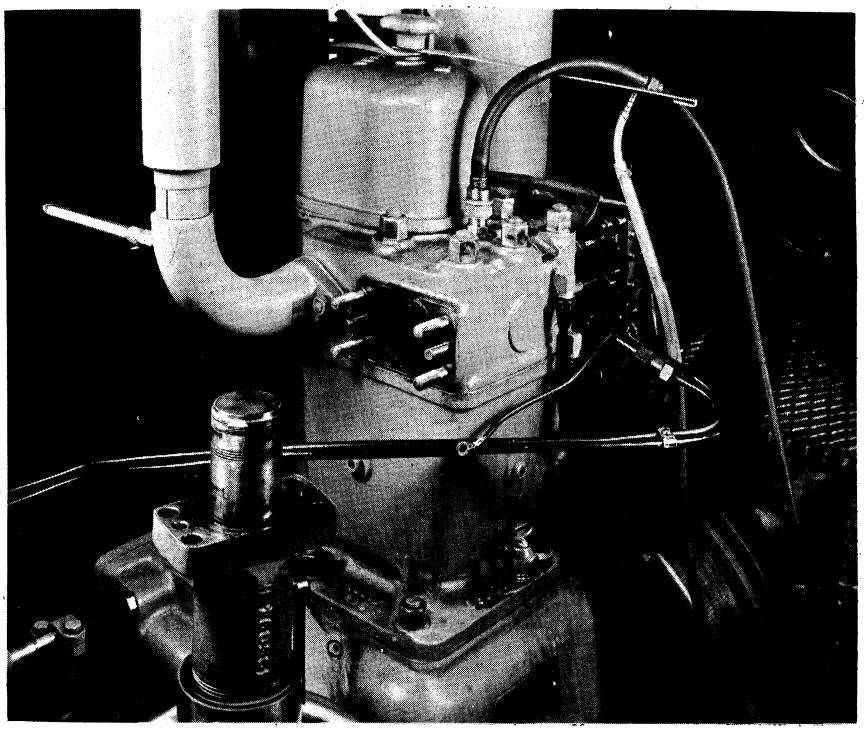

Fig. 13. Photograph of Compression Plug with Palladium

the palladium. After leaving the engine, the exhaust gases are passed through a glass-wool filter and exhausted above the roof of the building.

The effect of radiation on the combustion process is being measured by running two series of tests. In the first series, the cold (nonradioactive) palladium is placed in the engine and in the second series, the identical tests will be run after the palladium has been made radioactive. In each of the series, the following four tests will be run:

1) The minimum compression ratio at which the engine will fire
2) An indicated cetane number of the fuel
3) Pressure-time measurements of the combustion process
4) Fuel consumption and efficiency.

The minimum compression ratio at which the engine will fire is determined by bringing the engine up to standard operating temperature and motoring at some compression ratio lower than that necessary to cause firing with the fuel by-pass valve open (i.e., no injection of fuel into cylinder). The compression ratio is then increased in small increments once each minute. During the last five seconds of each minute the fuel by-pass valve is closed, causing fuel to be injected. The compression ratio at which the engine fires

once during the five-second injection period is considered to denote the minimum compression ratio at which the engine will fire. This procedure is quite arbitrary but has been accurately duplicated on cold runs and should indicate any change caused by the irradiation.

The actual cetane number of the fuel cannot be readily measured, since it depends on using reference fuels of known cetane number, and it would be expected that the irradiation would have the same effect on the reference fuels as on the regular fuel. However, the same fuel will be used on both the cold and hot runs, and any change in cetane number should be indicated. However, the magnitude of the increase in cetane number will have to be approximated. The ASTM test procedure for measuring cetane numbers is being followed in this test.

Pressure-time measurements are being made with a Control Engineering catenary diaphragm-type pick up, using a DuMont oscillograph, type 304-H. This oscillograph is equipped with a 35 mm camera, and pictures of the pressure-time diagram on the scope will be taken at various definite operating conditions on both the cold and hot runs. These will be compared to observe any effects caused by radiation.

The final test is the measurement of fuel comsumption and efficiency in the usual manner and a comparison of the results for the cold and hot runs. This is perhaps the least sensitive of the four tests, but any large effects would be observable and of much significance.

At the present writing, the palladium disc has been received from the manufacturer and the series of tests with the unactivated palladium has been completed. The runs with irradiated palladium are scheduled for February 4, 1952.

2. Jet Engines

a) Introduction. The purpose of this phase of the research program is the study of the effects of radiation on combustion processes applicable to the jet engine.

Investigations have been carried on in the past concerning the effects of several variables on the normal propagation rates of Bunsen flames and the blow-off velocities of simple, spherical-type flame holders. Some of the variables investigated have been pressure, temperature, type of fuel, fuel-air ratio, and system configurations. The results of these investigations have

been reported in University of Michigan External Memoranda 74[1] and 81[2]. It has been proposed that a radioactive source be placed in close proximity to the above-mentioned type of flames to determine the effect of radiation on the normal flame propagation velocity and the blow-off velocity. These two properties, i.e., the normal propagation velocity of laminar Bunsen-type flames and the blow-off velocity of inverted V-type flames stabilized by flame holders, are of fundamental importance to the design and operation of jet engines.

b) Description of Apparatus and Experimental Procedure. The apparatus used for the investigations carried on in the past consisted of a combustion chamber made of welded mild steel, 1/2 inch thick, with dimensions of 10 x 12 x 20 inches. A fuel-air mixture is introduced through a 1-5/8 inch diameter stainless-steel tube located in the bottom of the combustion chamber. At the top of the stainless-steel tube a nozzle assembly is located which holds any one of several interchangeable nozzles of different diameters. A photograph of the combustion chamber appears in Fig. 14 and an exploded view of the tube, nozzle assembly and nozzles appears in Fig. 15. The fuel-air mixtures are prepared in storage bottles at known mixture concentrations and at a pressure of 240 psig. The mixture flows through a pressure-regulating valve, then through one of three rotameters of different capacities, where the temperature and pressure of the flow are kept at about atmospheric conditions, i.e., conditions under which the rotameters were calibrated. The mixture then flows through a metering valve and then to the combustion chamber. After combustion takes place, the burned mixture is evacuated either by means of a Nash Hytor vacuum pump or a 3000 psi air ejector. A schematic drawing of the complete apparatus appears in Fig. 16 and a photograph of the apparatus is shown in Fig. 17. Flames are observed through two laminated plate glass windows 1-1/2 inches thick.

The pressure of the mixture at the rotameters and in the combustion chamber is measured by two 117-inch Meriam-type mercury manometers. The temperature of the mixture at the rotameters is measured by a mercury thermometer with the bulb immersed in the line. The temperature of the mixture at the nozzle exit in the combustion chamber is measured by a sliding Chromel-Alumel thermocouple assembly introduced through the side of the combustion chamber.

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[1]University of Michigan External Memorandum 74, September 1950. USAF Contract W33-038-ac-21100, Project MX-772, "Blowoff Velocities of Spherical Flameholders", by A. Weir, Jr., D. E. Rogers, and R. E. Cullen.

[2]University of Michigan External Memorandum 81, December 1950. USAF Contract W33-038-ac-21100, Project MX-833, "The Effect of Pressure on the Propagation Rate of Bunsen Flames in Propane-Air and Ethylene-Air Mixtures", by R. E. Cullen.

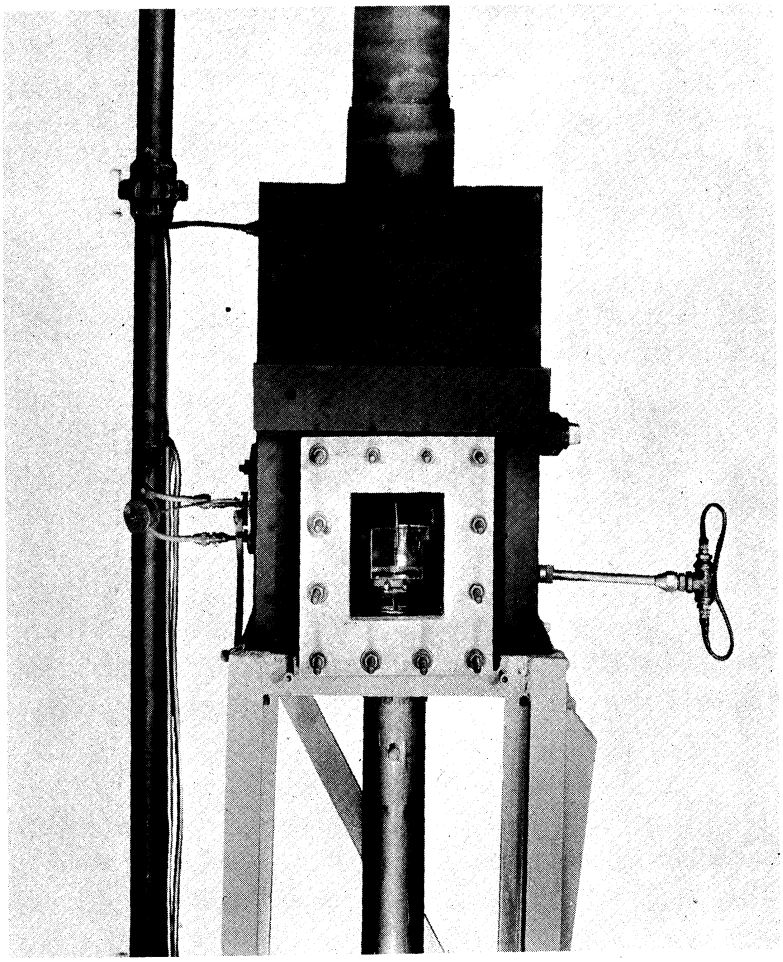


Fig. 14. Photograph of Combustion Chamber

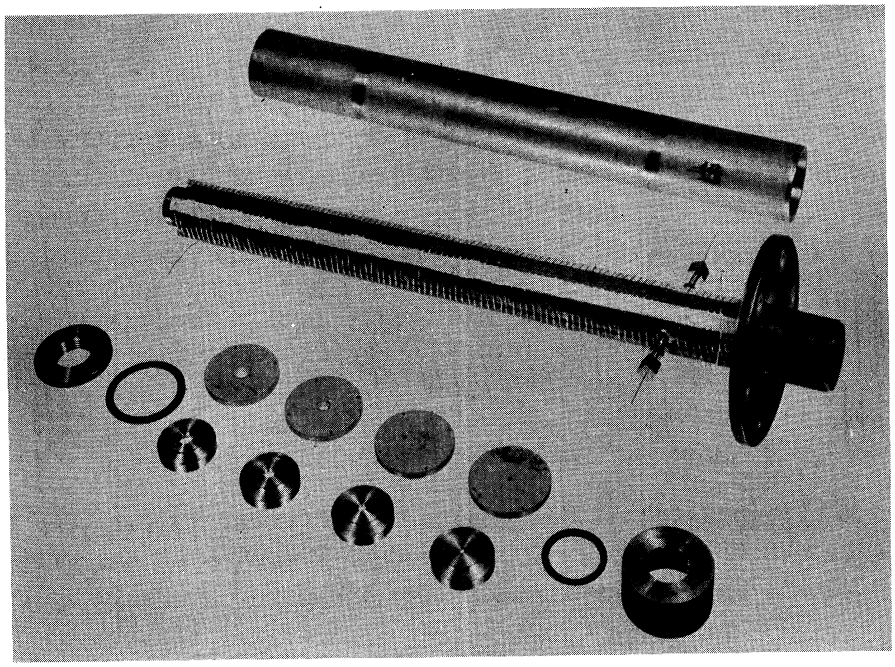


Fig. 15. Exploded View of Burner Tube and Nozzles

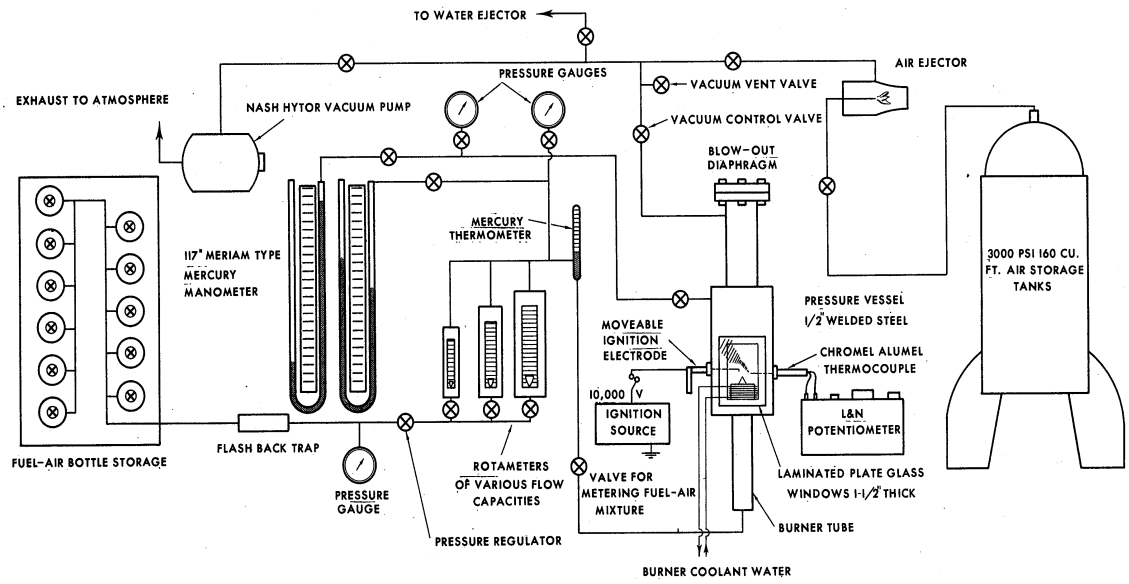


Fig. 16. Schematic Drawings of Apparatus used in Flame Studies

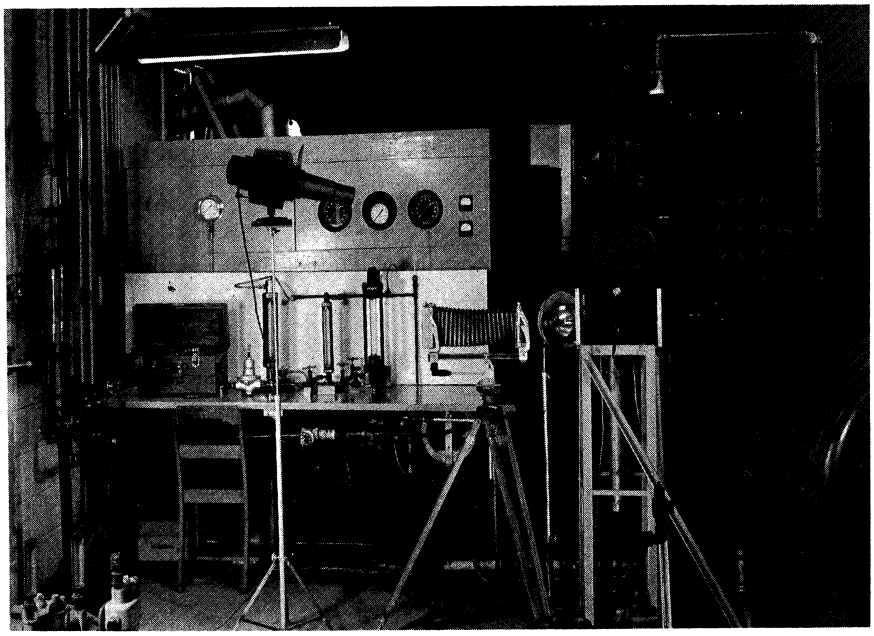


Fig. 17. Photograph of Apparatus for Study of Flame Phenomena

To determine the normal propagation velocity of the Bunsen flames, a 1-1/4-inch-diameter nozzle is used and flames are observed burning at pressures ranging from 13 inches mercury to about 2 inches mercury absolute. Photographs of flames are taken with an Eastman Graphic View camera. The image of the luminous reaction zone on the negative is made approximately full size. The photographs are then projected to about six times full size on white tracing paper and the inner boundary of the luminous reaction zone is recorded. The flame surface area is then computed by means of graphical integration. The normal flame-propagation velocity is obtained by the Gouy area method

$$V_f = Q/A_f ,$$

where V_f is the average normal flame-propagation velocity over the entire flame surface, Q is the volumetric rate of flow of the unburned mixture prior to entering the reaction zone, and A_f is the surface area of the inner limit of the reaction zone.

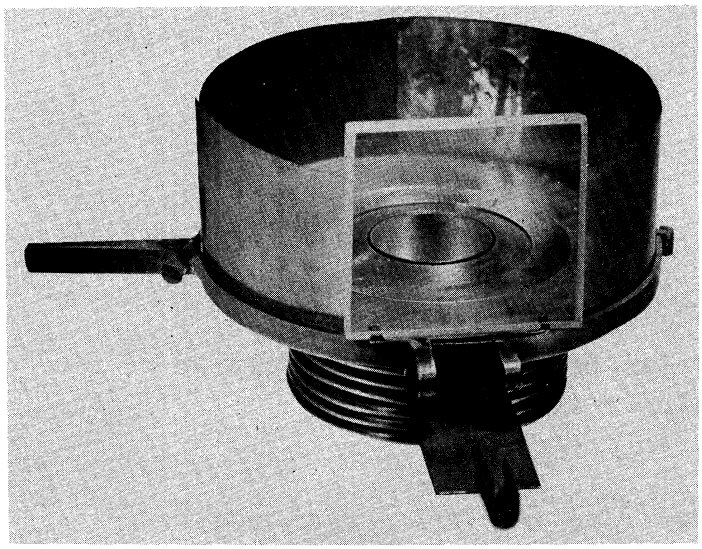
To determine the blow-off velocity of inverted V-type flames, a spherical 1/8-inch-diameter steel flame holder is used to stabilize the flame; the flame holder is situated 3/16 inch above a 3/8-inch-diameter nozzle. A fine wire of three twisted strands of .004-inch nichrome wire threaded through a No. 80 drill hole in the sphere is used to support the sphere.

In the experimental procedure the flame is ignited at atmospheric pressure, the flow rate in increased and the pressure reduced simultaneously until blow-off occurs at some pressure ranging from 23 inches mercury to 10 inches mercury absolute, at which instant the necessary readings of the flow instruments are taken to determine the mass velocity at blow-off.

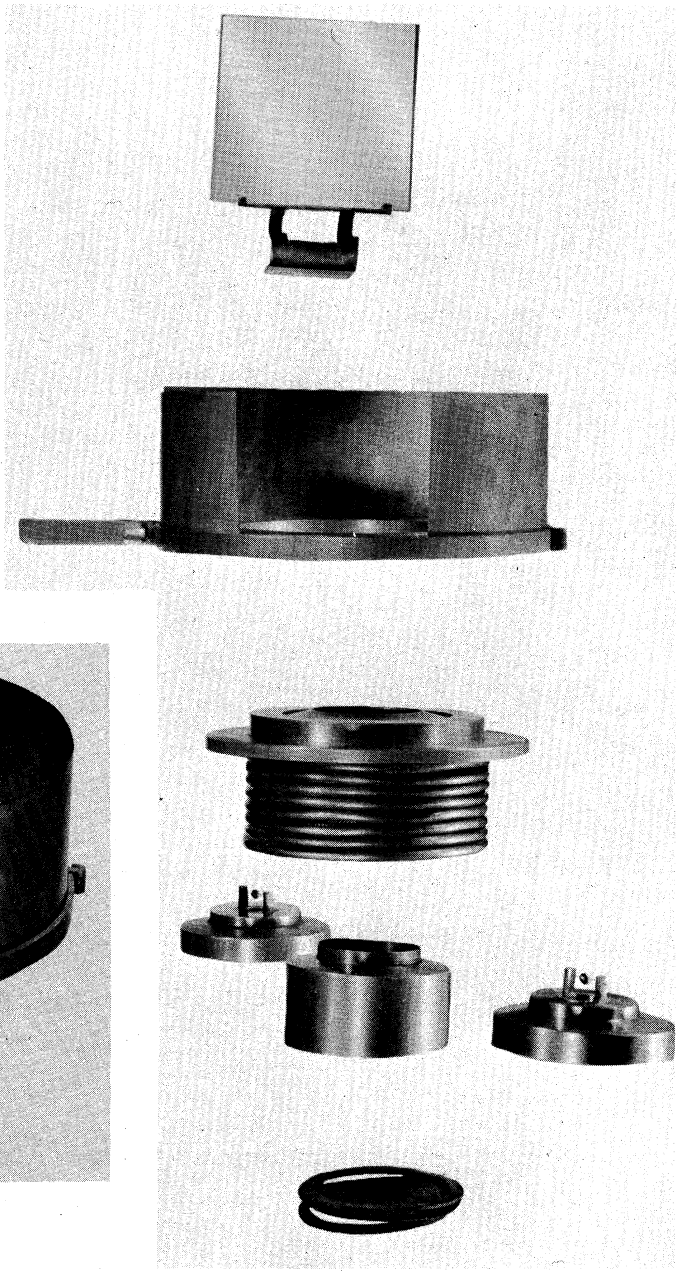
The radioactive isotope selected for the initial tests is palladium-109 in a palladium foil .001 inch thick, 6-1/4 inches long, and 1-3/4 inches wide. Two brass mounts were fabricated to hold the palladium foil about the two types of flames studied. The mounts consist of circular rings and circular hinged clamps to hold the foil about the flame. The foil is backed up with brass shim stock .005 inch thick. For the experiment using the Bunsen-type flame, a window made of Vicor glass is used, through which the flame is viewed. The window is necessary to eliminate the unsteady burning resulting from the asymmetrical arrangement of the palladium foil and shim stock about the flame. The window is not needed in the experiment with the blow-off of a V-type flame.

Fig. 18a is a photograph of the assembly used for the Bunsen-type flame, while Fig. 18b shows the exploded view. Fig. 19a is the assembly used for the V-type flame and Fig. 19b shows the exploded view.

The fuel used for the experiments was commercially pure propane, a fuel closely resembling the type of hydrocarbon fuel used in jet engines. For the



(a) Assembled View

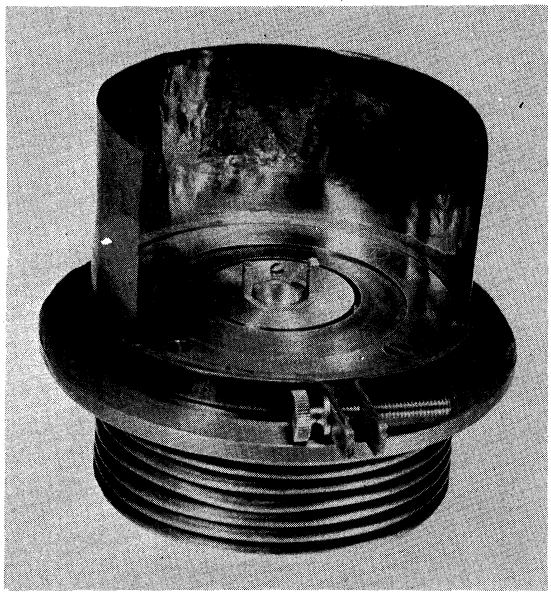


(b) Exploded View

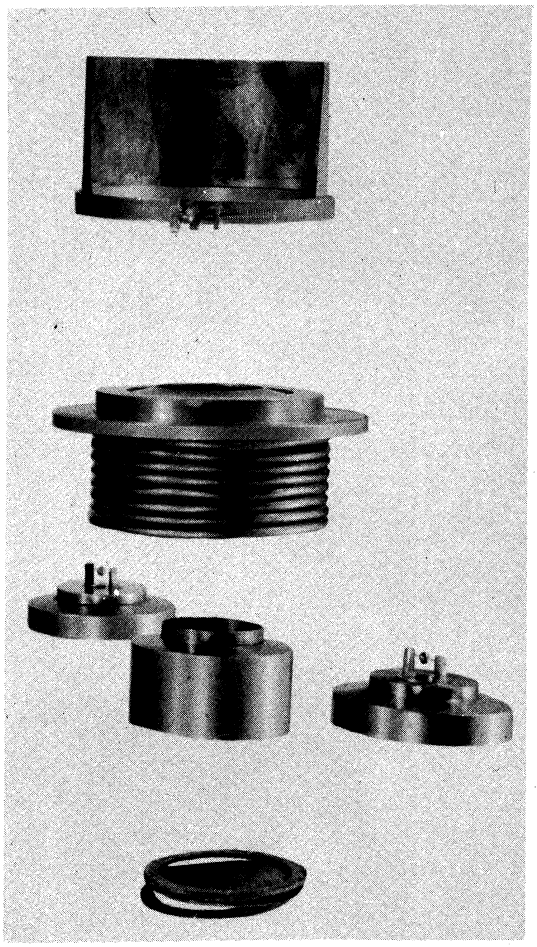
Fig. 18a and 18b. Photograph of 1-1/4-inch-Diameter Bunsen Burner and Palladium Foil Attachments

blow-off velocity experiments, mixture ratios of .067, .080, and .100 lb propane/lb air were used. From previous experiments reported in University of Michigan External Memorandum 74[1], maximum blow-off velocities were obtained with mixture ratios of .080 lb propane/lb air while lower blow-off velocities were obtained with richer and leaner mixtures.

[1]University of Michigan, External Memorandum 74, September 1950. USAF Contract W33-038-ac-21100, Project MX-772, "Blowoff Velocities of Spherical Flameholders", by A. Weir, Jr., D. E. Rogers, and R. E. Cullen



(a) Assembled View



(b) Exploded View

Fig. 19a and 19b. Photograph of 3/8-Inch-Diameter Nozzle and Spherical Flame-holder with Palladium Foil Attachments

In all cases experiments were also performed with an unactivated palladium foil to be used as a datum for comparison with the experiments made with the activated foil.

c) Experimental tests with radioactive palladium foil. The activated palladium foil was taken from the Chalk River Pile in Ottawa, Canada, at about 10 A.M. on January 14, 1952, and did not reach the laboratory until 6 P. M. on January 15, 1952, because of poor flying conditions. The Bunsen-flame experiment was performed first, the foil having an average activity of 6.6 curies during that experiment. Next, the blow-off velocity experiments with the three different fuel-air mixtures of 0.067, 0.080, and 0.100 lb propane/lb air were performed in that order with the average foil activities of 5.6, 5.3, and 4.9 curies, respectively.

d) Experimental results. The results of these tests are shown in Figs. 20 through 23. They indicate that there is no measurable increase in the normal flame-propagation velocities and blow-off velocities at any of the pressures at which flames were studied. It should be stated that the differences in the two curves in Fig. 20 are all within the range of experimental variation from run to run. Especially at low pressures, where the differences in the two curves are more pronounced, the location of the exact extent of the luminous flame surface is inaccurate because of the diffuse appearance of the reaction zone at those pressures. With the blow-off velocity experiments of Figs. 21, 22, and 23, the data show no appreciable difference so that only one curve was faired through the points.

Figs. 24 and 25 show photographs of typical flames burning at reduced pressure. Figure 24, taken from University of Michigan External Memorandum No. 81[1], shows the effect of pressure on the shape and reaction zone thickness of ethylene-air Bunsen flames burning at various reduced pressures. Fig. 25, taken from University of Michigan External Memorandum No. 742, shows the effect of pressure on a typical inverted V-type flame stabilized by a spherical flameholder. The appearance of the flames in Figs. 24 and 25 are quite similar to the flames observed in this study.

e) Discussion. From the free-radical theory of flame propagation, it would be expected that the formation of free radicals such as atomic hydrogen and the OH radical are important to the mechanism of flame propagation. It is believed that these radicals formed in the reaction zone diffuse into the unburned mixture, raising the energy level of the molecules entering the reaction zone. If the gas entering the reaction zone is highly ionized, this effect could be increased, causing an increase in the normal propagation velocity of the flame. Furthermore, it is generally true that there is a functional relationship between blow-off velocities of flames and propagation velocities of flames. Fuels with greater normal flame speeds also have greater blow-off velocities. Similarly, increasing the temperature of the unburned gases causes increases in both flame speeds and blow-off velocities. It is apparent from the results that the beta radiation from the palladium foil caused no measurable increase in the flame speeds or blow-off velocities. Had the activity of the palladium foil been higher, the unburned gas would possibly have been more highly ionized and an effect would be noticed.

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[1]University of Michigan External Memorandum No. 81, December 1950. USAF Contract W33-038-ac-21100, Project MX-833, "The Effect of Pressure on the Propagation Rate of Bunsen Flames in Propane-Air and Ethylene-Air Mixtures", by R. E. Cullen.

[2]University of Michigan External Memorandum No. 74, September 1950. USAF Contract W33-038-ac-21100, Project MX-772, "Blowoff Velocities of Spherical Flameholders", by A. Weir, Jr., D. E. Rogers, and R. E. Cullen.

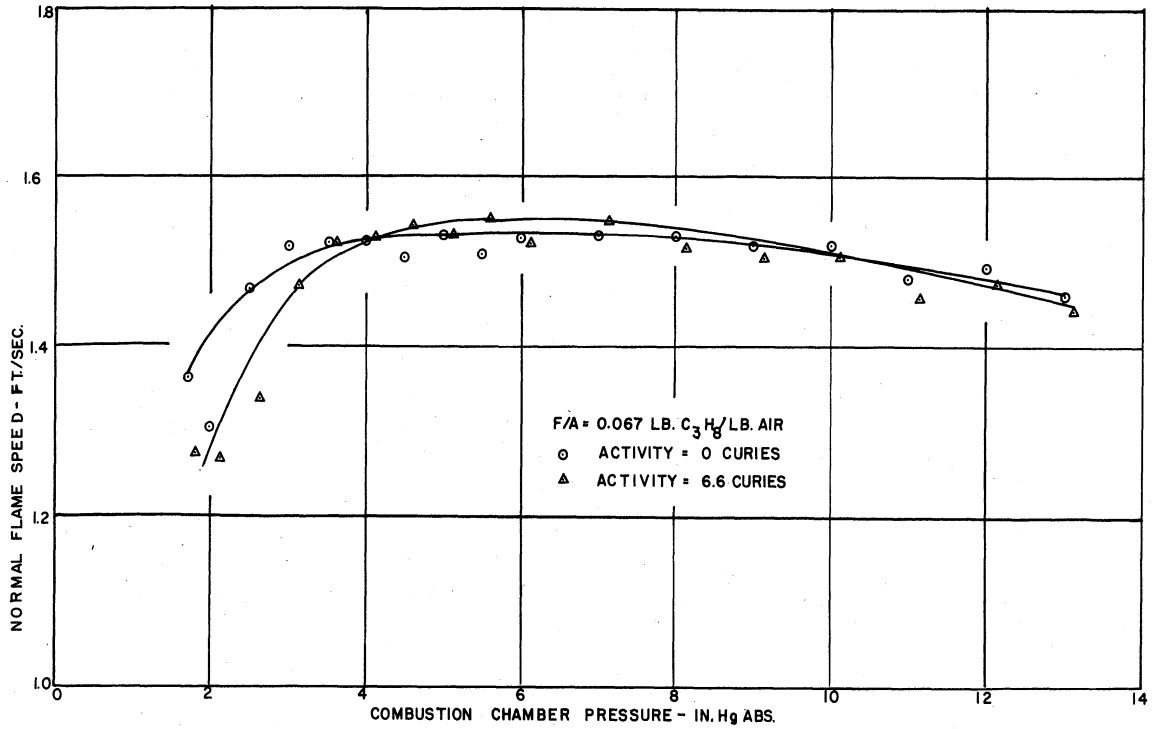


Fig. 20. Propane-Air Flame Propagation Experiments (Bunsen Burner Diameter = 1.25 inch. 1 mil Palladium Foil Mounted as Shown in Fig. 5).

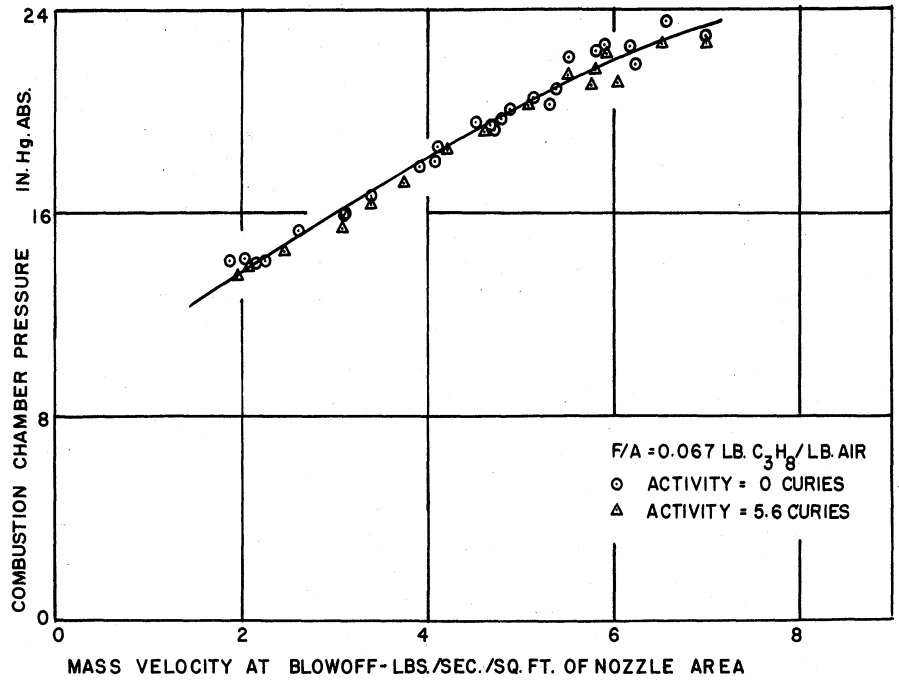


Fig. 21. Propane-Air Flame Stabilization Experiments (1/8" dia Steel Spherical Flameholder Located 3/16" above 3/8" dia Nozzle. 1-mil Palladium Foil Mounted around Flame as Shown in Fig. 6).

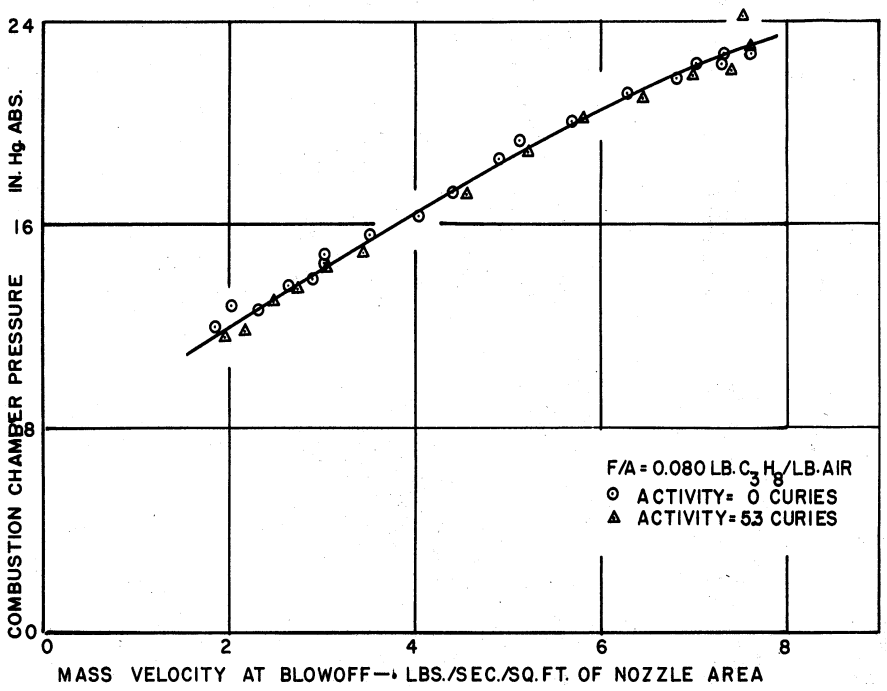


Fig. 22. Propane-Air Flame Stabilization Experiments (1/8" dia Steel Spherical Flameholder Located 3/16" above 3/8" dia Nozzle. 1-mil Palladium Foil Mounted around Flame as Shown in Fig. 6).

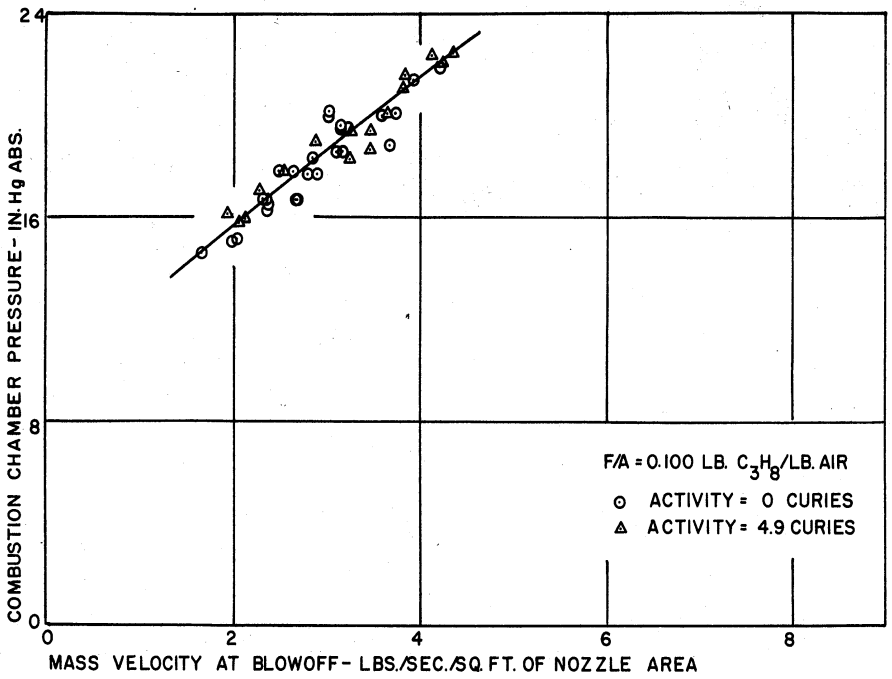
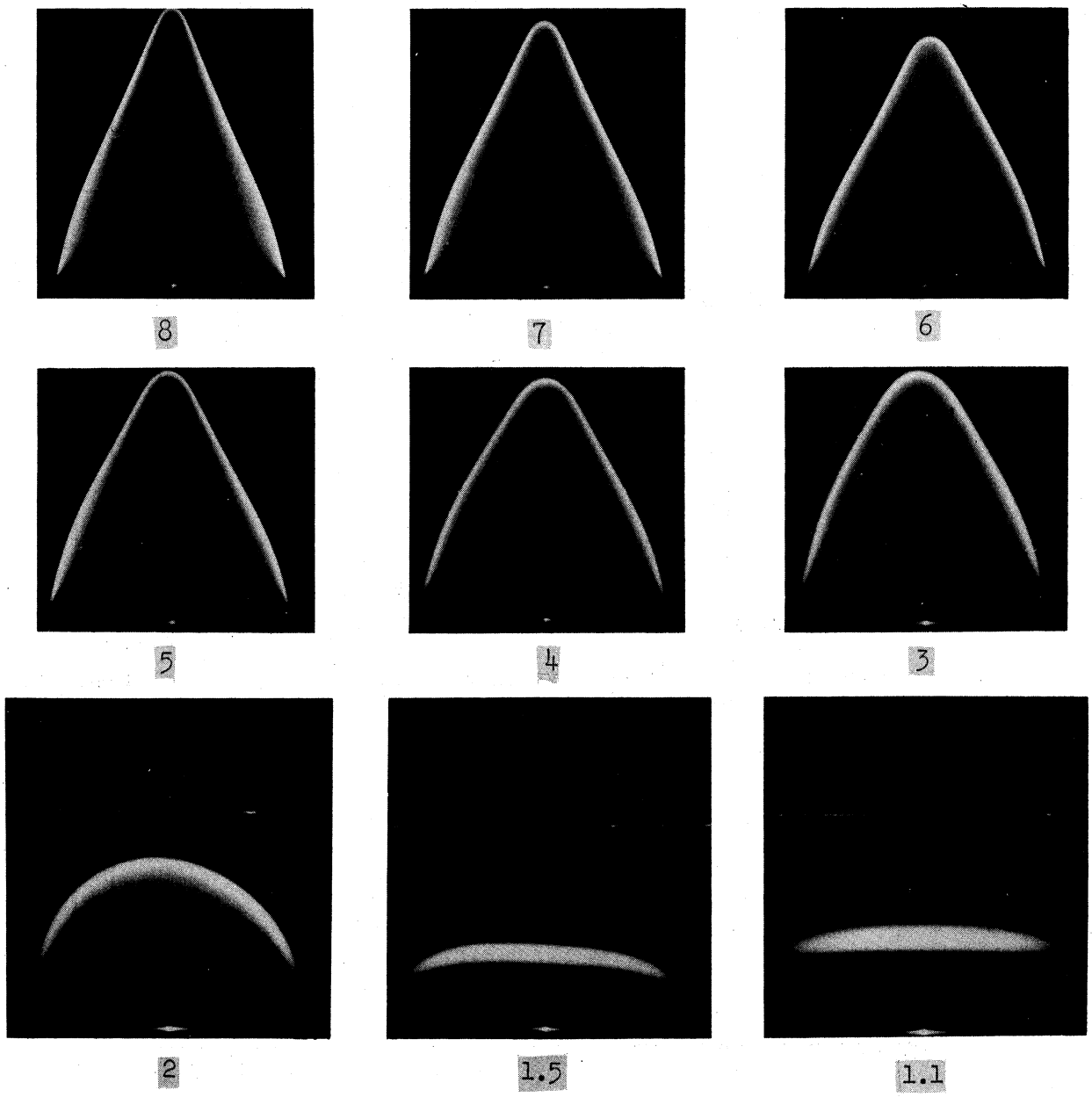


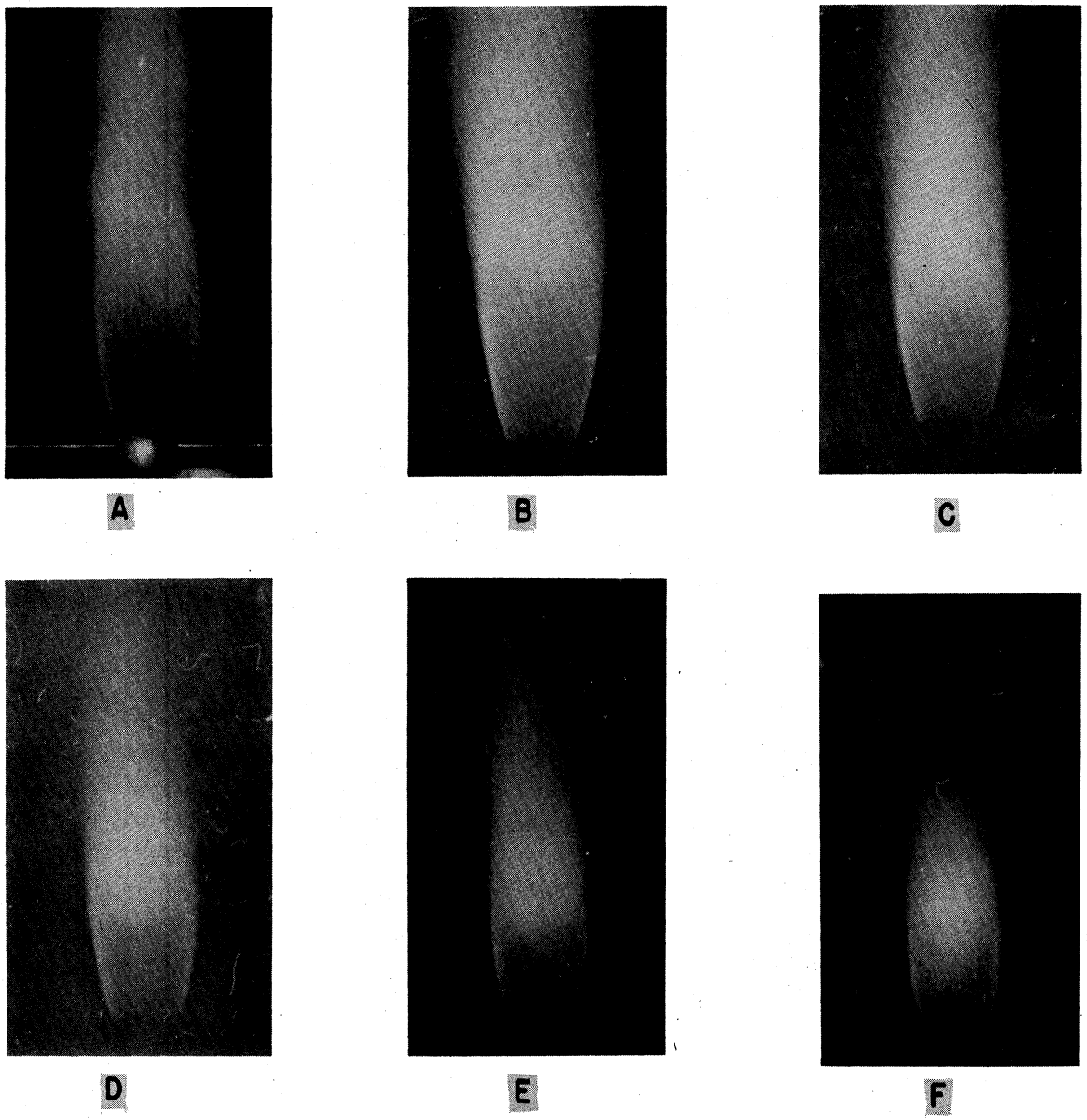
Fig. 23. Propane-Air Flame Stabilization Experiments (1/8" dia Steel Spherical Flameholder Located 3/16" above 3/8" dia Nozzle. 1-mil Palladium Foil Mounted around Flame as Shown in Fig. 6).



Ethylene-Air Bunsen Flames at Reduced Pressures, with the 1-1/4 Inch Burner; Fuel Air Ratio 0.0836 lb Ethylene/lb Air; Diameter of Burner Exit 1-1/4 Inch; Numbers under Photographs Indicate Pressure in Inches of Hg Absolute.

Fig. 24. Photographs of Typical Bunsen Flames at Reduced Pressures (Taken from University of Michigan External Memorandum 81)

f) Future program. The next experiment to be performed will again utilize palladium irradiated at Chalk River. The length of the foil will be increased to about 10-1/2 inches. In addition, an activated palladium sphere will be used to stabilize the flame. Perhaps the close proximity of the flame



Fuel-Air Ratio = 0.080; Nozzle Diameter = 3/8"; Sphere Diameter = 3/16"; Photograph No. A, B, C, D, E; Abs. Pressure-mm of Hg 680, 465, 412, 359, 333, 307.

Fig. 25. Photographs of Typical V-Type Flows Stabilized on a Spherical Flameholder and Burning at Various Reduced Pressures (Taken from University of Michigan External Memorandum No. 74)

to the radioactive sphere in conjunction with the surrounding foil will prove a more effective means of ionizing the gases. Also the activity of the foil will be increased by a factor of at least four if the shipment is not delayed for two half-lives as in the case of the first shipment.

PART II

THE EFFECT OF RADIATION ON CHEMICAL REACTIONS (SUBPROJECT M943-C)

Personnel

Subproject Supervisors: J. J. Martin, Associate Professor of Chemical Engineering; L. C. Anderson, Professor of Chemistry and Chairman of Department. D. E. Harmer, Research Assistant; J. G. Lewis, Research Assistant.

A. Introduction

The basic objective of this work was stated in Progress Report I; namely, to study the promotion of chemical reactions by the use of radioactive fission products. In the course of the study answers to two questions are being sought: When does radiation accelerate the rate of approach to thermodynamic equilibrium in a chemical reaction or cause a displacement of the equilibrium point, and how does the extent of the reaction vary with the quantity of radiation? When the answers to these questions have been found, it is believed it will be merely an application or developmental procedure to determine whether radiation from fission products can be effectively and economically utilized in the promotion of reactions of industrial importance.

Among the reactions which have been shown by previous investigators to be affected by radioactivity are the synthesis of ammonia, polymerization of some liquids, and dehydrogenation of some hydrocarbon gases. These reactions have, for the most part, been carried out under the influence of alpha and beta radiation. Comparatively little has been published about the effect of gamma radiation. Since fission products yield considerable amounts of gamma radiation and since the project has had available for the past several months a 1000-curie, cobalt-60 gamma source prepared at Brookhaven National Laboratory, a number of experiments have been conducted using this source of radiation. Also as this report is being written, a small piece of palladium foil containing 3 curies of Pd-109 has been received after irradiation at Chalk River, and some reactions have been studied under the influence of the 1 Mev beta radiation from this source.

B. Experimental Results

The first reactions to be studied under the influence of gamma radiation were conducted in small 9 mm by 20 cm pyrex tubes. The procedure was simply to seal the chemical reactants in the tubes and place them in the cobalt-60 vault for approximately 24 hours. At the end of this period the tubes were removed for observation and tested for any changes that might have taken place.

In the case of a number of hydrocarbon gases, including isobutane, butene-2, ethylene, butane, isobutylene and acetylene, the analysis involved breaking the sealed tubes in a closed system and observing the pressure change which resulted. It was found that none of these hydrocarbon gases originally sealed at atmospheric pressure and room temperature underwent any appreciable change in pressure, and this fact has been interpreted to mean that no reaction occurred.

In the case of a tube of liquid butane which was examined after 24 hours of gamma radiation, an appreciable amount of unsaturated gas was found by absorption in sulfuric acid and the presence of hydrogen was confirmed by combustion with copper oxide. This result indicated that significant decomposition had occurred. Since gaseous butane gave no reaction, it appears that the ability of the material to absorb and utilize radiation depends upon the density of the material, as would be expected.

Stoichiometric mixtures of nitrogen and hydrogen charged to the small tubes showed no measurable change after being exposed to the cobalt-60 source for 24 hours. However, in this case, as well as in the case of the hydrocarbon gases, the amount of reactant charged was very small and detection of any reaction was not considered very accurate. Therefore, large pyrex tubes of approximately 100 ml capacity were fitted with stopcocks and charged with nitrogen and hydrogen for further studies. Some of these samples were subjected to 24 hours of gamma radiation from the cobalt-60, and some of them were placed 6 inches from the target of a 200 KVP X-ray machine operating at 20 ma and 155 KVP for 3 hours. After radiation the gases in these large tubes were drawn through Nessler's test reagent for ammonia, which is capable of detecting a small fraction of one per cent of ammonia in the sample. In no case was any appreciable amount of ammonia formed. These experiments included tests in which nitrogen and hydrogen were dried before charging and also tests in which these gases were saturated with water vapor before charging.

In the next set of experiments sample tubes were constructed which had glass stopcocks at one end and ground-glass-joint caps at the other end. The caps permitted charging solid catalyst, or any other solid, into the tubes (see Fig. 26, page 33). A doubly promoted ammonia catalyst was obtained from the Pennsylvania Salt Mfg. Co. and prepared by reduction in hydrogen at 450° to

500°C. This reduced catalyst was transferred under nitrogen to the reaction tubes which were then evacuated and filled with a stoichiometric mixture of nitrogen and hydrogen. Using both dry and wet reactant gases, no appreciable amount of ammonia was detected by Nessler's test after these samples had the usual 24 hours in the cobalt-60 vault. It was noted in these experiments that the catalyst tended to adsorb gases very readily since it was quite porous. It is conceivable that very small amounts of ammonia might have remained on the catalyst, though the consistently negative results seem to indicate absence of ammonia. Because of the necessity of using stopcock grease on the ground-glass joints, the reaction tube with the catalyst could not be heated to very high temperatures after radiation to assure the liberation of minute amounts of ammonia.

In an attempt to determine whether gamma radiation might be favoring the decomposition of ammonia rather than its synthesis, a sample tube was charged with pure ammonia. Analysis was made on an Orsat-type gas analyzer using 10 per cent sulfuric acid as the absorber for ammonia gas. After 24 hours of gamma radiation no appreciable amount of unabsorbed gas remained, indicating no decomposition of ammonia. Experiments are now in progress to determine whether the presence of a catalyst might have any effect on the decomposition of ammonia when irradiated with gamma rays at room conditions.

It might be pointed out that the results of thermodynamic calculations for the system nitrogen, hydrogen, and ammonia at room temperature and atmospheric pressure, as given later in this report, show that equilibrium in the absence of radiation greatly favors ammonia. Therefore, if any appreciable decomposition is to be found, the equilibrium point must change. Furthermore, it seems probable that if decomposition does take place, it will vary directly with the quantity of radiation supplied, because the radiation must satisfy the positive free-energy-change requirement of the reaction. On the other hand, the extent of the synthesis reaction to make ammonia might well be independent of the quantity of radiation once the reaction is started, for the negative free-energy change forms an ideal setting for the initiation of a chain reaction.

In another experiment on the radiation of a liquid with gamma rays, it was found that a sample of acrylonitrile (practical grade) in a sealed tube formed many small nuclei of solid polymer during the first part of the irradiation; and at the end of 24 hours of irradiation, a hard white solid was obtained.

Monomeric styrene showed evidence of polymerization by a gradual increase in viscosity during irradiation. The viscosity continued to increase after the irradiated styrene was removed for the cobalt-60 vault while unirradiated check samples remained unchanged. Results obtained up to the time of this writing have not been consistent enough to warrant any general conclusions, although the viscosity increase after radiation seems to indicate that the long induction period ususlly found in thermal polymerization of plastics may be shortened in the presence of gamma radiation. Further experiments have been planned.

Since it is known from the literature that beta particle energy is more readily absorbed than high energy photon radiation, an investigation was started to find out whether the electrons produced by the Compton effect might be used as a means of imparting energy to chemical reactants. An increase in count was observed with Geiger counters when thin metal sheets were placed in front of the counter windows and gamma radiation passed parallel to the surface of the window. As an application of these observations, nitrogen and hydrogen were charged to reaction tubes containing copper gauze or brass sheets or aluminum dust. Although no ammonia was detected after the usual 24-hour gamma radiation from cobalt-60, it is felt that the possibility of using these secondary emissions should be investigated further, especially at higher pressures where absorption of the emitted electrons would be nearly complete. It is also planned to investigate other types of secondary radiation, particularly that which emanates from materials which fluoresce or phosphoresce when activated by beta or gamma rays.

More recent experiments involving beta radiation directly have not given any positive results. In one experiment nitrogen and hydrogen were charged to a two-liter flask (Fig. 27) along with a piece of palladium foil containing

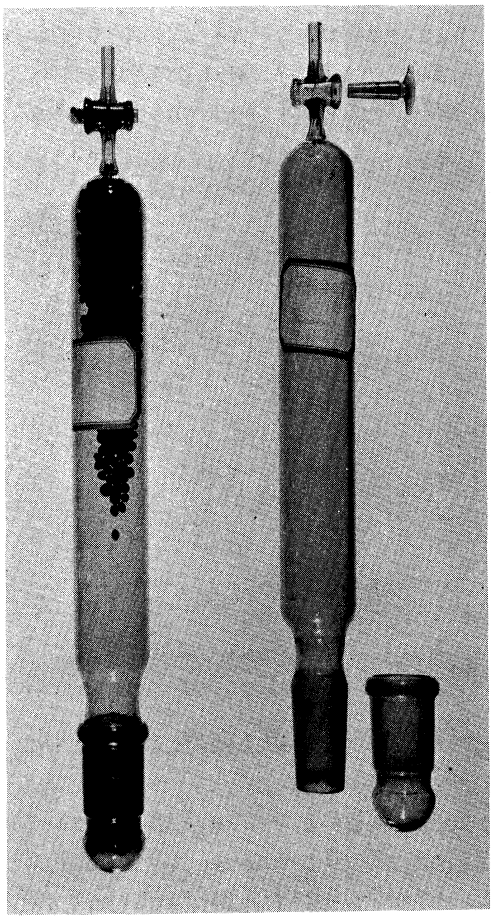


Fig. 26. Pyrex Glass Tubes for Irradiation of Gases at Low Pressure. Tube on the left is charged with a solid ammonia catalyst.

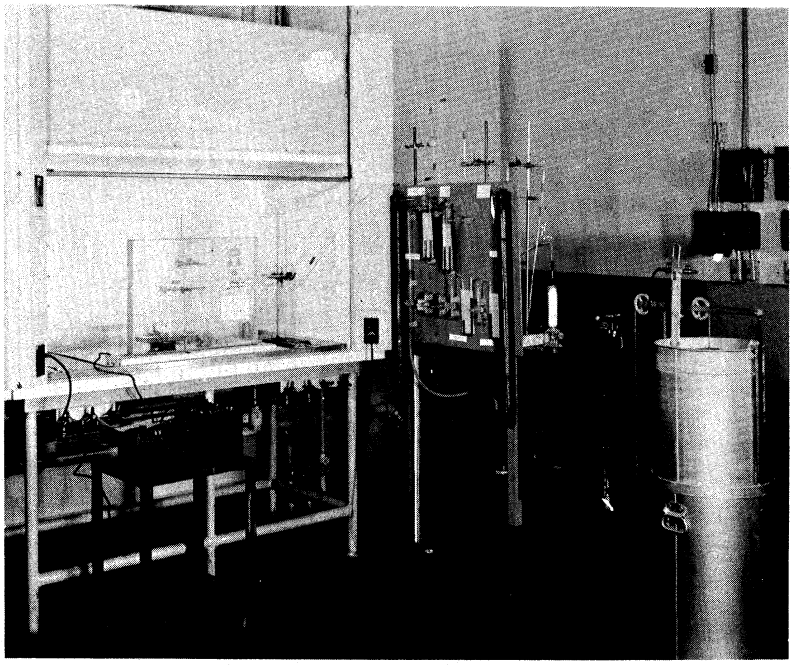


Fig. 27. Low-Pressure System for Gases in the Presence of Radioactive Catalysts. Flask behind lucite shield contains palladium foil with a small percent of Pd-109.

some palladium-109, whose total activity was about 3 curies. At the end of about 12 hours no ammonia could be detected by Nessler's test. In another experiment a stainless steel reactor tube (Fig. 28) was charged with the palladium foil

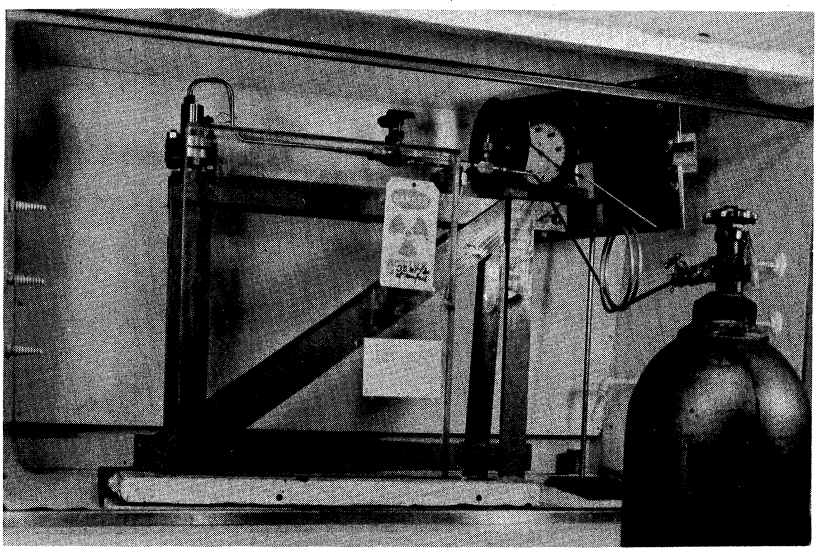


Fig. 28. Batch Reaction System Utilizing High-Pressure Reactor.

whose activity was now approximately 1 curie, and a stoichiometric mixture of nitrogen and hydrogen was added at a pressure of about 55 atmospheres. At the end of about 11 hours the Nessler test indicated that no appreciable amount of ammonia had been produced at room temperature by the beta radiation. This is the first experiment to be conducted at a pressure higher than atmospheric, and considerable confirmation and extension of this work remains to be done.

C. Design and Construction of New Equipment

The preliminary experiments to date indicate that high density of the chemical reactants is most favorable to the promotion of a reaction by radiation. This means that, in the case of gases, high pressures appear to give the greatest promise, both because of greater absorption of radiation and because of the increased conversion at equilibrium in many systems.

As an example of a reaction in which elevated pressure increases the normal equilibrium percentage of desired product, the ammonia synthesis is considered in detail. The equilibrium molar percentages of ammonia present in a reaction mass originally consisting of nitrogen and hydrogen in the stoichiometric ratio are given in Table IV.

TABLE IV

THEORETICAL PERCENTAGES OF AMMONIA IN A SYSTEM INITIALLY CONSISTING OF A STOICHIOMETRIC MIXTURE OF NITROGEN AND HYDROGEN
(Results of Thermodynamic Calculations)

	Temperature (°F)	Pressure (psia)	Percentage of Ammonia at Equilibrium
1	70	147	100-
2	70	1500	100-
3	450	14.7	35.9
4	450	1500	72.1
5	932	4400	26.4

(Data from Curtis,[1] Dodge,[2] and Hougen and Watson[3].

Lines 1 through 4 represent conditions of temperature and pressure attainable in the apparatus which will soon be available in this laboratory. Line 5 represents conditions approximating those in industrial ammonia reactors. It is evident from Line 2 that it is desirable to operate an ammonia reactor at as high a pressure and low a temperature as possible in order to obtain the maximum conversion to desired product at equilibrium. However, if the usual catalysts are employed for the reaction, the rate of reaction decreases with

1 Curtis, H. A., Fixed Nitrogen. New York, Reinhold (1932).
2 Dodge, B. F., Chemical Engineering Thermodynamics. New York, McGraw-Hill (1944).
3 Hougen, O. A., and Watson, K. M., Chemical Process Principles. New York, Wiley (1947).

temperature decrease in such a way that at room temperature there is no conversion whatsoever. Now it seems possible that radiation may speed up the reaction and permit lower operating temperatures either with or without catalyst. Similar remarks could be made for the methanol synthesis and for some other reactions of commercial importance.

By constructing a reaction system in which both temperature and pressure can be varied over considerable ranges, it is thought that much more information can be secured concerning the kinetics and equilibria of the systems to be investigated. In addition, it is possible that some reactions which would escape detection if carried out under milder conditions may be detected by operation under severe conditions.

The experimental reaction system being constructed is centered around a high temperature, high pressure reaction vessel. Fig. 29 is a photograph of the completed vessel, and Fig. 30 is a working drawing for the construction of the

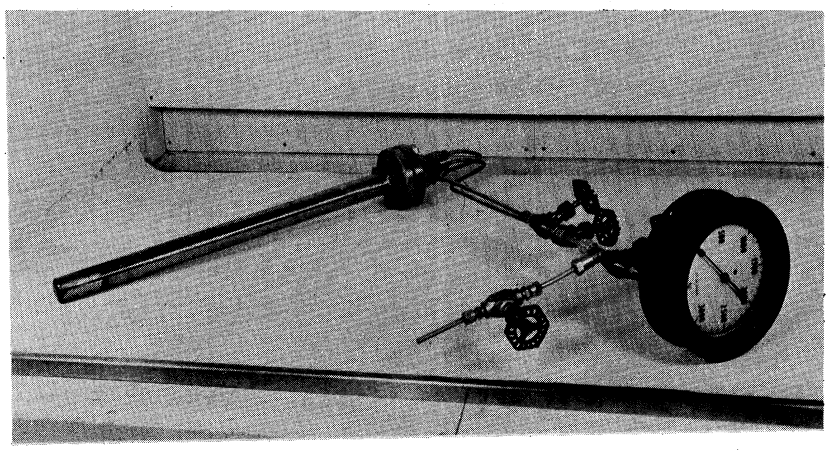


Fig. 29. Stainless-Steel Reactor and Auxiliary Fittings for Use with Gases at High Pressure.

vessel. Accessory equipment will be provided to permit the reactor to be operated either batch-wise or as a part of a continuous-flow reaction system, as shown on the flow sheet in Fig. 31. The reactor itself was designed according to the ASME code to operate at maximum conditions of 2000 psi and 650°F and was given a hydrostatic test at 2800 psi and room temperature. The reactor is constructed of stainless steel, and fits snugly into the 1-1/2-inch diameter access opening in the cobalt-60 vault. It will be necessary to limit the temperature of the outside of the reactor to approximately 350°F in order to avoid damage to the vault. It is thought that higher temperatures might cause warping of the aluminum case enclosing the cobalt or warping of the stainless-steel inner shell of the vault, or might even cause softening of the lead shielding. Provisions must be made for cooling the chemicals coming from the reactor since the accessory supply and control tubing will be of aluminum. The accessory equipment may be operated at 2000

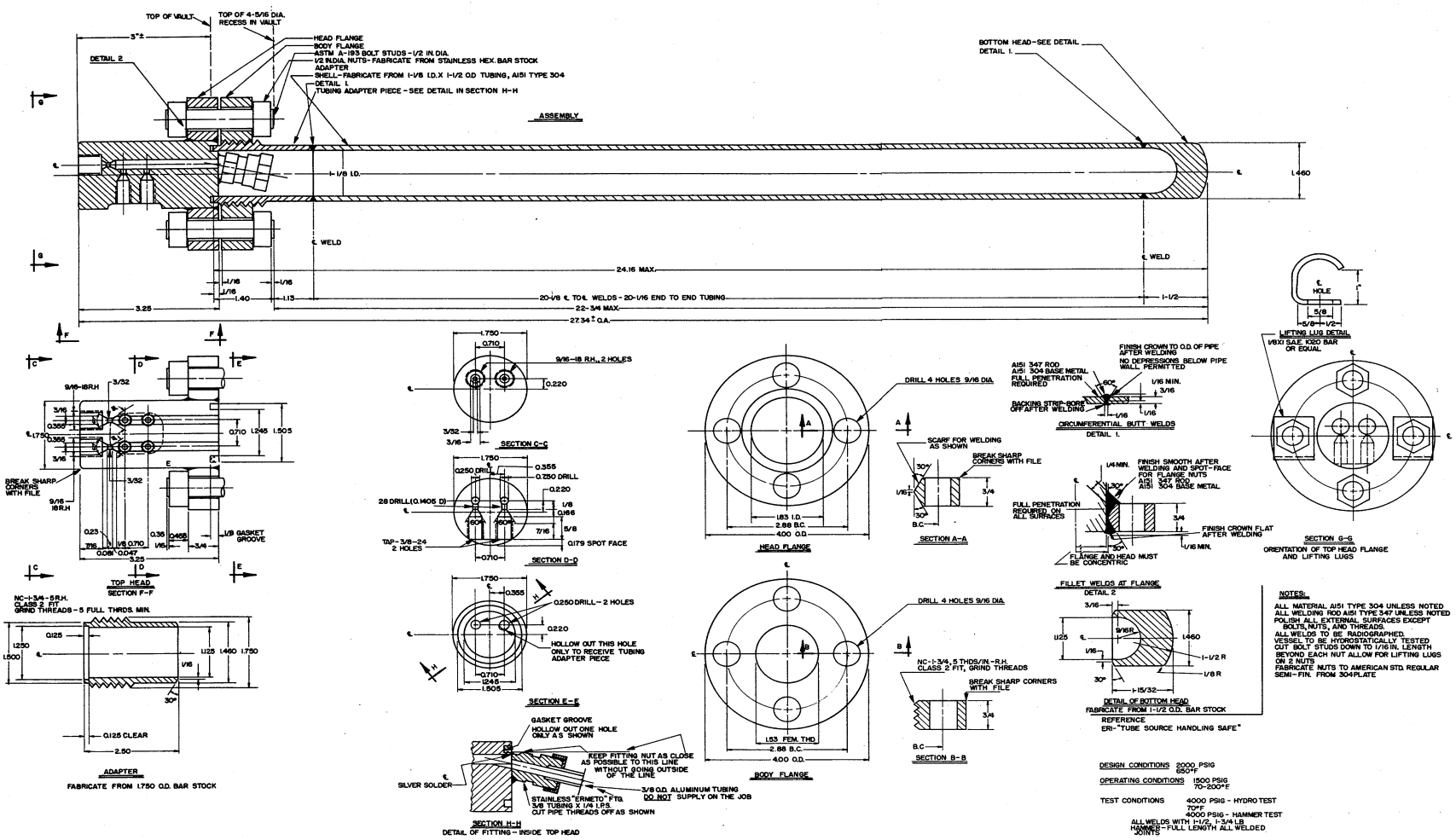


Fig. 30. Working Drawing for the Construction of the Reactor

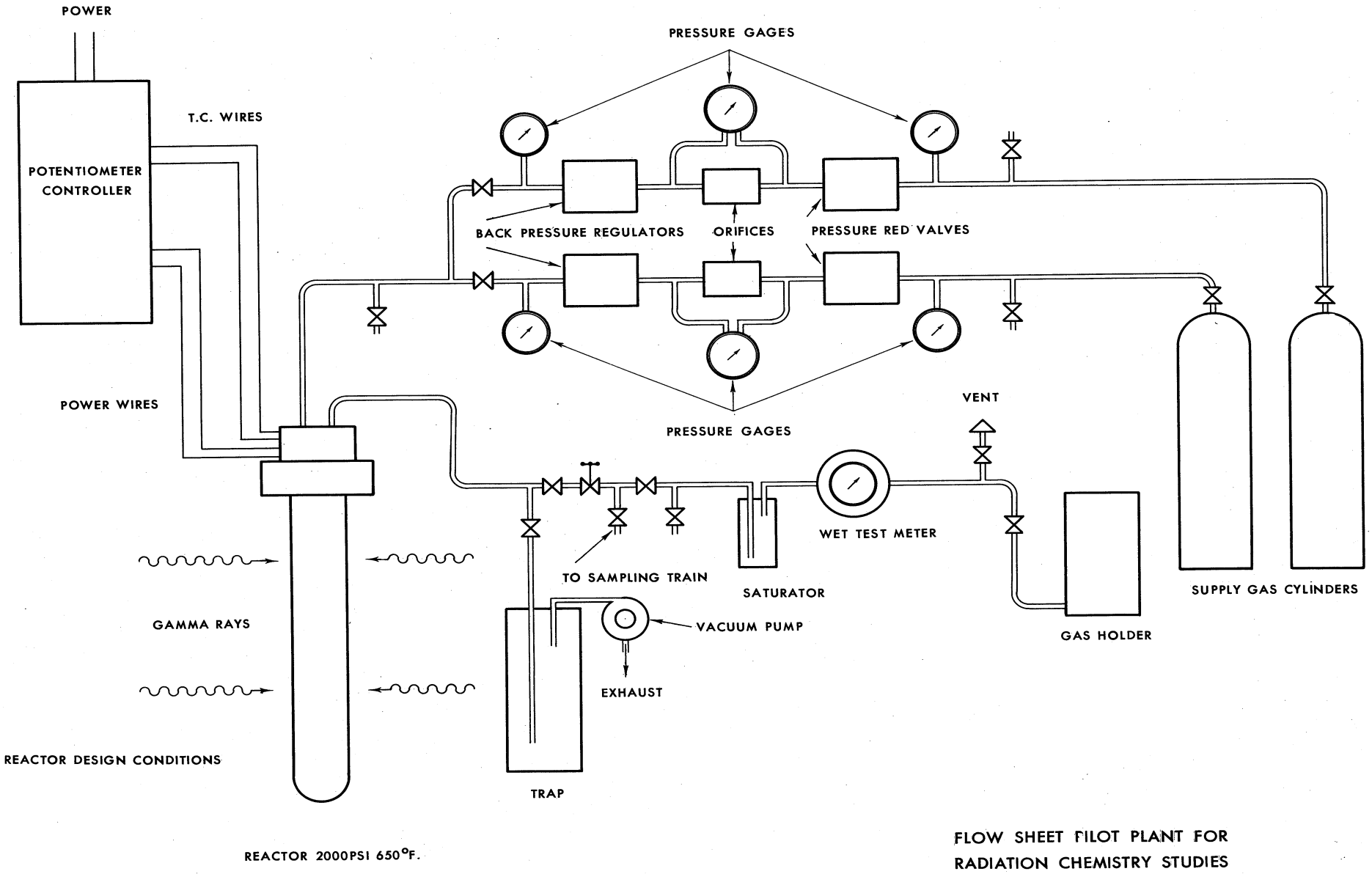


Fig. 31. Flow Diagram for a Continuous System Employing the High-Pressure Stainless-Steel Reactor

psi at room temperature, with the exception of the sampling, analytical and low-pressure metering apparatus. These three types of equipment will be operated at atmospheric pressure.

Fig. 32 shows the internal fittings for the reactor, which are patterned after some types of commercial reactors. The apparatus is designed in such a way that it may readily be removed from the reaction vessel shown in Fig. 30. Internal heat exchangers are provided in the form of baffles, so that incoming gases may cool the walls of the pressure vessel. A thermal radiation shield may be added if necessary in order to maintain the desired temperature in the interior and in order to prevent thermal radiation from the electric heater element from overheating the wall of the reactor. The heating element will be wrapped around the constriction just above the catalyst holder. Spiral countercurrent heat exchangers are provided to exchange the heat between the incoming and outgoing streams.

It may be discovered that the systems being investigated are so transparent to gamma radiation that little use is made of the energy of the radiation even at higher pressures. Since electrons in passing through a given thickness of material will lose a much higher percentage of their energy than will gamma rays, it may prove desirable to cause the gamma rays to eject electrons from a suitable material placed in the reacting system. If the rate of chemical reaction depends upon the rate of ionization, the rate of reaction should be increased by this secondary emission.

Fig. 33 is a drawing of a special packing which may be readily installed in or removed from the reactor vessel. This special packing consists of a series of washers placed parallel to the maximum component of the gamma radiation from the surrounding cylindrical source of cobalt-60. This packing should cause electrons produced in the washers by Compton-scattering of gamma rays to be dispersed in the reacting medium. The arrangement of the packing in washers was governed by the thought that a perferred orientation of the scattered electrons may exist and that this orientation may be different from the direction of the incident gamma radiation.

Although the reaction system described above was designed to fit into the arrangement of the existing cobalt-60 vault for studies using gamma radiation, it will be equally possible to use the entire system for studies in which a source of radioactivity is placed inside the reaction vessel itself. The latter arrangement would be particularly desirable when beta sources are used. The reaction vessel has now been completed and batch tests at room temperature will probably be conducted in this apparatus while construction on its internal fittings and the necessary equipment needed to study flow reactions is proceeding.

The greatest share of the materials needed to complete the construction of the experimental unit is now on hand, while most of the remainder of the materials should be secured within three months.

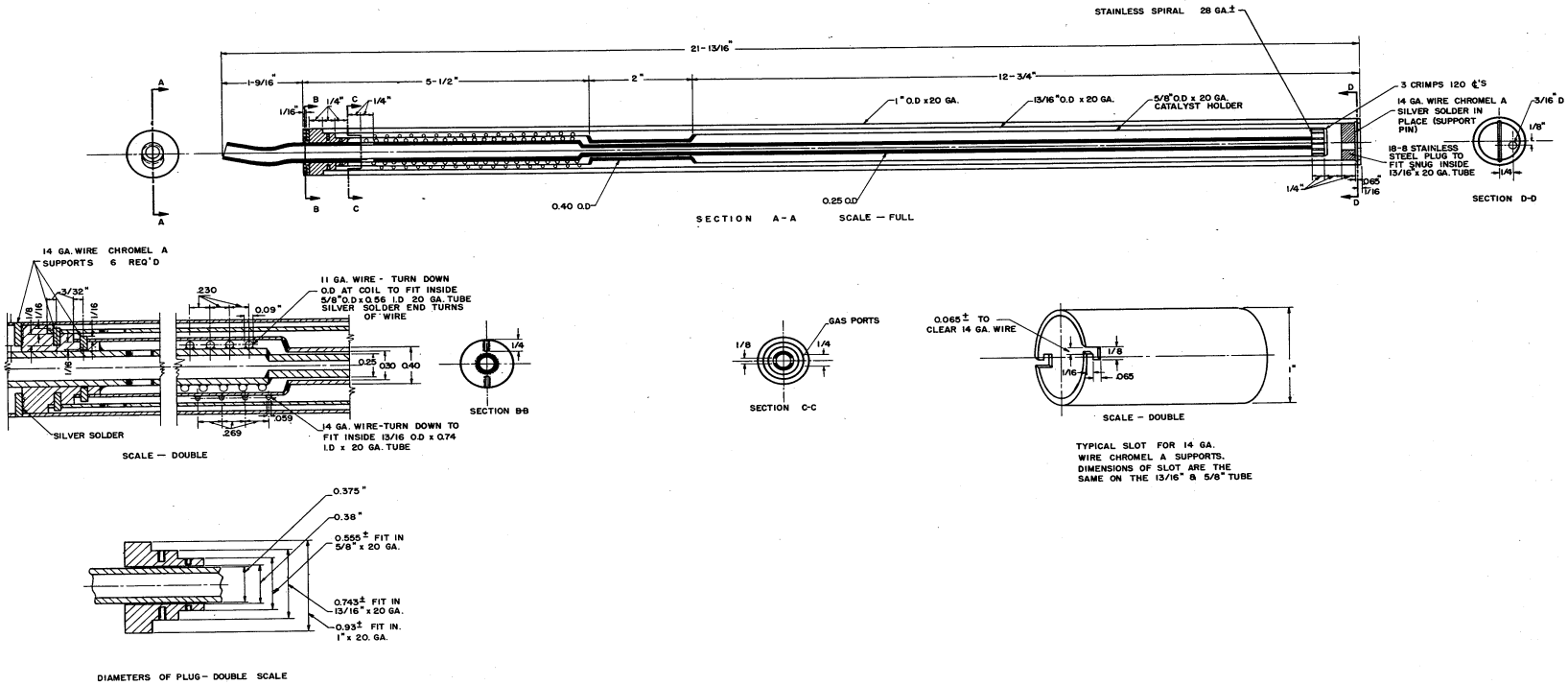


Fig. 32. Catalyst Holder for Reactor

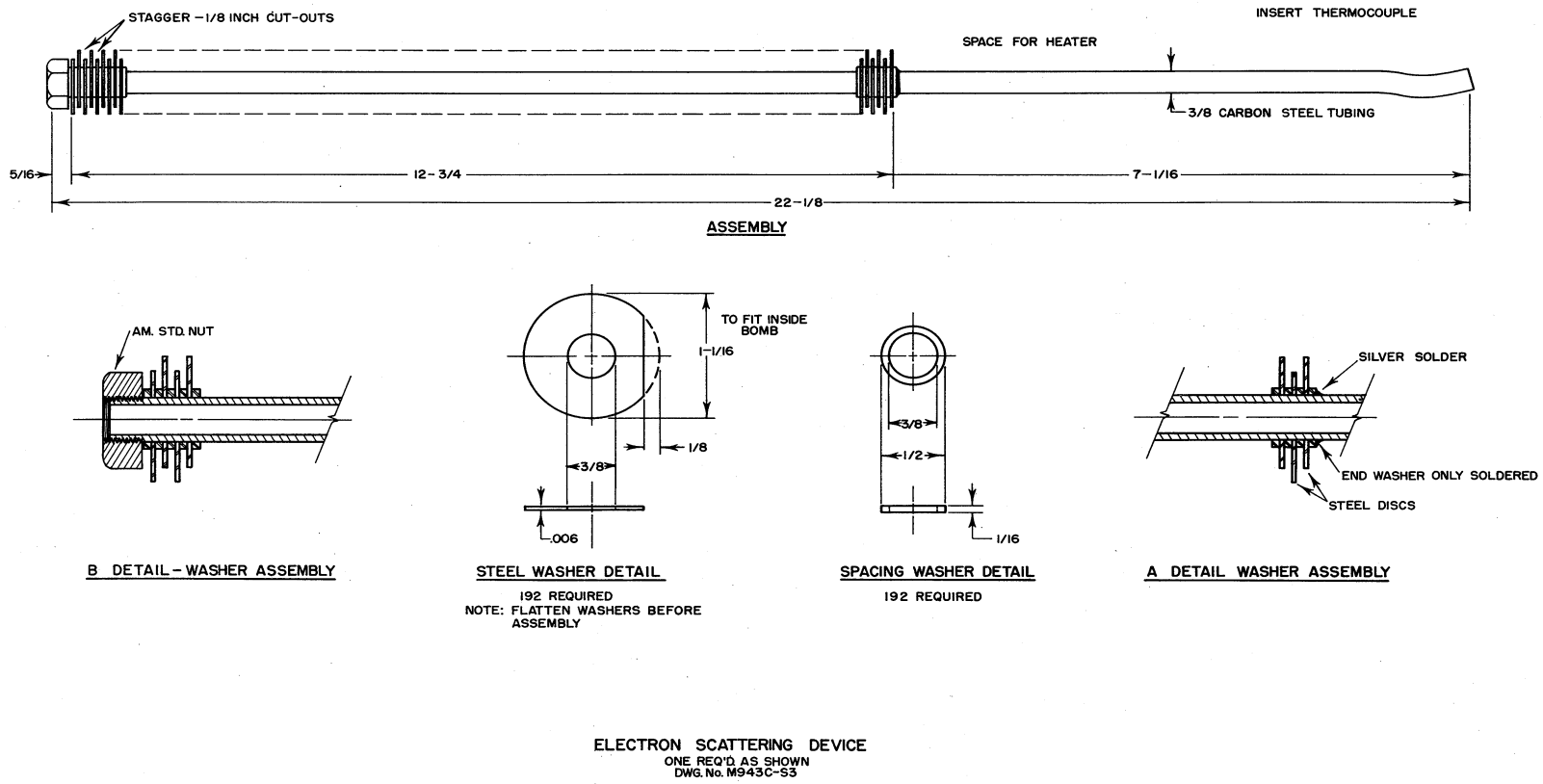


Fig. 33. Washer Packing for High-Pressure Reactor

D. Future Program

It is planned to continue the studies on the ammonia synthesis reaction, varying the temperature and pressure over wide ranges and using the new stainless-steel reactor. Since many commercial reactors operate at 4400 psia and 500°C, it is quite reasonable that no positive results were found at room temperature and atmospheric pressure. It is expected that studies at higher temperatures and pressures will prove more enlightening and will indicate the range of usefulness of beta and gamma radiation in promoting the ammonia synthesis, as well as other reactions, such as the methanol synthesis and Fischer-Tropsch reactions.

While the higher-pressure gas reactions are being studied, further work will be carried on with liquids at lower pressures. It is planned to study some systems in which chain reactions have been postulated or might be possible. The polymerization of liquids such as styrene and acrylonitrile are worthy of further study. Also some reactions between different liquids and solids, such as the Diels-Alder reaction will be investigated in this program.

Gamma radiation will continue to be tried for all reactions studied, and an increasing amount of beta radiation will be utilized. It is planned to try some P-32 as the beta source, as well as Pd-109. Both of these will be charged inside the reacting system, as their penetrating power is quite low. Secondary particle and photon radiation will also be studied thoroughly.

PART III. PRESERVATION OF FOODS AND DRUGS

A. SUBPROJECT M943-D PRESERVATION OF VARIOUS FOODS BY IRRADIATION

Personnel

Subproject Supervisor: L. E. Brownell, Associate Professor of Chemical and Metallurgical Engineering. H. S. Dombrowski, Research Assistant; M. E. Gluckstein, Research Assistant.

1. Introduction

The work previously reported in Progress Report I has been continued. Samples of many types of foods have been subjected to irradiation by 200 KVP x-rays and gamma rays from the 1000 curie cobalt-60 source. Preliminary studies have been made on the effect of preparation and packaging techniques, such as chemical pretreatment and vacuum and pressure packaging. Considerable data have been obtained on meat, milk, and peas. Orange juice, green beans, cottage cheese, spinach, apple juice, bananas, melons, and berries are some of the other foods investigated. Some data have been obtained on the irradiation of microorganisms, such as bacteria and molds, and on the irradiation of packaged pharmaceuticals. Short-term (five weeks) animal-feeding experiments with irradiated milk have been completed.

Research has been concentrated on materials which are believed to have the greatest possibility of being successfully processed by irradiation. At the present time, the cold sterilization of heat-sensitive drugs by gamma irradiation appears to be a process which might be commercially feasible in the near future.

2. Experimental Studies on the Irradiation of Foods

a) Procedures. Foods have been irradiated while exposed to air, after packaging in polyethylene (Visqueen*) film, and after sealing in glass tubes. Highly specialized or intricate procedures have been avoided.

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* Courtesy of Visking Corporation, Chicago, Illinois.

Polyethylene film formed into bags has been used to package samples of all products tested. This method of packaging is simple and rapid. The sample is placed in the polyethylene bag, excess air is pressed out and the open end is heat-sealed by fusion with a gas flame. Pressure applied to the liquid or air in the bag serves to test for leaks. The bags can not be heat-sterilized, because polyethylene softens and melts at temperatures above 200°F. Vacuum- and pressure-packaging techniques are not applicable because the film is quite permeable to air and other gases but is a good barrier to water vapor.

The use of nonporous, rigid-wall containers permits the use of vacuum and pressure packaging of liquid and solid samples. Glass test tubes were used as containers for this purpose. Test tubes of about 1-1/8" x 10" and 5/8" x 6" were used in this work. The tubes were usually sealed with a good grade of rubber stoppers. Some glass ampules have been used. They were made from 5/8" x 6" test tubes by drawing the top portion into a narrow neck. The ampules were sealed by melting the narrow glass neck after evacuation. In this way, a completely impervious seal was obtained. Such tubes were used for work in which the samples were frozen.

For samples that were evacuated or packaged under nitrogen, carbon dioxide, or other gas atmospheres, the following procedure was used: A short nipple of glass tubing was placed through a rubber stopper in the test tube. The nipple was connected by means of a short length of amber rubber tubing to a specially designed all-glass pressure-vacuum system. A rotary oil-filled vacuum pump was used to evacuate the system, and gas was supplied from commercial cylinders. Self-draining glass bulbs were placed in the line to prevent froth or foam from entering the pump. Two vapor traps were provided. Mercury manometers were used to read the pressure. A pressure approximately equal to the vapor pressure of water at 20°C is the minimum attainable with most foods, and therefore a manometer sufficed as a pressure-measuring device. Two glass stopcocks were used to control the rate of evacuation (see Fig. 34).

Vacuum-packed samples were prepared by evacuation to a constant manometer reading, and the rubber connection tube was sealed with a screw clamp. To ascertain that the air has been removed, the tube was held in the system at the reduced pressure for several minutes before the tube was sealed. This allowed the water vapor from the sample to "sweep" out the system.

Where a gaseous atmosphere other than air was desired, the procedure was initially the same. After the evacuation, however, the gas was introduced directly from the storage cylinder. The system was evacuated again and the process repeated. A total of three evacuations and refillings were used. In the final filling, the pressure was set at preselected values between the vapor pressure and two atmospheres. The valving arrangement permitted the lines from the gas cylinder to be evacuated between cycles. Where it appeared desirable to

A–ROTARY VACUUM PUMP
B–GAS CYLINDER
C¹–DEWAR FLASKS FILLED WITH DRY ICE,ACETONE
C²–DEWAR FLASKS FILLED WITH ICE WATER
D–VAPOR TRAPS
E–ABSOLUTE MANOMETER
F–DIFFERENTIAL MANOMETER
G–3-WAY GLASS STOPCOCK
H–SELF-DRAINING FOAM TRAPS
I–SAMPLE TUBE
J–SPARGING TUBE
(use with liquids only)
K–MICROMETRIC FLOW VALVE

COMBINATION PRESSURE
VACUUM SYSTEM
FOR PREPARING FOOD SAMPLES

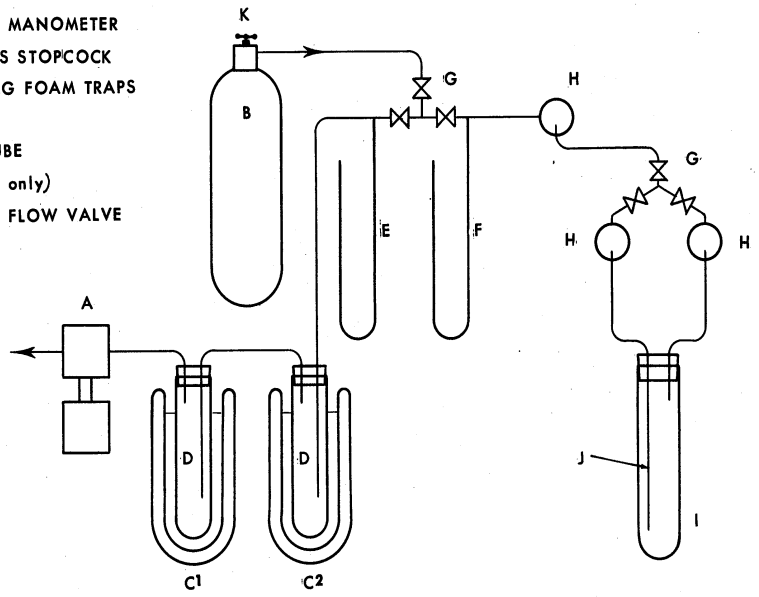

Fig. 34. Diagram of Apparatus for Evacuating Samples

saturate a liquid sample with the inert gas, a fritted glass sparging tube was used. This tube dispersed the gas in the sample in the form of fine bubbles.

To aid in the study of the effect of irradiation on frozen foods, a small-sized Dewar flask was constructed. Because of the size limitation of the cobalt-60 source (limiting dimensions,1-1/4" diameter x 12"), the largest sample tube that could be used in conjunction with the Dewar was a 5/16" test tube. About 15 cc of refrigerant was used to keep the contents of the tube frozen. By using acetone and dry ice (T = - 78°C) as the refrigerant, a sample of 15 cc of orange juice was kept frozen for more than 8 hours. It was necessary when using this technique to freeze the sample before sealing in order to prevent breakage of the sample tube. Ampules formed from 6-inch test tubes were used in this work. The sample tubes were filled to within 1/2 inch of the top to allow sufficient volume for expansion of the liquid sample during freezing.

Pretreatment before irradiation was usually limited to a blanch in boiling water. The material to be blanched was placed in boiling water for a short preselected time and then cooled rapidly with tap water. With some samples sodium bicarbonate was added to the blanch water before boiling. In cases where it was thought that storage properties might be improved by the presence of a reducing agent, the fruit or vegetable was soaked in a dilute ascorbic acid solution (about 1 mg/ml). This was done after blanching. Specific treatment of the samples prior to irradiation consisted of various combinations of the

procedures mentioned. Juices for irradiation were extracted from fresh fruit immediately before irradiation. A Waring blender or a pressure-type hand juicer was used, depending upon the nature of the food.

When it was desired to store samples of milk and to take periodic bacteria counts, the samples were irradiated in 15 ml serum bottles with diaphragm stoppers. In this way, the contents could be sampled with a hypodermic syringe without destroying or contaminating the sample.

Samples were irradiated in the cobalt-60 vault or by means of the 200 KVP x-ray machine previously described in Progress Report I. To reduce the effects of heat from the x-ray tube a small centrifugal blower was used to circulate air over the sample.

Pretreatment of meat samples consisted of cutting to size, trimming, and packaging. Only commercial cuts were used. Dairy products, with the exception of raw milk, were purchased from local retail markets. The raw milk was received from a local creamery.

b) Beef. Over fifty samples of beef were subjected to cobalt-60 gamma radiation. The samples were irradiated in both polyethylene bags and test tubes. No bacteriological or nutritional assays were made. The samples irradiated in polyethylene bags and in air-filled test tubes underwent considerable color change during storage at 32°F, 50°F and at room temperature. An evacuation or an evacuation followed by filling with an inert gas eliminated this effect.

In Fig. 35, are shown samples of various foods, including round steak of beef (top of figure) which has been irradiated by gamma rays from the cobalt-60 source for 24 hours (approximately 2.4×10^6 r*) and stored at room temperature in a partially evacuated glass container for 49 days. Fig. 36, a color photograph** of similar food samples, and shows the bright red color retained by the beef steak irradiated for 40 hours and stored in a glass container for 32 days at room temperature. The control shown in this figure (top right) was an adjacent slice of beef. It received the same treatment as the irradiated sample except that it was not irradiated. Mold colonies developed on the control samples and the putrid odor of decaying flesh was noted.

c) Green Peas. Samples of fresh green peas have been irradiated for dosages up to 24 hours with various pretreatments, including blanching, treatment with sodium bicarbonate solution, and freezing. Samples of freshly hulled

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* Initial approximate calibration of cobalt-60 source as received from Brookhaven was 100,000 roentgens/hr. A more precise calibration will be made before the next progress report is written.

** Color photographs are not in all copies of this report.

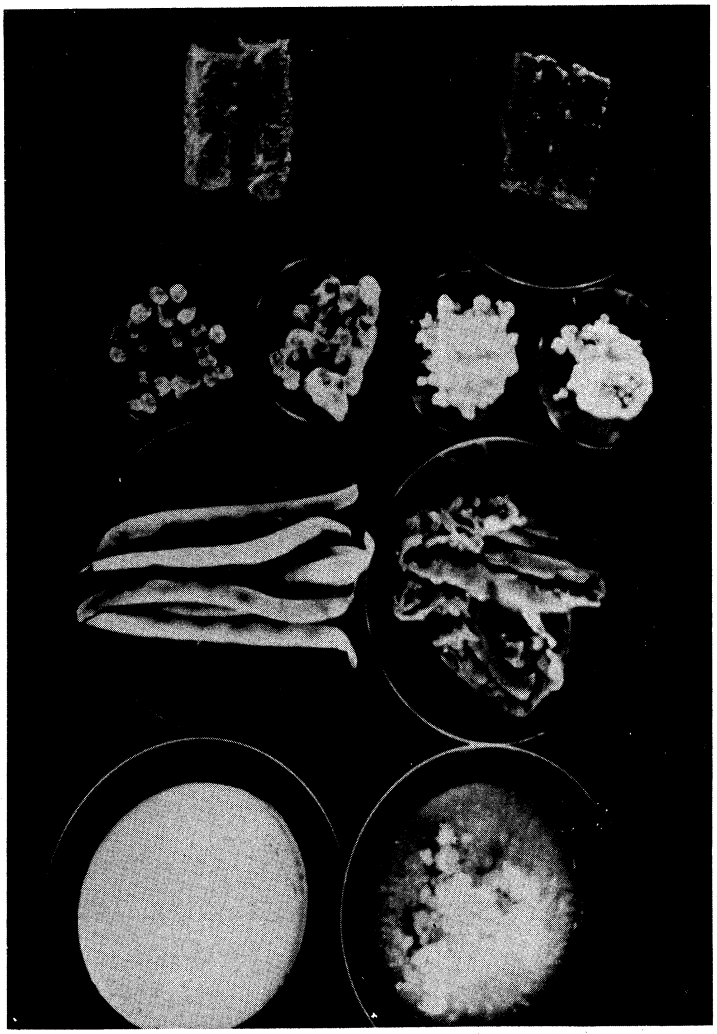

Fig. 35. Irradiated Food Samples (at Left) and Controls (at Right)

green peas irradiated for at least 8 hours exhibited improved storage properties compared to the control samples. However, the irradiated peas bleached out over periods of from four to fourteen days of storage. The texture of the peas was unchanged, and the peas retained their shape. Samples irradiated with no pretreatment and stored exposed to air retained their green color for relatively longer periods than blanched samples. However, the unblanched samples underwent changes in texture in about fourteen days. The irradiated blanched samples exhibited superior storage properties as far as texture was concerned, but they bleached out in relatively short times. The use of dilute sodium bicarbonate solutions (0.1% and 1%) did not retard this action. Irradiated commercially frozen peas did not bleach out, but after thawing they had a softer texture than the fresh peas. All samples sealed in polyethylene bags retained their firmness, but those exposed to air shrank and softened. Irradiation periods of 4 hours or more appeared to be effective in improving storage properties.

Reproducible results, however, were not obtained for irradiation dosages of less than 8 hours.

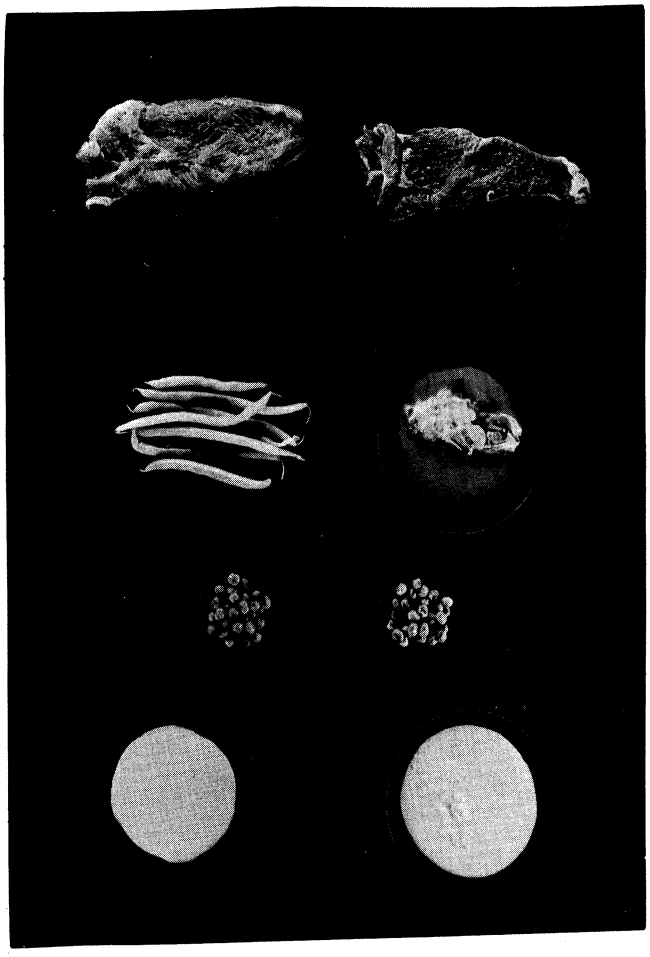

Fig. 36. Color Photograph of Irradiated Foods (at Left) and Controls (at Right)

Samples of green peas (from fresh pods) are shown in Fig. 35 (second row, left) after 47 days' storage at room temperature. The irradiated sample (first dish) received a total dosage of 2.4×10^6 r and appears to be preserved. The control (2nd dish) shows a white mold growth indicating spoilage.

In the color photograph (Fig. 36) commercially frozen green peas are shown after 6 days' storage at room temperature. Both samples were packaged in polyethylene bags, and the irradiated sample received a dosage of 24 hours. The irradiated peas (shown at left) retained a brighter green appearance than the control peas (shown at right). A similar sample of irradiated frozen peas was checked for flavor immediately after irradiation. Peas prepared in this manner and a control sample were cooked separately for 4 minutes in water and tasted. The irradiated peas had a good flavor. They were quite sweet and tasted as if completely cooked, whereas the control tasted slightly undercooked. The controls had a slight taste of bicarbonate, which may have been used in processing for freezing. The irradiated sample did not taste of bicarbonate. The cooked irradiated peas were not as bright a green as the cooked control peas. The irradiated peas had a softer texture after cooking than did the cooked control peas. The control peas were cooked for an additional 4 minutes and compared

again with the cooked irradiated peas. The irradiated peas and the control now were more nearly similar in flavor, color, and texture. The irradiation treatment seems to have an effect approximating that of cooking for 4 to 5 minutes.

d) Green String Beans. All methods of packaging and pretreating peas have been applied to beans. In addition two new treatments have been tried. In an attempt to control the yellowing caused by storage, one sample was blanched in conjunction with soaking in a 1 mg/ml ascorbic acid (Merck, USP) solution. The treatment with ascorbic acid did not affect yellowing. The other method tried was storage of a sample of irradiated beans under nitrogen gas, which resulted in a diminished rate of bleaching, but the discoloration was not entirely stopped. The beans were stored in a polyethylene bag in a desiccator, which was periodically flushed with nitrogen gas from a cylinder. The sample has been stored for 90 days with no apparent change in structure. The color, however, bleached to a greyish green in about 30 days and remained unchanged. The approximate minimum dosage for preservation of green beans was 10 hours of cobalt-60 irradiation. Figs. 35 and 36 show blanched green beans irradiated for 24 hours and the corresponding controls. The blanching was accomplished by placing the beans in boiling water for about 15 seconds. The beans were packaged in polyethylene bags. The irradiated beans gradually lost their color, changing from the original green to the yellowish color shown in Fig. 36. The irradiated beans remained firm, while the beans used as the control developed a mold growth and decomposed rapidly, as shown in Figs. 35 and 36. Samples of commercially frozen green string beans were irradiated for 24 hours and checked for flavor. As in the case with frozen peas, the color of the irradiated cooked string beans was not as bright a green as that of the cooked controls, and the irradiated samples had a softer texture. The flavor of the irradiated beans was not as good as that of the control beans. The irradiated beans had a flavor that was somewhat "grassy" or similar to that of more mature string beans. The irradiated beans tasted somewhat sweeter than the control.

e) Cottage Cheese. The growth of mold on cottage cheese was prevented by gamma irradiation. Samples were packaged in polyethylene bags and irradiated up to 24 hours. Although the irradiated samples did not mold, they did develop a sharp odor in about 7 to 10 days. The dosage required to achieve sterilization was about 10 hours. Fig. 35 shows extensive mold growth on the control sample of cottage cheese (second row, 4th dish). The irradiated sample (Fig. 35, second row, 3rd dish) is uncontaminated with mold and apparently has been preserved. Samples of cottage cheese irradiated for 24 hours however, had a poor flavor, strong and somewhat "smoky" and "goaty".

f) Milk. More than 100 samples of raw and pasteurized milk were irradiated. The results showed that milk may be sterilized by irradiation and stored at room temperature (70°F) for more than 6 weeks without spoilage from microorganisms. Some bacteriologic assays have been completed on samples of

irradiated raw and pasteurized milk. The data collected thus far show that

(1) dosage of at least 10 hours of gamma irradiation is required for the complete sterilization of pasteurized milk, and
(2) approximately 24 hours of gamma irradiation is required for complete sterilization of raw milk.

These conclusions were drawn from Tables V and VI, which present the data on the spoilage of milk. Consecutive numbers in Table VI represent a sample and control pair. A common occurrence in all the irradiated samples stored in polyethylene bags was the development of a brownish discoloration of the milk during storage. Samples stored in impervious evacuated containers underwent less discoloration. Table VII gives the data on the bacteria counts of a series of raw milk samples. The criterion for acceptability for consumer use is set at a count not to exceed 20,000 organisms per ml and a coliform negative test. Since the storage properties and taste are of prime importance, the samples were tested after 72 and 120 hours from the start of the gamma irradiation period. The data of Table VII are plotted in Fig. 37. Fig. 37 is a plot of the residual number of organisms plus one (N + 1) as a function of irradiation time in hours (T). The use of N + 1 as the ordinate permits extrapolation to N = 0 (N + 1 = 1). This represents the mathematical requirement for complete sterility.

TABLE V

BACTERIA COUNTS IN IRRADIATED RAW AND PASTEURIZED MILK

Sample Number	Irradiation Time	Approximate Dose * (rep)	Initial Count Org./ml	Count Post-Irradiation	Coliform Test	Remarks
M27	23.5 hrs	2.4×10^6	4.5×10^6	0	no test	raw milk
M8	17 hrs	1.7×10^6	815	0	no test	pasteurized milk
M157	17 hrs	1.7×10^6	7.75×10^6	100	negative	raw milk

Taste tests on irradiated pasteurized milk have shown that most individuals can detect an off-flavor in milk which has received only two hours of radiation. The off-flavor is best described as a burnt or sharp taste. Some have described it as a "chalky taste".

Fig. 35 shows milk in a watchglass (lower left of figure) which had been packaged in a sealed glass tube, irradiated for 24 hours and stored for 31 days at room temperature in the sealed tube. The control (at the lower right of figure) was treated the same way except that it received no irradiation. The control separated into curds and an almost clear whey.

* See footnote, page 46.

TABLE VI

OBSERVATIONS ON IRRADIATED MILK STORED AT ROOM TEMPERATURE

Sample Number	Time to Curdle	Remarks	Storage Temperature	Type	Irradiation Time	Container
M1	---	Not curdled after 6 weeks	77°F	Pasteurized	24 hrs	Test tubes
M2	48 hrs	Large white curds	77°F	Pasteurized	0	Test tubes
M3	6 wks	Brown discoloration, small curds	77°F	Pasteurized	24 hrs	Polyethylene bags
M4	72 hrs	Large white curds	77°F	Pasteurized	0	Polyethylene bags
M8	8 wks	Brown discoloration, small curds	77°F	Pasteurized	17 hrs	Polyethylene bags
M7	24 hrs	Large white curds	77°F	Pasteurized	0	Polyethylene bags
M24	3 wks	Red growth on milk (blood colored)	77°F	Raw	24 hrs	Polyethylene bags
M23	24 hrs	Large white curds	77°F	Raw	0	Polyethylene bags
M40		Opened after 8 weeks; no spoilage evident	Room temperature	Pasteurized	24 hrs	Evacuated test tubes
M41	48 hrs	White curds	Room temperature	Pasteurized	0	Evacuated test tubes

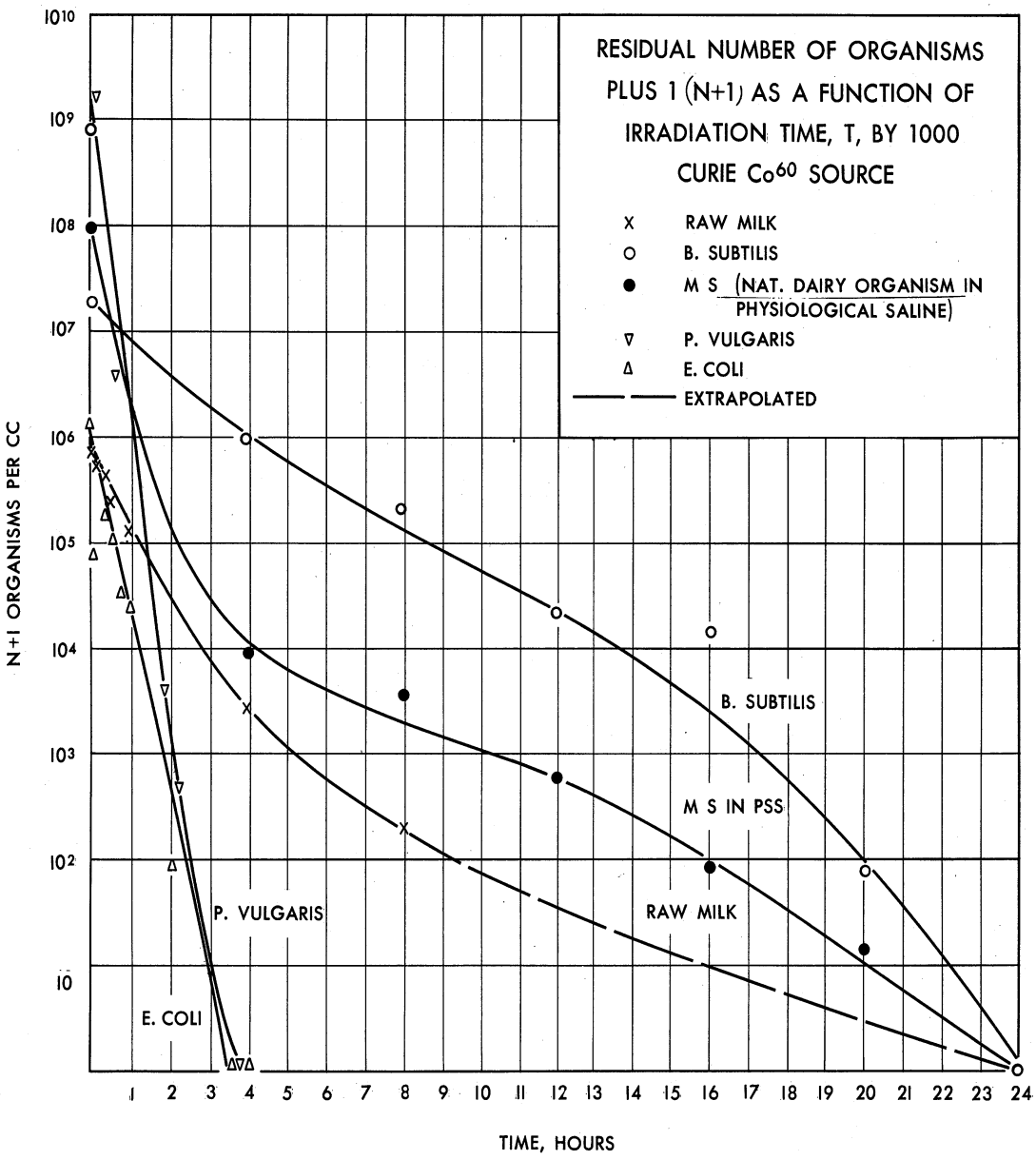
RESIDUAL NUMBER OF ORGANISMS PLUS 1 (N+1) AS A FUNCTION OF IRRADIATION TIME, T, BY 1000 CURIE Co60 SOURCE
X RAW MILK
O B. SUBTILIS
● M S (NAT. DAIRY ORGANISM IN PHYSIOLOGICAL SALINE)
∇ P. VULGARIS
Δ E. COLI
EXTRAPOLATED
N+1 ORGANISMS PER CC
10^10
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10
B. SUBTILIS
M S IN PSS
RAW MILK
P. VULGARIS
E. COLI
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24
TIME, HOURS

Fig. 37

TABLE VII

EFFECT OF COBALT-60 IRRADIATION ON RAW MILK STORED AT 50°F

Sample Number	Radia-* tion Time (hours)	Count (organisms/cc)** Housing in Storage 0	72	120	Coliform Test Housing in Storage 0	72	120	Time of Ir radiation (hours)
†148	0	755,000			+			0
149	.06	600,000	>20,000		+	+	+	.08
150	.25	447,500	>20,000		+	+	+	.25
151	.5	275,000	>20,000		+	+	+	.50
152	1	130,000	>20,000		+	+	+	1.00
153	2	42,500	>20,000		+	+	+	2.00
154	3	16,425	19,700	>20,000	+	+	+	3.00
155	4	2,450	2,705	97.65	-	-	+	4.00
156	8	290	310	6,615	-	-	-	8.00
ǂ157	17	790	730	1,225	-	-	-	17.00

* Bacteriological methods and technique to be discussed in Section C of Part III.
**No storage-time test made where count is greater then 20,000 or coliform test is shown positive on previous run.
† Control Sample.
ǂ Plated after 12 hours' storage at 32°F.

Fig. 36 shows similar samples in color. The milk samples in this case were stored in partially evacuated glass containers at room temperature for 30 days. The sample (at the left) was irradiated for 24 hours and appears well preserved except that it is more cream colored. The control (at the right) soured and formed curds, but the whey was not clear as in the previous test.

A considerable amount of research on raw and pasteurized milk was performed in cooperation with Michigan Memorial-Phoenix Project 20, supervised by Dr. C. A. Lawrence, who was assisted by John Graikowski. All but a few of the bacteria counts were made by this group. A copy of their report is included in this section.

g) Bananas. A number of banana samples were exposed to x-rays for dosages of from 10,000 to 1,000,000 roentgens. The results of this work indicated that the softening of a banana may be retarded by x-radiation. The amount of retardation increased with increasing dosages. This work also showed that x-radiation does not prevent the characteristic blackening of the skin. In fact, the radiation seemed to sensitize the skin to blackening. In those cases where long periods of irradiation were used (5 to 10 hours), the skin blackened during the irradiation; for the shorter irradiation periods, the area exposed

to the x-radiation was the first to blacken during storage. Small sections of hard, whole green bananas were cut and paraffined to prevent dehydration. The sections were then irradiated in the cobalt-60 vault for dosages in the same range as those in x-ray work. The results were similar. Those irradiated for 12 hours or more were completely blackened on removal. The sections irradiated for less than 12 hours blackened prior to the controls during storage. The nature of the blackening process was believed to be an oxidation catalyzed by ionizing radiation. In an effort to prevent this several samples were soaked in a 1 mg/ml ascorbic acid solution and irradiated. The results were negative.

The inhibition of softening of bananas during storage by x-radiation is shown in Fig. 38. Although the skin has completely blackened, the softening of the flesh of the fruit has been inhibited roughly in proportion to the dosage.

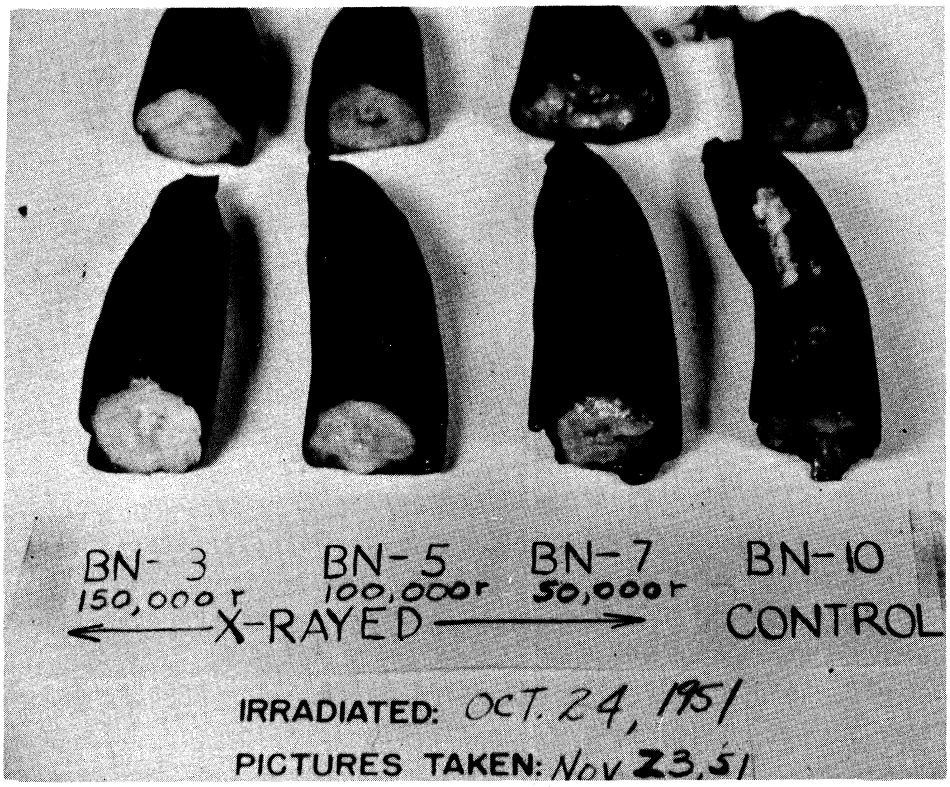

Fig. 38. Photograph of X-Irradiated Bananas

h) Apple Juice. The fermentation of apple juice to form cider and vinegar is a common process in the fruit industry. Gamma irradiation for periods of not less than 5 hours prevented fermentation. The juice was prepared from fresh apples by pulping in a Waring blender, followed by filtering through cloth or filter paper. Polyethylene bags were used for the samples in a number of experiments. These bags, however, proved unsatisfactory since they allowed air to diffuse into the sample, which resulted in extensive color changes. Coagulation also occurred in the samples stored in polyethylene bags. Irradiated

samples stored in test tubes were kept for periods in excess of 60 days. There was no visible evolution of gases which would be indicative of fermentation and the color of the samples did not appear changed from that of the fresh state. The controls in almost all cases where the sample was stored in test tubes fermented in 2 to 7 days. No flavor checks were made of the irradiated samples which were opened, however, no off-odors were noticed.

i) Orange Juice. Orange juice was sterilized by gamma radiation. The minimum dosage required was about 5 hours in the cobalt-60 vault. The juice was prepared by squeezing fresh oranges through a hand-pressure-type squeezer. Coarse particles and all of the seeds were removed by this method and further filtration was not used.

The taste of the irradiated juice was not satisfactory. In all cases the flavor was similar to that of heat-processed canned juice. Efforts to eliminate this effect by irradiation in the frozen state and/or in inert atmospheres have not been successful.

j) Spinach. In this series of experiments, an attempt was made to prevent or delay the fermentation of fresh spinach by irradiation with x-rays. Irradiation of large samples of spinach in the cobalt-60 vault was not practical because of the small volume available in the vault; therefore, the x-ray generator was used for these tests (200 KVP and 15 ma).

Preliminary studies involving the irradiation of relatively small samples of spinach in 1000 ml beakers with dosages up to about 500,000 roentgens appeared promising. The irradiated samples fermented in about 5 days as compared to less than 2 days for the control. The irradiated samples retained their green, leafy appearance until fermentation commenced, while the control darkened and compacted rapidly before and during fermentation. These samples were irradiated soon after they were received from the cold room (40°F) of the University of Michigan Food Service Building. They were irradiated at room temperature for a maximum of approximately 6 hours and placed with the control sample in a refrigerator held at 50°F.

Separate bushel baskets of spinach were irradiated for a maximum of 27 hours or an equivalent dosage of approximately 500,000 roentgens. The spinach as received from University of Michigan Food Service represented two consignments. All baskets were emptied, thoroughly mixed and repacked in order to obtain a uniform and comparable group of samples. Two baskets were retained as controls while two were irradiated under the x-ray tube. The samples to be irradiated were stacked one above the other, giving the upper sample a total dosage of about 300,000 roentgens and the lower sample a dosage of about 100,000 roentgens. The samples were not cooled during the irradiation period. Thermometers inserted after irradiation recorded a temperature of 100°F for all

samples. Room temperature during irradiation was 80°F. The samples were then placed in the Food Service's cold room (40°F) and temperatures recorded during a 10-day period. The temperature of all samples, i.e., the irradiated spinach and the control samples, declined to within a few degrees of the cold-room temperature and remained at this temperature for the duration of the test. There were small differences in the temperatures of the individual samples; however, these were not significant and did not reveal any apparent difference between the irradiated and control samples. Fermentation odors of all samples became evident after about 2 days in the cold room.

It appeared from the results of this test that fermentation had started in all samples before irradiation had been completed. Tests were repeated using cooling to minimize fermentation during irradiation. Three one-half bushel samples of fresh spinach were packed into fiber waste paper baskets to form two irradiation samples and a control. One of the samples was covered with an inch layer of crushed ice and irradiated for 43 hours for a total dosage of approximately 400,000 roentgens. A second sample placed under the first received a dosage of about 130,000 roentgens. Thermocouples were inserted in all samples and temperatures recorded during irradiation and for 4 days after the irradiation period. All samples, including the control, were cooled by ice during the irradiation period. The temperature of the high-dosage sample averaged about 57°F during irradiation, while the average temperatures of the low-dosage sample and the control were approximately 65°F. After irradiation, the ice-cooling was discontinued, and the temperatures of all samples rose to about 90°F (13° above room temperature). The odor of fermentation appeared in all samples at this point. The rate of fermentation appeared to decline approximately 4 days after the end of the irradiation period as noted by a decline in the temperatures of all the samples. The temperature of the high-dosage sample started to decline about 12 hours later than either the low-dosage sample or the control.

From the data on this run the cooling provided appeared insufficient to retard fermentation during the irradiation period. The sample receiving the greater dosage fermented during irradiation, although at a slower rate than the control, as indicated by its lower temperature for the same amount of cooling. Furthermore, it continued to ferment for some time after fermentation of the control was observed to decline. The sample receiving the lesser dosage gave results similar to the control in all respects, indicating that the amount of radiation it received was insufficient to affect it.

A further study was made in which spinach was irradiated in four one-quart thermos bottles provided with a small test tube of ice to maintain the temperature of the sample at 32°F during the irradiation period. By using thermos bottles, it was possible to place the sample nearer the x-ray source, thus obtaining a higher dose rate and a shorter irradiation period for

a given total dosage. The bottle was inverted midway through the irradiation period to irradiate all portions of the sample more uniformly.

One sample was irradiated for a total dosage of 130,000 roentgens. A plot of the time-temperature relation for this sample and a control is shown in Fig. 39.

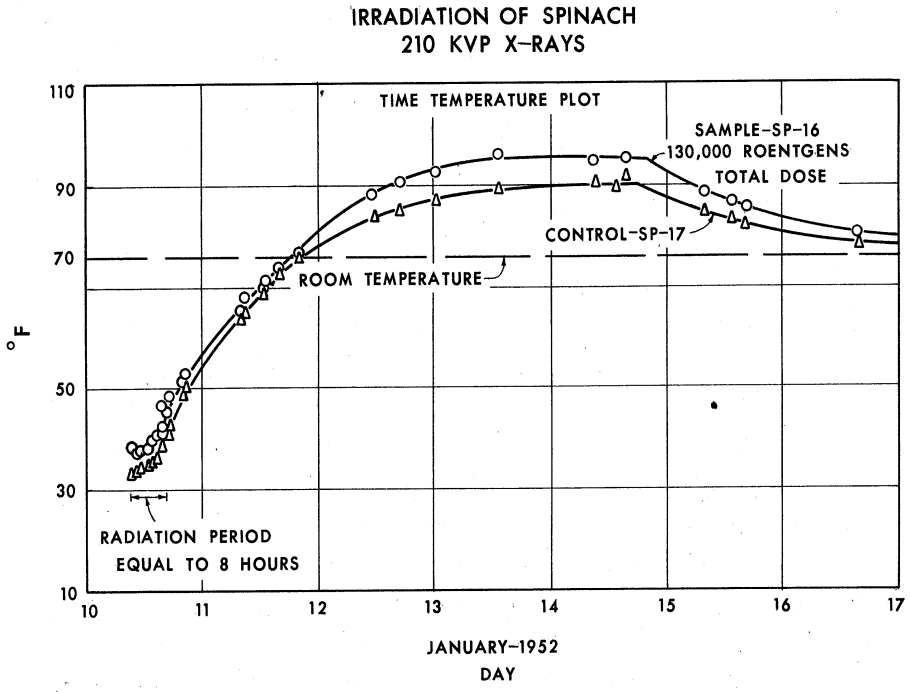

Fig. 39.

Both samples fermented as shown by the rise in temperature above room temperature. Apparently, the ice-cooling was not sufficient to keep the samples at 32°F during the irradiation period, and further tests are planned using greater radiation dosages and different cooling techniques.

k) Other Work on Foods. Other food products tested include melons, various berries, pears, plums, peaches, and grapes. Of these, grapes have been tested most extensively, and the results so far are not encouraging. Undesirable color changes and liquification occur on samples irradiated up to 24 hours in the cobalt-60 vault. The samples were packaged in polyethylene bags.

Tests on the other fruits mentioned have given either negative or irreproducible results. All other tests except those with berries and one test with plums were performed with the 200 KVP x-ray machine. Irradiation in the cobalt-60 vault could not be accomplished because of the size limitation.

A sample of tomato juice, prepared by pulping fresh, washed tomatoes in a Waring blender and filtering through cheesecloth was irradiated for 24 hours in the cobalt-60 vault and stored under nitrogen gas. The sample developed a growth of microorganisms after 14 days.

3. Conclusions and Discussion of Tests with Irradiated Foods

a) Meats. The experiments on the gamma irradiation of various meats have shown that it is possible to prevent spoilage by bacterial decay with dosages in excess of 16 hours in the cobalt-60 vault (approximately 1.6×10^6 r). The color change (darkening) that occurs during long-time storage of these foods when packaged in polyethylene containers may be prevented by vacuum packaging techniques or by storage under nitrogen or other inert atmospheres. It is believed that this color change is an oxidation process, since methods of packaging that allowed oxygen to diffuse into the meat lead to these color changes.

The limiting effective dosage for sterilization and long-time storage has not been rigidly established. Dosages below 2 hours are definitely ineffective, but dosages between 4 hours and 7.5 hours have given mixed results. Samples irradiated more than 16 hours in the cobalt-60 vault exhibit good storage properties. The temperature effect on the irradiated meat has not been detectable from visual observation. Spoilage of the samples was determined visually. Those factors considered as evidence of spoilage include the evolution of gases, excessive darkening, odor, and the formation of mold colonies. When polyethylene bags were used as the storage container, the presence of strong, putrid odors could be detected in the unirradiated sample after only short storage times (2-3 days at 67°F). No such odor could be detected in samples preserved by irradiation.

Taste tests on irradiated and stored meat indicate that there is a slight flavor change. In cooking the samples there is a noticeable off-odor early in the cooking process. It persists for several seconds and is replaced by the normal odor of broiling meat. All samples to be tested were pan-broiled in a very small amount of vegetable shortening. When the beef is irradiated as chopped or ground meat rather than whole pieces, no differences in effect or properties have been detected.

It was learned that experiments on the irradiation of meats were being conducted at the research laboratory of Swift and Company. Professor L. E. Brownell and Dr. F. H. Bethell visited the Swift laboratory in Chicago on October 2, 1951. The research at Swift is under the guidance of Dr. W. M. Urbain and has been in progress for about two years. Approximately 20,000 samples of meat have been irradiated primarily with various forms of beta irradiation. A few samples have been exposed to gamma irradiation at Brookhaven. Some two or three generations of rats have been kept on a diet of irradiated meat with no noticeable effects, except in one instance signs of vitamin-E deficiency were observed. This deficiency was corrected by feeding vitamin E as a supplement. It was the opinion of Dr. Urbain that there was no question about the fact that meat could be preserved by irradiation; and,

to date, there has been no evidence of toxic or harmful effects. Representatives of Swift and Company believe that flavor change is one of the biggest problems of developing a commercial process of irradiation of meat. Professor Brownell brought samples of beef irradiated for 24 and 44 hours in the 1000-curie source to the meeting in Chicago. These samples were cooked by a laboratory cook at Swift and Company along with samples of beef preserved by beta radiation in the research conducted at Swift and Company. One of the samples of Swift and Company was cold canned rare roast beef. All samples of irradiated beef were cooked for about 15 or 20 minutes by frying in a neutral shortening. The meat had a slightly scorched or musty flavor. This off-flavor was most noticeable in the sample irradiated with gamma rays for 44 hours. Swift's research workers mentioned that they had tried various forms of spiced meats, such as salami, in an attempt to mask the flavor change with seasoning but had found that the irradiation developed some very unusual flavors when spice was present in the meat.

Apparently, one of the big problems in the use of irradiation for food preservation will be the prevention of undesirable flavor changes. An effort will be made to investigate the maximum irradiation dosage that can be used without any detectable flavor change. It is believed that low-temperature irradiation may be helpful in minimizing these flavor changes. Some experiments will be made at low temperatures. Providing the problem of flavor change can be satisfactorily solved, many other problems still exist. Possible toxicity, of course, must also be checked thoroughly. Various types of containers and pretreatments must be investigated to determine the optimum conditions for storage of irradiated food. Other problems, such as the possible loss of nutritive value, public acceptance of the food, etc., will have to be evaluated.

b) Milk. Sterilization of both pasteurized and raw milk has been obtained with gamma irradiation. Although the irradiated milk has shown good long-time storage properties two undesirable changes have been noted. Samples stored for long periods in polyethylene bags have developed a yellow-to-brown discoloration. This was reduced by storing the milk in glass containers which were not permeable to air. Undesirable flavor changes were found in milk that was irradiated for 2 hours or more. It may be necessary to sacrifice complete sterilization and long-time storage properties in favor of shorter irradiation periods to minimize flavor changes. Milk is sensitive to all forms of processing, and off-flavors develop quite readily. If the problem of flavor changes can not be overcome a pasteurization by irradiation to reduce the bacterial count rather than sterilization by irradiation may be successful.

c) Green Peas. So far the processing of green peas by gamma irradiation appears most promising from preservation, appearance, and flavor standpoints. No objectionable off-taste or changes in color have been noticed,

and relatively short dosages of about 8 hours in the cobalt-60 vault (approximately 8.0×10^6 r) have been found effective in preservation. No special packaging techniques are required; however, best results were obtained with peas irradiated in the frozen state in vapor-tight containers and kept under refrigeration during storage.

d) Spinach. Both spinach and milk spoil by fermentation processes. Since milk has been preserved by gamma irradiation, it was thought that a similar preservation might be effected with spinach. Several attempts to prevent fermentation of spinach using 220 KVP x-rays have so far been unsuccessful. Further tests are planned using higher radiation dosages, such as are comparable to the dosages used in sterilizing milk. One of the difficulties is the problem of preventing fermentation of the spinach during the irradiation period.

e) Other Foods. With other foods tested the general conclusions which can be made to date are that gamma irradiation for about 24 hours in the 1000 curie cobalt-60 vault will sterilize the food. However, such dosages of gamma rays bring about flavor changes. Methods of preventing these flavor changes have not yet been investigated.

B. ANIMAL FEEDING EXPERIMENTS

Personnel

Chief Investigator: Dr. F. H. Bethell. Dr. A. H. Kretchmar, Research Assocatie.

1. Introduction

The early encouraging results with the preservation of milk by gamma irradiation indicated the necessity of determining any possible deleterious changes as a result of irradiation. Dr. Tuttle of the Washington, D. C., office of the Atomic Energy Commission, Division of Biochemistry and Medicine, suggested that at least preliminary tests should be run immediately. A cooperative research test was performed with the AEC, Biological Effects of Irradiation Laboratory, University of Michigan. The facilities of the Fission Products Laboratory were used to irradiate milk prepared by the staff of the Biological Effects of Irradiation Laboratory.

2. Feeding Experiments

The results reported in this section have been prepared by Dr. F. H. Bethell, Chief Investigator, and Dr. A. H. Kretchmar, Research Associate, Biological Effects of Irradiation Laboratory.

"It is to be emphasized strongly that this experiment can be regarded only as exploratory and that any conclusions which may be drawn from the data are preliminary and contingent upon further testing, both with feeding experiments and more refined methods of assay for individual components of the diet. This would require, for example; microbiological assays for amino acids and vitamins, and the setting up of long-term feeding and breeding experiments.

"Briefly stated, twelve young adult Wistar female rats were divided into two groups of six animals each. Group I was fed, ad libitum, irradiated whole milk. (All milk samples were irradiated in polyethylene bags for 24 hours in the cobalt-60 source. The dosage was 24 hours (approximately 2.4 x 10^6 rep). This material was reconstructed Klim irradiated as a four times concentrate and diluted by addition of salt solution just before feeding to give a milk solution containing 9.9% fat, 9.6% protein, and 13.5% lactose, and the following concentration of salts: 1.5 mgm Fe/100 ml; 0.3 mgm Cu/100 ml; 0.1 mgm Mn/100 ml. In addition, the diet included a leaf of lettuce once a week.

3. "Conclusions

(1) Rats consume the same amount of irradiated milk as of nonirradiated milk. It is therefore suggested that no great change in caloric value can have resulted from the irradiation. Several average daily intakes illustrate this point as shown in Table VIII.

"(2) Reference to Fig. 44 shows that the irradiated milk supports growth as adequately as does nonirradiated milk. It is therefore suggested that, within the limits of this experiment, no great loss in essential amino acids or B vitamins can have resulted from the irradiation.

"(3) Table IX presents a summary of the hematologic data obtained at the termination of the experiment. The values for red blood cell count, hematocrit and hemoglobin are low normal for this strain, under the conditions of housing in this laboratory and fed ad libitum on Rockland Rat Diet (complete). Since minerals were supplied as a supplement it is suggested that the organic precursors required for blood cell synthesis are as adequately supplied by irradiated as by nonirradiated milk."

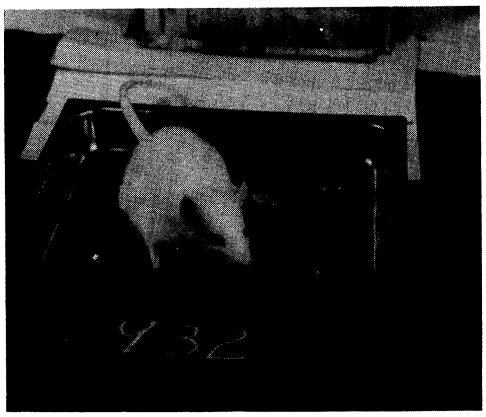

Fig. 40. Control Rat 932 at Beginning of Experiment

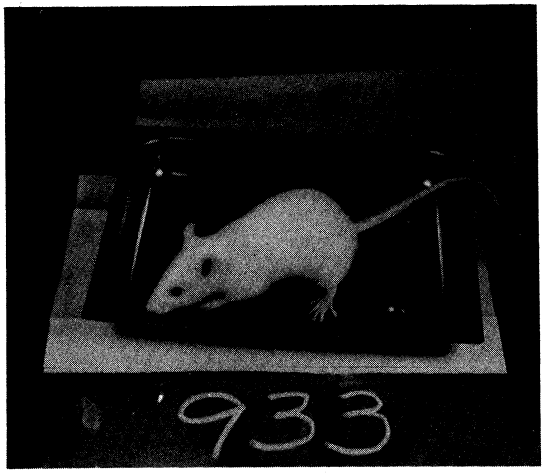

Fig. 41. Rat 933 at beginning of Experiment

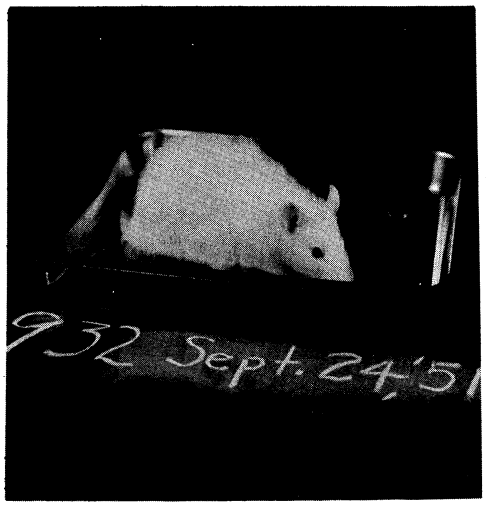

Fig. 42. Control Rat 932 after Five Weeks

Fig. 43. Rat 933 Fed Irradiated Milk for Five Weeks

TABLE VIII

AVERAGE CONSUMPTION OF MILK BY RATS IN ANIMAL FEEDING EXPERIMENT

8-13-1951	Irradiated Milk Consumed	I	23.7 g/day
	Nonirradiated Milk Consumed	II	25.3 g/day
8-15-1951	Irradiated Milk Consumed	I	24.8 g/day
	Nonirradiated Milk Consumed	II	27.8 g/day
9-13-1951	Irradiated Milk Consumed	I	41.7 g/day
	Nonirradiated Milk Consumed	II	39.3 g/day
9-15-1951	Irradiated Milk Consumed	I	31.0 g/day
	Nonirradiated Milk Consumed	II	30.6 g/day

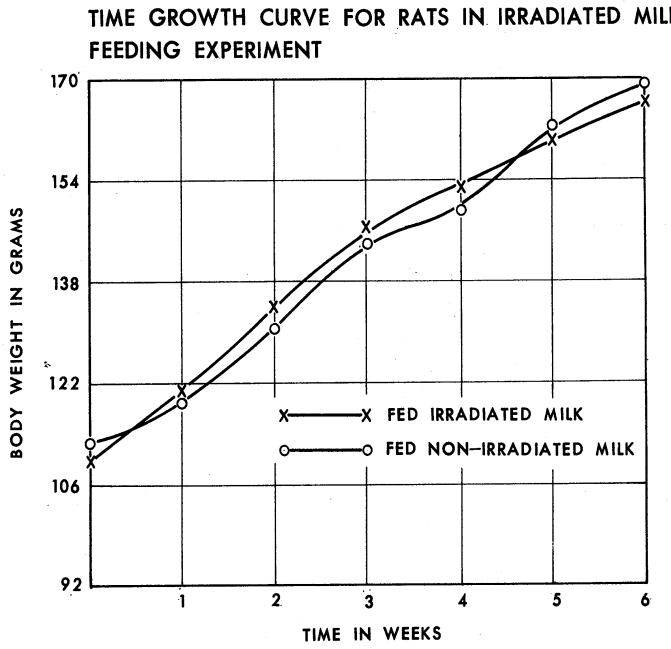

Fig. 44

TABLE IX

AVERAGE HEMATOLOGIC DATA ON RATS IN ANIMAL FEEDING EXPERIMENTS

	WBC cells/cu mm	RBC cells/cu mm	Ht in per cent	Hb in grams	Eos in cells/cu mm
Fed Irradiated Milk	9.93×10^3	9.2×10^6	47.1	14.8	116
Fed Nonirradiated Milk	9.64×10^3	9.3×10^6	46.8	14.9	88

C. COOPERATIVE RESEARCH WITH MICHIGAN MEMORIAL-PHOENIX PROJECT 20

1. Introduction

Engineering Research Institute Project M943 and Phoenix Project 20 have a mutual interest in the effects of gamma irradiation on microorganisms, engymes, and other biological factors. The Engineering Research Institute Project is attempting to find industrial uses of the radioactive fission products and the Phoenix Project is interested in the humanitarian uses of atomic energy.

Phoenix Project 20 is supervised by Dr. C. A. Lawrence, Assistant Professor of Bacteriology in the Medical School, who is assisted by John Graikowski. Since Phoenix Project 20 has the use of bacteriological laboratories,

equipment, and personnel, and since the Engineering Research Institute has the use of a 1000-curie cobalt-60 source, it was considered advisable to pool the resources on a joint research effort. Irradiations are performed by Engineering Research Institute Project M943, and bacteriological tests are performed by Phoenix Project 20. The results of these tests are to be made available to both Engineering Research and Phoenix. As the tests on microorganisms are related to the bacteria counts made on irradiated milk, the report of the Phoenix Project 20 is included at this point:

2. "The Lethal Effects of Gamma Radiations (Cobalt-60) Upon Microorganisms

"a) Experiment No 1 The Effects of Gamma Radiation (Cobalt-60) on Proteus vulgaris, Eschericha coli and Lactobacillus arabinosus

"Cultures and Methods: E. coli and Proteus vulgaris were grown on beef extract agar slant at 37°C for 18 hours. The growths from the surface of the slants were suspended in 15 ml sterile saline solution (0.85% NaCl) containing 0.5% gelatin (pH 6.2). The suspension was then filtered through sterile glass wool and distributed in sterile Pyrex culture tubes.

"The culture of L. arabinosus was grown in Difco Micro-Inoculum Broth for 18 hours at 37°C, at which time the organisms were centrifugated, the supernatant medium discarded, and the packed cells resuspended in sterile saline-gelatin solution. This culture was also distributed in Pyrex test tubes, as above, but, since the suspension appeared homogenous, the material was not filtered through glass wool.

"Approximately 5 ml of each culture in 15 x 150 Pyrex culture tubes (with plastic screw-caps) was exposed to gamma radiation (cobalt-60). Since previous calibrations of the temperature within the cobalt-60 vault indicated it to be comparable to the temperature of the room in which it was stored, the control culture suspensions which were not irradiated were maintained at room temperature in the laboratory during the period of irradiation of the test suspensions.

"Dilution of Culture Suspensions, Plating and Counting. With but occasional exceptions, dilutions of the treated and untreated organism suspensions with subsequent plating in agar was carried out within one hour after the cultures were removed from the vault (cobalt-60).

"In the preliminary dilution series of the treated and untreated organism suspensions 0.2 ml of the samples were added directly to the center of sterile petri dishes and 0.2 ml placed in 100 ml of sterile saline solution. Two-tenths of the latter dilution was then added to additional sterile petri

dishes, and 0.2 ml of the same suspension was put in a second bottle containing 100 ml saline. This same method of plating and dilution was carried out for an additional, or third series of tests, in which the untreated control sample was plated for bacterial counts. All cultures in which plates were prepared were made in duplicate.

"To the 0.2 ml of suspension in individual petri dishes was added approximately 15 ml of melted and cooled (45°C) nutrient agar. The plates were tilted and swirled gently to mix the suspension in the medium and the agar then allowed to solidify. When the latter was solid, the plates were incubated at 37°C for 24 to 48 hours and the numbers of colonies developing in the media were counted with the aid of a Quebec Colony Counter.

"The results of the first experiment against Proteus vulgaris, E. coli and L. arabinosus are presented in Table X.

TABLE X

COUNTS OF P. VULGARIS, E. COLI AND L. ARABINOSUS AFTER 24 HOURS IRRADIATION

	P. Vulgaris	E. Coli	L. arabinosus
Sample (irradiated 24 hrs)	0	0	0
Control (unirradiated)	1,593,750,000	850,000	65,000

"b) Experiment No. 2 The Effects of Gamma Radiation (Cobalt-60) on Proteus vulgaris, Escherichia coli and Lactobacillus arabinosus

"Since the results of the preliminary experiment (No. 1) indicated that an exposure of the three test organisms to cobalt-60 for a period of 24 hours caused 100 per cent 'destruction' of all the organisms, a second experiment was carried out on fresh suspensions of all three cultures, which were treated with gamma irradiation for a shorter period of time, 5 hours.

"The cultures used and the method by which the test was carried out are essentially a duplication of the procedures given in Experiment No. 1. The results of the present study are given in Table XI.

TABLE XI

COUNTS OF P. VULGARIS, E. COLI AND L. ARABINOSUS AFTER 5 HOURS IRRADIATION

	P. Vulgaris	E. Coli	L. arabinosus
Sample (irradiated 5 hrs)	0	0	0
Control (unirradiated)	1,898,750,000	1,138,000,000	25,000

"c) Experiment No. 3 The Effect of Gamma Radiation (Cobalt-60) and Roentgen Radiation (X-rays) Upon the Bacterial Flora of Milk[1]

A quart of pasteurized whole milk was purchased on the market and distributed in polyethylene tubes. One of the samples was irradiated with cobalt-60 for 2 hours, another sample was exposed to x-rays for a period of 1 hour and 32 minutes. A control sample was placed outside and adjacent to the cobalt vault but not exposed to radiations of the isotope. A second control consisted of testing the pasteurized milk for bacterial counts before the experiments were started. The methods of dilution and plating, and subsequent counting are essentially the same as outlined in Experiment No. 1. The results of this study are given in Table XII.

TABLE XII

COUNT OF BACTERIAL FLORA IN MILK AFTER IRRADIATION

Sample	Numbers of Organisms per ml
M-14 (2 hrs irradiation with Co^{60})	155
M-12 (1 hr 32 min irradiation with x-rays)	165
M-11 (2 hrs standing outside of cobalt vault - control)	250,000
M-10 (Control - count on milk before irradiations)	805

[1] This experiment was suggested by Professor L. E. Brownell of the Engineering Research Institute under a grant of the U. S. Atomic Energy Commission (AEC) Contract No. AT(11-1)-162, Project M943. The bacteriology was carried out under the present Phoenix Project Grant, No. 20.

"d) Experiment No. 4 The Effect of Gamma Radiation (Cobalt-60) on the Bacterial Flora of "Raw" Milk[1]

"While a definite reduction and inhibition of bacterial growth was obtained in Experiment No. 3, in which pasteurized milk was exposed to cobalt-60, it appeared desirable to determine whether a similar effect could be obtained by exposing 'raw' milk to the radiation. In the present study the time of exposure of the milk, contained in polyethylene bags, was extended to a period of 24 hours. Appropriate controls on the unirradiated samples were maintained as indicated in Experiment No. 3. The results of this study are presented in Table XIII.

TABLE XIII

COUNT OF BACTERIAL FLORA IN "RAW" MILK AFTER IRRADIATION

Sample	Numbers of Organisms per ml
M-27 (24 hrs irradiation with Co^{60})	0
M-26 (24 hrs standing outside of cobalt vault - control)	4,500,000
M-28 (Control - count on milk before irradiation)	20,000

"e) Experiment No. 5 The Effect of Gamma Radiation (Cobalt-60) on Proteus vulgaris and Escherichi coli, with Reference to Lethal Activity in Proportion to Time of Irradiation

Since the two preliminary or exploratory tests (Experiments 1 and 2) indicated that exposures of 5 and 24 hours of pure cultures of P. vulgaris and E. coli to cobalt-60 resulted in 100 per cent "destruction" of the organisms, it appeared desirable to determine the effects of shorter durations of exposures of the same cultures to the isotope. The preparation of the culture suspensions for irradiation, dilutions, and plate counting were carried out as outlined in Experiment No. 1. The number of surviving organisms following irradiation, the number in the unirradiated control specimens, and per cent reduction noted in the treated samples are given in Table XIV.

[1] See footnote, page 66.

TABLE XIV

COUNT OF P. VULGARIS AND E. COLI AS A FUNCTION OF IRRADIATION DOSAGE

Time of Irradiation	P. Vulgaris	Per Cent Reduction	E. Coli	Per Cent Reduction
0 (unirradiated control)	1,666,000,000	0	1,976,250,000	0
50 min	4,500,000	99.7	1,600,000	99.8
1 hr 45 min	3,680	99.99	500	99.99+
2 hr 25 min	55	99.99+	0	100.00
3 hr 40 min	0	100.00	0	100.00
5 hr	0	100.00	0	100.00

"f) Experiment No. 6 The Effect of Gamma Radiation (Cobalt-60) on Spore-Forming Microorganism, Bacillus subtilis, A Gram-Positive Sporulating Bacillus Isolated from Milk (M-S)[1], Penicillium notatum and Aspergillus niger

"With the evidence obtained in the preliminary experiments (Nos. 1, 2 and 5) that cobalt-60 has a definite "lethal" action on nonsporulating bacteria (P. vulgaris, E. coli), studies were outlined to determine the effects of cobalt radiations upon microorganisms of the spore-forming varieties. Cultures used in the present investigation included a 4-day-old culture of B. subtilis and a 22-hour culture of a spore-forming bacillus originally isolated from a milk sample. Both these organisms were grown on agar slants (beef extract agar). In addition to the latter bacteria, suspensions were made of 22-day old cultures of the molds P. notatum and A. niger which were grown on Sabouraud's Maltose agar slants. Suspensions of the organisms were made as indicated in experiment No. 1 of this report. Microscopic examination of each specimen before irradiation revealed an abundant number of spores of the organisms. In the plating procedure in which the molds were tested, Sabourand's Maltose agar was used. The bacteria were plated, as indicated previously, in beef extract agar. The latter organisms (colonies) were counted after 24 and again after 48 hours incubation at 37°C. The mold counts were made following 5 days' incubation at room temperature (24-26°C). The results of this study are presented in Table XV.

[1] The culture of the spore-forming bacillus was isolated and obtained through the courtesy of Dr. F. W. Barber, National Dairy Research Laboratories, Inc., Oakdale, Long Island, New York.

TABLE XV

COUNT OF SPORE-FORMING MICROORGANSIMS AFTER IRRADIATION

Time of Irradiation	B. subtilis	Per cent Reduction	M-S	% Red.	P. notatum	% Red.	A. niger	% Red.
0 (Control)	100,000	0	6,987,000	0	125,000	0	75,000	0
4 hrs, 25 min	19,000	81.0	7,800	99.9	0	100	10	99.98
7 hrs, 15 min	3,250	86.75	4,240	99.95	5	99,99	5	99.99

"Fig. 45 shows a photograph of petri dishes with a normal mold growth of P. notatum and Asp. niger on the control but no growth on samples irradiated for 4 hours.

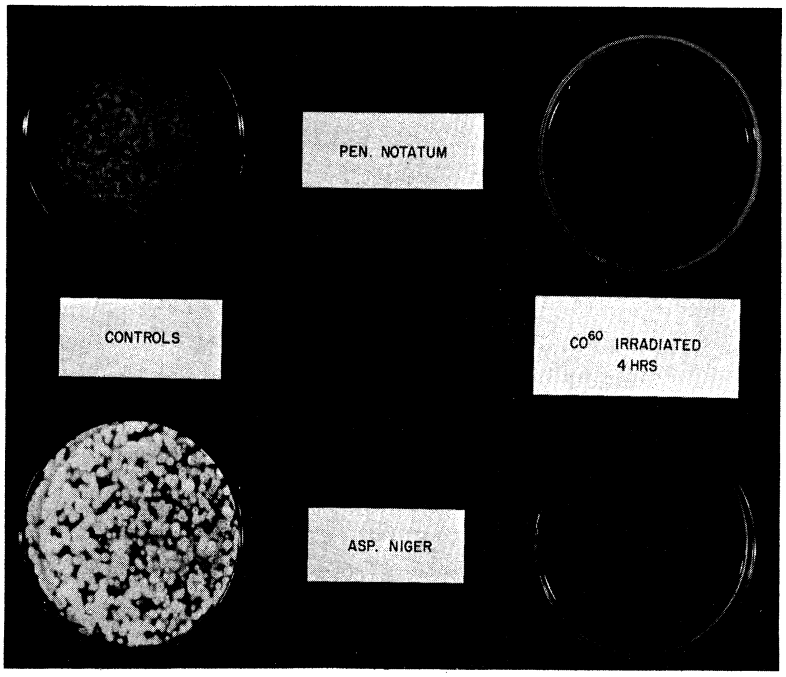

Fig. 45. Photograph Showing Effects of Gamma Irradiation on Molds

"g) Experiment No. 7 The Effect of Gamma Radiation (Cobalt-60) on Spore-Forming Bacteria, Bacillus subtilis and a Gram-Positive Sporulating Bacillus Isolated from Milk (M-S)[1], with Reference to Lethal Activity in Proportion to Time of Irradiation

[1] See footnote, Page 68.

"Inasmuch as complete 'destruction' of the two spore-forming bacteria B. subtilis and M-S was not obtained in the previous experiment (No. 6), following 7 hours exposure to cobalt-60, a second study was outlined in which frequent sampling was carried out on the same organisms following irradiation with the isotope over a period of 24 hours. The suspensions used in this investigation were prepared in saline-gelatin (0.5%) solution from beef extract agar slants that had been incubated at 37°C for 96 hours. Two ml quantities of the specimen were removed from the irradiated samples at 4-hour intervals, and the latter, as well as the unirradiated controls were stored in the refrigerator (2°C) until plated. Plating was carried out 2 hours after irradiation of the final samples (24 hour). Dilutions of the samples, plating, and counting were essentially the same as described in experiment No. 1. Table XVI presents the results of this experiment.

TABLE XVI

COUNT OF SPORE-FORMING BACTERIA VERSUS IRRADIATION DOSAGE

Time of Irradiation (hrs)	B. subtilis	Per cent Reduction	M-S	Per cent Reduction
0 (unirradiated)	18,000,000	0	97,250,000	0
4	960,000	94.6	8,750	99.99
8	202,000	98.8	3,280	99.99
12	20,500	98.8	550	99.99+
16	14,450	99.99	80	99.99+
20	75	99.99+	10	99.99+
24	0	100.0	0	100.0

"Fig. 46 shows the difference in growth of colonies of M-S organisms after different irradiation dosages.

"Since complete destruction of several cultures of bacteria were obtained in the irradiation studies up to this point, presentation of some of these data in a graph was carried out to represent the numbers of survivals of the test organisms as plotted against the dosage of cobalt-60 used in treating the suspension (irradiation time). Fig. 37 (shown previously, page 52) presents a graph of the irradiation results compiled against P. vulgaris, E. coli, B. subtilis and the Gram-positive sporulating bacillus isolated from milk (M-S) representing the results of experiments Nos. 5 and 7.

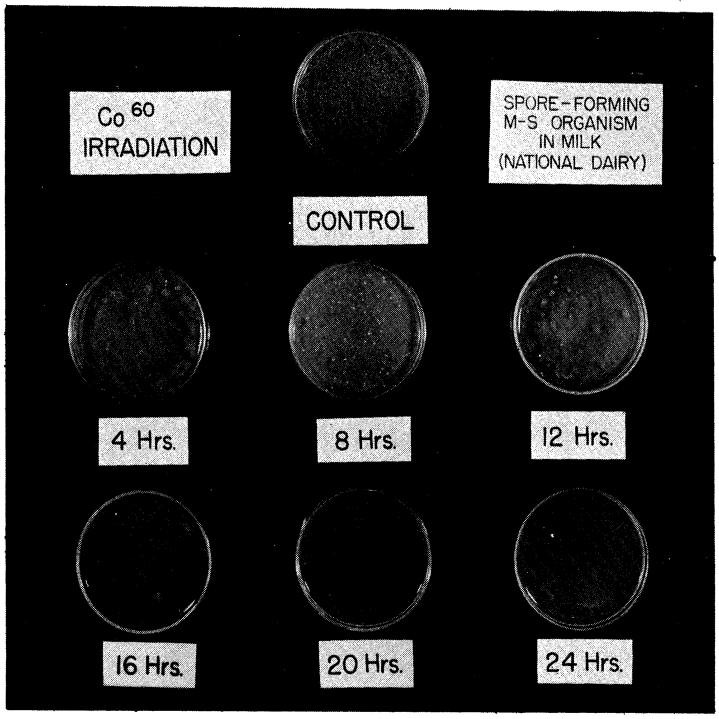

Fig. 46. Photograph Showing Effects of Gamma on Spore Forming (M-S) Microorganisms in Milk

"h) Experiment No. 8 The Effect of Gamma Radiation (Cobalt-60) on Spores of Molds, Aspergillus niger and Penicillum notatum

"With the evidence obtained in experiment No. 6, that the spore-forming fungi, A. niger and P. notatum, were markedly reduced in numbers following irradiation with cobalt-60, a second study was carried out on these two mold species in which the time intervals of irradiation with the isotope was spaced at shorter periods than before and sampling made at hourly intervals up to and including 4 hours.

"The spores of the molds, grown on Sabourauds agar slants for 24 days, were removed with saline-gelatin (0.5%) solution and irradiated in the latter medium. Immediately after radiation, diultions of the specimens were made and plating carried out in Sabouraud's maltose agar. Colony counts were made after the plates had been incubated at room temperature (24-26°C) for 5 days. Table XVII presents the results of this experiment (see page 72).

"i) Experiment No. 9 The Effect of Gamma Radiation (Cobalt-60) on Spore-Forming Bacteria, Bacillus subtilis and a Gram-Positive sporulating Bacillus Isolated from Milk (M-S)[1], with Short Periods of Irradiation

"Experiments Nos. 6 and 7 present the results of cobalt-60 irradiation upon the spore-forming bacilli, B. subtilis and the organism M-S that was

1 See footnote, page 68.

TABLE XVII

EFFECTS OF IRRADIATION ON MOLDS

Time of Irradiation Hours	A. niger	Per cent Reduction	P. notatum	Per cent Reduction
0 (unirradiated)	45,000,000	0	11,250,000	0
1	125,000	99.66	15,000	99.86
2	5,000	99.98	125	99.99
3	25	99.99	0	100.0
4	0	100.0	0	100.0

isolated originally from milk. In the latter studies a significant reduction in numbers of total bacteria and their spores were noted when samples were first removed at the 4-hour period. The present experiment was carried out to determine the effects of the radiation upon the same organisms when the latter were exposed for shorter periods of time. These time intervals of selecting specimens ranged from 15 minutes through 4 hours.

"In addition to irradiating the M-S culture suspended in saline-gelatin solution, a suspension of the organism was also prepared in evaporated (sterile) milk which was reconstituted to its original concentration with an equal amount of distilled water. The results of this investigation are given in Table XVIII.

TABLE XVIII

EFFECT OF SHORT PERIODS OF IRRADIATION ON SPORE-FORMING BACTERIA

Time of Irradiation	M-S in Milk	% Red.	M-S in Saline-Gelatin	% Red.	B. subtilis in Saline-Gelatin	% Red.
0	24,402,000,000	0	7,406,000,000	0	232,000,000	0
15 min	23,600,000	99.90	67,500,000	99.5	131,000,000	43.55
30 min	7,700,000	99.91	7,500,000	99.95	57,000,000	75.44
1 hr	1,600,000	99.99+	2,100,000	99.99+	34,000,000	85.11
2 hr	427,000	99.99+	621,000	99.99+	22,800,000	90.20
3 hr	292,000	99.99+	432,000	99.99+	18,400,000	91.87
4 hr	217,000	99.99+	237,000	99.99+	9,400,000	95.95

"j) Experiment No. 10 The Effect of Gamma Radiation (Cobalt-60) on Spore Suspensions of Bacillus subtilis and a Gram-Positive Sporulating Bacillus Isolated from Milk (M-S)[1]

"In the previous studies carried out on the effects of cobalt-60 radiation on the spore-forming bacilli (experiments Nos. 6, 7, and 9), no attempt was made to provide a spore suspension of the organisms in the apparent absence of vegetative forms of the respective organism. Conceivably, therefore, much of the initial reduction in numbers of organisms in the samples may be attributed to the effects of irradiation upon the vegetative forms of the bacteria with a subsequent "leveling off" in numbers which might be due to the resistance of the spores to gamma irradiation. This latter point was the subject of the following experiment. Sixteen-day-old cultures of B. subtilis and M-S bacteria, grown on beef extract agar, were washed from the slants with saline-gelatin solution. The suspensions were then heated in a water bath at 70°C for 30 minutes to destroy the vegetative forms of the organisms. The latter samples were subjected to gamma irradiation for a total period of 24 hours, during which samples of the treated suspensions were removed at intervals of 1 hour and 45 minutes, and again at 4 hours. Dilutions, plating and colony counts were made as indicated in experiment No. 1. The results of this study are presented in Table XIX.

TABLE XIX

EFFECT OF LONG PERIODS OF IRRADIATION ON SPORE-FORMING BACTERIA

Time of Irradiation	B. subtilis	Per cent Reduction	M-S	Per cent Reduction
0 (unirradiated)	27,500,000	0	1,000,000	0
1 hr 45 min	3,900,000	49.31	220,000	78.91
4 hrs	890,000	96.83	17,000	96.62
24 hrs	0	100.0	0	100.0

"k) Experiment No. 11 The Effects of Gamma Radiation from Cobalt-60 and X-rays upon Yeast (Saccharomyces cerevisiae)[2]

"A strain of yeast (Saccharomyces cerevisiae) was grown on Sabouraud's Maltose agar slants at 37°C for 48 hours. The culture was removed from the

[1] See footnote, page 68.

[2] The studies on the effects of radiations on yeasts were designed and conducted by Miss Gladys Torres, graduate student in the Department of Bacteriology.

surface of the slant in saline-gelatin (0.5%) solution and the suspension centrifuged. The supernatant liquid was removed and discarded. The packed cells were resuspended in saline-gelatin and again centrifuged. After removal of the supernatant fluid the cells were again suspended in fresh saline-gelatin solution, and this material was used in the irradiation studies. Control unirradiated samples were kept at room temperature until completion of the irradiation experiments at which time all test samples were diluted and plated in Sabouraud's medium. The details of dilution, plating, and counting the colonies are described in experiment No. 1. The results of this study are presented in Tables XX and XXI.

TABLE XX

LETHAL EFFECTS OF COBALT-60 ON YEAST CELLS

Time of Irradiation (hrs)	Saccharomyces cerevisiae Counts (colonies per ml)	Per cent Reduction
0 (unirradiated)	2,750,000,000	0
1	200,500,000	92.72
2	26,250,000	99.05
3	32,000	99.99+
4	25,000	99.99+

TABLE XXI

LETHAL EFFECTS OF X-RAYS ON YEAST CELLS

Dosage in Roentgens	Saccharomyces cerevisiae Counts (colonies per ml)	Per cent Reduction
0	3,250,000,000	0
86,000	55,250,000	98.30
132,000	10,000	99.99+
218,000	9,000	99.99+
344,000	5,500	99.99+

"Remarks: From the data given in Tables XX and XXI it is evident that the degree of lethal effects from gamma rays emitted from cobalt-60 and x-rays are comparable. While there appears to be some differences in the counts in the initial irradiation samples (1 and 2 hours, respectively), the percentage reduction variation are within the limits of error encountered in the test procedure.

"1) Experiment No. 12 The Effect of Gamma Radiation (Cobalt-60) on Yeast (Saccharomyces cerevisiae): With Reference to Age of Culture and Susceptibility to Radiation Action

"There are several references in the literature indicating that the age of cultures of microorganisms plays an important part in the degree of susceptibility of the organisms to irradiation effects. In general, the evidence available suggests that "young" cultures (24-48 hrs) are more susceptible to the lethal effects of x-rays than are "old" cultures of the same organisms. To determine whether or not the same correlation in difference of age of a culture of yeast cells to the effects of irradiation to gamma rays (cobalt-60) may be noted was determined in the present experiment. The culture used in the study was the same strain of Saccharomyces cerevisiae used in the previous experiment (No. 11). The age of the organism suspension prepared for radiation and unirradiated control was 8 weeks. No attempt was made to prevent the culture medium from desiccating during the incubation at 37°C for the period indicated. Suspension of the organisms in saline-gelatin, washing of the cells, etc., was carried out as indicated in the previous experiment (No. 11). The results of this study are presented in Table XXII. Included in the Table are the data given in Table XX, in which a 48-hour culture of the organism was exposed to the same radiation source. The latter results are included here for comparison purposes.

TABLE XXII

AGE OF YEAST CULTURE VERSUS SUSCEPTIBILITY TO LETHAL EFFECT OF COBALT-60

Time of Irradiation (hrs)	Saccharomyces cerevisiae Counts (colonies per ml) 48-hr culture	% Red.	1,344*-hr culture	% Red.
0 (unirradiated)	2,750,000,000	0	42,250,000	0
1	200,000,000	92.72	12,000	99.99+
2	26,250,000	99.05	0	100.0
3	32,000	99.99+	0	100.0
4	25,000	99.99+	0	100.0

* 8-week-old culture.

"Remarks: Examination of the data given in Table XXII reveals that contrary to several published reports on the resistance of "old" cultures of yeast cells to radiation effects, the present results indicate that the converse is true, i.e., the "young" cultures of the organisms are more resistant to the effects of gamma rays than are the "old" organism suspensions. The latter is not only based on the initial reduction noted following exposure for 1 hour as reported in Per cent Reduction, but also on the analysis of the unirradiated control, and the first counts at the 1-hour period will indicate the far greater mortality rate of the "old" sample as compared to the "young" culture (48 hour).

3. "The Effects of Radiations on Vitamins Essential for the Nutrition of Microbial Growth

"a) Experiment No. 1 The Effect of Gamma Radiation (Cobalt-60) on Niacin (Nicotinic Acid)

"A study was made to determine the effects of gamma radiation (cobalt-60) upon one of the vitamins that is essential as a growth factor for Lactobacillus arabinosus. The microbiological assay was performed on the irradiated and unirradiated (control) medium using the L. arabinosus strain No. 17-5.

"Methods: A reference stock solution of niacin, previously dried to a constant weight in a desiccator was prepared in 500 ml of absolute alcohol. This stock solution was kept at 5°C when not in use.

"The culture used in this study was obtained from the American Type Culture Collection,,Washington, D. C. The organism was maintained in "stab" agar culture of Difco Micro-assay agar. Transplants of the stock culture were made at weekly intervals and maintained at 5°C after an initial incubation at 37°C for 24 hours. For assay procedures, transplants were made from the "stab" culture to 10 ml of Difco Micro-Inoculum Broth. The latter cultures were incubated at 37°C for no more than 24 hours after which the suspension was centrifugated under aseptic conditions. The supernatant broth was discarded and the packed cells resuspended in saline (0.85% NaCl) solution (10 ml). One drop of this suspension was added to each assay tube containing the vitamin-deficient medium and varying amounts of the supplement, niacin substrate. The suspension of organisms was delivered to the assay tubes with a sterile tuberculin syringe and a No. 19-gauge needle.

"The Difco dehydrated Niacin Assay Medium used in this study is composed of the following ingredients:

Difco Niacin Assay Medium (Niacin Deficient)

Bacto-Vitamin Free Casamino Acids	12 g
Bacto-Dextrose	40 g
Sodium Acetate	20 g
l-cystine, Difco	0.4 g
d-l tryptophane	0.2 g
Adenine	0.02 g
Guanine	0.02 g
Uracil	0.02 g
Thiamine Hydrochloride	0.0002 g
Calcium Pantothenate	0.0002 g
Pyridoxine Hydrochloride	0.0004 g
Riboflavin	0.0004 g
p-Aminobenzoic Acid	0.0002 g
Biotin	0.0000008
Dipotassium Phosphate	1.0 g
Monopotassium Phosphate	1.0 g
Magnesium Sulfate	0.4 g
Sodium Chloride	0.02 g
Ferrous Sulfate	0.02 g
Manganese Sulfate	0.02 g

"The above formula is reconstituted in the amount of 75 grams to 1000 ml of distilled water by heating just to boiling and then 5 ml amounts distributed in test tubes. The medium was sterilized by autoclaving for no more than 15 minutes at 15 pounds (121°C) steam pressure.

"Irradiation: For the irradiation study, 10 ml of the niacin stock solution was added to a 15 x 120 mm screw-capped pyrex test tube (sterile) and the solution exposed to cobalt-60 for 24 hours. A similar sample was maintained in the laboratory at room temperature as a control. The pH of the solutions was 5.1. Microbiological assays on the solutions were performed as soon as possible after the irradiated sample was removed from the cobalt vault.

"Microbiological Assay for Niacin: Both the irradiated and unirradiated controls were diluted 1:500 with sterile distilled water in a sterile 500 ml volumetric flasks. Amounts equivalent to the following concentrations of niacin were placed aseptically in duplicate tubes containing 5 ml of the niacin-deficient assay medium: 0.00, 0.05, 0.10, 0.20, and 0.30 gamma. The volume of liquid in the tubes was then brought up to 10 ml with sterile distilled water. After thoroughly shaking the solution in the tubes, there was added to each one drop of the culture of L. arabinosus, as indicated earlier in this test.

"Turbidimetric measurements were made on the solutions after 20 hours incubation at 37°C. A photoelectric colorimeter (Lumetron, Model G, Photo Volt

Company, New York) was used for the latter procedure using the No. 530 filter. Two blanks were used, one containing 5 ml of niacin assay medium and 5 ml of distilled water but no bacteria inoculum, and another blank, prepared as before, but inoculated with the test organism and incubated as indicated above. The noninoculated control also served as a control on the aseptic technique that is necessary to carry out the microbiological assay of vitamin solutions. Table XXIII presents the results of the preliminary test on the effects of cobalt-60 on niacin.

TABLE XXIII

THE EFFECTS OF COBALT-60 IRRADIATION ON NIACIN (24-HOURS EXPOSURE)

Original Amount of Niacin Added	Assay Concentration of Niacin in Gammas	
	Nonirradiated	Irradiated
Control (no niacin)	0.00	0.00
0.050	0.058	0.048
0.100	0.100	0.083
0.200	0.200	0.125
0.300	0.300	0.300

"Results: The galvanometer readings were converted into optical density by the relation L = 2-log G. A standard curve was drawn using the values obtained from the assay of the nonirradiated sample of niacin. Values of the various concentration levels from the irradiated sample were then read from the standard curve.

"Within the limits of experimental error of this test procedure, the data in Talbe XXIII indicate that exposure of the niacin solution to cobalt-60 for a period of 24 hours will not appreciably alter the vitamin insofar as its growth-promoting ability for L. arabinosus is concerned.

"Additional studies are contemplated in which other vitamins, in solution and in the dry state, will be exposed to gamma and x-rays.

"b) Experiment No. 2 The Effect of Gamma Radiation (Cobalt-60) on a Chemically Defined Medium which Supports the Growth of Lactobacillus arabinosus

"The data given in the preceding experiment (No. 1) indicate that exposure of an aqueous solution of niacin to 24 hours' irradiation to cobalt-60 did not destroy the growth-promoting component of the vitamin for L. arabinosus. Since there are twenty ingredients, including vitamins, etc., in the basal niacin-deficient medium (Difco), as listed in the previous experiment, it appeared desirable to determine whether or not irradiation of the latter would

result in the inactivation of some of these components in the formula, or the production of toxic substances therefrom which would inhibit the growth of L. arabinosus in the presence of its essential vitamin, namely, niacin.

"Method: Fifteen milliliters of double-strength Niacin medium (Difco -see preceding experiment) were irradiated, with sufficient niacin added, for 6 hours with cobalt-60. The amount of niacin added to the medium provided a concentration of 0.1 gamma which was to be tested in the final dilution in each assay tube. Following irradiation, 5 ml of the solution was added per assay tube and the latter volume diluted to 10 ml with sterile distilled water. Thus, the concentration of the medium was the same as in the determination of niacin in the preceding experiment, and in this study the concentration of niacin per tube was 0.1 gamma, or the amount indicated above.

"Each tube of medium was inoculated with one drop of a 20-hour washed culture of L. arabinosus and allowed to incubate at 37°C for 20 hours. Turbidimetric measurements were made using appropriate blank controls. The optical densities of the control and irradiated samples are given in Table XXIV. Duplicate sets of tubes were prepared in each instance (Tubes 1 and 2).

TABLE XXIV

THE EFFECT OF COBALT-60 ON A BACTERIOLOGICAL MEDIUM

Tube No.	Nonirradiated	Irradiated
1	0.143	0.131
2	0.137	0.125

"Results: From the data given in Table XXIV it is evident that irradiation of the Niacin Assay Medium to which was added the essential vitamin, niacin, for the propagation of L. arabinosus, did not result in any apparent change in the medium that could be detected in the microbiological assay or growth of the organism.

4. "Effects of Gamma Radiation (Cobalt-60) on Various Proteolytic Enzyme Systems

"a) Methods:

The following are the methods that were employed in the preparation of the enzyme suspensions for irradiation and the determination of their activity:

"1) Pepsin: The pepsin used in the experiments was a recrystallized product of porcine origin obtained from Armour and Company. For irradiation, 40 milligrams were dissolved in distilled water adjusted to a pH of 1.1 with hydrochloric acid. Twenty ml of this suspension were irradiated in 12 x 60 mm pyrex-glass test tubes. At indicated time intervals, 3 ml samples were withdrawn. An unirradiated control containing the same amount of enzyme suspension was kept for the entire period of irradiation. Proteolytic activity was determined 20 hours after irradiation. A preliminary test performed immediately after the last irradiation indicated that there was little loss in activity after storage of the solutions at 5°C.

"The hemoglobin method of Anson[1] was used to determine proteolytic activity. The hemoglobin substrate was prepared by dissolving 2.6 grams of Bacto-Hemoglobin (Difco) in 100 ml of distilled water. One ml of a 1:1000 solution of aqueous merthiolate was added as a preservative. The hemoglobin solution was stored at 5°C when not in use. Prior to the determination, 1 ml of 0.3N hydrochloric acid was added to 4 ml of the hemoglobin solution in 15 x 150 mm test tubes. This acid-hemoglobin solution was allowed to stand at 25°C for at least an hour, after which 1 ml of the enzyme solution was added. The tubes were thoroughly shaken and then allowed to stand at 25°C for exactly 20 minutes. At that time 10 ml of 0.3N trichloroacetic was added to precipitate the nonhydrolyzed protein. After standing for at least 10 minutes, the suspension was filtered through a No. 3 Whatman filter paper.

"To 5 ml of the trichloracetic acid filtrate, 10 ml of 0.5N sodium hydroxide were added, followed by 3 ml of phenol reagent diluted 1:2 and prepared according to the method of Folin-Ciocalteau.[2] The color developed was read within 10 minutes in a photoelectric colorimeter with a No. 650 filter and a blank adjusted to 100 per cent transmission. The blank was treated in the same manner as outlined above, except 1 ml of distilled water pH 1.1 was substituted for the enzyme solution.

"2) Trypsin: The solution of trypsin used for irradiation was prepared as follows: 20 milligrams trypsin (Difco 1:250 a pancreatic extract) was dissolved in 50 ml of distilled water adjusted to a pH of 2.7 with hydrochloric acid. Ten ml of this enzyme solution was irradiated in 12 x 60 mm pyrex-glass test tubes. Individual test tubes were withdrawn at the indicated time intervals. A control was kept at room temperature during the period of irradiation.

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[1] Anson, M. L., "The Estimation of Pepsin, Trypsin, Papain and Cathepsin with Hemoglobin." J. Gen. Physiol. 22 79-89 (1938).

[2] Folin, O., and Ciocalteau, V., "On Tyrosine and Tryptophane Determination in Proteins." J. Bio. Chem. 23 627-650 (1927).

"Proteolytic activity was determined after the last irradiation. For determination, the casein method[1] was used. The casein solution was prepared by dissolving 1 gram of Borden's casein in 100 ml of phosphate buffer at a pH of 7.4. This solution was stored at 5°C and made up fresh each week. Prior to the determination, 1 ml casein solution was placed in test tubes and allowed to stand in a water bath adjusted to 37.5°C, for at least 20 minutes. After this period, 1 ml of the enzyme solution was added to the tube, the solution shaken thoroughly, and replaced in the water bath for exactly 20 minutes. The nonhydrolyzed protein was then precipitated by the addition of 3 ml of 5% trichloro-acetic acid. After standing at room temperatuer for 1 hour, the suspension was centrifuged in order to remove the precipitate.

"To 1 ml of the supernatant, 1 ml of 0.0025 M copper sulfate, 8 ml of 0.5 M sodium hydroxide, and 3 ml of Folin-Ciocalteau reagent diluted 1:2 were added. The per cent transmission was read in a photoelectric colorimeter using a No. 650 filter and appropriate blank.

"3) Chymotrypsin: The chymotrypsin used in this experiment was a recrystallized product of bovine origin obtained from Armour and Company. Twenty milligrams of the enzyme was dissolved in 50 milliliters of distilled water adjusted to a pH of 2.7 with hydrochloric acid. Four milliliters of the enzyme solution was irradiated in 5 x 35 mm test tubes. Individual tubes were withdrawn at the indicated time intervals. A control was kept at room temperature during the time of irradiation.

"The activity of the chymotrypsin was determined after the last irradiation. The casein method was also used for the assay of the latter enzyme.

"4) Papain: For this experiment 500 milligrams of Papain (Merck) was dissolved in 100 ml of distilled water and the solution filtered. In order to activate the enzyme, 10 drops of 2 N sodium cyanide was added to 1 ml of the enzyme solution. After standing at room temperature for 10 minutes 18.9 ml of distilled water was added to the latter. The solution was further diluted by addition of 10 ml of distilled water containing 5 drops of 2 N sodium cyanide. The pH of the enzyme solution was 5.7.

"For irradiation, 5 ml was placed in 5 x 35 mm test tubes and individual tubes were withdrawn at the indicated time intervals. A control tube containing unirradiated enzyme solution was kept at room temperature during the period of irradiation of the test sample.

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[1] Northrop, J. H., Kunitz, M., and Herriott, R. M., Crystalline Enzymes 2nd Ed. New York, Columbia University Press, 1948, pp. 308-309.

"Proteolytic activity was determined after the last irradiation. The method used for the determination was the same as that used for trypsin.

"b) Results of Studies on Effects of Cobalt-60 in Enzyme Systems:

"The per cent transmission of the various solutions was converted to optical density (L) by the formula, L = 2 - log G. Tyrosine values were read from a standard curve prepared previously, using known concentrations of tyrosine. The amount of tyrosine liberated by the hydrolysis of the proteins by the enzymes was taken as an index of the activity of the enzymes. The results of the various experiments are expressed as milligrams of tyrosine released. Tyrosine values were plotted against time of irradiation and are presented in Fig. 47.

TABLE XXV

EFFECTS OF GAMMA IRRADIATION ON ACTIVITY OF PEPSIN

Time of Irradiation	Milligrams of Tyrosine
0	0.0860
51 min	0.0720
1 hr 46 min	0.0535
2 hr 26 min	0.02875
3 hr 41 min	0.00240
4 hr 28 min	0.00076

TABLE XXVI

EFFECT OF GAMMA IRRADIATION ON ACTIVITY OF TRYPSIN, CHYMOTRYPSIN, AND PAPAIN

Time of Irradiation (hrs)	Milligrams of Tyrosine		
	Trypsin	Chymotrypsin	Papain
0	0.0945	0.0945	0.0635
1/2	----	0.0835	0.04815
1	0.0762	0.0505	0.0275
2	0.017	0.00345	0.01625
3	0.0040	0.00225	0.019
4	0.0035	0.00075	0.017

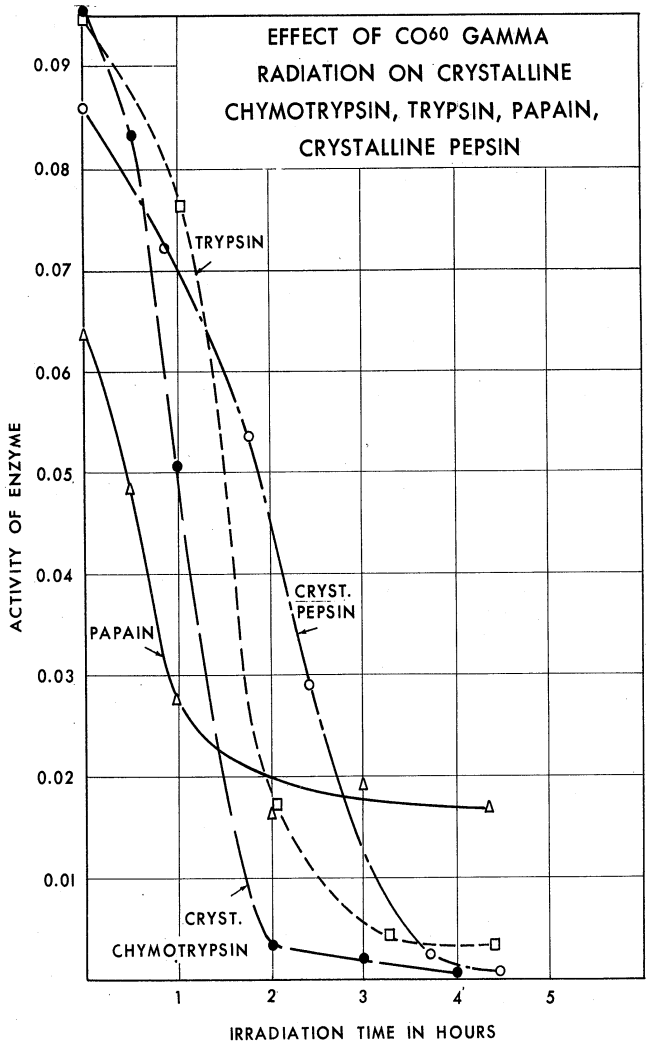


Fig. 47

D. STERILIZATION OF PACKAGED PHARMACEUTICALS

1. Introduction

The sterilization of heat-sensitive pharmaceuticals, especially those used for injection, appears to be one of the most promising uses of radioactive fission products.

The Upjohn Company of Kalamazoo, Michigan, has done considerable research using a two-million-volt electron accelerator to sterilize packaged pharmaceuticals. Professor L. E. Brownell visited the Upjohn research laboratories on October 19, 1951, and discussed the irradiation of drugs with Dr. O. R. Woods of this company. The research group at Upjohn seemed to be quite encouraged with the results which they have obtained to date but were

at liberty to divulge only limited information. Professor Brownell suggested the possibility of cooperative research; however, as yet, no such program has been established.

A similar discussion was held at Parke, Davis and Company of Detroit, Michigan, who have also made preliminary tests on the sterilization of pharmaceuticals by irradiation. The Parke, Davis and Company's representatives expressed great interest in the possibilities of such processes and in cooperative research relative to such an investigation. It was agreed that in a cooperative preliminary experiment, Parke, Davis and Company would supply samples of packaged pharmaceuticals for experimental study and would assay these samples and that the University of Michigan would perform the irradiation with the 1000-curie cobalt source. The results of these tests would be available to both parties and will be reported to the Atomic Energy Commission.

The pharmaceuticals selected for the initial experiment are shown in Fig. 48. A wide variety, including antitoxins, hormones, and vitamins, were tried. All were in aqueous form except Antuitrin G, which was a dry powder. Fig. 49 shows some of the samples irradiated for 24 hours as compared to the controls. The glass containers browned appreciably as a result of irradiation. The report of the experiment and the results and conclusions, as prepared by Dr. John Controulis of Parke, Davis and Company, follow.

2. Report by Parke, Davis and Company

"Three samples of each product were contaminated [with B. subtilis] and reserved as controls. Another group of three samples of each product were contaminated and irradiated for one hour. A third group of three samples of each product were contaminated and irradiated for 24 hours. A similar set of three groups of three samples of each product were not contaminated, but one group was left as control, another for one-hour irradiation, and the third for 24-hour irradiation. The contaminated set of samples was used for sterility evaluation and the uncontaminated set for the biological activity evaluation.

"a) Assay of Irradiated Samples:

"1) Activity Assay: The uncontaminated irradiated and control samples were submitted to various assay sections within the plant to ascertain whether or not any deterioration of the pharmaceutical had occurred as a result of irradiation. The methods of assaying the various products were as close as possible to the accepted assay procedures regularly used in manufacturing.

"Calcium Gluconate samples were observed visually to detect any breakdown of the solution.

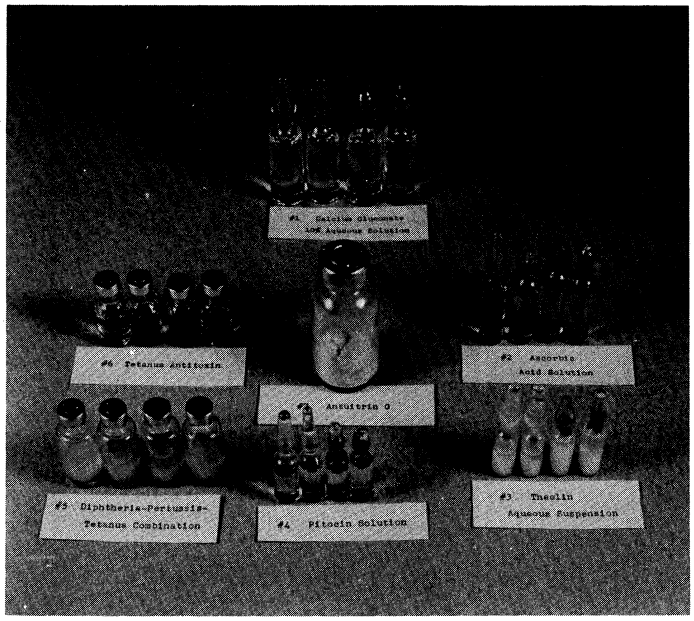


Fig. 48. Photograph of Drug Samples before Irradiation

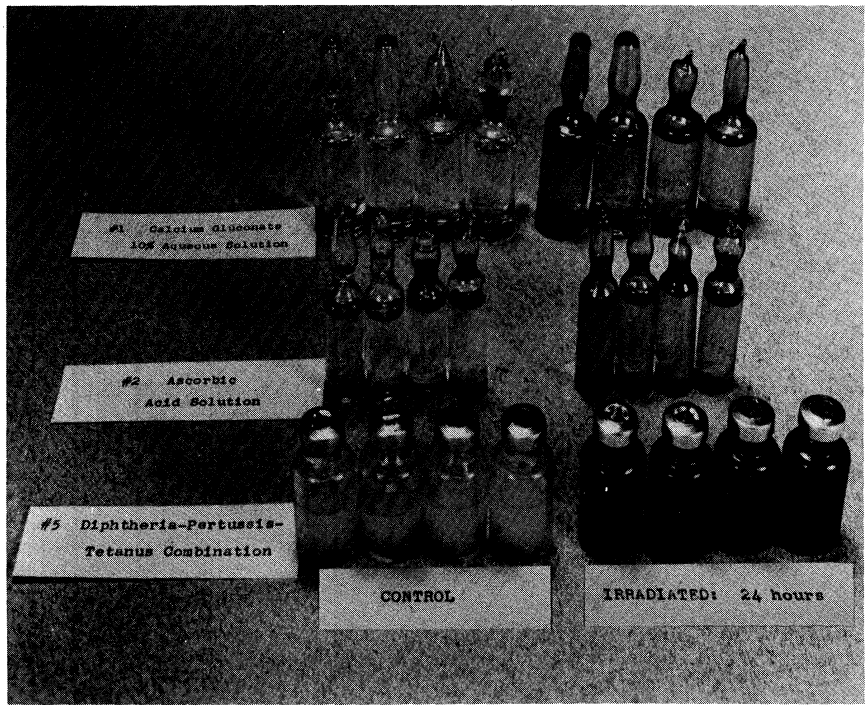


Fig. 49. Photograph of Drug Samples after 24 Hrs Gamma Irradiation in Cobalt-60 Vault

"Ascorbic acid samples were submitted to the Vitamin Section for colorimetric assay of the active ingredient.

"Pitocin samples were assayed by Mr. Rowe for oxytoxic activity.

"Theelin samples were assayed via ultraviolet absorption spectrum to determine any structural change in the estrone molecule.

"Tetanus Antitoxin samples were assayed in the Biological Department for tetanus units.

"Diphtheria-Tetanus-Pertussis samples were assayed via pertussis agglutination and also for Tetanus-combining power.

"2) Sterility Assay: The contaminated irradiated and control samples were submitted to Mr. Marshall of the Biological Department for sterility assay.

"b) Results:

"The results of the activity assays and the sterility assays appear in Table XXVII (see page 87).

"c) Conclusion:

"From the tabular data it is seen that gamma radiation accomplished sterilization in the case of each and every product at the 24-hour exposure level. While there may have been a statistically important reduction in the number of viable organisms after one hour of exposure to the radiation, there were always enough organisms present to render the samples nonsterile at this lower dosage. As to the effects of the irradiation upon biological activity, the following observations have been made:

"No. 1. There appeared to be no detrimental effect on the 10% Calcium Gluconate solution. (Since no reliable assay is readily available for Calcium Gluconate, evaluation is based on the visible appearance of the solution, turbidity, amorphous deposit, or crystalline growth being signs of deterioration usually noted with this product.)

"No. 2. The assays of irradiated Ascorbic Acid solutions showed no statistical departure from that of the unirradiated sample.

"No. 3. While there is a decrease in Theelin content of the irradiated samples compared to the control, the lower values are still above the label claim.

"No. 4. Pictocin is the most clear-cut example of a pharmaceutical drastically affected by gamma radiation.

"No. 5. Since the triple combination of Diphtheria-Tetanus-Pertussis is a difficult product to assay, Pertussis agglutination and Tetanus-combinating power were used to determine the effect of the irradiation. While the irradiated samples showed the same response as the unirradiated controls, the value for

TABLE XXVII

RESULTS OF GAMMA-RAY STERILIZATION

Sample Number and Product	Tests	Uncontaminated Irradiation Time 0 hr	1 hr	24 hr	Contaminated Irradiation Time 0 hr	1 hr	24 hr
1. Amp No. 239 Calcium Gluconate	Activity	OK	OK	OK			
	Sterility				C*	C	S*
2. Amp. No. 315 Axcorbic Acid	Activity (mg/cc)	275.5	283.3	273.0			
	Sterility				C	C	S
3. Amp No. 304 Theelin Aq. Suspension	Activity (mg/cc)	2.70	2.21	2.19			
	Sterility				C	C	S
4. Amp No. 160 Pitocin	Activity	100%	50%	0.02%			
	Sterility				C	C	S
5. Biol. No. 2091 Diph. Tet. Pert. Combination	Agglutination	+	+	+			
	Tetanus Combining Power (Units/ml)	0.8	0.8	0.8			
	Sterility				C	C	S
6. Tetanus Antitoxin	Activity (u/ml)	1900	1900	1500			
	Sterility				C	C	S
7. Antuitrin G	Activity						
	Sterility				C	C	S

* C = Contaminated
S = Sterile

Tetanus-combining power of the control was much lower than it should have been. Therefore, this evaluation is probably not a valid one and, in further studies, this point will have to be cleared up.

"No. 6. In the case of the Tetanus Antitoxin, the radiation dosage used for sterilization proved to cause appreciable and serious deterioration of the product. However, it may be possible to achieve sterilization at a lower dosage, say 12 hours, and cause simultaneously only an unimportant or no loss of activity.

"No. 7. While the radiation accomplished sterilization of this lot of Antuitrin G, a difficult one to sterilize, insufficient material was at hand to irradiate and check for retained potency. This point can be ascertained in the second stage of study of the method. However, it is of significance to note that the irradiation effected sterilization even though the product was in the dry state.

"As for the discoloration of the glass, irradiation for one hour caused a slight but definite tinting of the glass. This discoloration is strong enough to be recorded by the camera when one-hour irradiated samples were photographed along side unirradiated samples. In the case of the 24-hour irradiated samples, the discoloration was severe, the glass darkening to a deep amber or brown color.

"Comparing the gamma-radiation method with either the x-radiation or electron-radiation methods, it is readily seen that the gamma-radiation offers the most promise. The x-radiation method failed to achieve sterilization, even at high dosages, when the pharmaceuticals were irradiated in finished package form. General Electric researchers refused to attempt sterilization via electron-radiation of the finished packages and achieved sterilization only by irradiating thin polyethylene envelopes containing the pharmaceuticals. In addition, the gamma-radiation appears to warrant the least investment in equipment and operating cost when compared with the x- or electron-radiation methods.

"d) Future Plans:

"To overcome the first serious obstacle in the way of adopting gamma-radiation as a sterilization method, glass companies are being contacted to see if a noncoloring glass is or can be made available commercially. If such a glass proves to be satisfactory from the standpoints of withstanding radiation without coloring, of fabrication into ampules, and of economic considerations, then a duplication of a present study will be made using products contained in the new glass. Should this second study prove practical, then estimates of necessary equipment (operational and protective), of "hot" materials as the source and of costs of the program will be drawn up for consideration by the company. If such approval is granted, then more thorough tests, evaluations, clinical trials, etc., should be carried out before actual installations and constructions are begun.

"Should any of the above steps show the method to be impractical, it is the desire of the writer to cooperate with Dr. Brownell in publishing the data gathered thus far. To this end, approval of the publication committee will be solicited."

PART IV

EXPLORATORY RESEARCH ON NEW IDEAS (SUBPROJECT M943-E)

A. Introduction

The organized research program for the initial investigations consists of subprojects M943-A, B, C, and D. It is realized that some of the possible uses of the fission products being investigated will prove negative. Also it is hoped that new and better ideas regarding uses for the fission products will develop as the investigation progresses. With this in mind, subproject M943-E was created to provide means of exploratory research on new ideas prior to establishing an organized research program. Several ideas have been considered, but as yet exploratory research has been conducted on only two such ideas. The first exploratory study has been made on the alloys and ceramic glazes which can be produced from strontium so as to provide a more satisfactory method of using strontium-90. The second of these ideas has been the conversion of a 220-KV x-ray machine, which was donated to the University by St. Joseph's Mercy Hospital, into an electron gun to be used in studies of uses of beta radiation.

B. Experiments on the Utilization of Strontium-90 as a Source of Beta Radiation

Supervisors: L. Thomassen, Professor of Chemical and Metallurgical Engineering; L. E. Brownell, Associate Professor of Chemical and Metallurgical Engineering. R. Dean Pierce, Research Assistant.

High-velocity electrons (beta radiation) are known to influence chemical reactions. A convenient source of this radiation would be of importance in the further investigation of the promotion of chemical reactions by radiation. The effect of the beta rays is considered to be activation or ionization of the reacting molecules. Many of the fission products from a nuclear reactor emit such radiation, but most of them simultaneously emit gamma radiation. Strontium-90 is present in relatively large quantities in fission products and has the desirable property of not emitting gamma rays. Furthermore, when strontium disintegrates it forms yttrium-90 which, in turn, also emits only beta particles, forming zirconium-90 which is stable. Strontium-90 has a relatively long half-life (25 years) which makes it suitable for

sustained operations. The maximum energies of the beta particles emitted by strontium and yttrium, are 0.61 and 2.35 mev, respectively.

If these radiations are to be utilized for industrial operations, it is imperative that the radioactive materials be mechanically fixed in such a way that they would not be carried along with the reaction products of a chemical reaction and thereby become a health hazard. This is a serious problem in the case of strontium-90-yttrium-90 because the high energy particles may produce recoils. These recoil energies then may give rise to the phenomenon of "creep" resulting in the spread of radioactive material.

Experiments have been carried out along two lines: (1) to utilize strontium as an alloying constituent, (2) to incorporate it in a glaze on porcelain or a similar refractory material. Alloys and glazes of strontium were investigated using ordinary strontium rather than strontium-90. Three alloys were made up of ordinary strontium: one with 25 per cent by weight strontium, the balance magnesium; and two with 25 and 50 per cent strontium, the balance aluminum.

All three samples had a metallic lustre which was retained on exposure to air. Samples were pulverized and screened to obtain two grades of fines: one + 100 mesh - 48 mesh, the other - 100 mesh. The six samples were exposed to air for several weeks but showed no significant increase in weight, nor did they change in appearance. Quantitative experiments to find their resistance to oxidation at higher temperatures are under way.

Qualitative experiments on the acid resistance of the aluminum alloy with 25 per cent strontium showed that it was comparatively resistant to hydrochloric acid.

The advantage of a strontium glaze on a ceramic over the metal in an alloy is that the ceramic may be used at relatively high temperatures. The amount of radioactivity which may be obtained from such a glaze is significant. Computations of the amount of radiation that can be expected from a thin layer of strontium oxide in a glaze have been made on the basis of values stated in the Oak Ridge Isotope Catalog, assuming a layer 0.002 in. thick and neglecting absorption. Such a layer is equivalent to about 12 curies per square foot of surface. To obtain a higher energy output per unit volume, porous media such as small ceramic spheres, Raschig rings, etc., may be glazed. One cubic foot of 1/2 in. diameter spheres would accommodate, under the above-mentioned conditions, about one kilocurie of radioactive strontium. The use of an equivalent layer of 0.002 in. of strontium oxide is conservative, as thicker glazes may be produced and actually were obtained in the initial experiments.

In order to fix the radioactive strontium in a glaze, preliminary experiments were conducted on a method in which ordinary strontium chloride

reacts with a highly refractory unglazed type of porcelain. This procedure has been reported to be successful, but it requires very high burning temperatures and has been temporarily abandoned for this reason, although it would seem to give a very high concentration of strontium on the surface.

A second series of experiments has been tried following conventional procedures of making a glaze on less refractory porcelain. The glaze consists of the conventional materials, flint, feldspar, and clay, with strontium carbonate substituted for calcium carbonate. These compositions gave adherent glazes having a thickness of about 0.010 inch and containing up to 50 per cent of strontium oxide. Although these components show crazes, they seem to be mechanically strong.

The next step planned in this research is to compound a slightly radioactive glaze and investigate whether the strontium will be held securely by the glaze and thus not present any contamination hazards by being carried along with chemical reaction products.

C. Electron Accelerator

Supervisors: H. J. Gomberg, Assistant Director of Michigan Memorial-Phoenix Project and Assistant Professor of Electrical Engineering; W. Kerr, Instructor of Electrical Engineering.

A preliminary study is being made of the feasibility of constructing an electron accelerator to operate in the range of 100 to 200 kilovolts. The high-voltage power supply of a 200-kilovolt x-ray machine is available, and with some modification can be used as the source of high voltage for such an accelerator.

The planned accelerator would project a beam of electrons through a thin window of metal foil into a chamber or onto a target which could be at a pressure of the order of one atmosphere.

For electrons initially accelerated to 100 kilovolts, the energy loss in traversing a 0.05-mil window of nickel foil has been computed to be of the order of 7.7 kiloelectron-volts.

It is planned to construct an accelerator using a single-potential cathode of the type described by Pierce[1]. This cathode can be made to produce

[1] J. R. Pierce, "Rectilinear Electron Flow in Beams", Journal of Applied Physics, vol. II, pp. 548-554, August, 1940.

convergent beams of a desired size and intensity[1]. The resultant beam may then be accelerated so as to pass through a thin window as described above. Available x-ray-type tubes can probably be modified for use in such an accelerator. The chief problem would seem to be that of determining the effect upon the cathode focussing field of the potential distribution along the enclosing glass envelope. The proposed accelerator is shown in Fig. 50.

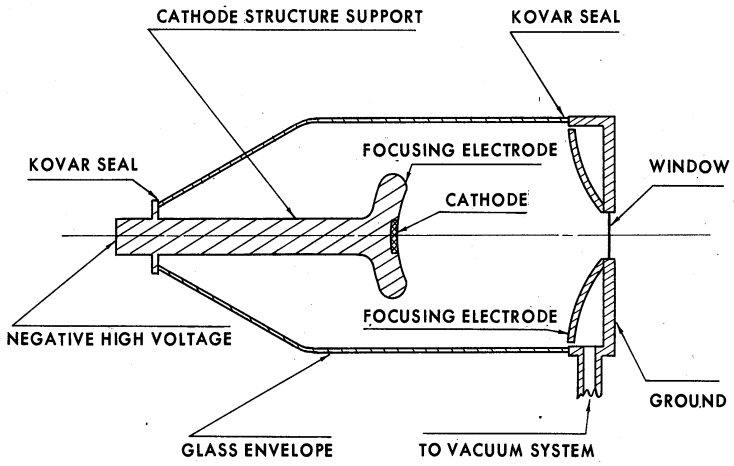
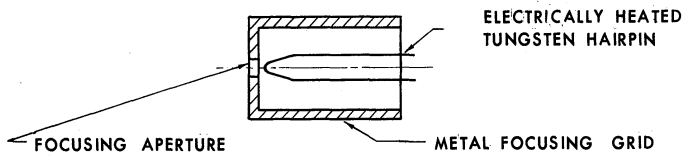


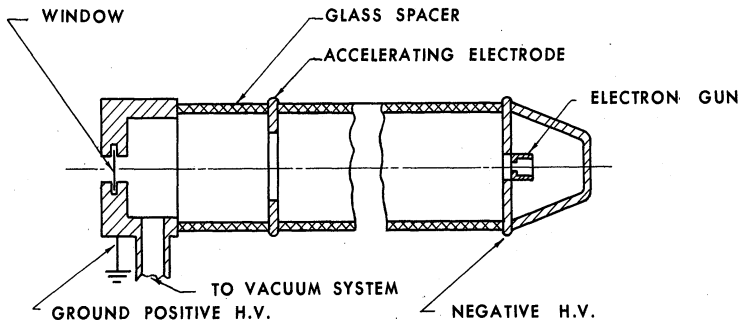
Fig. 50. Pierce-Type Electron Accelerator

If this design should not prove feasible, an alternate design would consist of an electron gun of the type which uses a heated tungsten ribbon or "hairpin" as a source of electrons. The electron beam is then determined by a small circular aperture in a cylindrical focusing "grid". The accelerator barrel would be made up of about five metal accelerating electrodes connected to a suitable voltage-dividing network. Cylindrical glass spacers would be used between the accelerating electrodes. In either design the window end of the accelerator would be at ground potential. Nickel foil of 0.05-mil thickness would be used as a window through which electrons would be ejected into the chamber containing the material to be bombarded. A vacuum system consisting of an oil diffusion pump and a mechanical fore-pump will be used to evacuate the accelerating space continuously. This design is illustrated in Fig. 51.

[1]R. Helm, K. Spangenberg, and L. Field, "Cathode-Design Procedure for Electron-Beam Tubes", Electrical Communication, vol. 24, n. 1, March 1947.



ELECTRON GUN



ELECTRON ACCELERATOR

Fig. 51. Electron Accelerator with Hairpin-Type Gun

PART V

OPERATION OF FISSION PRODUCTS LABORATORIES (SUBPROJECT M943-F)

Personnel

Advisors: L. E. Brownell, Associate Professor of Chemical and Metallurgical Engineering; H. J. Gomberg, Assistant Professor of Electrical Engineering; W. W. Meinke, Assistant Professor of Chemistry; L. Thomassen, Professor of Chemical and Metallurgical Engineering. Assisted by: J. V. Nehemias, Health Physicist; M. E. Gluckstein, Research Assistant.

A. Introduction

The high-level laboratory, as shown in Fig. 7 of Progress Report I has been in full operation since September. The low-level laboratory including the office and clothes-change area has just been completed. Operational Procedures which will provide maximum safety without appreciable restrictions to research have been established. Additional monitoring equipment has been installed in the laboratory. A safe with a combination lock has been installed to hold classified documents needed for reference in the course of research.

Mr. John V. Nehemias has been added to the laboratory staff as of November 1, 1951. He is a graduate physicist with more than two years experience as a Health Physicist at Brookhaven National Laboratory. His primary responsibility in the laboratory is to act as Health Physicist and assume the responsibility for the safe operating procedures in the research work.

Mr. Nehemias will keep the Radiation Policy Committee of the University of Michigan informed as to operational procedures and will check such procedures with either Dr. G. M. Ridenour, Radiological Safety Officer, or Mr. A. H. Emmons, Associate Radiological Safety Officer of the Radiological Safety Department. Mr. Nehemias will also act as Research Associate on the various research programs and may conduct research investigation when he is not occupied with problems relating to health physics.

B. Operational Procedures

The following operational procedures have been established for the 1000-curie cobalt-60 source which is used in the high-level laboratory:

1) Personnel Monitoring. Every person in the laboratory, with the exception of visitors with no occasion to approach the source, is required to wear a film badge at all times, both on the same floor as the source and on the floor above.

Persons dealing with the source are required to wear two pocket chambers and a finger chamber. A permanent record of chamber and film exposure is maintained.

2) Shielding. The source will remain at all times in its lead vault, which has sufficient shielding to reduce radiation at all points to tolerable levels when the closure plug is in place. The lead vault and auxiliary equipment are shown in Fig. 52.

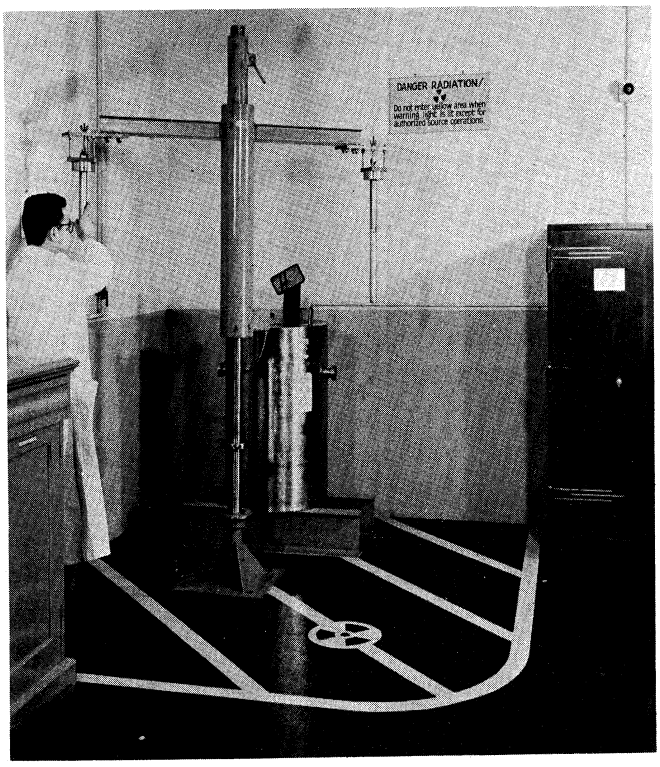


Fig. 52. Photograph of Cobalt-60 Vault (open) and Auxiliary Equipment

3) Operation Precautions.

(a) Flashing red warning lights visible from both floors are lit by means of a limit switch (at end of BX cable on vault, Fig. 52) whenever the plug is removed.

(b) An audible signal is actuated by the same limit switch upon removal of the plug.

(c) Those areas in which a tolerance dosage rate of 10 mr per hour is exceeded are indicated by yellow stripes on the floor (see Fig. 52).

(d) Warning signs stating

> DANGER RADIATION!
>
> Do not enter the yellow area when warning light is lit except for authorized source operations

are conspicuously posted on both floors (see Fig. 52)

(e) That area of the roof over which tolerance dosage rate is exceeded will be fenced off.

(f) Unauthorized persons will be kept safely distant from the source by laboratory personnel. When the laboratory is not in use, the door will be kept locked.

4) Routine Survey. Surveys to detect possible changes in the radiation field will be made at intervals of six months or less.

Dust samples will be collected to detect possible ruptures of the aluminum sheath around the cobalt at intervals of one week or less.

5) Waste Disposal. No waste-disposal problem is anticipated.

C. Palladium-109 Beta Source in Flame Burner

1) Personnel Monitoring. Each person involved in the experiment is required to wear a film badge throughout the test. Finger film rings are worn by those whose activities involve working with the source.

2) Shielding

(a) Shipping: The shipping container was designed and fabricated by Chalk River Laboratory.

(b) Unloading and Source Assembly: The manipulations involved in removing the foil from its shipping container and assembling the mountings were performed remotely, using a two-inch lucite shield.

(c) Operation: The experiment itself is performed in a half-inch steel pressure chamber faced with one-inch glass windows. The source is several inches from the chamber walls. This chamber provides sufficient shielding for operations involved in the experiment.

(d) Storage: Between phases of the experiment and while being held for decay, the source is stored in a "cave" of two-inch lucite, suitably labelled.

3) Operation Precautions. Opening and unloading the shipping container and assembling the source and mountings was performed using tongs, vises, clamps, wrenches, etc., at least two feet long. When possible, the operator remained behind the lucite shield.

4) Routine Survey. Constant radiation monitoring is maintained with a Geiger-tube survey meter during the operation. Ionization chamber surveys are made when indicated. Samples of the water used in the vacuum-pump system and of dust in the ejected air are taken and subjected to radioanalysis.

When the operation is complete, a smear survey is made of the inner walls of the pressure chamber.

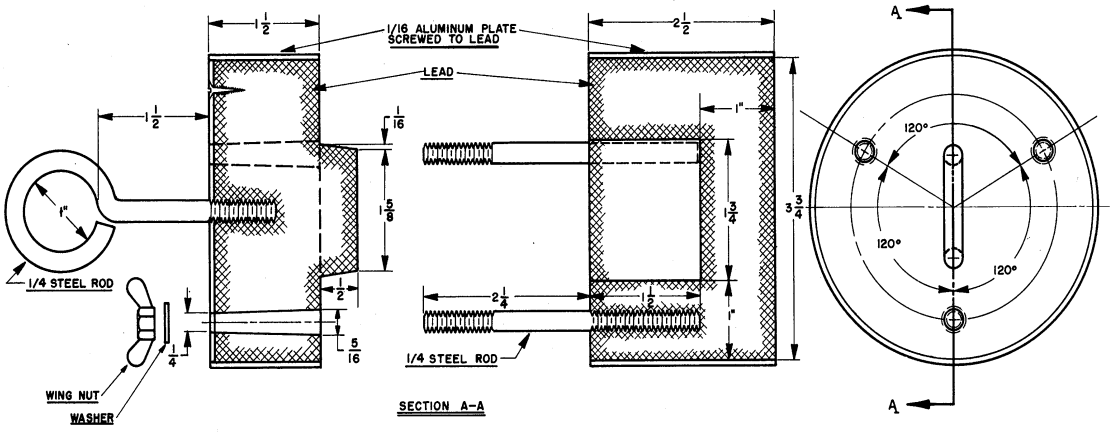
5) Waste Disposal. The exhaust from the vacuum system is released through a fiber-glass filter into the atmosphere, distant from any inhabited area.

D. Palladium-109 Beta Source in Diesel Engine

1) Personnel Monitoring. Each person involved in the experiment will be required to wear a film badge throughout the test. Finger film rings will be worn by those whose activities involve working with the source.

2) Shielding.

(a) Shipping: The source will arrive in a one-inch-thick lead shipping container, designed and fabricated by the Fission Products Laboratory. Fig. 53 is a drawing of this container.



DRAWING OF CONTAINER FOR PALLADIUM-109

Fig. 53

(b) Unloading and source assembly: The manipulations involved in removing the source from its shipping container and assembling the mounting will be performed remotely, using a two-inch lucite shield.

(c) Operation: During the experiment, the source will be housed in the combustion chamber of a diesel engine, in close proximity to the quarter-inch iron walls. A lead shield of one-brick thickness (one and one-half inches) will be available if needed for Bremsstrahlung. Otherwise, the operator will be shielded by two-inch lucite.

3) Operation Precautions. Proposed precautions have been discussed under Shielding and Routine Survey.

4) Routine Survey. Constant radiation monitoring will be maintained with a Geiger-tube survey meter during the operation. Ionization-chamber surveys will also be made as indicated.

Samples of dust in the exhaust gases will be taken as indicated for radioanalysis.

5) Waste Disposal. The exhaust from the engine will be released to the atmosphere, after passage through a fiber-glass filter, well above the peak of the laboratory roof.

A radioanalysis of the filter will be made.

D. Dust Monitoring Equipment

The continuous dust monitoring equipment referred to in the procedures is shown in Fig. 54. Air is drawn through the filter paper at end of hose by a commercial vacuum cleaner (Filter Queen). The rate of flow of air through the filter is regulated by a rheostat control on the motor (7-1/2 amp powerstat). A flow rate of about eight cubic feet per minute is used in this laboratory. This flow rate enables quantities of the order of 10^{-12} microcuries per cubic centimeter of air to be detected without difficulty from weekly samples.

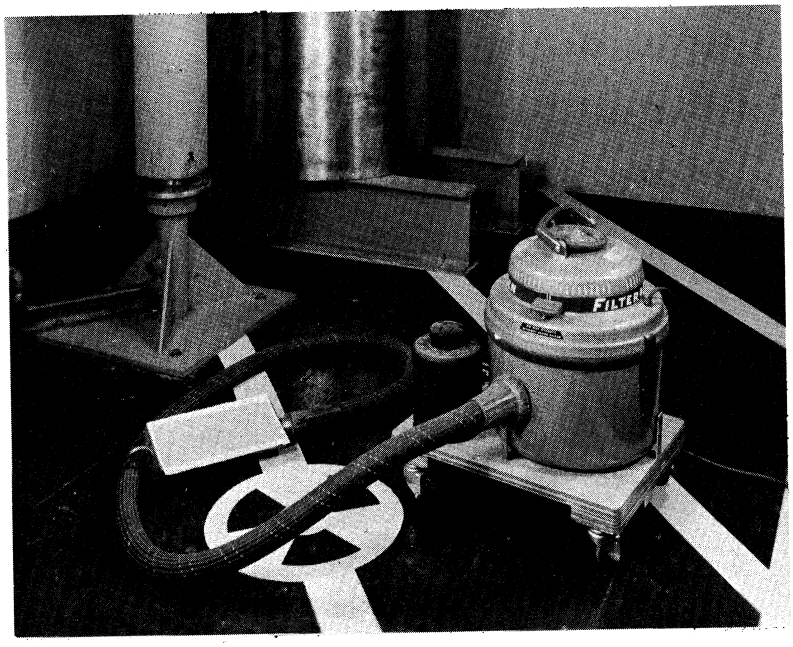


Fig. 54. Photograph of Dust Monitoring Equipment

The tolerance concentration for cobalt-60 agreed upon at the International Conference at Chalk River in 1949 is 2×10^{-9} microcuries per cubic centimeters. Therefore, with this equipment it is possible to measure five ten-thousandths of tolerance. If shorter collection intervals are indicated by the usage of the laboratory, this sensitivity can be maintained by increasing the flow rate.

E. Health Physics Report of Procedures Used in Handling Palladium-109 in Flame Burner Experiments

The flame burner experiment using pile-irradiated palladium foil around a flame was the first operation requiring the manipulation of major quantities of radioactive materials by project personnel. It is of interest to report the progress of the operation from the radiation-safety point of view.

The shipment was transferred from a commercial plane at Windsor (Canada) airport to a University vehicle and rushed through customs to Willow Run. A survey at the point of transfer (Fig. 55) indicated a safe handling time of over an hour. With the container in place at the rear of the station wagon, no radiation was dectectable at the front seat. Poor flying weather had delayed the shipment so that it had decayed from 32 curies at the time of removal from the pile to 8 curies at the time of arrival at Windsor airport.

Fig. 55. Monitoring Shipment of Palladium-109 at Transfer from Plane to Station Wagon.

Immediately upon arrival at Willow Run the shipping container was placed upon a sturdy, paper-covered table, behind half-inch lucite shields. However, when the container was opened and surveyed (Fig. 56), the presence of an appreciable amount of penetrating radiation was detected. The shielding was then increased to two inches of lucite on one side and a one-and-one-half inch lead-brick wall on the second side.

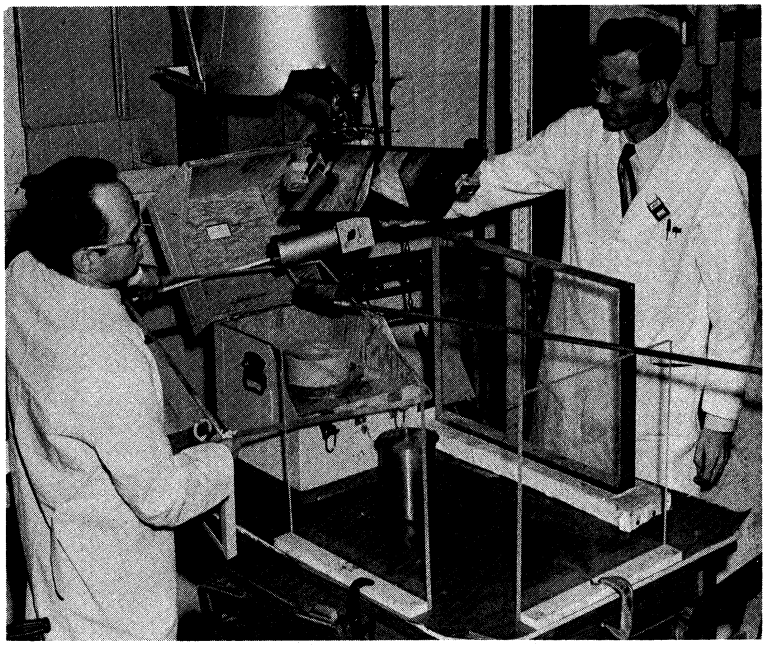


Fig. 56. Determination of Presense of Penetrating Radiation with Lucite Shield

From behind this increased shielding, the foil was placed around a brass ring against a piece of brass shim stock (Fig. 57) and locked into position by closing the outer ring and screwing it tight (Fig. 58). All these operations were performed remotely, using 3 ft tongs and screwdrivers. In this manner it was possible to avoid serious exposures during the process. This assembly, when completed (Fig. 59) was set in place with tongs around the flame burner in the furnace (Fig. 60). The walls of the furnace provided sufficient shielding to reduce radiation levels at all operating positions to less than 20 milliroentgens per hour.

When this first phase of the experiment was completed, the assembly was removed to the shielded area. Then the source was transferred to the smaller assembly designed for the second half of the experiment and reinstalled in the furnace without incident. When the experiment was completed, the source was removed from the assembly and carried in its shipping container to the Fission Products Laboratory for use in a chemical reaction experiment.

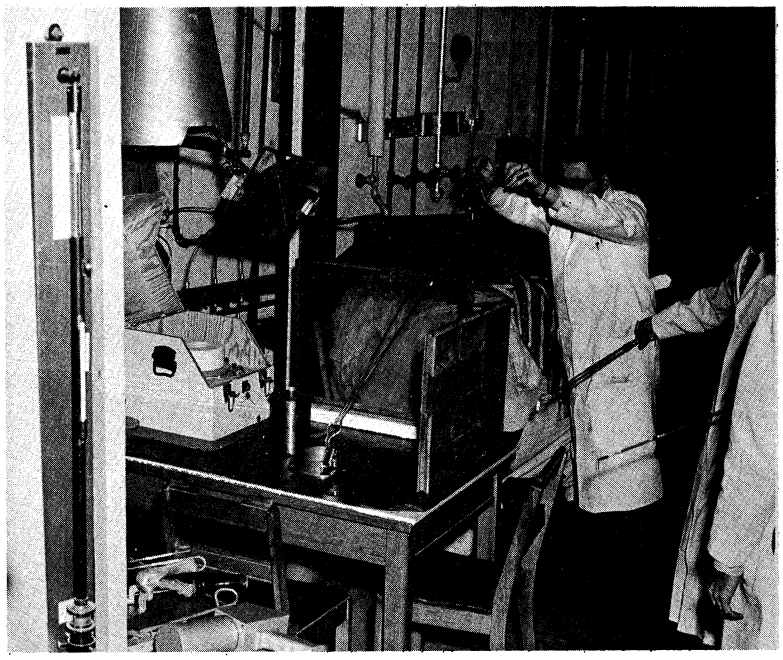


Fig. 57. Insertion of Foil between Ring and Brass Shim Stock

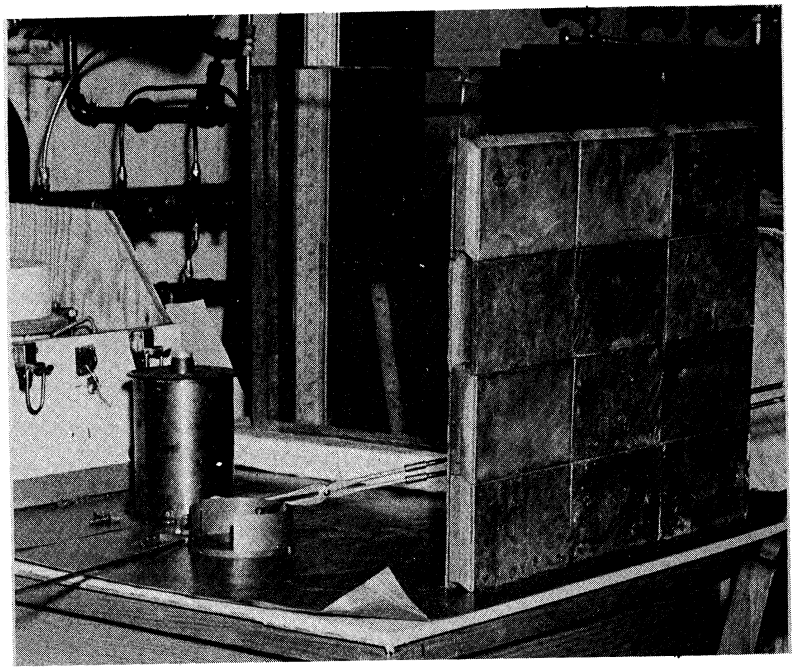


Fig. 58. Tightening of Outer Brass Ring

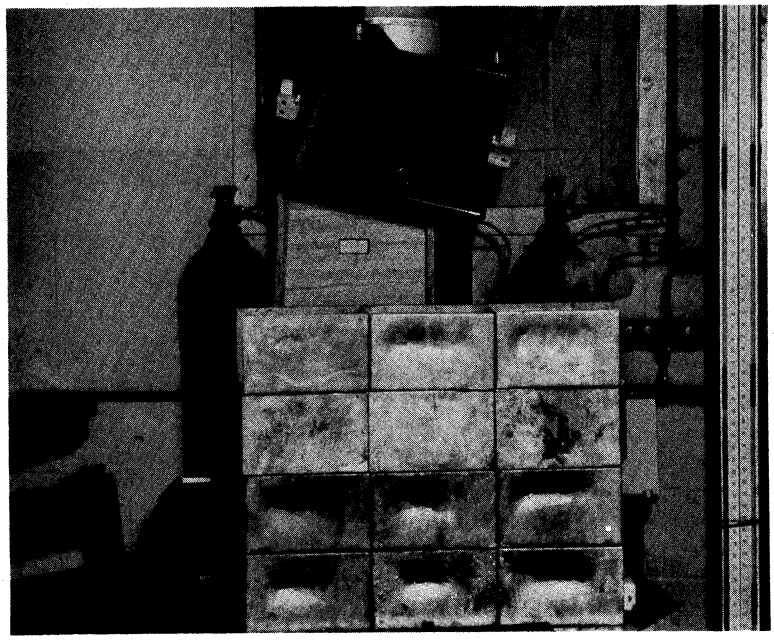


Fig. 59. Mirror-View of Completed Assembly Ready for Insertion into Flame Burner

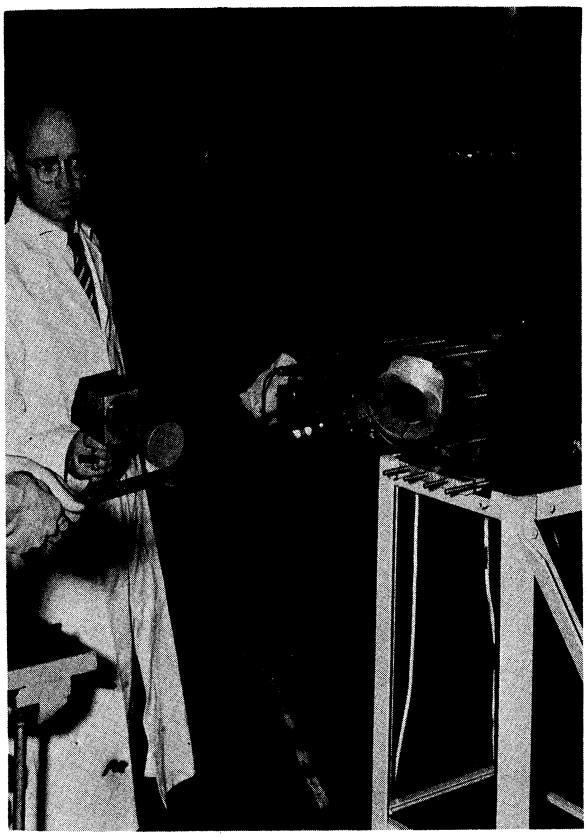


Fig. 60. Installation of Completed Assembly into the Flame Burner

The entire operation, including the setting-up of the chemical experiment, was performed by five persons. An average whole-body exposure of 80 milliroentgens and an average hand exposure of 95 milliroentgens was received by the group.

Fig. 61 is a graphical presentation of dosage measurements as obtained at the time of the experiment when the foil was rated at about 7 curies. Application of these data to other planned experiments would, of course, require scaling these values up or down in proportion to the number of curies present.

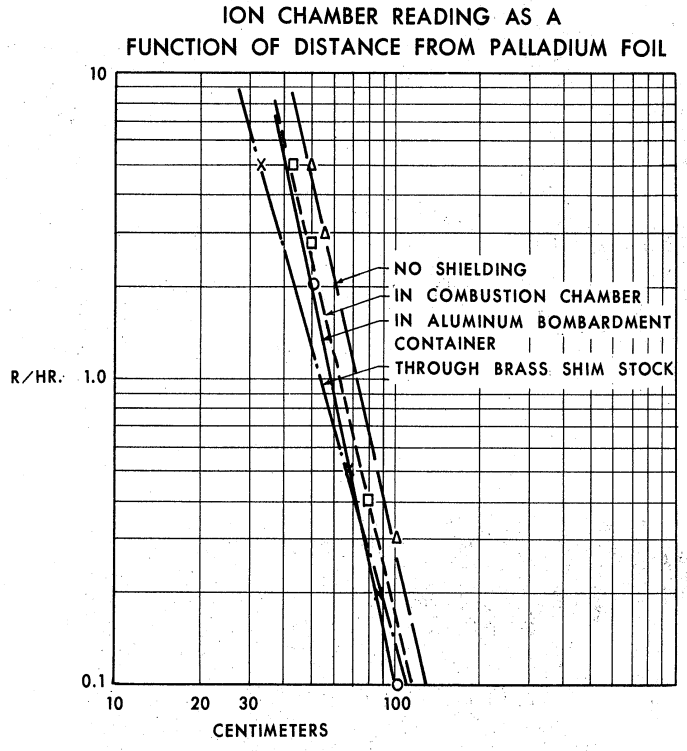


Fig. 61

A serious quantity of loose contamination was detected on everything that came in contact with the palladium foil. Several such spots read 20 milliroentgens per hour. This contaminant was found to be intimately associated with the foil itself, as the radiation characteristics of the contaminant were identical with those of the foil after the palladium had undergone several half-lives of decay. A study of these characteristics and a chemical analysis should provide an indication as to the nature of the contaminant. This information will be made available when the study is finished.

F. Modified Pneumatic Lift for Cobalt-60 Source

Since the completion of Progress Report I, the installation of the auxiliary equipment for the cobalt-60 vault has been completed. Fig. 52 shows

the completed installation. The lift shown is a modified form of the design presented in the previous report. Fig. 62 shows a sectional drawing of the modified lift. The essential differences from the original projected design are the conversion to a pneumatic lift and the use of a T-shaped arm to balance the load. Raising and lowering of the lift is accomplished by means of two variable-flow, quick-opening valves. A single pipe line serves as the inlet and outlet to the lift cylinder. This line has a pipe tee at the top. The valves are connected to each of the arms of the tee. One of the valves is connected to the air line, the other is vented to the room. The lift may be raised, lowered, or held at any position on its shaft by manipulation of these valves. When the plug is raised clear of the vault, the entire arm and cylinder assembly may be rotated through 360°. In this way, the operator is always in a safe position and the open time of the vault is reduced to a minimum.

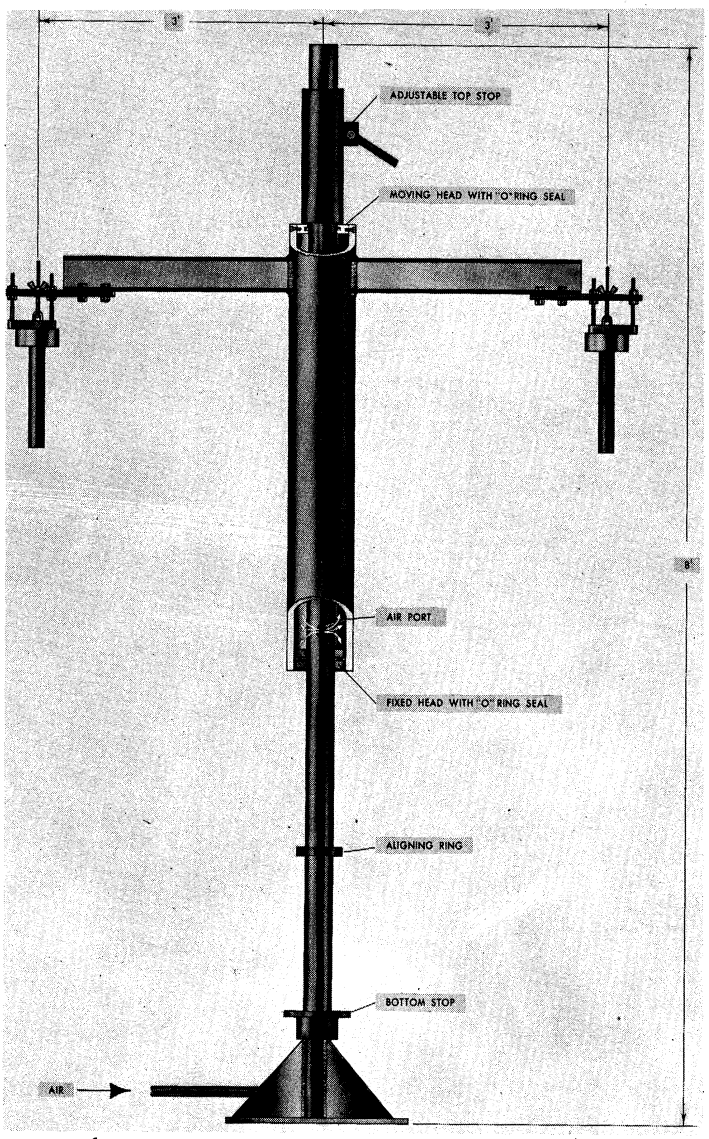


Fig. 62. Sectional Drawing of the Modified Pneumatic Lift

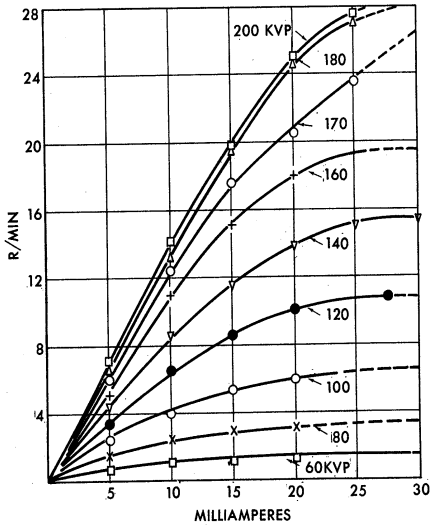
An air compressor has been installed to supply air for the lift and for general laboratory purposes.

G. Calibration of 210 KVP X-Ray Machine

A calibration of the 210 KVP Westinghouse X-Ray Machine has been completed. The results are shown in Figs. 63, 64, and 65. The data are in agreement with the inverse square law in that the radiation rate in roentgens per minute is proportional to the reciprocal of the square of the distance from the source.

The procedure followed in the calibration was essentially the same as that used in the calibration of the 150 KVP x-ray machine as reported in Progress Report I. An instantaneous roentgen ratemeter (Victoreen Model 510) was used.

PLOT OF ROENTGENS PER MINUTE (R/MIN.) VS CURRENT IN MILLIAMPERES AT CONSTANT KILOVOLTAGE(KVP) FOR 210 KVP X-RAY MACHINE. DISTANCE=39" FROM SOURCE



PLOT OF DOSAGE RATE IN ROENTGENS PER MINUTE(R/MIN) VS KILOVOLTAGE(KVP) AT CONSTANT CURRENT(MILLIAMPERES) FOR 210 KVP-X-RAY MACHINE. DISTANCE TO SOURCE=39".

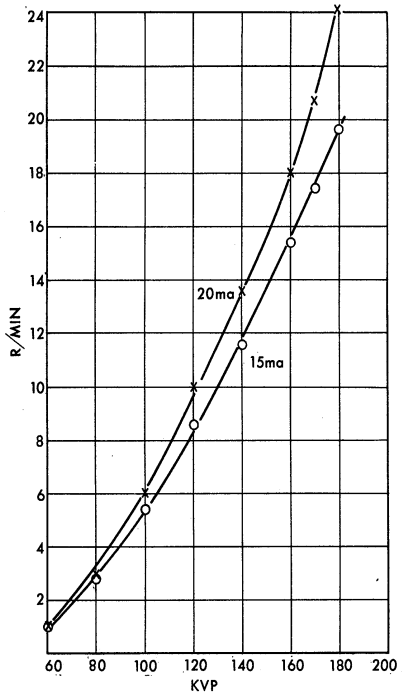


Fig. 64

DOSE IN ROENTGENS PER MINUTE (R/MIN.) VS DISTANCE TO SOURCE ℓ, IN INCHES FOR 210 KVP X-RAY AT INDICATED OPERATING CONDITIONS

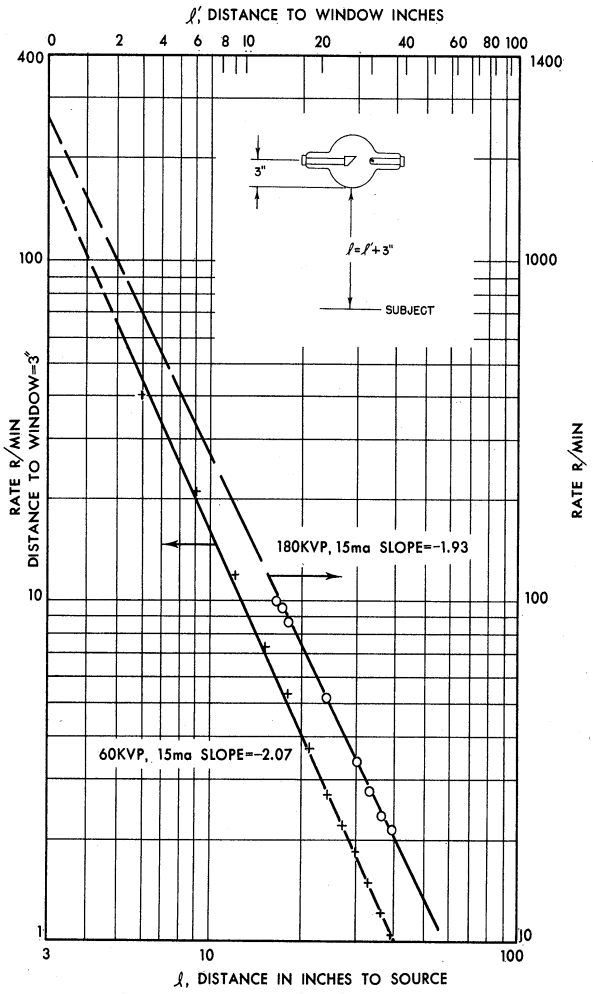


Fig. 65

The instrument was equipped with a probe capable of measuring up to a maximum of 100 roentgens per minute.

The experimental apparatus used is shown in Fig. 66. The lead plate which formed the lower absorber was ruled with a rectangular coordinate system of lines 1 inch apart. The approximate center of the plastic window of the x-ray machine head was located, and the head of the machine was adjusted so that the window was horizontal. A plumb line was dropped from the center of the window to the ruled lead plate to locate the origin on the coordinate system. The probe was clamped on a ring support so that the tip was centered over the origin. An arbitrary distance of 36 inches from the window was selected and the machine set in operation at 60 KVP. The current in the x-ray tube was then adjusted in increments of 5 ma, between 5 and 30 ma and the corresponding readings

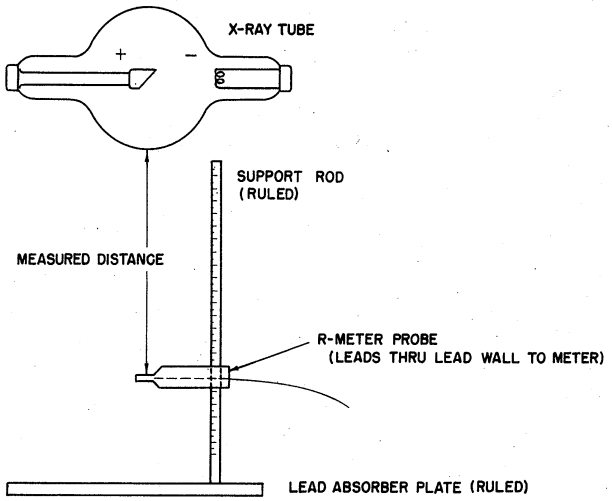


Fig. 66. Drawing of X-Ray Calibration Set-Up

on the Roentgen ratemeter recorded. After the runs for a particular distance and voltage were completed, the probe was raised a few inches and the sequence repeated. After the full range of the probe was reached, the probe was returned to the 36-inch position and the run repeated for a new KV setting. In this way, the entire range from 60 to 200 KVP was investigated. Some of the calibration data are given in Table XXVIII.

TABLE XXVIII

CALIBRATION DATA FOR 210 KVP X-RAY MACHINE

KVP Setting	Milli-amperes	Distance to Window	x*	y*	Rate Roentgens/min.
140	5	36"	0	0	4.3
140	10	36"	0	0	8.6
140	15	36"	0	0	11.5
140	20	36"	0	0	13.8
140	25	36"	0	0	15.0
140	30	36"	0	0	15.2

*Measured from origin on lead sheet; ruled with x-y coordinates.

H. Surface Dosage Consideration of Beta-Emitting Foils

As explained in Progress Report I, the personnel operating the Fission Products Laboratory under subproject M943-F acts as a service group to the other

research groups in that it obtains and handles the radioactive materials, checks problems of health physics, performs necessary calculations, and other services. For example, it supplied information on surface dosage of beta-emitting foils to the research group under M943-B. The following section of the report was prepared to supply this information and is included here, as it may be of interest to other research groups.

1. Variation of Surface Dosage with Foil Thickness. The experiments for subproject M943-B conducted at the Willow Run Research Center to study the effects of ionizing radiation upon flame stability and propagation rate required an intense source of beta radiation. These experiments, as planned, could be completed in from ten to twelve hours. To satisfy these requirements and still minimize health hazards, it was decided to use pile-irradiated palladium foil as the beta source. The only isotope detectable a few hours after removal from the pile, assuming the presence of no major contaminents is,

$$Pd^{108}\ (n,\gamma)\ Pd^{109}.$$ [4]

Palladium-109 emits a pure beta of 0.95 mev maximum energy, with a half-life of about thirteen hours. Palladium is one of the noble metals, and has no known effect on human metabolic function.

As the radioactive material is, in this case, a heavy metal, it is apparent that self-absorption will be an important factor in computing surface dosage and in estimating foil thickness. It is of interest, therefore, to compute the fraction of total beta energy reaching the surface as a function of foil thickness.

Using the Fermi energy distribution theory and equation,

$$W = A\ E^{1/2}\ (1 + 2E)\ (1 + E)^{1/2}\ (E_m - E)^2\ dE,$$ [5]

where W is the probability for the emission of beta particles in the energy range E to (E + dE), E the energy, E_m the maximum energy and A a normalization constant dependent upon activity, one can compute an energy distribution for a 0.95 mev beta emitter[6] such as palladium-109. Fig. 66 is, then, the energy distribution of beta particles emitted by such an isotope. For instance, from the curve one would expect more particles of about 0.35 mev than any other

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[4]National Bureau of Standards. Nuclear Data, U. S. Dept. of Commerce, NBS Circular 499.

[5]Novey, T. B., et al. "Theoretical Study of Beta Absorption Curves and Correlation with Feather Method of Beta Energy Determination", Formerly Paper 2.6; based on Report CC-579, 1943.

[6]Pollard and Davidson, Applied Nuclear Physics, New York, Wiley, 1942.

energy about twice as many as one would expect to find at energies of 0.10 or 0.65 mev. The vertical scale represents numbers that are proportional to frequency of occurrence for each energy chosen, such that

$$\int_0^{E_m} W dE = 1. \quad [5]$$

Considering a foil one-half mil thick and proceeding to a foil of one-mil thickness, it is of interest to compute what fraction of the additional activity (i.e., in the second half mil) will penetrate the first half mil and reach the surface. The first half mil acts simply as an absorber in this computation.

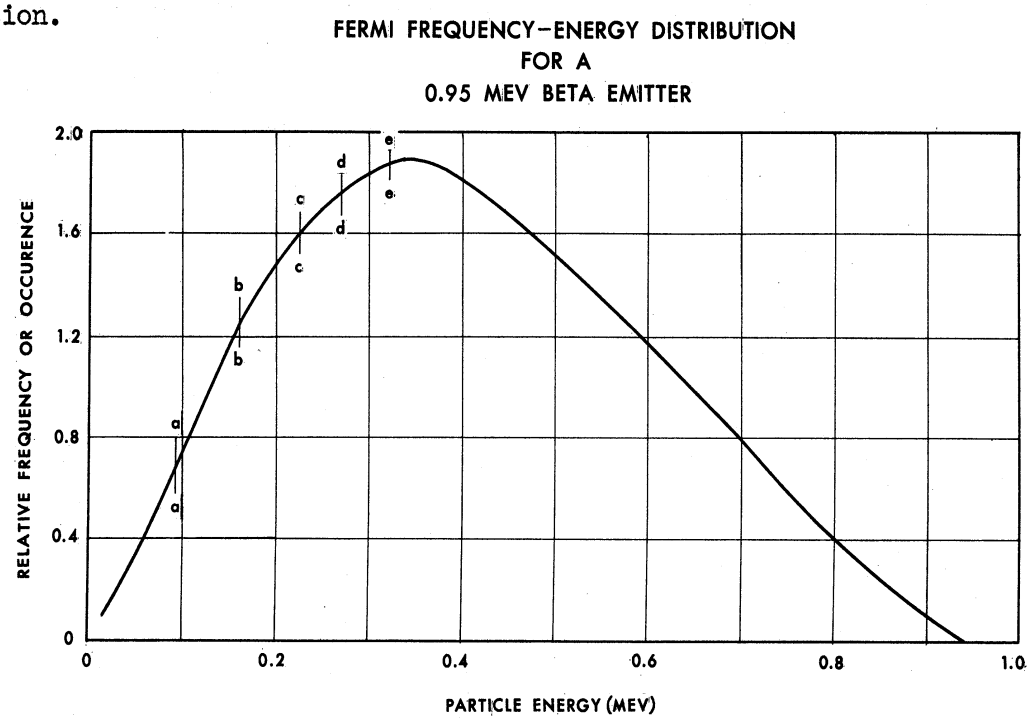

Fig. 67

From known range-energy relationships,[2] it can be established that no particles leaving the second half mil with energies less than that indicated by a-a in Fig. 67 will penetrate the first half mil and reach the surface. A third half mil would be subjected to a total of one mil of palladium as an

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[5]Novey, T. B., et al. "Theoretical Study of Beta Absorption Curves and Correlation with Feather Method of Beta Energy Determination", Formerly Paper 2.6; based on Report CC-579, 1943.

[2]Lawrence, J. H. and Hamilton, J. G., Advances in Biological and Medical Physics, New York, Academic Press, 1948.

absorber. In this case particles of energy less than that indicated by b-b would be stopped completely. This process continues with the addition of further thicknesses until the maximum range, about 13 mils,[1] is reached. After that none of the particles emitted by any added palladium would reach the surface. In this manner the spectrum may be divided into energy intervals "oa", "ab", "bc", etc.

Considering again the case of a one-mil foil, it is of interest to compute the contribution to surface dosage from the second half mil. As has been stated, those particles in the energy interval "oa" will be absorbed completely. Particles of higher energy than a-a will be subjected to an average of 1-1/2 half mils of absorber and will be partially absorbed. As absorption is known to be linear with distance for small energy intervals,[7] relative absorption curves can be plotted for each interval. Fig. 68 is such an absorption curve for energy interval "de". It is readily seen that, with 1-1/2

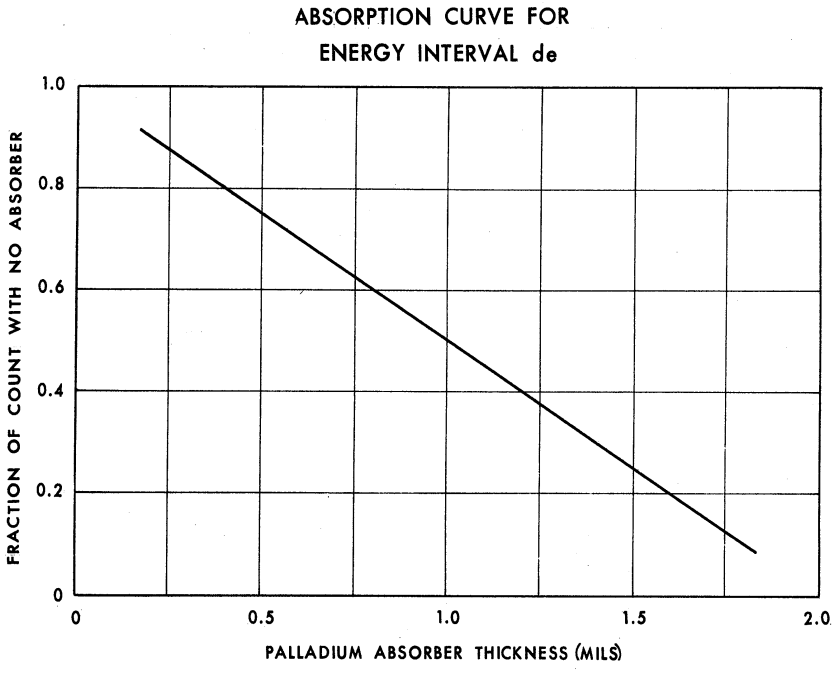

Fig. 68

half mils of absorber, only 63 per cent of the particles leaving the second half mil in the energy interval "de" will reach the surface. Finding this fraction for each energy interval, weighting the fractions according to the area of the interval, and summing the fractions yields a number which is the fraction of a total radiation originating in the second half mil reaching the surface. Fig. 69 is a plot of these values as a function of thickness.

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[1]Feather, N., An Introduction to Nuclear Physics, New York, Cambridge University Press, 1936.

[7]Rutherford, E., et al. Radiations from Radioactive Substances, New York, Cambridge University Press, 1930.

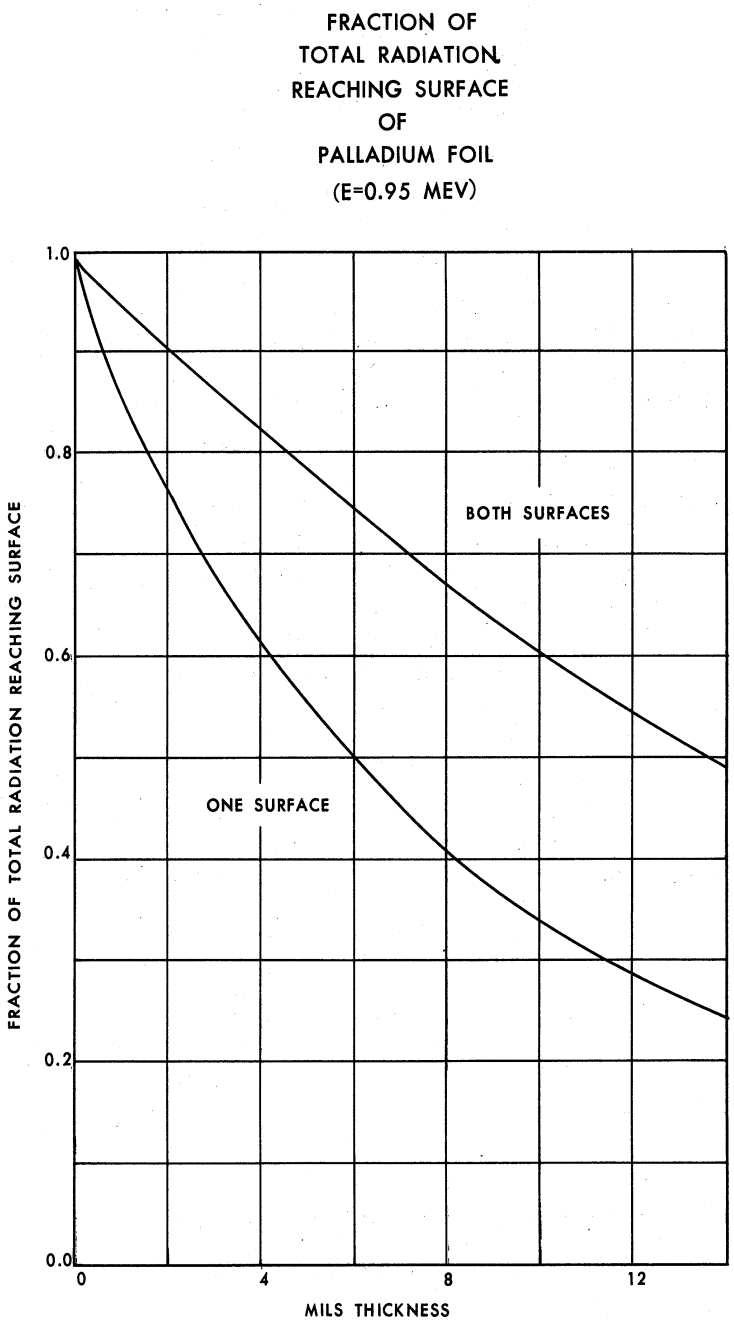

Fig. 69

If the radiation from both surfaces is useful, as in the case of a foil immersed in solution, the same computation can be repeated with the thickness of absorber reduced in each case by a factor of two. A plot of these results is also shown in Fig. 69.

By simply summing the fractions in Fig. 69 as a function of thickness, one can compute the fraction of total radiation originating within the foil reaching the surface. This information is plotted in Fig. 70.

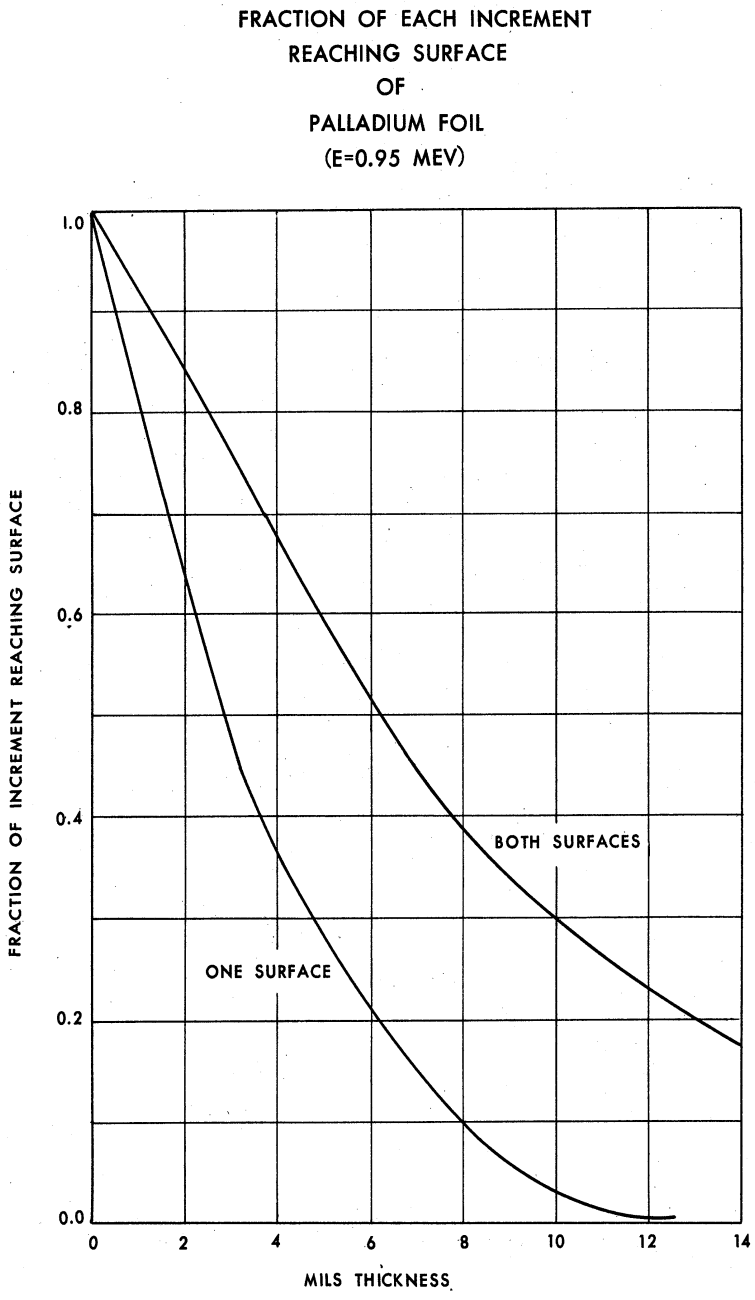

Fig. 70

2. Computation of Absolute Dose Rate. To compute absolute surface dose rate, one may use the relationship

$$S = 8.7 \times 10^{-7} \, nRk,^{2}$$

[2]Lawrence, J. H. and Hamilton, J. G., Advances in Biological and Medical Physics, New York, Academic Press, 1948.

where S is the surface dose rate in roentgens equivalent physical (rep) per hour, n the number of particles per square centimeter of surface per second, R the average range in centimeters of material, and k the specific ionization (ion pairs per centimeter of path) in air.

Applying this relationship to a specific problem to a piece of palladium foil, 6 in. x 1-3/4 in. x 1 mil thickness, containing twenty-five curies, the following calculations may be made:

To find n:

$$n = \frac{\text{disintegrations per second}}{\text{area of foil in centimeters}}$$

$$= \frac{25 \text{ curies } (3.7 \times 10^{10}) \text{ disintegrations per second per curies}}{(6 \times 1.75) \text{ sq in. } (2.54)^2 \text{ sq cm per sq in.}}$$

$$= 1.36 \times 10^{10} \text{ particles per sq cm per second}$$

To find R: Range is linearly related to energy in this region,[1] and average energy has been found to be 0.42 of maximum energy.[5] Thus:

$$\text{Rave} = 12.6 \text{ mils } \left(\frac{2.54}{1000}\right) \text{ (centimeters per mil) } (0.42)$$

$$= 0.0134 \text{ cm}$$

To find k: Consult tables.[6]

$$k = 57 .$$

Substituting these values,

$$S = (8.7 \times 10^{-7})(nRk)$$

$$= (8.7 \times 10^{-7}) (1.36 \times 10^{10}) (13.4 \times 10^{-3}) (57)$$

$$= 9050 \text{ Rep per hour at surface due to } Pd^{109}$$

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[1]Feather, N., An Introduction to Nuclear Physics, New York, Cambridge University Press, 1936.

[5]Novey, T. B., et al. "Theoretical Study of Beta Absorption Curves and Correlation with Feather Method of Beta Energy Determination", Formerly Paper 2.6; based on Report CC-579, 1943.

[6]Pollard and Davidson, Applied Nuclear Physics, New York, Wiley, 1942.

3. Attenuation in Air. For health and safety reasons, one is also interested in knowing the dose rate as a function of distance from the foil. The dotted curve in Fig. 71, shows the attenuation one would expect from air absorption alone. One would expect a further reduction of intensity with distance, approximating an inverse square reduction at distances greater than about 40 cm. The solid curve of Fig. 71 represents the predicted variation of dose rate with distance.

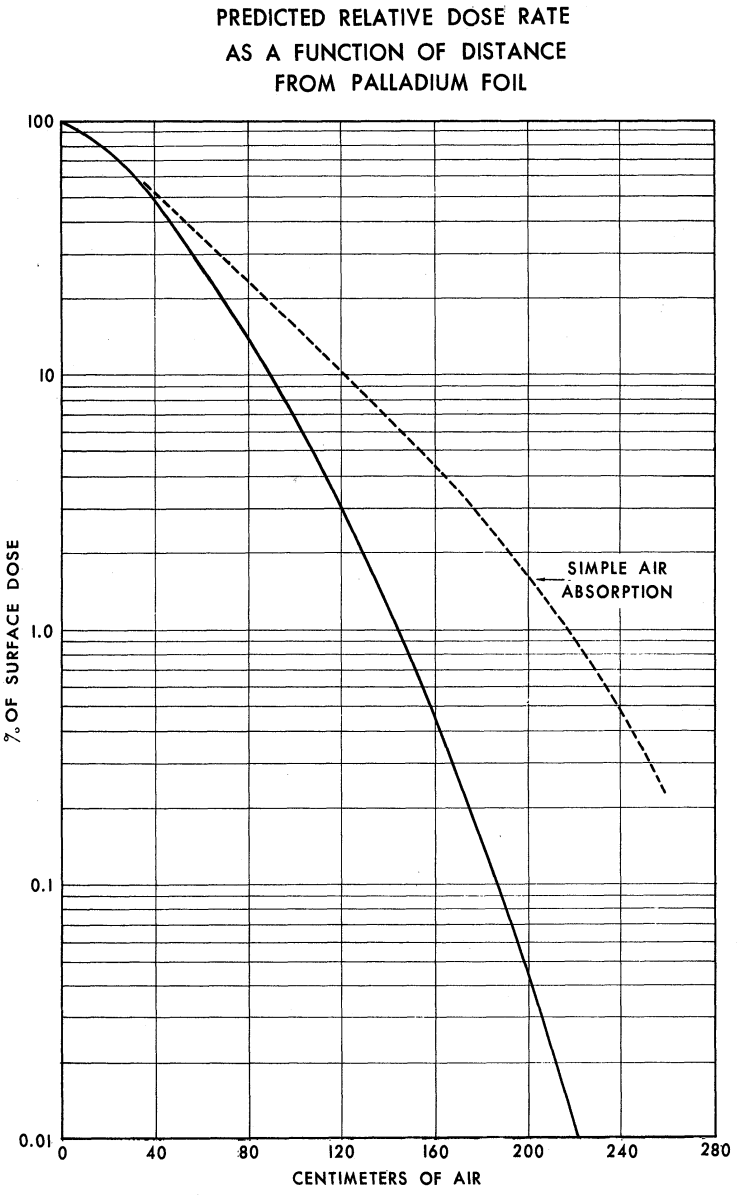

Fig. 71