ENGINEERING RESEARCH INSTITUTE
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COO-91

PROGRESS REPORT 3

UTILIZATION OF THE GROSS FISSION PRODUCTS
(Unclassified)

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Project M943

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ABSTRACT

This report describes the work accomplished since Progress Report 2 (COO-90), dated January 31, 1952. Additional experimental results have been obtained on all subprojects. These results are described in this report and are abstracted as follows:

On subproject M943-A (Fundamental Study of Effect of Radiation on Combustion Engine Performance), tests have been made with beta-particle radiation from a phosphorus-32 source and from palladium-109 sources. Data indicate that beta-particle radiation has little effect on combustion conducted at less than atmospheric pressure. The next experiments on this subproject will involve reactions at high pressures.

On subproject M943-B (Performance of Combustion Engines under the Influence of Radiation), positive results have been obtained with the experimental diesel engine. In a test using about 41 curies of palladium, a slight reduction in the ignition delay period, a slight reduction in the compression ratio required for ignition, a slight reduction in the specific fuel consumption were obtained and are attributed to the effect of beta-particle radiation. All these effects contribute to the improved performance of the engine. Additional experiments are planned using larger amounts of radiation. Experiments on this subproject with an experimental burner confirm the results of subproject M943-A that ionizing beta-particle radiation has little effect on combustion conducted at atmospheric pressure.

On subproject M943-C (Effects of Radiation on Chemical Reactions), experiments have been conducted on the polymerization of styrene and natural oils and additional information has been obtained on the ammonia synthesis reaction. Additional experiments will be conducted at high temperatures and pressures.

On subproject M943-D (Radiation of Biological Materials), the flavor of irradiated foods has been studied. Green vegetables may be irradiated without undesirable flavor change, while products containing animal proteins generally undergo undesirable changes in flavor as a result of gamma radiation. Small amounts of chemical additives can be used with fresh meats to prevent these undesirable flavor changes.

On subproject M943-E (Exploratory Research), ceramic glazes containing strontium-90 have been tested at room temperature and at high temperature. At low temperature the ceramic glazes retain the radioactive strontium-90, but additional experiments will be required to develop satisfactory glazes for high-temperature service. Glazes with larger amounts of activity will be tested.

On subproject M943-F (Operation of Fission Products Laboratories), several calibrations of the 1000-curie cobalt-60 gamma-ray source were made. The best average value was taken as 79,000 rep in air as of June 30, 1952. Preliminary designs for a 10-kilocurie source were developed. An order was placed by project M943 with Eldorado Mining and Refining Ltd. of Ottawa, Canada, for 5 kilocuries of cobalt-60 in the form of cobalt rods. An additional

5 kilocuries was purchased by Michigan Memorial-Phoenix Project No. 41, and the two sources will be pooled to give a maximum gamma-ray flux.

In cooperative research Michigan Memorial-Phoenix Project No. 41 has further investigated the effects of gamma radiation on enzymes, bacteria, and viruses. With Parke, Davis and Company, a glass was tested which does not discolor and which meets the normal requirements for ampules and containers of pharmaceuticals. In other cooperative tests, blood fractions have been irradiated for the Division of Laboratories of the Michigan Department of Health, with the goal of sterilizing blood fractions so as to destroy the virus of serum hepatitis, which has recently appeared as a problem in the field of medicine.

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PROGRESS REPORT 3

UTILIZATION OF THE GROSS FISSION PRODUCTS

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

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.

This project is concerned with the effects which may be produced on combustion processes by radiation from radioactive sources. The general approach is to study the basic phenomena of combustion and the characteristic changes which may be produced by radiation. Conditions are simplified wherever possible in order to reduce to a minimum the number of variables in effect during a given experiment.

1. Experiments at Low Pressure

The first specific approach was an investigation of the effects of radiation on mixtures of hydrogen-oxygen and methane-oxygen at pressures too low for explosion to occur. The gases were admitted to a previously evacuated pyrex bulb of 15-cm inside diameter. Pressures of from about 0.5 cm Hg to about 3 cm Hg were used. Changes in pressure resulting from reaction were measured with a McLeod gauge. For a more complete description of apparatus, see Progress Report 2.[1]

a. Reactions in the Glow Discharge. A direct-current glow discharge was passed through the gas, and the resulting change in pressure was measured at two-minute intervals with a McLeod gauge. This set of experiments were improved over those described in Progress Report 2 in the following respects:

1). It was found that the introduction of 100 grams of anhydrous magnesium perchlorate (dehydrite) into the reaction bulb would give a much more reproducible pressure-time curve. The dehydrite removes the water vapor as fast as it is formed so that the actual rate of reaction is measured. Formerly the pressure drop resulting from reaction was partially masked by the formation of the water vapor. Thus the long "tails" on the curves shown in Progress Report 2, pages 6-13, were the result of a slow diffusion of water vapor into the cold trap through a long connecting tube. The "humps" in certain of the curves relating to methane-oxygen mixtures were also caused by this phenomenon.

2). In these experiments the current in the glow discharge was held to a fixed value by means of a Powerstat. Little difficulty was experienced in keeping the current constant to within $\pm$ 0.2 milliamp. During a few runs with hydrogen-oxygen mixtures the current became very erratic near the end of the run because of the reduced pressures in the reaction bulb. Fig. 1 shows a typical set of pressure-time curves for different discharge currents in hydrogen-oxygen.

The first radioactive material used was phosphorus-32, which has a half-life of 14.3 days and emits beta particles of 1.70-Mev maximum energy. The phosphorus-32 was obtained from Oak Ridge and was contained in a bakelite disc, 1 inch in diameter and 0.19 inch thick. Reported dose rate at time of removal from the pile was 3188 rep per hour and the calculated source strength was 83 millicuries.

The phosphorus source was mounted in the reaction chamber and the runs were repeated with the glow discharge at 6 milliamps. In Fig. 2 curves A refer to typical runs without a radioactive source present, curves B refer to runs with the phosphorus-32 present, and curve C refers to a run made with a palladium-109 source consisting of a palladium foil which had been irradiated in the Chalk River pile. The calculated strength of the source was 20 curies. Palladium-109 has a half-life of 14 hours and a maximum beta particle energy of 1 Mev.

It was decided that the effects of the two sources above on the hydrogen-oxygen mixtures were either nonexistent or so small as to lie within the experimental error under the conditions of the experiment. The experiment was then conducted with a mixture of methane and oxygen. Fig. 3 shows a

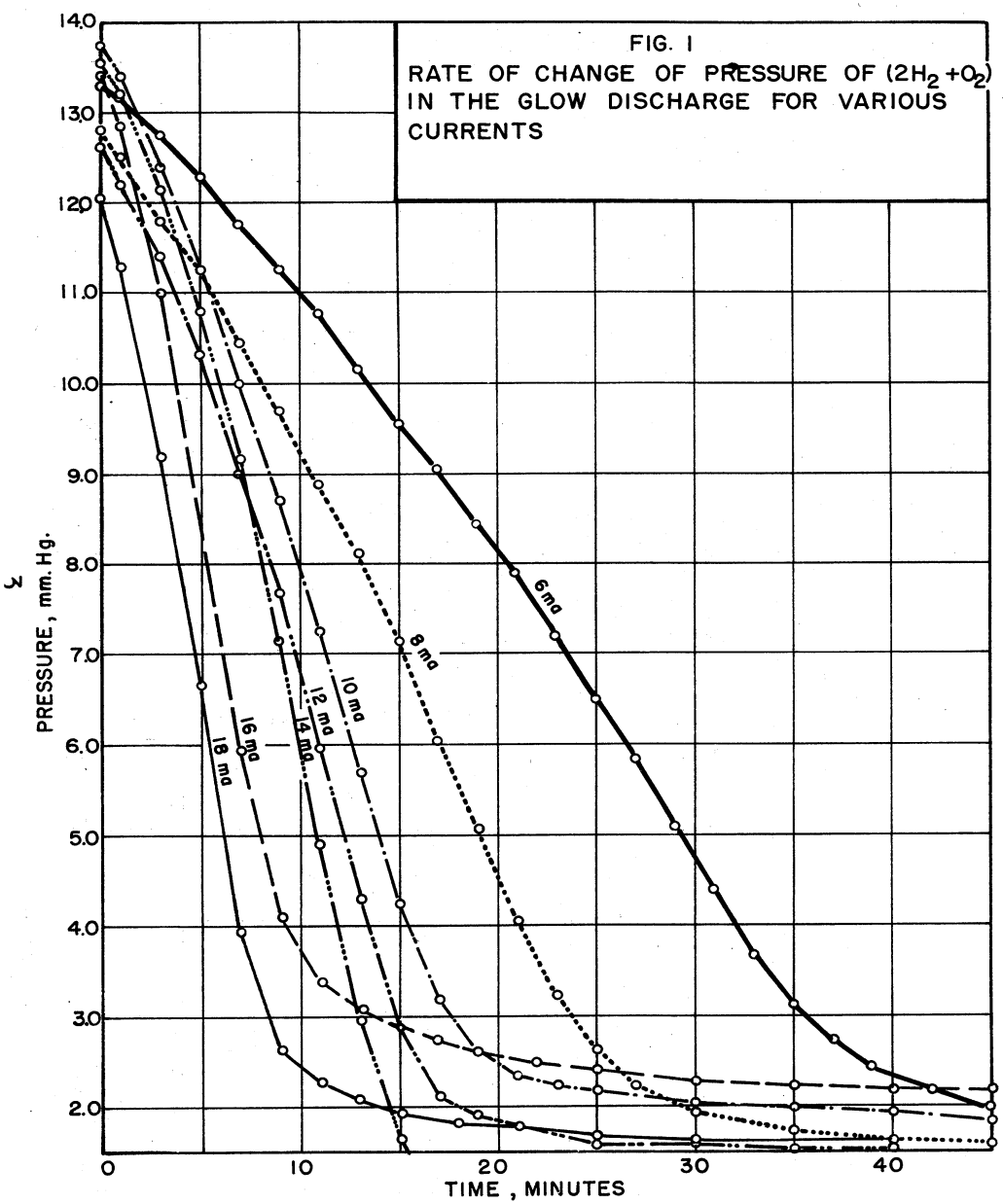


FIG. 1
RATE OF CHANGE OF PRESSURE OF ($2H_2 + O_2$) IN THE GLOW DISCHARGE FOR VARIOUS CURRENTS
PRESSURE, mm. Hg.
TIME, MINUTES
6 ma
8 ma
10 ma
12 ma
14 ma
16 ma
18 ma

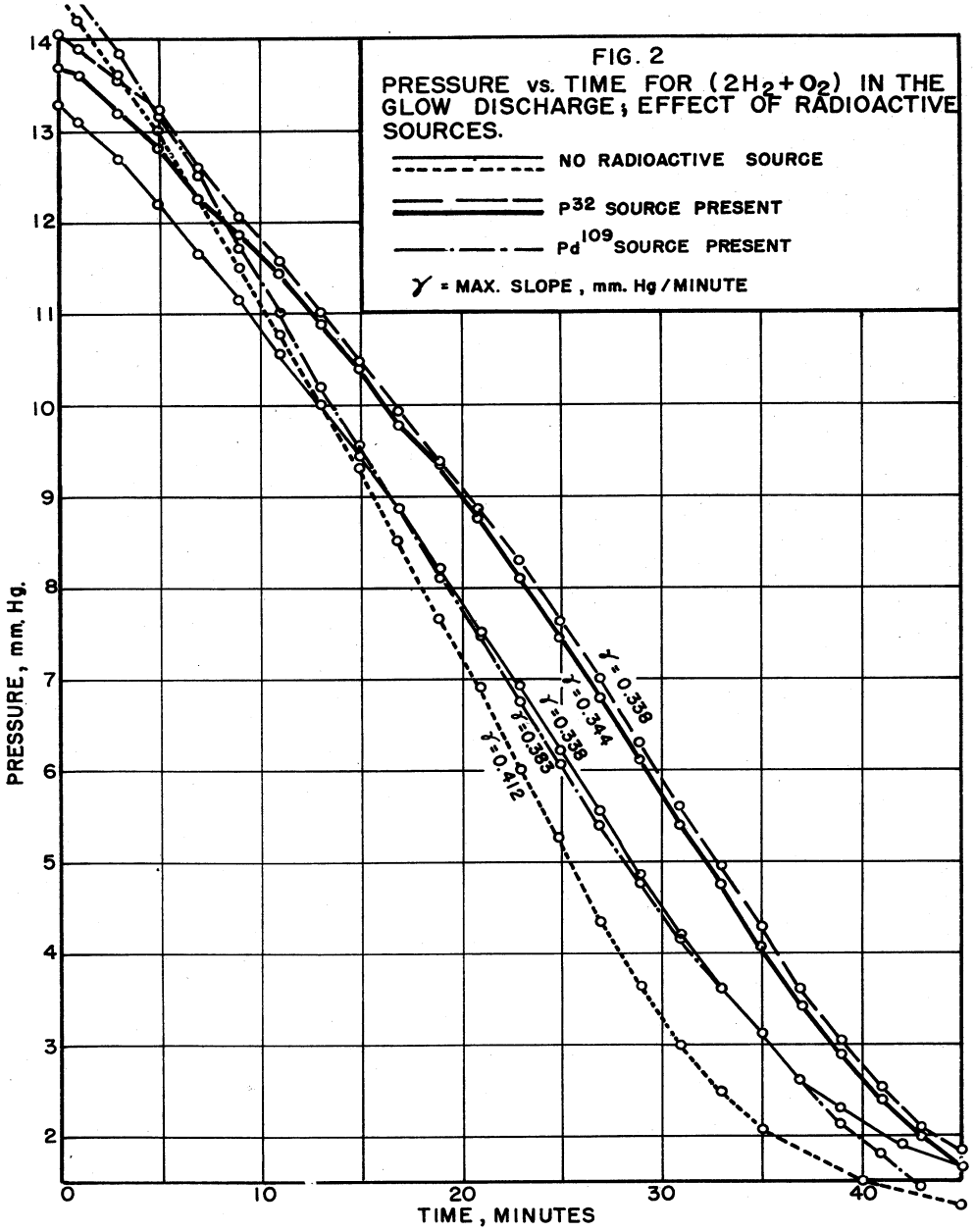


FIG. 2
PRESSURE vs. TIME FOR ($2H_2 + O_2$) IN THE GLOW DISCHARGE; EFFECT OF RADIOACTIVE SOURCES.
NO RADIOACTIVE SOURCE
P^{32} SOURCE PRESENT
Pd^{109} SOURCE PRESENT
γ = MAX. SLOPE, mm. Hg/MINUTE
γ = 0.338
γ = 0.344
γ = 0.338
γ = 0.383
γ = 0.412
PRESSURE, mm. Hg.
TIME, MINUTES

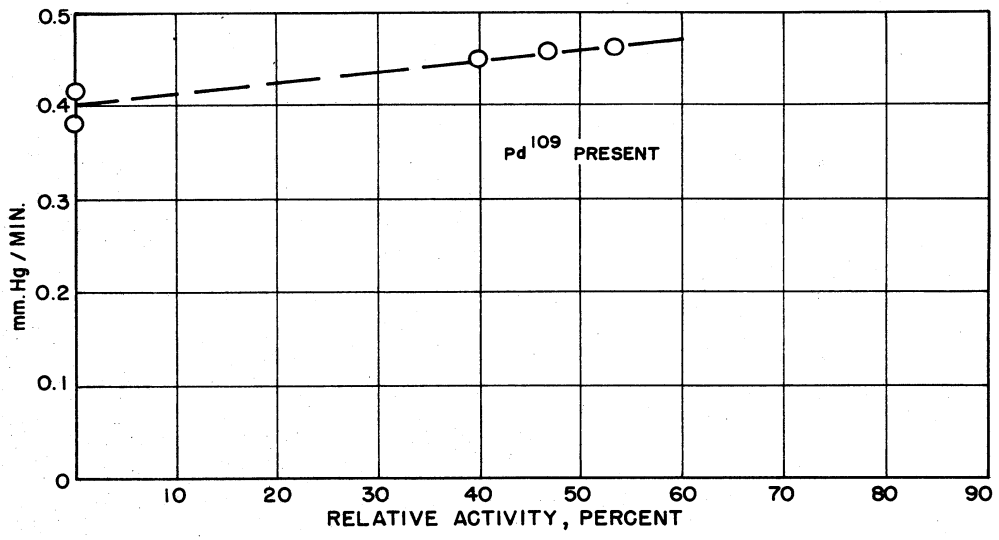
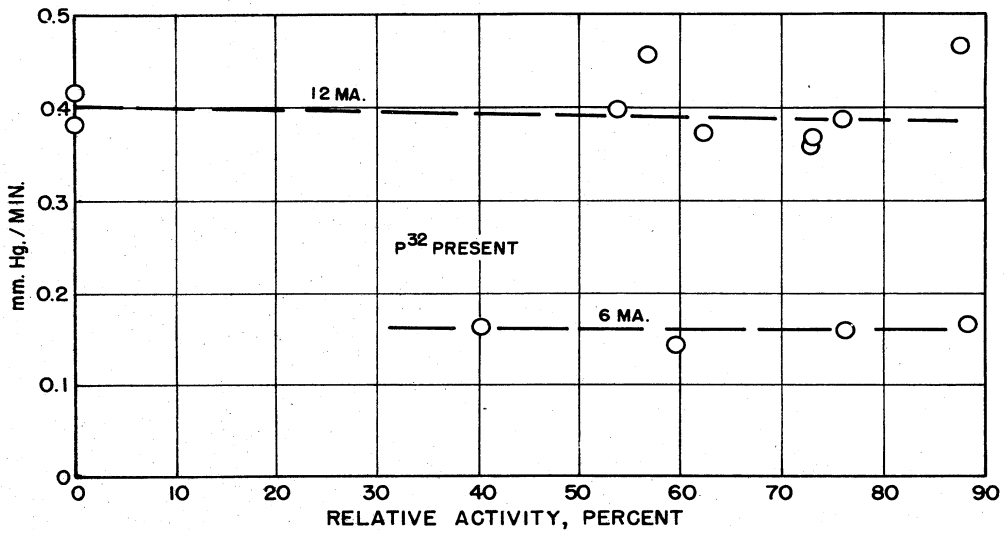


FIG. 4
REACTION RATIO DUE TO GLOW DISCHARGE IN ($CH_2 + 2O_2$)
AS A FUNCTION OF RELATIVE SOURCE STRENGTH



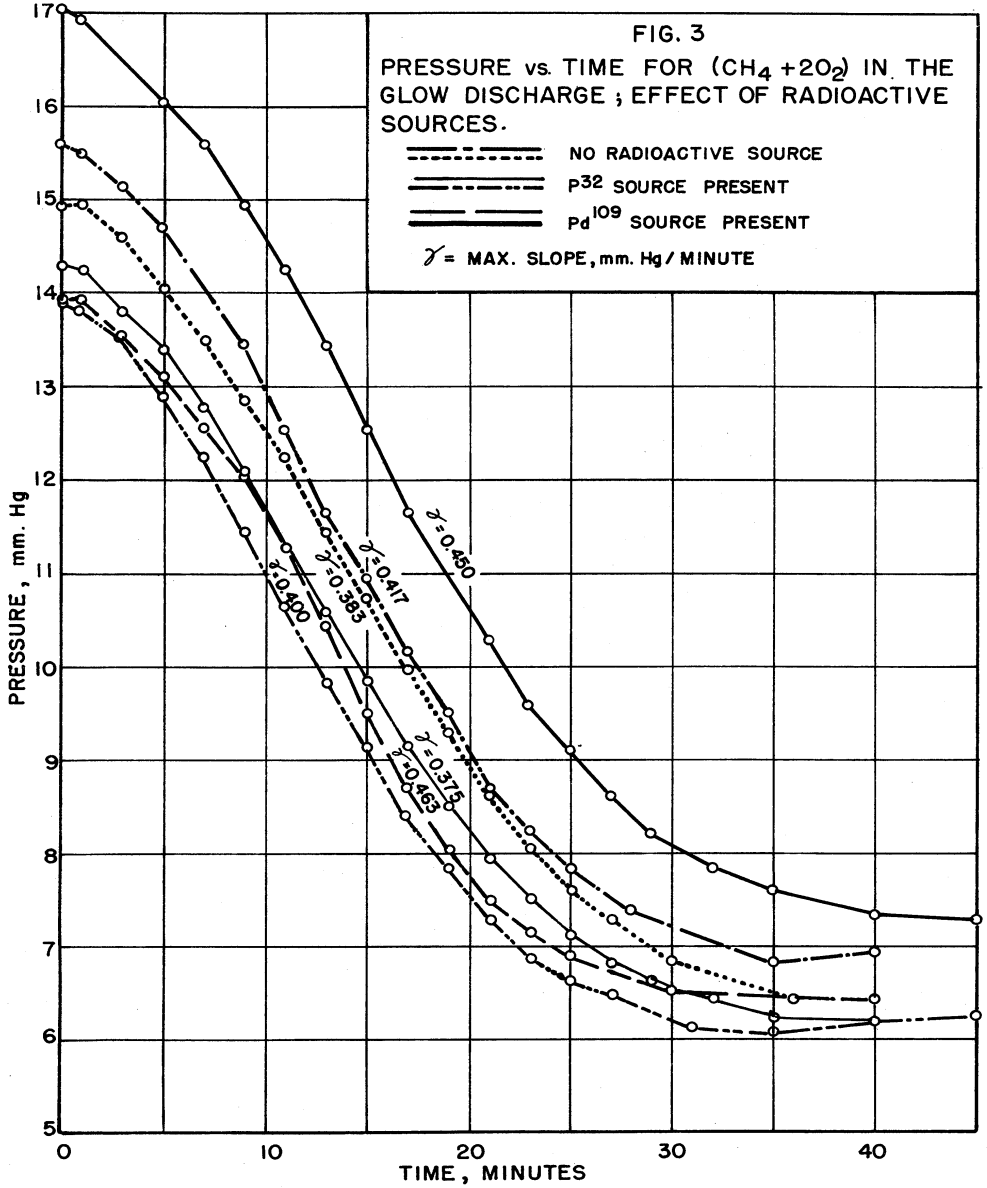


FIG. 3
PRESSURE vs. TIME FOR ($CH_4 + 2O_2$) IN THE GLOW DISCHARGE ; EFFECT OF RADIOACTIVE SOURCES.

comparison of the pressure-time curves with no radioactive source, with the phosphorus-32 source, and with the palladium-109 source. Fig. 4 is a plot of the maximum slope of the pressure-time curves versus the relative activity of the radioactive sources. The effects of the two sources are not directly comparable because of the differences in their energy spectra, matrix materials, and geometry. It might be thought that curve B indicated an accelerating effect for palladium-109. However, a comparison with observed experimental errors shows that no such conclusion is warranted. The spread in the points of curve A bears this out.

b. Use of Aluminum Foil as "Moderator". From a consideration of the range and general behavior of beta particles in air (see part 2), it was thought possible that the experiments described in 1 above failed because of inadequate absorption of the beta-particle energy in the gas mixture. An attempt was therefore made to increase the relative proportion of slow betas, which are several-fold more effective in producing ionization than the high-speed beta particles. For this purpose the reaction vessel was packed losely with shredded aluminum foil 0.8-mil thickness (5.5 mg/cm^2). The range of a 1-Mev beta particle in aluminum is about 470 mg/cm^2. It was reasoned that the high-energy beta particle would pass through many thicknesses of the foil, and be decelerated in the process. At least part of the low-energy portion of the path would then lie in the gas between the foils. The glow discharge was not used in this experiment. Instead, pressure readings were taken with the McLeod gauge to see if any pressure drop was produced in the hydrogen-oxygen mixture by the radiation.

The source used in this case was a palladium foil which had lost most of its activity from palladium-109 through decay, but still had an appreciable concentration of silver-111, which has a half-life of about 8 days and a maximum beta-particle energy of about 1 Mev. The strength of the source was calculated to be 0.6 curie.

A slow pressure drop was actually observed, but since it leveled off in time and since it also occurred without any radioactive source present, it was concluded that adsorption of hydrogen on the large surface area of the aluminum foil was responsible. This adsorption masked any effect the radiation may have produced.

Because of the absence of any measurable results in the experiments described above, it is clear that a new approach must be tried in order to increase the effectiveness of the beta radiation materially.

2. Review of the Problem

It seems desirable to review the fundamental nature of the problem in some detail in order to account for the failure of the foregoing experiments and to design a new set of experiments which will have a better chance of success. Such a review must consider both the nature of the interaction of beta radiation with matter (with the emphasis on gases) and the basic mechanism of explosions in gaseous mixtures.

a. Behavior of Beta Radiation. The basic experimental facts of beta radiation are well known and will merely be enumerated here for convenience in what follows.

The energy distribution of beta particles from radioactive material is continuous up to a definite maximum energy and varies approximately as indicated in Fig. 5.

According to the theoretical calculations[25] the peak of the curve should occur at 0.4 times the maximum energy and the mean energy at about 0.42 times the maximum.

It is known that beta particles produce ionization in gases and, in fact, that the energy loss per ion produced is about 35 electron-volts. Since the ionization potentials of oxygen and nitrogen are only about 14 volts, it is clear that considerable energy must be absorbed in some other way. While some of this energy may be accounted for by the kinetic energy imparted to the ions, it is reasonable to suppose that some energy goes to produce excited states in the neutral molecules and to the splitting of molecules into free radicals. This idea is supported indirectly by the work of Essex and Smith[5] with alpha particles. It may be expected that the effects of alpha and beta particles on matter should be qualitatively similar. Essex and Smith found that when ammonia was bombarded by alpha radiation, not more than about 30 per cent of the decomposition was attributed to ionization.

The work of Lind and Vanpee[7] also supports the idea that ionization is only one means by which charged particles lose energy. They found that when hydrogen-oxygen mixtures were irradiated with alpha particles, the rate of reaction was increased by the addition of xenon to the mixture. Yet the ionization potential of xenon is less than that for either hydrogen or oxygen, so that there could be little possibility of the reactant gases being ionized by collisions of the second kind with xenon. It was concluded that a major share of the reaction was produced by means other than ionization of the reactants.

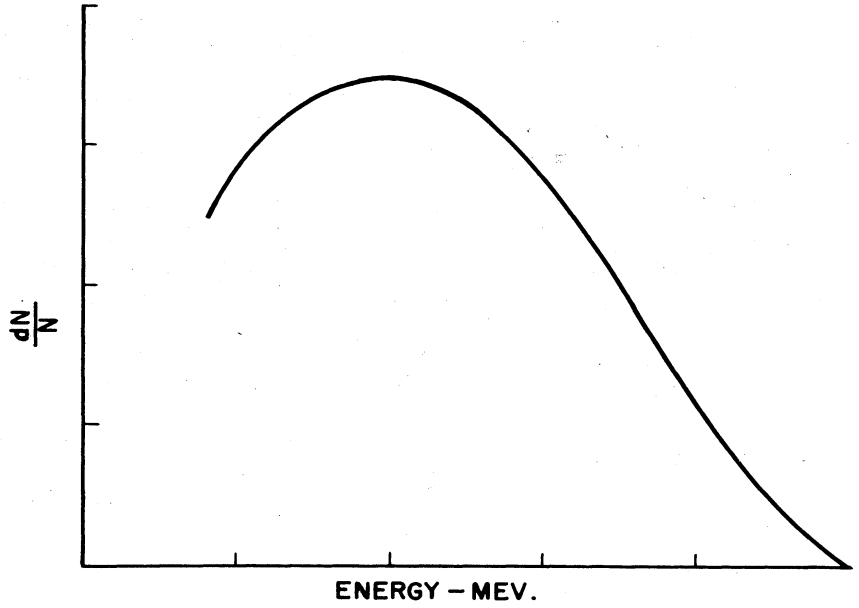
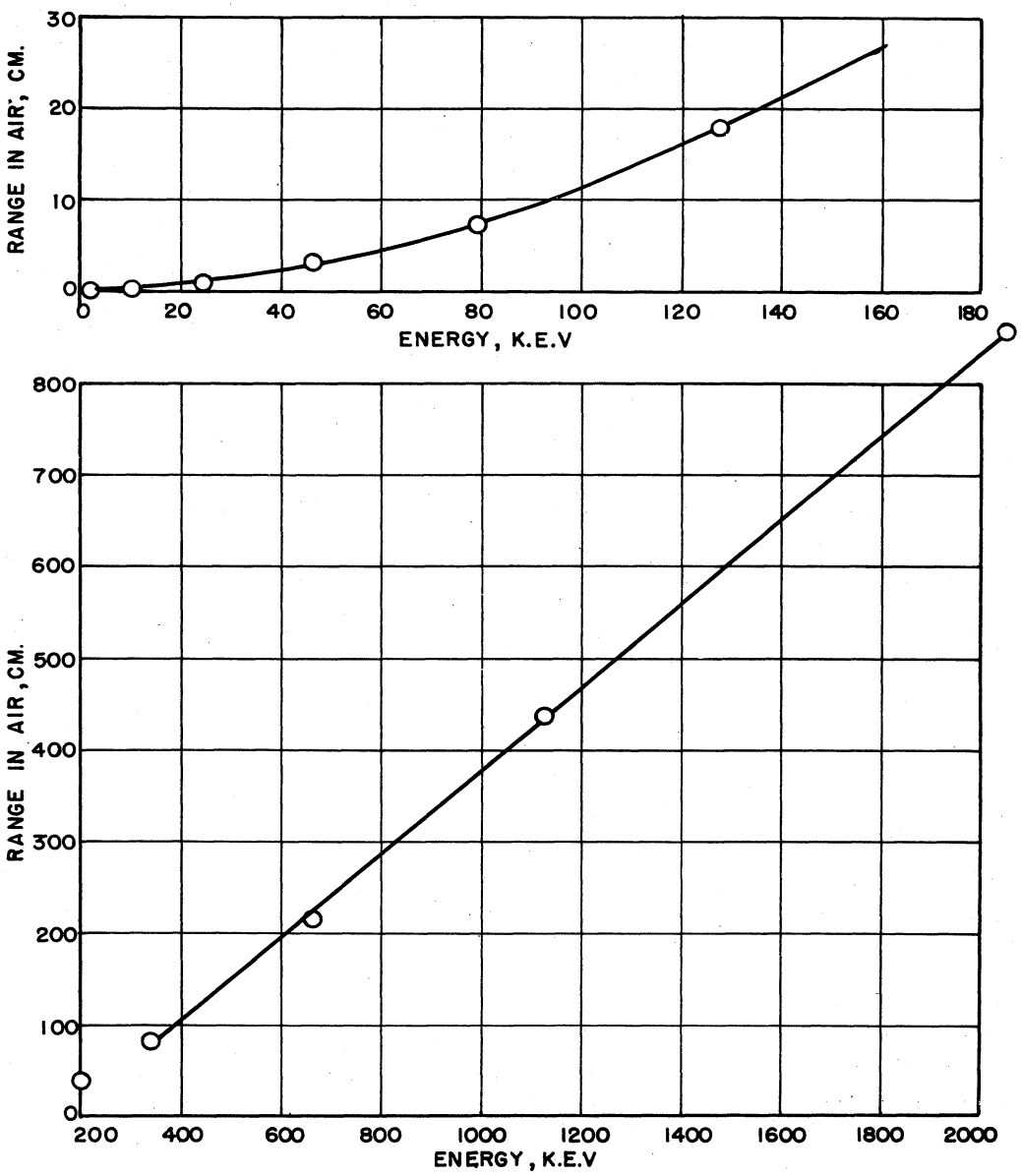


FIG. 5

ENERGY SPECTRUM OF A BETA SOURCE

FIG. 6

RANGE OF BETA PARTICLES IN AIR AS A FUNCTION OF ENERGY.



Above about 0.2 Mev the range of beta particles in air is practically a linear function of energy (Fig. 6)*. Specific ionization is much greater for low energies than for high, so that the greatest specific ionization occurs near the end of the path.

Bohr[8] and Bethe[9] have derived theoretical expressions for the rate of energy loss of a charged particle moving through matter. They show that to a good approximation the rate of loss is directly proportional to the concentration of electrons. This is supported experimentally by the work of Otvos[6], who found that for the hydrocarbon series, probability of ionization by carbon-14 radiation increased with the number of electrons. He also found that for iso-electronic gases a multi-atomic gas (CH_4) showed a higher ionization probability than a monatomic gas (Ne).

Another experimental phenomenon of possible importance is that of "reflection" of beta particles by solids. Burtt[10], working with very thin sources backed by various materials, found the increase in number of counts, due to scattering, appeared to be directly proportional to the atomic number of the backing material. For heavy elements, such as palladium or tungsten, he found the number of counts was increased by as much as 79 per cent over the number obtained with an unbacked source. He also found that the per cent reflection was independent of the maximum beta energies of sources having a maximum beta energy greater than 0.6 Mev. On the basis of the energy distribution of beta particles from a single source (Fig. 5), it would seem that the per cent reflection for monoenergetic beta particles must be independent of energy above 0.2 Mev.

From the considerations above, it is possible to draw some conclusions as to the conditions under which beta radiation will be most effective. It is clearly essential that nearly the entire path should lie inside the reaction region for the major part of the beta particles.

The maximum energy of the beta radiation from phosphorus-32 is 1.7 Mev and the average energy is about 0.7 Mev. From Fig. 6 the corresponding ranges in normal air at S.T.P. are about 7 meters and 2.5 meters, respectively. For palladium-109 the maximum energy is about 1.0 Mev and the average energy about 0.4 Mev. The corresponding ranges are 3.8 meters and 1.1 meters, respectively.

Since rate of energy loss is proportional to the electron concentration, which is in turn proportional to the concentration of a given gas,

*From data published by James E. Cork, Radioactivity and Nuclear Physics, 2nd Ed. D. VanNostrand Co. Inc., New York, 1950. By permission.

it follows that path length should be approximately inversely proportional to pressure at a fixed temperature.

If we calculate the range of an average beta particle from palladium-109 at a pressure of 1 cm, we find a value of about 830 meters. Using a figure of 35 electron-volts per ion pair and a vessel diameter of 15 cm, we can predict that such an average electron will produce no more than a single ion pair within the reaction chamber under the conditions described in part A. This compares with a total of some 11,000 ion pairs which the particle is capable of producing. In the experiments described the number of ion pairs is still further reduced by two additional factors: (a) the specific ionization of a high-energy particle at the beginning of its path is much less than the average number of ion pairs per cm which was used in the above calculation, and (b) the gases used in the experiment included methane and hydrogen, which have fewer electrons per molecule and hence presumably less stopping power than the molecules of air. Thus, it appears that in the experiment described in part A only a very small fraction of the capabilities of the beta radiation was utilized. The introduction of the aluminum foil was an attempt to remedy this situation by absorbing some of the energy of the betas and thus increasing the specific ionization along the paths of the beta-particles within the chamber.

We are interested in obtaining the highest possible concentration of excited molecules of all types within the reaction space. These excited molecules cannot be detected directly at present, but we assume that the ion-pair concentration provides a measure of the concentration of the excited molecules.

Increasing the dimensions of the reaction vessel is of no value. While it will permit more efficient use of the energy of the beta particles, it will not increase the ion concentration except near the end of the path.

The most obvious way of increasing the electron concentration is to increase the pressure. Thus, at 20 atmospheres of air the maximum range of phosphorus-32 beta particles would be about 35 cm and the average range about 12.5 cm.

Another way of increasing electron concentration is applicable in an experimental system where total pressures may be limited and where an inert gas may be used. This is to use a gas having a large number of electrons per molecule. Thus, for example, the gas hexafluoro ethane is very stable and has 66 electrons per molecule as compared with 14 per molecule of nitrogen.

It may also be possible to increase the ion concentration by increasing the number of electron trajectories due to the same source within the volume of the reaction vessel. This might be accomplished by lining the

reaction vessel with one of the heavy metals. According to the figures of Burtt[10], certain of the heavy metals will reflect about 80 per cent of the impinging beta particles, while iron reflects only about 40 per cent. Thus after two reflections not more than 16 per cent of the original beta particles would remain in a steel vessel, while with a heavy metal liner 64 per cent would be retained, discounting other effects. In practice these figures will not be attained, because those beta particles arriving at the wall with much less than 0.2-Mev energy will be absorbed in a much higher proportion. Also, there is probably some energy loss in the walls.

From the point of view of cost and convenience, lead seems to be the most logical choice for the liner in an experimental explosion system. On the basis of the previous paragraph, it might appear that the smallest vessel dimensions would be most suitable. It must be remembered, however that a high-level radioactive source may be embodied in a large mass of material which may itself absorb a large share of the reflected beta particles if the walls are too near. For example, the phosphorus-32 was contained in a bakelite matrix which would absorb most incident beta particles. Hence, the reaction vessel should be large enough so that the source material does not intercept a large solid angle at the walls.

It should be emphasized that the methods proposed above for increasing the concentration of excited molecules in a reaction chamber have been considered from the point of view of attaining the desired result in a set of highly simplified basic experiments. These methods are not necessarily regarded as directly applicable to practical combustion devices in their present form. Rather, the approach to a specific practical application would require that one work from the principles lying behind the methods described above, keeping in mind the limitations imposed by the particular device under consideration.

b. Gaseous Explosions. The chain theory of explosions in gases postulates that reactions take place through the agency of "chain carriers", i.e., some type of active molecule or radical.[11] These carriers take part in a reaction "cycle" with the constituents of the gaseous mixture and are eventually regenerated. More than one type of carrier may be involved in the cycle. If in the course of one cycle a single carrier of the original type is reproduced, the reaction chain is said to be "straight". If more than one of the original carrier is reproduced in a given cycle, the chain is said to be "branched". In the latter case it is clear that if there are no competing reactions the concentration of chain carriers will rapidly approach infinity and explosion will occur. An example of a branched chain cycle is that proposed by Lewis and Von Elbe[14] as the basic chain-branching mechanism involved in the explosion of hydrogen-oxygen mixtures:

$$H + O_2 \rightarrow OH + O \quad (1)$$

$$O + H_2 \rightarrow OH + H \quad (2)$$

$$OH + H_2 \rightarrow H_2O + H \quad (3)$$

If in the course of a cycle $(1 + \alpha)$ of the original chain carriers are regenerated, then α is said to be the chain branching coefficient. If n is the concentration of chain carriers, then αn is the rate per cycle at which n is increasing. In the above example $\alpha = 2$.

If the possibility exists of competing reactions which destroy the chain carriers, then the phenomenon of the explosion limit may be observed. These are said to be "chain-breaking" reactions. They may involve reactions in the gas phase or they may be the result of diffusion of the chain carriers to the wall and subsequent destruction.

Hydrogen-oxygen mixtures exhibit the phenomenon of explosion limits. For a given composition and temperature (above 400°C), it is found that explosion occurs within certain sharply defined limits of pressure. If the pressure is below or above these limits, there is no explosion. Furthermore, a third limit may be observed for certain proportions of hydrogen-oxygen and certain temperatures; i.e., if the pressure is steadily increased above the second limit value, a pressure will eventually be attained above which explosion again occurs.

It may be assumed that whenever the rate of chain branching exceeds the rate of chain breaking, an explosion will occur. The conditions under which the rate of branching just equals the rate of chain breaking must define the explosion limit.

A reaction is said to be of first order with respect to the chain carriers if, in each step of the cycle, a chain carrier reacts with one of the major components of the gas mixture. The reaction is of second order if one of the steps involves the interaction of two chain carriers.

Consider first a first-order chain branching reaction. Let

I = rate of initiation of chain carriers

αn = rate of generation by chain branching

βn = rate of loss by chain breaking.

then

$$\frac{dn}{dt} = I + \alpha n - \beta n.$$

When a steady state is reached,

$$\frac{dn}{dt} = 0$$

and

$$n_o = \frac{I}{\beta - \alpha} .$$

The explosion limit is then defined by

$$\alpha = \beta$$

It is important to note that I, the rate of carrier initiation, cannot affect the conditions of the explosion limit for a first-order chain branching reaction. If the branching reaction is of second order, then

$$\gamma n^2 = \text{rate of branching}$$

and

$$\frac{dn}{dt} = I + \gamma n^2 - \beta n = 0$$

$$n_o = \frac{\beta \pm \sqrt{\beta^2 - 4\gamma I}}{2\gamma}$$

For $4\gamma I > \beta^2$, n_o becomes imaginary and there is no steady-state condition. The explosion limit is then defined by

$$\beta^2 = 4\gamma I .$$

Egerton and Warren[16,17] have developed a comprehensive theory to explain the entire explosion region of hydrogen-oxygen mixtures under the condition of constant temperature throughout the entire volume of the gas. These results are based in part on the previous work of Frost and Alyea[12], Grant and Hinshelwood[13], Lewis and Von Elbe[14], and Willbourn and Hinshelwood[15]. In all of this work explosion was produced by an elevated temperature maintained throughout the volume of the gas by a furnace.

The chain breaking reaction which defines the second limit under these conditions is believed to be the ternary collision:

$$H + O_2 + M \rightarrow HO_2 + M \quad , \tag{4}$$

where M is any molecule capable of acting as a third body to remove the excess energy of the reaction.

The first limit occurs at very low pressures and is defined by the diffusion of H, O, and OH to the walls, where they are destroyed. The third limit is due to the increased probability at higher pressures of a straight chain reaction which regenerates a chain carrier:

$$HO_2 + H_2 \rightarrow H_2O + OH \quad , \tag{5}$$

All the reactions mentioned above are of first order, and presumably the corresponding explosion limits could not be influenced by radiation.

Egerton and Warren[16] report that when the inner surface of the reaction vessel is coated with B_2O_3, then a second-order chain reaction becomes significant. They attribute this to a failure of the mechanism which destroys HO_2 at the walls, so that it becomes available for the reaction.

$$H + HO_2 \rightarrow OH + OH \quad , \tag{6}$$

such a reaction should be subject to radiation effects.

When the explosive mixture is ignited by a spark instead of in a furnace, conditions are radically different. The chain carriers are produced in large concentrations in a very small region and the effective temperature in the spark is extremely high. If the pressure is high, as it must be if the radioactive source is to be efficiently used, then conditions would tend to favor second-order reactions.

Mole[18] has carried through a mathematical analysis of the ignition of gaseous explosions. He has assumed that the reactions are of second order and he shows that in such a case explosions can always be produced if the initial concentration of chain carriers is high enough.

Linnet and Frost[19] studied ignition of hydrogen-oxygen below 500 mm pressure. In their analysis they assumed the first-order chain branching reaction described above, but did not take into account any second-order reaction or the homogeneous chain breaking reaction (reaction 4). They assumed that chain carriers were lost by diffusion out of the spark region. The agreement between their experimental results and theory is best at low pressures, as might be expected.

The combustion of hydrocarbons is exceedingly more complex, although the occurrence of both slow combustion and detonation in hydrocarbon-air mixtures indicates that both straight and branched chain reactions must take place. Some suggestions have been made as to the nature of these reactions[20-24] but their validity is as yet far from certain.

From the above considerations, there appears to be a good possibility that intense beta radiation can alter some of the characteristics of gaseous explosions, under proper conditions. Measurements of the explosion limits require less equipment than measurements of flame speeds or pressure peaks in an explosion, so this type of measurement will be tried first. Additional measurements of other types may be desirable at a later date.

3. Projected Explosion System

In line with the conclusions reached in part B, an explosion system has been designed to take full advantage of the radiation from beta sources. The components of this system are on order and will be assembled as soon as they are available. Fig. 7 is a schematic diagram of the proposed explosion system. It will consist of an explosion bomb and the necessary accessories, which will be connected through a superpressure check valve to an ordinary medium-pressure feed system. Gases will be admitted to the bomb through constant-pressure diaphragm-type regulators and needle valves. Pressure measurements will be made with precision gauges which will be in addition to the gauges of the regulators. A mechanical pump will be used to evacuate the system prior to each run. All exhaust outlets to the system will be fed into the exhaust manifold of the Fission Products Laboratory.

The bomb will consist of a manganese-steel superpressure reaction vessel manufactured by the American Instrument Company, together with the necessary superpressure accessories. Its dimensions are:

outside diameter	6-7/8 inches
inside diameter	4-1/16 inches
inside depth	40 inches

It will be provided with one electrical connection and two pressure connections. One of the pressure connections will be attached to the feed and exhaust system, while the other will be connected to a rupture disc and blow-out assembly which will rupture on explosion.

In order to increase the amount of beta-particle reflection at the walls, a thin cylindrical lead liner will be constructed for the reaction

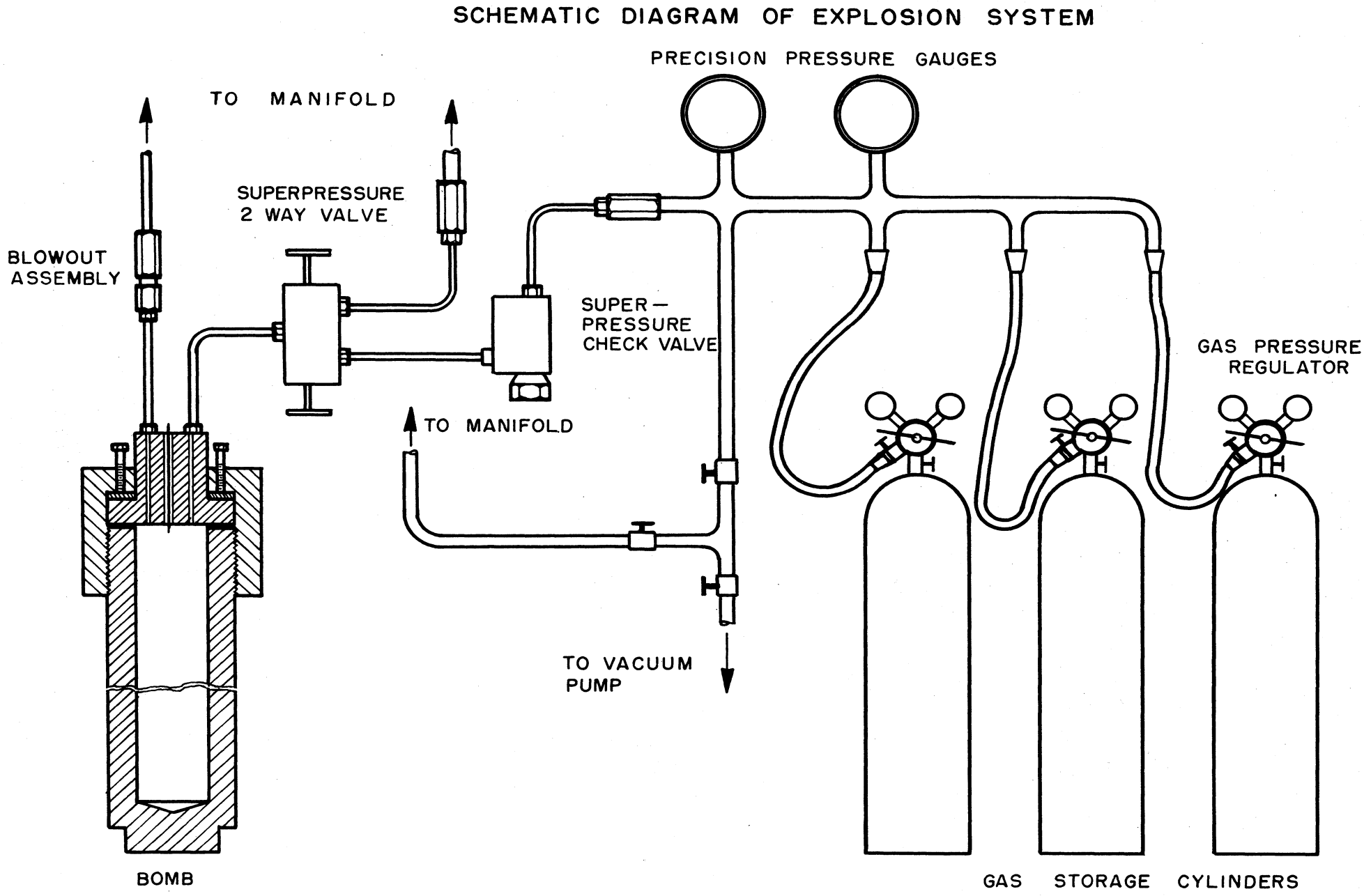

SCHEMATIC DIAGRAM OF EXPLOSION SYSTEM
PRECISION PRESSURE GAUGES
TO MANIFOLD
SUPERPRESSURE
2 WAY VALVE
BLOWOUT
ASSEMBLY
SUPER—
PRESSURE
CHECK VALVE
GAS PRESSURE
REGULATOR
TO MANIFOLD
TO VACUUM
PUMP
BOMB
GAS STORAGE CYLINDERS

FIG. 7

vessel. According to the results obtained by Burtt, the lead should about double the percentage of reflections at the wall.

It is expected that ignition will be produced by means of a standard spark plug mounted inside the reaction vessel.

High-intensity palladium or phosphorus beta sources will be used in the reaction vessel. Use of the latter will depend on whether or not the phosphorus-impregnated bakelite which is the usual source material will withstand the conditions existing in the reaction vessel during explosion.

A tentative experimental procedure has been worked out as follows: hydrogen and oxygen will be admitted to the reaction vessel at fixed pressures; varying pressures of nitrogen will then be added and the mixture will be sparked. The pressure of nitrogen at which explosion just fails to occur determines the explosion limit for the given pressures of hydrogen/oxygen and the apparatus. By repeating the procedure for different pressures of hydrogen/oxygen, an explosion limit curve such as that in Fig. 8 may be obtained.

The set of experiments may then be repeated with high-intensity beta-particle sources present in the reaction vessel.

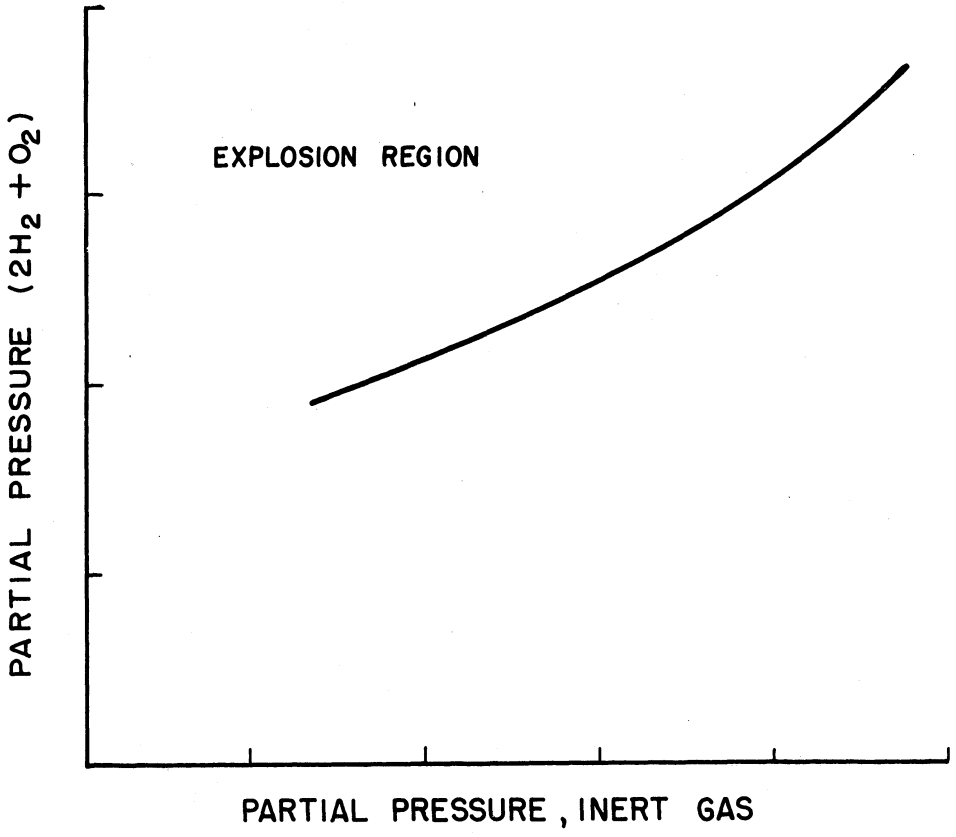


FIG. 8
EXPLOSION LIMIT CURVE

If the radiation is capable of producing any appreciable effect on the explosion limits of the gas, one should observe a measureable shift of the explosion-limit curve.

The apparatus described above should be adaptable to making measurements of flame-front velocities or to pressure-time measurements if these data appear desirable at a later date.

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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

a. Introduction. The effect of beta radiation on the combustion process of reciprocating internal-combustion engines is being investigated under actual operation. At present, the work is limited to diesel engines, but an investigation into the spark-ignition engine is also planned. To date, one test has been completed.

b. Description of Test. On February 4, 1952, a test was conducted on a single-cylinder variable-compression-ratio Wauhesha CFR diesel engine. The source of radiation was palladium-109 in the form of a palladium disc 1-1/2 inches in diameter, 1/8 inch thick, having a mass of 33 grams, which was attached to the fore of the compression plug in the combustion chamber above the main cylinder. The test procedure was to run two series of tests, a cold test with the inactive palladium disc present and an identical test with the irradiated palladium disc. The effects of the radiation were indicated by differences between the two series of tests.

In each series, the following four tests were conducted (details of these tests have been described in Progress Report 2):

1) Indicated cetane number of the fuel.

2) Minimum firing compression ratio.

3) Pressure-time measurements of the combustion process.

4) Specific fuel consumption.

The palladium was irradiated in the Brookhaven pile. It was removed from the pile at 7 A.M. on February 4, 1952, and flown to Detroit's Willow Run Airport; thence it was transported by car to the Automotive Laboratories of the

University of Michigan. The tests were conducted between 2 P.M. and 5 P.M. on the same afternoon. The average activity during the tests was 41 curies. Only a small fraction of this activity was effective, however, since all beta particles beneath the outer 13 mils of palladium metal expend their energy in the metal disc itself due to the short range of the beta particle in palladium. The radiation flux from the disc is thus dependent on this surface beta-particle radiation and on the brehmstrahlen due to the thick beta-particle source.

c. Results. (1) Indicated cetane number of the fuel: The basis of this test is to determine the compression ratio at which ignition will occur at top dead center when injection takes place 13° before top dead center. Following is a summary of the results.

Date	Jan. 21	Feb. 4	Feb. 13
Palladium condition	unirradiated	irradiated	unirradiated
Air Temperature, °F	149	152	150
Cooling-water temperature, °F	208	206	207
Fuel temperature, °F	84	80	82
Barometer, inches mercury	---	28.74	29.31
rpm	1100	1100	1100
cc fuel/min	15	15	15
Injection timing	13°BTDC	13°BTDC	13°BTDC
Ignition timing	TDC	TDC	TDC
Compression ratio scale reading	1.437	1.429	1.424
Compression ratio	14.48	14.57	14.60

It proved difficult to obtain accurate results on this test, and results cannot be duplicated precisely. This test would indicate that beta-particle radiation does not change the cetane number, but the accuracy of the test is not great enough to preclude the possibility of there having been a small change in cetane number.

(2) Minimum firing compression ratio: The procedure was to close the fuel injection value for five seconds each minute, and determine the lowest compression ratio at which the engine fires, all other conditions being held constant. Results are as follows:

Date	Jan. 21	Feb. 4	Feb. 13
Palladium condition	unirradiated	irradiated	unirradiated
rpm	900	900	900
Air temperature, °F	149	150	150
Cooling-water temperature, °F	208 1/2	207	208
Compression ratio scale reading	1.78	1.83	1.79
Minimum firing compression ratio	11.72	11.41	11.66

This test indicates that the engine will fire at a slightly lower compression ratio when the combustion takes place in the presence of beta-particle radiation. Results could be duplicated quite closely, and the test is believed to be fairly reliable. The percentage reduction in minimum firing compression ratio is 2.5 per cent.

(3) Pressure-time measurements: Measurements of the pressure-time relation in the combustion chamber were made by photographing the pressure-time trace on the oscillograph, the pickup device being of the Control Engineering catenary-diaphragm type. Photographs were made with irradiated palladium on February 4, 1952, and a cold run was made on February 13, 1952. Fig. 9 shows a typical pressure-time trace. The light dots are the calibration points for pressures, the ordinate of these dots corresponding to a pressure to 500 psi.

From these photographs the pressure at which ignition occurred was determined. Results are given below:

Compression Ratio	rpm	cc fuel/min	Pd^{109} Unirradiated	Pd^{109} Irradiated	Per Cent Decreased
13 to 1	1100	15	490	480	2.0
16 to 1	1100	15	671	614	8.5
19 to 1	1100	15	795	716	9.9
19 to 1	1200	22	722	598	17.2
16 to 1	1200	22	590	488	17.3
13 to 1	1200	22	494	477	3.4

It is noted that in every case the pressure at which ignition occurred was lower when the combustion took place in the presence of beta radiation. A decrease in the pressure at which ignition occurs means that there has been a shorter ignition-delay period. It is further observed that the percentage decrease in ignition pressure increases as the compression ratio is increased, and that it also increases as the quantity of fuel injected increases.

(4) Specific fuel consumption: Measurements were made of specific fuel consumption. Two runs were made with the irradiated palladium and two with the unirradiated palladium. The results of all runs are presented in order to show the variation in readings and also the averages for purposes of comparison.

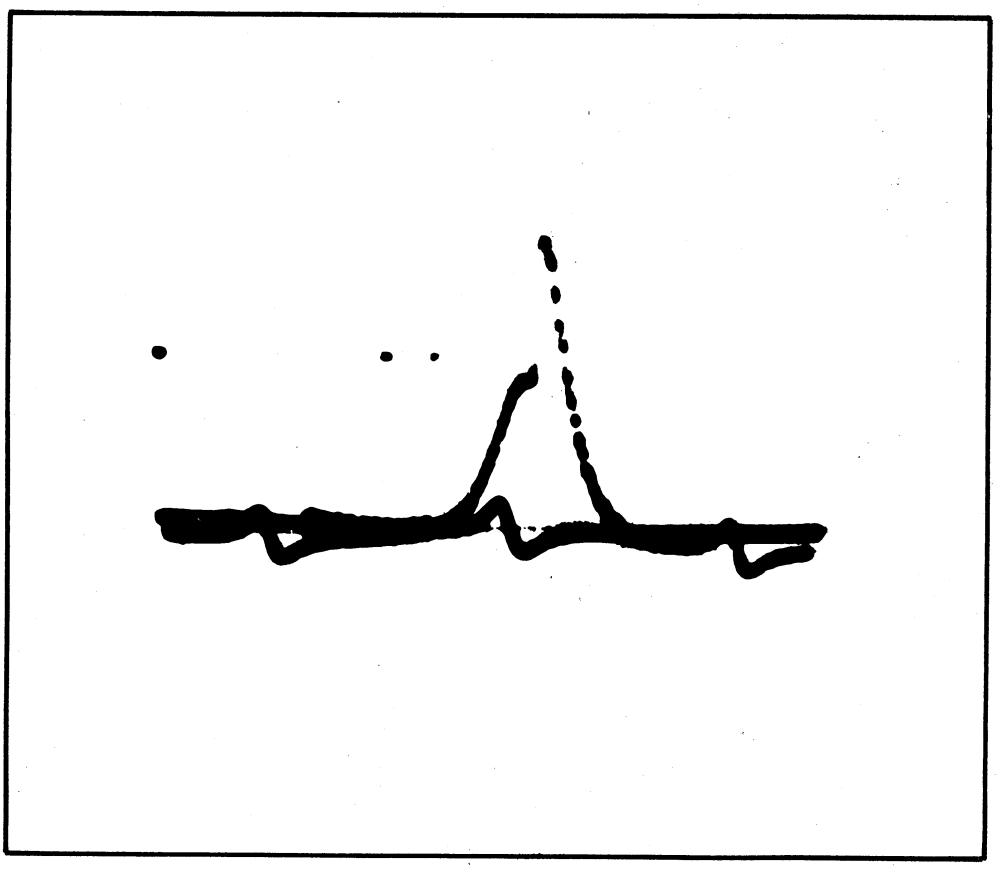

Fig. 9. Typical Pressure-Time Trace.

Compression Ratio	rpm	Palladium Unirradiated 1st Run cc *	lbs**	2nd Run cc*	lbs**	Palladium Irradiated 1st Run cc*	lbs**	2nd Run cc*	lbs**
13 to 1	1100	14.9	1.162	15.0	1.148	15.6	.997	15.3	1.052
16 to 1	1100	14.9	1.061	14.8	.979	15.1	.999	15.5	.967
19 to 1	1100	14.9	.926	14.5	.934	14.9	.901	15.1	.880
19 to 1	1200	23.4	.728	22.4	.711	22.1	.723	22.7	.715
16 to 1	1200	22.7	.767	22.3	.762	22.6	.759	22.7	.738
13 to 1	1200	22.6	.875	22.2	.866	22.1	.810	22.4	.809

These results vary appreciable, and individual readings may be in error by several per cent. However, it is significant that the specific fuel consumption is, in every case except one, less for the runs with beta-particle radiation. In general, this decrease in specific fuel consumption seems to be greater at the lower compression ratios.

Average Specific Fuel Consumption

Compression Ratio	rpm	Pd unirradiated lbs fuel/hp-hr	Pd irradiated lbs fuel/hp-hr	Difference	Per cent Change
13 to 1	1100	1.155	1.025	.130	11.2
16 to 1	1100	1.020	2.983	.037	3.6
19 to 1	1100	.930	.890	.040	4.3
19 to 1	1200	.719	.719	.0	0
16 to 1	1200	.765	.748	.017	2.2
13 to 1	1200	.870	.810	.060	6.9

* cc fuel/min
** lbs fuel/hp-hr

d. Discussion of Results. In Progress Report 2 the combustion process in the diesel engine, as well as the possible effects of radiation on the combustion process were described. The ignition-delay period is the time which elapses between the injection of fuel into the cylinder and the ignition of this fuel. A reduction in the ignition-delay period would result in less detonation and more controlled burning, which would lead to higher efficiency. An increase in the rate of combustion would also lead to higher efficiency. Both a decrease in the ignition-delay period and an increase in the rate of combustion were anticipated as effects of the beta-particle radiation.

All the observed changes as a result of beta-particle radiation are in the direction of improved combustion. The slight change in the minimum firing compression ratio indicates a greater ease of combustion and possibly a shorter ignition-delay period. The effects of beta radiation as observed from the pressure-time measurements are a reduction in the ignition-delay period, and the fuel-consumption tests show a slight reduction in specific fuel consumption due to the beta-particle radiation. In these respects the results are in accordance with those anticipated.

There are two factors which should be borne in mind regarding these results, however. These results are based on a limited number of runs and must be verified by additional experimental evidence. Another factor is that precise duplication of results is difficult to achieve in internal-combustion engine work, especially in view of the period of time which elapses between the runs.

e. Future Program. It is believed that the experiments should be repeated in the presence of a greater amount of radiation. This can be acheived

in two ways, namely, using a greater mass of palladium (increasing the surface area) and more intense irradiation of the palladium. Greater intensity of irradiation is possible in the Chalk River pile, although a limit is placed on the size of the piece which can be irradiated. It was decided that a number of 1/4- inch diameter rods projecting into the combustion space would be an effective means of increasing the mass of the palladium and yet limiting the pieces to a size which can be accommodated in the Chalk River pile.

These rods have been purchased and cold runs are being conducted at the present time. An improved exhaust filtering system has been designed as well as an improved technique in taking the pressure-time photographs. The tests with the irradiated palladium rods are scheduled for the end of July.

2. Jet Engines

a. Introduction. The investigations performed since February, 1952, are a continuation of the experiments which were reported in Progress Report 2. These investigations have attempted to establish whether or not beta-particle radiation affects the combustion processes that are of basic importance to the design and operation of jet engines; namely, the normal flame propagation rate and the flame stability limits.

b. Apparatus and Experimental Procedure. The apparatus used for the experiments has been described in Progress Report 2. Photographs of the apparatus and a description of the experimental procedure appear in section B2(b) of Progress Report 2.

c. Experimental Tests with Radioactive Palladium. Two series of experiments were performed, one on February 19, 1952, and the second on March 2, 1952. In the first series of experiment flame stability was measured A 0.195-gm palladium sphere, 1/8 inch in diameter, containing palladium-109 was used in conjunction with a foil containing palladium-109, 0.001 inch thick, 1-3/4 inches wide and 10 inches long surrounding the flame. The activity of the foil at the beginning of the experiments was approximately 33 curies; the activity of the sphere was approximately 1.8 curies at the same time. The radiation intensity from the palladium sphere was measured at several distances and extrapolated to 1 cm, giving a value of 1300 milliroentgens per hour. The combined radiation from the foil and the sphere was used in the flame-stabilization experiment, with three different fuel-air ratios of 0.067, 0.080 and 0.100 lb propane/lb air. The foil was also used as the source of radiation for the flame-propagation experiment, with a fuel-air mixture of 0.067 lb propane per lb air.

In the second series of experiments a 5-mil palladium foil 1-3/4 inches wide and 12 inches long was used. The foil was received from the Chalk River pile at about 10 P.M. on March 1. As a result of an accident in the handling, however, one half of the foil was damaged. The foil was deformed so that it could not be mounted in the retaining ring used for the flame-propagation experiment, because it would physically interfere with the flame, thus distorting the flame cone. Therefore, the flame-propagation experiment was not attempted and the foil was used only for the flame-stabilization experiment, where the interference with the reaction zone was not nearly so critical. The activity of the foil was approximately 76 curies at the beginning of the experiment. The same procedure was used as in the experiments performed on February 19, with the exception that a spherical steel flameholder, 1/8 inch in diameter, was used.

d. Experimental Results. Fig. 10 is a plot of all the data taken to date relating to the effect of beta-particle radiation on flame-propagation rates. These data include the control experiment, the earlier experiment with a foil activity of 6.6 curies, and the more recent experiment with a foil activity of 36 curies. The results indicate that there is no appreciable difference in any of the three sets of data other than that attributed to experimental scatter.

Figs. 11, 12, and 13 are plots of all the data taken using a 1/8-inch diameter steel flameholder to stabilize the flame. These data include the control experiments, the earlier experiments using the foil with an average activity of 5.6, 5.3, and 4.9 curies, respectively, and the latter experiment with the foil activity of 72, 70 and 76 curies, respectively. These results again indicate there is no measureable difference in the flame stability limits with the three sets of data.

Figs. 14, 15, and 16 show the results of the experiments made using a 1/8-inch diameter spherical palladium flameholder containing palladium-109 in conjunction with a 1-mil foil of palladium-109 surrounding the flame. The data from Figs. 14 and 15 indicate that there is no appreciable difference between the runs using the activated palladium and the control experiment. There does appear to be a slight increase in blow-off velocities at any given pressure for the rich fuel-air mixture of 0.100 lb propane/lb air. The effect appears to be more pronounced at higher pressures. However, because some experimental difficulties were involved in obtaining data at higher pressures with the run made using the activated palladium, no comparison can be made near the upper pressure limit.

e. Conclusions. From the results it can be concluded that beta-particle radiation from a palladium foil surrounding a Bunsen flame at a distance of 1-1/2 inches with activities as high as 36 curies has no effect on the normal

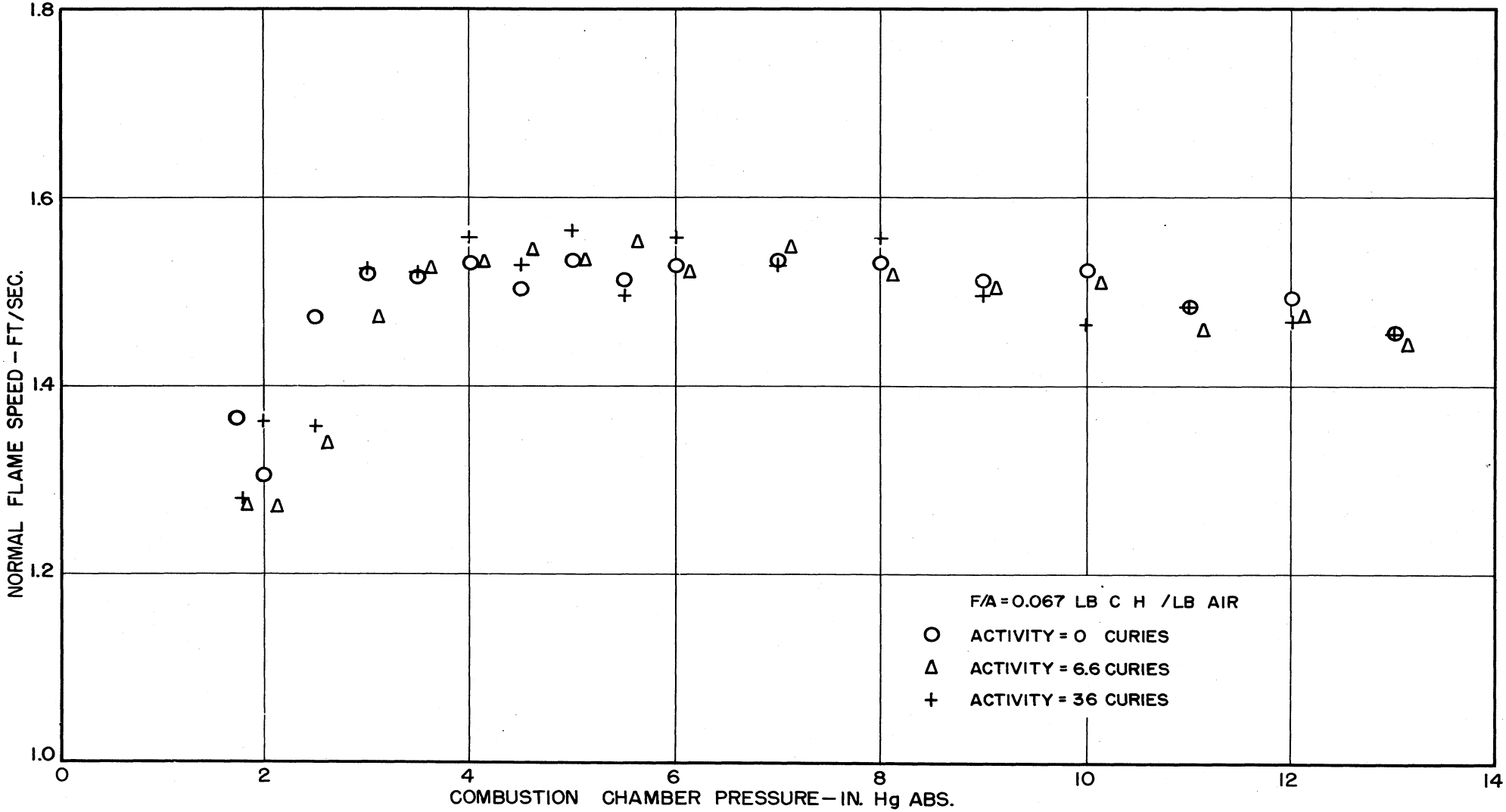

FIG. 10. FLAME PROPAGATION RATES WITH PALLADIUM FOIL OF DIFFERENT ACTIVITIES MOUNTED ABOUT A BUNSEN-TYPE FLAME.

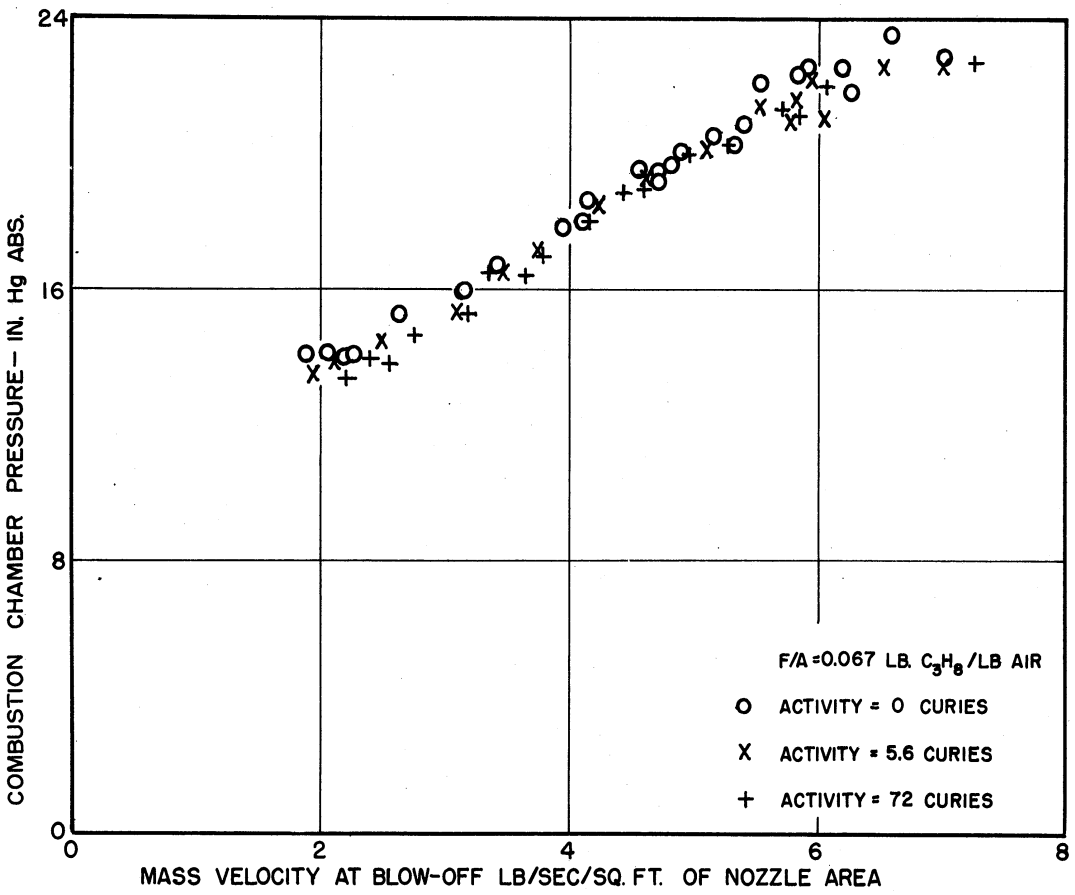

FIG. 11. FLAME STABILIZATION EXPERIMENT WITH PALLADIUM FOIL OF DIFFERENT ACTIVITIES MOUNTED ABOUT A V-TYPE FLAME STABILIZED WITH A STEEL SPHERICAL FLAME HOLDER.

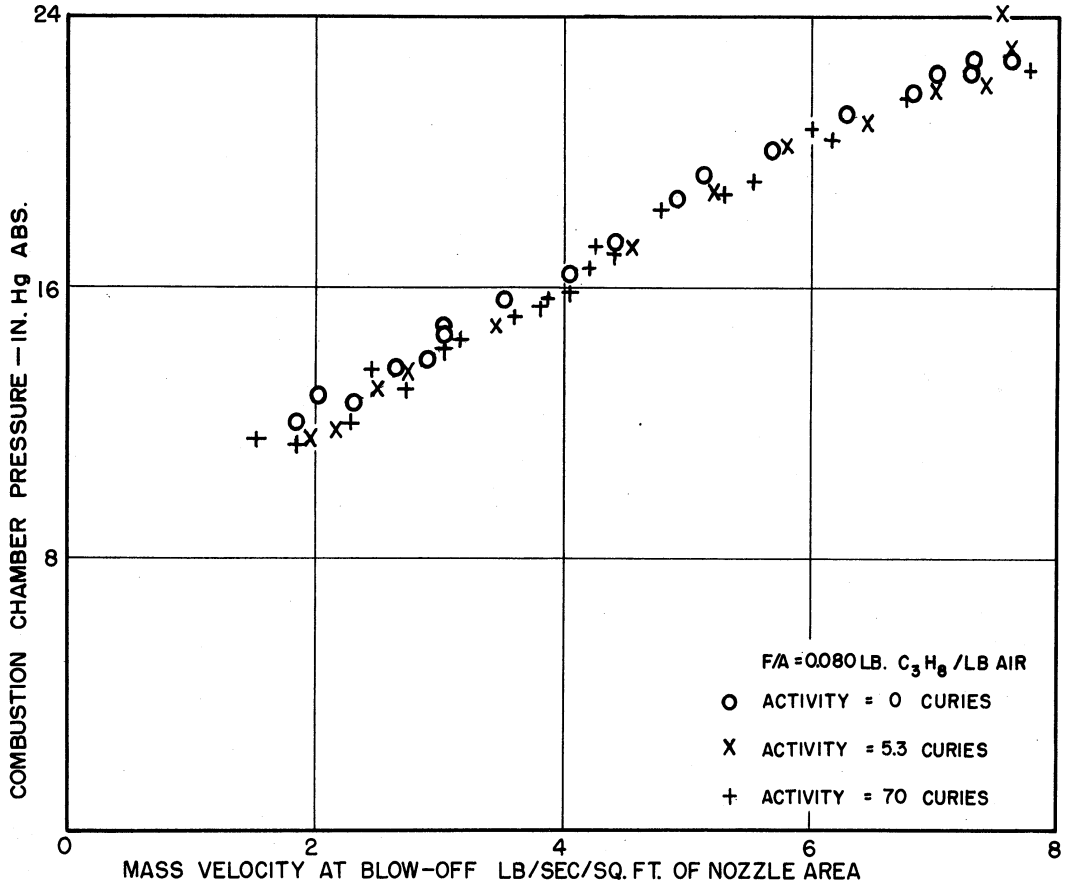

FIG. 12. FLAME STABILIZATION EXPERIMENT WITH PALLADIUM FOIL OF DIFFERENT ACTIVITIES MOUNTED ABOUT A V-TYPE FLAME STABILIZED WITH A STEEL SPHERICAL FLAME HOLDER

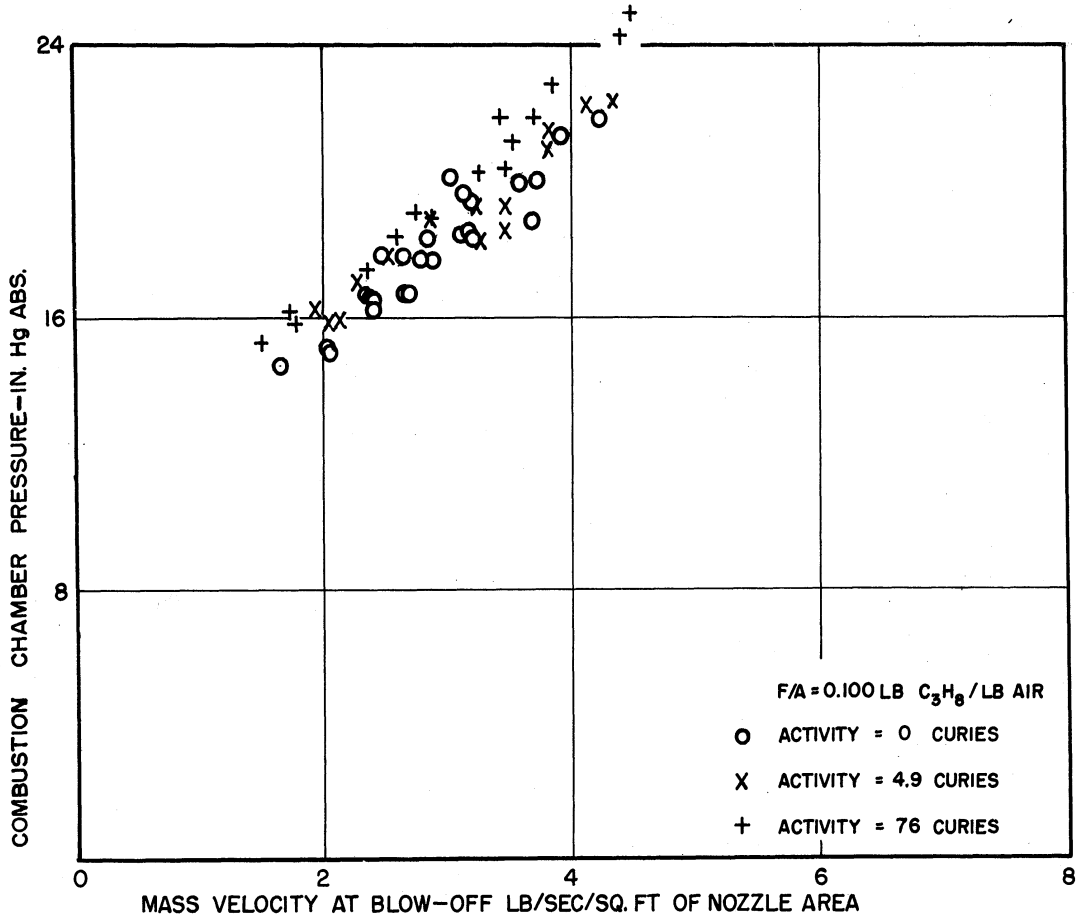

FIG. 13. FLAME STABILIZATION EXPERIMENT WITH PALLADIUM FOIL OF DIFFERENT ACTIVITIES MOUNTED ABOUT A V-TYPE FLAME STABILIZED WITH A STEEL SPHERICAL FLAME HOLDER.

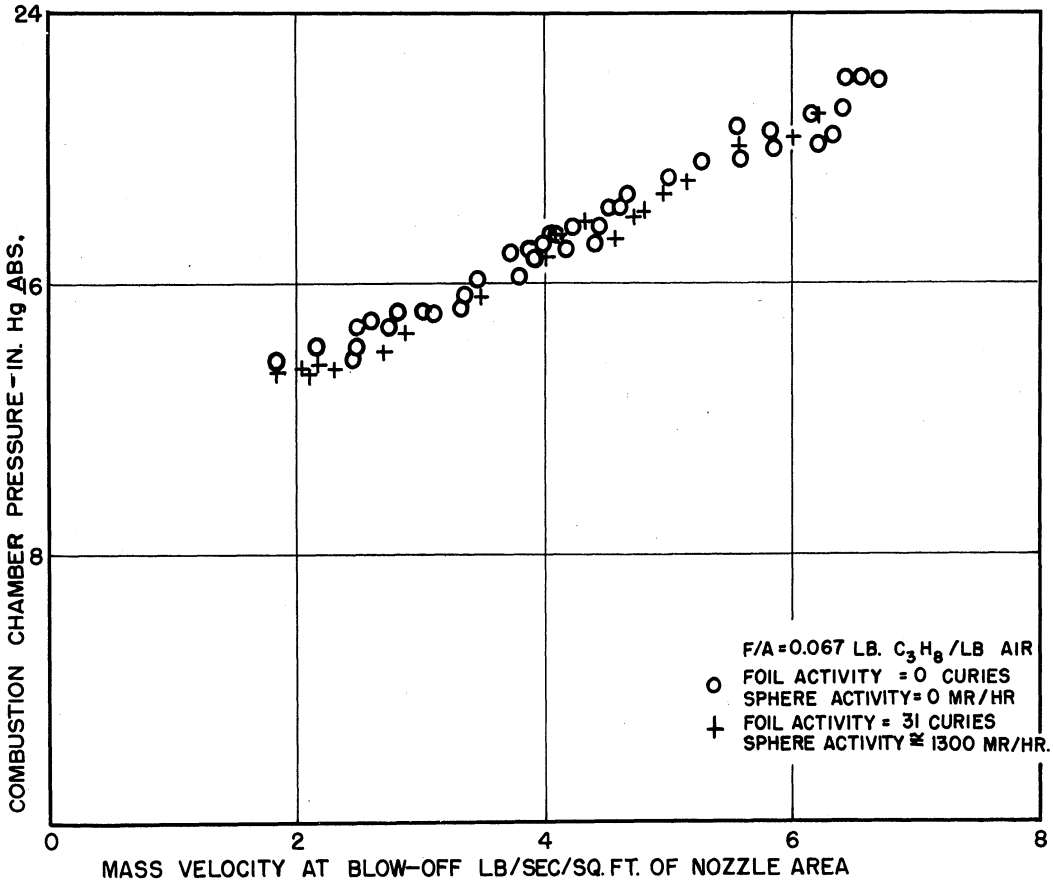

FIG. 14. FLAME STABILIZATION EXPERIMENT WITH PALLADIUM FOIL OF DIFFERENT ACTIVITIES MOUNTED ABOUT A V-TYPE FLAME STABILIZED WITH A PALLADIUM 109 SPHERICAL FLAME HOLDER.

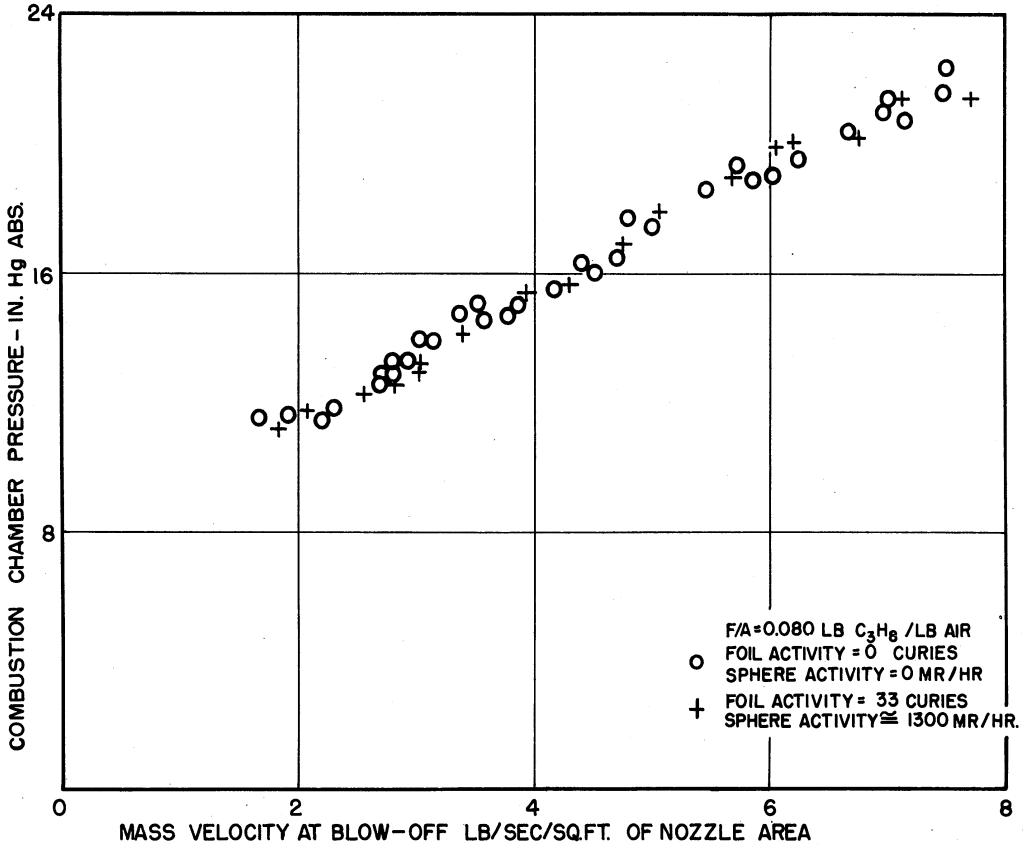

FIG. 15. FLAME STABILIZATION EXPERIMENT WITH PALLADIUM FOIL OF DIFFERENT ACTIVITIES MOUNTED ABOUT A V-TYPE FLAME STABILIZED WITH A PALLADIUM 109 SPHERICAL FLAME HOLDER

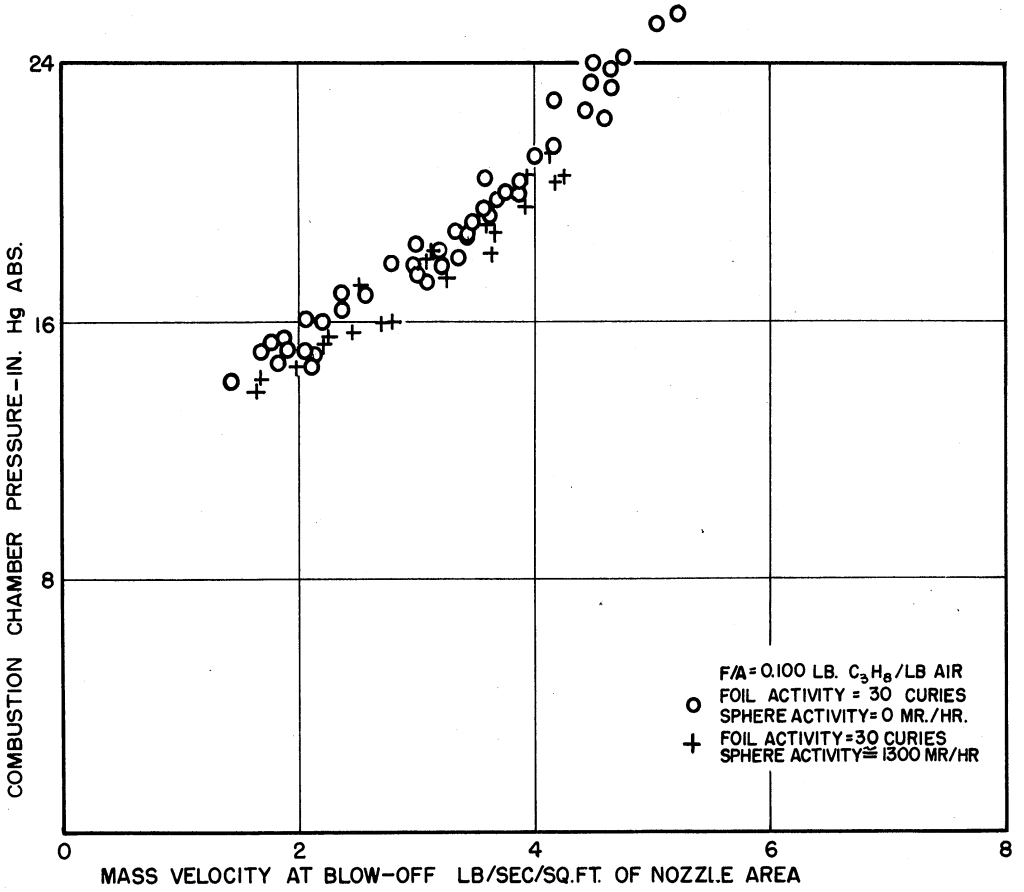

FIG. 16. FLAME STABILIZATION EXPERIMENT WITH PALLADIUM FOIL OF DIFFERENT ACTIVITIES MOUNTED ABOUT A V-TYPE FLAME STABILIZED WITH A PALLADIUM 109 SPHERICAL FLAME HOLDER.

flame-propagation rates at subatmospheric pressures in the region from 13 inches mercury absolute to about 2 inches mercury absolute. The fuel used was a mixture of 0.067 lb propane/lb air.

It can be also concluded that beta-particle radiation from a palladium foil at a distance of about 1-1/2 inches surrounding a V-type flame stabilized from a spherical steel flameholder, 1/8 inch in diameter, has no effect on the stability of a flame burning in propane-air mixtures of 0.067, 0.080 and 0.100 lb propane/lb air, with foil activities as high as 76 curies.

Concerning the experiment using a 1/8-inch diameter palladium flameholder, it can be concluded that the only possible change effected in the stability limits was a slight increase with a fuel-air mixture of 0.100 lb propane/lb air. The stability limits of flames burning from the two leaner mixtures were unaltered.

f. Future Experiments. It appears from the results of the above experiments that the only possible change effected by beta radiation was in the flame-stability experiment using the activated source in close proximity to the reaction zone, i.e., at the flameholder. Therefore, two avenues of approach are indicated:

(1) Institute a more complete series of experiments using flameholders made of palladium-109, perhaps in the form of grids to increase the total flux of radiation.

(2) Institute a series of experiments using gaseous hydrocarbon fuels that have been irradiated by a source of beta or gamma radiation placed immediately beneath the flame nozzle.

The experiments performed under (2) above would include the investigation of flame-propagation rates and flame-stability limits as previously carried out. One additional type of experiment appears to be of interest, inasmuch as the experimental equipment is available. It would include the effect of radiation on detonation velocities, detonation limits, and the susceptibility of detonation in combustible mixtures by means of shock waves of various intensities.

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

Personnel:

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Research Assistants: D. E. Harmer, J. G. Lewis, J. R. Hallman, S. A. Stolton.

A. INTRODUCTION

The objective of this work is indicated by the title of this subproject and has been discussed in earlier reports. Recently, the work has been concentrated on more nearly quantitative determinations of some effects of radiation, but studies are also being continued with the objective of finding which reactions are affected by radiation. The effects of gamma rays from the cobalt-60 source have been receiving the most attention. More work will probably be done on beta radiation in the future.

1. Styrene

In the last progress report made for this project[1], it was reported that the viscosity of samples of monomeric styrene after irradiation in the cobalt-60 gamma source continued to increase after the samples were withdrawn from the source, while unirradiated check samples remained unchanged. This effect has been investigated more fully.

Others working in this field have produced a high degree of polymerization of styrene at room temperature with radiation[2]. The concern of this project, however, has been to study the effects of radiation on the progress of the conventional thermal polymerization of styrene. It is thought that industrial applications for radioactive materials may be more quickly realized by these methods.

In original experimental runs, relative viscosity was used as a qualitative measure of the degree of polymerization of unstabilized styrene which had been heated at 125°C. Samples of styrene were given 24 hours of

radiation in the kilocurie cobalt-60 gamma source. They were then subjected to thermal polymerization at 125°C. Viscosities of these samples were measured by observing the time required for stainless-steel ball bearings to fall through a given layer of the material. The viscosities of irradiated samples were found to be two to three times as great as the viscosities of corresponding unirradiated control samples. This relationship appeared to hold over the entire range of viscosities which could be measured by the method described.

The diameters of the glass tubes containing the styrene and falling ball were somewhat variable, since ordinary Pyrex tubing was employed, so a large variation in results was obtained. However, the data were regarded as being good evidence that radiation had produced a noticeable effect on the polymerization rate. When 12 or 48 hours of radiation were given, the viscosity increase of the irradiated sample was correspondingly less or more than that of the 24-hour runs, as would be expected. Runs made on styrene stabilized with tertiary butylcatechol also showed an increase in reaction of the irradiated sample over the unirradiated control. The samples used for this particular series of experiments were irradiated and polymerized under their own vapor pressure.

Since the experiments just described indicated that the irradiation of the monomeric styrene was giving a noticeable radiocatalysis to the subsequent thermal polymerization, data were sought to determine more quantitatively the effect of the prepolymerization irradiation period on the percantage conversion at various times of thermal polymerization.

A standard method for the purification of the styrene monomer has been developed. All glassware which would be used to handle the dried monomer was first heated to remove moisture and volatile impurities. The monomer (E. K. Co. No. 1465 stabilized with tertiary butylcatechol) was then run through a 10 x 400-mm tube packed with fresh Ascarite to act as a drying agent and to remove as much as possible of the phenolic inhibitor by salt formation. The dried styrene was placed in an all-glass distillation apparatus (see Fig. 17), after which the inlet tube was sealed with a torch. A stopcock was provided for evacuation of the apparatus, and was placed so that the liquid styrene did not come in contact with it or with the stopcock grease. The system was evacuated and the stopcock was closed. The distillation was carried out by a process of evaporation from the surface of the styrene, rather than by ebullition with the accompanying danger of spray being carried over through the Vigreaux column into the receiver. It was found that such a distillation could be carried out by immersing the boiling flask in hot water, while cooling the receiver in a dry-ice, chloroform, carbon tetrachloride bath. Under these conditions the vapor came over at a temperature of 60°-75°C, and was frozen immediately in the receiver. When

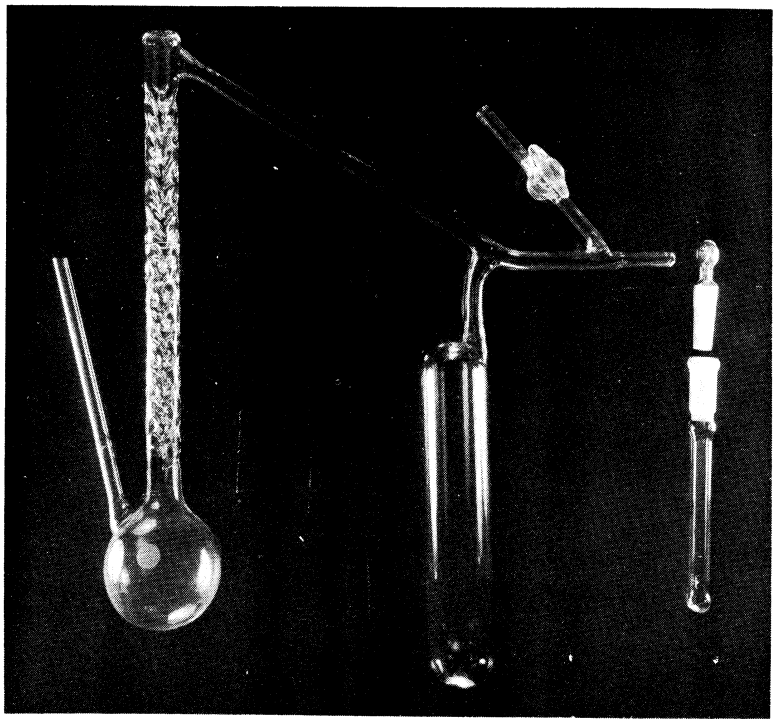

Fig. 17. Glass apparatus for vacuum distillation of styrene, and glass-stoppered tube for irradiation and polymerization of styrene.

about three-fourths of the material had been distilled, air was admitted to the system, and the frozen distillate was brought to room temperature.

The sealed ends of the glass apparatus were broken open in order to pour the purified monomer, under air, into cleaned and dried glass-stoppered sample tubes, (See Fig. 17). The design of the distillation apparatus allowed the monomer to be poured out through the exit tube without coming into contact with the vacuum stopcock, while the undistilled material was retained by the shoulder of the boiling flask.

When the tubes of samples could not be used immediately, they were stored either in ice or in a dry-ice bath to prevent appreciable polymerization. In runs made up to the time of this report, the irradiated samples were given 24 hours in the cobalt-60 gamma source, corresponding to about 1,920,000 rep in air. During the irradiation, the samples were stoppered in an air atmosphere, and were at room temperature. Polymerization of control and irradiated samples was carried out in an oil bath maintained at 125° $\pm$ 2°C.

At the end of the polymerization period, the sample tubes were cooled to room temperature in a water bath, wiped clean, and weighed. The polymer solution was poured into absolute ethyl alcohol, and the tube, with the small amount of solution that remained in the tube, was weighed again. The known weight of polymer mixture was determined by the difference in weight of the original and emptied tube. This known weight of solution was allowed to remain in the alcohol for a few minutes. The resulting precipitate was then placed in a weighed Gooch crucible and extracted in a Soxhlet extractor with absolute alcohol for about 24 hours. The crucible and polymer were then placed in a vacuum dessicator at 68-75°C for 24 hours. At the end of this procedure, the cooled crucibles were weighed again with the remaining polymer. It is felt that by this process, the significant part of the unpolymerized styrene and the absorbed alcohol was removed, so that the percentage which had polymerized could be ascertained.

Some samples were polymerized to such an extent that they could no longer be poured into the alcohol. A slight modification of the above procedure was employed for these samples. The glass sample tube was broken, so that the solidified polymer mixture could be removed. The rod of polymer mixture was then crushed, weighed, and dissolved in benzene. The benzene solution could then be poured into alcohol, and the polymer precipitated as before.

From data thus obtained for a series of different times, a graph of per cent conversion versus time of polymerization at 125°C for irradiated (24 hours in the cobalt-60 gamma source) and the unirradiated samples was drawn (Fig. 18). Examination of this plot shows that some amount of polymerization has taken place during the irradiation process. The amount which was polymerized during irradiation is somewhat greater than that obtained by other investigators[2] This difference may be attributed to the fact that the styrene used in the data for this report was polymerized in an air atmosphere. The upper line of the graph, representing the irradiated styrene, shows that such samples are always polymerized to a greater extent than the corresponding unirradiated samples. This fact is especially important in the highest percentages polymerized, since it is here that the reaction rate is the slowest. This graph does not correspond exactly to data published by the Dow Chemical Company[3], but the slightly slower rate of polymerization of the control samples may be attributed to traces of inhibiting impurities, or to a different amount of dissolved air in the samples.

2. Exploratory Investigation of Reactions Influenced by Radiation

A considerable proportion of the work of this laboratory has been devoted to the search for chemical reactions which may be promoted to an

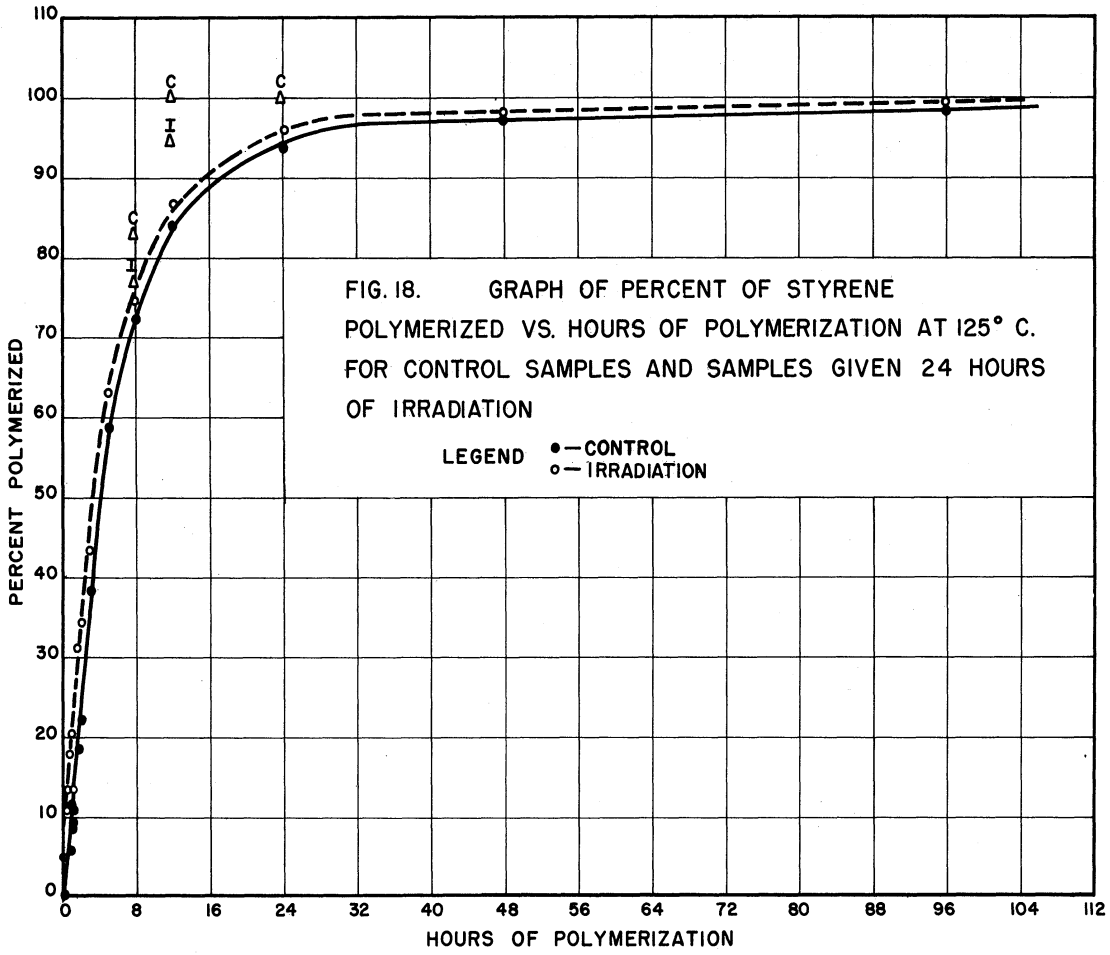

FIG. 18. GRAPH OF PERCENT OF STYRENE POLYMERIZED VS. HOURS OF POLYMERIZATION AT 125° C. FOR CONTROL SAMPLES AND SAMPLES GIVEN 24 HOURS OF IRRADIATION

industrially important extent by radiation. Several different reactions have been investigated, using macro methods of chemical analysis in attempts to detect changes produced by radiation. From the data included in this report it can be seen that some reactions have given much more promising results than have others. Below are described some experiments which were conducted in order to find easily detectable effects of radiation on chemical systems.

Acetylene was irradiated with 1.9×10^6 rep in air of cobalt-60 gamma radiation. The acetylene was dissolved in acetone which had first been absorbed in a dried portland cement, asbestos mixture. The cakes of cement were placed in a stainless-steel pressure vessel designed to hold 2000 psig and fitted with an aluminum rupture disc designed to burst at 150 psig. This assembly was known as the "bomb". (The pressure vessel is that described on p. 37 of Progress Report 21 of this laboratory.) The bomb was evacuated with the cement in place, flushed with nitrogen, and evacuated again. Then 105 grams of acetone was added and acetylene introduced until an equilibrium pressure of 5 psig was reached. The bomb thus charged was irradiated for 24 hours, after which the volatile contents of the bomb were recovered by immersing the bomb in hot water and heating the discharge pipes with infrared lamps. Subsequently, the bomb was evacuated while being heated in the manner just described. During the heating period all effluent material was passed through dry-ice traps.

The condensed liquid was distilled in a Podbielniak column, where it was observed that the overhead temperature was not significantly different from that of acetone during the distillation. About 0.2 gram of a brownish, waxy solid was recovered from the pot of the column. This material was insoluble in acetone.

Some natural oils, donated by the Wyandotte, Michigan, plant of the Archer-Daniels-Midland Company, of Minneapolis, Minnesota, were subjected to radiation by beta rays from palladium-109. Watch glasses were arranged on a wire frame. One drop of each oil to be observed was weighed on a separate watch glass. A thin polyethylene film was placed over the watch glasses, and the palladium-109 foil, encased in manila paper, was placed over the film of polyethylene. The oils were allowed to stand at ambient room temperature for one week in the apparatus described above. Control samples were run on the same oils. The control samples received no radiation, but were treated the same as the test samples in other respects. A summary of the results is given in Table I.

TABLE I

IRRADIATION OF NATURAL OILS WITH PALLADIUM-109 BETA RAYS

Material	Percentage Gain in Weight		Remarks	
	Test	Control	Test	Control
raw linseed	+7.8	+0.53	tough, rough film	no change
boiled linseed	+5.7	+7.0	tough, rough film	tough, rough film
degummed soya	+5.8	+0.39	tough, smooth film	no change
castor	+1.3	-0.58	no change	no change
refined menhaden	+9.3	+4.5	tough, smooth film	no change
cotton-seed pitch	-0.36	-0.19	no change	no change

Some of the gains in weight observed may have been caused by oxidation in the air. It is interesting to note that on the irradiated samples of raw linseed, degummed soya, and refined menhaden, hard, tough films appeared while on the unirradiated samples of these same oils, no films appeared.

A preliminary investigation was conducted on the effect of radiation on the increase in viscosity of dry, refined, degummed soya oil. Samples of the oil were placed in sealed containers and irradiated for 24 hours by gamma rays from cobalt-60. Immediately after a sample was removed from the cobalt-60 vault, it was placed in an apparatus designed to maintain the temperature and pressure of the sample at some fixed values. Nearly all runs reported were conducted at 1 mm absolute pressure. One set of runs was conducted at 575°F, and one set was conducted at 600°F. The results are tabulated in Table II, as follows:

TABLE II

CHANGES IN VISCOSITY OF SOYA OIL AFTER IRRADIATION AND SUBSEQUENT HEATING

The starting material used in each run was dry, refined, degummed soya oil.

Run No.	Irradiation	Temperature, °C	Time of Heating, Hr	Viscosity G-H	Viscosity Poises	Pressure, mm mercury, absolute
11	24 hr, Co^{60}	300	6	N+1/3	3.5	1
12	--	300	6	J	2.5	1
13	24 hr, Co^{60}	300	6	L	3.0	1
14	24 hr, Co^{60}	300	6	L	3.0	1
15	--	300	NG-air leak	--	---	>12
16	24, hr, Co^{60}	300	6	L	3.0	1
17	--	300	6.3	L	3.0	1
18	--	300	6.2	J	2.5	1
19	--	300	6	J	2.5	1
26	24 hr, Co^{60}	316	6	Z4	63.4	1
27	--	316	NG-air leak	--	----	>10
28	24 hr, Co^{60}	316	6	Z4+1/4Z5	69.4	1
29	--	316	NG-boiled over	--	----	1
30	--	316	6	Z1+2/3Z2	33.0	3
31	--	316	6	Z3+1/2Z4	54.8	3-4

The samples heated at 300°C after irradiation had viscosities of 3.0 to 3.5 poises, whereas those not irradiated but otherwise treated in the same way had viscosities of 2.5 to 3.0 poises. This difference in viscosities was approximately 20 per cent of the lower value. The samples heated at 316°C after irradiation were generally more viscous than those not irradiated but otherwise treated in the same manner. Some relatively mobile liquid distilled off the samples tested at 316°C. Because of the difficulties experienced in controlling the material balances at this temperature, further work under these conditions is indicated and planned.

Equipment has been constructed and is now being tested for the study of the kinetics of the ammonia synthesis at approximately 930°F. These tests will be conducted both in the presence and in the absence of gamma radiation from cobalt-60. Construction is also proceeding on equipment to test gaseous reactions in the presence of cobalt-60 gamma radiation at 2000 psig and temperatures up to 930°F. It is expected that operation at conditions of temperature and pressure approaching those used in the industrial processes for the manufacture of ammonia and methanol may provide useful additional information regarding the influence of radiation on these reactions.

B. FUTURE WORK

1. Experimental Studies

Future work being planned with styrene includes investigations of the variables of polymerization temperature, irradiation time, and length of time elapsed between irradiation and thermal polymerization. Apparatus has now been constructed to permit thermal polymerization and irradiation of the styrene simultaneously in the gamma source. With this new arrangement, it may be possible to ascertain whether the effects of temperature and of radiation on this reaction may be combined to give a reaction rate greater than the combined individual effects, or whether the two effects will operate independently.

The use of impure samples of styrene, in which an induction period for the reaction is to be expected, may yield information about the usefulness of radiation to shorten this induction period. When sufficient data have been obtained in the case of styrene, those effects which appear to increase the rate of reaction will be investigated in other vinyl systems, and for other olefins.

It is planned to conduct additional tests on the oxidation and/or polymerization of natural oils under the influence of gamma radiation. More information regarding the changes taking place could be secured by subjecting

the treated samples to some of the tests ordinarily used in testing natural oils, such as iodine number, unsaponifiable material, free fatty acids, and melting point.

The work on the kinetics of gaseous reactions at high temperatures will be continued. The influence of radiation will be studied in the presence of catalysts ordinarily used for the reactions in question.

2. Need for Modified Gamma Source

The available space within the shielding of the present source of gamma radiation is so limited that the introduction of chemical equipment into the radiation field is severely handicapped. The demand for use of the present source is great, and only one small sample at a time can be accommodated. As a consequence, the work on chemical reactions is retarded, particularly by the necessity of designing and constructing highly specialized equipment needed to conduct the usual operations of chemistry within a space 1-1/2 inches in diameter by 13 inches long. If the working space within the shielding of the present source were to be increased, the flux density in the resulting volume would be reduced materially below the level now available.

The productive effort of this laboratory would be measurably increased if more working space were available. However, in order to maintain the intensity of gamma radiation in the working space at some value approximating that now available, a source of greater intensity would be needed. Such an arrangement would permit more frequent scheduling of experiments, each of which would require less time to prepare. In addition, experiments needed to explore the maximum effects of radiation upon given reactions could be scheduled for the long, continuous exposures required without interfering with other experiments.

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PART III. SUBPROJECT M943-D, IRRADIATION OF BIOLOGICAL MATERIALS

Personnel:

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A. INTRODUCTION

The biological materials that have been irradiated and studied include foods and food fractions; various systems of microorganisms such as molds, yeast and bacteria; pharmaceutical products such as antibotics, hormones and vitamins; and biological products such as various blood fractions.

B. IRRADIATION OF FOODS

The development of a process which would utilize the fission products to sterilize foods by gamma radiation involves many problems. First it is necessary to demonstrate that food can be sterilized and preserved by a radiation treatment. Progress Reports 1 and 2 of Project M943 presented data which showed that food can be sterilized by gamma radiation. Various systems of microorganisms such as molds, yeasts, bacteria, etc., were completely destroyed by radiation. Additional systems will be investigated. How long food may be preserved after sterilization depends to a great extent upon other factors such as method of packaging, temperature of storage, absence or presence of oxygen and undesirable enzymes, the tendency of the food to undergo chemical reactions, etc. These problems are quite complex and must be considered separately for different types of foods. In addition to sterilization and preservation, other important factors must be considered such as food value, loss in vitamins, the possibility of producing oxic compounds, and undesirable flavor and color changes.

Animal feeding experiments will be required to establish food values and to assure that no toxic compounds are produced. Some preliminary tests along these lines are reported in Progress Report 2. However, the present 1000-curie cobalt-60 source is inadequate for this purpose and further tests of this nature will necessarily be postponed until a larger source having

sufficient capacity to irradiate adequate food for significant animal feeding experiments is available. Although irradiation of food no doubt results in some loss of vitamins, the preliminary experiments indicated that irradiated food supports normal growth without any observed undesirable effects. It must be emphasized that these tests were only of preliminary nature and are not conclusive.

The research since the writing of Progress Report 2 has been devoted primarily to the investigation of flavor changes in irradiated food, since it has already been fairly well established that foods can be sterilized and since the feeding tests must be postponed.

1. Flavor Changes in Irradiated Foods

a. Foods with Little or No Flavor Change. In the following tests, in which flavor was the primary subject of study, it should be pointed out that usually the sample was tested soon after irradiation so as to distinguish flavor changes caused by radiation from flavor changes developed in storage. Therefore, in these tests very little data were obtained on the storage characteristics.

Some foods can be irradiated without the development of any "off" flavors. In general, green vegetables are free from flavor changes.

1). Green asparagus: Several samples of fresh green asparagus stored under vacuum and at room temperature have been irradiated for 24 hours (equivalent to an air dose of about 1.9×10^6 rep). Taste tests run on samples stored at room temperature for periods of from one to seven days showed no loss in color, flavor, and texture. The taste could not be differentiated from that of fresh asparagus. The cooking of the asparagus, as in the case of all other vegetables, was accomplished by boiling in lightly salted water until tender.

2). Fresh spinach: Spinach that was irradiated under vacuum in a test tube for 24 hours (1.9×10^6 rep in air) and tasted immediately after irradiation had no off flavor or color. The taste could not be differentiated from that of a sample of unirradiated, fresh, cooked spinach. A sample irradiated under vacuum and then stored for 18 days at room temperature wilted during the storage period. An unirradiated control stored under the same conditions developed a green mold and lost fluid on storage.

3). Green peas: Samples of both fresh and frozen green peas have been irradiated. The frozen peas show both color change (as bleaching) and structural degradation (as softening after irradiation). The color change was less pronounced in an oxygen-free atmosphere, but the structural changes still

continued. However, peas irradiated in the jacket showed no color or structural changes after 24 hours of irradiation (1.9×10^6 rep in air). Chronologically, this was the first experiment in which the color change of green peas was inhibited. An extract of peapods in water was prepared and a sample of peas was soaked in the extract before it was irradiated. Examination of these peas immediately on removal from the source showed no color change. The peas were then cooked and tasted. The taste was the same as that of fresh peas. Since the peapods have a high chlorophyll content, it was decided to determine whether or not a solution of chlorophyllin A would produce the same effects as the peapod extract. Results indicated that this was the case. Several more samples have been treated in this manner, cooked, and tasted. The color, flavor, and texture were not different from those of fresh peas. Storage for more than several days has not been investigated.

4). Green beans: Green beans soaked in chlorophyllin A, packaged in evacuated test tubes, and then irradiated for 24 hours (1.9×10^6 rep in air) do not decompose or extensively decolorize. Samples treated in an identical manner but not irradiated developed a growth of mold and decomposed after 72 hours of storage. When the irradiated sample was opened, no strong of "off" odors were observed. The flavor of the cooked samples was not identical to that of fresh beans, but approximated that of frozen beans. Experiments made on several successive days with the same initial batch of beans showed a definite degradation of flavor as the age of the bean prior to radiation increased. One sample stored for 22 days and then tasted was free from "off" flavors.

5). Green broccoli: Samples of fresh and frozen broccoli have been irradiated for 24 hours (1.9×10^6 rep in air) with cobalt-60 gamma radiation. As reported in Progress Report 2, broccoli which has been irradiated while exposed to air will bleach to a yellow-green color. Evacuation of the sample coupled with blanching does not inhibit this change to any great extent. Commercial frozen broccoli, packaged in evacuated test tubes, bleached to a yellowish color when irradiated for 24 hours (1.9×10^6 rep in air). The irradiated product had a sweet odor and a good taste; however, the flowers of the irradiated samples were not as firm as those of the control. The samples were tasted immediately after irradiation. Fresh samples of broccoli were soaked in a dilute solution of chlorophyllin A (approximately 20 mg/ml). The color was bright green and the texture in all cases was firm. The taste was good, although not as good as the control. Except for one case where the texture had degraded and the flavor was poor, the results were reproducible.

6). Fresh carrots: Fresh whole carrots were irradiated under vacuum for 24 hours (1.9×10^6 rep in air) with cobalt-60 gamma rays, and were preserved with little or no change in color, flavor, or texture. Several observers tasted irradiated carrots after cooking and reported their flavor to be sweeter than that of a fresh control. Samples stored for 76 days at room temperature

showed a slight bleaching on opening. The taste and texture, however, were not impaired. Control samples stored for more than 10 days developed a white mold and lost fluid.

7). Bacon and ham: Samples of commercially prepared bacon were irradiated for 24 hours (1.9×10^6 rep in air). Immediately after irradiation, a sample was fried and tasted. The taste could not be distinguished from a freshly fried sample of unirradiated bacon. Samples stored at room temperature for 14 days also showed no change in flavor. The temperature during storage was sufficiently high to melt a portion of the bacon fat. Similar results have been obtained with smoked ham. The maximum storage time used with irradiated ham was 6 days. The flavor on grilling was slightly different from a fresh sample cooked as a control, but was still completely palatable and tasty.

b. Foods Undergoing Flavor Change. 1). Foods of animal origin: Foods of animal origin appear to be particularly sensitive to flavor change. Milk, fresh meats, eggs, cheese, etc., all undergo undesirable flavor change as the result of irradiation. This flavor change seems to be of two general types: (1) proteins develop an animal or "goaty" odor and flavor, and (2) fats become rancid. Various chemical additives have been used in an attempt to prevent these undesirable flavor changes, with some success.

a). Milk and milk products: Attempts to control the flavor change in irradiated milk by means of chemical additives have not been successful. The materials tried were sodium nitrite, carotene, ascorbic acid, thiourea, and hydroquinone. In all cases the milk tasted had an "off" flavor. Samples were prepared from both whole and skim milk. The skim milk was centrifuged to remove all cream. After irradiation for 24 hours (1.9×10^6 rep in air), it had a "goaty" off odor and flavor somewhat similar to that of irradiated fresh meats. Cream removed from milk was irradiated and the product had a rancid odor and taste similar to oxidized fat. Fresh creamery butter gave the same result. Cottage cheese was irradiated for 24 hours and the irradiated product had a very pronounced "off" odor and flavor of a "goaty" nature similar to the irradiated skim milk. The flavor and odor were so poor that the product could not be considered edible.

b). Fresh meats: A variety of fresh meats, including beef, chicken, and pork, were irradiated and checked for flavor. All developed a characteristic "off" odor and flavor as a result of radiation. Chemical additives were used to prevent this flavor change, with some success.

Chicken breasts were treated with thiourea and with ascorbic acid by soaking the meat in a dilute solution (approximately 10 mg/ml) and were irradiated for 24 hours (1.9×10^6 rep in air). The samples were opened immediately on removal from the radiation source, washed with water to remove some of the preservatives, and boiled. An unirradiated sample was boiled as a control.

When tasted, the sample treated with thiourea could not be distinguished from the fresh control. The sample treated with ascorbic acid had a sharp flavor that was definitely inferior. Several repeat runs verified these results. Samples treated with thiourea and stored at room temperature for six days and then cooked still maintained good flavor. It is realized that thiourea and some of the other chemicals used are toxic to varying degrees and are not approved by the Food and Drug Administration. It was reasoned however, that data obtained with these materials might lead to the development of more suitable chemicals.

Ground and sliced beef also was treated with thiourea and with ascorbic acid solutions prior to radiation in the same manner as in the experiments with chicken breasts. The results were similar to those obtained with chicken. Beef receiving no chemical treatment had a very poor taste after irradiation and the samples treated with ascorbic acid gave about the same results. The sample treated with dilute thiourea solution, however, had a very good taste after 24 hours irradiation (1.9×10^6 rep in air). The irradiated, treated sample was tasted both raw and after cooking and both samples were free of the characteristic animal "off" flavor found in irradiated untreated meats.

2). Produce: a). Cauliflower: Samples of raw cauliflower were packaged in glass tubes, evacuated, and irradiated for 24 hours (1.9×10^6 rep in air). The container was opened shortly after irradiation and the sample had a pungent sour odor. The texture appeared to be the same. Upon cooking the pungent sour odor was replaced by the normal odor of cooking cauliflower. The sample was palatable but definitely of poorer quality than the control.

b). Radishes: Irradiation of radishes for 24 hours (1.9×10^6 rep in air) resulted in some bleaching of the samples and a loss in crispness. The odor and flavor of the irradiated samples were of poor quality and seemed characteristic of old or partially spoiled radishes.

c). Orange juice: A variety of attempts were made to preserve orange juice without flavor change. Orange juice was irradiated while frozen, evacuated, and under atmospheres of nitrogen and also carbon dioxide. Various chemical additives were used, including ascorbic acid, carotene, hydroquinone, thiourea, and formic acid. In no case was it possible to prevent flavor change. The flavor of the irradiated juice was not unpleasant, but it did not have the fragrance of fresh juice. Irradiated orange juice tastes somewhat like cooked orange juice. The irradiated juice is sterile after about 2×10^6 rep and has been kept without refrigeration for more than sixty days.

A lyophilized pure orange juice product in pellet form (prepared by the Dry-Freeze Corporation of Chicago, Illinois) was irradiated and then reconstituted. Irradiation periods of not more than 4 hours (320,000 rep) yield a product

which is not different from the control. Greater dosages of radiation result in a flavor different from that of the control.

2. Discussion of Results and Conclusions

Some foods such as green vegetables, spinach, asparagus, green peas, broccoli, and others, including carrots, can be sterilized by gamma radiation without undesirable flavor changes. It was found that fresh meat could be protected from flavor changes by using small amounts of protective chemicals. Work in this direction has been continued with support by Michigan Memorial-Phoenix Project 41 and since the writing of the main portion of this report, the personnel of Phoenix Project 41 have found it possible to prevent these flavor changes with small amounts of additives that would meet the approval of the Food and Drug Administration. Therefore, fresh meat can probably be added to the list of foods that can be sterilized by gamma radiation without undesirable flavor changes. If animal feeding experiments establish that food sterilized by gamma radiation will meet the requirements of FDA in all respects as food for human consumption, a commercial method of processing these foods by gamma radiation could be developed providing suitable gamma sources were available.

The three principal steps that must be taken before food can be preserved by gamma radiation on a commercial basis are: (1) food sterilized by gamma radiation must be proven acceptable to FDA and to consumer tastes; (2) industry must show interest by supporting research work to develop the process in terms of commercial feasibility; and (3) the AEC must make suitable fission product gamma-ray sources available in sufficient quantities and at reasonable costs.

3. Future Experiments

Project M943-D is inactive as of July 1, 1952. The work in this field, however, will be continued with the support of the Michigan Memorial-Phoenix Project and whatever additional industrial grants and contracts that can be obtained. The Michigan Memorial-Phoenix Project will conduct animal feeding experiments with counsel from FDA.

C. COOPERATIVE RESEARCH WITH MICHIGAN MEMORIAL-PHOENIX PROJECTS 20 and 41

Engineering Research Institute Project M943 and Phoenix Projects 20 and 41 have a mutual interest in the effects of gamma radiation on microorganisms, enzymes and other biological factors. The Engineering Research Insititute project is attempting to find industrial uses of the radioactive fission products, while the Phoenix project is interested in the humanitarian uses of atomic energy.

Phoenix Project 20 and the experiments in bacteriology of Phoenix Project 41 are supervised by Dr. C. A. Lawrence, Assistant Professor of Bacteriology in the Medical School, who is assisted by John Graikoski. Dr. H. B. Lewis, Chairman of the Department of Biological Chemistry, will supervise the animal feeding experiments of Phoenix Project 41, and Dr. L. E. Brownell, Associate Professor of Chemical Engineering, will supervise the experiments on food preservation of Phoenix Project 41. Since Phoenix Projects 20 and 41 have 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. Radiations are performed by Engineering Research Institute Project M943, and bacteriological tests are performed by Phoenix Projects 20 and 41. The results of these tests are to be made available to both Engineering Research and Phoenix. The results reported here are a continuation of the work reported on Progress Report 21 (COO-90, January, 1952).

1. Experiment No. I. The Effects of Concentration, Cystine and Sodium Pyruvate, on Enzymatic Activity Following Gamma Irradiation (Cobalt-60)

The following set of experiments demonstrates the protection offered by alteration of conditions of irradiation.

In one case various concentrations of enzymes were irradiated. In those instances where extreme concentrations were used the enzyme solution had to be diluted after irradiation in order to conduct the assay. Conditions of irradiation and assay procedure are as outlined previously.

In testing the effect in the presence of cystine and sodium pyruvate, one per cent solutions were mixed with an equal quantity of enzyme solution, giving a final concentration of 0.5 per cent of the chemical during irradiation. Concentration of enzymes used during irradiation is as indicated for the individual experiments.

TABLE III

PROTECTIVE EFFECT OF ENZYME CONCENTRATION, CYSTINE AND SODIUM PYRUVATE, ON PEPSIN FOLLOWING IRRADIATION WITH COBALT-60

Time of Irradiation	Dose (rep in air)	MGM/ml of Tyrosine		
		H_2O	0.5% Sodium Pyruvate	0.5% Cystine
0	0	0.30	0.3	0.30
1	80,000	0.181	0.25	0.30
2	160,000	0.105	0.181	0.30
3	240,000	0.016	0.090	0.285
4	320,000	0.00	0.044	0.256

conc. pepsin = 0.25 mgm/ml
pH of solution = 1.1
4 ml irradiated in 10 x 70-mm test tubes
Temp.: 25°-26°C

TABLE IV

PROTECTIVE EFFECT OF ENZYME CONCENTRATION ON PEPSIN FOLLOWING IRRADIATION WITH COBALT-60

Irradiation Time (hrs)	Dose (rep in air)	MGM/ml of Tyrosine		
		10 mgm/ml	1 mgm/ml	.01 mgm/ml
0	0	0.30	0.158	.012
1	80,000	0.30	0.095	0
2	160,000	0.30	0.085	0
3	240,000	0.30	0.58	0

pH of solution = 1.1
4 ml in 10 x 70-mm tubes - irradiated
Temp. 25°-26°C

TABLE V

PROTECTIVE EFFECT OF ENZYME CONCENTRATION ON TRYPSIN DURING IRRADIATION

Time (hrs)	Dose (rep in air)	MGM/ml of Tyrosine		
		50 mgm/ml	0.5 mgm/ml	0.05 mgm/ml
0	0	.167	.275	.105
1	80,000	.147	.256	.005
2	160,000	.160	.14	.005
3	240,000	.153	.04	.003
4	320,000	.147	.015	.003

		20 mgm/ml	10 mgm/ml	1 mgm/ml	0.1 mgm/ml
0	0	.254	.30	.275	.230
1/2	40,000	.254	.263	.137	----
1	80,000	.254	.256	.048	.141
2	160,000	.254	.285	.005	.005
3	240,000	.254	.285	.005	.004

4 ml irradiated per tube 10 x 75 mm
pH 2.4

TABLE VI

PROTECTIVE EFFECT OF SODIUM PYRUVATE
ON TRYPTIC ACTIVITY DURING IRRADIATION

Time (hrs)	Dose (rep in air)	MGM/ml of Tyrosine	
		H_2O	Na Pyruvate 0.5%
0	0	0.23	0.30
1/2	40,000	0.185	0.256
1	80,000	0.167	0.212
2	160,000	0.019	0.212
3	240,000	0.01	0.212
4	320,000	0.01	0.212

conc. enzyme = 0.2 mgm/ml
pH 2.54

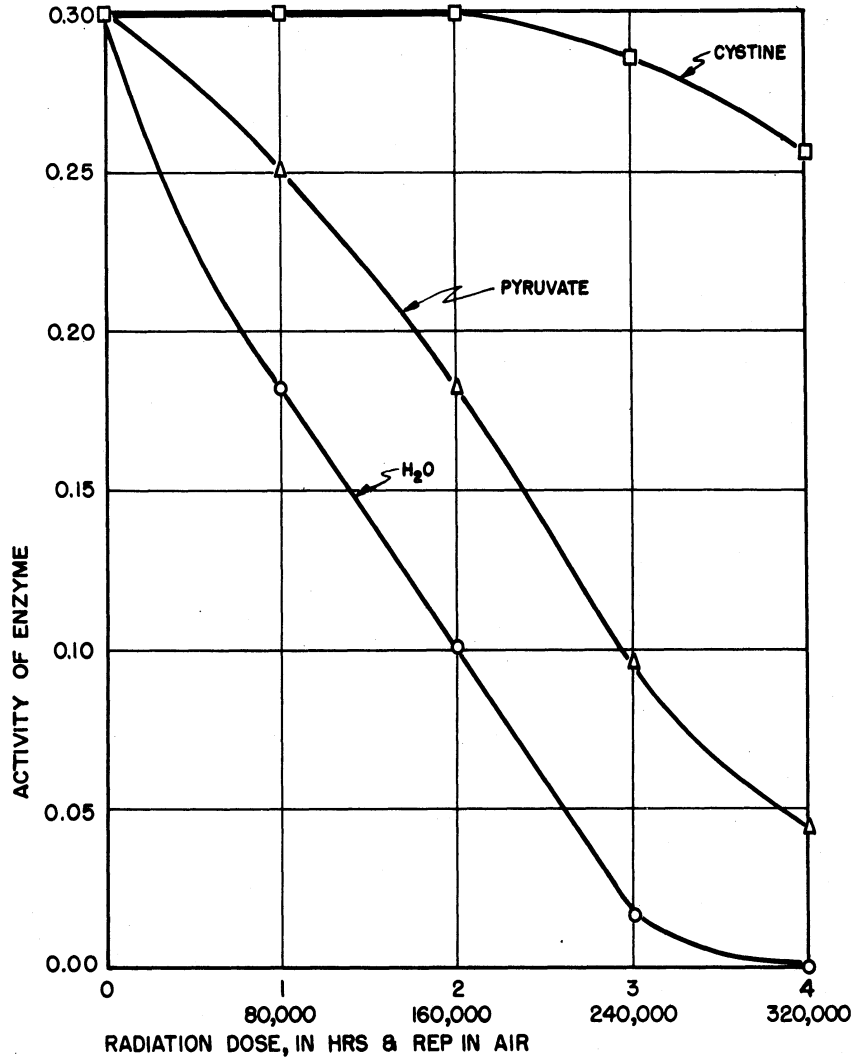


FIG. 19. PROTECTIVE EFFECT OF *l*-CYSTINE AND SODIUM PYRUVATE ON PEPSIN DURING IRRADIATION.

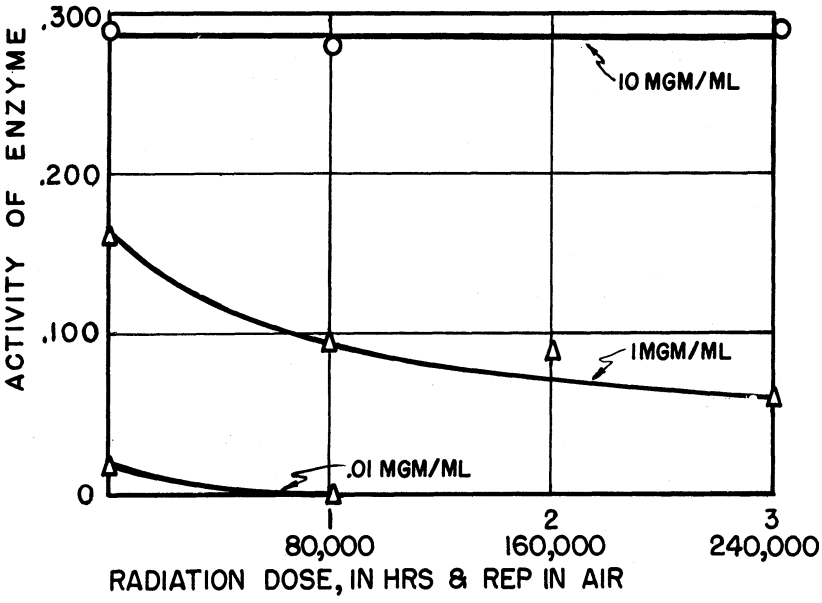
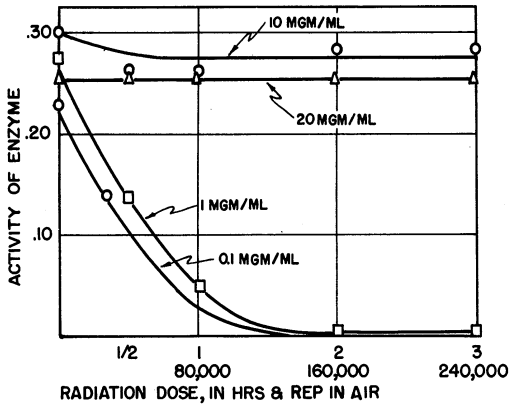


FIG. 20. EFFECT OF ENZYME CONCENTRATION ON PEPTIC ACTIVITY FOLLOWING IRRADIATION WITH GAMMA RAYS FROM COBALT-60



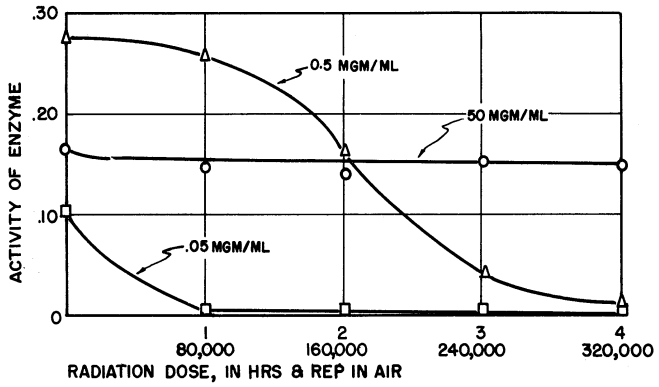


FIG. 21. EFFECT OF CONCENTRATION ON ACTIVITY OF TRYPSIN

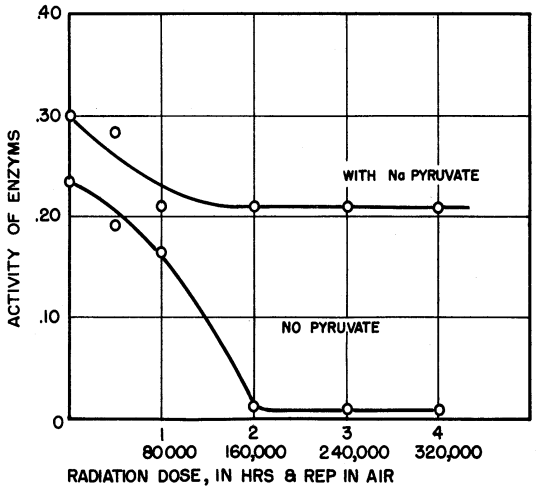


FIG. 22. PROTECTION EFFECT OF 0.5 PER CENT SODIUM PYRUVATE ON TRYPSIN DURING IRRADIATION

2. Experiment No. II. The Effect of Gamma Radiation (Cobalt-60) on the Phosphatase Enzyme Activity of Raw Milk.

Phosphatase, an enzyme present in raw milk, is used as an index of pasteurization because it is readily inactivated by heat. In order to test what effect, if any, gamma radiation from cobalt-60 has on phosphatase activity, raw milk obtained from a local dairy was irradiated for various time intervals.

Phosphatase activity was measured by its action on disodium phenyl phosphate. The amount of phenol liberated was measured photometrically.

Procedure.[2] The buffer-substrate was made by dissolving 1.09 gm of disodium phenyl phosphate and 11.54 gm of sodium veronal in 1 liter of distilled water, saturated with chloroform. This substrate was kept at 5°C when not in use. For the test, 10 ml of substrate was added to 16 x 120-mm test tubes and one drop of chloroform was added to keep down bacterial growth. To each of duplicate tubes, 0.5 ml of raw milk was added, after which the tubes were incubated at 37.5°C for more than 18 hours but less than 24 hours. They were then removed and cooled to room temperature. To the tubes was added 4.5 ml of Folin Ciocalteau reagent diluted 1:2 and allowed to stand for three minutes. The suspension was filtered using a No. 3 Whatman filter. To 5-ml amounts of filtrate was added 1 ml of 14 per cent sodium carbonate and the tubes were then placed in a boiling-water bath for 15 minutes. After cooling and filtering, the color which developed was read in a photoelectric colorimeter (Lumetron-Mod. 400-G) using a No. 630 filter.

Concentration of phenol was determined from a standard curve prepared previously, using known concentrations of phenol, the results being expressed as mgm/ml of phenol.

Irradiation. For irradiation, 2 ml of milk was suspended in 10 x 95-mm tubes and exposed for the various time intervals indicated. Samples were irradiated at room temperature. A control was maintained at room temperature during irradiation of the samples.

For dilutions, raw milk was mixed with indicated amounts of sterile distilled water and 10 ml were irradiated in 60 x 150-mm test tubes for 19 hours. The results of this study are presented in Tables VII and VIII.

Results. From the data given in the tables, it will be noted that the phosphatase activity in raw milk is not affected to any appreciable extent even after 24 hours (1,920,000 rep in air) of radiation. Diluting milk out to 1:1000 did not increase the sensitivity of phosphatase to radiation.

TABLE VII

PHOSPHATASE ACTIVITY IN RAW MILK AFTER IRRADIATION WITH COBALT-60

Irradiation Time, hrs	Radiation Dose, (rep in air)	Phenol, mgm/ml
0	0	0.55
1.75	140,000	0.55
4.50	360,000	0.48
11.25	900,000	0.51
24.0	1,920,000	0.43

TABLE VIII

PHOSPHATASE ACTIVITY IN VARIOUS DILUTIONS OF MILK AFTER IRRADIATION WITH COBALT-60

Sample* No.	Dose (rep in air)	Phenol, mgm/ml Dilutions of Milk** 0	1:10	1:100	1:1000
Control 1	0	0.354	0.364	0.134	0.059
Control 2	0	0.398	0.374	0.148	0.059
Irradiated 1	1,920,000	0.484	0.364	0.148	0.050
Irradiated 2	1,920,000	0.498	0.364	0.096	0.059

* 10 ml amounts irradiated
**Diluted with sterile distilled water

3. The Effect of Gamma Radiation on Various Microorganisms

a. Experiment No. III. Effect of Gamma Radiation (Cobalt-60) on Trichomonas Foetus. For this experiment 1-ml amounts of a 4-day culture of Trichomonas foetus, suspended in the original fluid part of Schneider's media, were irradiated in 10 x 75-mm tubes for the indicated time intervals. Motility and numbers were ascertained by "hanging drop" preparations before and after irradiation. After irradiation, 0.1 of ml was inoculated into duplicate tubes of Schneider's medium and checked for growth after 3-4 days at 37.5°C by "hanging drop". All irradiations were carried out at room temperature. An appropriate control which did not receive any radiation but which was treated in the same manner as the irradiated samples was also used. The results of this study are given in Table IX.

Results: From the data given in the table it will be noted that growth of the Trichomonas is prevented in 5 hours (400,000 rep in air) but not in 3 hours (240,000 rep in air) of irradiation. Another experiment indicated that growth is prevented in 4 hours (320,000 rep in air) but not in 3 hours (240,000 rep in air). Motility is not immediately affected by irradiation except in the higher doses. The cells move more slowly and there is evidence of cell destruction. No attempt was made to estimate the number of cells which were inactivated or destroyed.

TABLE IX

EFFECT OF GAMMA RADIATION (COBALT-60) ON TRICHOMONAS FOETUS

Irradiation Time (hrs)	Radiation Dose (rep in air)	Motility		
		Before Irradiation	After Irradiation	Growth 4 days, 37.5°C
0 (control)	0	+	+	+
1	80,000	+	+	+
3	240,000	+	+	+
5	400,000	+	+	0
7	560,000	+	+	0
Time (Min)				
0 (Control)	0	+	+	+
15	20,000	+	+	+
30	40,000	+	+	+
60	80,000	+	+	+
Time (hrs)				
0(Control)	0	+	+	+
1	80,000	+	+	+
4	320,000	+	+	n.g.
5	400,000	+	+	(+) in one (0) in one

b. Experiment No. IV. The Effect of Gamma Radiation (Cobalt-60) on Viruses*. Several previous studies have indicated the effects of radiation on viruses. Hodes, Webster, and Lavin[3] were able to inactivate rabies virus with ultraviolet radiation and yet retain its immunizing properties more effectively then chloroform- or phenol-treated virus. Huber and his associates[4,5] reported on the effects of high-intensity electrons on a number of viruses and also demonstrated that an effective immunizing rabies virus could be prepared by the use of the Capacitron. Their studies indicated that this form of radiation was generally more effective in destroying the infectivity of several viruses than were radiations from beta, gamma, or x-rays. In general, there is agreement that viruses are more resistant to the lethal effects of radiations than are bacteria, mold, yeasts, and protozoa.

Experimental: Psittacosis (strain 6BC) and Human pneumonitis (strain SF) viruses were selected for the present study. A freshly harvested embryonic chick allantoic fluid preparation of each virus, visibly free of particulate matter, was diluted 1:2 with sterile physiological saline solution and divided into two equal portions. One portion of the latter was exposed to gamma radiation emitted from the cobalt-60 source. The other half served as the control and was held at room temperature, equivalent to that in the cobalt vault. All the virus material was maintained in cotton-plugged, pyrex test tubes. At various time intervals, aliquot amounts of the irradiated, as well as the control, virus suspensions were removed from the tubes and stored immediately at -46°C until tests for inactivation were undertaken.

Tests for complete inactivation of virus and infectivity titration were accomplished by intracerebral inoculation of 10 to 14-gram albino mice. Six to ten mice were used in testing each specimen of irradiated virus. Infectivity titrations were performed by preparing a series of tenfold dilutions of virus in veal infusion broth, six mice being inoculated with each dilution of virus. After the mice were inoculate, they were observed for signs and symptoms of virus infection, time of deaths, and survivals in the irradiated and unirradiated control series. In the latter group the animals invariably died within a period of 5 days following inoculation. The animals surviving the experiment were observed for a period of 10 to 12 days before being discarded. Table X presents the results of this study.

Results: From the data given in the table it will be noted that both viruses were completely inactivated after exposure to cobalt-60 for 3 hours or longer. Exposure of the suspensions for 1 hour was insufficient to destroy the same agents. That the inactivation obtained at 3 and 6 hours was due to the specific action of the gamma radiation and was not the result of thermo

* This study was carried out in collaboration with R. D. Francis, graduate student in the Department of Bacteriology of the Medical School.

inactivation is indicated by the fact that there was no decrease in the LD_{50} titers of the control specimens, which were also maintained at room temperature during the maximum exposure period employed.

Although the present work is only of a preliminary nature, it serves to indicate that the virus preparations employed here can be inactivated in a relatively short period of exposure to gamma rays emitted by cobalt-60. Further study will be required to determine if this method of inactivation alters the antigenicity of the viruses under consideration.

TABLE X

EFFECT OF GAMMA RADIATION (COBALT-60) ON
PSITTACOSIS AND HUMAN PNEUMONITIS VIRUSES

Irradiation Time (hrs)	Radiation Dose (rep in air)	Psittacosis		Human Pneumonitis	
		Irradiated	Control LD^*_{50}	Irradiated	Control LD^*_{50}
0	0		$10^{-6.5}$		$10^{-5.0}$
1	80,000	Active		Active	
3	240,000	Inactive		Inactive	
6	480,000	Inactive	$10^{-6.6}$	Inactive	$10^{-5.2}$

*LD_{50} = Dilution which will give 50 per cent mortality in animals tested.

c. Experiment No. V. The Effect of Gamma Radiation (Cobalt-60) on a Virus of Bacteria (Staphylococcus Bacteriophage). Much of the work on the effects of radiation on bacteriophage (bacterial viruses) has been concerned with the action of radiation on mutagenic or gene changes in the genes[6,7,8]. Brasch and Huber[9] found that by exposing bacteriophage to the Capacitron, these viruses were destroyed by lower dosages than that required for the destruction or "sterilization" of animal viruses. In the present investigation, a broth culture of a Staphylococcus aureus bacteriophage was exposed to gamma radiation from cobalt-60 for varying periods of time to determine the lethal effects of the latter on this particular agent.

Experimental: The bacterial-free broth culture of phage was distributed in 2-ml amounts in several cotton-plugged, pyrex serological tubes. A control

tube was kept at room temperature through the period that the remaining tubes were placed in the cobalt vault. At the termination of the exposure period, all tubes were stored in the refrigerator and tested the following day for bacteriophage activity.

To test the activity of the several samples of phage, beef extract agar was poured into several sterile Petri dishes. After the medium had solidified, approximately 2 ml of a 1:100 saline dilution of a 24-hour broth culture of S. aureus was placed on the surface of the medium. The suspension was flooded over the entire surface of the agar and the excess removed with a sterile pipette. The plates were then placed at 37°C with the covers of the dishes removed, and the suspension of organisms allowed to dry on the surface of the agar for a period of 30 minutes. Upon returning the cultures to the laboratory, the dishes were tilted on the laboratory table by placing one edge of the plates on a section of rubber tubing. One drop of each suspension of treated bacteriophage, as well as of the unexposed control, was placed at the upper edge of the agar medium and the drop allowed to flow down the agar surface by gravity. The plates were then incubated at 37°C and the activity of the bacteriophage on the staphylococcus noted at the end of 24 hours. Fig. 23 presents the photographs of the results of this experiment.

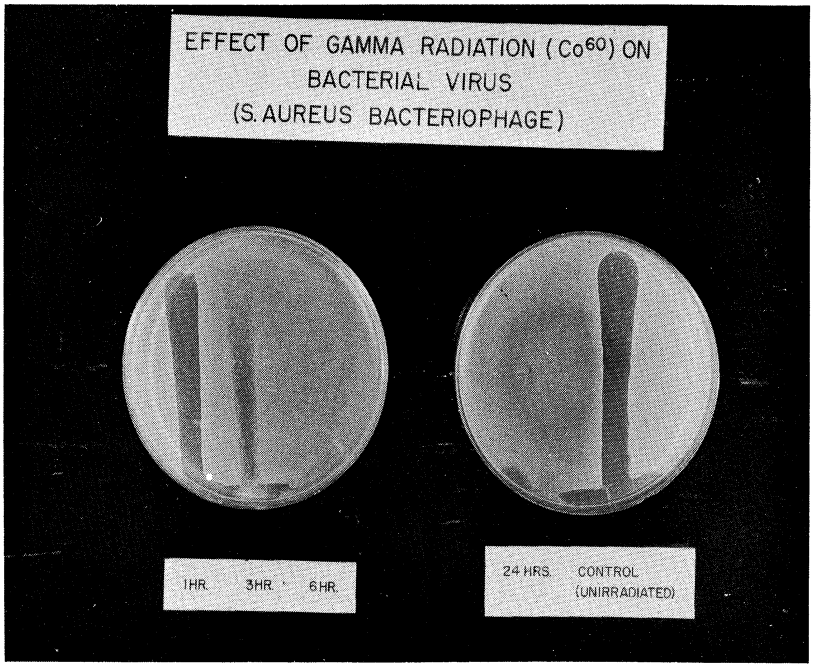


Fig. 23. Effect of Gamma Radiation (Co^{60}) on Bacterial Virus (S. Aureus Bacteriophage).

Results: It is evident from the data presented in the photographs of the agar plates that 1 hour (80,000 rep in air) of gamma irradiation of the bacteriophage resulted in no appreciable change in the virus compared to the control. It should be stressed, however, that this preliminary test is a very crude technique for detection of phage activity. By using a more refined procedure (dilution) in subsequent studies, partial destruction of the bacteriophage at the 1-hour (80,000 rep in air) period may be detected. The photographs indicate that after 3 hours (240,000 rep in air) of gamma irradiation of the phage, approximately 50 per cent of the particles may have been destroyed, and following 6 to 24 hours (480,000 to 1,920,000 rep in air) of gamma irradiation complete destruction of the bacteriophage can be expected. Subsequent experimentation on this phase will include attempts to photoreactivate irradiated bacteriophage[10,11].

d. Experiment No. VI. The Effect of Gamma Radiation (Cobalt-60) on a Human Strain of Mycobacterium Tuberculosis. A study was made of the effects of cobalt-60 on a mouse-virulent strain of M. tuberculosis (Olsen strain) which was originally isolated from a human case of clinical tuberculosis infection.

Experimental: A suspension of the tubercle organisms was prepared by removing 5 loopfuls of a heavy growth of the organisms from a Petragnani's slant that had been incubated at 37°C for a period of 11 weeks. The organisms were added to a small screw-capped bottle, containing glass beads and 5 ml of sterile saline solution (0.85% NaCl). The contents of the container were shaken vigorously in order to break up the bacterial culture, following which an additional 30 ml of saline solution was added to the container. The bacterial suspension was distributed in 5-ml amounts into 15-ml vaccine bottles provided with rubber stoppers. A control bottle was maintained at room temperature throughout the period that several of the suspensions were irradiated with gamma rays from cobalt-60. Following the irradiation period, all samples were placed in the refrigerator. A portion of each sample was placed on the surface of duplicate tubes of Petragnani's medium 24 hours after irradiation. The latter were incubated at 37°C for a period of 2 months and the presence or absence of growth of the tubercle bacilli noted.

Two days after the radiation experiment, 0.1-ml amounts of each suspension, including the control, were injected intravenously by the tail-vein route, into each of 5 normal albino mice (this injection series was carried out by Dr. D. W. Smith, Postgraduate Fellow, and A. A. Grover, Graduate Student in the department of Bacteriology). The animals were kept on a normal Rockland mouse-food diet throughout the experiment, i.e., for a period of 53 days, at which time the animals surviving the experiment were sacrificed by ether anesthesia. One of the control animals died during the second week following injection. However, upon autopsy it was determined that this animal died from some cause

other than the injection of the tubercle organisms. Two additional animals in the control series were found dead on the 45th day of the experiment; however, the condition of the deceased animals was such (canabalism by the surviving animals in the same cage) that autopsy of these did not appear advisable. This was also true of three animals that died on the 42nd and 43rd days, respectively, of the experiment in the 3-hour (240,000 rep in air) radiation series. Nevertheless, of the animals that were autopsied at the termination of the experiment and from the data obtained from the Petragnani's cultures, certain conclusions could be drawn from this preliminary experiment. The results of this study are presented in Table XI.

TABLE XI

THE EFFECT OF GAMMA RADIATION (COBALT-60) ON M. TUBERCULOSIS (OLSEN STRAIN)

Hrs Irradiation of TB Culture	Radiation Dose (rep in air)	Growth on Petragnani's Medium	Animals Inoculated	Animals Surviving Test	Gross Path.	Acid-fast Stains
0 (control)	0	Positive	5	2	Pos.	Pos.
1	80,000	Positive	5	5	Pos.	Questionable
3	240,000	None	5	2*	Neg.	Neg.
6	480,000	None	5	5	Neg.	Neg.
24	1,920,000	None	5	5	Neg.	Neg.

*See remarks in "Results" of this experiment on pages 58 and 59.

Results: The control animals which received unirradiated tubercle bacilli showed definite evidence of tuberculosis of the lungs. Slide smears and acid-fast stains of the lungs showed definite evidence of acid-fast bacilli. The animals surviving in the 1-hour (80,000 rep in air) irradiated suspension series showed some evidence of gross pathology of the lung tissue; however, diligent search of acid-fact preparations of the same tissues failed to show evidence of tubercle bacilli. The remaining animals in the 3-hour, 6-hour, and 24-hour (240,000, 480,000, and 1,920,000 rep in air) groups showed no evidence of gross pathology of the lungs or spleen, nor were there any acid-fast bacilli detected

in the same smears of these tissues. Observation of the Petragnani's slant showed a luxuriant growth of the tubercle bacilli in the control and also a good growth, although less in amount than the control, in the 1-hour (80,000 rep in air) irradiated suspension. The tubes containing suspensions from the 3-hour, 6-hour, and 24-hour (240,000, 480,000, and 1,920,000 rep in air) irradiated series were completely devoid of colonies at the termination of the experiment. Although the suspensions were all inadvertently destroyed during the intervening period of the experiment, thus preventing a study of the possible immunizing properties of the killed vaccines, additional studies of these organisms are contemplated in which this subject will be taken into consideration.

4. The Effect of Gamma Radiation on Toxins, Antisera, and Antibiotics

a. Experiment No. VII. The Effect of Gamma Radiation (Cobalt-60) on Tetanus Toxin*. Tetanus toxin is a labile protein substance, and a wide variety of mild chemical and physical agents render it nontoxic for experimental animals. Physical agents found to be effective are mild-heating and radioactive materials such as radium; also ultraviolet light and chlorophyll have been noted to have an inhibiting action on the toxin. Efrati[12] studied the effects of x-rays on the same system and Larson, Evans, and Nelson[13] and Vincent[14] described the action of surface-active soaps on the toxin. The activity of various oxidizing substances on the toxin has been reported by Halter[15], Lippert[16], and Jungeblut.[17]

Experimental: The present report deals with the effects of gamma radiation from cobalt-60 on the toxic fraction of tetanus toxin and also on the immunizing activity of the toxin following irradiation. A purified toxin (50% pure), furnished through the courtesy of Dr. R. C. Pittenger of the Lilly Research Laboratories, Indianapolis, Indiana, was used for the radiation study. This toxin was capable of killing guinea pigs in a dilution of 30×10^6 when 1 ml was injected subcutaneously. The toxin was diluted 1:25 with sterile 15% gelatin containing 1:10,000 merthiolate as a preservative. A 0.2-ml portion of the diluted toxin was placed in sterile serological tubes plugged with cotton and the latter irradiated for the periods designated in Table XII. Control tubes were maintained at room temperature throughout the irradiation period of the samples exposed to cobalt-60. After irradiation, all tubes were placed in the cold room and titrations in mice carried out 24 hours after the original toxin had been diluted for this experiment. The pH of the solution was 7.4. Toxicity for mice was determined by injecting 0.1 ml of the toxin solutions subcutaneously. The results of this study are given in Table XII.

* This study was carried out in collaboration with Ray DeHaan, graduate student in the Department of Bacteriology of the Medical School.

TABLE XII

EFFECTS OF GAMMA RADIATION (COBALT-60) ON TOXICITY OF TETANUS TOXIN

Number of Mice		Hrs	Radiation	Survival Time Hours	
Control	Irradiated	Irradiated	Dose (rep in air)	Control	Irradiated
2	6	1/2	40,000	24;24	24;24;24; 24;24;24
2	6	1-1/2	120,000	24;24	All survived
2	6	6	480,000	24;24	All survived
6	6	24	1,920,000	24;24;24 24;24;24	All survived

Results: From the data given in Table XII it is apparent that gamma radiation for 1-1/2 hours (120,000 rep in air) would detoxify the calculated amount of 25,000 MLD's of the tetanus toxin. This may be attributed to the presence, in part, of hydrogen peroxide which is produced by gamma radiation. The activity of oxidizing substances has been shown by a number of workers to have detoxifying action on the same system.

Immunizing activity of the detoxified toxin: Guinea pigs were injected with 0.2 ml of the detoxified toxin which received a 1-1/2-hour (120,000 rep in air) radiation dose. This amount of material before detoxification contained approximately 30,000 MLD's for guinea pigs. The animals received 3 injections of the treated toxin over a period of 3 weeks. They were then allowed to rest for 1 week before being challenged with 100 MLD's of untreated toxin. Of the 4 animals used in this study, all died at approximately 72 hours, which also was the time of deaths for the controls. It is apparent from this preliminary study, therefore, that accompanying detoxification of tetanus toxin, under the conditions of this experiment, there is also a loss in immunizing or protecting activity of the toxin.

b. Experiment No. VIII. The Effect of Gamma Radiation (Cobalt-60) on Agglutinating and Precipitating Antibodies.* Brasch and Huber[18] have described the relative ineffectiveness of high-energy electrons on antibodies to diphtheria toxin in dosages that afforded sterile serums. Thus, it appeared desirable to include the effects of gamma radiation from cobalt-60 on two antibody systems in the present study.

* This study was carried out in collaboration with Jack Battisto, graduate student in the Department of Bacteriology.

Experimental: Effects of gamma radiation on red blood cell agglutinins
Into each of 5 serological tubes was placed 0.5 ml of serum containing antibodies to mouse red blood cells (this serum had a 1:10,000 concentration of merthiolate as a preservative and was kept at 5°C when not in use). A control tube of the serum was maintained at room temperature while several of the tubes (cotton-plugged) were being irradiated in the cobalt vault. The sera were then stored in the refrigerator for several days, or until tests were made on their agglutinating properties. The sera were diluted with saline solution (0.85% NaCl) in twofold dilutions, following which 0.5 ml of a 2% suspension of mouse red blood cells was added to each tube. The latter were then held at 37°C for 2 hours and readings of the extent of agglutination in each recorded. These readings were made as (++++) for complete agglutination, (++) for partial agglutination and (-) for no agglutination. The results of this experiment are presented in Table XIII.

Results: From the data given in the table it is evident that exposure of the red blood cell agglutinating serum to cobalt-60 for a period up to 6 hours has no appreciable effect on the agglutinating activity of the serum (the difference of a (++) reaction in the latter series in the 1:128 dilution and the corresponding value in the 1:256 dilution of the control tube, is well within the range of the experimental error of this technique). Exposure of the serum for 24 hours to the radiation resulted in an appreciable drop in titer of the agglutinating serum, from the original 1:256 dilution down to 1:16 concentration.

Experimental: Effects of gamma radiation on type I pneumococcus soluble-specific-substance (SSS) precipitins. To each of 5 serological tubes was added 0.5 ml of serum containing antibodies to pneumococcus Type I polysaccharide (SSS). (This serum contains no preservative but was kept at -40°C.) One tube (cotton-plugged) was maintained at room temperature, while the remaining tubes were irradiated in the cobalt vault. After irradiation, all tubes were stored in the refrigerator for several days, or until determinations were made on the precipitin titers of the various samples. To each precipitin tube was added 0.1 ml of serum. An 0.1-ml amount of tenfold dilutions of the homologous Type I pneumococcus polysaccharide was then "layered" over the top. The formation of a ring of precipitate at the interface of the two solutions was read as positive (+). The results of this experiment are given in Table XIV.

Results: From the data given in the table it is evident that precipitating antibodies used in this study are extremely sensitive to the gamma rays emitted from cobalt-60, which is in contrast to the findings of the relative resistance of red blood cell agglutinins reported above. Thus, in the present series 1-hour (80,000 rep in air) irradiation of the serum resulted in complete loss of precipitating activity of the antibodies. These studies are of a preliminary nature and will be investigated further in subsequent experiments.

TABLE XIII

THE EFFECT OF GAMMA RADIATION (COBALT-60) ON RED BLOOD CELLS AGGLUTINATING ANTIBODIES

Irradiation Time	Radiation Dose (rep in air)	RBC control (no anti-serum)	Agglutinating Titers (Dilution of Antiserum) 1:2	1:4	1:8	1:16	1:32	1:64	1:128	1:256	1:512
0 (Unirradiated Control)	0	-	++++	++++	++++	++++	++++	++++	++	++	-
1 hr	80,000	-	++++	++++	++++	++++	++++	++++	++	-	-
3 hrs	240,000	-	++++	++++	++++	++++	++++	++++	++	-	-
6 hrs	480,000	-	++++	++++	++++	++++	++++	++++	++	-	-
24 hrs	1,920,000	-	++++	++++	++++	++	++	-	-	-	-

TABLE XIV

THE EFFECT OF GAMMA RADIATION (COBALT-60) ON TYPE I PNEUMOCOCCUS POLYSACCHARIDE PRECIPITATING ANTISERUM

Irradiation Time	Radiation Dose (rep in air)	Serum-Saline Control (no antigen)	Antigen Titers (Dilution of antigen) from 1 mg/ml						
			10^{-0}	10^{-1}	10^{-2}	10^{-3}	10^{-4}	10^{-5}	10^{-6}
0 (Unirradiated Control)	0	-	-*	+	+	+	+	+	-
1 hr	80,000	-	-	-	-	-	-	-	-
3 hrs	240,000	-	-	-	-	-	-	-	-
6 hrs	480,000	-	-	-	-	-	-	-	-
24 hrs	1,920,000	-	-	-	-	-	-	-	-

*Presumably a "prezone" reaction in which the undiluted antigen fails to show a precipitation with the antiserum.

c. Experiment IX. The Effect of Gamma Radiation (Cobalt-60) on the Potency of Several Antibiotics. Brash and Huber[18] demonstrated that various drugs could be effectively sterilized by electrostatic impulses from the Capacitron with no detectable harmful effects on the chemical or biological activity of the materials. Brownell,[19] moreover, reported on the results of a series of collaborative studies carried out with Parke, Davis and Company in which gamma radiations from the present cobalt-60 source (1000 c) also effectively sterilized several pharmaceutical products. The latter items selected covered a wide range of molecular weights and included those which were heat-sensitive and gave some difficulty in sterilization by other methods. In the study, the samples were contaminated with B. subtilis and then irradiated in glass-sealed or rubber-capped containers. The results indicated that gamma radiation from cobalt-60 caused complete sterilization in all instances of exposure for 24 hours (1,920,000 rep in air). Calcium gluconate and ascorbic acid appeared to be unaffected by this amount of exposure, insofar as the chemical nature of the compounds was concerned. There was about 20 per cent loss in activity in the irradiated hormone "theelin" and almost complete loss in activity of the hormone "pitocin". Antitoxins for tetanus and a mixture of tetanus-diphtheria toxin with pertussis vaccine appeared to be unaffected as the result of 24 hours (1,920,000 rep in air) of irradiation, insofar as their biological activities were concerned.

Experimental: In the present study several antibiotics were irradiated in the dry state for 24 hours (1,920,000 rep in air) with gamma radiation from the same source used by Brownell and the Parke, Davis group. Following irradiation the powders (along with control samples) were dissolved in sterile distilled water in appropriate concentrations and cylinder assay tests carried out for potency of the various antibiotics, using Staphylococcus aureus as the test organism.

Tests for potency of the treated, as well as the untreated, controls were carried out three days after irradiation. The antibiotics were diluted in distilled water and the solutions were stored in the refrigerator. Additional tests for potencies were made on the same solutions 6 days and again 90 days after irradiation. The results of these tests are presented in Table XV.

Results: In summarizing the data given in Table XV it may be noted that, in general and within the limits of experimental error of these preliminary tests, there appears to be no appreciable overall differences in potencies between the irradiated and unirradiated samples of the several antibiotics. With the exception of penicillin, which appeared to retain its potency while in solution for 90 days (both in the irradiated and unirradiated series) the remaining solutions showed some losses in potencies which appeared comparable in degree in the irradiated as well as the unirradiated control solutions. The footnote under the table indicates the physical appearances of the powders or

crystals soon after irradiation. A slight change in color, from white to grey, was noted in the irradiated samples of styreptomycin and chloromycetin. However, there did not appear to be any differences in the degree of solubilities of these samples as compared to the unirradiated ones.

TABLE XV

THE EFFECT OF GAMMA RADIATION (COBALT-60) ON THE POTENCY OF SEVERAL ANTIBIOTICS

Antibiotic*	Potencies - Days After Irradiation (same solutions) 3 days		6 days		90 days	
	Irrad.	Unirrad.	Irrad.	Unirrad.	Irrad.	Unirrad.
K-Penicillin "G"	37mm**	37mm	37mm	37mm	37mm	37mm
Streptomycin KCl	24mm	24mm	18mm	16mm	13mm	13mm
Aureomycin	31mm	31mm	25mm	20mm	23mm	24mm
Chloromycetin	23mm	23mm	14mm	21mm	16mm	16mm
Terramycin	29mm	24mm	17mm	20mm	21mm	21mm

*Physical appearance of the antibiotic powders, or crystals, immediately after removal from the cobalt-60 vault following 24 hours exposure to the gamma rays:

Penicillin: No color change, no apparent change in solubility in water.
Streptomycin: Slight change to gray of treated powder: no change in solubility.
Aureomycin: No change in color, no apparent change in solubility.
Chloromycetin: Slight change to grey in treated crystals; no apparent change in solubility.
Terramycin: No change in color; no apparent change in solubility.

**Figures represent zones of inhibition against S. aureus in the cylinder assay test of solutions prepared for treated and untreated powders.

D. IRRADIATION OF PHARMACEUTICALS

Preliminary tests on the radiation sterilization of pharmaceuticals have been reported in Progress Report 2. Since then, a meeting of manufacturers

of antibiotic products was held in Washington, D. C., on February 14, 1952, regarding the Radiation Sterilization of antibiotics. Professor L. E. Brownell of the University of Michigan attended the morning session of this meeting. The papers presented at this meeting acquainted the various manufacturers of antibiotics with the possibility of radiation sterilization and brought to light work which had been performed in the various laboratories.

Dr. E. M. Weber, of the Charles Pfizer Company, reported that his company has studied the radiation sterilization of antibiotics using the Van de Graaff accelerator, the Capacitron, electron generator, and gamma radiation from cobalt-60. These studies indicated that there is a close parallel in beta-particle and gamma radiation, both with regard to dosages required for sterilization and with regard to side effects. Sterility after irradiation was determined according to established FDA tests. The products were studied for changes in both chemical and physical behavior after irradiation. The changes studied were (1) loss in potency, (2) change in toxicity, (3) time and temperature stability, (4) gross appearance, and (5) effect of high temperature on stability of crystalline forms. Containers were checked for changes. Discoloration of the glass was observed as well as disturbance of the silicone film used on the inner surface of some vials.

In some of the tests, freeze-dried suspensions of a variety of organisms were used to contaminate different samples of several antibiotic products of the Pfizer Company. The contaminated samples were placed in polyethylene bags and radiated with the Capacitron using a total of 1.5 million rep. All samples were found to be sterilized by this dosage. Increasing this dosage fivefold gave no loss in potency of the various antibiotics. Stability tests were made at temperatures of 0°, 25° and 37°C for nine months and indicated no change in stability. The sodium and potassium salts of penicillin G were heated to 100°C to check the stability, and it was found that these crystalline salts lost some of their stability as a result of irradiation. The irradiated samples were somewhat darker in color than the controls; the discoloration appeared to be a function of the amount of impurity present. No change in acute or chronic toxicity was noted when using five times the radiation dose required for sterilization. Glass containers suitable for radiation sterilization appear to be a real problem, however, because it is not believed the products marketed in the brownish glass resulting from irradiation will be able to compete on an equal basis with products marketed in clear glass containers.

Dr. G. C. Bond, of the Upjohn Company reported that their company has conducted a variety of experiments using their own Van de Graaff accelerator and have clinically evaluated products sterilized by radiation. These are the first reported clinical tests using irradiated products. Radiation was found to be lethal to all types of microorganisms including spores, and the dosage

required for sterilization was found to vary with the materials tested. It was found that each drug product had to be considered individually and a suitable radiation dosage and procedure must be developed for each product. Following this procedure, it was possible to sterilize a wide variety of pharmaceuticals without deterioration of the product. The pharmaceuticals which were sterilized included three multivitamins, three preparations from animal tissue, two steroids, three hormones, one crystalline powder, and one blood product. Tests indicated that the two antibiotic preparations lost some activity as a result of irradiation. A long-range stability test showed no difference between the irradiated products and the control. Acute and chronic toxicity tests indicated no difference between the irradiated product and the control. A very slight change in solubility in some of the products after irradiation was noticed, but this change in solubility is so slight it is not considered important. Some products were discolored by radiation, but in many cases the discoloration faded after storing. Radiation had no effect on syringeability of injectable suspensions, on pH, or on infrared spectra of several products. In the clinical tests, six products (penicillin, multivitamins, and purified gland extracts) were thoroughly studied after radiation sterilization and evaluated clinically. No differences were observed between the product sterilized by radiation and the control from the standpoint of administration, patient acceptability, or therapeutic effectiveness.

Dr. Henry Welch, Head, Division of Antibiotics, U.S. Food and Drug Administration stated the position of F.D.A. regarding the use of radiation as a sterilization process. The F.D.A. would have no hesitancy about granting a well-documented request for recognition of the process, providing it had been shown that the process has no effect on potency, stability, or toxicity and performs the desired function of sterilization. The F.D.A. has no objections to accepting antibiotics packaged in colored glass providing other requirements are met.

Discussions from the floor indicated that most of the manufacturers of pharmaceuticals considered the discoloration of glass to be one of the real disadvantages of radiation sterilization and that from the standpoint of sales the discolored-glass containers would have poor appeal to the market. The general attitude after presentation of the papers was that radiation sterilization of pharmaceuticals was feasible and offered a new method of sterilizing products difficult to sterilize by conventional methods.

Cooperative Tests with Parke, Davis and Company

As a result of the Washington meeting, it was decided that the next step with pharmaceutical products should be the investigation of a suitable glass which would be free from discoloration. Samples of three glasses were received from Dr. H. P. Hood of the Corning Glass Works: The first a colorless

glass (EXP. 733 BB); the second, a lightly tinted blue-green (EXP. 733 BI); and the third, a pale olive color (EXP. 733 BJ). The tinted glasses were described as being of lower cost than the clear glass. These glasses were given a radiation dosage of 4,000,000 rep in air and the colorless glass (EXP. 733 BB) showed no appreciable discoloration. The Parke, Davis and Company's Chemical Laboratory tested this glass and found that the glass tubing can be fabricated into ampules without special technique. They also reported that this glass was found to meet Type 1, USP Standards. As a result of these tests the following comment was received from Dr. John Controulis, Assistant Director, Products Development Department, Parke, Davis and Company:

> "... given a new product incapable of sterilization by ordinary methods, we (Parke, Davis and Company) would consider radiation sterilization on an experimental basis. But as for production use of the method, it appears that more optimal data on the availability of and technology of the use of fission products on an industrial basis are necessary before Parke, Davis and Company would be willing to embark on such a program."

E. STERILIZATION OF BLOOD PRODUCTS

Experiments on the sterilization of various pharmaceutical products have indicated that gamma radiation can be used successfully but may have a limited application because it will have to compete with other methods of sterilization. However, there is one group of products, the blood fractions, which has not been satisfactorily sterilized by any other method. These products require sterilization to protect against the disease, serum hepatitis. Gamma radiation in this particular instance may offer the solution to an important problem in public health for which, at present, there is no simple solution. The layman is well acquainted with the importance of research in the crusades against diseases such as cancer, poliomyelitis, tuberculosis, etc., but has only slight knowledge about this new disease, serum hepatitis. Therefore, some background will be given so that the importance of this study can be realized.

1. Description of Serum Hepatitis by American Red Cross

Some background information for the layman is given in Public Affairs Pamphlet No. 145, "Blood's Magic For All", by Alton L. Blakeslee, reprinted in 1949 by the American Red Cross, which states on page 21:

"Blood, as whole blood, plasma, and the array of blood products, was one factor in a brilliant wartime medical record— the survival of 97.3 per cent of all wounded American Servicemen. But V-J Day and peace brought...the ending, on September 15, 1945, of the Red Cross wartime blood donor program.

"Doctors and surgeons immediately missed the once-abundant supplies of blood.... There was a temporary supply of plasma and some of the fractions, mainly the anti-measles injections, from surplus Red Cross plasma. The armed forces turned back 4,777,980 units—250 cc each—of plasma after the war. The Red Cross distributed most of this plasma and anti-measels gamma globulin to physicians through state health departments.

"Of the total surplus plasma, 3,593,773 of the 250-cc units have been made available to physicians. But it soon was discovered that it had to be handled with caution.

"For, unfortunately, this plasma had been produced before new facts were learned about a liver disease, infectious hepatitis [now recognized as homologous serum hepatitis], that causes a form of jaundice. It hadn't been realized that people who once had had the disease could carry the hepatitis virus in their blood. The virus might remain dormant for years, but still be capable of causing the disease in someone else.

"In the wartime blood collections, many donations were pooled for efficiency and economy in processing plasma. Plasma processed from 25 to 50 donations would be mixed in one batch. If just one of these persons had the virus, the whole batch might unknowingly be contaminated. Although only one person in several thousand had the virus, many batches of plasma became contaminated. The virus resisted both the drying and refrigeration in handling the plasma.

"Studies showed that only about 4 to 5 per cent of all the plasma produced in the Red Cross program did contain the virus, but it might give jaundice to some one receiving it. As soon as this peril became known, the Red Cross, the American Medical Association, and private physicians immediately warned that the surplus plasma should be used only as a life-saving measure, and with due consideration that any one bottle of it might contain the virus. The Red Cross is discontinuing the distribution of this surplus plasma, using it and any outdated plasma for blood fractions."

However, this has not solved the problem because it has also been found that certain blood fractions also transmit the disease of serum hepatitis.

2. Description of Serum Hepatitis by California State Department of Public Health

A technical description of the disease is presented in "A Manual For the Control of Communicable Diseases in California", compiled by the California State Department of Public Health, 1951. The description of the disease and control measures are given below:

"Description of Disease

"Identification: Clinical and laboratory manifestations [of homologous serum hepatitis] are practically indistinguishable from infectious hepatitis...The chief points of difference are: the incubation period, and the mode of transmission. The relationship between the two conditions is in doubt; some investigators consider them to be caused by the same agent.

"Etiologic agent: A specific virus or viruses, relatively heat-resistant.

"Source of infection: Blood or blood products from an infected person.

"Mode of transmission: By parenteral (intravenous, intramuscular, or subcutaneous) inoculation of infected blood, plasma, or serum; or by administration of prophylactic or therapeutic agents from syringes and needles contaminated with traces of blood from infected persons.

"Incubation period: Estimated at two to six months, probably averaging 12 to 14 weeks.

"Period of communicability: The virus has been found in the blood long before the onset of symptoms in experimentally inoculated volunteers. How long an individual may harbor the virus subsequent to infection is unknown. It is probable that some persons become 'carriers' without having experienced a clinically recognized attack.

"Susceptibility and resistance: Susceptibility is high; about one out of five persons who have received injections of infected blood or its derivatives may be expected to manifest the disease

in a clinically recognizable form.

"Prevalence: Not determined because of difficulty of distinguishing serum hepatitis from infectious hepatitis. Estimates of the incidence among recipients of pooled blood products have varied from around 2 per cent to as much as 7 per cent. A high incidence has also been found in certain clinics where therapeutic parenteral injections were given."

"Control Measures

"General:

"Administration of whole blood, and particularly of pooled blood serum or plasma, only in cases where adequate indications exist for its therapeutic usefulness or necessity. Pooling increases the likelihood that the blood product will contain the infectious agent.

"Thorough heat sterilization of syringes and needles, and of stylets for finger-puncture, prior to each use. It should be realized that in administering intramuscular injections traces of blood commonly return into the syringe, thus contaminating it.

"The regulations pertaining to the production and distribution of biologics in the State of California require that all human plasma and serum intended for use in human beings be adequately exposed to ultraviolet light radiation or by some other approved method to insure the destruction of the virus of hepatitis. [Ultraviolet light has since been found to be inadequate.]

"The infected individual, contacts, and environment:

"Recognition of the disease: The development of acute hepatitis in the absence of cases of the naturally occurring disease, and with a history of having received blood transfusions two to six months previously is suggestive. A group of cases among patients in a clinic where parenteral therapy is commonly employed (e.g., a syphilis clinic) would also arouse suspicion."

"Concurrent disinfection: None.

"Terminal disinfection: None.

"Quarantine: None.

"Immunization: Passive immunization by administration of immune serum globulin is under study.

"Investigation of source of infection: Of importance in determining the frequency of this disease and revealing errors in technique of parenteral inoculations and in use of human plasma and serum unprotected by required radiation."

A discussion of viral hepatitis is given in a report by John Graikoski, Research Assistant, Michigan Memorial-Phoenix Project 41, as follows:

"The term viral hepatitis has been proposed by Neffe[20] to those forms of hepatitis caused by hepatotropic, filterable, infectious agents which have not been identified by specific serological means, but produce a liver injury as their outstanding clinical manifestation. The assumption is made that viral hepatitis is caused by a filterable agent; since all evidence seems to point to an infectious agent for these syndromes.[20,21] At least two forms of viral hepatitis are now recognized: (1) infectious hepatitis (catarrhal jaundice, hepatitis epidemica, infective hepatitis), and (2) homologous serum hepatitis (late post-arsphenamine jaundice, syringe jaundice, transfusion jaundice, yellow vaccine jaundice).[21] The former refers to the sporadic or epidemic forms of the naturally occurring disease. The latter is designated to the hepatitis produced in patients which have received parenteral injections of blood and blood products containing the virus.[21]

"Homologous serum jaundice is an artificial disease in that it is transmitted only by the parenteral inoculation of plasma, serum and serum products.

"Numerous outbreaks of serum hepatitis have occurred following immunization against yellow fever and measles using convalescent plasma and transfusions of pooled blood.[21] In the U. S. Army there occurred an epidemic in 1942 of 51,337 cases of serum hepatitis following immunization against yellow fever with an icterogenic vaccine.[22] Also, the disease can be transmitted by improperly sterilized syringes and needles used in anti-syphilitic therapy and in blood drawing.[21] There also has been a case of serum hepatitis following tattooing.

"The virus causing serum hepatitis in man has not been transmitted to laboratory animals, although attempts have been made to transmit it to rhesus monkeys, rabbits, mice in embryonic stage, and newborn cotton rats, pigs, Syrian hamsters, guinea pigs, horses, young chicks, embryonating hens' eggs, and tissue culture.[21]

Consequently, most of the work has been done with human volunteers, as in infectious hepatitis.

"The transmission of the virus has been demonstrated in volunteers after the parenteral inoculation of serum obtained from patients which had received icterogenic yellow fever vaccine.[23] The incubation period of the vaccine varies from 40 to 160 days. In this respect it differs from that of infectious hepatitis, which has an incubation period of 10 to 40 days.[24] Serum hepatitis also differs from infectious hepatitis in that it has not been demonstrated in the feces of patients with the acute phase of the disease and the disease has not been transmitted via oral routes.[20,21]

"The various characteristics of the agent of serum hepatitis, such as heat and chemical resistivity and passage through bacterial filters, are identical with those of infectious hepatitis, with the possible exception of greater heat resistance. In addition, it has been demonstrated that the virus can be inactivated by ultraviolet radiation,[24] of wavelength 2537Å, a fact which has found intensive but not always successful use in controlling the disease in plasma and serum products.

"Serum hepatitis and infectious hepatitis are clinically and pathologically indistinguishable after the onset of symptoms. The epidemiological characteristics of the disease seem to be the same as those of infectious hepatitis, although the incidence seems to be greater in older age groups, but this may be due to the receiving of larger volumes of blood in stricken conditions.

"Homologous serum hepatitis has been demonstrated in convalescent patients who were re-inoculated with serum from experimentally induced serum hepatitis. No cross immunity was shown to exist between serum hepatitis and infectious hepatitis, a fact highly suggestive of the differences of the two etiological agents.[21]

"Table XVI shows the various similarities and dissimilarities between serum hepatitis and infectious hepatitis. However, the exact relationship between infectious hepatitis and homologous serum jaundice is not known. The differences in the two diseases are based on length of incubation period, failure to transmit serum hepatitis orally, and failure to detect the agent in the feces. On the other hand, the clinical and pathological pictures are undistinguishable. Whether these similarities and dissimilarities are sufficient to warrant the classification as two distinct entities remains to be determined.

TABLE XVI

Comparison of Behavior of Viruses of Infectious Hepatitis and Homologous Serum Jaundice in Experimentally Infected Volunteers[21]

	Virus	Infectious Hepatitis	Serum Hepatitis
1.	Filterable	Seitz SK	Seitz SK
2.	Resistance to heat	56°C, 30 min	56°C, 30 min
3.	Susceptible Host	Man	Man
4.	Incubation (Days)	15-34	56-134
5.	Route of Infection	Parenteral or oral	Parenteral
6.	Virus in stool	Acute phase	Not demonstrated
7.	Virus in serum	Acute phase	Incubation period acute phase
8.	Immunity	Present	Present
	a. Homologous	None apparent	None apparent
	b. Heterologous	None apparent	None apparent

3. Cooperative Research with Michigan Department of Health

The problem of serum hepatitis is particularly important to the Division of Laboratories of the Michigan Department of Health. This laboratory is one of the larger laboratories manufacturing blood products and is the only laboratory that produces antihemophilic globulin blood fraction. This antihemophilic blood fraction has saved a great many lives in Michigan and elsewhere and is a tremendously useful tool in surgery. However, it has been found to be a carrier of serum hepatitis and the procedures of sterilization using ultraviolet light which were developed by the Michigan Department of Health have not proven completely satisfactory.

The possibility of gamma radiation for sterilization of blood fractions is of particular interest, since the utilization of ultraviolet light has proven inadequate. A plan of study for this research has been prepared by the Associate Director, Dr. H. D. Anderson, and members of the Biologic Products Section Staff of the Division of Laboratories of the Michigan Department of Health and is as follows:

"STUDY PLAN"

"Title of Study: Sterilization of biological products with gamma radiation.

"Estimated Time: 1 to 2 years - Present capacity of the radiation laboratory at the University of Michigan would permit our use of the facilities for a maximum of 48 hours per week. The present capacity of their radiation vessel would be approximately 6 bottles of 20-cc capacity, and the radiation time for each bottle will vary from 2 to 24 hours, so that the number of individual trials will be limited unless a larger and more powerful radiation source is obtained.

"Objective: The purpose of this study will be to explore the possibility of the use of gamma radiation for the sterilization of biological products in final containers, and at a later date, if the data so indicate, to attempt to sterilize biological products in bulk bottles. This laboratory will perform the necessary tests to assure stability, potency, safety, and sterility of each product tested.

"Reason or Need for Study: The current data available from studies with human volunteers indicate that the present method of using ultraviolet light is inadequate for the destruction of the virus of homologous serum jaundice in heavily infected pools of plasma; consequently, it seems advisable to study the use of a higher energy source, such as gamma radiation.

"Recent reports[25] from the New York State Laboratories indicate that the probable dosage necessary to sterilize the homologous serum jaundice virus will be of the order of 4 million rep, and they are undertaking a study involving three dosage levels of 2, 4 and 8 million rep, respectively. The New York study will employ a sample of the highly infectious pool of plasma which has been employed in the ultraviolet radiation studies by the National Institutes of Health.

"The plan is for the Michigan Department of Health and the University of Michigan to collaborate in an experiment designed to evaluate the use of gamma radiation for the sterilization of biological products, [particularly blood products] and we feel that such a collaborative study should be undertaken as soon as possible.

" Brief Outline of Plan: It is difficult at this time to give a complete outline of the study, since much of the later work will depend upon the early results which are obtained. It is proposed that in the course of this study the radiation times required to destroy common strains of bacteria and laboratory viruses will be determined in several types of suspending media, including the human blood products. We will also investigate chemical and physical changes which may result from gamma radiation, particularly in the blood products. If a procedure is developed to the point that it is practical for laboratory use, it might well be employed in the sterilization of certain bacterial media where heat or filtration causes adverse changes. This procedure might also be employed in the preparation of diagnostic antigens, the preparation of bacterial and virus vaccines, and many of the animal serum products. It would appear to be particularly useful if products could be sterilized in the final containers with no adverse effects.

"In an effort to begin these studies immediately and to perform certain essential experiments, involving a minimum of cost, we propose to determine the effects of various radiation dosages on several of the plasma fractions. Our first experiment will involve the irradiation of antihemophilic globulin, immune serum globulin, and normal serum albumin. Samples of these products will be irradiated for periods of 1 hour, 3 hours, 6 hours and 24 hours, respectively; then tests will be performed to determine changes in solubility, thermostability, biological potency, and sterility of each product.

"At a later date, these products will be inoculated with various types of bacteria and virus to determine the dosages required to sterilize each product. It is also proposed that should the process appear useful it will be used first in addition to routine ultraviolet radiation, and if no damaging effects are observed, the products will be subjected to suitable animal tests and later clinical trial. Finally, products radiated with only the gamma radiation will be tested clinically."

This indirect procedure is necessary when working with the virus of serum hepatitis as this is a disease of man alone and cannot be given to laboratory animals. The seriousness of the disease prevents experiments with humans. Following the above procedure, it is believed that the effect of gamma radiation can be demonstrated without infection.

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PART IV. SUBPROJECT M943-E, EXPLORATORY RESEARCH

Personnel:

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

A. PREPARATION OF STRONTIUM-90 BETA SOURCE

A great many proposals have been made for possible uses of beta radiations. These radiations are known to produce some effect on certain chemical reactions and are known to produce biological and bacteriological effects. Studies are being made to determine whether these effects can be utilized industrially. Some research groups are investigating the use of beta radiation to catalyze chemical reactions in both liquid and gaseous systems. Other research studies are being made in an attempt to improve combustion processes with beta-ray bombardment, and radiation sterilization is being extensively investigated.

The utilization of strontium-90 as a beta-ray source is being studied by this project. A primary purpose of this contracted research is to investigate possible uses of fission products. Strontium-90 is one of the most abundant fission products, with a rather long half-life of 25 years. It is one of the few pure beta-ray emitters, and its daughter product yttrium-90 is also a pure beta-ray emitter. Yttrium-90 disintegrates, with a 65-hour half-life, to stable zirconium-90. The energies of the radiations from strontium-90 and yttrium-90 are rather high, having maximum energies of 0.61 and 2.35 Mev, respectively.

An isotope source of beta radiation must compete against electron accelerators. These accelerators have certain advantages which are not so readily available from isotope sources. Accelerators can be turned on or off at will, the energy of the emitted particles can be quite carefully controlled, and tremendous doses can be delivered on localized areas. Radioactive beta-ray sources also have certain advantages; these sources do not require a heavy-duty power supply and they can be built in any desired shape to introduce radiation in the required manner. As even high-energy beta particles have short ranges, it is very necessary to have the beta-ray source and the target close to each other. Certainly no electron gun could provide the intimate

contact and great surface for reaction that would be introduced in a porous reactor bed such as ceramic Raschig rings, Berl saddles, etc., containing radioactive materials. It is believed that these radioactive sources have the greatest possibility of being usable over wide ranges of temperatures and pressures in ceramic sources.

Some difficulties result when attempts are made to utilize strontium-90 as a radiation source. Metallic strontium is an active alkaline earth, reacting readily with water or air. In order to protect the source and the samples, the strontium-90 must be made more stable. Stability can be achieved by combining strontium with other materials. Another complicating property exhibited by strontium-90 is that of "creep", the escape of radioactive materials from the body of the source. It is suggested that this creep may be a result of the recoil of nuclei on the emission of beta particles. It appears that if the molecular binding of a source were strong enough, no escape of radioactive nuclei would be noticed.

Two proposals have been considered for the elimination of these difficulties in using strontium-90. Strontium can be alloyed with more stable metals or it can be incorporated in a ceramic glaze. Each of these proposals has been investigated. Common, nonradioactive strontium was alloyed with magnesium and with aluminum. These binary alloys contained nominally 25% strontium and 75% aluminum, 50% strontium and 50% aluminum, and 25% strontium and 75% magnesium.

Samples of the alloys were crushed to a -100 Tyler mesh. These fine samples were carefully weighed and set aside in order to observe any weight change resulting from oxidation. Samples were stored at 70°F, 400°F, and 750°F. The samples stored at 70°F and 400°F showed only very slight oxidation after several weeks. There was no change noticed in the appearance of these samples. The sample maintained at 750°F oxidized rapidly. The magnesium strontium sample oxidized almost completely the first day. No inhibiting of the oxidation rate was observed as the oxidation proceeded.

Crushed samples (-100 mesh) were exposed to dilute reagents. The reagents used were all 0.1 N; they included sodium hydroxide, hydrochloric acid, nitric acid, and sulfuric acid. The strontium-aluminum alloys were rather resistant to dilute sulfuric and nitric acids, but reacted vigorously with sodium hydroxide and rather rapidly with hydrochloric acid. The strontium-magnesium alloy appears quite resistant to sodium hydroxide but reacts rapidly with the acids.

Further testing of strontium alloys is not being considered at the present time. Emphasis has been placed instead on the preparation of radioactive glazes. Glazes can be made to be very inert. A glaze covers only the

surface of a body. This is a desirable feature for a strontium-90 source because the beta rays have short ranges.

The preliminary work in the investigation required the selection of a body to be glazed and the development of a high strontium glaze. A perfect glaze must have the same coefficient of expansion as the body it covers. A clay was selected as the body to be used in the first experiments, and a glaze was developed which provided a fair fit to the body. The clay was prepared as small "cookies", samples about 1-1/2 inches square and 1/4 inch thick. The glaze was applied to one side of the cookie over an area of about 1 square inch. The glazes are about 0.02 inch thick. This glaze, which contains 20 per cent by weight of strontium, matures at temperatures above 2150°F.

The first tests were designed to determine the ability of glazes to prevent creeping of strontium-90. Smear tests were used to detect the presence of loose radioactive material on the glazes. Smear tests were made by carefully wiping the surface of the glazes with special blotting paper and determining the quantity of radioactive material picked up on the paper.

The first radioactive glazes were prepared by adding an extremely small fraction of strontium-90 to common strontium, resulting in cookies containing approximately 10^{-8} curie each. After a storage of several weeks at room temperature, no radioactive material could be detected by the smears. After the negative results were obtained with these samples, cookies were prepared using the same glaze but containing 10^{-5} curie of strontium-90 per cookie. Smears taken on these cookies showed traces of radioactivity. Data have been obtained by taking smears from these cookies stored for various lengths of time and at various temperatures up to 1000°F. Some of the results are listed in Table XVII; however, it must be emphasized that these results are quite qualitative. Smears cannot pick up all the radioactive material that is loose, although checks indicate that the first smear picks up most of the loose material. It cannot be expected however that the same proportion of the free radioactive material will be picked up on each test. Also the various cookies display somewhat different properties as a result of variances in the firing and cooling.

These tests indicate that even at temperatures up to 1000°F very little radioactive material escapes from the glazes containing 10^{-5} curie of strontium-90. As a result of these tests, attempts will be made to improve the quality of the glaze with the hope of using these glazes with 10^{-2} curie of strontium-90. Next the compositions and techniques used in preparing chemical ware will be considered.

Cookies containing larger fractions of strontium-90 in their glaze will be prepared, although the present facilities will limit the intensities

TABLE XVII

RESULTS OF SMEAR TESTS ON GLAZED COOKIES
CONTAINING RADIOACTIVE STRONTIUM-90

Sample Number	Time of Storage	Temperature of Storage, °F	Sr^{90} on Smear Expressed as Parts per Million of Sr^{90} on Cookie
A-6	7 days	80	2.0
A-8	7 days	80	5.2
A-2	8 days	80	6.1
A-2	6 weeks	80	3.5
A-4	6 weeks	80	4.4
A-3	96 hours	325	4.8
A-4	96 hours	325	5.8
A-1	96 hours	475	3.4
A-5	96 hours	475	2.4
A-3	36 hours	550	2.2
A-4	36 hours	550	33.3
A-1	30 hours	750	4.3
A-5	30 hours	750	11.5
A-2	85 hours	900	39.7
A-5	60 hours	1000	7.6

that can be handled safely. It has been calculated that, using the present glaze, approximately 30 curies can be placed on 1 square foot of surface. A porous bed of 1/2-inch spheres would accommodate approximately 85 square feet of surface in a cubic foot of packing. However, somewhat thinner glazes would possibly be more practical, as many of the beta rays would be stopped

by 0.02 inch of glaze. A glaze 0.002 inch thick would stop only a very small portion of the emitted beta particles.

Samples prepared in the future should be carefully investigated at many temperatures. Tests should include studies of the heat shock properties, tensile and compressive strengths, impact strength, and chemical inertness.

It is believed that the other research groups will soon be requiring beta sources from the fission products to complete the studies on the possible industrial uses of the fission products. Project M943-E will become inactive as of July 1, 1952, but the work on the preparation of strontium-90 beta sources will be continued in conjunction with the research being conducted on combustion engines and chemical reactions.

PART V. SUBPROJECT M943-F, OPERATION OF FISSION PRODUCTS LABORATORIES

Personnel:

Advisors: L. E. Brownell, Associate Professor 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. Nehemias, Health Physicist; M. E. Gluckstein, Research Assistant; E. C. Coleman, Research Assistant.

A. INTRODUCTION

The high- and low-level laboratories have been operating in routine manner since publication of Progress Report 2. The operational procedures proposed at that time have been implemented successfully. Several dose-rate determinations of our cobalt source have been made, the best calibration indicating a value of about 79,000 rep/hr for the dose rate in air on June 30, 1952. In addition, much time and effort has been spent on the preliminary plans and design of a versatile 10-kilocurie cobalt-60 source.

B. DOSE-RATE DETERMINATIONS

The dose rate within the present kilocurie cobalt cylinder has been studied by various means. The results are summarized in Table XVII.

1. Ferrous Sulfate

Preliminary calibration of the cobalt-60 gamma-ray source was made by the ferrous sulfate method of Weiss[1] and others. Our data yield a dosage of 62,300 rep/hr dose rate in water (using a value of 15.4 micromoles per liter per 1000 rep as recommended by Weiss). This result is being checked by further calibrations.

TABLE XVIII

SUMMARY OF DOSE-RATE DETERMINATIONS

Method	Medium	Dose (rep/hr)
Ferrous Sulfate (preliminary)	water	62,300
Chloroform	air	74,100
Film and Ionization Chamber	air	79,200 ± 9,000
Geometrical Estimate	air	78,300
Best Average Value in air as of June 30th, 1952		79,000

2. Chloroform

The rate of formation of hydrochloric acid by hydrolysis within a chloroform-water mixture[2,3] at known gamma dose rates (in air) was determined by pH measurements. The comparable rates of formation within the source yielded the value 74,100 rep/hr for one set of measurements.

3. Film and Ionization Chambers

Dupont Adlux film and Victoreen r-meter chambers were compared with ionization chambers ("cutie pie" type) calibrated for gamma radiation. Readings were taken at a point of suitable dose rate, outside and about a foot above the center of the source. The lower dose rate was demanded by the limited range of the "cutie pie" meters. At this position and dose rate the "cutie pies" and r-meters yielded essentially identical results. The films were calibrated at this position.

The dose rate within the source itself was too high for "cutie pie" measurement; therefore the film and r-meter were used. These produced similar results, the mean being

79,200 ± 9,000 rep/hr.

4. Geometrical Estimate

A point source of one millicurie of cobalt-60 will yield 13 r/hr at the surface of a 1-centimeter sphere (about 12.6 square centimeters). On the basis of reactor measurements and decay rate, it is estimated that the present laboratory source contains 900 curies of activity. This 900 curies would yield 900 x 1,000 x 13, or 11.7×10^6 r/hr at one centimeter. If the source is finite, the 1-centimeter surface is distorted but the same total gamma flux passes through this surface. Assuming that one can select an approximate isodose surface equivalent to this 1-centimeter surface, one would expect the dose rate to vary inversely with area (inverse square law, in the limiting case of a point source).

The laboratory source is a hollow cylinder, 6 centimeters in diameter and 35 centimeters long. A surface 1 centimeter distant from the source can be approximated by two concentric cylinders of 4- and 8-centimeter diameters, 37 centimeters long, joined at the ends by plane surfaces of 4-centimeter major radii and 2-centimeter minor radii. This surface differs appreciably from an isodose surface only in the location of the outer cylinder; i.e., the flux 1 centimeter outside the cylinder is slightly less than the flux of 1 centimeter inside. The outer radius is probably not more than 5 per cent too large. It should be borne in mind, however, that this treatment does not take into account: (1) secondaries produced in the measuring medium or the cobalt itself, (2) bremsstrahlen produced within the cobalt from its beta radiation, and (3) Compton energy degradation within the cobalt. The computation yields:

2π (4) 37 sq cm outside cylinder
2π (2) 37 sq cm inside cylinder
2π (64 - 16) sq cm ends

1695 sq cm total area of isodose surface

The ratio of areas then yields a calculated value of

$$\frac{12.6 \text{ sq cm}}{1695 \text{ sq cm}} \quad (11.7 \times 10^6) = 87{,}000 \text{ rep/hr.}$$

Correcting for absorption within the cobalt (1/4 inch thick) yields 78,300 rep/hr.

Proper weighting of these measurements gives a figure of 79,000 rep/hr for the dose rate in air of our cobalt-60 source on June 30, 1952. Fig. 24 indicates the dosage in air to which material irradiated previously has been subjected. From the figure we see that the dose in air was about 90,000 rep/hr when the source was first put into use (July, 1951).

The dose rate in water solutions or tissue is of greater interest, but is less well-known. On the basis of our present data, it appears to be somewhat smaller than the value in air, but further study is proposed Recent work[4] suggests that the value in tissue would be about 0.95 of the dose in air and in our present source.

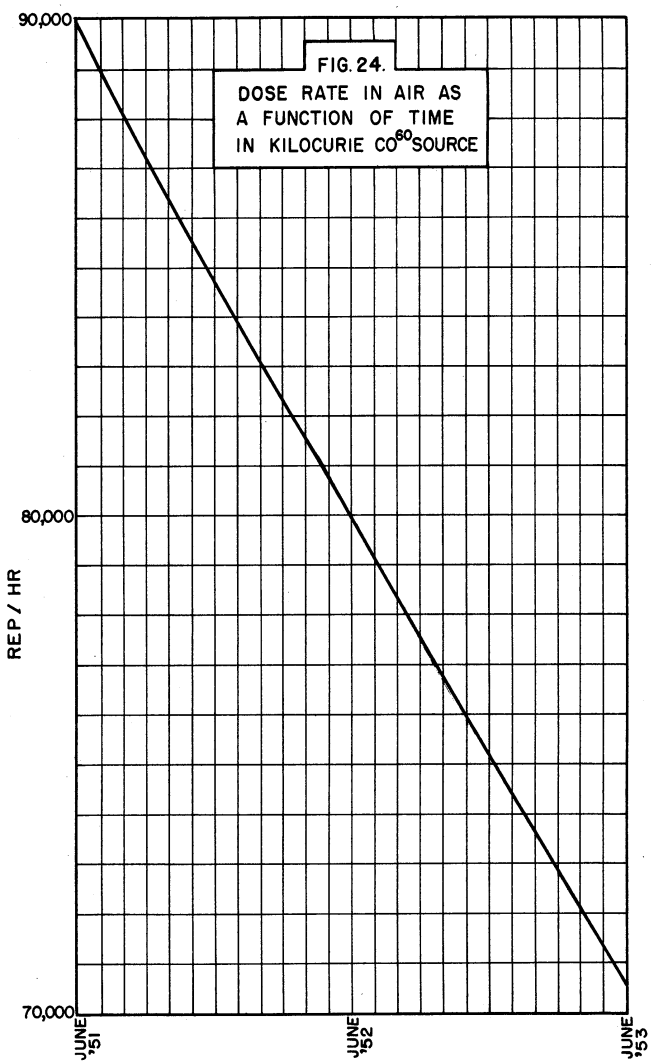


FIG. 24.
DOSE RATE IN AIR AS A FUNCTION OF TIME IN KILOCURIE CO^{60} SOURCE

C. 10-KILOCURIE COBALT-60 SOURCE

At present the experiments being conducted with gamma radiation are to a large extent limited by the availability of irradiation time, the low intensity of the gamma-radiation field, and the limited working space inside the

source (1-1/4 inches by 10 inches). The 1000-curie cobalt-60 source is being used 7 days a week, 24 hours a day, and there is a long waiting list. With the present source, sterilization experiments require, for the most part, irradiations of about 24 hours—almost a maximum for work where side reactions such as spoilage, etc., are competing with the sterilization. Even worse, the radiation available from this source is near the threshold for only the most advantageous chemical reactions and in the case of most reactions is far too small. Finally, the space limitation of the present source has made it excessively difficult or impossible to perform many chemical experiments and has put the food experimenters at a distinct disadvantage.

It was thus realized that the amount of research possible in our laboratories was definitely limited by our present gamma-ray source. The only way to remedy this situation was to obtain a larger, more versatile source of gamma radiation. Consideration of the types of experiments proposed showed the need for a source of about 10 kilocuries. For added versatility it was decided that this source should be in the form of a number of rods which could be set into a cylindrical pattern or into a layer pattern, whichever was desired.

A few comments regarding the efficiency of using gamma radiation sources might be mentioned. As the intensity of the gamma source is increased, it can be used more efficiently, as a greater percentage of the gamma field emmited can be used. For example, a 1-curie source is practically useless in promoting chemical reactions or sterilizing biological materials because the field is of such low intensity, whereas, a 1-kilocurie source can be used for these purposes, as has been demonstrated in this laboratory and others. However, a 10-kilocurie source is about 30 times as useful as a 1-kilocurie source. Other factors such as geometry, of course, must also be considered.

Project M943 received approval from the Atomic Energy Commission to purchase only 5 kilocuries for a new radiation source. This was considered a compromise and also was not considered adequate. The problem was presented to the Michigan Memorial-Phoenix Project with the request that consideration for assistance be given to this attempt to secure an adequate gamma source for the research program. Although the Michigan Memorial-Phoenix Project had previously refused such a request, reconsideration was given and as some new funds were available, 5 additional kilocuries were purchased on Phoenix Project 41.

The Brookhaven National Laboratories were approached about furnishing this irradiation, but scheduling difficulties prevented their making a delivery within a year. Furthermore, the present flux of the Brookhaven pile practically precludes the procurement of a source as large as 10-kilocuries within a reasonable period.

Hence, the possibility of obtaining this irradiation in the Chalk River pile was explored, and the many arrangements for the irradiation have been made

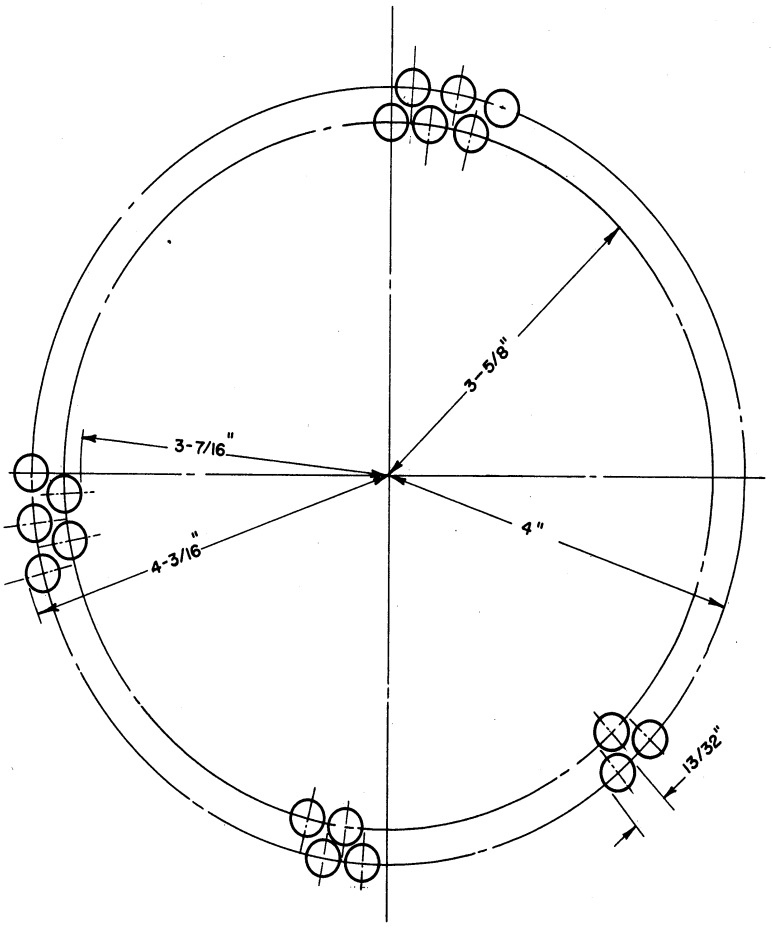
with the help of Mr. C. H. Hetherington of the Sales Department of Eldorado Mining and Refining (1944) Limited of Ottawa and the many Chalk River personnel involved.

The source will consist of 100 cobalt rods, 1/4 inch in diameter and 10 inches long. These high-purity cobalt rods were fabricated and machined to close tolerances by the Kulite Tungsten Company, 723 Sip Street, Union City, New Jersey (Mr. J. Kurtz, the sales manager, expedited our order). The rods were encapsulated in 1/8 inch nominal size 3SH18 aluminum pipe, 0.269 inch I.D., with a wall thickness of 0.068 inch and an O.D. of 0.405 inch. The aluminum pipe was obtained from the Central Steel and Wire Company of Chicago. Encapsulation was performed by the Eldorado Mining and Refining (1944) Limited of Ottawa, Ontario, Canada. The ends of the rods were welded with Alcan 2S welding rod, machined clean, and tested for leaks by dropping the complete rod into hot water.

The rods are to be inserted into the Chalk River reactor during July. Initial specifications called for the 10-kilocurie source to be formed in a radiation period of 4-1/2 months using the 100 rods. The curie was defined as the activity obtained from calculations based on a total amount of "nvt" exposure, with consideration given to irradiation efficiency. The further restriction was made that no one rod of the 100 irradiated was to be more than twice as active as any other rod — thus insuring a fairly even radiation field in the assembled source.

Tentative plans for the source arrangement, housing, and shielding have been made. For our purpose the best geometrical arrangement for the 100 rods seemed to be a double circle about 6-1/2 inches in I.D., as shown in Fig. 25. This configuration minimizes absorption of the gamma radiation by the cobalt rods and still furnishes high flux areas both inside and outside the circle.

Fig. 26 shows a tentative arrangement for irradiation chambers. The maximum usable space inside the source will be 6-1/2 inches in diameter and the overall flux inside is estimated to be about 4 times the flux, inside the present source, or about 320,000 rep/hr. Variation over the area should not exceed 15 per cent. The two rows of circles surrounding the source represent experimental ports of varying diameters with the indicated fluxes expressed as multiples of the flux of the present source. These values are based on extension of the geometrical estimate in the previous section. To insure uniform radiation, containers placed in the ports will be rotated by small synchronous motors geared down to a slow speed. The large (12-inch diameter) radiation port will be used for experiments requiring accessory equipment such as frozen food irradiations, etc. In addition, provision will be made for temperature control of the entire source surroundings and sufficient insulation will be included to make it possible to hold a reasonable temperature range.



PRELIMINARY ARRANGEMENT FOR 100 COBALT RODS
FIG. 25.

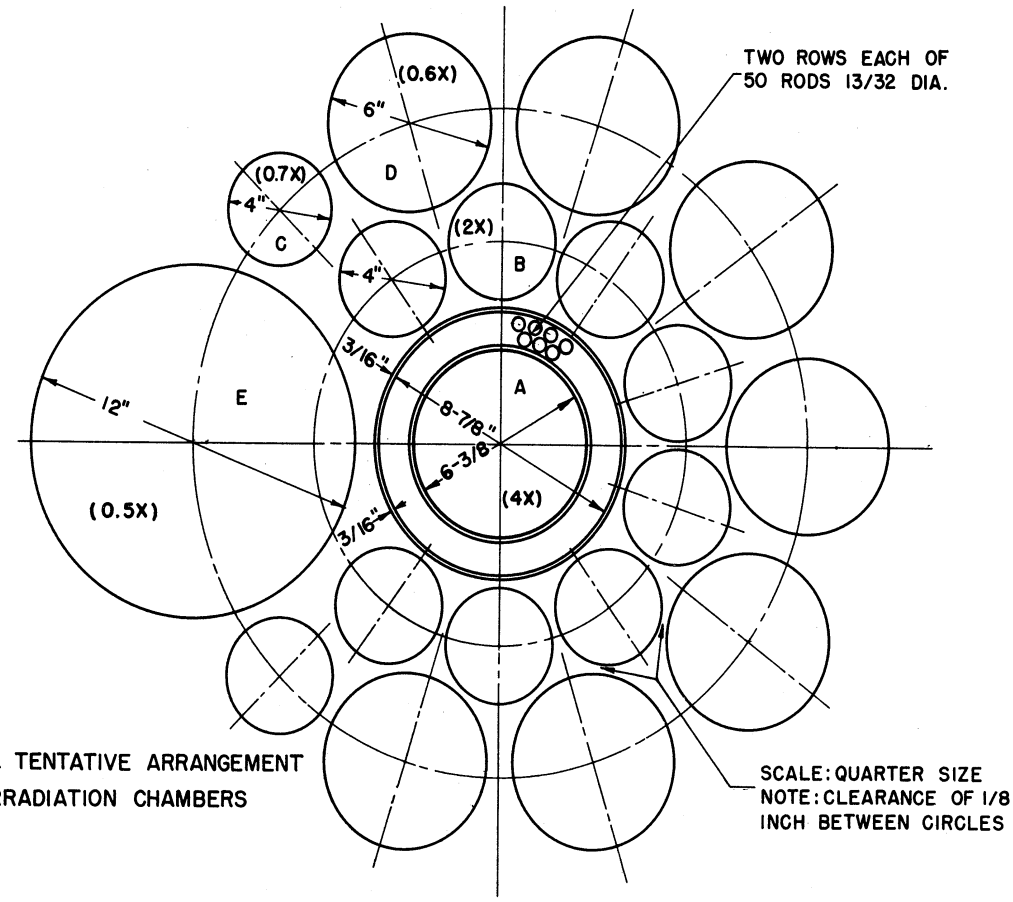


FIG. 26. TENTATIVE ARRANGEMENT
FOR IRRADIATION CHAMBERS

Fig. 27 shows a plot of the predicted dose rate (in air) as a function of distance from preliminary arrangement of the 100 cobalt rods. Manipulation of this large amount of material is, of course, a difficult problem. The details are being worked out at the present time and our solution will be presented in the next progress report.

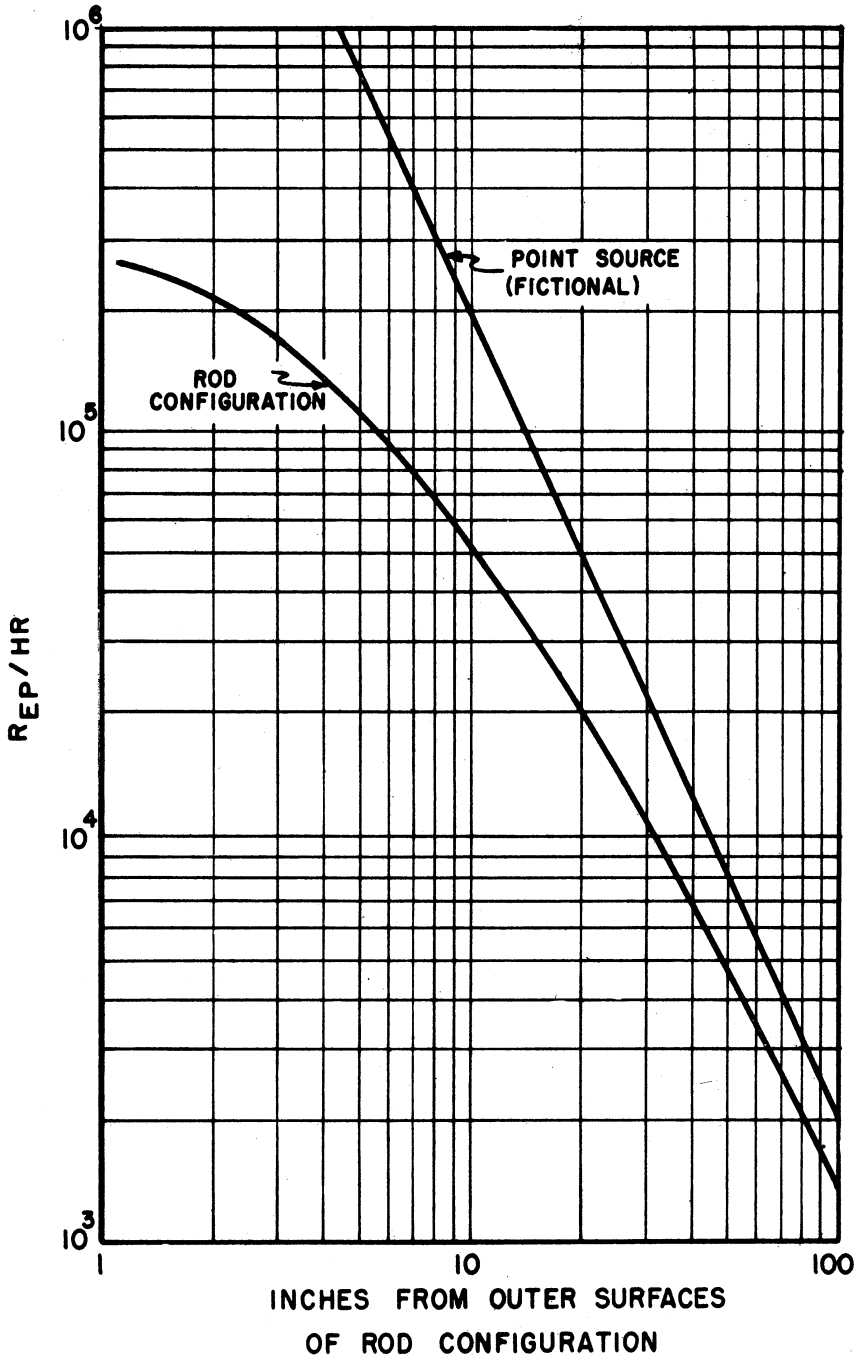


FIG. 27. PREDICTED DOSE RATES (IN AIR) AS A FUNCTION OF DISTANCE FROM PRELIMINARY ARRANGEMENT OF 100 COBALT RODS (10,000 CURIES)

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