# ENGINEERING RESEARCH INSTITUTE UNIVERSITY OF MICHIGAN ANN ARBOR

#### COO-124

#### PROGRESS REPORT 4

## UTILIZATION OF THE GROSS FISSION PRODUCTS (Unclassified)

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#### Project M943

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#### PREFACE

This report presents the results of research performed during the period of June 1, 1952, to December 31, 1952, on Project M943 of Engineering Research Institute, University of Michigan under AEC Contract No. AT(11-1)-162.

Results of research supported by other funds such as Michigan Memorial-Phoenix Projects 20, 28, 41, and 54 and results of cooperative studies made with the Michigan Department of Health also have been included in Part III of this report. It should be stressed that there is no relationship between these studies and Project M943 except that the personnel have a common interest in the effects of radiation and the possible uses of radioactive materials and use the same source of gamma radiation. The results of these other studies have been included because this information is believed to have significance in the possible utilization of the fission products. If the results of these other studies were not reported here, they ordinarily would not be brought to the attention of AEC and its contractors until some of the results appeared in the professional literature, perhaps a year or more later because of delays in preparation of manuscripts, reviews, and publications. To protect the authorship of research personnel of the Michigan Memorial-Phoenix Project, no part of their results is to be reproduced without permission of the respective authors.

#### ABSTRACT

This report describes the work accomplished since Progress Report 3 (COO-91), dated June 30, 1952, was published. The experimental studies described in this report may be summarized as follows:

The fundamental study of the effect of radiation on combustion engine performance (formerly subproject M943-A, now M943-2) has continued with the design and installation of equipment to study combustion under explosive conditions over a pressure range up to a maximum of several thousand pounds per square inch.

The study of the performance of combustion engines under the influence of radiation (formerly subproject M943-B, now M943-3) has been continued, additional tests have been performed with an experimental diesel engine using large amounts (up to 1200 curies) of palladium-109 in the form of seven palladium rods inserted into the combustion chamber. These tests indicated that radiation had little influence on the performance of the diesel engine, contrary to previous results in which lesser amounts of radiation seemed to produce an improvement in engine performance. Another test will be performed in an attempt to establish the reason for the difference in these results. In the experiments on the performance of jet burners, it was found that radiation had little or no influence on the performance of the burner. Another experiment will be made using an appreciably larger amount of radiation and a burner of different design.

The study of the effect of radiation on the promotion of chemical reactions (formerly subproject M943-C, now M943-4) has produced a number of interesting results. It has been found that gamma radiation readily promotes the chlorination of benzene by substitution to produce benzene hexachloride. Gamma radiation also promotes the chlorination of toluene. Ethylene can be polymerized under the influence of gamma radiation to produce a polymer having properties which differ from the conventional polymer produced by existing methods. Sulfurous acid can be converted to sulfuric acid by passing oxygen through sulfurous acid in the presence of gamma radiation. This reaction and the chlorination reactions will be investigated more thoroughly.

Cooperative research investigations were made with other projects. Blood fractions were irradiated for the Michigan Department of Health in an attempt to find a means for destroying the virus of serum hepatitus. Preliminary results were favorable and these studies are to be continued. Irradiation of meat contaminated with trichinae larvae was performed for Michigan Memorial-Phoenix Project 54. It was found that the cycle of trichinosis can be broken by a light dose of gamma radiation (about 12,000 rep). Irradiating processed meat for Phoenix Project 41 indicated that sodium nitrate and nitrite aid in protecting the flavor of meat exposed to radiation. Peroxide values were determined for irradiated fats and it was found that pork fats undergo the greatest increase in peroxide value. Peroxide values were not consistent with organoleptic tests. An animal feeding experiment will be initiated early in 1953 on Phoenix Project 41.

A new 10,000-curie cobalt-60 gamma source has been designed and the cobalt rods for this source are being irradiated in the NRX reactor at Chalk River. The new source will be used in a radiation room or cave and will be "shut off" by lowering the cobalt rods into a pit of water 16 feet deep. It is expected that this new source will be in operation early in 1953.

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#### PROGRESS REPORT 4

UTILIZATION OF THE GROSS FISSION PRODUCTS

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

A. SUBPROJECT M943-2 (FORMERLY M943-A), FUNDAMENTAL STUDY OF EFFECT OF RADIATION ON COMBUSTION ENGINE PERFORMANCE

#### Personnel:

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

#### 1. Review

The research of this subproject is directed toward a fundamental investigation of the effectiveness of using the radioactive fission products to influence the processes of gaseous combustion. The present experiment involves an investigation of the explosion limits of gaseous mixtures. In this experiment a mixture of combustible gas, oxygen, and nitrogen will be "sparked" in a pressure vessel or "bomb" at pressures of the order of 20 atmospheres. The pressure of nitrogen will be varied until the mixture just fails to explode. In this manner the explosive limit for a given pressure of combustible gas and oxygen will be determined. This procedure will be repeated for different initial pressures of combustible gas and oxygen, so that an explosion limit curve, represented schematically in Fig. 1, can be plotted.

Once a normal explosion limit curve has been established for the apparatus, the experiments will be repeated in the presence of an intense beta source.

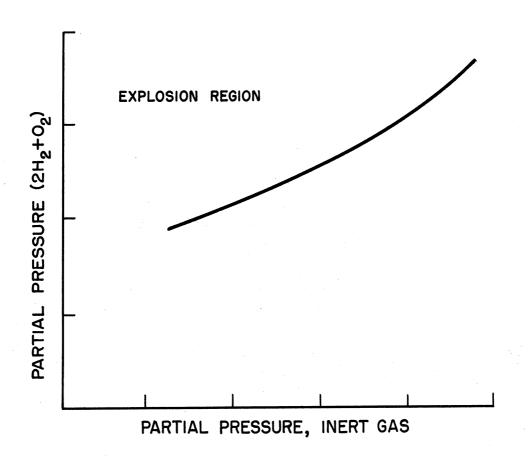


Fig. 1. Explosion limit curve.

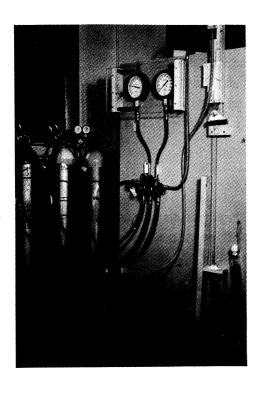
The explosion limits are determined by the rates of production and removal of chain carriers within the gas. It is expected that the ionization created by high-intensity beta radiation will increase the concentration of chain carriers. Positive results should then appear as a shift of the explosion limit curve as a result of the increased concentration of carriers.

#### 2. Experimental Equipment

During the period covered by this progress report, work has progressed on the installation of the explosion system described in Progress Report 3.

Most of the components have been received from suppliers or have been fabricated in University shops. The assembly of these parts is almost completed, and at present the equipment is being tested. The pressure gauges are being calibrated in the Sohma Precision Instruments Laboratory of the University Chemical Engineering Department.

The flow sheet of the system is essentially the same as that shown in Fig. 7 of Progress Report 3 except that minor modifications have been introduced. Figure 2 shows the instrument pannel with manifold, gauges, and feed lines leading to the gas cylinders. Figure 3 shows the Aminco high-pressure vessel and connections.



feed system.

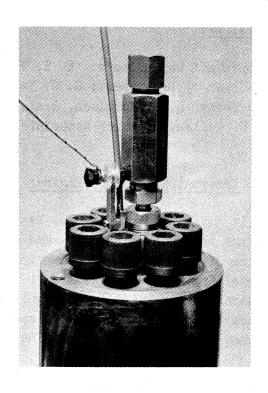


Fig. 2. Instrument panel and Fig. 3. Head of reaction vessel showing electrical connection, feed line, and blowoff assembly.

An ignition system has also been designed and built. It consists of a standard automotive storage battery, coil, and breaker gap turned by an induction motor driving a V-belt. The spark gap is a standard automotive spark plug mounted on the inside of the compression cap of the reaction vessel.

A filter for the exhaust must be designed to retain any radioactive material carried by the exhaust gases. The original plan was to exhaust from the rupture disc assembly directly into the laboratory exhaust manifold, but this plan was discarded as not feasible because of the danger of damaging the manifold system. The filter for the exhaust will not be required for the earlier experiments, which are to be performed without radioactive materials.

Barring unforeseen difficulties, it is believed this experimental equipment will be in operation early in 1953.

B. SUBPROJECT M943-3 (FORMERLY 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, Assistant Professor of Aeronautical Engineering. R. E. Cullen, Research Associate; M. E. Gluckstein, Research Associate.

#### 1. Internal Combustion Engines

- <u>a. Introduction.</u> Previous progress reports have outlined the nature of the research being performed on the effects of beta radiation on the performance of reciprocating combustion engines. To date, work has been limited to diesel engines. The results of the first test, reported in Progress Report 3, indicated a decrease in the ignition delay period when the combustion took place in the presence of beta radiation. In this report the results of a second test on the diesel engine are presented.
- <u>b.</u> <u>Description of Test.</u> On August 12-15, 1952, a second test was conducted with a beta radiation source in the combustion chamber of the one-cylinder CFR diesel test engine. The source of radiation was seven palladium rods, 1/4 inch in diameter by 1-1/4 inch long, which projected into the combustion chamber from the compression plug, as shown in Fig. 4. The back of the compression plug was water-cooled in order to keep the temperature of the palladium rods within safe limits.

These rods were placed in the NRX reactor at Chalk River for two weeks; they were removed from the reactor at 8 a.m. on August 11 and taken immediately to Ann arbor by car. At 1 a.m. on August 12, the first test was conducted. Identical runs were conducted on the afternoons of August 12, 13, and 14, in order to determine any dependence on beta-ray intensity. The intensity of radiation decreased by a factor of 14 between the first and last run.

In each run, readings were taken for two compression ratios and two fuel rates, giving four different operating conditions. The following data were obtained for each operating condition:

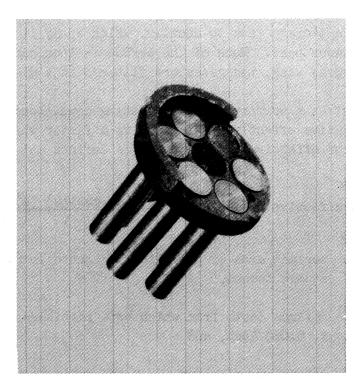


Fig. 4. Radioactive palladium rods and holder

- (1) pressure-time relations determined by photographing the trace on an oscilloscope with a catenary-diaphragm-type pickup;
- (2) specific fuel consumption; and
- (3) minimum firing compression ratio.

In the experimental tests it was difficult to duplicate exactly all the operating variables from one run to the next. Of these, the most difficult variable to regulate was the injection timing. The injection is regulated by adjusting a micrometer screw, which changes the point in the cycle at which the injection of fuel takes place. However, the point in the cycle at which injection occurs is measured by a flashing light arrangement operating on the flywheel. This measurement involves another rather sensitive adjustment. The net result is that there is sometimes a question as to whether or not a measurement has been duplicated exactly. The question of timing accuracy arises from the fact that from one day to the next, for the same micrometer setting, the flashing light would indicate a different time of injection. The best adjustment possible was made in each case, but there remains the possibility of some variation in the time of injection from one run to another.

One change was made on the filter equipment prior to this test. A new exhaust filter system was devised, which consists of a spray washer. Water

is circulated from a storage tank to nozzles, which spray it on screens through which the exhaust gases pass. Much of the carbon is thus carried away by the water. After the spray wash, the gases are filtered in a glass-wool filter.

Table I gives a summary of the operating conditions for the four series of tests. The "relative effective radiation" is a factor which indicates the relative intensity of effective radiation and is defined as:

relative effective radiation = 
$$\frac{\text{(total curies) (a) (1)}}{v}$$

where

a = surface area of palladium in square inches = 6.19 square inches,

v = volume of palladium = 0.457 cubic inch.

TABLE I

SUMMARY OF OPERATING CONDITIONS
FOR TESTS USING PALLADIUM RODS IN DIESEL ENGINE

| Run<br>No. | Date    | Time of<br>Start<br>of Run | Barometer, in. Hg | Total Curies at Start of Run | Relative<br>Effective<br>Radiation |
|------------|---------|----------------------------|-------------------|------------------------------|------------------------------------|
| I          | Aug. 12 | 1:40 a.m.                  | 29.15             | 740                          | 130.8                              |
| II         | Aug. 12 | 2:00 p.m.                  | 29.25             | 410                          | 72.3                               |
| III        | Aug. 13 | 3:20 p.m.                  | 29.27             | 133                          | 23.4                               |
| IV         | Aug. 14 | 4:30 p.m.                  | 29.19             | 52                           | 9.2                                |

c. Summary of Results. The most significant aspect of the pressure-time measurements is the crank angle at which ignition occurs. Assuming that injection occurs at 13° BTDC (as pointed out previously, this was difficult to regulate accurately), the crank angle at which ignition occurs is an important parameter. For a fixed time of injection, the earlier the time of ignition the shorter the ignition delay period, which is in general beneficial in diesel engines. Table II gives the ignition crank angle for each test. Also listed in this table of results is the specific fuel consumption. These results are presented with identical operating conditions in chronological order.

TABLE II

SUMMARY OF RESULTS OF TESTS
USING PALLADIUM RODS IN DIESEL ENGINE

| Operating Conditions Specific |                    |      |                   |  |                              |
|-------------------------------|--------------------|------|-------------------|--|------------------------------|
| Compression<br>Ratio          | Fuel Rate, ccm/min | Rpm  | Run<br><u>No.</u> | Ignition<br>Crank Angle                  | Consumption, lbs fuel/hp-hr  |
| 15 to 1                       | 15                 | 1100 | I<br>III<br>VI    | 5° BTDC* 2° BTDC 4° ATDC** 4° ATDC       | 1.40<br>1.26<br>1.24<br>1.30 |
| 12.7 to 1                     | 15                 | 1100 | I<br>III<br>V     | 8° ATDC<br>6° ATDC<br>6° ATDC            | 1.45<br>1.53<br>1.41<br>1.36 |
| 12.7 to 1                     | 23                 | 1100 | I<br>III<br>IV    | 9° ATDC<br>7° ATDC<br>9° ATDC<br>8° ATDC | .862<br>.855<br>.844<br>.873 |
| 15 to 1                       | 23                 | 1100 | I<br>III<br>IV    | 6° ATDC<br>4° ATDC<br>5° ATDC<br>4° ATDC | .789<br>.824<br>.786<br>.815 |

<sup>\*</sup> BTDC - before top dead center

The minimum compression ratio for firing is given below. This test is conducted by motoring the engine for 55 seconds with no fuel injection, followed by a 5-second period in which fuel is injected. The lowest compression ratio at which the engine will fire during the 5-second fuel injection period is considered to be the minimum firing compression ratio.

<sup>\*\*</sup> ATDC - after top dead center

| Run No. | Minimum Firing<br>Compression Ratio |  |  |
|---------|-------------------------------------|--|--|
|         | ·                                   |  |  |
| I       | 12.23                               |  |  |
| II      | 12.37                               |  |  |
| III     | 12.37                               |  |  |
| IV      | 12.37                               |  |  |

<u>d.</u> <u>Discussion of Results.</u> Examination of these results shows no appreciable affects of the radiation. As pointed out previously, it was difficult to maintain all the operating conditions exactly, which accounts for the variations in results from one day to the next. However, any large-magnitude effects would have been observable in spite of these variations.

The lack of detectable results is somewhat surprising in view of the improved combustion noted on the first test, in February, 1952. This arrangement of palladium rods was chosen in order to obtain a larger exposed area of palladium, and thus permit reduction of the size of the palladium rods so as to accommodate the NRX reactor at Chalk River. By this means a relative effective radiation at the start of this test of 130.8 was obtained, as compared to 5.7 at the start of the February test. It was anticipated that this increased intensity of radiation would produce a greater effect, but the results indicate no effect of beta radiation. It can only be concluded that either the first or the second test gave an inaccurate answer to the question of the effect of beta radiation, and another test will be necessary to establish a positive answer. In speculating on possible reasons why this test might have failed to indicate the effects of beta radiation on combustion, the following observations made during the test are of interest.

- (1) The palladium rods projected into the combustion chamber and certainly affected the spray pattern, the mixing of the air and fuel, and the subsequent vaporization of the fuel. It is possible that these factors limited the ignition of the fuel rather than the usual ignition delay period. It is this ignition delay period which the beta radiation would be most likely to influence.
- (2) After a short time of operation, the palladium rods were covered with a fairly heavy deposit of carbon (which tends to verify (1) above), which might have decreased appreciably the number of beta particles leaving the surface.

Both these factors have been anticipated, but it was felt that the greater intensity of radiation which this design afforded would offset them. However, it is possible that the combination of these effects cancelled the effects of the radiation.

It is also of interest that in this experiment an appreciable amount of radioactive material was collected in the exhaust filter system. This was probably the result of incomplete combustion caused by poor mixing, so that the exhaust contained much unburned carbon, which was collected on the filters. Some of this carbon may have flaked off the rods, carrying radioactive palladium with it. These results emphasize the importance of a good exhaust filtering system.

- $\underline{e}$ . Future Experiments. It is considered advisable to conduct at least one more experiment on this diesel engine, incorporating two important changes.
  - (1) Identical pieces of palladium will be available, one irradiated and the other in its normal condition. The tests will be run on the same day, thus eliminating the effects of operating conditions varying from one day to the next. In this way each test will be compared with a cold run under exactly the same operating conditions.
  - (2) The palladium will be in the form of a cylinder around the outside of the combustion chamber, thus leaving the spray pattern and mixing unaffected. There will also be a disc of palladium on the end of the compression plug. Thus, the entire combustion chamber will be surrounded by a radiating surface. The larger size pieces involved will result in a lower intensity of radiation (because they cannot be accommodated in the Chalk River Pile), but this seems the best procedure to avoid disturbing the injection and mixing and yet maintain a large radiating surface. This plan also permits the longest possible mean free path of the beta particle, which seems to be desirable from the point of view of maximum ionization in the chamber.

The palladium for this test has been ordered, and the test will be undertaken as soon as it arrives.

A spark-ignition CFR engine is being readied for a test similar to that performed on diesel CFR engines. The engine is being equipped with a surge chamber for inlet and discharge, in order to achieve more uniform operating conditions. These tests will be undertaken as soon as work on the engine is completed.

#### 2. Jet Engines

a. Introduction. The investigation reported in Progress Reports 2 and 3 has been continued. Analysis of these results has shown that moderately high levels of activity ( $\leq$ 100 curies) have no appreciable effect on normal flame speed or blowoff velocity. This has been attributed to two major causes: (1) insufficient activity and (2) poor geometry for absorption of the radiated energy by the gas stream. The work on flame speeds and blowoff of flames from a spherical flameholder has been discontinued.

The major effort has been directed to the design and construction of a miniature ram-jet burner. This burner is equipped with a removable gold liner and a movable flameholder. The liner will be irradiated with neutrons to give a source of approximately 5000 curies of gold-198. Runs will be made with various fuels and at various simulated altitudes.

<u>b. Theory.</u> The interaction of beta radiation and the gas stream gives rise to molecular species other than those initially present. These species include free radicals, ions, and possibly new compounds whose formation is catalyzed by the radiation. If the radiation is to have any effect on the combustion process, the number and kind of specie introduced must be appropriate to catalyze and maintain reactions similar to those found in the normal combustion process. Theoretical discussions of the mechanism of combustion usually are highly idealized and cannot be applied to the systems being considered; however, some of the theory employed in the design of this burner can be presented qualitatively.

If a beta particle with energy of 1 mev is considered, its range in air at 1 atmosphere is approximately 350 cm. Since the fuel-air mixtures being used do not differ appreciably in density from air, this value will be used as the range of the electron in the fuel-air mixture.

Since the formation of one ion pair corresponds to the absorption of approximately 35 ev, a single 1-mev beta particle traveling its entire range will give rise to 31,500 ion pairs or 90 ion pairs per cm of path. Thus 1 curie of activity yields very nearly 3.3 x 10" ion pairs per cm per second. Since the ions so formed have a definite life expectancy before they are destroyed by a combination or collision, it is necessary to devise a system whereby as many ions as possible will reach the reaction zone. Such a design, however, must be consistent with the geometry and flow characteristics of the system being considered.

For flame-speed studies, a radioactive grid located immediately preceding the combustion zone would fulfill the required conditions; however, due

to other difficulties, to be discussed in the succeding section, such an investigation is not feasible at this time.

In the design of a conventional ram-jet burner a drag-producing device, known as a flameholder, is placed in the gas stream. Combustion then takes place in the turbulent zone downstream of the flameholder. This turbulence causes the reaction to take place in a volume of reactants, rather than on a surface of reaction, as in the Bunsen flame case. Some experimental work performed at the University of Michigan Aircraft Propulsion Laboratory on cylindrical ram-jet combustion chambers with ceramic walls at 3200°F emitting thermal energy by a regenerative process has shown that this system gives more efficient combustion. 1

It is therefore felt that if the radiation source surrounds the reaction volume, any significant effect on the combustion process can be ascertained. This type of design would permit an efficient use of the electrons emitted by the source. Briefly, the reactants would be irradiated before and during the combustion process. This geometry offers a multiple path for reflection of the initially emitted electrons and therefore permits a large number of the ion pairs formed to reach the reaction volume. Under the flow condition and configuration usually found in ram-jet burners, the proportion of ion pairs entering the reaction zone has been roughly calculated as 10-7 mole of active specie per mole of reactant per curie of activity. Thus a source of 1000 curies would give  $10^{-l_1}$  mole per mole. The assumptions made in calculating these values are conservative. They are as follows:

- (1) Only one-step ion-formation reactions, i.e., no chain reactions, are considered.
- (2) Eighty per cent of the electrons reaching a wall are reflected, i.e., if 100 electrons are initially emitted and reach a wall, 80 are reflected on the first pass, 64 on the second pass, etc.
- (3) The energy lost by inelastic collisions is of the same order of magnitude as the energy absorbed in passing through the gas.

In order to ascertain the effect of the radiation on the combustion processes, it was decided to utilize the measurement of the combustion chamber parameter  $S_a$ , or specific air impulse.1,2,3

It is the purpose of a ram-jet combustion chamber to produce thrust by adding heat to the internal air flow, thereby increasing its momentum. For simplicity let us assume a combustion chamber that is essentially a parallelwalled duct exhausting into the atmosphere. The fuel is mixed prior to

Station 1, so that the mixture is homogeneous at Station 1, where combustion begins. The products of combustion are exhausted at Station 2. Writing the equation for conservation of momentum across the combustion chamber for one-dimensional flow,

$$p_1A_1 + \rho_1A_1V_1^2 - D_f = p_2A_2 + \rho_2A_2V_2^2,$$
 (1)

where  $D_{\mathbf{f}}$  is the loss due to flameholder drag and skin friction.

Since the units of the above equation are in pounds force, we can introduce the concept of stream thrust, F. The stream thrust at Station 1, minus the losses due to friction may be equated to the stream thrust at Station 2, or

$$F_1 - D_f = F_2 . (2)$$

Using the equation of state and writing in terms of the Mach number, M, Eq (1) becomes

$$A_1p_1 (1 + \{M_1^2\}) - D_f = A_2p_2 (1 + \{M_2^2\}) = F_2.$$
 (3)

The mass flow W  $\,=\,$   $\rho AV$  can be written as follows, using as the speed of sound

$$C = \sqrt{\chi} Rg_{c}T:$$

$$W = \frac{pM \chi Af_{c}}{C}.$$
(4)

Solving for Ap at Station 2 and noting that the mass flow W =  $W_a$  (1 + f), where  $W_a$  is the air flow and f the fuel-air ratio:

$$A_{2}P_{2} = \frac{W_{a} (1 + f) C_{2}}{M_{2} \chi_{2} g_{c}}$$
 (5)

From the conservation of energy and the equation of state

$$C_2 = \frac{C_{s_2}}{\left[1 + (\sqrt{-M_2^2})/2\right]^{1/2}} \tag{6}$$

where  $C_{\rm S2}$  is the velocity of sound corresponding to the stagnation temperature at Station 2.

Combining Eq (3), (5), and (6), the following relation is obtained:

$$\frac{F_2}{W_a} = \begin{bmatrix} \frac{C_{s_2} (1 + f)}{\sqrt{g_c}} \end{bmatrix} \begin{bmatrix} \frac{1 + \sqrt{M_2^2}}{M_2 + \sqrt{-1} M_2^{2/1/2}} \end{bmatrix}.$$
 (7)

Noting that the quantity in the right-hand brackets of Eq (7) would normalize to 1 at M = 1 if divided by  $\sqrt{2(\zeta+1)}$ , Eq (7) is multiplied by

$$\frac{\sqrt{2(f+1)}}{\sqrt{2(f+1)}}$$

to obtain the following:

$$\frac{F_2}{W_a} = \frac{\begin{bmatrix} c_{s_2} (1+f) \sqrt{2(f+1)} \\ f_{s_2} \end{bmatrix}}{\sqrt{g_c}} \frac{1+\sqrt{M_2^2}}{M_2 \sqrt{2(f+1)} \left[1+\frac{f-1}{2} M_2^2\right]}$$
(8)

or defining two terms in the brackets,

$$\frac{F_2}{W_a} = \left[S_a\right] \left[\phi(M)\right]. \tag{9}$$

The quotient of the stream thrust and the air flow rate then is dependent on two parameters:  $S_a$ , or the specific air impulse, which is a function of the stagnation temperature at the exit of the burner (Station 2), and  $\phi(M)$ , which is a function of the exit Mach number. The function  $\phi(M)$  is plotted against Mach number in Fig. 5.  $S_a$ , then, is a criterion of the amount of heat added to the stream, i.e., the completeness of combustion for a given fuel and fuel-air mixture. Furthermore,  $\phi(M)$  is a parameter describing the flow characteristics of the burner, or how completely the heat of combustion is converted to momentum of the exhaust gases.

Solving Eq (9) for  $S_a$ ,

$$S_{a} = \frac{F_{2}}{W_{a} \phi(M)} = \frac{A_{2}p_{2} + \frac{WV_{2}}{g_{2}}}{W_{a} \phi M}$$

$$= \frac{A_{2}p_{2} + \frac{GA_{2}V_{2}}{g_{c}}}{\phi(M) \frac{GA_{2}}{1 + f}}$$
(10)

From the foregoing considerations,  $S_a$  is then a combustion-chamber parameter that can be compared with the theoretical maximum value of  $S_a$  to give a realistic combustion efficiency, indicating how completely the combustion chamber is converting chemical energy into kinetic energy of the exhaust gases. Since the combustion chamber is primarily a thrust-producing device, the calculation of combustion efficiency should reflect any condition which detracts from its ultimate performance.

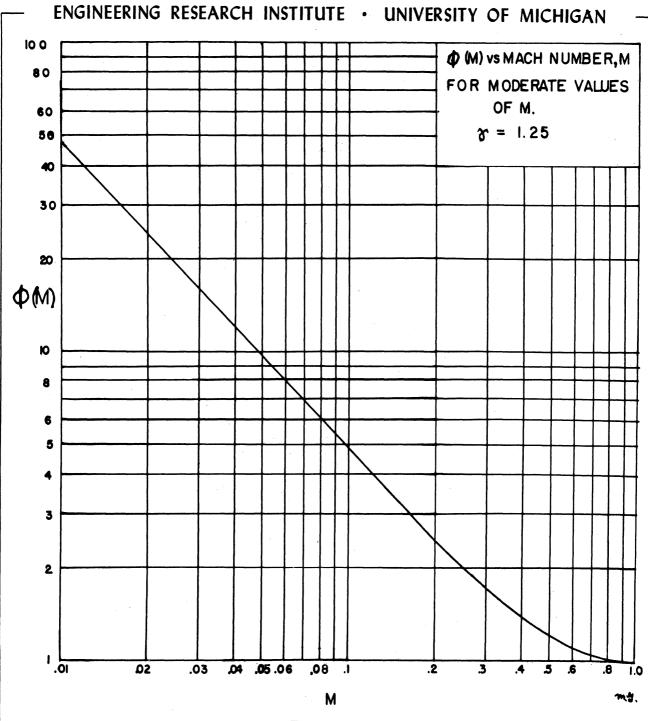


Fig. 5

The combustion-chamber parameter  $S_a$  as defined by Eq (10) then may be computed from relatively simple experimental measurements. From the measurement of the inlet stream thrust,  $F_1$ , at Station 1 and the measurement of the combustion-chamber drag due to the flameholders and skin friction, the exit stream thrust,  $F_2$ , the numerator of Eq (10) may be calculated. Then, by means of the mass continuity and state equations, the exit Mach number,  $M_2$  and thus  $\phi(M)$  may be obtained.  $S_a$  can then be determined and compared with the theoretical maximum  $S_a$  to compute the combustion-chamber efficiency for any particular condition of burner operation.

#### List of Symbols

A = Combustion-chamber cross-sectional area,  $ft^2$ 

C = Velocity of sound, ft/sec

 $C_{\rm S}$  = Velocity of sound corresponding to the stagnation temperature

 $D_{f}$  = Combustion-chamber drag, lbs force

f = Fuel-air ratio, lbs fuel/lb air

F = Stream thrust, lbs force
G = Mass velocity, lbs/sec-ft<sup>2</sup>

 $g_c = Acceleration of gravity, ft/sec^2$ 

M = Mach number, M = V/C

m = Molecular weight

p = Static pressure, lbs/ft<sup>2</sup>

 $R = Gas constant = R_{11}/m$ 

 $R_U$  = Universal gas constant in gas law, pv = n  $R_U$  T

 $S_a = Specific air impulse, lbs/(lb air)(sec)$ 

V = Velocity, ft/sec

W = Mass flow, lbs/sec

Wa = Air flow rate, lbs/sec

 $\gamma$  = Ratio of specific heats

 $\rho$  = Density, lbs/ft<sup>3</sup>

 $\emptyset(M)$  = Mach number function

<u>c</u>. <u>Results</u>. The principal effort during the period covered by this report has been devoted to the design of an experimental ram-jet burner. In developing this design, it was necessary to collect data on the characteristics of these burners under various operating conditions and geometric configurations.

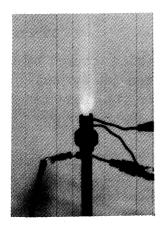
Some effort was devoted to a possible design of equipment to study flame speeds under the influence of beta radiation. It was demonstrated that a source of radiation in the form of a fine gauze located upstream from the reaction zone would be more efficient than the method described in Progress Reports 2 and 3. Gold was selected to be the active material, since it has a longer half-life and a larger absorption cross section than palladium. In view of the previous results, it was felt that a source of approximately 1000 curies should be used. The choice of this type of source required that suitable handling techniques be developed so that the actual determination of the flame speed is not impeded.

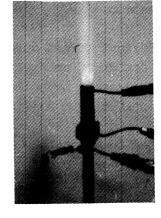
The experimental work was of a qualitative nature, involving the evaluation of various sizes of gauze that could be used. It is necessary that any gauze introduced upstream not alter the symmetry of the flame. This requires that the gauze be not less than 80 mesh and that its surface be free of imperfections, creases, tears, and obstructions. Utmost care must be exercised in the

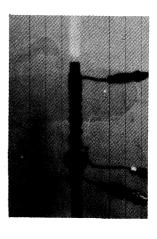
placement of the gauze in the nozzles or the flame will not be symmetrical. It was determined from examination of the data collected with the various gauzes that these two requirements could not be economically satisfied simultaneously; therefore, work on flame speeds has been discontinued.

To design a ram-jet burner for use in determining the effect of radiation on combustion, it was necessary first to examine the performance of models under different operating conditions. Such models have been built and tested, and these results utilized in the design of the test burner.

The model burner consists of a straight length of 3/8-inch schedule 40 pipe equipped with a flange to accommodate removable burner sections. A flat cross flameholder of approximately 30 per cent blockage is used. The reaction is initiated by a miniature spark plug located immediately upstream of the flame holder. A pressure tap is located diametrically from the spark plug. The burner sections are 0.500 inch in diameter and from 1.5 to 6 inches long. Figure 6 (a, b, and c) shows this model in operation with three different-length burner sections. Water is passed through coils surrounding the burner to cool the walls. The mass velocity, G, and the fuel-air ratio, F/A, are constant for each of the burner lengths shown.







а

L = 1.5''  $G = 1.4 lb/ft^2/sec$ 

F/A = 0.100Fuel = propane L = 3.0"
G =  $1.4 \text{ lb/ft}^2/\text{sec}$ F/A = 0.100

Fuel = propane

L = 6.0"
G =  $1.4 \text{ lb/ft}^2/\text{sec}$ F/A = 0.100

Fuel = propane

Fig. 6. Photographs of propane-air flame.

In making each run, the fuel is ignited at the minimum flow rate possible and the flow rate is then increased until the flame blows out or the burning becomes "rough". By "rough" burning is meant unstable, noisy, or irregular burning. Table III presents data from runs 21 and 25 at the condition just preceding blowoff or rough burning. The values of rotameter pressure indicated with an asterisk (\*) were read directly with a single-leg pot-type

TABLE III

TYPICAL DATA FOR JET BURNER TEST

| Run Number:  | 21      | 25                |
|--|---------|-------------------|
| Date:  | 10/2/52 | 10/2/52           |
| Fuel:  | Propane | Propane           |
| (1) Burner diameter, in.                             | •500    | .500              |
| (2) Burner length, in.                               | 6.0     | 1.5               |
| (3) Fuel/air ratio                                   | 0.067   | 0.067             |
| (4) Barometer, in. Hg (uncorrected)                  | 29.35   | 29.35             |
| (5) Ambient temperature, °C                          | 22      | 22                |
| (6) Rotameter reading                                | 146     | 149               |
| (7) Rotameter temperature, °C                        | 23.4    | 23.5              |
| (8) Rotameter pressure, psig                         |         |                   |
| (9) (8) $\times$ 2.036 = Rotameter Pressure, in. Hg  |         |                   |
| (10) Barometer, in. Hg (corrected)                   | 29.23   | 29.23             |
| (11) $(10)$ + $(9)$ = Rotameter pressure in.         |         |                   |
| Hg abs.  | 31.9*   | 32.5 <del>*</del> |
| (12) Rotameter temperature °R;                       |         |                   |
| $(7) + 273 \times 1.8$                               | 533.5   | 533.7             |
| (13) K/(12) K = 1.3571 F/A = .067                    |         |                   |
| 1.3626 .080  |         |                   |
| 1.3708 .100  | .002544 | .002543           |
| (14) Rotameter $\rho = (11) \times (13)$ ; $1b/ft^3$ | .08115  | .08252            |
| $(15) \ \sqrt{(14)} + \sqrt{\rho}$                   | .2849   | .2873             |
| (16) 3.31 x (15)                                     | •9430   | .9510             |
| (17) CFM of 0.073 air (rotameter calibration         | 2.81    | 2.89              |
| (18) (17) x (16) = $G lbs/sec-ft^2$                  | 2.650   | 2.748             |
| (19) P <sub>e</sub> (exit pressure in. Hg abs.)      | 29.23   | 29.23             |
| (20) $\Delta P_{\text{cold}}$ (in. $CCl_4$ )         | •55     | .505              |
| (21) $\Delta P_{\text{hot}}$ (in. CCl <sub>4</sub> ) | 3.11    | 1.26              |
|  |         |                   |

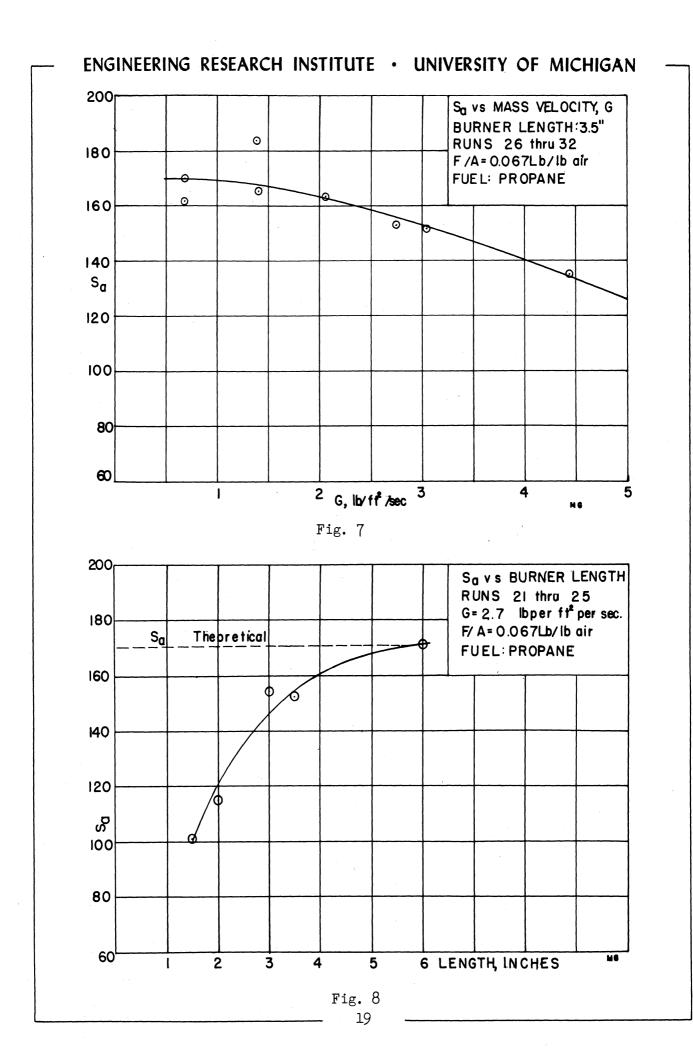
manometer. The value K in row 13 is a modified gas constant, corrected for the change in molecular weight due to the addition of fuel to the air. Since the rotameters were calibrated in CFM of a density of 0.073 lb/ft<sup>3</sup>, these readings must be corrected to CFM of fuel plus air at the rotameter conditions. Generally, for rotameters operating at densities not very different from that for which they

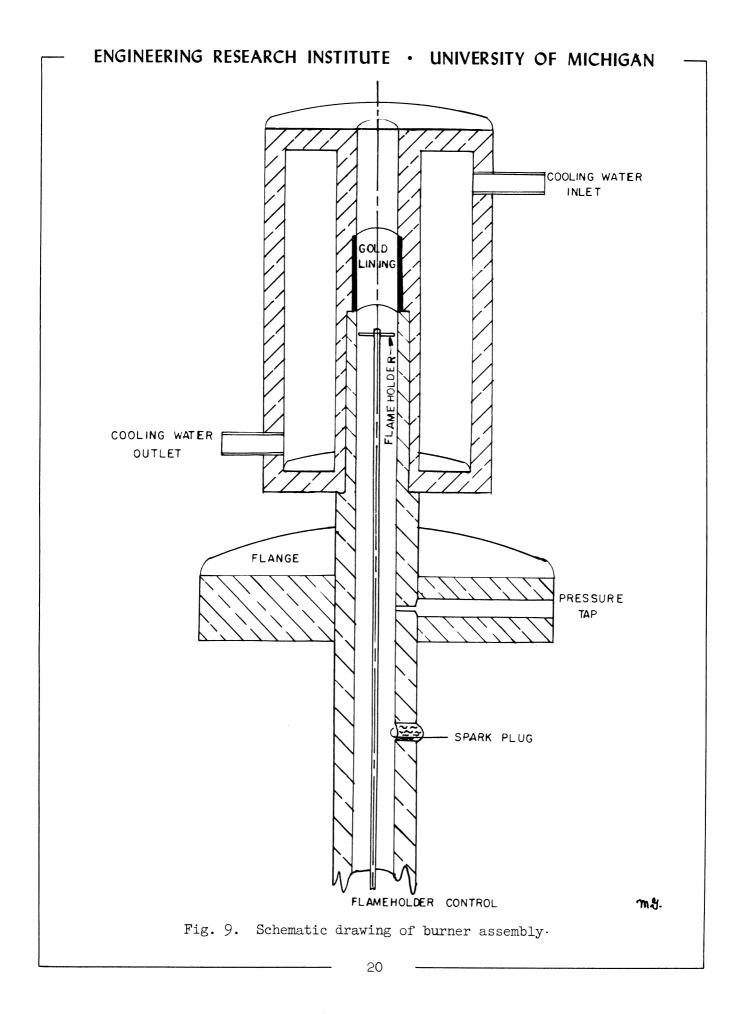
were calibrated, the corrected flow rate is equal to the calibration value times the square root of the ratio of the densities; i.e.,  $Q_2 = Q_1 \sqrt{\rho_2/\rho_1}$ . The value 3.31 in row 16 is a factor to convert CFM to G, the mass velocity.

The exit pressure,  $P_e$ , (row 19) is taken as the ambient atmospheric pressure. The values in rows 20 and 21 are values of pressure drop which enable the observer to determine the pressure drop due to burning; i.e.,  $\Delta P_{burning} = \Delta P_{hot} - \Delta P_{cold}$ . The value  $\Delta P_{cold}$  is the frictional drag term. By means of a master calculation sheet involving 63 steps in the calculation, the value of  $S_a$  can be calculated from the data and derived terms in Table III. The data from 32 runs have been calculated using propane fuel at fuel-air ratios of 0.067, 0.080 and 0.100. The results of selected runs are plotted in Figs. 7 and 8. In addition, some runs were made with ethylene-air mixtures to determine qualitatively the behavior of the various burners with this fuel.

After an analysis was made of the data obtained, a burner length of 3 inches was selected. (The length as defined here is from the flameholder to the exit point.) The radioactive source will be in the form of a gold insert to be placed in a groove in the walls of the burner tube. The flameholder position will be variable, so that it may be located at any position relative to the gold liner. Two interchangeable heads are provided, so that when the flameholder is located at the top or bottom of the gold liner, the distance to the exit may be kept at 3 inches. Figure 9 is a cross-sectional schematic drawing of the burner assembly. The completed assembly is shown in Fig. 10 and an exploded view in Fig. 11.

Due to the nature of the radiation emitted by the source (0.98 mev beta and 0.41 mev gamma), the problems of handling and shielding are more difficult than in the previous work. The entire burner will be mounted in the bottom of a vertical tank 30 inches in diameter and 6 feet deep. The tank will be shielded by sandbags. Prior to assembly the tank will be filled with water, and then the gold liner will be inserted and the head put in place. The tank will then be sealed and drained. All subsequent operations will then be conducted remotely. Water from the tank will be stored in a closed 220-gallon tank until it can be examined for contamination. If no contamination is found, the water will be discharged to the sewer. Provision will be made for repumping the water into the burner tank. Figure 12 is a schematic diagram of this system. Exhausted gases will pass through a Cambridge absolute filter before being discharged. The entire system will be connected to a vacuum pump, so that simulated altitude runs may be made.





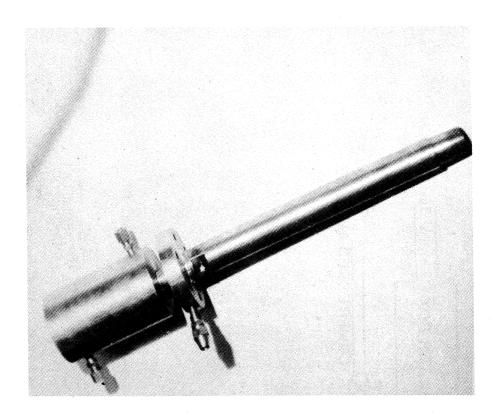


Fig. 10. Photograph of burner assembly.

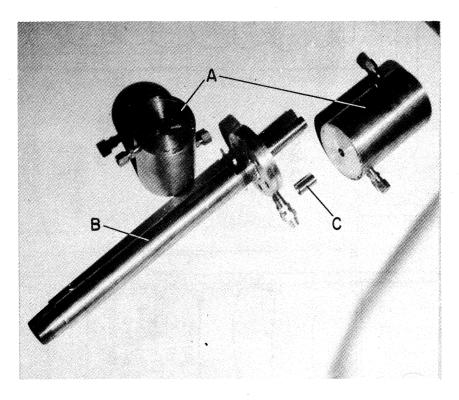


Fig. 11. Burner heads (A), burner tube (B), and gold liner (C) of burner assembly.

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## PART II. SUBPROJECT M943-4 (FORMERLY M943-C), THE EFFECT OF RADIATION ON CHEMICAL REACTIONS

#### Personnel:

Subproject Supervisors: J. J. Martin, Associate Professor of Chemical Engineering, L. C. Anderson, Chairman of the Department of Chemistry.

Senior Research Assistants: D. E. Harmer, J. G. Lewis.

Assistants in Research: D. J. Goldsmith, J. R. Hallman, R. L. Kinney, E. M. Rosen.

#### A. INTRODUCTION

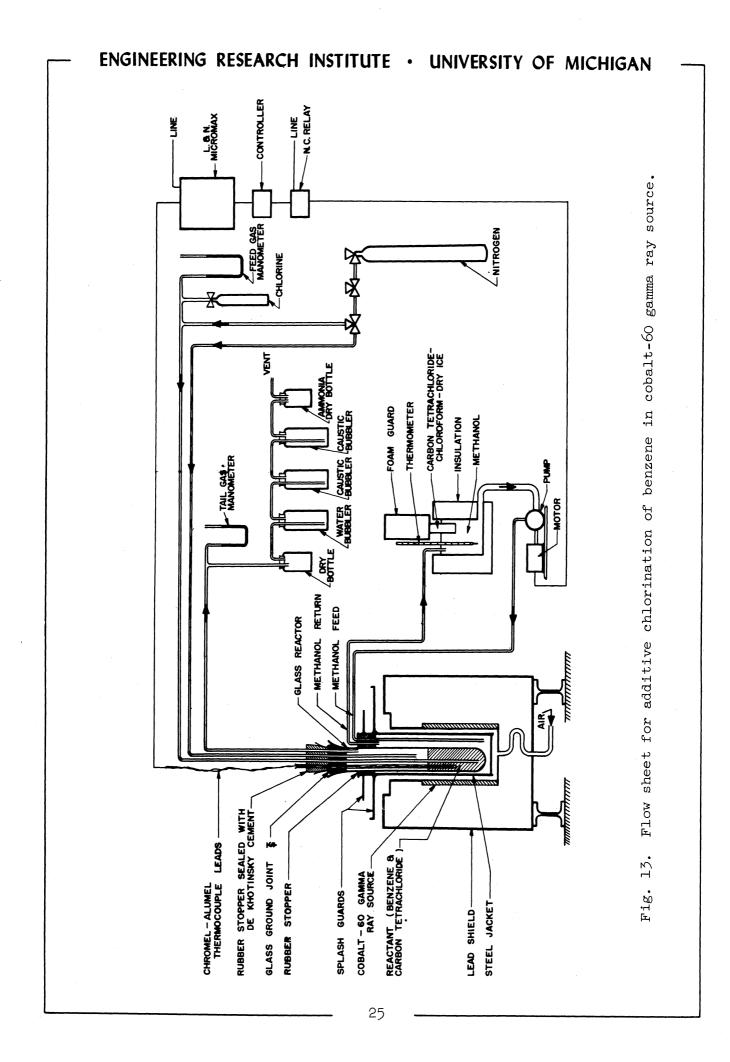
In the investigation of the effect of radiation on chemical reactions, two types of reactions are being studied: (1) chemical reactions of commercial, or possible commercial, importance that may be influenced by radiation, and (2) chemical reactions of a general kind which aid in explaining the mechanism of radiation effects.

#### B. EXPERIMENTAL WORK

In choosing chemical reactions for study of the effect of radiation, two different avenues of approach may be followed: A single reaction which has been found to be affected by radiation may be studied in detail, determining quantitatively the effect of all variables involved: or a large number of reactions may be tried in a preliminary sort of investigation to determine which reactions are affected appreciably by radiation. In general it is the latter approach which has been more closely followed so far. Of the reactions considered below, only the chlorination of benzene has received special attention.

#### 1. Halogenation

As used in this work, the term halogenation (chlorination, bromination, or iodination) refers to the reaction of an organic compound with a halogen. The products may be the result of addition or substitution processes or combination thereof.



<u>a.</u> Chlorination of Benzene. It is well known that ultraviolet radiation promotes the addition of chlorine to the benzene ring, Slator<sup>12</sup> having demonstrated this fifty years ago. Alyea<sup>1</sup> showed that alpha radiation produces a similar effect. Therefore, an experiment was undertaken to determine whether this reaction could be promoted by gamma radiation.

When chlorine is bubbled through benzene, an addition reaction takes place at a very rapid rate. In view of the importance of the product, benzene hexachloride, it was decided to make a more intensive study of this reaction. After preliminary runs had been made, apparatus was constructed and assembled in which the reaction could be carried out under controlled conditions. A schematic diagram of this system is presented as Fig. 13. For convenience of operation in the Fission Products Laboratory, most of the controls for the experimental work were located on the second floor, adjacent to the opening over the kilocurie cobalt-60 source. (See laboratory plan on page 14 of Progress Report 14.) Thus the gas cylinders, manometers, tail-gas absorption train, and temperature recorder-controller were located on the second floor (Fig. 14), while the kilocurie cobalt-60 source, glass reactor with its steel jacket, and the cooling system were located on the first floor. Glass tubing was used

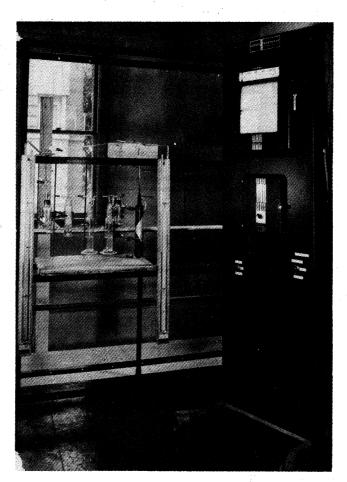


Fig. 14. Control equipment for chlorinations (Second floor, Fission Products Laboratory).

to carry gases between the reactor and the supply cylinders and absorption train.

The glass reactor consisted of a 25-mm glass tube long enough to extend completely out of the cobalt-60 source. The lower end of the glass reactor was closed, while the upper end was fitted with a standard ground joint. Four tubes, a chlorine inlet, a nitrogen inlet above the liquid level, a tail-gas exit, and a thermocouple well, were held in place in the inner ground joint by means of a rubber stopper sealed with resin cement (see Fig. 15). The assembled reactor (Fig. 16) was inserted in a steel jacket through a rubber stopper which also contained feed lines for the coolant liquid (Fig. 17). Splash guards were affixed to the steel jacket and compressed air was circulated through the vent hole at the bottom of the source container in order to prevent any possible damage to the cobalt-60 source in event of a spill or a chlorine leak. The tail gases from the reaction were absorbed in sodium hydroxide solution for analysis at the end of the experimental run (Fig. 18).

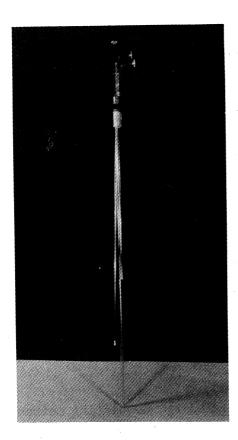


Fig. 15. Inner part of glass reactor tube, showing inlet and exit tubes and thermocouple well.

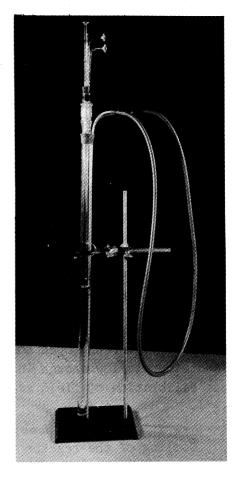


Fig. 16. Assembled glass reactor tube, containing liquid reactants.

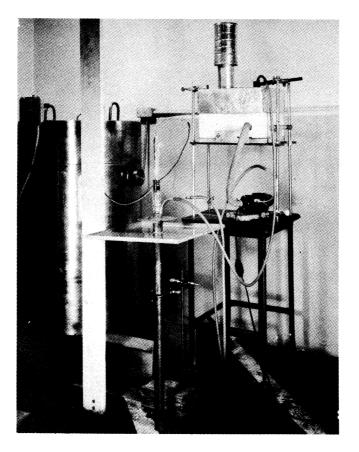


Fig. 17. Glass reactor tube in its steel jacket, connected to cooling system. Cobalt-60 vault is in the background.

The assembled glass reactor tube in its steel jacket was placed in the open kilocurie cobalt-60 gamma source and all glass connecting tubes were attached to the feed lines by extension tools fabricated for that purpose (Fig. 19). The design of the lead shielding of the kilocurie cobalt-60 gamma source at the University of Michigan is such that a high-intensity beam is located directly over the open source, but the radiation field at the sides of the container is low enough in intensity to allow loading operations in this area (see calibration of field around the open source, page 14 of Progress Report  $1^4$ ).

Cooling of the reactor was accomplished by circulating methanol chilled by dry ice. When dry ice was added directly to the methanol, the centrifugal circulating pump became filled with carbon dioxide gas, causing the pump to lose its prime. Consequently, it was found necessary to chill the methanol indirectly by passing it around a can containing a mixture of carbon tetrachloride, chloroform, and dry ice. This part of the cooling system was

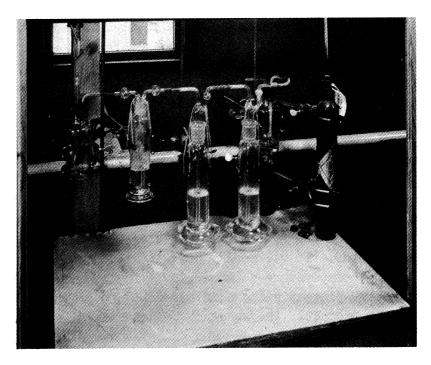


Fig. 18. Chlorine feed cylinder and tail-gas absorber bottles.

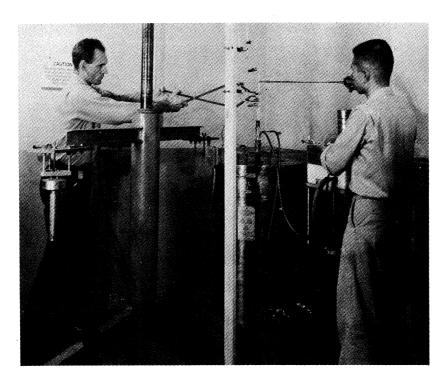


Fig. 19. When in place in the cobalt-60 gamma source, the glass reactor can be connected to feed lines by means of tools which allow the workers to stand in a field of low radiation intensity.

insulated with sections of polystyrene foam. The thermocouple of a Micromax temperature recorder-controller was placed inside the glass reactor tube. This controller alternately turned the pump on and off. The cold methanol was pumped into the steel jacket surrounding the reactor. The temperature control was greatly improved in later runs by circulating methanol continuously through the reactor cooling jacket. The temperature of the methanol was controlled by by-passing some of the methanol through the dry-ice chamber by means of a second pump. This pump was controlled by the Micromax regulator. The assembled apparatus was located on the first floor; its appearance from the second floor may be seen in Fig. 20.

Before each run, the entire system was purged with nitrogen in order to exclude all oxygen from the system. In early experiments, pure benzene was placed in the reactor. It was found, however, that the reaction then proceeded so rapidly that temperature control was virtually impossible, and the inlet lines soon became plugged with solid product. To prevent this complete solidification of the product, the reactant benzene was diluted with carbon tetrachloride. Mixtures containing as much as 30% benzene by volume produced

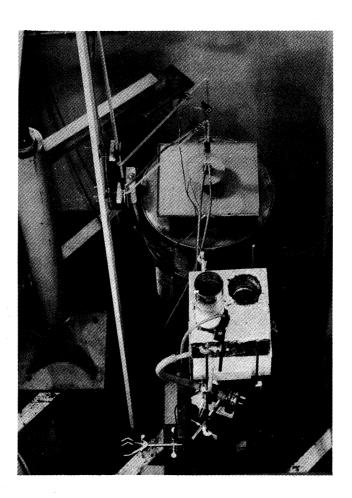


Fig. 20. Second-floor view of reactor and its glass feed lines and cooling system.

a semifluid slurry of solid product in the carbon tetrachloride. The reaction has been run to near completion in as little as 15 minutes when using benzene solution kept saturated with chlorine.

The solid product has been analyzed and was found to be nearly pure 1,2,3,4,5,6-hexachlorocyclohexane, or "benzene hexachloride".\* This compound is formed as a mixture of five stereoisomers, to which other investigators have assigned the first five letters of the Greek alphabet. The gamma isomer is known commercially as "Lindane" and possesses marked insecticidal properties which make it of considerable commercial value. The annual production of the mixed isomers in 1951 was about 125 million pounds?

Although the material may be used either as the unseparated mixture of isomers or in the purified gamma form, the insect-killing power is almost entirely dependent on the gamma isomer content. Table IV shows the gamma

TABLE IV \*\*

ANALYSIS OF SOME SAMPLES OF BENZENE HEXACHLORIDE

| Approximate Temperature of Reaction, °C | % Benzene<br>(by volume) | % CCl <sub>4</sub> (solvent)<br>(by volume) | <pre>% Gamma Isomer in Benzene Hexachlor- ide (from infra- red analysis)</pre> |
|---|--------------------------|---|--|
|   |                          |   |  |
|   |                          |   | 11.3 (over   |
| 20                                      | 10                       | 90  | chlorinated)   |
| 20                                      | 20                       | 80  | 12.5   |
| -10                                     | 10                       | 90  | 12.3   |
| -10                                     | 30                       | 70  | 12.8   |
|   |                          |   |  |

<sup>\*</sup> The authors are indebted to the Process Engineering Department of Wyandotte Chemicals Corporation and to Dr. L. E. Liggett of that company for an analytical method for total organic chlorides by the use of sodium diphenyl reagent.

Primary standard samples of the alpha, beta, gamma, and delta isomers were kindly supplied by the Hooker Electrochemical Company, Physical Chemical Laboratory. These samples were for the purpose of setting up analytical facilities in this laboratory.

<sup>\*\*</sup> The authors wish to express their gratitude to E. I. du Pont de Nemours and Company, Engineering Service Division, for furnishing the analyses for gamma isomer listed here.

isomer content of a few samples which were analyzed by infrared. It should be noted that the temperatures listed are only approximate. In each case the temperature went somewhat higher (as much as 5°C) at the height of the reaction. The continuous cooling system that was installed later brought this maximum temperature down several degrees. The percentages of gamma isomer are not significantly different from those obtained in processes from the addition of chlorine to benzene under the activation of ultraviolet light.

- <u>b.</u> Chlorination of Toluene. Recently, work has been undertaken to chlorinate toluene under conditions similar to those used for benzene. Although both ultraviolet light and gamma radiation activated the addition of chlorine to the benzene nucleus, it is known that in toluene ultraviolet radiation activates substitution of chlorine for hydrogen in the methyl group. Therefore, it is of great interest to determine whether gamma radiation will activate the side-chain substitution or the nuclear-addition reaction. If it is the latter reaction which is predominantly promoted when toluene is chlorinated in the presence of gamma radiation, it is possible that gamma radiation may serve as a new tool to promote reactions that cannot be accomplished by other means. Preliminary results at the time of this writing indicate that both reactions, the addition and the substitution, are taking place under gamma irradiation; however, separation and analysis of reaction products is still in progress. The reaction between toluene and chlorine takes place at a rate comparable to that between benzene and chlorine under similar conditions.
- c. Bromination and Iodination Reactions. Because of the success in the promotion of chlorine addition to benzene, experiments have been tried using bromine and iodine. Evaporation of a solution of bromine in benzene which had been given about 16 hours of gamma irradiation in the kilocurie cobalt-60 source left a residue of a small quantity of crystals in a liquid having noticeable lachrymatory properties. At the time of writing, this small yield of reaction product had not been identified. A solution of iodine in benzene which was given about 25 hours in the kilocurie cobalt-60 gamma source gave no noticeable residue on evaporation. No change in color of either the bromine or iodine solution could be observed during the irradiation. A solution of linseed oil, benzene, and iodine was found to become lighter in color when given about 21 hours of irradiation in the kilocurie cobalt-60 gamma source.

#### 2. Oxidation

<u>a. Using Elementary Oxygen.</u> Sulfur dioxide and oxygen in a gaseous mixture of stoichiometric proportions were irradiated while under pressures of about 50 psig and temperatures of about 300°F. No sulfur trioxide was detected by iodometric-acidimetric titration procedures.<sup>3</sup>

Oxygen was bubbled through liquid sulfur dioxide held in the cobalt-60 vault at temperatures of about 250°F until the total pressure reached about 800 psig. No visible drop in total pressure was observed when introduction of oxygen was stopped. However, when the reaction products were absorbed in water, considerable amounts of sulfate were precipitated by the addition of barium chloride.

When sulfur dioxide and oxygen were agitated in aqueous solution at atmospheric pressure in the presence of gamma radiation, considerable amounts of sulfate were detected in the resulting solution. These results indicate the formation of sulfuric acid by the oxidation of sulfur dioxide dissolved in water. This can be compared with the results of Alyeal and Backstrom2 on the oxidation of sodium sulfite in aqueous solution in the presence of ultraviolet radiation.

<u>b.</u> <u>Using Other Oxidizing Agents</u>. From time to time, when space has been available in the cobalt-60 source, single experiments were made on mixtures in which there was reason to believe, on the basis of other experience, that reaction might occur. In addition to investigating the possibility of brominating or iodinating benzene, as mentioned above, attempts have been made to react sulfur with benzene, pyridine, and naphthalene. Changes in odor after irradiation indicated that some reactions had apparently taken place, but these reactions have not been investigated further up to this time.

While other workers have also used elementary oxygen in reactions carried out in radiation fields, the use of other oxidizing agents has not been reported. Solutions of potassium permanganate in acetone and in an acetone-benzene mixture have been irradiated in the kilocurie cobalt-60 gamma source. The solutions containing benzene were decolorized and yielded a dark precipitate which appeared to be manganese dioxide. No change was apparent in irradiated solutions of permanganate in acetone.

Potassium iodide solutions were also irradiated in the cobalt-60 gamma source, using neutral solutions, some slightly acidic with acetic acid, and some 1 N with hydrochloric acid. Visible amounts of iodine were present in all samples irradiated for 30-40 hours. Titratable quantities of iodine (with 0.1 N thiosulfate) were present in the irradiated acidic samples. Irradiated aqueous acidic (acetic acid) potassium iodide solution displayed a measurable potential with respect to an unirradiated control solution, indicating a difference in concentration. Also, a measurable difference was detectable when each solution was measured separately with respect to a calomel half-cell.

#### 3. Polymerization

Acetone has been reported to decompose into free methyl and acetyl radicals.  $^{13}$  Acetone was therefore tried as a reaction initiator under gamma

irradiation. Irradiation appeared to change the odor of mixtures of acetone with ethanol and acetone with ethyl ether to some extent. It was consequently thought that some amount of ester may have been produced. However, distillation analysis on the Podbielniak column failed to yield any larger amount of any material other than the reactants. Mixtures of acetone and ethylene have been irradiated while under pressures of about 1000 psig. Yields of 1 to 5% by weight of white powder have resulted from these runs, and also with ethylene alone held at 900 psig and irradiated for about 50 hours with cobalt-60 gamma radiation.

Solutions of 1% glacial acetic acid in styrene have been irradiated, and viscosity measurements indicate that the acetic acid activates the polymerization of styrene under gamma irradiation. Liquid isobutylene was irradiated for some time under its own vapor pressure. A small amount of liquid residue was isolated. This material had a terpene-like odor. When a mixture of 50% (by volume) of styrene in linseed oil was irradiated for about 20 hours in the kilocurie cobalt-60 gamma source it showed slightly increased viscosity and a slightly bleached color. A similar mixture of linseed oil and acrylonitrile was given 39 hours in irradiation time, producing a thick, thixotropic paste. Odor indicated that some monomer was still present in this mixture.

#### 4. Other Reactions

Certain similarities in the electron configurations of the  $\text{Cl}_2$  molecule and the  $\text{H}_2\text{O}_2$  molecule led to the trial of hydrogen peroxide as a reactant with benzene. In some runs acetone was used as a mutual solvent. In one such run a crystalline product with a sharp melting point was obtained, but subsequent runs produced explosive products, which were probably organic peroxides. This work has been temporarily discontinued.

The reaction of benzene and chlorine is promoted by both ultraviolet and gamma radiation. It is therefore of some interest to compare the behavior of systems which have been subjected to known amounts of radiation from each of these two ranges of frequencies. For purposes of comparison, the benzophenone-isopropanol reaction was selected, since under certain conditions of concentration, sunlight, and geometrical arrangement of apparatus, this solution will show a visible precipitation of the bimolecular reduction product, benzopinacol, in about 5 hours. A sample of this solution failed to show any such reaction after about 50 hours of gamma irradiation in the kilocurie cobalt-60 source. Apparently this reaction is not appreciably promoted by gamma radiation. The energy absorbed by this system in the kilocurie cobalt-60 gamma source was estimated to be 0.15 cal/ml-hr. This value is based in part on the calibration of the cobalt-60 gamma source as given in Progress Report 3.6 In addition, the rate of energy absorption was calculated for this system when exposed to sunlight. The fraction of the solar constant in the range 3300Å to 3400Å was

estimated. The sun was assumed to be a black-body radiator, and the Planck radiation function was employed, as given by Jahnke and Emde. <sup>10</sup> It is further assumed that no absorption of incident solar radiation occurs in this frequency range, either by the atmosphere or by the glass container. When all radiation in the range of 3300Å to 3400Å was assumed to be absorbed by the reaction mixture, a value of 0.39 cal/ml-hr was found. It will be noted that the rates of energy absorption for this system in sunlight and in the source are of the same order of magnitude.

#### C. FUTURE WORK

The investigations of this laboratory indicate that the reactions which are most likely to be accelerated by ionizing radiation are those which proceed by a free-radical chain mechanism. The evidence to date also indicates that dense phases (liquids or gases under very high pressures) are necessary to absorb appreciable amounts of gamma radiation to produce sufficient effects for the products to be measured by the usual macro-methods of chemical analysis. Future work will, therefore, follow lines consistent with these observations.

It is expected that reactions of liquids at atmospheric pressure and varying temperature, such as the benzene chlorination, will receive continued emphasis, as will reactions of gases under pressure such as the polymerization of olefins or the hydrogen-carbon monoxide reactions. Every effort is being made to control the temperature of the reactions more closely, since this is so important in comparing reactions with and without radiation. Reactions which normally take place at high temperature are especially important in this study; it is desirable to determine whether gamma radiation can cause them to proceed at a rapid rate at a lower temperature.

If other reactions are found to proceed as rapidly under gamma radiation as the chlorination of benzene, these reactions will probably be given special consideration and studied in some detail. However, the broad approach of trying a number of reactions is still very much in effect in the program at Michigan.

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#### PART III. COOPERATIVE RESEARCH WITH OTHER PROJECTS

#### A. INTRODUCTION

As explained in the preface, cooperative research has been performed with other groups, primarily the Michigan Department of Health and the Michigan Memorial-Phoenix Project. In these studies the samples to be irradiated were prepared by other groups. The irradiations were made in the Fission Products Laboratory and then the samples were analyzed by the personnel of the other research groups. The results, made available to both groups, are described in this part of this report.

## B. COOPERATIVE RESEARCH WITH THE MICHIGAN DEPARTMENT OF HEALTH

The work with the Michigan Department of Health involved the gamma irradiation of several human blood plasma fractions: Antihemophilic Globulin, Immune Serum Globulin and Normal Serum Albumin. One of the purposes of this study was to investigate the feasibility of sterilization of these blood fractions so as to destroy the virus of serum hepatitis. This serious disease is transmitted by contaminated blood and blood products (see Progress Report 3).

In the experimental work, two vials of each plasma fraction were exposed to gamma radiation in the one-kilocurie cobalt-60 source for dosages of 0.08 mega rep in air (1 hour), 0.24 mega rep in air (3 hours), 0.48 mega rep in air (6 hours), and 1.92 mega rep in air (24 hours).

Messrs. K. B. McCall and F. C. Bloom, of the Biologic Products Section, Division of Laboratory, Michigan Department of Health, have prepared the following report dated October 1, 1952, and entitled, "Sterilization of Biological Products with Gamma Radiation", which presents the results of their experiments.

!! · · ·

# 1. Normal Serum Albumin (Human) Lot A36

Two mice weighing 18-20 grams each were injected intraperitoneally with a 0.5-ml dose of albumin, and observed for 7 days for gains in weight.

"TABLE V
SAFETY TEST OF GAMMA-IRRADIATED SERUM ALBUMIN

| Ra    | adiation Dose   | <u>Gain in W</u> | eight,(g) |
|-------|-----------------|------------------|-----------|
| Hours | Mega Rep in Air | 1                | 2         |
| 1     | 0.08            | 2.5              | 2.0       |
| 3     | 0.24            | 2.9              | 1.8       |
| 6     | 0.48            | 2.5              | 2.8       |
| 24    | 1.92            | 5.1              | 4.4       |

"TABLE VI
PYROGEN TEST OF GAMMA-IRRADIATED SERUM ALBUMIN

|        | Rabbit  |        |                  | Temperature C | hange in °F |
|--------|---------|--------|------------------|---------------|-------------|
| Sample | No.     | Weight | Dose             | Low           | High        |
| 0 hr.  | MM 819  | 1.6 kg | 4.8 ml           |               | 0.8         |
| 0 111. |         | _      |                  |               |             |
| 11     | MM 860  | 1.9    | 5 <b>.</b> 7     |               | 1.0         |
|        | MM 873  | 1.7    | 5.1              |               | 1.2         |
| l hr.  | ML 703  | 1.8    | 5.4              |               | 1.0         |
| tt .   | ML 719  | 1.6    | 3·3 <del>*</del> |               | 0.6         |
| 11     | ML 763  | 1.6    | 3.3 <del>*</del> |               | 2.0         |
|        | 144 (O) | 1.0    | J• J             |               | 2.0         |
| 3 hr.  | ML 785  | 1.5    | 4.5              |               | 0.7         |
| 11     | ML 730  | 1.6    | 3.3 <del>*</del> |               | 0.9         |
| 11     | ML 727  | 1.9    | 3·3 <del>*</del> | 0.6           | 0.2         |
|        |         |        |                  |               |             |
| 6 hr.  | ML 736  | 1.6    | 4.8              |               | 1.0         |
| M .    | ML 702  | 1.8    | 3 <b>.</b> 0*    |               | 1.0         |
| 11     | ML 771  | 2.0    | 3.0*             |               | 0.2         |
|        |         |        |                  |               |             |
| 24 hr. | IN 919  | 1.7    | 5.1              |               | 1.5         |
| 11     | IN 927  | 1.7    | 2.6 <del>*</del> |               | 1.3         |
| 11     | IN 975  | 1.6    | 2.4 <del>*</del> |               | 1.3         |
|        |         |        |                  |               | -           |

<sup>\*</sup> Partial Dose (Full Dose = 3 ml/kg)

"TABLE VII
ELECTROPHORETIC ANALYSIS OF GAMMA-IRRADIATED SERUM ALBUMIN

| Hours of Exposure | Composition of Albumin | Solution, per <u>1</u> | cent<br>2 |
|-------------------|------------------------|------------------------|-----------|
| 1                 | 96                     | 3                      | 1         |
| 3                 | 95                     | 4                      | 1         |
| , 6               | 97                     | 3                      | 0         |
| 24                | A93                    | 6                      | 1         |
|                   | D97                    | 9                      |           |
|                   | 95                     | 4                      | 1         |
| 0                 | A95                    | 5                      | *         |
|                   | D97                    | 2                      | 1         |

"TABLE VIII
STABILITY TEST OF GAMMA-IRRADIATED NORMAL SERUM ALBUMIN

| TT TT                   |            | Hours Treated | with Gamma       | Radiation        |                   |
|-------------------------|------------|---------------|------------------|------------------|-------------------|
| Hours Heated<br>at 57°C | 0*<br>N.U. | 1<br>N.U.     | 3<br><b>N.U.</b> | 6<br><b>N.U.</b> | 24<br><b>N.U.</b> |
| •                       |            |               |                  |                  |                   |
| 0                       | 6          | 9             | 8                | 10               | 11                |
| 24                      | 7          | 11            | 11               | 13               | 32                |
| 48                      | 7          | 13            | 13               | 18               | 69                |
| 72                      | 7          | 14            | 14               | 20               | 98**              |
| 96                      |            | 15            | 15               | 20               |                   |

<sup>\*</sup> Stability data for control sample obtained 3-17-52; all other data obtained beginning 7-21-52.

# "2. Immune Serum Globulin

Two mice weighing 18-20 gm each were injected intraperitoneally with a 0.5-ml dose of globulin and observed for 7 days for gain in weight. Results were satisfactory.

<sup>\*\*</sup> Not accurate; could not open diaphragm enough to balance against standard. N.U. = Nephelometric units.

"TABLE IX
SAFETY TEST OF GAMMA-IRRADIATED IMMUNE SERUM GLOBULIN

| Sample | Gain in 1    | Weight, (g)  |
|--------|--------------|--------------|
| l hr.  | 7.3          | 5 <b>.</b> 8 |
| 3      | 5 <b>.</b> 5 | 6.4          |
| 6      | 5.3          | 7.1          |
| 24     | <b>6.</b> 3  | 5.5          |

## "TABLE X

# INFLUENZA, NEUTRALIZATION TEST OF GAMMA-IRRADIATED IMMUNE SERUM GLOBULIN

(Type A, Fml strain virus used; 3-week-old mice - 5 per dilution.)

| Sample                | Dilution at 50% Endpoint | Treated<br>Untreated | Treated Control Serum | Untreated<br>Control Serum |
|-----------------------|--------------------------|----------------------|-----------------------|----------------------------|
| Control serum #11g141 | 1:179                    |                      |                       | 1:1.8                      |
| Untreated globulin    | 1:320                    |                      |                       |                            |
| 1-hr. treatment       | 1:249                    | 1:0.8                | 1:1.4                 |                            |
| 3-hr. treatment       | 1:269                    | 1:0.8                | 1:1.5                 |                            |
| 6-hrtreatment         | 1:285                    | 1:0.9                | 1:1.6                 |                            |
| 24-hr. treatment      | 1:135                    | 1:0.4                | 1:0.8                 |                            |

#### "TABLE XI

# DIPHTHERIA ANTIBODY TITRATION (FRASER TEST) OF GAMMA-IRRADIATED IMMUNE SERUM GLOBULIN

| Sample  | $\underline{\mathtt{Units/ml}}$ |
|---|---------------------------------|
| Control serum #11g141 Untreated globulin 1-hr. treatment 3-hr. treatment 6-hr. treatment 24-hr. treatment | 1.6<br>1.4<br>1.4<br>1.4<br>1.4 |
|   |                                 |

## "3. Antihemophilic Globulin

"TABLE XII

NITROGEN DATA ON GAMMA-IRRADIATED ANTIHEMOPHILIC GLOBULIN

|             | Soln. |            | Pro      | otein       |                  |          |
|-------------|-------|------------|----------|-------------|------------------|----------|
| Irradiation | Time, | Insoluble, | Soluble, |             | % Total          | Clotting |
| Time, hrs   | Sec   | mg         | mg       | ½ Insoluble | <u>Clottable</u> | Time     |
|             |       |            |          |             |                  |          |
| 1           | 45    | 6.19       | 156      | <b>3.9</b>  | 45.2             | 105 sec  |
| 3           | 40    | 4.00       | 187      | 2.1         | 43.2             | 160      |
| 6           | 60    | 6.13       | 175.4    | 3.4         | 37.8             | 70       |
| 24          | 45    | 32.6       | 137.8    | 19.2        | 19.5             | 155      |
| 0           | 75    | 49.4       | 232.8    | 17.6        | 43.4             | 185      |

# "4. Summary of Data

"Normal Serum Albumin (Human) Lot A36. A safety test was quite satisfactory. There was a slight increase in weight gain through the 6-hour sample. A very marked gain is shown in the 24-hour sample.

"The pyrogen test shows a gradual increase from the O-hour to the 24-hour sample. This might be indicative of some change in the product.

"The electrophoretic data are very comparable to the untreated control. The data do not indicate any particular change in mobilities.

"The stability test (57°C) does show a very definite change from the untreated sample. Judging from these data, 6 hours of treatment with gamma radiation would be about the maximum to use.

"Immune Serum Globulin. The safety test is good. All test animals show a very marked gain in weight.

"The Influenzae neutralization test shows that at the dilution of 50% endpoint' there is a slight drop in dilution for the 24-hour sample. The treated serum compared to the untreated shows a gradual drop in dilution through the 24-hour sample, which shows the maximum. The same thing is true for the treated serum compared to the control serum.

"The diphtheria antibody titration shows that here again there is a slight loss in antibody in the 24-hour sample.

"Antihemophilic Globulin. The data to consider here are the columns headed '% Insoluble' and '% Total Clottable'. It is apparent that gamma irradiation through 6 hours increases the solubility but decreases the clottable protein. Since clottable protein is probably the most important factor with this product, it is apparent that irradiation for a period longer than 3 hours would not be desirable.

"In general, it can be stated that the effect of gamma irradiation upon the chemical and physical properties of Normal Serum Albumin, Immune Serum Globulin, and Antihemophilic Globulin is not significant if the exposure time is 3 hours or less. From these data it can be recommended that further studies be made."

#### C. COOPERATIVE RESEARCH WITH MICHIGAN MEMORIAL-PHOENIX PROJECTS

## 1. Breaking the Cycle of Trichinosis by Gamma Irradiation

H. J. Gomberg, Assistant Director of the Michigan Memorial-Phoenix Project, and S. E. Gould, M. D., Pathologist of Wayne County General Hospital and Clinical Professor of Pathology of Wayne University, College of Medicine, have conducted a series of investigations on the effect of gamma radiation on trichina larvae. These experiments were first undertaken using x-radiation in the U. S. Atomic Energy Commission Laboratory on Biological Effects of Irradiation, University of Michigan. The results of the study using x-radiation have been described in an article, "Effects of X-Rays on Trichina Larvae", American Journal of Pathology (in press).

This research has been continued with support of Phoenix Project 54 on a cooperative basis with the Fission Products Laboratory. Gamma irradiation from the one-kilocurie cobalt-60 source was performed by the Fission Products Laboratory and the results of the experiments were made available to both groups. Before these results are presented, it may be appropriate to review briefly the problem of trichinosis for the benefit of those not of the medical profession and perhaps unfamiliar with the problem.

- a. <u>Trichinosis in Man.</u> S. E. Gould, M. D., is the author of a text, <u>Trichinosis</u>, of 356 pages and a number of articles on this subject in the medical literature and is one of the leading authorities on the subject. The following historical record of the disease and autopsy surveys were described by Gould in Postgraduate Medicine as follows:
- 1) "Historical. In 1835, James Paget, then a 21-year-old first-year medical student at St. Bartholomew's Hospital in London, encountered, in the dissecting room, small whitish calcified specks in the skeletal muscles

of a cadaver. He sought out someone who had a microscope, examined the specks and found them to be made up of cysts (Fig. 21), each of which contain a worm. The worm was then named <u>Trichina spiralis</u> but is now known as <u>Trichinella</u> spiralis.



Fig. 21. Portion of diaphragm, from a man of 59, showing calcified trichina cysts.

"In 1846, a Philadelphia physician, Joseph Leidy, while eating a pork sandwich, noticed minute specks in the meat which struck him as being similar to those he had seen in the muscles of a human subject that he had dissected a few days previously. He examined the rest of the pork microscopically and found it to be full of trichinae (Fig. 22). This discovery, for the first time, definitely associated the parasite with the pig.

"It was not, however, until 1860 that the disease was known to produce fatal human infection. In that year, a German servant girl in Dresden, Germany, was brought into the hospital where she died thirty-three days after the onset of an illness which was diagnosed as typhoid fever. Zenker, the pathologist, examined the skeletal muscles of this patient at autopsy for evidence of degeneration characteristic of typhoid fever, but found instead dozens of living worms in every microscopic field. This startling picture led him to undertake a remarkable bit of detective work. He visited the home of this girl and found that a pig had been butchered just before Christmas-time, that every member of the household had eaten of various products made from this pig, and that all persons subsequently became ill with a disease variously diagnosed. He found similar living worms in the unused meat. With the aid of Virchow and other investigators, he then conducted experiments to work out the life cycle of the parasite (Fig. 23).

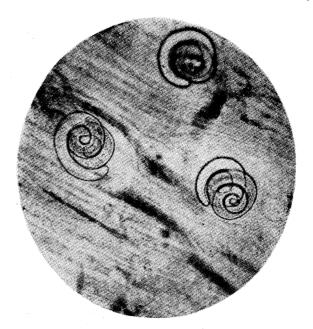


Fig. 22. Trichina larvae in muscle, seven months after infection x45. Unstained prepration mounted in glycerin

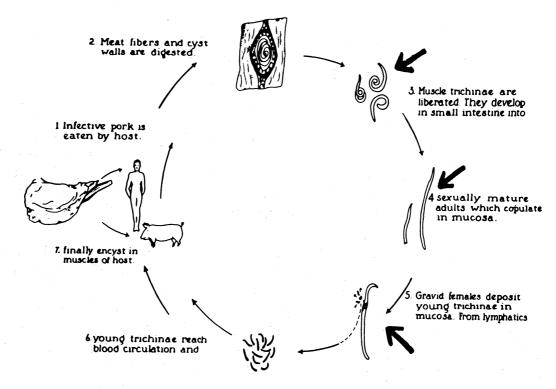


Fig. 23. Life cycle of Trichinella spiralis

"When pork, containing living trichinae, is eaten by man, or when scraps of pork (in garbage) are eaten by the pig, the muscle and the cyst walls are digested in the stomach of the host, liberating the larvae. These larvae, which are of male or female sex, mature in the small intestine of the host, and toward the end of the first week copulate. The adult male later dies but the adult female, burrowed in the mucosa of the small intestine, during the next eight weeks or so, liberates her living young worms into the mucosal lymphatic vessels from where they reach the heart, then the general blood stream, and finally are filtered out of the blood capillaries to settle and encyst in the skeletal muscles of the host (Fig. 24).

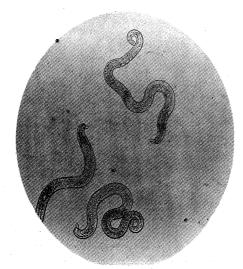


Fig. 24a. Adult male trichinae (stage
found in small
intestinal tract
of host early in
infection). x65.

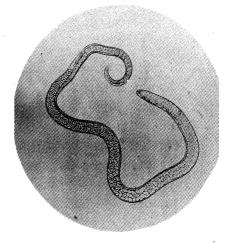


Fig. 24b. Adult female trichinae containing numerous larvae within its uterus. x65

"Shortly after the discovery of this first fatal infection, in 1860, several epidemics (Table XIII) occurred in Germany, the most notable of which took place in 1865 in the town of Hedersleben, having a population of 2,100. There 327 persons fell ill with this disease and 101 died, a mortality rate of 30 per cent. This stirred the medical profession in Germany to urge the adoption of measures for control of the disease. The net result was that a system of microscopic inspection of all hogs slaughtered in Germany was instituted...

Fig. 24c. Portion of gravid adult female trichina imbedded in mucosa of duodenum of experimental white rat, six days after infection. x55.



Fig. 24d. Portion of adult worm shown in Fig. 24c. x250. Note larva present within vagina of mother worm, from which it will soon be extruded into tissues (intestinal mucosa) of host.



Fig. 24e. Muscle trichina. x135. From white rat, six days after experimental infection. x35.

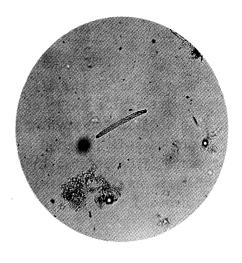




Fig. 24f. Beginning coiling of trichina larvae in muscle of white rat, eighteen days after experimental infection. x35.

Fig. 24g. Myositis in human trichinosis four weeks after infection. Note degeneration of muscle, edema and inflammatory cell infiltration. x70.

"TABLE XIII
EARLY EPIDEMICS OF TRICHINOSIS IN GERMANY

| Year | Town   | Persons Ill | Deaths — | Mortality Rate, Per Cent |
|------|--|-------------|----------|--------------------------|
| 1849 | Wegeleben Hettstädt, pop. 4,000 Hedersleben, pop. 2,100 Emersleben | 164         | 27       | 16.5                     |
| 1863 |  | 158         | 27       | 17.1                     |
| 1865 |  | 337         | 101      | 30.0                     |
| 1833 |  | 403         | 66       | 16.4                     |

2) "Autopsy Surveys. Within recent years in the United States a number of surveys of the incidence of trichinosis in autopsies have been made, and these show an average of 16 per cent infection (Table XIV). This means that at least 16 per cent of persons in the United States, at the time of death, have trichinosis. In our series of over 1,200 autopsies, we found a total incidence of trichinous infection of 22 per cent (Table XV). Notice in Table XV that in succeeding decades the incidence became progressively greater, so that in the 80's we found an incidence of 33 per cent. It stands to reason that the older the person the more chances he has had of exposure to this infection.

"TABLE XIV

INCIDENCE OF TRICHINOSIS IN THE UNITED STATES AT AUTOPSY

| <u>Year</u> <u>1</u> | Author               | Locality                          | Method                              | exams.    | No.      | d            |
|----------------------|----------------------|-----------------------------------|-------------------------------------|-----------|----------|--------------|
| 1931 Queer           |                      |                                   |                                     |           |          |              |
|                      |                      | Rochester,<br>N. Y.               | Digestion                           | 344       | 59       | 17.1         |
| 1934 Riley<br>Sch    | ,<br>neifley         | Boston, Mass. Minneapolis, Minn.  | Digestion<br>Compression            | 58<br>117 | 16<br>20 | 27.6<br>17.1 |
| 1936 Hinma           | <u> </u>             | New Orleans,<br>La.               | Digestion                           | 200       | 7        | 3.5          |
| 1936 McNau<br>And    | ight,<br>lerson      | San Francisco,                    | Digestion                           | 200       | 48       | 24.0         |
| 1937 Magat           |                      | Rochester, Minn.                  | Compression                         | 220       | 17       | 7.7          |
| 1937 Pote            |                      | St. Louis, Mo.                    | Compression and microscopic section | 1060      | 163      | 15.4         |
|                      | (reported Scheifley, | Denver, Colo.                     | Digestion                           | 431       | 70       | 16.2         |
| 1938 <b>Sch</b> ei   | •                    | Minneapolis,<br>St Paul,<br>Minn. | Compression                         | 118       | 15       | 12.7         |
| 1938 Evans           | k<br>Kanada          | Clevland, Ohio                    | Digestion and compression           | 100       | 36       | 36.0         |
| 1938 Walke<br>Bre    | er,<br>eckenridge    | Birmingham, Tuscola, Ala.         | Digestion and compression           | 100       | 33       | 33.0         |
| i                    | Lapeyre              | Los Angeles, Calif.               | Digestion                           | 170       | 31       | 18.2         |
| 1939 Harre           | •                    | Durham, N. C.                     | Digestion                           | 1+14      | 0        | 0.0          |
| Joh                  | nston                |                                   | Compression                         | 6         |          | 0.0          |
|                      |                      |                                   | Digestion and compression           | 55        | 3        | 5.4          |
| 1939 Hood,           | Olson                | Chicago, Ill.                     | Digestion                           | 208       | 12       | 5.8          |
|                      |                      |                                   | Digestion and compression           | 220       | 25       | 11.4         |
| 1939 Sawit           | Z                    | New Orleans,<br>La.               | Digestion and compression           | 400       | 24       | 6.0          |
| 1940 Catro           | <b>n</b> ,           | Ann Arbor,<br>Mich.               | Digestion and 270 compression       | 300       | 71,      | 14.7         |
| 1940 Oosti           | ng                   | Dayton, Ohio                      | Digestion and 1/3 compression 48    | 134       | 27       | 20.1         |

"TABLE XIV (cont.)

|      |         |                             |                           | No. of | Posit | ive  |
|------|---------|-----------------------------|---------------------------|--------|-------|------|
| Year | Author  | Locality                    | Method                    | exams. | No.   | %    |
| 1941 | Meleney | Nashville,<br>Tenn.         | Digestion and compression | 209    | 21    | 10.0 |
| 1942 | Wright  | Various cities in 37 states | Digestion and compression | 5362   | 853   | 15.9 |
| 1942 | Gould   | Eloise, Mich.               | Digestion                 | 90     | 11    | 12.2 |
| •    |         |                             | Digestion and             | 1141   | 266   | 23.3 |
|      |         |                             | compression               |        |       |      |
|      |         | T                           | otals 1931 to 1942        | 11,287 | 1,801 | 16.0 |

"TABLE XV

# INCIDENCE OF TRICHINOSIS, ACCORDING TO AGE AT AUTOPSY (1,231 autopsies)

| Age<br>(in Years) | Persons<br>Examined | Persons With Trichinosis | Per Cent With Trichinosis |
|-------------------|---------------------|--------------------------|---------------------------|
| All ages          | 1,231               | 278                      | 22.6                      |
| Under 10          | 2                   | 0                        | 0                         |
| 10-19             | 6                   | .O                       | 0                         |
| 20-29             | 28                  | 1                        | 3.6                       |
| <b>30-</b> 39     | 84                  | 10                       | 11.9                      |
| 40-49             | 188                 | 32                       | 17.0                      |
| 50 <b>-</b> 59    | 293                 | 72                       | 24.6                      |
| 60 <b>-</b> 69    | 335                 | 78                       | 23.3                      |
| 70-79             | 219                 | 61                       | 27.9                      |
| 80-89             | 70                  | 23                       | 32.9                      |
| 90-99             | 4                   | 1                        | 25.0                      |
| Unknown           | 2                   | 0                        | 0                         |

"At autopsy larvae may be sought by the usual microscopic method of sectioning, and by the compression and digestion methods. In our series we found 2 per cent of infections when a single block of diaphragmatic muscle was sectioned. By compression of a 1.0-gm portion of diaphragm we detected 11 per cent, but when 10 portions of 1.0 gm each were examined we detected 29 per cent of infections. By the digestion method alone we found 16 per cent. When both methods were used we were able to recover trichinae from over 30 per cent of bodies examined. In a large part, therefore, the incidence of infection as determined at autopsy will depend on the methods used and the thoroughness with which these methods are applied.

"The muscle taken from the patient or from the subject at autopsy may be examined under the microscope for presence of the organisms. In the digestion method, artificial gastric juice (1 per cent pepsin and 1 per cent hydrochloric acid) is added to the ground muscle in a container. The mixture is stored in an incubator-room overnight to digest the muscle and the cyst walls. The larvae, however, resist digestion. When the mixture is then filtered, the organisms (Figs. 25 and 26) are recovered (Table XVI).

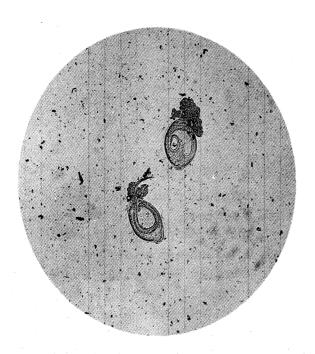
# "TABLE XVI

# RECOVERY OF T. SPIRALIS AT AUTOPSY

| Method of Examination  | Muscle<br>Examined | Gm of<br>Muscle | No. of Autopsies | Per Cent<br>Positive |
|------------------------|--------------------|-----------------|------------------|----------------------|
| 1. Microscopic section | diaphragm          | 0.25            | 331              | 2.4                  |
| 2. Compression         | pectoralg          | 1               | 1,004            | 11.1                 |
| <del>-</del>           | diaphragmg         | 1               | 1 <b>,</b> 139   | 13.5                 |
|                        | diaphragmt         | 1               | 1,031            | 19.7                 |
|                        | diaphragmt         | 10 x 1          | 317              | 29.3                 |
| 3. Digestion           | pectoralg          | 48              | 1,102            | 16.1                 |
|                        | diaphragmg         | 47              | 1,231            | 16.4                 |

 $\sigma$  = ground muscle

t = tendinous portion



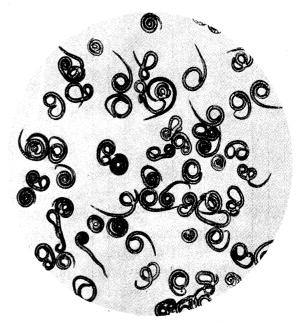


Fig. 25. Trichina cysts taken from Fig. 26. Trichina cysts obtained from digested human diaphragm. x 35. digested human diaphragm. x35.

50

3) "Diagnostic Methods. Methods that may be used for diagnosis of the disease include examination of the blood for eosinophilia; immunological tests (intradermal test, blood precipitin test, and complement-fixation test); demonstration of larvae in the host, in the blood, in the cerebrospinal fluid, or in skeletal muscles; and demonstration of larvae in the suspected meat (Table XVII).

#### "TABLE XVII

#### DIAGNOSTIC TESTS IN TRICHINOSIS

- A. Eosinophilia in blood
- B. Immunological tests
  - 1. Interadermal
  - 2. Blood precipitin
  - 3. Complement-fixation
- C. Demonstration of larvae
  - 1. In the host
    - a. blood
    - b. cerebrospinal fluid
    - c. skeletal muscle
  - 2. In suspected meat

"The leukocyte count in generally increased in the disease, with eosinophilia as the most important single sign (Fig. 27). Many times eosinophilia is present in persons who have eaten lightly infected meat, even though they develop no symptoms. This finding is encountered frequently in epidemics, in which the symptoms of various patients may be severe, moderate, mild, or absent.

"The eosinophilia appears earliest at about the tenth day after eating the infected meat and increases to a peak in the third week. The severity of the disease is not paralleled by the severity of the eosinophilia. In other words, the degree of eosinophilia is not an index to the severity of the disease. In the presence of a complication the level of eosinophilia often falls; when the complication disappears, the eosinophilia often returns. A sudden, rapid drop in number of eosinophils to one per cent or zero is usually a grave prognostic sign, many times foreboding death of the patient within a day or two.

"The duration of eosinophilia is usually several months. After six months, ordinarily, the eosinophilia has disappeared.

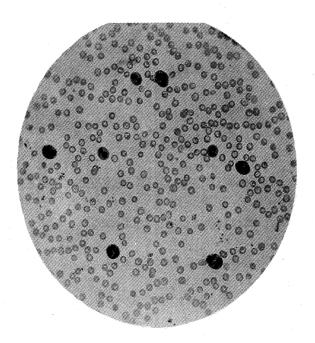


Fig. 27. Eosinophilic polymorphonuclear cells from blood of a patient with trichinosis. x320.

"For the intradermal test, the material used is made from an extract of the ground larvae. The reaction first appears on the sixteenth day of infection. Blood precipitin and complement-fixation tests become positive on or after the thirthieth day..."

In the Bulletin of the New York Academy of Medicine<sup>8</sup> Gould describes the method of infection, symptoms, treatment and prevention, and incidence of trichinosis in the United States as follows:

4) Method of Infection. ...Man acquires the infection almost exclusively from the ingestion of pork containing viable trichinae. The pig acquires the infection primarily from the consumption of bits of raw trichinous pork in uncooked garbage. The meat fibers and the walls of the trichina cysts are digested in the stomach of the host, liberating the living trichina larvae which develop within a few days into sexually mature adult forms in the small intestine. Beginning at the end of the first week of infection and continuing for a number of weeks thereafter, the gravid females, imbedded in the mucosa of the bowel, deposit their living larvae into the mucosa, whence they enter the lymphatics and then the blood stream, traverse the capillaries of the lungs to reach the left side of the heart and are then thrown into the general systemic circulation whence they finally enter the striated muscle fibers. At the time of birth, the larvae measure approximately 100 microns in length and 6 microns

in diameter, the diameter therefore being somewhat less than that of a red blood cell.

- "Symptoms. Within the skeletal muscles the larvae enlarge and set up an inflammation which is responsible for many of the symptoms of the disease. At 17 days after infection, the larva begins to coil and at about 35 days after infection, it reaches its maximum growth and then begins to encyst within the muscle fiber which has undergone degeneration. The degeneration of the muscle is attended by considerable inflammation and edema and in heavy infections the inflammation and edema may be so marked as to give rise to violent symptoms. In such severe infections, trichinosis may truly be a 'terrible' disease. Indeed, movement may be so painful that the patient may lie in one position constantly for days, afraid even to move his eyes or his tongue, to speak, or even swallow. The heart is regularly invaded by the parasite and an inflammation is set up within the myocardium. In moderately severe or in heavy infections, the myocarditis may be dangerous since it may cause sudden death or may lead to heart failure. After some weeks, depending on the severity of the disease, the inflammation in the heart, as in other striated muscles, subsides. Within the skeletal muscles the parasite, if not destroyed, becomes well encapsulated and may remain viable for many years. Within the heart, however, the parasite is regularly destroyed even though it has set up a severe inflammation. Encystment of Trichinella spiralis within the myocardium never occurs. Another danger of trichinosis is that which results from capillary damage and petechial hemorrhages within the brain. Encephalitis and meningitis sometimes are found in acute fatal cases. The incubation period varies from 2 to 28 days with an average of 10 days. The disease has been divided into the following stages: (a) intestinal, (b) muscular, and (c) convalescent. Most of the symptoms are due to the effects of invasion of the newly-born larval parasites. Particular mention may be made of supraorbital edema and fever, of the diagnostic sign of eosinophilia, and of electrocardiographic evidence of myocarditis."
- 6) "Treatment and Prevention. Treatment is largely symptomatic. Bedrest should be prescribed until all danger of heart failure or of complications has passed. There is no specific remedy except that of prevention. [ACTH and cortisone have recently been administered to lessen the severity of the symptoms.] At the present time, prevention is largely up to the ultimate purchaser or consumer of pork and consists principally in thorough cooking or sufficient heating of the pork and pork products. The Federal government recommends that pork should be boiled at least 30 minutes for each kilogram of weight (2.2 lb). Another approximate guide for thick cuts of ham or pork is to cook them one-half hour per pound. All portions of cooked pork should be white. Trichinae may also be killed within pork by methods of freezing. In order to destroy by freezing living trichinae within cuts of meat not exceeding six inches in thickness, govermental regulations require that a temperature of not less than 5°F be maintained for at least 20 days. Incidentally, this temperature is within the range of deep-freeze cabinets, such as are used in the home.

- "Incidence of Trichinosis in United States. In 1932, the meat of 14 million hogs was subjected to microscopic examination in Prussia and trichinae were found in only 0.001 per cent of the animals or in 9 hogs per million. In 1933, the meat of 14 million hogs was similiarly examined and only 0.0008 per cent or 8 animals per million were found trichinous. During the years 1898 to 1906, the meat of 8 million hogs was microscopically examined in the United States and 1.41 per cent, or over 14,000 hogs per million, were trichinous. The average incidence of trichinosis among hogs in the United States at present is as follows: 0.4 per cent of hogs which are fed cooked garbage, 0.8 per cent of hogs fed grain, 6.4 per cent of hogs fed uncooked garbage, and 15 per cent of animals fed slaughter-house offal. A conservative estimate of the average incidence of trichinosis among hogs in the United States at the present time is 1.5 per cent. (In 1944 over 90 million hogs were slaughtered in this country. If 1.5 per cent of these were trichinous, and if each hog furnished meat for a about 100 meals, there would be consumed within the United States in that year 135 million meals of pork containing trichinae. Fortunately, of course, in most instances the trichinae will have been killed.) Too often, however, live trichinae will be ingested and it is no wonder, therefore, that in recent surveys of autopsies in the United States, an average of 16 per cent of the population should have been found to harbor trichinae. This figure is low, for when more thorough examinations are made, the incidence at autopsy has been found to be as high as 36 per cent. It would be a conservative estimate to state that approximately 25 per cent of the general population of this country develop trichinous infection during their life-time. In other words approximately one person in every four becomes infected with this parasite. Fortunately the infection is subclinical in the majority of cases. It is estimated that at least 5 per cent of those who become infected show signs of illness but that only about 1 per cent of all who are infected become sufficiently ill to confine them to bed with clinical trichinosis. The mortality rate from clinical trichinosis in the United States is 5 to 6 per cent. It has been said that 'trichinosis is one of the major health problems in the United States' and that 'the United States has the greatest problem of trichinosis of any country in the world'. At the present time, the public is not sufficiently protected against the dangers of this disease..."
- b. Effect of Irradiation With Cobalt-60 on Trichina Larvae. H. J. Gomberg and S. E. Gould, M.D., have submitted the results of this study to a technical journal for publication. The manuscript of this article is presented here to describe the effects of gamma radiation on trichina larvae and to show the possible importance of radiation from fission products as a means of breaking the cycle of trichinosis and thereby eliminating the disease. No part of the following manuscript by H. J. Gomberg and S. E. Gould is to be reproduced without their permission.

"In a previous study of the effect of 200-kilovolt x-rays on trichina larvae<sup>1</sup>, it was found that a dose of 750,000 roentgens or more was required

to kill trichina larvae in vitro, but that much smaller doses were effective in rendering irradiated larvae sterile or in preventing them from maturing to adult forms. When trichina larvae were irradiated in vitro, the sterilizing dose was approximately 5000 roentgens. When a layer of trichinous muscle 1.45 cm thick was irradiated, the sterilizing dose was approximately 10,000 roentgens and the maturation-inhibiting dose was approximately 15,000 roentgens. In the present study our purpose was to determine the radiation dose measured in roentgens from a cobalt-60 source that would produce the same effects. In recent studies<sup>2</sup> it has been shown that the biological effect produced by radiation varies rapidly with photon energy even though total ionization in air (roentgen dose) is kept constant.

"Alicata and Burr<sup>3</sup> found that exposure of trichinous meat to 12,000 roentgens of cobalt irradiation produced sterility in 60 to 100 per cent of the adult female worms recovered from the intestinal tract of experimental rats 6 days after the latter were fed the irradiated larvae. After 30 days, 4 of 6 experimental rats 'showed no larvae' in their muscles, indicating that the larvae fed to these rats were sterile.

"Trichinous rat muscle and isolated trichina larvae obtained by the digestion of trichinous rat muscle were placed in lusteroid tubes for irradiation. The techniques, up to this point, are similar to those used in our previous study.  $^{\rm l}$ 

"The filled lusteroid tubes were then placed inside a cylindrical cobalt-60 irradiation unit for exposure periods ranging from 4 minutes to 13 hours. The radiation source is constructed so that only the 1.17- and 1.33-mev gamma photons are effective. The 0.3-mev beta rays are absorbed by an aluminum jacket around the cobalt cylinder. The inside diameter of the aluminum-jacketed cylinder is 1.5 inches, and the cylinder is 13.5 inches long. The cylinder, in turn, is inside a lead shield about 10 inches thick, with a plugged access hole at the top of shield. The plug is in line with the cylinder so that, with the plug removed, samples to be irradiated can be lowered into the jacketed cobalt irradiator. At the present time, the radiation intensity inside the cylinder is 1280 roentgens per minute. The cobalt has a half-life of 5.3 years. Radiation sources similar to the unit at the University of Michigan have been described in detail elsewhere.

"Measurements were made to determine the maturation-inhibition dose and sterilizing dose (maturation with loss of ability to reproduce) for both

<sup>\*</sup> The kilocurie cobalt source at the University is located in the Waste Fission Products Laboratory, an activity devoted to research on use of radioactive fission by-products and supported by the U.S. Atomic Energy Commission and the Michigan Memorial-Phoenix Project.6

isolated larvae and larvae in trichinous rat muscle. Measurements of the lethal dose were made only on the isolated larvae.

"In determining the maturation-inhibiting and sterilizing radiation levels, doses ranging from 5120 roentgens to 20,480 roentgens were applied. Where larvae were irradiated in muscle, they were subsequently isolated by artificial digestion of the muscle with 1 per cent pepsin and 1 per cent hydrochloric acid. The radiation effect was measured by reduction in the ability of the irradiated larvae to mature in 6 days in the intestinal tract and by the absence of trichina cysts in the muscle tissue of test rate 30 days after ingestion of trichina larvae. Each rat was tube-fed with 5000 larvae. For each radiation dose level, whether applied to isolated trichinae or trichinous rat muscle, at least 6 test animals were used. After 6 days, 3 of the 6 rats were sacrificed and the contents of the small intestines examined for adult worms. The number of adult worms was calculated. 8 When adult worms were present, 50 female worms were examined for the presence within them of embryos.

"The other 3 rats were sacrificed at the end of 30 days. In addition to examination of the contents of the small intestines for adult worms, blocks of tissue from the tongue, diaphragm and muscle of a hind leg were taken for microscopic section and the carcass was then digested for recovery of muscle trichinae. When muscle trichinae were found, the number present in the entire carcass was calculated. In all, 160 rats were used to obtain the data on trichina sterilization and maturation-inhibition with cobalt-60 irradiation.

"The lethal dose for isolated larvae was determined from motility measurements on larvae irradiated at 269,000, 512,000, 768,000, and 1,024,000 roentgens. At the end of each period of irradiation and at 2 hours and 24 hours following irradiation, 200 of the irradiated larvae were examined on a warm stage (43.3°C, 110°F) for evidence of motility; in each case the percentage of motile forms was recorded. Nonmotile (dead) larvae when killed by irradiation often appear tightly coiled under the microscope, so that the mere examination for coiled and uncoiled states does not determine viability. Death due to irradiation does not usually cause the larvae to uncurl as do heat and other killing methods. In addition to microscopic examination 5000 irradiated larvae were fed to 6 test rats as in the case of sterilizing and maturation-inhibiting dose experiments.

"The dose of cobalt-60 radiation necessary to cause sterilization of trichinae while they are still encysted in rat muscle is indicated in Fig. 28. Complete sterilization is reached by a radiation dose of 12,800 roentgens. To achieve the same effect with isolated larvae irradiated in vitro, about 10,000 roentgens are sufficient, as shown in Fig. 29. While these curves are based on the degree of infection found in muscle 30 days after feeding 5000 irradiated larvae, the examination for larval young of 50 adult female worms recovered

from the gut 6 days after irradiation lends to the same conclusions. Data based on this method of examination are presented in Table XVIII.

#### "TABLE XVIII

PERCENTAGE OF ADULT FEMALE TRICHINA WORMS CONTAINING YOUNG LARVAE
RECOVERED FROM GUT OF RAT 6 DAYS AFTER FEEDING COBALT-60
IRRADIATED LARVAE

Each Percentage Is Based on 50 Adult Female Worms Examined, unless Otherwise Specified.

| Radiation Dose  |     | Test Animal        |     | Λ             |            |
|-----------------|-----|--------------------|-----|---------------|------------|
| in Roentgens    | 1   | 2                  | 3   | Average       |            |
|                 |     | - April California |     |               |            |
| 0               | 100 | 100                | 100 | 100           | Irradiated |
| 12,800          | 0   | 2                  |     | 1             | Trichinous |
| 15,360          | 0   | 0                  |     | 0             | Muscle     |
|                 |     |                    |     |               |            |
| 0               | 100 | 100                | 100 | 100           |            |
| 10,240          | 2   | 0                  | 0   | 0.66          |            |
| 10,240          | 2   | 12                 | 0   | 7             | Irradiated |
| 12,800          | 0   |                    | · 2 | 1.0           | Isolated   |
|                 |     |                    |     | (Based on 25) | Trichinae  |
| 15 <b>,</b> 360 | 0   | 0                  | 0   | 0             |            |
| 15,360          | 0   | 0                  | 0   | 0             |            |

Figures 30 and 31 are photomicrographs obtained by courtesy of S. E. Gould, Wayne County General Hospital, Eloise, Michigan. They indicate the effects of gamma radiation on adult female trichinae. Figure 30 is an unusual photograph, showing a normal adult female in the act of giving birth to a larval young. A considerable number of other young can be seen in the body of this female. Figure 31 is a similar photograph of an adult female which has been exposed to a sterilizing dose of gamma radiation (about 12,000 rep) while in the larval stage. Although this female was not killed by this dose and grew to normal adult size, she is incapable of reproducing.

"By irradiation, it is possible to prevent larvae from growing to mature adults when introduced into a suitable host, even though the larvae show normal motility in the warm-stage test. The amount of irradiation required to do this is somewhat higher than that for sterilization. When trichinous muscle

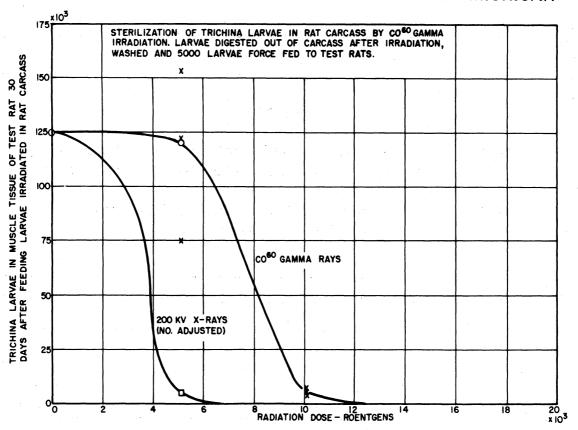


Fig. 28.

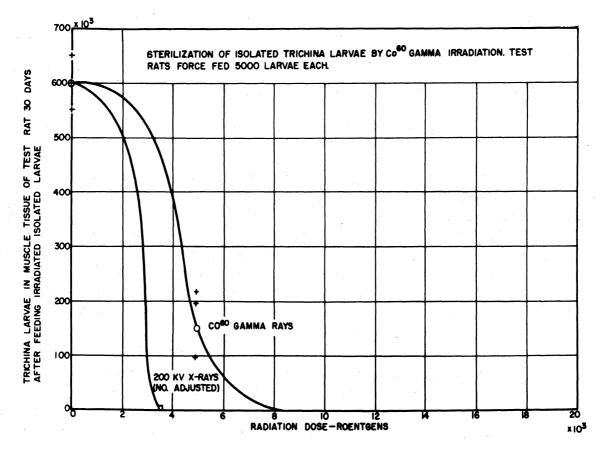


Fig. 29.

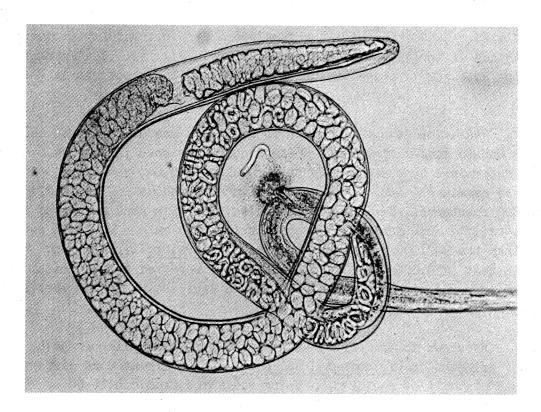


Fig. 30. Normal adult female trichina giving birth to a larval young.

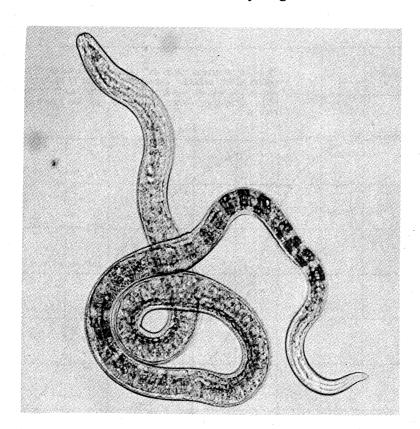


Fig. 31. Adult female trichina which has been exposed to a sterilizing dose of gamma radiation.

is irradiated, 18,000 roentgens are required to inhibit maturation completely. The variation in inhibition with dose is shown in Fig. 32. The maturation-inhibiting dose for trichina larvae in vitro is 15,000 roentgens as shown in Fig. 33.

"Radiation dosages required to kill were measured only on isolated larvae. Substantially higher doses are required as seen in Fig. 34. Complete kill as determined by the motility test requires 750,000 roentgens. If the larvae are examined 2 hours after completion of irradiation, the killing dose is 400,000 roentgens. From previous results, there is good reason to believe that the killing dose for irradiation of trichinous muscle would be substantially higher than the 700,000 roentgens observed for in vitro irradiation. When larvae from this test (250,000 to 1,000,000 roentgens) were fed to test rats, no adult trichinae were recovered from the gut after 6 days and no larvae were recovered from muscle after 30 days.

"As shown in the curves and tables, the life cycle of the causative agent for trichinosis (<u>Trichinella spiralis</u>) can be arreated at different stages by the use of ionizing radiation. Using radiation from cobalt-60 or 200-kilovolt x-rays, it has been possible to prevent reproduction, inhibit maturation, or kill trichina larvae by irradiating them while they are encysted in rat muscle.

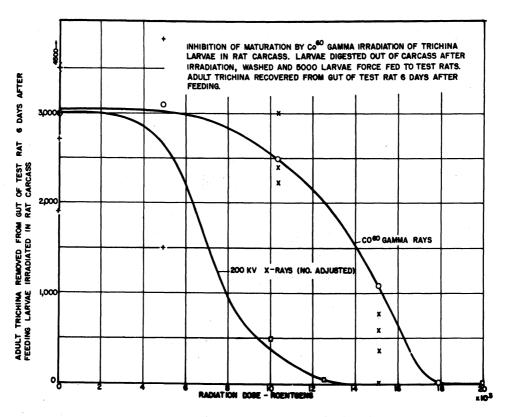


Fig. 32.

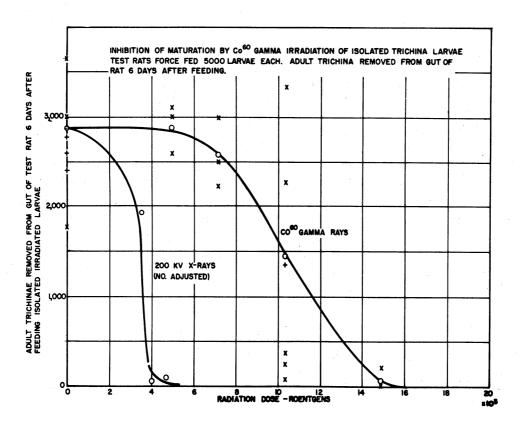


Fig. 33.

"Work is continuing on the irradiation of pork as a possible method of controlling trichinosis. Further studies will proceed along the following lines:

- a) Irradiation of larvae encysted in pork. This work is under way.
- b) Study of the possible use of fission products as low-cost sources of high-energy gamma radiation.
- c) Test of the feasibility of irradiating large volumes of meat such as sections of or whole hog carcasses and still achieving a reasonably uniform radiation dose throughout the meat.
- d) Study of undesirable side effects produced by irradiation.

Undesirable flavor changes occur in many foods when preserving doses of radiation (about two million roentgens)<sup>6</sup> are used. However, one-hundredth of that dose is more than adequate to prevent maturation of encysted trichinae. Preliminary tests of pork irradiated with doses up to 38,400 rep have shown negligible flavor change.<sup>7</sup> A detailed report on this work will be published soon.

"It is also planned to explore further the possibility of disease control through radiation sterilization of the causative agent while it is outside the body. Control of tapeworm, for example, might be possible using an approach similar to that used on the trichinae."

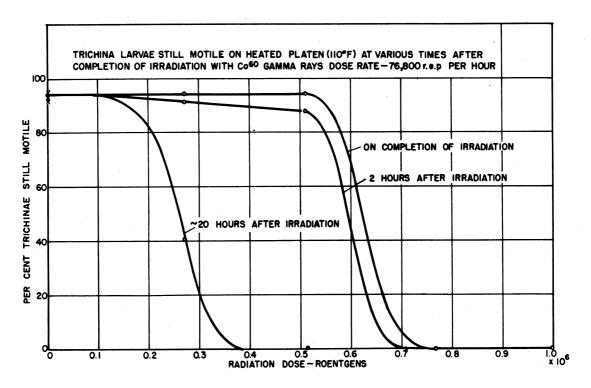


Fig. 34a.

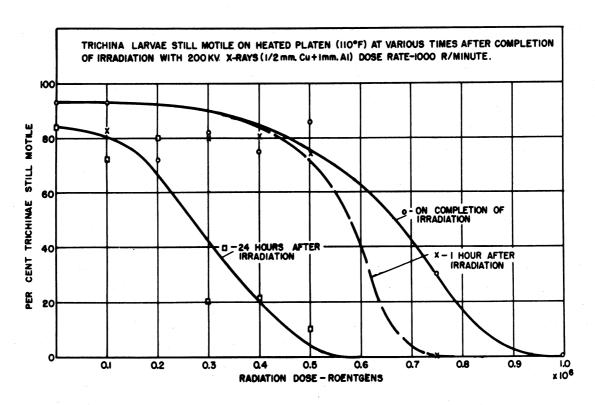


Fig. 34b.

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#### 2. Irradiation of Foods

#### Personnel:

L. E. Brownell, Associate Professor of Chemical and Metallurgical Engineering; L. L. Kempe, Assistant Professor of Bacteriology, Medical School, and Assistant Professor of Chemical and Metallurgical Engineering, Engineering School; J. T. Graikoski, Research Assistant.

Since Progress Report 3, some research on foods exposed to gamma radiation has been continued by Michigan Memorial-Phoenix Project No. 41. Part of the research has been devoted to investigation of the flavor of irradiated meats. There has also been an investigation of the formation of peroxides in irradiated fats.

a. Organoleptic Tests of Irradiated Cured Meats. One of the problems encountered in the sterilization of food products with ionizing radiation is the development of "off-flavors" during irradiation. These off-flavors are more pronounced in animal products such as meat, milk, etc., than in vegetables and fruits. It has been observed that "cured" or treated meats such as ham, corned beef, and bacon undergo less flavor change as a result of irradiation than do fresh meat.

Scores of samples of different brands of corned beef, bacon and cured hams were tested organoleptically after irradiation with gamma rays from the 1000-curie cobalt-60 source. In all cases samples received sterility doses (approximately 2 x 10<sup>6</sup> rep in air). Samples were irradiated in glass tubes sealed with paraffin-lined rubber stoppers. All organoleptic tests were carried out by a panel of 3 to 6 individuals accustomed to the taste of irradiated foods. The irradiated samples were tested in both the cooked and uncooked state. The corned beef was cooked for about one hour in distilled water, while ham and bacon were fried over moderate heat. Table XIX summarizes some of the results obtained.

Generally speaking the results indicated good flavor for most of the samples tested. The smoky odor of ham was decreased by irradiation. Also in some samples a definite but slight tallowiness was detectable in the fatty protion of the samples, which disappeared in most cases upon cooking. The elimination of the tallowiness appeared to depend on the extent of cooking of the sample.

The irradiated corned beef was not as deep red in color as the control, but had an appealing color. It was thought that the irradiated sample was more tender than the control.

TABLE XIX

ORGANOLEPTIC TESTS OF IRRADIATED CURED MEATS

| <b>a</b> 1                 | Raw       | •   | Cooked |  |  |  |
|----------------------------|-----------|---|--------|--|--|--|
| Sample                     | Odor      | Taste                                     | Odor   | Taste                                    |  |  |
|                            |           |   |        |  |  |  |
| Ham, smoked 5 varieties    | Smokiness | Slight<br>Tallowiness                     | Good   | Good; Slight tallowiness on some samples |  |  |
| Corned Beef<br>3 varieties | Good      | Good                                      | Good   | Good                                     |  |  |
| Bacon                      | Good      | Slight tallowiness of fat in some samples | Good   | Good                                     |  |  |

It would seem from these test that some of the off-flavors present in irradiated meats are the result of the action of radiation on the lipid components present. Samples of lean freshly ground beef were mixed with various concentrations of beef fat and it was found that those samples with the greatest percentage of fat developed the greatest off-flavor.

Since cured meats contain various concentrations of sodium nitrate and sodium nitrite, it seemed desirable to treat samples of fresh meat with these chemicals in order to check whether or not these agents are responsible for the protective effect. Samples of ground beef were therefore treated with various concentrations of sodium nitrite and sodium nitrate alone or in combinations. The ground meat was thoroughly mixed with a minimum amount of an aqueous solution of the chemicals and the sample was then allowed to stand for about two hours in order for diffusion to take place. It was found that a final concentration of 100 ppm was necessary for definite flavor protection and that sodium nitrite is mainly responsible for the protective action. In testing the extent of protection, samples of irradiated treated and untreated meat were diluted with quantities of fresh meat of the same batch before cooking and tested organoleptically. Results of a typical test are shown in Table XX. Table XX indicates that a certain degree of flavor protection was afforded by pretreatment with sodium nitrite. Increasing the concentration of sodium nitrite increased the protective effect but meat so treated had a red color even after cooking, which was thought to be undesirable. It was also observed that the raw beef treated with sodium nitrite developed a gray-brown color before irradiation, with color reversion after irradiation. Upon cooking the meat turned to the characteristic gray of cooked meat with a very slight indication of a reddish tinge.

TABLE XX

# INFLUENCE OF SODIUM NITRITE ON FLAVOR OF IRRADIATED RAW BEEF

| Dilution   | Organoleptic Tests | of Irradiated Beef |
|------------|--------------------|--------------------|
| with       | Untreated          | Treated with       |
| Fresh meat |                    | 100 ppm NaNO2      |
|            |                    |                    |
| 0          | Off-flavor         | Off-flavor         |
| 1/10       | Off-flavor         | Fair               |
| 1/20       | Off-flavor         | Good               |
| 1/30       | Good               | Good               |

In testing various batches of meat using sodium nitrite as a protective agent, variable results were obtained. With some batches no off-flavors were detected organoleptically after pretreatment with 100 ppm of NaNO<sub>2</sub>; with others variable amounts of off-flavor were obtained, even though care was taken to treat all samples equally.

It is difficult to explain these different results except on the basis of the condition of the meat before treatment. It is a well-established fact that the compositions of meat, especially the lipid fractions, depends on the feed which the animal receives. Even different cuts from same animal have been shown to vary in the composition of the fat. Also the previous conditions of storage, such as temperature, have a definite effect on the quality of the meat. It would be highly desirable to have a more accurate means of measuring the quality of meat than by organoleptical tests.

# <u>b. Effect of Gamma Radiation on Natural Fats. Preliminary Experiments on Peroxide Values of Animal Fats.</u>

(1) Review. In organoleptic tests on irradiated meats and meat products, it was observed that samples receiving sufficient doses for sterility developed a slight but definite "tallowiness". This is often referred to as the "oxidative" type of rancidity in contrast to the "hydrolytic" type resulting from the action of microorganisms or enzymes on the fat producing free fatty acids. The oxidative type of rancidity is believed to be caused by the absorption of oxygen by the unsaturated linkages with the formation of peroxides or peroxide-like components with subsequent break-down into lower homologs. These lower homologs, such as aldehydes, ketones, and lower-chain fatty acids, are thought to impart the "off-flavors" present in rancid fats. Although much work has been done on the rancidity of edible fats, the exact compounds responsible fot the "off-flavors" have not been firmly established.

Many methods have been devised to test the rancidity of edible fats; oxygen stability, peroxide determination, and the Kreis test are the most widely used. The peroxide value of fat is expressed as the milli-equivalents of oxygen absorbed by 1000 grams of fat, whereas the Kreis reaction is the action of an etheral solution of phlorozlucinol on the fat in question. In the Kreis test a definite red color indicates rancidity and is believed to be caused by the presence of epihydrin aldehyde (CH2CHCHO). Efforts have been made to correlate organoleptic tests with chemical tests in order to determine degree of rancidity, since organoleptic techniques are subject to human variables. However, the true significance of such correlations is open to question.

The subject of the effect of ionizing irradiation on fats has not been studied extensively. With nonionizing irradiation a certain amount of work has been performed. In general, nonionizing irradiation is reported to produce increases in the peroxide value, in the acid value, in the substance responsible for the Kreis reaction, and in viscosity; and a decrease in iodine number. I

With ionizing irradiation an early report indicates that the dielectric constant, acid value, and iodine number showed the greatest change when olive oil was exposed to x-rays and radium. 5 In investigations of alpha particles on fatty acids both decarboxylation and dehydrogenation were observed.6,7 In preliminary experiments it has been reported that gamma rays (cobalt-60) have little effect on the iodine number and acid value of oleic acid exposed at ordinary temperatures and pressures. 8 X-radiation has been reported to induce the auto-oxidation of linoleic acid.9 Using electrons from a Van der Graff accelerator, a definite increase in the apparent peroxide value of butter occurs. 10,11 The effect occurs even at low temperatures, but the storage life can be extended by addition of anti-oxidants. A recent publication reports that polymerization, bond breakage, and oxidative changes occur in fish and vegetable oils when subjected to a high-intensity electron burst from the capacitron. 12 An increase in peroxide oxygen also occurred, with further increase on subsequent storage. Inert atmospheres and anti-oxidants suppressed the formation of peroxides. Correlation between organoleptic and chemical reactions of irradiated oils was not possible. Organoleptic tests showed that rancidity was not as pronounced as the peroxide values would indicate.

(2) Experimental Tests. For experimental determination of peroxide values the ferric thiocyanate method as described by Loftus-Hills and Thiel<sup>13</sup> was used. Others have modified this method for use with a photoelectric colorimeter. In the present study the method of Loftus-Hills and Thiel was used but was modified so that determinations could be made with a photoelectric colorimeter. Essentially, the method consists of the oxidation of the ferrous to the ferric ion in presence of ammonium thiocyanate, and reading the intensity of the red color produced. This method, although not widely used in this country is considered to be more sensitive than the iodiometric method of Lea; it gives consistantly higher peroxide values, 15 i.e., values approximately 2 times those

obtained by iodiometric methods. Solutions used in the procedure were prepared as follows:

Hydrated barium chloride (0.4 gm) is dissolved in 50 ml of distilled water and the solution is added slowly to 0.5 gm of ferrous sulfate dissolved in 50 ml of distilled water. The resulting solution is acidified with 2 ml of lON HCl. After the precipitate of barium sulfate settles, the solution is decanted and protected from light. Stability of the reagent must be checked before use.

A solution of ammonium thiocyanate is made by dissolving 30 gm in distilled water and diluting to 100 ml.

A standard curve for use with the colorimeter is obtained by dissolving bright iron wire in iron-free 10N HCl, and oxidizing with hydrogen peroxide. The excess  $\rm H_2O_2$  is boiled off and the solution adjusted to give 10 mg iron and 1 ml 10N HCl per 100 ml. In preparation of the curve various dilutions are made from the standard iron solution and readings are obtained by the same procedure as is used in fat analysis. Values for known concentrations of iron are plotted on semi-logarthmetic paper.

A solvent for fat extraction of 70 volumes of benzene and 30 volumes of methanol was used. Good solubility of natural fat occurred except for small parts of protein and connective tissue which remained undissolved. Dissolved fat was determined by weight.

In the analysis of the fat, 1 gm of fat was dissolved in an appropriate volume of solvent. To each of three matched tubes containing 9.9 ml of solvent was added 0.1 ml of this solution, followed by 0.05 ml of ammonium thiocyanate and 0.05 ml of ferrous chloride. After thorough mixing the tubes were placed for 2 minutes in a water bath adjusted to 50°C. The tubes were then cooled and immediately read in a photoelectric colorimeter (Lumetron Model 400-G) using a No. 530 filter. A blank consisting of all reagents except the fat was used. This blank compensated for slight amounts of ferric ion present in the test reagents and slight oxidation of ferrous ion due to other factors. The blank determination varied from 95 to 100 per cent transmission. Concentration of ferric ion per 10 ml of solvent was obtained from the standard curve prepared previously.

Peroxide values were calculated from the following relationship:

Peroxide value m.e. 
$$0_2/\text{kgm fat} = \frac{\text{mgm Fe}^{+3}/10 \text{ ml solvent}}{\text{gm fat x 55.84}}$$

For irradiation purposes, samples of beef and pork fat free of protein material were selected from a local market. A few samples of bacon fat and lard were also included in the study. About 30 gm of samples were exposed to

gamma radiation (cobalt-60) in rubber-stoppered glass tubes. Controls were handled in the same way except they received no irradiation. The analyses were conducted immediately after the last irradiation.

(3) Results. Results of some preliminary experiments are presented in Tables XXI, XXIII, XXIII, and Fig. 35. Since the methods were being established when these runs were made, the results should be used only for comparison between irradiated and control fats.

TABLE XXI

EFFECT OF GAMMA RADIATION ON SOME ANIMAL FATS

| Fat Sample             | Dose   | Peroxide Valuem.e. 02/kg fat |                |  |  |
|------------------------|--|------------------------------|----------------|--|--|
|                        | (Rep in Air)                                     | Control                      | Irradiated     |  |  |
| Pork<br>No. 1<br>No. 2 | 1.83 x 10 <sup>6</sup><br>2.07 x 10 <sup>6</sup> | 0                            | 48.54<br>58.75 |  |  |
| Beef<br>No. 1<br>No. 2 | 1.83 x 10 <sup>6</sup><br>1.83 x 10 <sup>6</sup> | 0                            | 26.62<br>1.97  |  |  |
| Bacon Fat              | 2.07 x 10 <sup>6</sup>                           | 0                            | 34.21          |  |  |
| Lard                   | 2.07 x 10 <sup>6</sup>                           | 5.01                         | 15.04          |  |  |

#### TABLE XXII

PEROXIDE VALUES OF BEEF SUET EXPOSED TO VARIOUS DOSES OF GAMMA RADIATION (COBALT-60)

|          | Danie                  | Peroxide Value    |       |   |       |  |
|----------|------------------------|-------------------|-------|---|-------|--|
| Sample   | Dose                   | After Irradiation |       | 7 Days at                               | 25°C  |  |
|          | (Rep in Air)           | Control           | Irrad | Control                                 | Irrad |  |
| Beef Fat | 7.68 x 10 <sup>4</sup> | 0                 | 17.55 | 0 /                                     | 36.64 |  |
|          | 2.30 x 10 <sup>5</sup> | 0                 | 16.36 | 0                                       | 47.82 |  |
|          | $9.22 \times 10^{5}$   | 0                 | 10.69 | 0 - 2 - 2 - 2 - 2 - 2 - 2 - 2 - 2 - 2 - | 57.48 |  |
|          | 1.83 x 10 <sup>6</sup> | 0                 | 3.45  | 0                                       | 39.08 |  |

#### TABLE XXIII

# PEROXIDE VALUES OF PORK FAT STORED AT 4°C AFTER IRRADIATION WITH VARIOUS DOSAGES

| Sample   | Dose                                     |               | Peroxide Value<br>Days at 4°C |       |                |        |        |
|----------|--|---------------|-------------------------------|-------|----------------|--------|--------|
|          | (Rep in Air)                             | 0             | <u> </u>                      | 8     | 13             | 17_    | 22     |
| Pork Fat | 0  | 0             | 0                             | 0     | 0              | 0      | 0      |
|          | 3.80 x 10 <sup>4</sup>                   | 0.90          | 2.18                          | 2.24  | 6.77           | 9:98   | 26.57  |
|          | $7.68 \times 10^4$                       | 7.23          | 12.18                         | 13.93 | 31.64          | 48.68  | 60.35  |
|          | $7.68 \times 10^4$<br>$1.69 \times 10^6$ | 91.76         | 99.67                         | 97.87 | 99.67          | 108.02 | 120.25 |
|          | 140                                      |               |                               | T - T | - T            | ٦ .    |        |
|          |  |               |                               |       |                |        |        |
|          | 120                                      |               |                               |       |                |        |        |
|          | FAT)                                     |               |                               |       | 1.69 x 106 REP |        |        |
|          | ₩ 100                                    | $\rightarrow$ |                               |       |                | -      |        |

EQUIV.

PEROXIDE VALUE (M.

Fig. 35. Increase in peroxide value of pork fat stored at 4°C and receiving different dosages of gamma radiation.

DAYS AT 4°C

7.68 x 10<sup>4</sup> REP

CONTROL

The peroxide values obtained were found to vary with different lots of fat (Table XXI). This is believed to be caused by variations in the samples of fat such as moisture content and degree of unsaturation of fats. Table XXII shows an apparent decrease in peroxide value with increased dosage, which is difficult to explain. The same result was also observed in another experiment using a different lot of beef suet. With pork and beef fat there is a definite

continuous increase in peroxide oxygen, on storage (Tables XXII, XXIII, Fig. 35). It was of interest to note that the control of the pork samples stored at 4°C developed a high degree of rancidity as evaluated organoleptically, after 22 days, whereas irradiated samples maintained a characteristic type of tallowiness for a much longer period of time before developing rancidity. This probably resulted from a decrease in the original concentration of microorganism by radiation, which depressed the hydrolytic type of rancidity.

Organoleptic tests of irradiated samples did not prove that they were objectionable. Samples had a definite tallowiness which was recognized as being pronounced more by odor than by taste. Samples of irradiated fat studied did not give a Kreis reaction (production of red color in etheral solution of phloroglucinol).

In conclusion, it can be stated that irradiated fats definitely show an increase in peroxide oxygen. The exact relationship to organoleptic tests awaits further study. In the interest of utilizing radiation for food sterilization, the stability of irradiated fats must be determined and the relationship with toxicity of such products ascertained. Toxicity studies of oxidized fats have shown them to produce a disease called "oxidized fat syndrome" in dogs and rats. 16,17

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#### 3. Animal Feeding Experiments

#### Personnel:

These experiments will be under the supervision of Professor H. Lewis, Chairman of the Department of Biological Chemistry, Medical School and H. C. Eckstein, Professor of Biological Chemistry. Irradiation of foods will be performed by L. E. Brownell, Supervisor of Fission Products Laboratory, and J. T. Graikoski, Research Assistant. The description of the proposed experiment was prepared by L. E. Brownell.

a. Preliminary Tests. Preliminary animal feeding experiments with food exposed to gamma radiation were conducted by F. H. Bethell, M.D., Chief Investigator, and A. H. Kretchmar, M.D., Research Associate, in the Biological Effects of Irradiation Laboratory, University of Michigan. These experiments were reported in Progress Report 2. The results showed no difference in health, growth, etc., between the animals fed irradiated reconstituted whole milk and those fed the controls. All milk samples were irradiated in polyethylene bags in the 1000-curie cobalt-60 source, receiving a dose of slightly over two million rep (in air). The milk was reconstituted Klim prepared as a four-times concentrate and diluted just before feeding by the addition of a salt solution. This milk constituted the entire diet, with the exception of one nonirradiated leaf of lettuce added each week. It was emphasized in Progress Report 2 that this experiment was only exploratory and that any conclusions from the data were preliminary and contingent on further testing, both with feeding experiments and with more refined methods of assay for individual components of the diet. This would require the setting up of long-term feeding and breeding experiments.

Although these preliminary feeding experiments were negative, the results in the previous section indicated that appreciable quantities of peroxides were formed as a result of irradiation of fats and that the amount of peroxide was greater for fats containing larger percentages of unsaturated fatty acids. Apparently one undesirable effect of the formation of peroxides in the destruction of the tocopherols in the fats, which results in loss of vitamin E and the characteristic symptoms of vitamin E defficiency. Although this could be corrected by feeding supplements of vitamin E, the present state of knowledge would indicate the undesirability of irradiation the major portion of the unsaturated fatty acids in the diet. Various tests by other research groups have indicated partial destruction of some of the other vitamins as a result of irradiation. Ascorbic acid and some of the B complex vitamins appear to be sensitive to radiation. This would indicate the undesirability of irradiating the vitamin supplement of a diet.

However, there seems to be no information which would indicate the undesirability of irradiating the proteins, the principal food constituent responsible for growth, or the carbohydrate, usually the principal energy constituent of a diet. It is believed that this is the question which should be investigated in the long-term feeding experiment. Such an experiment should not be used to establish vitamin losses or formation of peroxides which can be investigated by simpler and shorter experiments. However, the fats which would normally be present in foods which might be irradiated for human consumption should be present in the diet of the animals in the feeding experiment to check the effects of irradiating a portion of the fats in the diet. Also, a portion of the water intake should be in the irradiated food, as some of the effects of irradiation have been attributed to the formation of hydrogen peroxide from water as a result of irradiation.

Some of these questions regarding the proposed long-term animal experiments have been discussed with the personnel of other laboratories that have conducted similar experiments using food sterilized by electron bombardment from accelerating machines. There has also been some discussion of the proposed experiments in correspondence with representatives of the Food and Drug Administration. The suggestion has been made that meat would be a more suitable and convenient food than reconstituted milk because of the difficulties of obtaining normal feeding with a completely liquid diet.

The feeding experiment to establish the wholesomeness of irradiated food should be independent rather than combined with an experiment which might involve the dosage of radiation required for sterility. If the animals are fed food in which the wholesomeness of the food is independent of the requirements of sterilization by radiation, using food such as canned milk or canned meat which is sterilized thermally, the variable of radiation dosage required to sterilize the foods will not influence the experiment. This precaution will prevent the possible loss of animals from food spoilage, which would destroy the experiment to investigate the wholesomeness of radiated food. Experiments to investigate further the dose of gamma radiation necessary to produce sterility in canned foods may be conducted simultaneously and separately and at less cost by microbiological studies rather than by animal feeding experiments. Perhaps the best reason for using thermally sterilized meat in the feeding experiments is to obtain a uniform control. If raw meat were canned in the laboratory and sterilized by gamma radiation, an inventory of irradiated food sufficient for feeding for two or more weeks would have to be kept on hand so as to allow for shutdowns, repairs, or other experiments with the radiation source. This would not be possible with the control food, which could be kept for only short periods because of the danger of spoilage. If canned and thermally sterilized food is used both for the control feeding and for feeding irradiated food, a more uniform control will result and problems of storage of perishable foods will be minimized.

<u>b. Proposed Experiment.</u> With these different considerations and after consulation with others interested in these experiments, it was decided that the animal feeding experiments should involve the feeding of four generations of rats. A diet should consist of irradiated proteins and carbohydrates supplemented with nonirradiated fats, vitamins, minerals, and some material to add bulk to the diet. The irradiated food may consist of 50 parts of canned meat such as Swift's Chopped Beef for Babies (see Table XXIV for typical analysis), 25 parts of carbohydrates such as corn starch, and 10 parts of dry casein. The nonirradiated supplement may consist of 10 parts of vegetable oil, 5 parts of a vitamin premix such as given in Table XXV, and 2-12 parts of a salt supplement such as given in Table XXVI. In addition, appropriate amounts of cod liver oil will be mixed with the diet at feeding to supply vitamins A and D and wheat germ oil will be given orally by droppers to be sure of the intake of vitamin E. Ground cellulose will be used to add some bulk to the diet.

#### TABLE XXIV

#### SWIFT'S BEEF FOR BABIES

| Protein   | 18.7% | Calcium    | 8 mg/100 gm.      |
|-----------|-------|------------|-------------------|
| Fat       | 3.0%  | Phosphorus | 130  mg/100  gm.  |
| Moisture  | 77.1% | Iron       | 2  mg/100  gm.    |
| Total Ash | 1.2%  | Thiamine   | 0.01  mg/100  gm. |
| Salt      | 0.6%  | Riboflavin | 0.21  mg/100  gm. |
|           |       | Niacin     | 3.39 mg/100 gm.   |

Preliminary experiments will be carried out using nonirradiated foods to determine the amounts of supplementary casein which will be required to assure satisfactory growth and reproduction. This will be done by starting with 36 young rats, 18 male and 18 female, divided into three equal groups. The first group will be given a diet of meat, carbohydrate, fat, bulk (ground cellulose), inorganic salts, and vitamins. The second group will receive the same diet supplemented with 5 per cent casein and the third group will receive the original diet supplemented with 10 per cent casein. The adult rats will be mated and the number and quality of the young determined.

At the successful conclusion of this preliminary experiment to determine the optimum diet, the feeding experiment using irradiated food will be commenced with 24 young rats, 12 male and 12 female, having a weight of 50 to 60 grams each. In this series 12 animals will be used as controls and 12 animals will be fed the identical diet fed the controls except that all the proteins and carbohydrates of the diet will be exposed to a sterilizing dose (at least 2 million rep) of gamma radiation. The adult rats will be mated and the first litter of animals will be discarded after weaning. The mating will be repeated and the second liter of animals will be used to continue the experiment of feeding irradiated food in the same manner as with the first generation. This

#### TABLE XXV

#### VITAMIN PREMIX

| Ingredients                  | Grams                                       |
|------------------------------|---|
| Thiamine                     | 0.400                                       |
| Riboflavin                   | 0.400                                       |
| Pyridoxine                   | 0.400                                       |
| Ca Pantothenate              | 2.200                                       |
| Niacin                       | 2.000                                       |
| Vitamin K                    | 0.500                                       |
| Folic Acid                   | 12.5 mg.                                    |
| Biotin                       | 5.0 mg.                                     |
| P-Aminobenzoic Acid          | 25.0  |
| Inositol                     | 20.0  |
| Choline Chloride             | 50.0  |
| Liver Powder (Wilson's N.F.) | 430.0                                       |
| Yeast (Anheuser-Busch 300)   | 215.0                                       |
| Corn Starch                  | 154.0                                       |
|                              | 899.9 Use as 5% of diet on dry solids basis |

#### TABLE XXVI

#### SALT SUPPLEMENT MIXTURE

Formulated per Hubbell, Mendel and Wakemen, Journal of Nutrition, 14,273 (1937)

| Calcium carbonate              | 54.300% |
|--------------------------------|---------|
| Magnesium Carbonate            | 2.500%  |
| Magnesium Sulfate              | 1.600%  |
| Sodium Chloride                | 6.900%  |
| Potassium Chloride             | 11.200% |
| Potassium Phosphate(monobasic) | 21.200% |
| Ferric Phosphate               | 2.050%  |
| Potassium Iodide               | 0.008%  |
| Manganese Sulfate              | 0.035%  |
| Sodium Fluoride                | 0.010%  |
| Aluminum Potassium Sulfate     | 0.017%  |
| Copper Sulfate                 | 0.090%  |

procedure will be repeated by breeding the second generation (second liter of first generation) to produce a third generation and the third generation (second litter of second generation) to give a fourth generation. During this period records will be kept for both the controls and the animals fed irradiated food. These records will include the rate of growth, the number of offspring, and the general health of the animals. After the second litter has been obtained from the second, third, and fourth generations the parents will be sacrificed for pathological study to check for any abnormalities. The original animals will be kept until the end of the experiment. (The maximum number of rats to be fed at any one time will thus be two generations.) When these experiments are completed, it may be desirable to perform some additional experiments using animals other than rodents, such as dogs or monkeys. However, such experiments with other animals would be of a simpler nature and used only to supplement the results obtained using rats as experimental animals.

In conducting the experiments the supplements will be intimately mixed with the other materials and the diets will be prepared fresh at least every four days and kept refrigerated in the interim. The room will be air conditioned by means of a 3-ton unit so as to maintain proper circulation of fresh air at a uniform temperature, but no attempt will be made to control the humidity. The animals will be caged separately in individual cages so that the food consumption can be determined accurately. These cages will be washed and sterilized at least weekly.

Other shorter experiments in which canned raw food sterilized by gamma radiation is fed, and experiments in which all the diet except the vitamin supplement is irradiated, may be conducted with the excess animals from the second, third and fourth generations of rats.

### 4. Irradiation of Soya-Alkyd Film

In cooperation with Phoenix Project 28 (supervised by Professor L. L. Carrick, with Mr. A. J. Permoda as research assistant), some soya-alkyd films were exposed to x- and gamma radiation. The purpose of these tests was to determine the effect of x- and gamma radiation on such films, which are used as protective coatings.

The following description of these tests is selected from the report entitled, "Effect of Gamma-, X-, and Ultraviolet Radiation on the Stress-Strain Properties of Soya-Alkyd Films", by L. L. Carrick, J. T. Banchero, and A. J. Permoda; Paper No. 2, Michigan Memorial-Phoenix Project 28, no part of which is to be reproduced without the permission of the authors.

"This report covers an exploratory investigation of the aging cycle and/or exposure to radiations of a number of the test soya-alkyd coatings and on one soya-oil coating that were used in the contemporary investigation mentioned above. An investigation into the effect of higher-energy shorter-wavelength radiations on protective-coating films was believed pertinent because of the well known accelerated weathering and aging of such films when exposed to ultraviolet radiations. If the shorter gamma and x radiations were appreciaably absorbed by these test coatings, aging should follow according to the Planck Equation:

Energy = Planck constant x Radiation frequency.

a. "Coating Preparation. All the alkyd pigmented coatings were ground in laboratory-sized pebble mills for 72 hours and had a minimum grind of 7 as determined by the Hegman Gage. The viscosity of the coating material was adjusted with solvent for easy application by means of the Baker film applicator. The drier consisted of 0.02% Mn and 0.06% Zn metal based on the vehicle solids.

"All coatings were deposited on tin-coated steel panels by an electrically driven draw-down apparatus and a film applicator in order to insure a uniform film deposition. The dry film thickness was approximately 1.2 mils. The soya-alkyd drying cycle consisted of 30 minutes of air drying followed by 35 minutes of force drying at 295°F in an electrically heated oven provided with air circulation. The coatings were stripped from the tin-coated panels by immersion in a mercury bath. The dislodged coatings were subsequently brushed clean and cut into 0.5-inch strips (lateral to application direction) Scotchtaped to paper in order to form grip tabs for the ensuing tests in a Gardner Laboratory tensile-tester having a grip separation of 2 inches. This tensile apparatus, which employs a tempered spring for load application, was used in all the contemporary investigations; the apparatus does not provide either a constant rate of load application or a constant rate of elongation, since the apparatus is constructed to give a constant rate of load application only with nonelongating films (a load application of 100 grams per 0.93 minute under the latter conditions was used in these tests). The stress-strain relationship for this apparatus, however, may be correlated by the following derived equation:

t = aS + be

t = time in minutes required to rupture film,

S = stress at rupture,

e = elongation at rupture,

a = constant varying with film thickness, and

b = constant varying with load application, which was fixed
 in these tests.

"Both the specimen condition and the stress-strain determination were conducted in a constant-temperature constant-humidity room at 77°F and 50% R.H. The film thickness was determined by a dial gage prior to testing. The reported values for breaking stress and elongation (Table XXVII) are the averages determined from the indicated number of strips. Considerable variation was often present between specimens, especially between unpigmented and also between the fresh films. Extreme variations in elongation were discarded according to a statistical method for a small number of samples.<sup>1</sup>

TABLE XXVII
STRESS-STRAIN DATA ON CONTROL TESTS FILMS

| Test | Film<br>Life,<br>Days | Number<br>of<br>Films | Detached<br>Film<br>Life,days | Breaking<br>Stress,<br>kg/cm <sup>2</sup> | Breaking Elongation % | Toughness (Area) ** | Thickness mils |
|------|-----------------------|-----------------------|-------------------------------|---|-----------------------|---------------------|----------------|
| (U   | Inpigmen              | nted, Soy             | a-Alkyd)                      |   |                       |                     |                |
| 1    | 3                     | 21                    | 1                             | 78.5                                      | 43.9                  | 31.5                | 1.13           |
| 2    | 20                    | 18                    | 5                             | 78.4                                      | 47.1                  | 32.6                | 1.23           |
| 3    | 72                    | 31                    | 65                            | 96.8                                      | 42.3                  | 35.8                | 1.29           |
| 4    | 185                   | 10                    | 172                           | 108.0                                     | 72.7                  | 72.1                | 1.36           |
| 5a   | 212                   | 13                    | 1                             | 79.0                                      | 42.2                  | 28.0                | 1.20           |
| 11*  | 77                    | 11                    | 1                             | 127.3                                     | 82.6                  | 88.8                | 1.38           |
| ( A  | lkvd-Ur               | ea, 7:3)              |                               | •   |                       |                     |                |
| 12   | 9                     | 15                    | 7                             | 157.5                                     | 27.8                  | 37.3                | 1.41           |
| 13   | 19                    | 10                    | 3                             | 202.8                                     | 17.5                  | 29.6                | 1.64           |
| 14   | 105                   | 18                    | 103                           | 234.3                                     | 12.0                  | 20.2                | 1.38           |
| 17   | 20                    | 9                     | ĺ                             | 217.5                                     | 10.4                  | 17.0                | 1.34           |
| 19   | 70                    | 26                    | 67                            | 209.0                                     | 9.6                   | 16.6                | 1.58           |
| (Δ   | lkvd_Ti               | 0. 0.17               | micron, 25%                   | ( PVC)                                    | •                     |                     |                |
| 21   | : 3                   | 30                    | 1                             | 103.9                                     | 44.2                  | 30.2                | 1.18           |
| 22   | 10                    | 14                    | 1                             | 116.3                                     | 45.3                  | 36.3                | 1.31           |
| 23   | 31                    | 8                     | _<br>1                        | 113.8                                     | 36.3                  | 29.1                | 0.92           |
| 24   | 60                    | 24                    | -<br>55                       | 121.0                                     | 33.6                  | 29.3                | 1.24           |
| 25   | 77                    | 11                    | 75                            | 133.2                                     | 30.0                  | 29.5                | 1.16           |
| 26   | 185                   | 21                    | 178                           | 135.5                                     | 35.4                  | 38.0                | 1.25           |
| (A)  | lkyd-Ti(              | 0.07                  | micron, 25%                   | PVC)                                      |                       |                     |                |
| 34   | 5                     | 11                    | 1                             | 83.1                                      | 49.5                  | 18.5                | 1.16           |
| (A)  | Lkvd-TiO              | 0.06                  | micron, 25%                   | PVC)                                      |                       |                     |                |
| 38   | 7                     | 11                    | 5                             | 81.2                                      | 34.5                  | 14.0                | 0.93           |
|      |                       |                       |                               | <del></del>                               |                       |                     |                |

TABLE XXVII (cont.)

| Test<br>No.  | Film<br>Life,<br>Days | Number<br>of<br>Films | Detached<br>Film<br>Life,days | Breaking<br>Stress,<br>kg/cm <sup>2</sup> | Breaking<br>Elongation<br>% | Toughness<br>(Area)<br>** | Thickness mils |
|--------------|-----------------------|-----------------------|-------------------------------|---|-----------------------------|---------------------------|----------------|
| (A)          | lkvd-TiC              | 0.17                  | micron, 10%                   | PVC)                                      |                             |                           |                |
| 42           | 3                     | 13                    | 1                             | 86.6                                      | 28.5                        | 21.5                      | 0.93           |
| 43           | 32                    | 9                     | 1                             | 94.8                                      | 31.3                        | 24.0                      | 1.00           |
| (A)          | Lkyd-TiC              | <sub>2</sub> , Ruti   | le, 0.4 micr                  | on, 25% PV                                | C)                          |                           |                |
| 47           | 10                    | 17                    | 8                             | 89.2                                      | 8.7                         | 6.3                       | 1.30           |
| 48           | <b>3</b> 0 .          | 12                    | 28                            | 127.2                                     | 19.5                        | 19.7                      | 1.13           |
| 49           | 105                   | 22                    | 103                           | 138.2                                     | 15.8                        | 17.6                      | 1.06           |
| (A)          | _k <b>y</b> d-CaC     | 03, 0.17              | micron, 25%                   | PVC)                                      |                             |                           |                |
| 50           | 6 .                   | 11                    | 2                             | 76.6                                      | 24.2                        | 13.6                      | 1.07           |
| 51           | 32                    | 10                    | 2                             | 79.0                                      | 19.4                        | 12.9                      | 0.97           |
| (A)          | kvd-CaC               | 0z. 0.09'             | 7 micron, 25                  | % PVC)                                    |                             |                           |                |
| 57           | 7                     | 13                    | 5                             | 82.3                                      | 44.2                        | 26.0                      | 1.09           |
| <b>(</b> Δ ٦ | kvd-CaC               | 0~. 0.07)             | + micron, 25                  | % PVC)                                    |                             |                           |                |
| 61           | 7                     | 11                    | 5                             | 83.8                                      | 51.1                        | 24.8                      | 1.10           |
| 62           | 63                    | 17                    | 6                             | 84.5                                      | 21.2                        | 13.6                      | 1.27           |
| ( A 1        | kwd_BaS               | n, n 5-0              | 2.0 micron,                   | 03% DVC)                                  |                             |                           |                |
| 68           | 10                    | 13                    | 8                             | 90.3                                      | 48.2                        | 27.8                      | 1.38           |
| 69           | 10                    |                       |                               | 81.1                                      |                             | 28.8                      |                |
|              |                       | 13                    | 1                             |   | 53.1                        |                           | 1.17           |
| 70           | 74                    | 13                    | 61                            | 110.0                                     | 47.0                        | 34.7                      | 1.10           |
| 71           | 106                   | 10                    | 104                           | 133.0                                     | 45.5                        | 47.4                      | 1.29           |
|              | kyd-BSW               |                       | 4 micron, 2                   | •   |                             |                           |                |
| 76           | 9                     | 14                    | 7                             | 81.2                                      | 4.7                         | 2.96                      | 1.25           |
| 77           | 74                    | 14                    | 66                            | 125.2                                     | 6.2                         | 6.30                      | 1.27           |
| 78           | 110                   | 8                     | 108                           | 119.2                                     | 18.6                        | 17.9                      | 1.48           |
|              | kyd-Carl              | bon Black             | c, 0.078 mic                  | con, 5% PVC                               | C)                          |                           |                |
| 83           | 3                     | 13                    | 1                             | 107.2                                     | 43.1                        | 41.5                      | 0.93           |
| 84           | 34                    | 13                    | 2                             | 87.0                                      | 45.5                        | 31.8                      | 1.09           |
| (A1          | kyd-Carl              | oon Black             | ., 0.078 mic                  | on, 10% PV                                | rc)                         |                           |                |
| 93           | 3                     | 9                     | 1                             | 115.0                                     | 47.8                        | 42.6                      | 0.91           |
| 94           | 9                     | 12                    | 1                             | 97.6                                      | 49.0                        | 35.1                      | 1.15           |
| 95           | 29                    | 26                    | 1                             | 119.5                                     | 48.1                        | 44.1                      | 1.07           |
|              | •                     |                       |                               | _ 80                                      |                             |                           | •              |

TABLE XXVII (cont.)

| Test  | Film<br>Life,<br>Days | Number<br>of<br>Films | Detached<br>Film<br>Life,days | Breaking<br>Stres,<br>kg/cm <sup>2</sup> | Breaking<br>Elongation | Toughness (Area) ** | Thickness mils |  |
|---|-----------------------|-----------------------|-------------------------------|--|------------------------|---------------------|----------------|--|
| 96  | 60                    | 26                    | 50                            | 134.8                                    | 52.9                   | 55.5                | 1.04           |  |
| 97  | 175                   | 6                     | 150                           | 137.0                                    | 51.3                   | 55.3                | 1.03           |  |
| (Al   | kyd <b>-</b> Car      | bon Blacl             | k, 0.31 micr                  | on, 10% PV                               |                        |                     |                |  |
| 106   | 3                     | 13                    | 1                             | 68.2                                     | 46.2                   | 24.3                | 1.52           |  |
| 107   | 30                    | 13                    | 1                             | 104.7                                    | 60.0                   | 39.0                | 1.37           |  |
| (Soya Oil-Carbon Black, 0.31 micron, 10% PVC) |                       |                       |                               |  |                        |                     |                |  |
| 116   | 3                     | 10                    | 1                             | 20.6                                     | 20.7                   | 1.12                | 1.17           |  |

<sup>\*</sup> This unpigmented soya-alkyd film was deposited, and then aged without baking at 77°F and 50% R.H. for 75 days. It then was baked under the standard schedule, stripped, and tensile-tested on the 77th day.

- "Discussion of Results. The drier content and the baking schedule selected for the soya-alkyd films were within the range recommended by the resin manufacturer for industrial application of this vehicle and were specifically selected for the test films because they conferred desirable stressstrain and hardness characteristics on these coatings. The graphs, Figs. 36-43 and data, Table XXVII. show the effect of the given amount of gamma or x-radiation on the test films. Existing facilities did not permit uninterrupted exposure to these radiations and required that a nonirradiated control be run simultaneously in order to ascertain the specific effect of the irradiation on the test films as separated from the normal aging effect. The gamma irradiation were accomplished in a cobalt-60 vault which was calibrated as having an intensity of approximately 100,000 roentgens per hour (radiation protection for humans advises against exposures beyond 0.3 roentgen per week over the entire body). The x-irradiations were accomplished with a W target employing 210 kv and 15 ma across the tube. The distance from the tube window to the film was under 12 inches, and the 500,000-roentgen exposures required approximately 11 hours of exposure time. Air was circulated over the films during exposure in order to maintain the films at room temperature.
- c. "Comparison of Irradiated and Non-Irradiated Test Films. Table XXVIII and Fig. 36 give data on the irradiated unpigmented alkyd vehicle. Both gamma and x-irradiation increase the tensile strength of the films somewhat, with a similar increase in film toughness, curve 7 vs 3, and curve 6 vs 2;

<sup>\*\*</sup> The toughness values of these films were determined with a planimeter as the area under the stress-elongation curve, and are a measure of the work required to rupture the film.

however, additional aging of this same alkyd vehicle exposed to x-irradiation reverses the trend in that the irradiated film is less tough than its control, curve 4 vs 5.

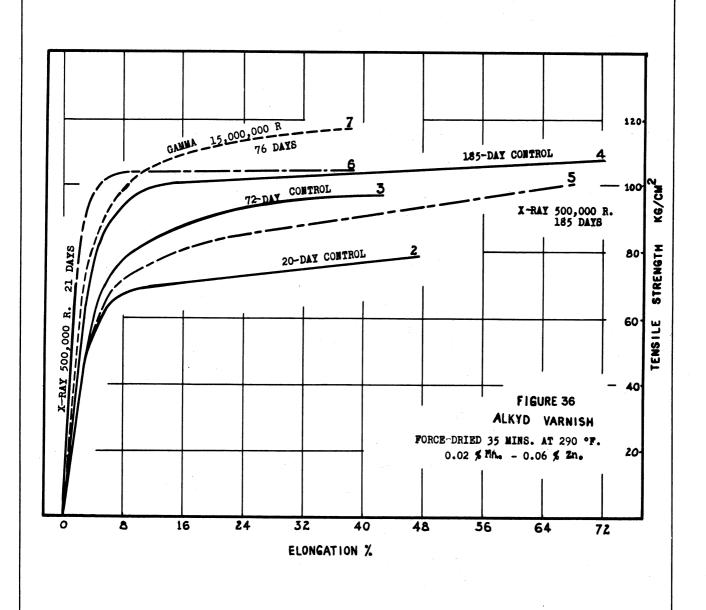
#### TABLE XXVIII

STRESS-STRAIN DATA ON GAMMA- AND X-IRRADIATED SOYA-ALKYD FILMS
Gamma irradiation in cobalt-60 chamber, 15,000,000 roentgens during 150 hours
X-irradiation, Cr target, 500,000 roentgens in approximately 11 hours.

| Test       | Film<br>Life,<br>Days | Irradia-<br>tion | Pigment                                | PVC<br>% | Breaking<br>Stress,<br>kg/cm <sup>2</sup> | Breaking Elongation, | Toughness<br>(Area) |
|------------|-----------------------|------------------|--|----------|---|----------------------|---------------------|
|            |                       |                  |  |          |   |                      |                     |
| 3          | 72                    | Control          | None                                   | 0        | 96.8                                      | 42.3                 | 35.8                |
| 7          | 76                    | Gamma            | None                                   | 0        | 117.3                                     | 38.1                 | 39.3                |
| 2          | 20                    | Control          | None                                   | 0        | 78.4                                      | 47.1                 | <b>32.</b> 6        |
| 6          | 21                    | $\mathbf{X}$     | None                                   | 0        | 103.6                                     | 38.5                 | 38.4                |
| 4          | 185                   | Control          | None                                   | 0        | 108.0                                     | 72.7                 | 72.1                |
| 5          | 185                   | X                | None                                   | 0        | 100.0                                     | 67.9                 | 57.5                |
| 3.07       | 10                    |                  | (Allered II 7.7)                       | 0        | 000 9                                     | 17 5                 | 29.6                |
| 17         | 19                    | Control<br>X     | (Alkyd-Urea, 7:3)                      | 0        | 202.8<br>236.5                            | 17.5<br>7.0          | 10.8                |
| 18         | 23                    |                  | (Alkyd-Urea, 7:3)<br>(Alkyd-Urea, 7:3) | 0        | 209.0                                     | 9.6                  | 16.6                |
| 19         | 70<br>70              | Control          | (Alkyd-Urea, 7:3)                      | 0        | 212.8                                     | 9.6                  | 16.6                |
| 20         | 70                    | Gamma            | (Alkyd-orea, 7:7)                      | 0        | 212.0                                     | 9.0                  | 10.0                |
| 25         | 77                    | Control          | TiO2-0.17 micron                       | 25       | 133.2                                     | 30.0                 | 29.5                |
| 29         | 75                    | Gamma            | TiO2-0.17 micron                       | 25       | 131.2                                     | 20.4                 | 20.2                |
| 23         | 31                    | Control          | Ti02-0.17 micron                       | 25       | 113.8                                     | 36.3                 | 29.1                |
| 28         | 16                    | X                | TiO2-0.17 micron                       | 25       | 116.8                                     | 36.2                 | 30.4                |
| 26         | 185                   | Control          | Ti02-0.17 micron                       | 25       | 135.5                                     | 35.4                 | 38.0                |
| 27         | 185                   | X                | Ti02-0.17 micron                       | 25       | 142.8                                     | 20.8                 | 23.8                |
| 61         | 7                     | Control          | CaCO <sub>3</sub> -0.07 micron         | 25       | 83.8                                      | 51.1                 | 24.8                |
| 62         | 63                    | Control          | CaCO <sub>3</sub> -0.07 micron         |          | 84.5                                      | 21.2                 | 13.6                |
| 63         | 25                    | X                | CaCO <sub>3</sub> -0.07 micron         | 25       | 97.4                                      | 32.3                 | 22.1                |
| 64         | 85                    | Gamma            | CaCO <sub>3</sub> -0.07 micron         | 25       | -   | able for hand        |                     |
| 04         | ٥٦                    | Gamma            | 000005 0.01                            | -/       |   |                      |                     |
| 70         | 74                    | Control          | BaSO4 (0.5-2.0) M                      | 23       | 110.0                                     | 47.0                 | 34.7                |
| 72         | 81                    | Gamma            | BaSO4 (0.5-2.0) u                      | 23       | 119.2                                     | 35.1                 | 30.9                |
|            |                       |                  |  |          |   |                      |                     |
| 77         | 74                    | Control          | BSWL-1.3 micron                        | 23       | 125.2                                     | 6.2                  | 6.3                 |
| 78         | 110                   | Control          | BSWL-1.3 micron                        | 23       | 119.2                                     | 18.6                 | 17.9                |
| <b>7</b> 9 | 81                    | Gamma            | BSWL-1.3 micron                        | 23       | 106.2                                     | 15.2                 | 13.8                |
|            |                       |                  | 00                                     |          |   |                      |                     |

TABLE XXVIII (cont.)

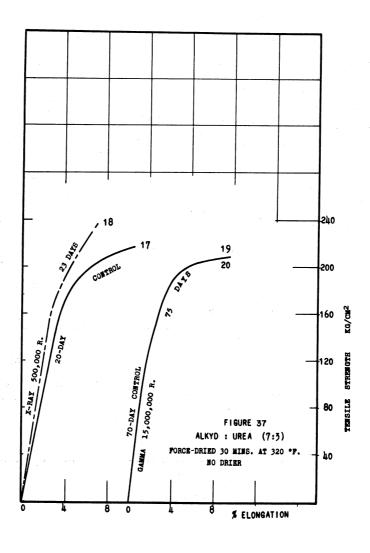
| Test | Film<br>Life,<br>Days | Irradia-<br>tion | Pigment                    | PVC<br>% | Breaking<br>Stress,<br>kg/cm <sup>2</sup> | Breaking Elongation, | Toughness<br>(Area) |
|------|-----------------------|------------------|----------------------------|----------|---|----------------------|---------------------|
| 95   | 29                    | Control          | C. Black 0.078/            | 10       | 119.5                                     | 48.1                 | 44.1                |
| 100  | 23                    | X                | C. Black 0.078 H           | 10       | 130.0                                     | 51.2                 | 52.9                |
| 96   | 60                    | Control          | C. Black 0.078 M           | 10       | 134.8                                     | 52.9                 | 55.5                |
| 98   | 65                    | Gamma            | C. Black 0.078 M           | 10       | 147.2                                     | 50.4                 | 57.9                |
| 97   | 175                   | Control          | C. Black 0.078 \mathcal{P} | 10       | 137.0                                     | 51.3                 | 55.3                |
| 99   | 175                   | Gamma            | C. Black 0.078 \mu         | 10       | 165.1                                     | 33.8                 | 46.8                |

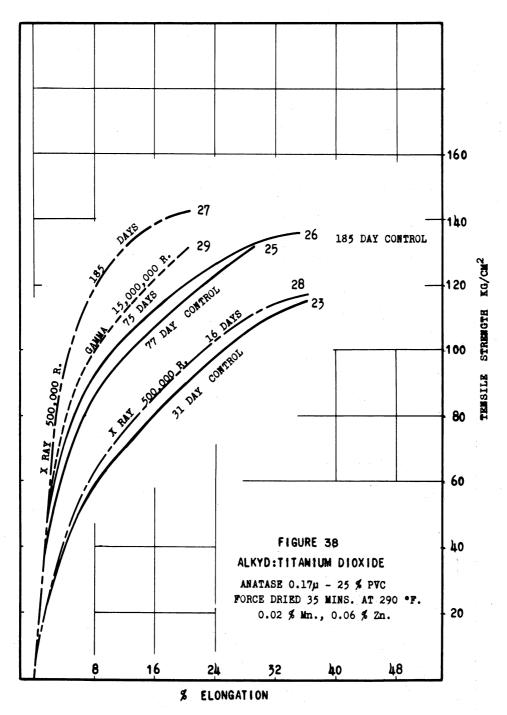


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"Figure 37 presents graphs on an unpigmented vehicle of soya-alkyd and urea, in a ratio of 7:3 by weight of nonvolatile content. This type of test vehicle presents difficulties in the duplication of results between films applied on different days, since this combination vehicle at approximately 50% nonvolatile as used for doctor blade application was very subject to a viscosity increase followed ultimately by gelling: this was conceivably due to an addition reaction occurring during can pre-aging, which subsequently affected the stress-strain properties accurding to the extent of its occurrence prior to application, the extent of which was difficult to duplicate. Gamma irradiation had no effect on the urea-soya-alkyd vehicle, curve 19 vs 20, although x-irradiation decreased elongation and toughness, curve 18 vs 17.

"Figure 38 supplements data in Table XXVIII on the TiO<sub>2</sub> (0.17-micron) pigmented test soya-alkyd with a 25% PVC, where gamma irradiation decreased elongation and toughness about 30%, curve 29 vs 25. X-irradiation had no immediate effect on these films, curve 28 vs 23, but exhibited an embrittling effect when these same films were further aged, curve 27 vs 26.



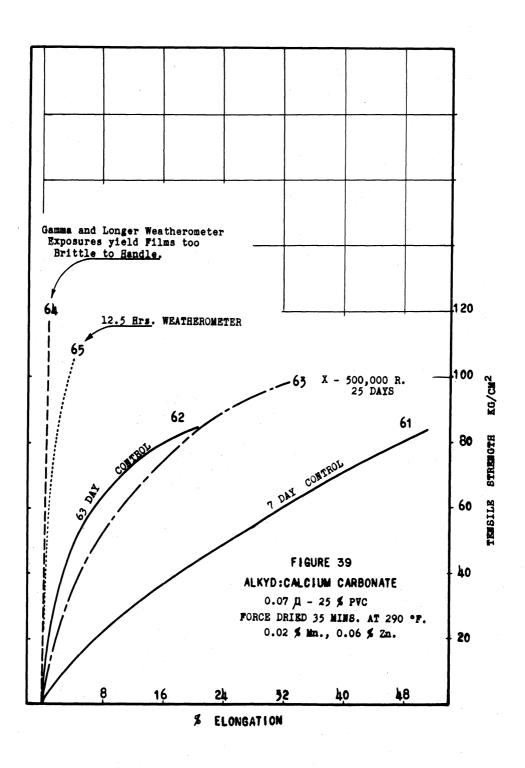


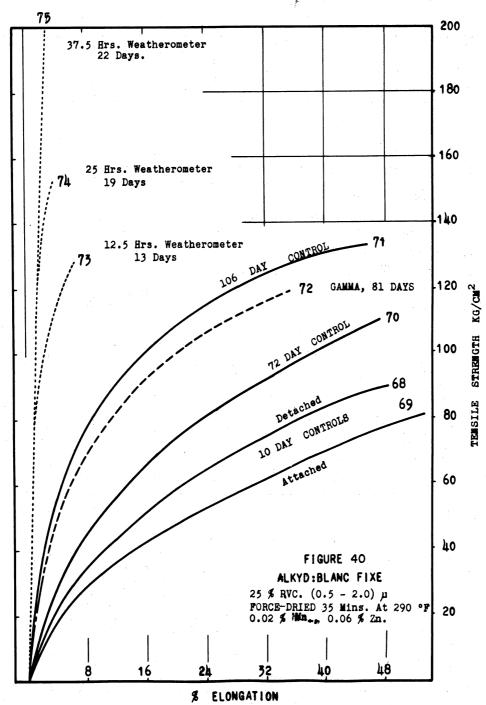
"Figure 39 supplements data in Table XXVIII on the CaCO<sub>3</sub> pigmented alkyd, where the x-irradiation had little effect on these films, curve 63 vs average of 61 + 62. Gamma irradiation embrittled the film to an extent which precluded testing, although the films could be cut with a razor blade if aged at a high humidity. Because of the high embrittling effect of the gamma irradiation on the CaCO<sub>3</sub> pigmented test alkyd film, the BaSO<sub>4</sub> and the BSWL pigmented alkyd films were included in supplementary irradiation studies in order to check whether pigmented absorptivity followed the general absorptivity of metals for these short-wavelength radiations according to:

absorptivity =  $c(Z\lambda)^3$  =  $KZ^3$  (under test conditions),

where Z = atomic number (20 for Ca, 22 for Ti, 56 for Ba, and 82 for Pb); and

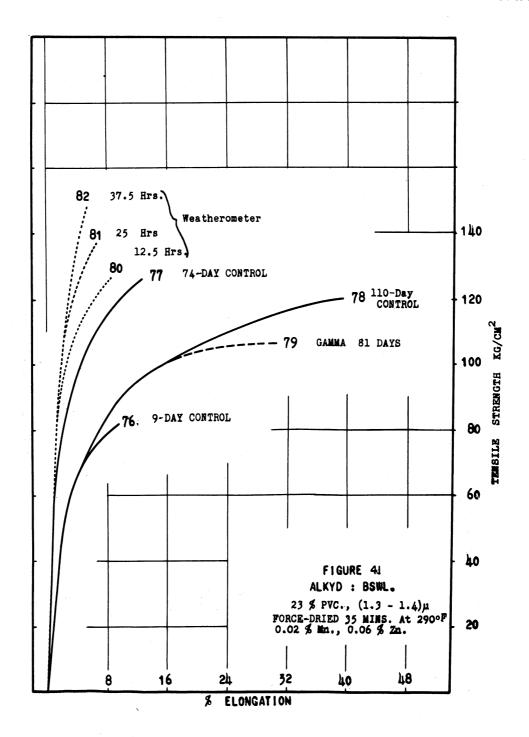
 $\lambda$  = wavelength.



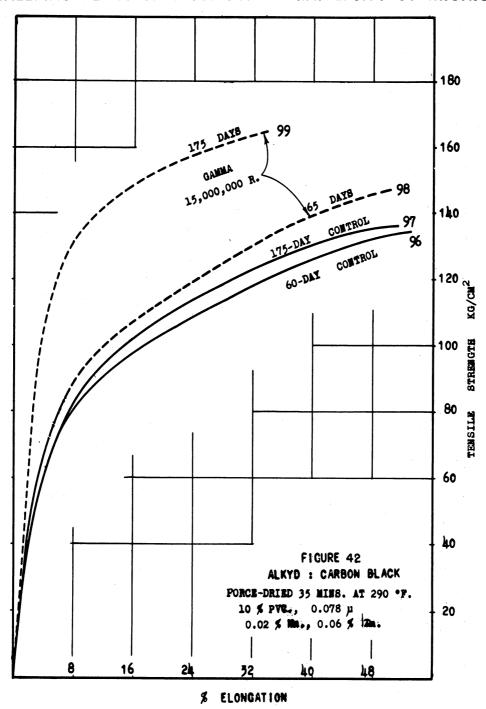


"Figure 40 on a BaSO4 pigmented alkyd shows that these films, when gamma-irradiated, follow the relative effect of this radiation on the unpigmented alkyd without reducing the toughness appreciably, curve 72 vs 70.

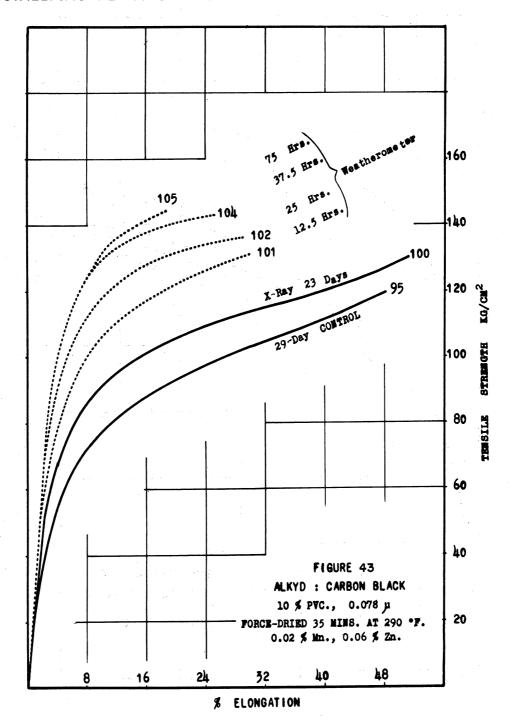
"Figure 41 on a BSWL pigmented soya-alkyd shows that gamma irradiation, curve 79 vs 77 and 78, does not significantly affect this type of film as regards to its stress-strain relationship, thereby indicating that generally the accelerated aging of pigmented alkyd films on exposure to gamma radiations does not follow the above absorptivity relationship for metals, which depends on the atomic number of the metallic atom.



"Figures 42 and 43 on a carbon-black pigmented soya-alkyd show that the test amount of gamma irradiation on these films follows the relative effect of this radiation on unpigmented soya-alkyd films, curve 98 vs 96, i.e., produces a slight increase in tensile strength and toughness. Additional aging of the gamma-irradiated films reduces their toughness below that of the unirradiated films, curve 99 vs 97. X-irradiation, in the test amounts, has a slight toughening action on carbon-black pigmented alkyds, as it does on unpigmented soya-alkyd films, curve 100 vs 95.



"As was mentioned previously, the test films did not receive uninterrupted exposure for 150 hours in the cobalt-60 vault, but were inserted for varying time intervals which totaled 150 hours during a 2-month period (arranged according to a priority irradiation schedule), which system was also adhered to for the x-irradiations. Between irradiations the detached films were retained in their thin plastic or glass envelopes in order to prevent handling damage: These were kept at normal room temperature of 70-77°F and under lighting conditions simulating those found in the average home or office. In order to estimate the effects of the irradiation exposures, the nonirradiated controls



were aged under comparable light and temperature conditions, with the final conditioning being done at 77°F and 50% R.H. All test films received a minimum of 24 hours aging at 77°F and 50% R.H. prior to testing on the tensile apparatus at the latter conditions of temperature and humidity.

d. <u>Summary</u>. "...The effect of gamma, x, and ultraviolet radiation on the stress-strain relationships of some baked, detached soya-alkyd surface-coating films has been investigated. The pigments used were: untreated calcium carbonate, anatase titanium dioxide, carbon black, blanc fixe, and basic sulfate white lead.

"Gamma and x-radiations in test amounts (500,000-15,000,000 roentgens) and test intensities did not materially affect the stress-strain relationships of these soya-alkyd films: generally the irradiated specimens exhibited a higher stress at the point of rupture than did the nonirradiated controls..."

"...Additional aging of the irradiated samples indicates that the radiation-induced reaction continues after cessation of radiation, in accord with a phenomenon which was previously noted, since oxidation is a general effect of radiation whenever free or combined oxygen is available to the system. Investigators on the effects of gamma and beta radiation on plastics note that the latter retain essentially the initial tensile strengths up to approximately  $10^7$ - $10^8$  roentgens.<sup>2</sup> ..."

#### e. Bibliography

- 1. Dean, R. B., and Dixon, W. J., "Simplified Statistics for Small Numbers of Observations," <u>Analytical Chemistry</u> 23, 636 (1951).
- 2. Burr, J. G., and Garrison, W. M., "The Effect of Radiation on the Physical Properties of Plastics," AECD-2078, 2-3, declassified June 25, 1948.

# PART IV. SUBPROJECT M943-7 (FORMERLY M943-F), OPERATION OF THE FISSION PRODUCTS LABORATORY

#### Personnel:

Advisors: L. E. Brownell, Associate Professor of Chemical and Metallurgical Engineering; H. J. Gomberg, Assistant Professor of Electrical Engineering; W. W. Meinke, Assistant Professor of Chemistry; L. Thomassen, Professor of Chemical and Metallurgical Engineering.

Assisted by: J. V. Nehemias, Health Physicist; E. W. Coleman, Research Assistant.

#### A. DESIGN OF A 10,000-CURIE GAMMA SOURCE

### 1. Need for 10,000-Curie Source

A major portion of the work on this subproject since the writing of Progress Report 3 has been devoted to the design and construction of facilities for the new 10-kilocurie cobalt-60 gamma source. The 1000-curie source received from Brookhaven in June of 1951 has proven a simple and useful laboratory tool and has been very valuable in the experiments which have been conducted during the past 18 months. During this period, this source of radiation has been in use 7 days a week, 24 hours a day, with but few exceptions. In fact the research has been limited by the availability of radiation time. Additional disadvantages are that only a small fraction of radiation from the 1000-curie source can be absorbed by a specimen placed in the radiation chamber and, further, that the small internal diameter, about 1-1/2 inches, has greatly limited the size of the sample which may be irradiated, making it difficult or impossible to conduct many experiments. Therefore, in the spring of 1952 the decision was made to secure another gamma source of greater flexibility.

First, it was decided that the design should be modified so that a greater percentage of the radiation would be usable. A modification so that samples can be placed around the exterior of the cylinder as well as within the cylinder was a step in this direction. The internal diameter of the source was increased so that larger samples can be irradiated either within the cylinder or on the outside of the cylinder. Chemical reactors for gas reactions under

pressure are used in some experiments. Equipment for this service is much easier to fabricate, easier to control, and less expensive if some flexibility in size is permitted. The glass reactor used in the chlorination of benzene, for example, is less than 1-1/2 inches in diameter and has six lines attached to it as shown in Fig. 16 of this report. This exemplifies the difficulties involved in limiting equipment to such a small diameter. In experiments with irradiated food, it is desirable to be able to place commercial-size cans in the irradiation chamber. It has been found that preservation of food by irradiation involves many problems other than sterilization, as experienced in the canning industry; i.e., irradiated food must be protected from oxidation, dehydration, etc. as in To date it has been necessary to limit all tests with the canning operation. food to small glass test tubes or plastic containers because the 1000-curie source will not accommodate the smallest-size "tin can". The gamma flux must have sufficient strength so that irradiation time can be short enough to assure that the sample will not spoil before sterilization can be accomplished. With these considerations in mind, it was decided that a radiation source of at least 10 kilocuries would be required and that this source should be in the form of a number of rods which could be set into a cylindrical pattern or into other patterns, as desired.

A few comments regarding the efficiency of using gamma radiation sources might be pertinent. As the intensity of the gamma source is increased, it can be used more efficiently; that is, a greater percentage of the radiation field 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, the new 10-kilocurie source is estimated to be about 30 times as useful as the present 1-kilocurie source because, in addition to having 10 times the total radiation flux, it is estimated that about 3 times as much of the total radiation field will be useful for experimental samples.

Upon inquiry it was found that, during the latter part of 1952, all reactors in the United States with a neutron flux sufficiently high to produce 10 kilocuries of cobalt-60 activity within a reasonable period of time had scheduled radiation services for several months in advance. Rather than postpone the procurement of a larger source, the possibility of irradiation in the Chalk River NRX reactor was explored and the arrangements made with the help of Mr. C. H. Hetherington, of the Sales Department, Commercial Products Division, Eldorado Mining and Refining Limited (1944) (this company is now called Atomic Energy of Canada Limited), of Ottawa, and the Chalk River personnel involved.

### 2. Design of Cobalt Rods for 10,000 Curies

The source consists 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. Kurt, Sales Manager, expedited our order). The rods were encapsulated in 1/8-inch nominal size 3S-H18 aluminum pipe, 0.269 inch I.D., with a wall thickness of 0.068 inch and an 0.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 Limited (1944) of Ottawa, Ontario, Canada. The ends of the rods were welded with Alcan 25 welding rod, machined clean, and tested for leaks by dropping the complete rods into hot water.

The rods were inserted into the Chalk River reactor during July. Initial specifications called for the 10-kilocurie source to be produced by 4-1/2 months of neutron bombardment of the 100 rods. The curie level was to be defined on the basis of calculations of total "nvt" exposure, with consideration given to irradiation efficiency. The further restriction that no one rod of the 100 irradiated was to be more than twice as active as any other insured a fairly even radiation field in the assembled source.

#### 3. Design of Cave for 10,000 Curies

Although it was relatively simple to select the optimum shape and size of the cobalt rods for the 10,000-curie source, it was much more difficult to decide how this source should be contained and shielded, and how experimental samples should be brought into the radiation field with a minimum of danger to operating personnel and a maximum of flexibility regarding experimental procedures. Several designs, including such ideas as lowering experimental samples down through charging tubes into the radiation field, the use of shielded parts, etc., were investigated and discarded. The eventual decision was that the most flexible method of using the radiation source was simply to have it in a radiation room or chamber. With such an installation it was necessary to provide some means of "shutting off" the source so that laboratory personnel could enter the room to set up experimental equipment. It was decided that the most convenient means of shutting off radiation from the area would be to submerge the source in a "well" of water. This permits the setting up of experimental equipment in a room free of hazardous radiation. After laboratory personnel leave the room, the source can be raised and the experimental irradiation performed. Figure 44 shows a cutaway or phantom view of the radiation "cave" for 10,000 curies of cobalt 60.

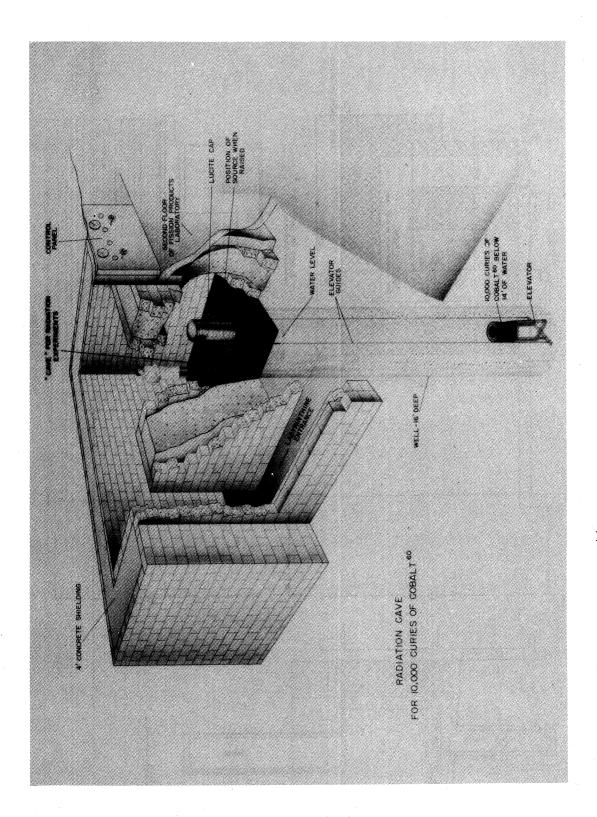
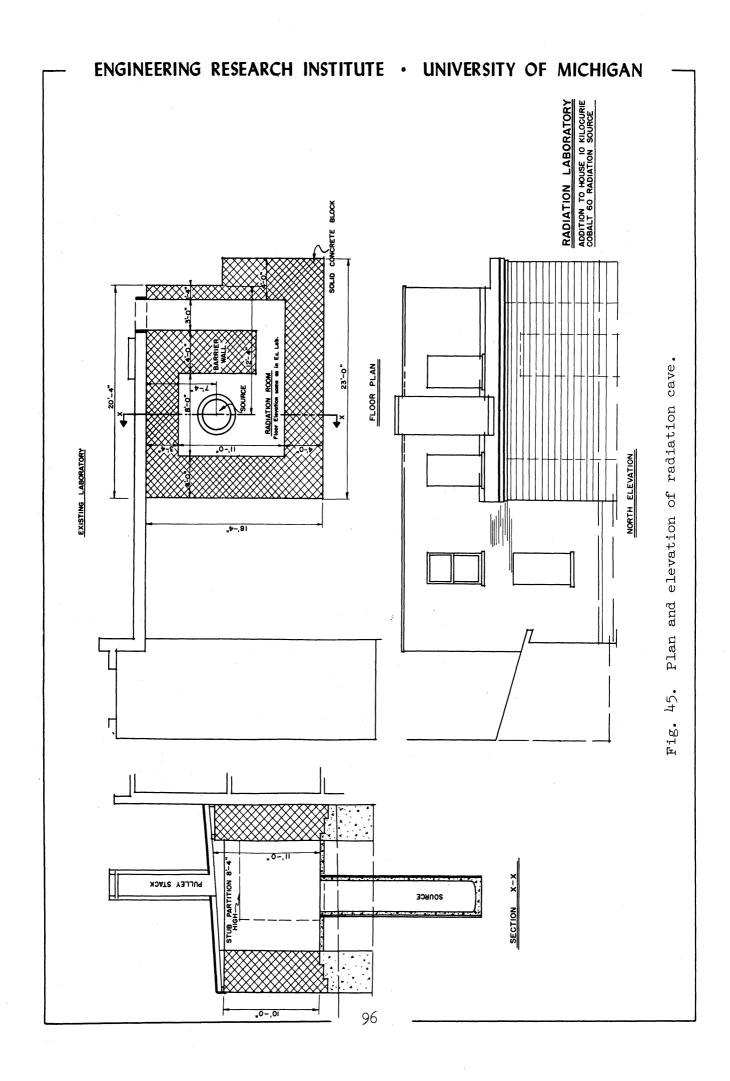


Fig. 44. Cutaway view of radiation cave.



#### 4. Details of Design of Cave

Figure 45 shows the plan and elevation of the radiation cave. The essential features are the 4-foot thick concrete walls necessary to shield laboratory personnel and the surrounding area from gamma radiation and the 16-foot well used for shutting off the source. The 4-foot barrier wall provides a simple labyrinthine entrance and prevents direct radiation from reaching the door. The barrier wall serves to diminish the radiation flux in the labyrinthine entrance so that a heavily shielded door is not required. A mechanical safety interlock makes it impossible to open the door to the radiation cave when the source is in the raised position: the locking bar, as shown in Fig. 46, cannot be moved to clear the door unless the cable has been completely unwound from the winch, as is the case when the source is at the bottom of the well.

Figure 47 shows the same interlock with the door in the unlocked position and the winch in the locked position. In this case the locking bar has been moved to enter a slot in the end plate of the winch. The clearance between this bar and the face of the drum is such that the bar cannot be slid into position until the cable has been entirely unwound from the drum. Also, as the door is opened after sliding the locking bar to lock the winch, the release mechanism blocks the bar so that it cannot be moved back until the door is again closed.

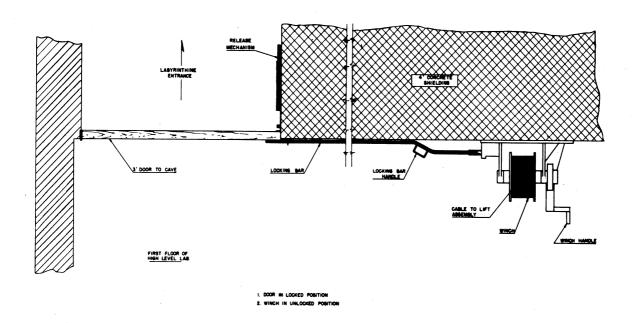


Fig. 46. Plan view of safety interlock with 10,000-curie cobalt-60 source in raised position.

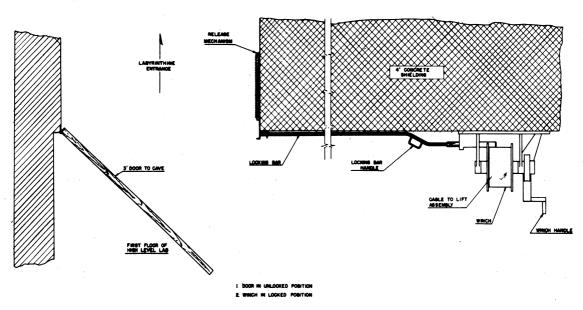


Fig. 47. Plan view of safety interlock with the 10,000-curie cobalt-60 source in lowered positions.

In addition to this interlock, a sliding panel on the safety light, immediately above the locking bar handle, serves to indicate the position of the 10,000-curie source. This device is not operated by the cable but by the 10,000-curie source itself. As the source approaches its uppermost position, it raises a vertical rod and moves the sliding panel, indicating the position of the source. If the vertical rod is in contact with the source, the source is in some raised position and the red warning light is lit. If, and only if, the source is down from the lucite cap, the vertical rod hangs free, allowing the green light to be lit.

#### 5. Details of Lift and Holder for Cobalt Rods

Figure 48 shows the lift assembly. Two 1/2-inch diameter 18-8 stainless-steel rods serve to raise and lower the cobalt source. A small pedestal is welded to the lift platform so as to raise the source in order that the bottom of the cobalt rods will be about 12 inches above the floor of the radiation cave when the source is in the raised position.

The holder for the rods, shown in Fig. 49, is constructed of fabric-reinforced phenolic plastic which serves to insulate the rods from contact with the elevator and from each other. The 100 rods are placed in two concentric circles spaced as close together as possible.

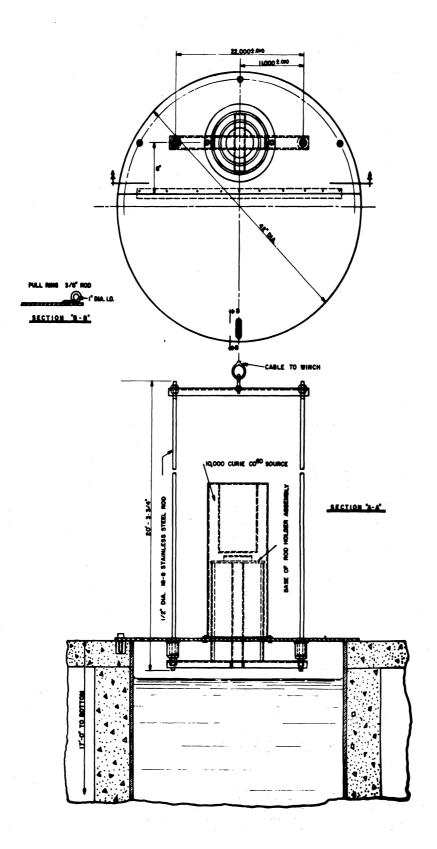
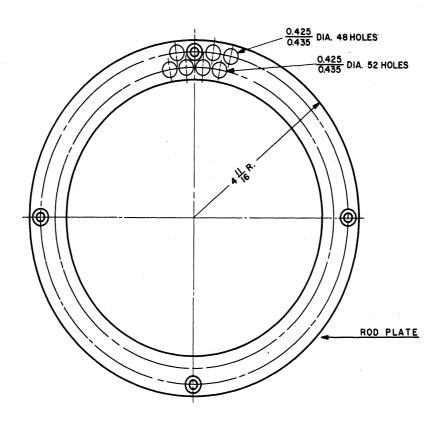


Fig. 48. Lift assembly.



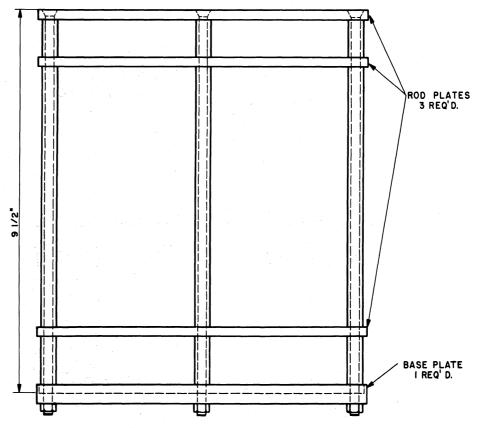


Fig. 49. Rod holder assembly.

### 6. Photographs of the Cave and Accessories of Cobalt-60 Source

A series of photographs were taken to aid in describing the construction and operation of the cave for the 10,000-curie source and the accessory equipment. Figure 50 is a photograph of the cave during construction. For ease in assembling and to minimize the difficulty of future disassembling, the 4-foot concrete walls were constructed mainly of solid 8 x 8 x 16-inch concrete blocks with shorter joints. Alternate rows were filled with poured concrete; however, the outside rows were all of block construction. The blocks were staggered vertically to minimize radiation through the mortar joints. Also, in each successive course the rows containing poured concrete were alternated. Figure 50 is a photograph of the masons laying the solid concrete blocks and leaving alternate rows for poured concrete.

Figure 51 is a photograph taken from the second floor of the Fission Products Laboratory and shows the walls of the cave just before the rafters were covered with roofing material. Figure 52 is a photograph of the completed cave with the barbed-wire barricade around the roof and the signs to warn against crossing the barricade.

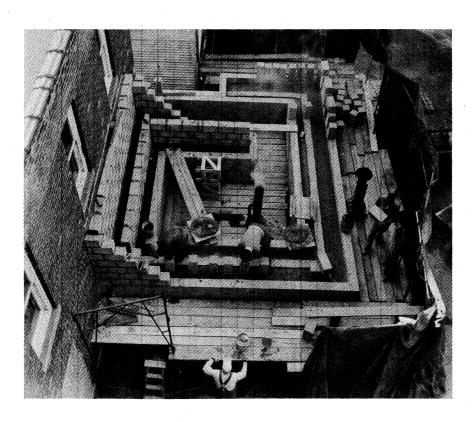


Fig. 50. Photograph of cave during construction.

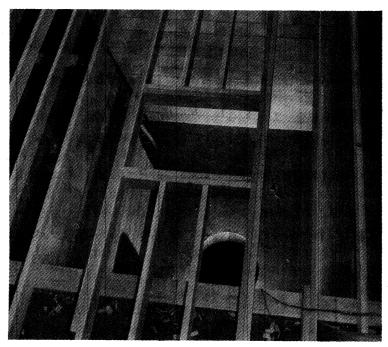


Fig. 51. Photograph of radiation cave just before completion.

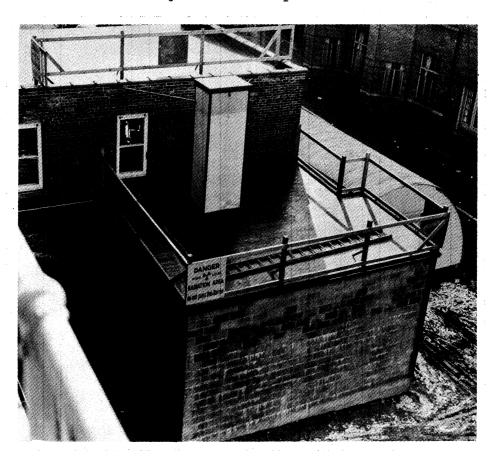


Fig. 52. Photograph of radiation cave completed.

Figure 53 is a photograph taken looking down into the well before it was filled with water and shows the elevator in the down position. In Fig. 54 the elevator is shown in the partly raised position with the aluminum holder for the rods in place. In Fig. 55 the elevator is shown in the raised position. The plastic (lucite) cap serves to prevent any potentially contaminated water from dripping off the rods and the rod holder and splattering on the experimental samples as the source is raised. The cup in the center holds samples placed in the high-flux position. Figure 56 shows one of the laboratory employees placing a No. 10 tin can in the lucite cap.

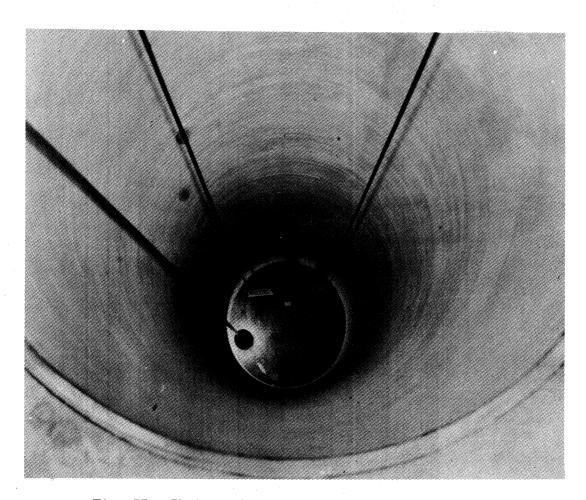


Fig. 53. Photograph of elevator in down position.

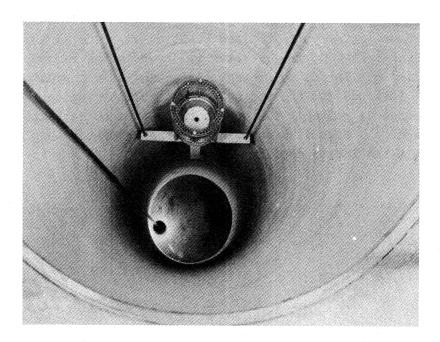


Fig. 54. Photograph of elevator in partly raised position.

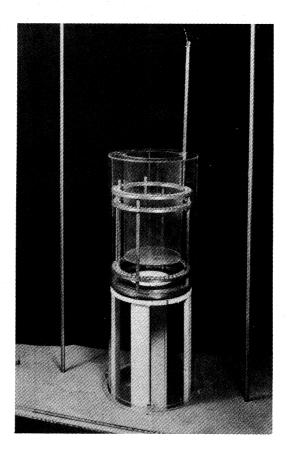


Fig. 55. Photograph of elevator in raised position with rod holder in lucite cap.

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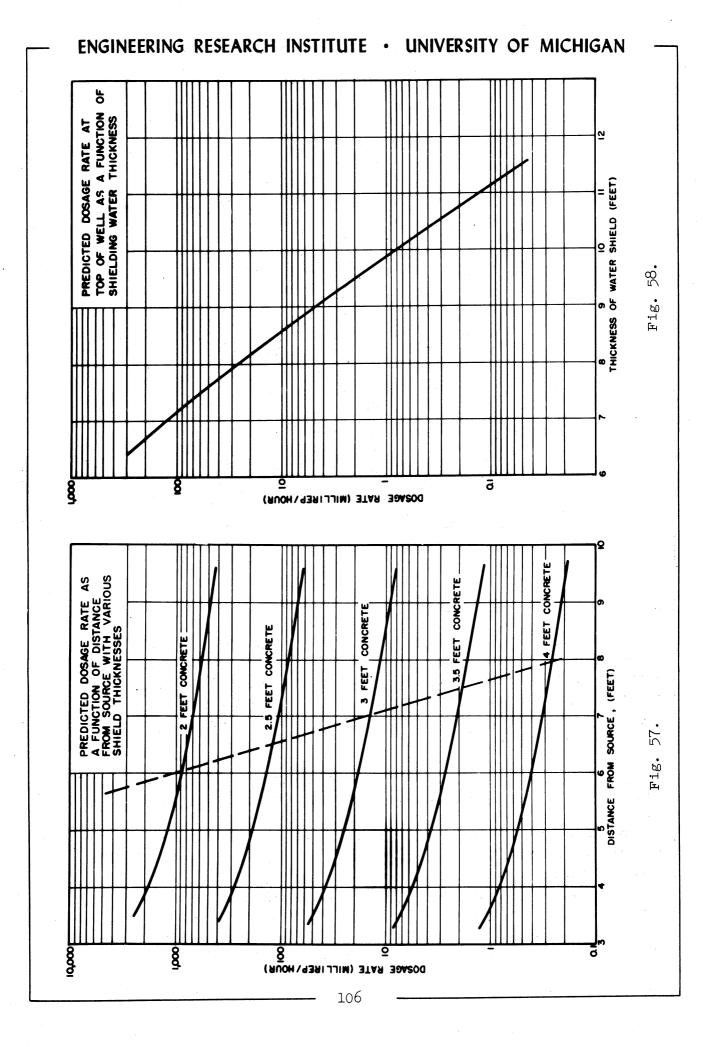


Fig. 56. Placing No. 10 tin can in lucite cap.

#### 7. Shielding Considerations

After consideration of the various experiments which were anticipated, it was decided that the radiation cave would be 8 feet square, which places the inner surface of the shield wall 4 feet from the center of the source. With the shield so situated, it was necessary to determine the thickness of concrete required to reduce the radiation levels at the outside surface to values safely below tolerance.

Figure 57 shows the effect of various thichnesses of concrete at various positions based on predicted dosage rates, 1 and shielding computations.2 The dotted lines indicates the dosage rate outside a concrete shield as a function of shield thickness for a design in which the inner face of the shield is 4 feet from the source. As the outer surface is accessible to the general public, a maximum design dosage rate of 1.0 mr/hr was considered permissible. However, there was also the consideration that it is convenient to lay concrete walls in 8-inch units of thickness; therefore, a thickness of 48 inches (4 feet) of concrete was selected for the shielding walls. The estimated dosage rate at the outer surface of such a shield wall would then be less than 0.3 mr/hr.



The source is to be lowered into a well of water 3 feet in diameter and of sufficient depth to make it possible for workers to enter the cave without hazard. To determine the well depth required, a set of calculations similar to those applied to the concrete shielding was made; the predicted dosage rates are shown in Fig. 58. On the basis of these calculations, a shielding thickness of 11 feet was considered sufficient. However, in addition to shielding thickness a total of an additional 5 feet of depth is needed, partly for manipulation space at the top and bottom of the well and partly to provide space for the lift mechanism. Therefore, a 16-foot well was prepared. On the basis of this design and the estimated dosage rates, it should be possible to effect substantial future increase of source strength with no increase in shielding requirements.

#### 8. Proposed Installation Procedures

The source is expected to arrive from Chalk River on or about February 1, 1953, in the 3-ton lead container shown in Fig. 59. This container will be set into place between the well and the barrier wall.

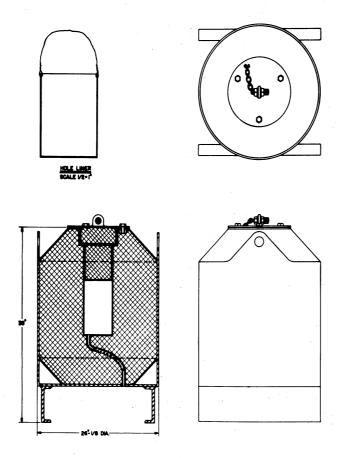


Fig. 59. Shipping container for 10,000-curie cobalt-60 source.

It will be necessary to connect the lifting apparatus both to the cover of the shipping container and the handle of the aluminum bucket which actually contains the source rods. The cover of the shipping container, which weighs about 300 pounds, will then be raised and set out of the way by personnel operating behind the barrier walls, using the lift and boom shown in Fig. 60. The bucket of source rods will then be lifted from the shipping container and lowered into the well. The expected dosage rate at the operators' position will be of the order of several hundred mr/hr during the actual transferring of the source rods. However, as practice runs indicate a maximum transfer time of less than 1 minute, no overexposures are anticipated for this operation.

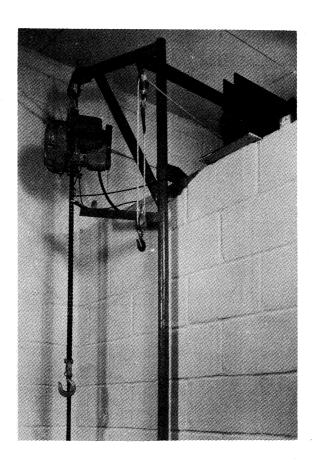


Fig. 60. Unloading lift for 10,000-curie cobalt-60 source.

When the source rods are on the bottom of the well, the source rack will be lowered into place. The rods are then to be picked up, one at a time, and placed in the rack with the 18-foot tongs. Negligible radiation levels are anticipated for this operation, which may take several days. As soon as the rods are in place in the source rack, the 10,000-curie cobalt-60 radiation cave unit will be ready for operation.

#### 9. Corrosion

The cobalt rods were jacketed in 3S-H18 aluminum prior to irradiation so as to prevent corrosion of the cobalt rods and contamination of the water in the well with cobalt-60 ions. However, it is also necessary to protect the aluminum jackets from corrosion.

Olsen<sup>3</sup> described some corrosion studies on aluminum-clad reactor-fuel elements in filtered water. Two types of protection were investigated: (1) element pretreatment by either anodizing or alodizing; and (2) solution control by using nitric acid to maintain a pH of 5.5 to 6.5 or the addition of 60 ppm sodium chromate as an inhibitor. The results of these tests indicated that "no pretreatment was effective in stopping pitting attack for more than four months as a maximum. Control of the pH tended to reduce pitting attack but did not stop it. However, the addition of sodium chromate was found to stop all pitting attack and at the same time reduce the overall corrosion rate to essentially zero."

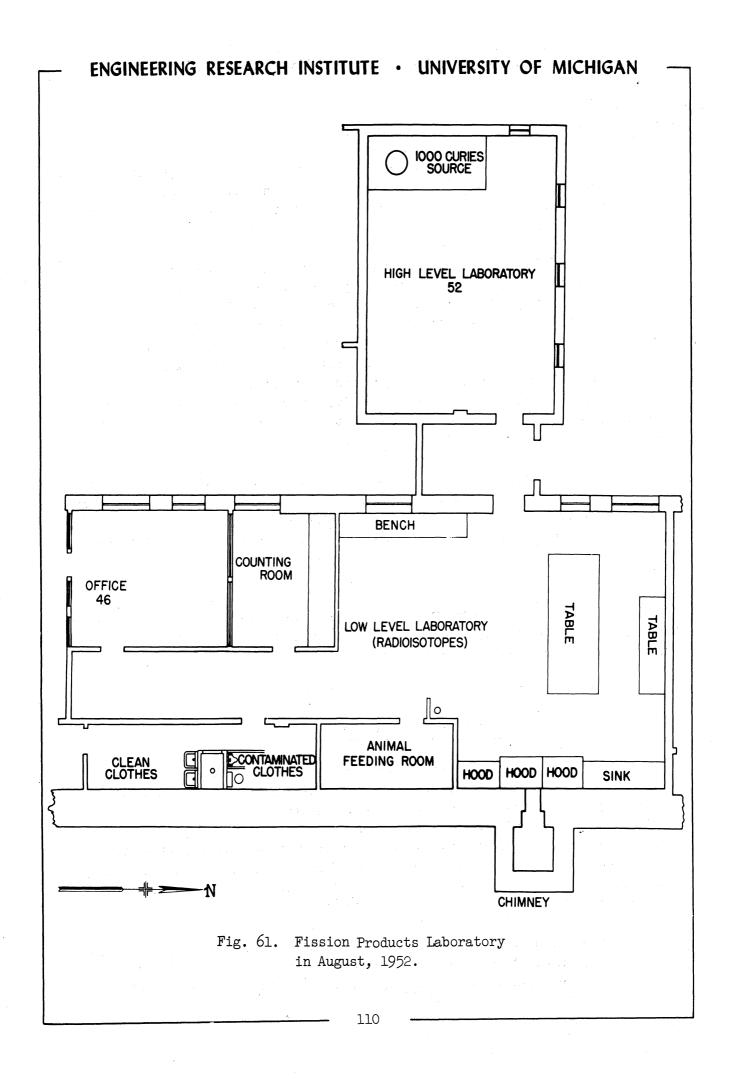
Methods of protecting the aluminum rods against corrosion were also checked with the Research Laboratory of the Aluminum Company of America. One method which this laboratory recommended was the use of deionized or distilled water in the well treated with 1000 ppm  $\rm Na_2CrO_4$  as inhibitor and 1500 ppm  $\rm NaH_2PO_4$  plus 1000 ppm  $\rm Na_2HPO_4$  as a buffer to hold the pH close to 6.8 (pH to be adjusted with  $\rm H_3PO_4$ ).

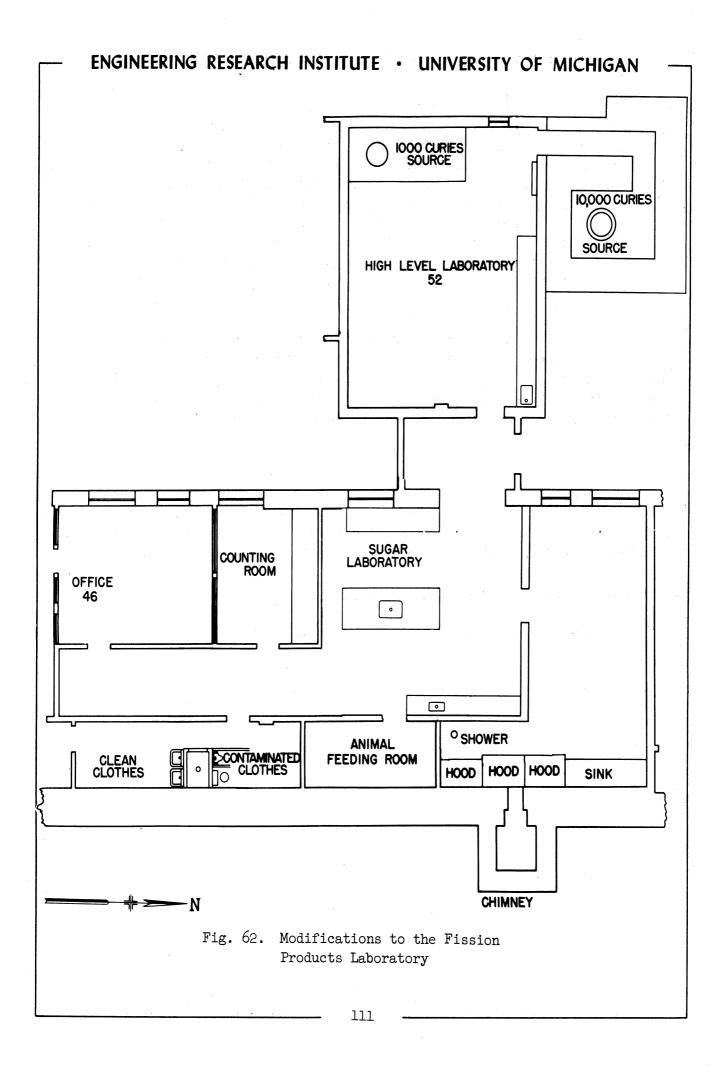
The method recommended by the Research Laboratory of the Aluminum Company of America will be used. Dummy rods (nonradioactive) will be kept in the well and inspected for corrosion.

#### B. CHANGES IN THE FISSION PRODUCTS LABORATORY

Figure 61 shows a plan of the radiation laboratories as described in Progress Report 1. The main parts of the Fission Products Laboratory are represented by the area designated by Nos. 46, 50, and 52. During the past six months a number of modifications have been made, as shown in Fig. 62. The principal addition has been the cave shown at the northwest corner of the high-level laboratory. The University has made various additions to the laboratory facilities at its own expense, so that at present there are sinks and benches on both the north and south sides of the second floor.

The reduction in budget planned for the fiscal year July 1, 1952, to June 30, 1953, made it necessary to decrease the scope of the experimental program and as a result all the laboratory space provided by the University for





use by the Fission Products Laboratory could not be utilized. As laboratory space is at a premium, a portion of this space has been reassigned to other projects. At the expense of other projects the low-level laboratory was partitioned approximately in half and the space which did not contain the fume hoods was assigned to Engineering Research Project 2047 and Phoenix Project 41 and the laboratory proper was converted to the Sugar Beet Laboratory. The store room which was formally located just east of this laboratory parallel to the hoods is being converted into an air-conditioned animal feeding room for the feeding experiments which will be conducted by Michigan Memorial-Phoenix Project 41. At present some space on the first floor of the high-level laboratory has also been assigned to the Phoenix Project 41.

No other changes are anticipated in the laboratory prior to June 30, 1953. At that time the space presently occupied by the Sugar Beet Laboratory may be reassigned to the Fission Products Laboratory if additional working space is required. If the research program is further reduced, the unused laboratory space may be allocated to other research projects.

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- 2. Morgan, G., at En, Conn, Isot Div. Circle B-3, 1948.
- 3. Breazeale, W. M. ORNL 1105 (declassified Aug. 8, 1952).

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| Abbot Laboratories                 | D. L. Tabern                           | 1   |
| Air Force Special Weapons Center   | Chief, Tech. Library Branch            | 1   |
| American Can Company               | O. F. Ecklund; H. A. Benjamin          | 2   |
| American Cyanamid Company          | F. M. Hall                             | 1   |
| American Society of Refrig. Engrs. | M. C. Turpin; T. J. Ammel; S. J.       |     |
|                                    | Williams                               | • 3 |
| Archer-Daniel-Midland Co.          | O. Graziani                            | 1   |
| Argonne National Lab.              | P. Fineman                             | 1   |
| Armour Research Foundation         | R. F. Humphreys                        | 1   |
| Assn. of Brit. Pharmaseu. Industry | Gordon Smith                           | 1   |
| The Atomic Center                  | M. M. Reiss                            | 1   |
| Atomic Energy of Canada Ltd.       | C. H. Hetherington                     | 1   |
| Bendix Aviation Corp.              | C. Branyan; D. Calking; J. F.          |     |
|                                    | Coneybear                              | 3   |
| Brookhaven National Lab.           | C. Williams; B. Manowitz; D. Ballantin | ne  |
|                                    | Library 6                              |     |
| Bureau of Supplies and Accounts    |  |     |
| (Code W)                           | Chief                                  | 1   |
| Canada Packers Ltd.                | W. F. McLean                           | 1   |
| Champion Spark Plug Co.            | H. F. Royal                            | 1   |
| University of Chicago              | L. S. Skaggs                           | 1   |
| Univ. of Chicago, Amer. Meat       |  |     |
| Inst. Foundation                   | H. R. Kraybill                         | 1   |
| University of Chicago, Food        |  |     |
| Research Institute                 | G. M. Dack                             | 1   |
| Coe Laboratories                   | W. S. Rice                             | 1   |
| Columbia University                | E. Gaden; W. A. Selke, C. G. King      | 3   |
| Continental Can Co.                | L. E. Clifcorn                         | 1   |
| Continental Grain Co.              | R. G. Luitiens                         | . 7 |

# Distribution List (cont.)

| Copper-Bessemer Corp.              | R. L. Boyer                         | 1   |
|------------------------------------|-------------------------------------|-----|
| Cornell Univ., Hotel Admin.        | J. J. Wanderstock                   | 1   |
| Corning Glass Works                | H. P. Hood                          | 1   |
| The University, Chem. Dept.        |                                     |     |
| (England)                          | F. S. Dainton                       | 1   |
| The Dow Chemical Co.               | W. H. Beamer                        | 1   |
| Dry-Freeze Corp.                   | R. F. Colton                        | 1   |
| Ebasco Services, Inc.              | W. F. Friend                        | 1   |
| El-Tronics, Inc.                   | E. M. Pollock                       | 1   |
| Electronized Chemicals Corp.       | W. Huber                            | 1   |
| Environment Health Center          | Library                             | 1   |
| Esso Labs., Res. Div.; Chem. Div.  | C. O. Tongberg; C. E. Morrell       | 2   |
| Federal Security Agency, and Food  | E. M. Nelson; H. Welch; E. P. Laug; |     |
| and Drug Admin.                    | F. A. Vorhes; G. Slocum             | 5   |
| Federal Security Agency, Milk      |                                     | . • |
| and Food Branch                    | J. D. Faulkner                      | 1   |
| Florida Citrus Mutual              | G. A. Little                        | 1   |
| Food Manufacture                   | T. Crosbie-Walsh                    | 1   |
| General Electric Co.               | W. W. Schultz; M. A. Edwards        | 2   |
| General Foods, Inc.                | R. R. Baldwin                       | 1   |
| G. and L. Beijer (Sweden)          |                                     | 1   |
| Hanford Works (General Electric)   | C. C. Gamertsfelder                 | 1   |
| High Voltage Engineering Corp.     | F. L. Foster, Jr.                   | 1   |
| Illinois Inst. of Technology       | E. H. Harvey                        | 1   |
| Walter Kidde Nuclear Laboratories, |                                     |     |
| Inc.                               | J. Silverman                        | 1   |
| Kingan and Company                 | R. E. Morse                         | 1   |
| Knolls Atomic Power Laboratory     | L. Dorfman                          | 1   |
| John Labatt, Ltd.                  | W. F. Read                          | 1   |
| Low Temperature Research Station   |                                     |     |
| (England)                          | R. S. Hannan                        | 1   |
| Massachusetts Inst. of Technology  | B. E. Proctor; S. A. Goldblith      | , 2 |
| University of Michigan Memorial    |                                     |     |
| Phoenix Project                    | R. A. Sawyer; H. G. Gomberg;        |     |
|                                    | G. G. Brown; H. B. Lewis;           |     |
|                                    | C. A. Lawrence; L. Kempe;           |     |
|                                    | H. C. Eckstein; S. A. Gould         | 10  |
| Michigan Department of Health      | G. D. Cummings; H. D. Anderson      | 2   |
| Michigan State College             | K. Wilson                           | 1   |
| Minneapolis-Honeywell Regulator    |                                     |     |
| Co.                                | W. E. Belcher, Jr.                  | 1   |
| Minute Maid Corp.                  | J. E. Melvin                        | 1   |
| National Dairy Research            |                                     |     |
| Laboratories, Inc.                 | F. W. Barber; F. J. Carleton        | 2   |
| National Research Corp.            | F. Maslan                           | 1   |
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| Nuclear Engineering Co.           | R. Simpson                    | 1   |
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| Nucleonics                        | C. J. Mosbacher, Jr.          | 1   |
| Oak Ridge National Laboratory     | C. Hochanadel; A. F. Rupp;    |     |
|                                   | Central Files X-10 Site       | 3   |
| Parke, Davis and Company          | L. Sweet; J. Controulis       | 2   |
| Pasteuray Corp.                   | H. W. Abshire                 | 1   |
| Charles Pfizer and Company        | J. F. Snell                   | 1   |
| Purdue University                 | N. W. Desrosier               | . 1 |
| Quartermaster Food and Container  |                               |     |
| Inst. (Army)                      | D. K. Tressler, K. T. Swartz  | 2   |
| Radio Corp. of America            | R. G. Picard                  | 1   |
| The Rath Packing Co.              | R. W. Rath                    | 1   |
| Rensselaer Polytechinic Institute | L. G. Bassett                 | 1   |
| Rohm and Hass Co.                 | J. F. Woodman; L. C. Engleton | 2   |
| Shering Corp.                     | W. Tarpley                    | 1   |
| A. E. Staley Mfg. Co.             | C. E. Smith                   | 1   |
| Southwest Research Institute      | J. P. O'Meara                 | 1   |
| Stanford Research Institute       | P. M. Cook                    | 1   |
| Stirling and O'Brien, Inc.        | D. B. Stirling                | 1   |
| Swedish Institute for Food        |                               |     |
| Preservation Research             | G. Borgstrom                  | 1   |
| Swift and Company                 | W. M. Urbain                  | 1   |
| Syracuse University               | B. P. Burtt                   | 1   |
| Technical Enterprises, Inc.       | A. Redniss                    | 1   |
| Tracerlab, Inc.                   | R. D. Zentner                 | 1   |
| Union Oil Co. of California       | J. E. Sherborne               | 1   |
| United Engineering Laboratories   | L. de Forest                  | 1   |
| U.S. Naval Supply Depot           | LDCR J. A. Corrick, RCO       | 1   |
| University of Notre Dame          | M. Burton                     | 1   |
| University of Notre Dame, Lobund  |                               |     |
| Institute                         | T. D. Luckey                  | 1   |
| The Upjohn Company                | O. R. Woods; G. C. Bond       | 2   |
| Vitro Corporation of America      | W. A. Bain                    | 1   |
| Wood Laboratory                   | G. Roger Wood                 | 1.  |
| Yale University                   | R. H. Bretton                 | 1   |
| Yale University, Sloane Institute | E. C. Pollard                 | 1   |
| Yardley of London Ltd. (Canada)   | R. F. Merrill                 | 1   |

