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PROGRESS REPORT 7

UTILIZATION OF THE GROSS FISSION PRODUCTS
(Unclassified)

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PREFACE

This report presents the results of research performed essentially during the period January 1, 1954, to June 30, 1954, on Project M943 of the Engineering Research Institute, University of Michigan, under AEC Contract No. AT(11-1)-162. Some research performed after June 30, 1954, has been included to complete the report on certain studies.

Results of other research studies in which the irradiation facilities of the Fission Products Laboratory have been used are also reported, because it is believed that these studies may provide additional data on the use of waste fission products. For this reason some of the studies supported by Michigan Memorial-Phoenix Projects 41, 43, and 71 and some of the research performed by personnel of the University of Michigan Hospital are included in this report. It is to be noted that there is no relationship between these studies and Project M943 except that the personnel have a common interest in the effects of radiation and possible uses of radioactive materials and that they use the same source of gamma radiation. The research of the Michigan Memorial-Phoenix Project is supported by funds of the University of Michigan obtained through contributions by individuals and corporations and is in no way supported by, or connected with, the Atomic Energy Commission. The results of the studies reported by the personnel of the Michigan Memorial-Phoenix Projects and of the University of Michigan Hospital in most instances will appear in the scientific literature at some future date. To protect the authorship of these individuals, no part of their results is to be reproduced without permission of the respective authors.

This report is expected to be the final comprehensive report in which studies performed on materials irradiated in the Fission Products Laboratory are described. The discontinuation of the studies on irradiated food supported by Project M943, the diversification of the support of research using the facilities of the Fission Products Laboratory, and the reduction in funds available for the preparation of these reports, have led to the termination of this type of report.

As this is a terminal, comprehensive report it is worthwhile to review very briefly what has been accomplished during 3 years of research. The original goal of Project M943 was to explore possible industrial uses of the waste fission products. To investigate the uses under consideration by research personnel of the University of Michigan, the Fission Products Laboratory was established and high-level, gamma- and beta-particle radiation facilities were designed, constructed and successfully put into operation. Extensive data were collected in three principal fields regarding the effect of radiation on: (1) performance of combustion engines, (2) promotion of chemical reactions, and (3) biological materials (particularly foods).

Six designs for commercial irradiation facilities have been studied and reported in Progress Reports 5, 6, and 7. The designs include: (1) the irradiation of pork to control trichinosis, (2) the irradiation of prepackaged meat to increase its refrigerator storage life, (3) the additative chlorination of benzene under gamma irradiation to produce an insecticide, and (4) three facilities using different sources of radiation to irradiate potatoes to increase the life of storage potatoes. In addition, designs have also been considered but not reported for such applications as: (1) the irradiation of grain to control insect infestation, (2) the polmerization of ethylene under gamma irradiation to produce polyethylene, and (3) the irradiation of surgical supplies for sterilization. All of these designs are believed to be feasible and represent possible commercial applications of the fission products.

The possible industrial uses of fission products are numerous and cannot be fully evaluated until additional data are available on the complex effects of radiation. Some of the effects of radiation have been explored and a few possible applications have been investigated in Project M943 but this study has only opened the door in certain fields and has left other fields untouched. Additional research by industry, government, and the universities is needed to provide more information on the uses for the fission products.

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ABSTRACT

Experiments were completed using 6200 curies of gold-198 and gold-199 to explore the possible effect of a beta radiation field on the spark ignition of a mixture of combustible gases. The effects of the beta source proved to be small and in the reverse direction from that expected; i.e., there was a small but definite depression of the explosion limit.

Further studies on the polymerization of ethylene under gamma radiation were made using higher temperatures and slightly higher pressures. At 220°C an oily liquid was obtained whereas tough or waxy solids had been obtained at lower temperatures.

Toluene was chlorinated under gamma radiation. The products obtained by dehydrohalogenation and subsequent oxidation of the chlorinated toluene confirm the hypothesis that chlorine adds to the ring of toluene. New glass reaction equipment has been constructed to study the kinetics of the chlorination reaction and analytical procedures have been developed.

A design was developed for a chemical plant to chlorinate benzene under gamma radiation for the production of insecticides. Sources of cesium-137, 6-month-old mixed fission products, and cooling reactor-fuel elements were compared. Estimated prices of producing the addition product using gamma radiation compared favorably with existing prices of the product prepared under ultraviolet radiation.

Low dosages of gamma radiation (50,000 to 80,000 rep) were found to increase the refrigerator storage life of raw, ground pork and beef and prepackaged coleslaw, as indicated by taste-panel tests. A radiation dosage sufficient to treat flour or wheat for insect infestation (25,000 rep) was observed to have no undesirable effects on the taste and baking qualities of wheat flour, whereas large radiation dosages (over 100,000 rep) were considered to have an undesirable effect.

Low dosages of gamma radiation (7,000 to 21,000 rep) were found to prevent the sprouting of storage potatoes. Taste-panel tests showed no off-flavor problem with such irradiated potatoes. Chemical analysis showed that irradiated, stored potatoes had a lower sugar content than the stored controls. The same dosages of gamma radiation also increase the storage life of onions. Although the radiation does not prevent sprouting of onions, the sprouts grow only a short distance and then cease growing.

A potato irradiation facility was designed in which a comparison was made between cesium-137, mixed fission products, and cooling reactor-fuel elements as radiation sources. The most economical design was that using fuel elements. Cost of the radiation facility was estimated to be less than $50,000 and cost of treatment less than $4.00 per ton.

Twenty-four elements that occur in foods have been tested for induced activity after irradiation in the high-level, cobalt-60 source. No activity was observed with a scintillation well counter.

Research reported by Michigan Memorial-Phoenix Projects includes studies on animal feeding, plant morphology, and elastomers. In short-term feeding studies using albino rats, no evidence of toxicity was observed in tube-fed rats on a diet irradiated with up to 45 megarep. Other pilot studies were made using small numbers of mice. In this experiment the second generation of Bagg-strain albino mice on the 4 megarep experimental diet failed to reproduce, whereas a mixed pigmented strain of mice on the same diet reproduced as did control groups for both strains. The number of mice in the experiment was too small for statistical significance. The long-term feeding and breeding studies using albino rats have been negative to date except for the possible significance of tumor development in 2 rats out of 62 on the irradiated diet. No tumors have been observed in the 62 controls.

Personnel of the University of Michigan Hospital have reported successful transplants in dogs of aortas and bones subjected to 2-megarep and 3-megarep doses of gamma radiation, respectively. These tests indicate the feasibility of using this process for human aortas and bones.

Research performed in cooperation with industry includes tests on coffee, seed walnuts, and raw fresh crabmeat. Low dosages of gamma radiation improved the refrigerator storage life of fresh crabmeat without undesirable flavor changes.

The Fission Products Laboratory has continued to be operated as planned and without incident. Plans have been made to install a new multi-kilocurie, cobalt-60 gamma source in the radiation "cave" of the Phoenix Laboratory during the summer of 1955.

PROGRESS REPORT 7

UTILIZATION OF THE GROSS FISSION PRODUCTS

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

Personnel:

Subproject Supervisor: R. A. Wolfe, Professor of Physics

Research Assistant: F. L. Tobey

A. INTRODUCTION

The research supported by Project M943 on the effect of radiation on the performance of combustion engines was concluded in June, 1953. However, as combustion is an important chemical reaction, this fundamental experiment has been completed during the past year as a supplement to the studies on chemical reactions.

The purpose of this experiment was to discover whether or not an intense beta-radiation field could exert any appreciable effect on the conditions under which a mixture of combustible gases would be ignited by a spark. If such an effect were present, it might point the way toward certain desirable results such as increased efficiency or the utilization of inferior fuels in combustion engines, etc.

It was decided to determine the explosion threshold for various pressures of a 2:1 mixture of hydrogen and oxygen when diluted with nitrogen.[1]* These thresholds were then compared with those obtained in the presence of a powerful beta source.

*Raised numbers refer to the reference list at the end of each section.

B. APPARATUS

1. Explosion System

The explosion system used to determine the thresholds has been described previously.[2]

2. Beta Source

The beta source used for the hot runs consisted of a five-mil gold foil; measuring two inches by three inches. The foil was mounted in a horizontal position at a distance of seven and three-quarter inches from the gap of the spark plug. Gold-198 and 199 were formed in the foil by irradiation in the Materials Testing Reactor at Arco, Idaho.

Basic data on the two radioactive isotopes of gold are as follows[3]:

Mass No.	Half Life	Neutron Cross Section	Maximum Energy, mev
198	2.69d	96b	0.97
199	3.3d	16000b	0.33

The estimated neutron-flux density at the position of the foil was $2x10^{14}$. The irradiation schedule is given below.

From	To	Duration hr	min
6-14-1950	6-14-2010		20
6-16-1835	6-19-1428	67	53
6-22-0403	6-24-0956	53	53

The activity of gold-198 at time of removal from the reactor was estimated to be 5950 curies on the basis of the above data. The activity of the gold-199 was determined by extrapolation backward from beta-absorption measurements made after the activity had dropped to a low level. Gold-199 activity obtained in this way amounted to 270 curies. Total beta activity at time of removal from the pile was estimated to be 6200 curies.

The dose rate in air at the position of the spark was measured with a Victoreen "R meter" using the 25 R chamber 48 days after removal from the pile. This value was then extrapolated back to find the dose rate in air during the course of the experiment (Fig. 1). These values were further corrected on the assumption that dose rate is a linear function of gas pressure.

A special mount was devised to support the gold in the reaction vessel. The mount was designed to be readily loaded in the canal at the M.T.R. and to permit easy transfer from the shipping container to the reaction vessel. The mount consisted of an arrangement of two mouse traps facing each other on a flat circular base. A "keeper" was used to keep the jaws open while the foil was being placed in position (Fig. 2). When the "keeper" was removed, the jaws clamped the foil in position (Fig. 3). A loop was provided for transferring gold and mount to and from the shipping container. The lead base of the mount increased the amount of beta reflection[4] and improved the stability of the arrangement during loading.

Figure 4 shows the arrangement for transferring the source from the shipping container to the reaction vessel. This consisted of a swinging arm, a sliding loop, a hook, and some cord. A mirror was used to position the mount above the reaction vessel.

Transportation of the source from Idaho to Michigan was arranged through the cooperation of the Commander, 10th Air Force. Special thanks are due Major Kellam, Deputy for Operations, Selfridge AFB, Mt. Clemens, Michigan, for arranging details of the operation. Without this aid a serious loss in activity of the source would have been incurred, before it could be put to use.

C. EXPERIMENTAL PROCEDURE AND RESULTS

1. Cold Runs

The difficulties reported on page 2, Progress Report 5[2], were found to be due to improper mixing. Some minor changes in experimental procedures eliminated these difficulties.

Hydrogen, oxygen, and nitrogen were admitted to the reaction vessel in that order. The ratio of hydrogen to oxygen was held at 2:1 throughout. After a suitable delay to allow mixing to take place, sparks from a 12 volt automotive spark coil were passed through standard spark plug in the reaction vessel for a duration of 30 seconds. Reactions near the explosion limit are quite mild and were detected by leaving one of the Bourdon gauges connected to the system during the sparking interval. This interval was somewhat arbitrary since delayed reactions were frequently observed. In a few cases two successive reactions were also observed.

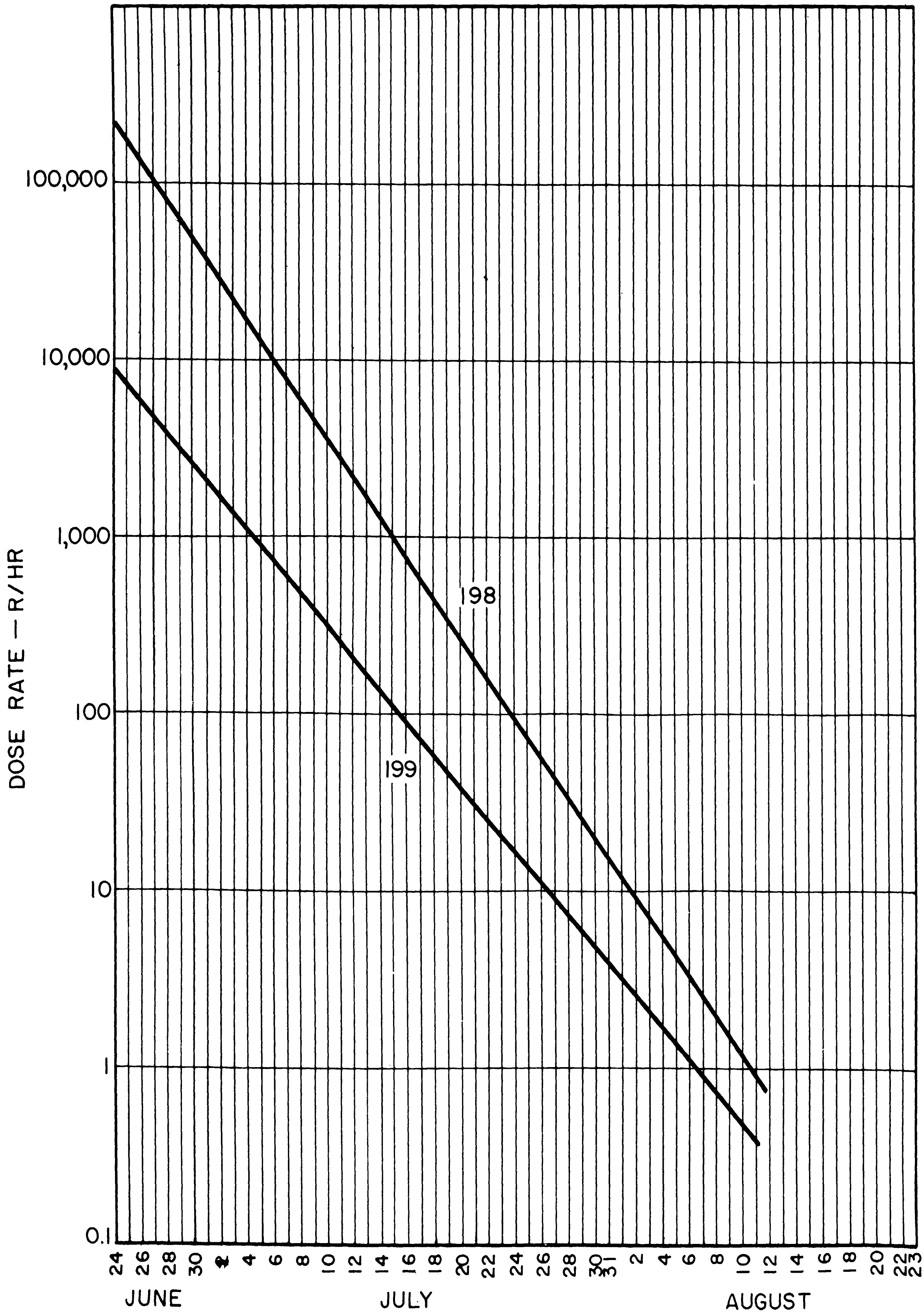

Fig. 1. Dose Rate at Position of Spark Gap due to Au-198 and Au-199.

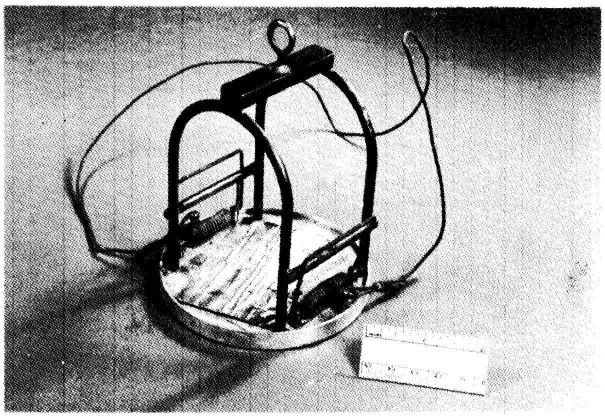

Fig. 2. "Mouse Trap" Set To Take Foil.

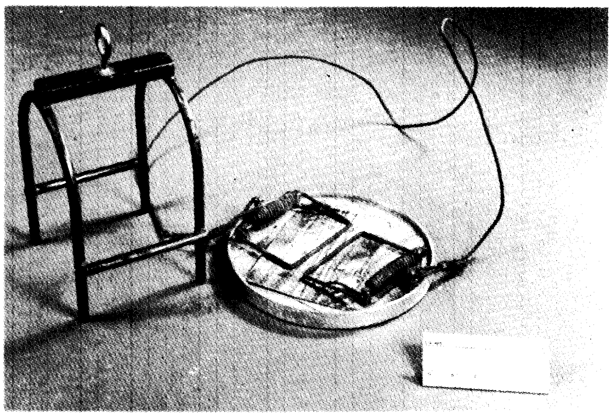

Fig. 3. "Mouse Trap" with Foil in Place.

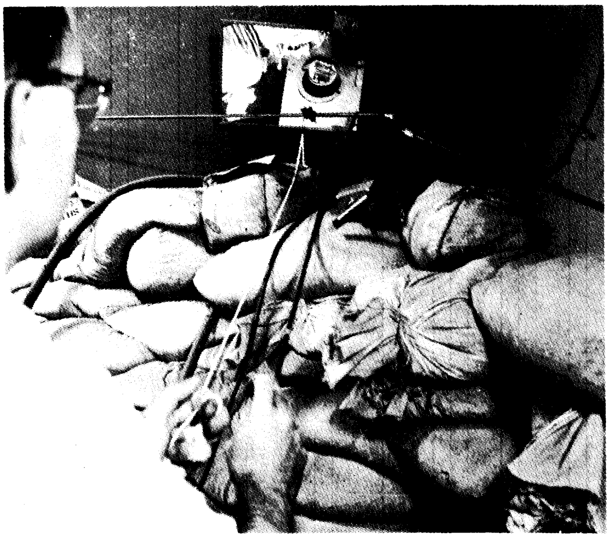

Fig. 4. Transferring Foil to Bomb.

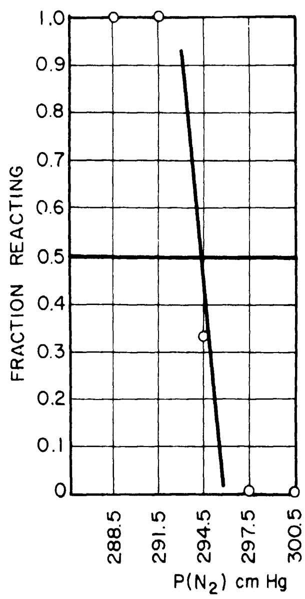

Fig. 5. Threshold Curve $P(2H_2 + O_2) =$ 70.3 cm.

To establish a point on the explosion limit curve a series of runs (at least 20) were made with varying pressures of nitrogen, while the hydrogen plus oxygen pressures were kept constant. The nitrogen pressures were separated into a group of ranges and the frequency of reaction determined for each range. These frequencies were then plotted as ordinates against the mean nitrogen pressure for each range as abscissae. In theory the resulting curve should have the shape of a reversed "S". The best curve as determined by eye was drawn through the plotted points. The point at which the curve crossed the 50 percent reaction ordinate was taken as the explosion limit. Figure 5 shows a typical threshold curve. Figure 6 is a plot of the explosion limit curve. The two points at the top end of the curve were made before and after the hot runs, showing that reproducibility is quite good.

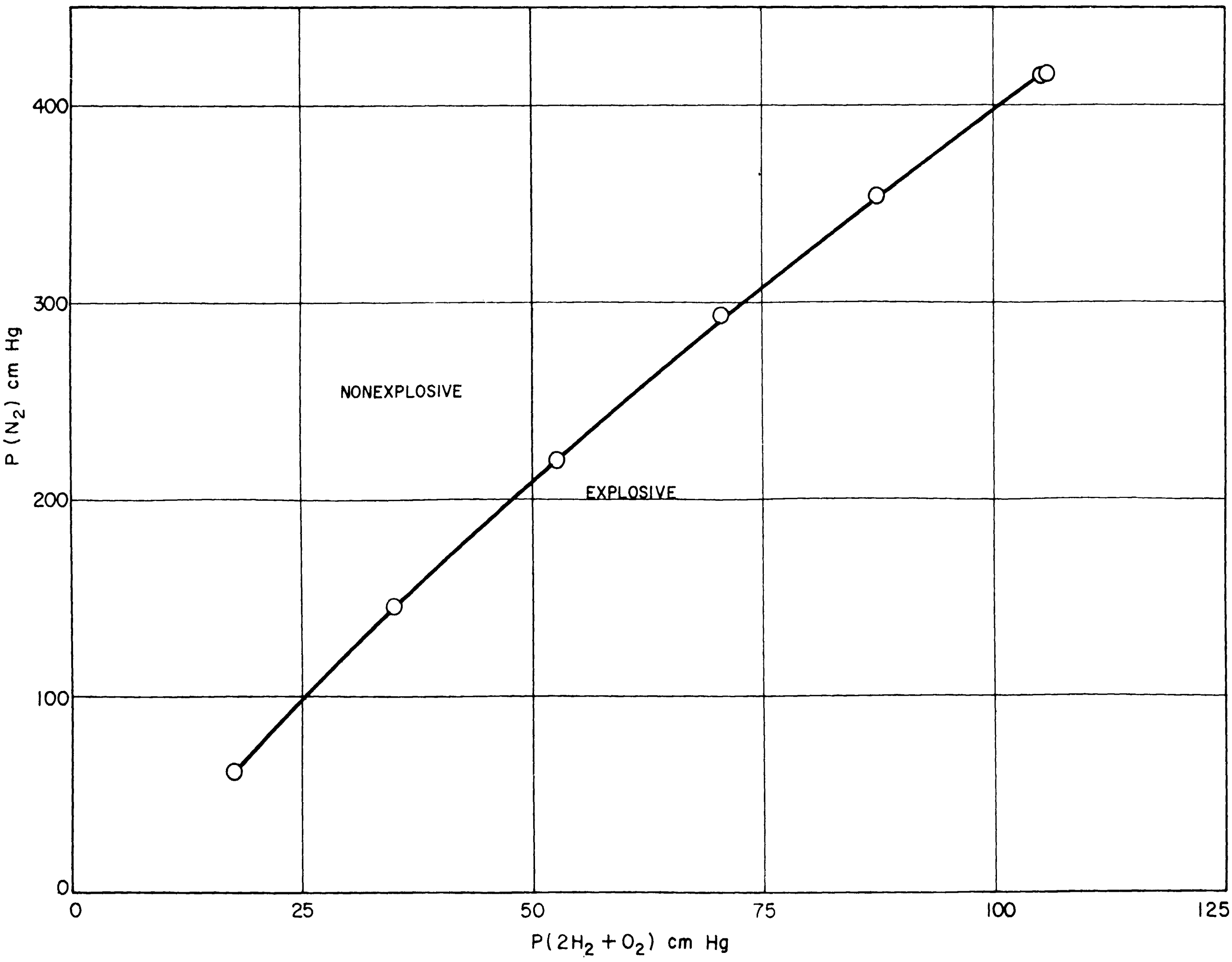

Fig. 6. Explosion Limit Curve for $(2H_2 + O_2)$ and N_2.

It should be pointed out that data taken up to March 8, 1954, indicated somewhat higher values for the explosion limits than those presented here. From March 8 to 11 there was an abrupt drop in the limit. The data taken before March 8 are consistent, although incomplete. Since the data taken after March 12 are consistent and have remained so, only these data have been used. It must be admitted, however, that no reason for the observed drop has been discovered.

2. Hot Runs

It was originally expected that if the beta source produced any effect on the explosion limit, it should raise it markedly. Hence, a relatively few hot runs at each hydrogen-plus-oxygen pressure will provide the necessary information, if reactions are produced well within the region of 100 percent failure for the cold runs (see Fig. 5). A complete study of the limits at several points was not feasible because the dose rate changes rapidly with time.

D. DISCUSSION AND CONCLUSIONS

Table 1 gives a comparison of the thresholds for the cold and hot runs. The values for the hot runs are much less precise than those for the cold runs because only a relatively small number of runs could be made. Note also the wide spread in dose rates for a particular point.

TABLE 1

THRESHOLDS FOR HOT AND COLD RUNS

$P(2H_2+O_2)$	$P(N_2)$ at Threshold Cold	Hot	Range of Dose Rate, R/hr
17.5	62.1	62.0	1.6×10^5 1.1×10^4
34.9	145.7	143.0	3700 3000
52.4	220.3	215.5	4600 370
70.3	294.2	284.0	4×10^4 7900
86.9	353.8	352.3	920 220
104.6	414.4	407.8	9.4×10^5 1.5×10^5
105.2	415.3	(Made after previous hot run)	

The effect seems to be more pronounced at the higher pressures and negligible at the low pressures. Figure 7 gives a comparison of the two series of runs used to find the thresholds for a pressure of hydrogen plus oxygen of 105 cm. It is clear that a number of failures occur in a nitrogen pressure range which had previously shown 100 percent reaction.

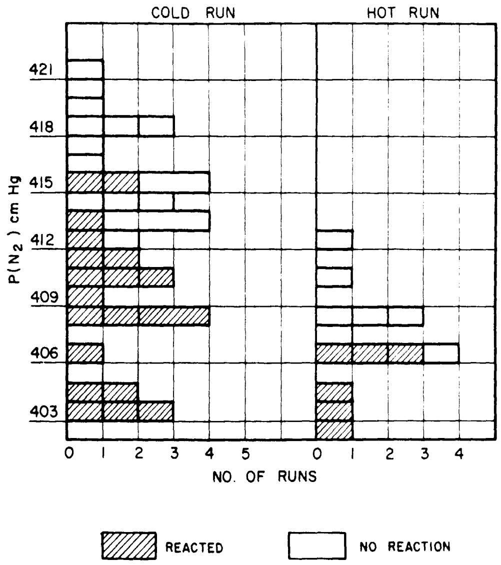

Fig. 7. Comparison of a Series of Hot and Cold Runs.

The value at 87 cm of hydrogen plus oxygen is the only one which was found as accurately as the values for the cold run. Note that the dose rates in this case are much below those pertaining to the other "hot" threshold data and that the threshold essentially coincides with that of the cold runs.

The effects of the beta source proved to be disappointingly small and in the reverse direction from that expected, i.e., there was a small but definite depression of the explosion limit.

E. REFERENCES

1. Brownell, L. E., et al., "Utilization of the Gross Fission Products", Progress Report 3 (COO-91), Univ. of Mich., Eng. Res. Inst. Proj. M943, June 30, 1952.

2. Brownell, L. E., et al., "Utilization of the Gross Fission Products", Progress Report 5 (COO-196), Univ. of Mich., Eng. Res. Inst. Proj. M943, September, 1953.

3. "Nuclear Data", National Bureau of Standards Circular 499 (1950).

4. Burtt, B. P., "Absolute Beta Counting", Nucleonics 5, No. 2, 28 (1949).

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

Personnel:

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

Senior Research Assistants: B. G. Bray, David E. Harmer

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A. INTRODUCTION

Since the preparation of Progress Report 6, work has been carried out on three major fronts: (1) The polymerization of ethylene under gamma radiation has been studied over a wide range of temperature. At the same time the copolymerization of ethylene and sulfur dioxide has received a small amount of additional attention. (2) The rate of reaction of chlorine with toluene under gamma radiation has been studied in more detail than reported by this laboratory earlier, and fractions of the product have been subjected to further tests to prove the presence of large amounts of the addition product of chlorine and toluene. (3) On the basis of reaction-rate data for the chlorination of benzene under gamma radiation, a preliminary design has been made of a plant to produce benzene hexachloride. A comparison is given between the estimated cost of the product from this gamma-activated process and product from the conventional ultraviolet process which is employed commercially.

The following sections of this report describe these studies in greater detail.

B. POLYMERIZATION OF ETHYLENE

The study of the effect of gamma radiation on the polymerization of ethylene as inaugerated by Lewis[2,3,4,21] has been considerably extended. Lewis carried out all polymerization runs at room temperature and at varying pressures and radiation dosages. In the work reported here, primary interest has been in varying the temperature of the polymerization reaction to determine its effect on the rate of the reaction and the nature of the products.

1. Equipment Used

The high-pressure stainless-steel bomb built by Lewis[21] was used for all polymerization runs. A set of heaters built by Lewis was placed around the bomb in all runs, regardless of whether heating was required, so that the radiation flux within the bomb would be the same in all cases. The bomb was loaded and emptied in the same manner described by Lewis.[21] The temperature was raised from 13 to 220°C, while the reacting pressure of pure ethylene was varied from 1075 to 1950 lb/sq in.

2. Visual Inspection of the Products

In the case of the three runs at the lowest temperatures (runs 145800, 145801, and 145807 described in Table 2) three distinct layers of solid polyethylene product were found in the bomb at the conclusion of the runs. The top layer was a spongy, white mass that adhered to the sides of the bomb with a physical consistency much like that of a fine porous sponge. The middle layer was a white curdy material, much resembling the curds in cottage cheese, and was directly opposite the center of the gamma radiation source; therefore, receiving the highest dose rate. The bottom layer was a hard solid layer that rested on the bottom of the bomb. This layer was from three quarters to two inches thick and had to be cut out of the bomb with a chisel.

Runs 145808 and 145812 yielded a finely divided white powder which adhered to the sides of the bomb. There was no particular variation in the physical appearance of this powder within the bomb. The powder did not always fall to the bottom of the bomb, but usually remained attached on the sides of the stainless-steel vessel.

The runs conducted at the highest temperatures (runs 145813 and 145814) were made at approximately 220°C and yielded a creamy white opaque liquid of high viscosity with an odor similar to that of used motor oil. After standing several days, a finely divided white powder precipitated our from the product and left a clear cream-colored liquid layer.

TABLE 2

IRRADIATION OF ETHYLENE

Run Number	Initial Pressure, psig	Reaction Temp, °C	Hours Irradiated	Averaged over Reactor: Dose Rate, kilorep/hr	Averaged over Reactor: Total Dose, megarep	Polymer, gm	Radiation Yield A, gm-moles reacted (metric ton)(megarep)
145800	1450	13	65.1	54.2	3.53	21.8	2370
145801	1260	13	91.75	53.8	4.94	39.7	3570
145807	1950	39	70.9	53.5	3.79	45.6	5860
145808	1500	88	71.33	26.6	1.90	1.4	438
145812	1400	90	91.93	47.0	4.32	3.8	542
145813	1425	220	77.8	53.0	4.12	6.3	1695
145814	1075	217	114.5	35.4	4.06	17.9	3860
145817*	940	25	98.25	37.8	3.72	15.1	2130
145824**	410	25	64.0	62.5	4.00	33.9	7030

*42 psig gaseous SO_2 added to reaction.

**20 gm liquid SO_2 added to reaction; vapor pressure was 42 psig.

3. Physical Properties of the Solid Polyethylene Product

The radiation yields (A values) given in Table 2 are in general higher than those reported by Lewis[21] but the dose rate to the reactor also was usually higher. The melting points and tensile strengths of the polyethylene prepared by polymerizing ethylene in a field of gamma radiation are listed in Table 3. The tensile strengths are somewhat higher than those reported by Lewis. These tensile strengths were made on an Instron tensile tester with a head and/or jaw speed of ten inches per minute. This speed was selected to eliminate the cold flow phenomeon observed in plastics in general and polyethylene in particular. The tensile strengths determined by Lewis were made on a Gardiner-Parks tensile testing machine where the cold-flow phenomeon was present.

The film for the tensile specimen was molded in a two compartment mold at 300°F at 1500 psi for 5 minutes. The sample was then punched from

TABLE 3

PROPERTIES OF IRRADIATED ETHYLENE

Run Number	Sample Section	Melting Point, °F Lower	Melting Point, °F Upper	Tensile Strength, psi	Elongation at Rupture, in./in.
145800	Middle	258.4	>700	3354	.24
145801	Middle	263.0	>700	3211	.23
145801	Top	259.0	>700	2947	.30
145807	Top	257.2	>700	2455	.24
145807	Bottom	260.4	>700	2600	.25
145808	Total	222.0	230.4	Too small sample	
145812	Total	217.2	221.7		
145813	Total	Liquid		Liquid	
145814	Total	Liquid		Liquid	
145817	*Total	>465			
145824	*Total	>450			

*Decomposes before melting at the temperature indicated.

the film with a standard sample die. The sample was somewhat larger than those used by Lewis.

Melting points of the polyethylene solids were determined on the laboratory melting-point bar[3] and in general are higher than those obtained by Lewis. They are, however, consistent with those of Lewis when account is taken of the difference in dose rates.

4. Other Work on Polymerization of Ethylene

Hayward and Bretton[17] at Yale University, working with a 400-curie gamma ray colbalt-60 source, have investigated the polymerization of ethylene under conditions ranging from 80 to 460°F and from 0.5 to 21 atmospheres. Both liquid and solid products were obtained.

Lewis[21] observed the effect of pressure on the polymerization of ethylene in both 1-kilocurie and 10-kilocurie sources at this laboratory. He observed little effect of pressure at constant temperature on the rate of polymerization.

A comparison of the ranges of temperature and pressure covered by Hayward and Bretton and by Lewis is shown in Fig. 8 of this report.

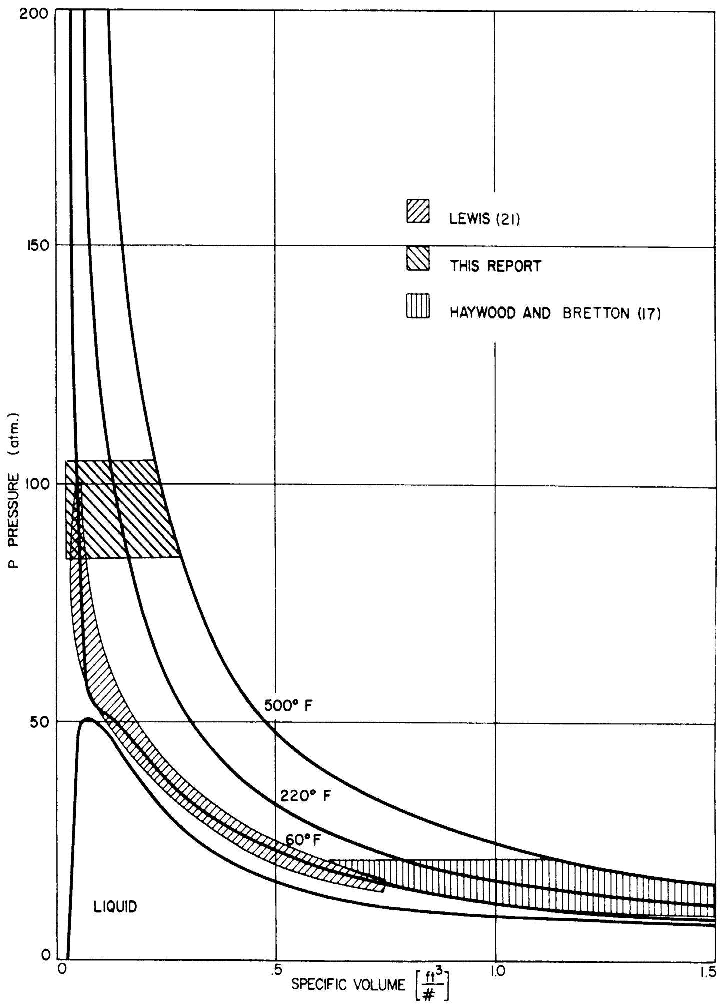


Fig. 8. Regions of Investigation of Polymerization of Ethylene under the Influence of Gamma Radiation.

C. POLYMERIZATION OF ETHYLENE AND SULFUR DIOXIDE

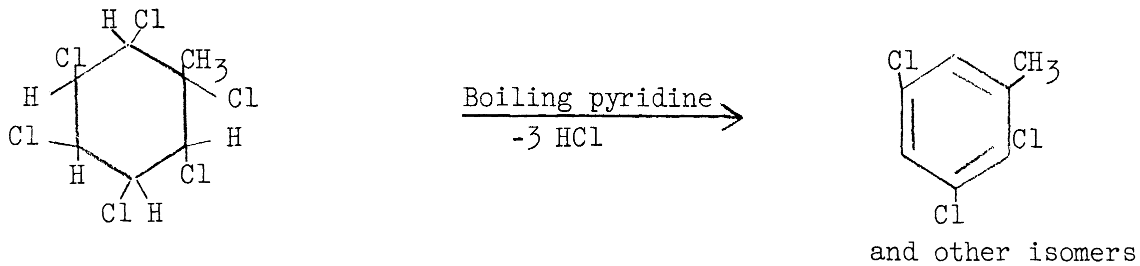
As reported by Lewis, the addition of sulfur dioxide to ethylene in the reaction bomb under gamma radiation produces a copolymer of the two compounds. The results of the two runs conducted in this study are reported in Tables 2 and 3. A fluffy white granular product was obtained which decomposed before melting at greater than 450°F; leaving a brown and black residue. All attempts to press this polymer into a film in order to obtain tensile test specimens resulted in decomposition of the product. The film produced contained black and brown decomposition specks and was very brittle.

These two runs are reported here in the interest of coupling with the results of prior work. The physical properties of these products are being investigated more fully and future work is palnned in the copolymerization of the two compounds.

D. CHLORINATION OF AROMATIC COMPOUNDS

1. Investigation of the Structure of the Primary Product Obtained from the Gamma Ray Activated Reaction of Toluene with Chlorine

(a) Dehydrohalogenation. The supposed hexachloro addition product which has been obtained by reaction of toluene with chlorine under gamma radiation[4] has been investigated more closely in order to establish whether it is indeed the postulated product. The first step in the investigation was the degradation to a supposed trichloro toluene by means of dehydrohalogenation in boiling pyridine.



Previously it was demonstrated that nearly one-half of the total chlorine content was eliminated during this reaction.[3] As starting materials for the reaction, mixtures of high-boiling products obtained from the chlorination runs in the 10-kilocurie-source equipment[4] were used and several dehydrohalogenations were carried out.

The initial materials used for one dehydrohalogenation were 176 gm of a material boiling at 123 to 154°C (50 to 60 microns of Hg) mixed with 89 gm of a material boiling at 125 to 158°C (65 to 75 microns Hg). The theoretical chlorine content for a hexachloro addition product of toluene and chlorine is 69.8 percent; the materials used contained 69.8 to 70.1 percent chlorine, respectively, indicating the presence of some higher chlorinated material in the latter of the two. This mixture of materials was added to about 1.5 l of redistilled pyridine and refluxed for 47 hours. On cooling, crystals, presumably pyridine hydrochloride, formed in the dark green liquid. The material in the flask was added to a large excess of dilute cold hydrochloric acid, and after standing, the upper aqueous layer was decanted off. The organic residue was heated to about 70°C and filtered; some gummy residue which seemed to contain most of the dark green coloration remained on the filter paper. The filtrate was a dark brown oil, density 1.4 gm/cc, weight 87.7 gm. Assuming the starting materials to be $C_7H_8Cl_6$ and the oil to be $C_7H_5Cl_3$, a yield of 53.3 percent is indicated for the dehydrohalogenation reaction.

The oily product from the dehydrohalogenation was fractionally distilled in a Podbielniak column at atmospheric pressure; a summary of these data is presented in Table 4. Each of the first nine fractions obtained by distillation was recrystallized from methanol, while the remainder, being nearly insoluble in methanol, was recrystallized from ethanol. Fractions three through eight gave two phase mixtures when the methanol was partially evaporated and the resulting residues cooled, so these phases were separated and crystals taken from each. Melting points were taken on a melting-point bar. Some fractions were analyzed for chlorine content by the use of sodium diphenyl reagent followed by a Volhard titration, but the values thus obtained are only approximate because of small sample size. Carbon-hydrogen analyses were also taken on certain samples.

It is apparent from the chemical analyses of these materials that both tri- and tetrachloro compounds are present, although the majority of material appears to be the expected trichloro compound. The presence of the tetrachloro material may arise from either or both of two causes; the dehydrohalogenation may have been incomplete, or the higher chlorine content of part of the starting material may have been carried through as an extra chlorine atom in some of the molecules.

The melting-point ranges found in many of these compounds indicate that separation was not complete. However, they indicate that one of the products is undoubtedly the known compound, 2,4,5-trichlorotoluene, whose melting point is given in the literature as 80 to 82°C[11,24,28] and whose boiling point has been given as 229 to 230°C.[30]

(b) Oxidation of the Dehydrohalogenated Material. A quantity of supposed hexachloro addition product of toluene and chlorine was dehydrohalogenated as before and then oxidized to obtain an acid. For this treatment, 60 gm of material boiling at 90 to 156°C (90 to 240 microns of Hg) were placed in 100 ml of pyridine and heated at just below the reflux temperature for 48 hours. Water was added to extract the pyridine hydrochloride and excess pyridine. The organic layer was neutralized with nitric acid, then 750 ml of 20% nitric acid were added. About one-half of this two-phase mixture was sealed into glass tubes and heated for 66 hours at 135 to 153°C. On cooling, large floculant crystals and a solid phase separated out from the solution.

TABLE 4

FRACTIONS OBTAINED BY THE DISTILLATION AND RECRYSTALLIZATION OF THE DEHYDROHALOGENATED PRODUCT FROM REACTIONS OF CHLORINE WITH TOLUENE UNDER GAMMA IRRADIATION

Fraction Number	Weight, gm	Boiling Range, °C (atm pressure)	Upper or Lower Layer from Methanol Mixture	Melting Point, °C	% by Analysis Cl	C	H
1	0.9	210.5-231.0					
2	1.3	228.0-238.5					
3	8.0	238.5-241.5	Lower	77.5	54.0	43.0	2.68
4	1.5	242.0-242.5	Lower	77.3-78.6			
			Upper	79.8			
5	9.3	241.5-242.0	Lower	77.5-78.0	54.2		
			Upper	77.8-78.6			
6	4.4	242.0-243.0	Lower	78.0-78.6	56.8	43.0	2.50
			Upper	76.4-77.8			
7	2.7	243.0-245.5	Lower	76.4-78.0			
			Upper	77.0			
8	3.0	245.5-257.0					
9	0.6	258.0-272.0		66.7-70.2			
10	1.3	273.0-277.0		80.2-88.8			
11	3.3	277.0-279.5		88.2-89.8	61.4	36.7	1.86
12	1.1	279.5		80.2-86.6			
13	1.1	277.0-278.5		87.6-88.8	61.1		
14	1.6	278.5		87.5-90.0	61.1		
15	3.1	Hold-up drained from column	Not recrystallized	94.0-96.8	61.4		

Theoretical values for percent of C, H, and Cl:

Trichlorotoluene: C = 43.00 %, H = 2.58 %, Cl = 54.4 %

Tetrachlorotoluene: C = 36.6 %, H = 1.75 %, Cl = 61.7 %

Tetrachloromethyl cyclohexadiene (incomplete dehydrohalogenation product): C = 36.2 %, H = 2.61 %, Cl = 61.2 %

The solids were filtered off from the nitric acid solution and dissolved in ammonium hydroxide solution. This solution was extracted with toluene to remove nonionic materials. The organic acid was then precipitated by addition of nitric acid, filtered, washed, and dried. The yield was 9.5 gm, or 42 percent overall, based on the original starting materials and assuming trichlorobenzoic acid as the product.

Identification of the final product required separation of the crude product into its various isomers. Since each isomer should differ in its acid strength, the first attempted separation was based on this property. A series of extractions was carried out on 4 gm of acid so that during each step one-half of the acid would dissolve in an aqueous ammonium hydroxide layer, while the other half would be retained in the organic phase. These extractions were carried out until 32 divisions were obtained. However, when the melting points of these 32 fractions were taken, long ranges indicated that practically no separation had been effected.

Since the differences in acid strength of these materials thus appeared to be quite small, it seemed advisable to turn to a chromatographic type of separation. Still making use of the acid strength characteristics of the material, ion-exchange chromatography was employed. A highly basic anionic resin, Dowex-1, 8 percent cross linked, 200 to 400 mesh, was used. Two columns were used in series, the first 400 x 10 mm ID and the second 20 x 6 mm ID, each being packed two thirds full of resin. The first column was jacketed and water at 55°C was circulated around it. Before use, the resin was backwashed and put through a chloride-hydroxide cycle twice.

After several experiments, it was found that the following conditions were satisfactory for operation of the column. The resin was prepared for use in the chloride form. The mixed isomers of the acid (about 1 gm) were introduced at the top of the column as the ammonium salt solution in 75% ethanol (10 ml volume). The column was washed with distilled water and elution was carried out with 0.01N and 0.1N hydrochloric acid in 75% ethanol (25% water). Twenty 5-ml samples were collected at a slow rate, 1 to 16 with 0.01N acid and 17 to 20 with 0.1N acid. Initially, dark bands were observed to move down the column, but these became indistinguishable as they approached the base of the upper section

It was found on evaporation of the solvent from the fractions that the first two contained nearly no solids, while numbers 3 through 9 contained about equal amounts of solids. Numbers 10 through 12 contained small amounts of impure solids, and the remaining fractions contained only traces of solids. The solid materials were recrystallized from hot water.

The degree of separation of isomers obtained is illustrated in Fig. 9, which is a graph showing melting ranges of the series of samples.

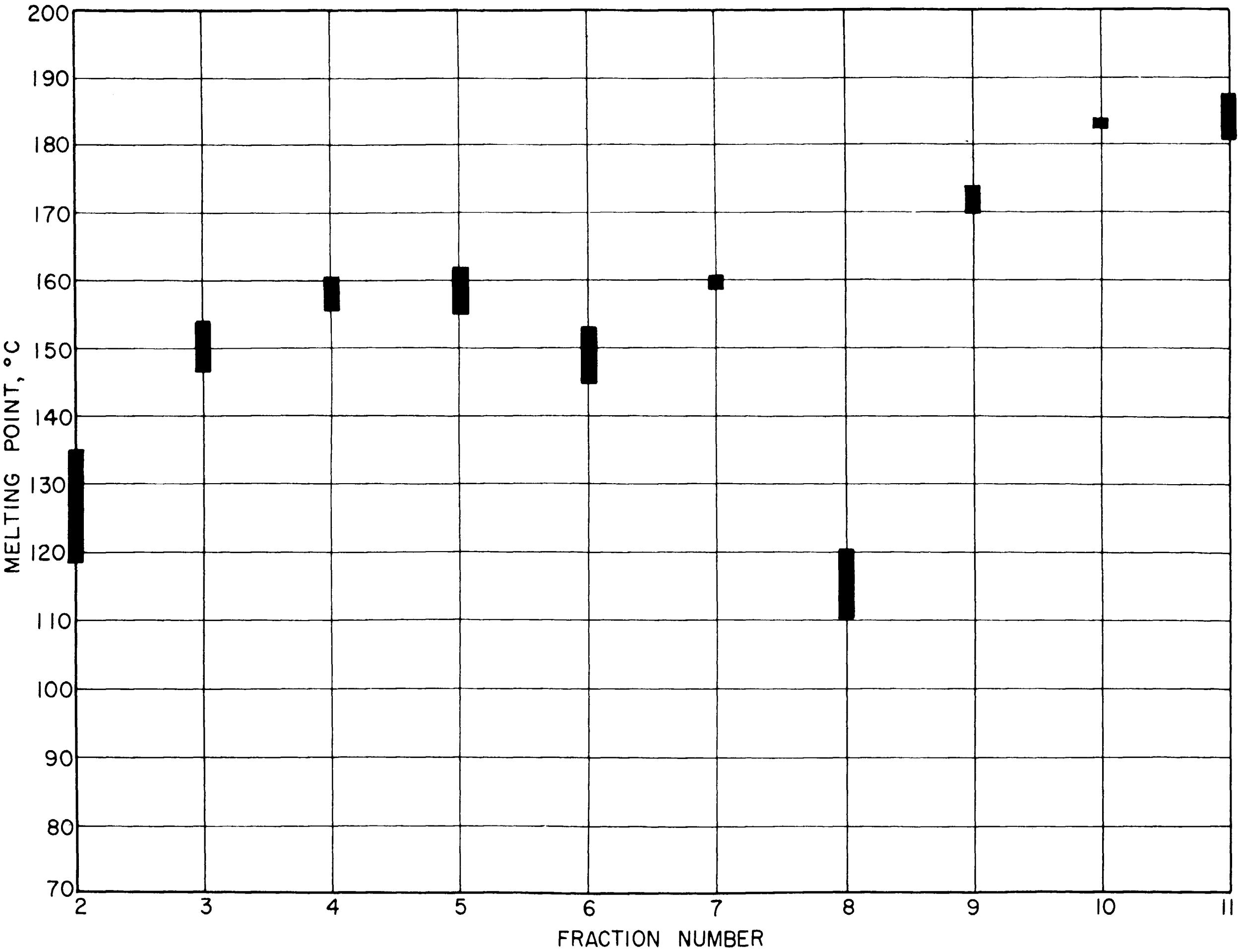


Fig. 9. Melting-Point Ranges for Fractions of Isomeric Trichlorobenzoic Acids Separated by Ion-Exchange Chromatography.

The melting-point maxima are accompanied by a decrease in temperature range showing an increase in purity of the material in that particular fraction. The much lowered melting points and long ranges of fractions 2 and 8 indicate possibly that these were collected at a point of transition from one chromatographic band to another. Fractions 7 and 10 exhibit the sharpest melting points and lie in a range close to that reported for isomers of trichlorobenzoic acid. The increased range but higher first melting point of fraction 10 could perhaps be attributed to the presence of amounts of a much higher melting isomer in this fraction. All melting points of this series of fractions are recorded in Table 5, together with chloride analyses and data for comparison with reported values for trichlorobenzoic acids.

Fractions 4 and 5 merit special attention. During the determination of melting points on a melting-point bar, these two fractions had the melting point listed when freshly contacted to the hot surface. However, when they were held below the melting temperature, part of the material

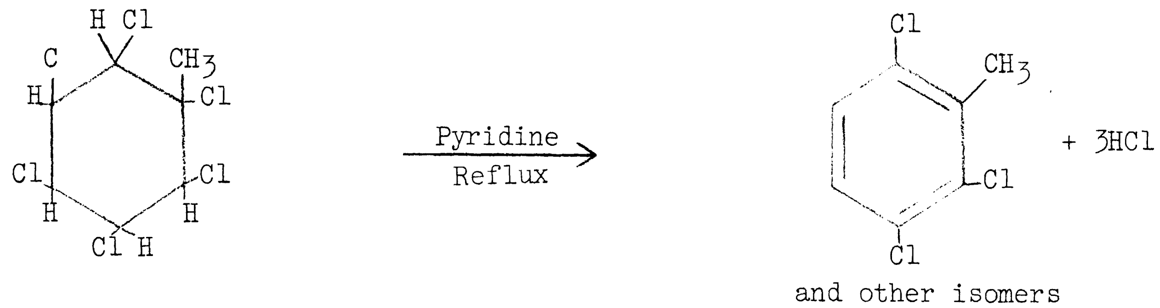
TABLE 5

FRACTIONS OBTAINED BY CHROMATOGRAPHIC SEPARATION OF ACIDS FROM THE DEHYDROHALOGENATED PRODUCT FROM REACTION OF TOLUENE AND CHLORINE

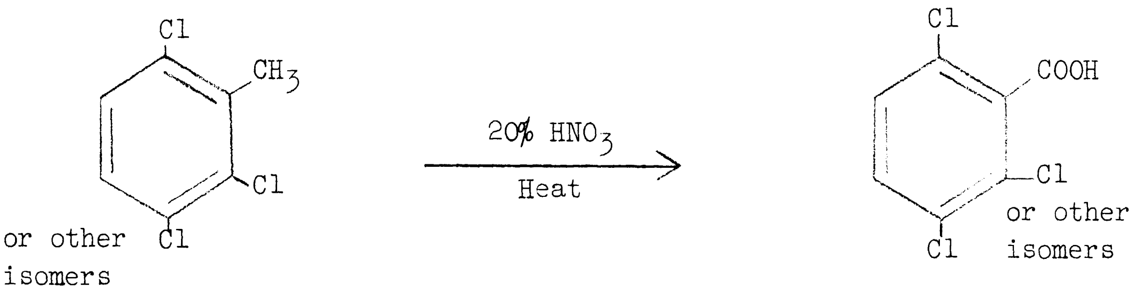
Fraction Number	Melting Point, °C	% by Analysis			Comparison Compound	Melting Point of Comparison Compound	% Calculated from Comparison Compound		
		Cl	C	H			Cl	C	H
2	118.7-135.0								
3	147.0-154.4								
4	156.0-160.6								
5	155.6-162.0								
6	145.6-153.2								
7	159.4-160.8	49	37.4	1.69	2,4,5 trichlorobenzoic acid	163	47.2	37.3	1.34
					2,3,6 trichlorobenzoic acid	163-4			
					2,4,6 trichlorobenzoic acid	164			
8	110.5-120.7								
9	170.2-173.8								
10	182.6-183.6	46			2,3,4 trichlorobenzoic acid	186-7	47.2		
11	180.8-187.2								

was found to sublime, leaving a material of a higher and sharper melting point. In a further investigation, quantities of fractions 4 and 5 were placed in a sealed melting-point tube. This tube was partially immersed in an oil bath at 180°C until a film of material had condensed on the cooler walls of the upper end of the tube. The tube was then sealed off at the center and melting points of the more volatile materials were taken. Table 6 is a summary of information on the products of this separation procedure and data for comparison with values reported for isomers of trichlorobenzoic acid.

Work on degradation of the product of the reaction of toluene with chlorine under gamma irradiation can be summarized by the following equations:



(1)



The products obtained by dehydrohalogenation and subsequent oxidation confirm the hypothesis that chlorine adds to the ring of toluene and further indicate that substitution of more halogen atoms for hydrogen atoms occurs in some of the molecules of the hexachloro addition compound.

2. Separation of Products of Chlorination of Toluene

(a) Attempted Crystallizations. Several unsuccessful attempts were made to obtain a crystalline solid from the syrupy form in which the hexachloro products were obtained. Carbon disulfide, methanol, petroleum ether, glacial acetic acid, and isopropyl alcohol were miscible with the chlorinated material; a mixture of 75% methanol in water formed a two-phase system when

TABLE 6

MELTING POINTS OF SEPARATED COMPONENTS OF FRACTIONS 4 AND 5 OF TABLE 5

Fraction	Melting Point, °C	Comparison Compound	Melting Point of Comparison Compound	% Calculated for Comparison Compound C	% Calculated for Comparison Compound H	% by Analysis C	% by Analysis H
4-sublimate	166.0-166.5	2,3,6 trichlorobenzoic acid 2,4,6 trichlorobenzoic acid	163-164 164	37.3	1.34	37.4 in mixture before sublimation	1.22 in mixture before sublimation
4-residue	163.2	2,4,5 trichlorobenxoic acid	163	37.3	1.34		
5-sublimate	166.0-166.5	2,3,6 trichlorobenzoic acid 2,4,6 trichlorobenzoic acid	163-164 164	37.3	1.34	37.4 in mixture before sublimation	1.69 in mixture before sublimation
5-residue	162.0	2,4,5 trichlorobenzoic acid	163	37.3	1.34		

mixed with the syrup, but repeated extraction procedures gave no evidence of separation of components of the chlorinated material or of production of crystalline solids.

(b) Analysis of Solids from the Reaction Mixtures. The liquid mixtures consist chiefly of unreacted toluene, benzyl chloride, and the chlorine addition products of toluene. The liquid product from several runs became cloudy after a period of standing and a small amount of very fine crystals slowly separated. The solids which formed were investigated further to determine whether they were crystalline 1,2,3,4,5,6-hexachloro-methyl-cyclohexane. They were separated from the mother liquors by centrifugation, placed on porous plate for oil removal, and recrystallized from methanol. Analysis for chlorine content showed variable composition (ranging from 72.1 to 72.8% chlorine) for materials from various runs and can probably be regarded as impure heptachloro products (theoretically 73.1% chlorine) arising from substitution of additional chlorine in the hexachloro product.

3. Investigation of Reaction Rates between Toluene and Chlorine

(a) Apparatus. A series of reactions between toluene and chlorine was carried out to obtain data from which the kinetics of this reaction could be determined in order to gain better insight into the nature of the activation produced by the gamma irradiation. The equipment used was a combination of certain parts described in previous reports. Gas handling and control equipment were similar to that outlined in Fig. 23 of Progress Report 6.[4] The glass reaction vessel shown in Figs. 62 and 63 of Progress Report 5[3] was used at various distances from the source and was connected to the glass lines leading to the gas-control apparatus located on the second floor.

In early experimental runs the amount of chlorine was ascertained by weighing the gas cylinder during the run, but it soon became apparent that this procedure was not accurate enough because of the inherent insensitivity of the type of balance usable under the large weight of the steel cylinder. An orifice-type meter was next used in which the pressure drop of the gas was measured as it passed through a calibrated glass orifice; results in this case also lacked reproducibility. Satisfactory measurements of the inlet gas flow were finally obtained with the use of a pair of "Rotometer" type flowmeters. These instruments were made of Pyrex with sapphire floats and were connected in series so that the two different ranges might be used without the need of stopcocks. They were calibrated against nitrogen, ethylene, butane, carbon dioxide, and ethane, according to the correlation of Martin.[23]

A four-way stopcock was used for taking samples of exit gas from the reaction; this made it possible to purge a gas absorption bottle with nitrogen, pass all the exit gas through it for a specified time, then purge out the connecting tubes with nitrogen again. The absorption bottles were provided with fritted-glass dispersion tubes to facilitate efficient absorption in the solution. The fritted tubes were calibrated frequently to assure that the pressure drop of the gas through each was not significantly different from the rest. During experimental runs, except for the periods during which samples were taken, the exit gas was vented to the atmosphere or passed through large absorption bottles containing 50% sodium hydroxide solution. Figures 10 and 11 illustrate the gas-handling equipment.

(b) Analytical Methods and Preliminary Results. The method used for analysis of exit gases, and hence, the choice of absorption solution, received considerable attention and several difficulties were encountered. Methods making use of liberated oxygen from hydrogen peroxide or of oxidation of iodine compounds were found to lack precision under the experimental conditions of this work. Since it can be shown theoretically that ferrous ion should be oxidized quantitatively to ferric ion by chlorine, an analytical procedure was devised making use of this reaction. In order to gain efficient solution of the gas, two liquids were used in each absorption bottle. A lower layer of reagent-grade carbon tetrachloride provided rapid absorption of excess chlorine in the exit gas, while an upper layer of aqueous acidic ferrous sulfate dissolved all hydrogen chloride in the gas stream. Examination of two absorption bottles in series showed that gas absorption was complete at the flow rates ordinarily used.

After absorption of the gas sample, the absorption bottle was shaken vigorously. This caused the dissolved chlorine in the lower layer to react completely with the ferrous sulfate of the upper layer. An aliquot portion of this layer was then titrated with ceric sulfate for ferrous ion content, which when subtracted from the original content gave the amount of chlorine absorbed. A second aliquot was oxidized with hydrogen peroxide to convert all ferrous ion to ferric ion and analyzed for chlorides by the Volhard method. Thus, free chlorine and total chloride could be determined from a single absorbed gas sample. Preliminary investigations indicated the method should be capable of giving precise results on free chlorine determinations. A series of experiments using the equipment previously described and using the ferrous absorption method were made. When the gas flowmeter had been properly calibrated and the reaction rates calculated, it was found that results were very scattered. Therefore, a very thorough investigation of analytical methods was imperative. Using samples of 99.5 to 99.8% pure chlorine gas, runs were carried out in which the gas passed through dry glass tubes only, and samples were taken in the usual manner. Several runs showed differences as great as 10 percent in the amounts of chlorine measured at the flowmeters, analyzed as free chlorine, and analyzed as chloride. When

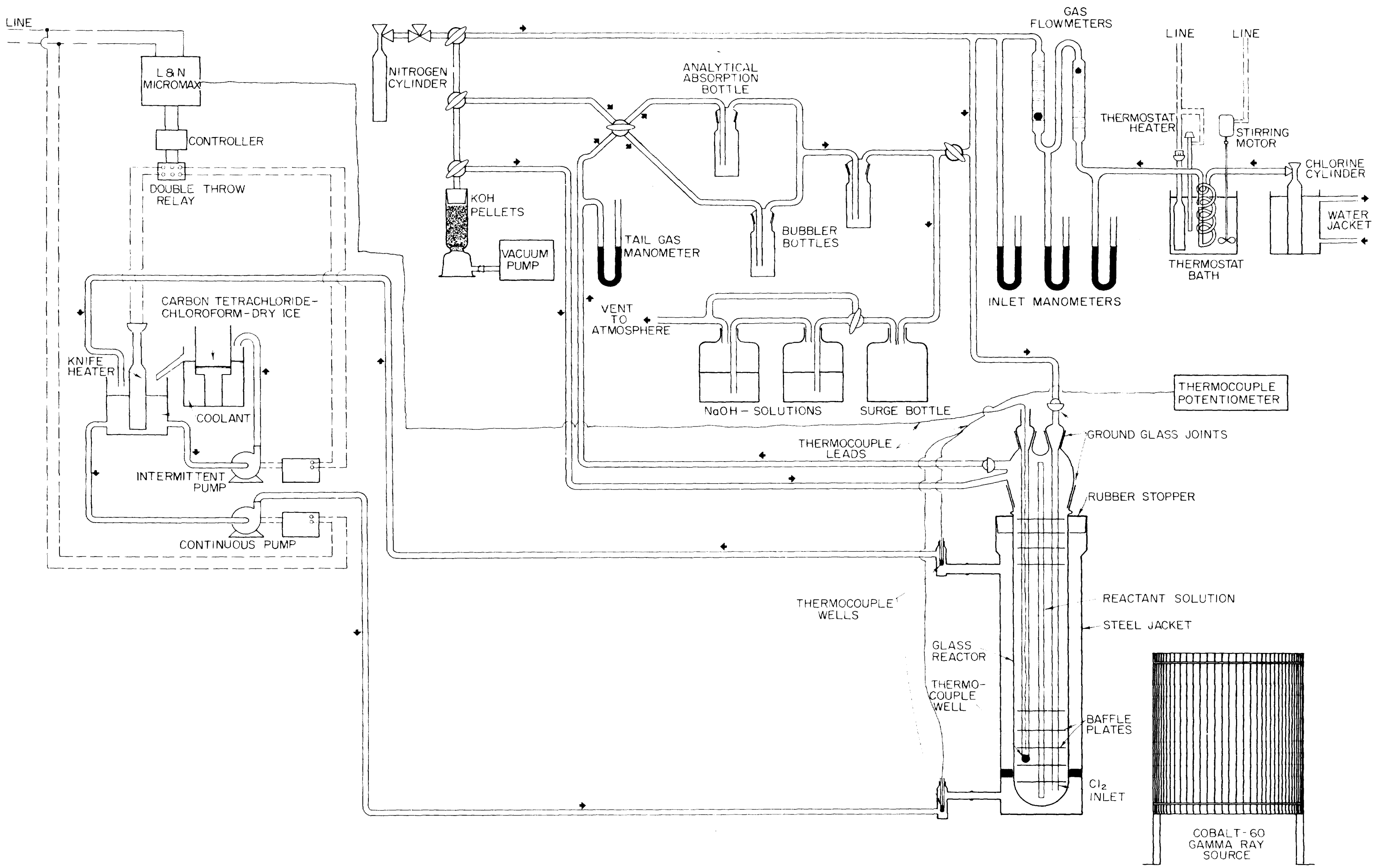


Fig. 10. Flowsheet for Equipment Used To Determine Reaction Rates between Toluene and Chlorine.

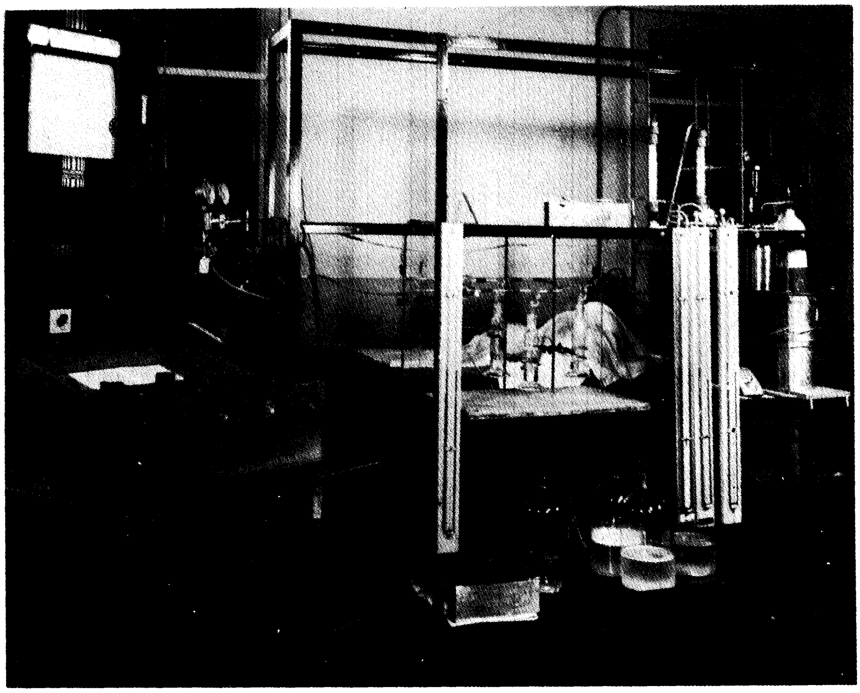


Fig. 11. Gas-Control Equipment Rack Used for Reactions between Toluene and Chlorine.

samples were absorbed in potassium iodide and titrated with sodium thiosulfate, a significantly different deviation from the flowmeter values was obtained. When potassium hydroxide was employed as abosrbent, followed by hydrogen peroxide reduction of the hypochlorite formed, the chloride ion titrated in this solution showed still a different deviation from the flowmeter value. A satisfactory explanation for the failure of the ferrous sulfate method of analysis has not been found, but it was abandoned in favor of a more precise method.

Absorption of chlorine in alkaline sodium arsenite has been published as an analytical method.[12,34,37] By use of 0.2N sodium arsenite in 3N potassium hydroxide, it was found that absorption of the exit gas stream was rapid and complete. For analysis, an acidified aliquot portion was titrated with ceric sulfate, giving free chlorine by difference. A second aliquot was acidified and analyzed for chloride ion by the Volhard titration method. Thus, free chlorine and total chloride can be determined from a single absorption sample. The arsenite solution is sufficiently stable to eliminate the need for an inert atmosphere and the use of carbon tetrachloride as an absorbent is avoided. Agreement between free chlorine and chloride values is good, with deviations of 1 percent or less. A somewhat larger discrepancy is noted between the flowmeter values and the results of these analytical methods. A small correction on calculated flowmeter values may be necessary.

(c) Equipment Changes. Further refinements of technique for kinetic runs have involved certain modifications in equipment. A new all glass reactor, illustrated in Figs. 12, 13, and 14, has been constructed. The stream of chlorine gas is caused to bubble back and forth across a series of five baffle plates. Above this is a space for foam separation. Further up the tube is a series of baffles, placed so that the exiting gases follow a spiral path throwing entrained spray against the outer walls of the tube. The chief advantages of this design are larger contact time between the gas and liquid phases, more efficient separation of entrained liquid, less open space for gas above the liquid with resulting decreased lag in the time required for gases to return for analysis, complete separation of parts for cleaning, and location of the thermocouple well in a position for most accurate indication of temperature.

A steel jacket has been constructed to fit the new glass reactor in order to provide for a very rapid linear flow of coolant along the walls of the reactor tube. New larger diameter tubes have been installed in the 10-kilocurie-source room to permit the coolant circulation equipment to be placed outside the source room without causing undue pressure drops in the lead lines. This combination of improved cooling equipment should make temperature control more precise and flexible.

In order to insure that the glass reaction vessel is always free from contaminants which might catalyze or inhibit the reaction, special cleaning procedures have been adopted and equipment constructed to facilitate these procedures. After cleaning with solvents and drying, the reactor is placed in a mixture of concentrated sulfuric acid and chromic acid for a few hours. It is then rinsed in distilled water and placed in a steam cleaning tower. About one and one-fourth liters of water is vaporized and passed through this tower so that all surfaces of the dissassembled reactor are thoroughly steamed. The parts of the reactor are then dried at 150°C in a special oven wherein the glass parts rest on a plate glass shelf and do not come in contact with any metal. When blown out with filtered air and cooled, the reactor is then assembled for use and filled with reactant liquid. This new equipment is now being used to study the kinetics of the chlorination reactions.

E. DESIGN OF A PLANT FOR THE CHLORINATION OF BENZENE UNDER GAMMA RADIATION

Benzene and chlorine react under a variety of conditions to produce either addition or substitution of chlorine to the benzene ring. In 1903, Slator[32] first showed that ultraviolet light selectively promotes the hexachloro addition product of chlorine and benzene, 1,2,3,4,5,6-hexachlorocyclohexane. Since 1903 the reaction has been studied under the influence of

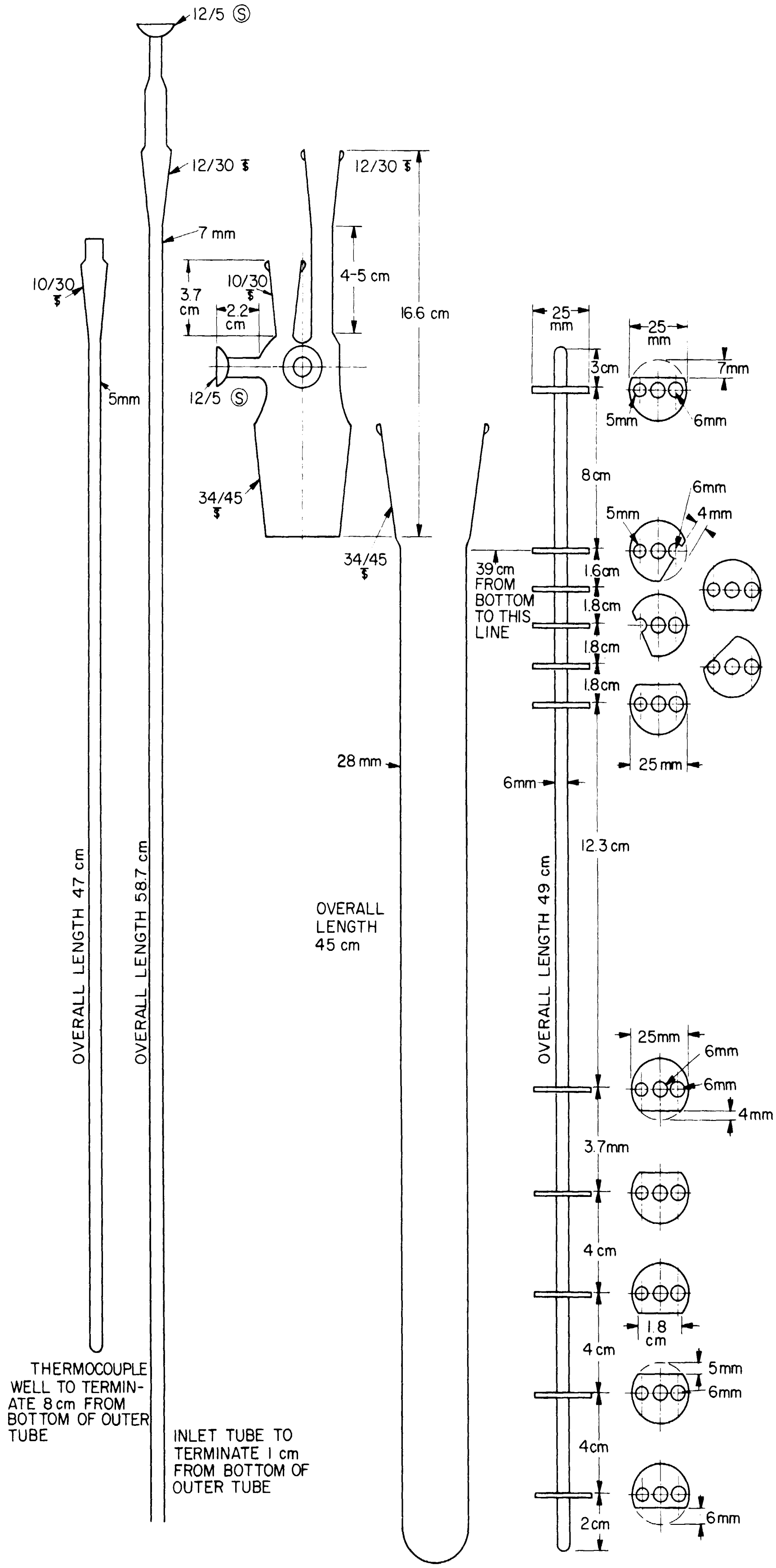


Fig. 12. Drawing of Glass Reactor Used in Determining Reaction Rates between Toluene and Chlorine.

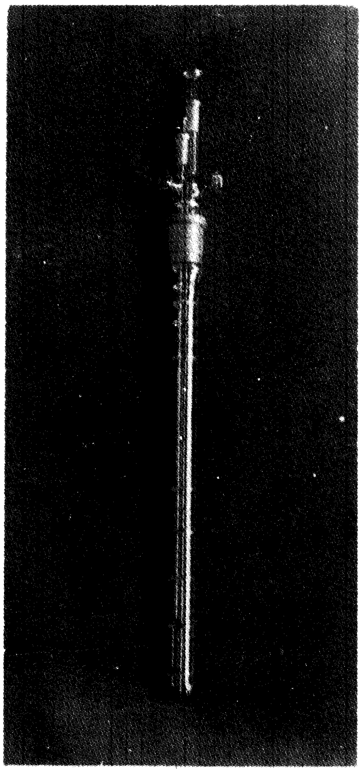


Fig. 13. Glass Reactor Used for Determining Reaction Rates between Toluene and Chlorine.

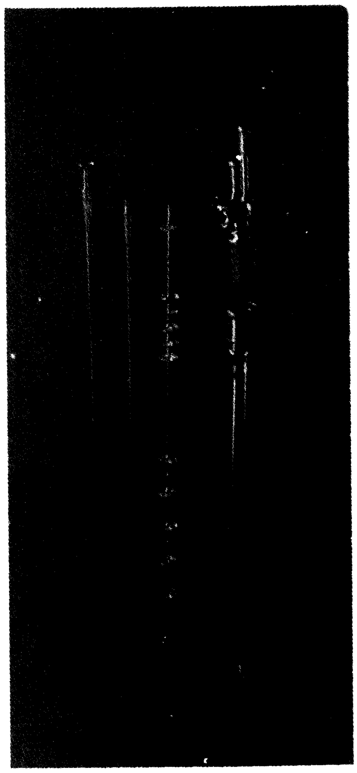


Fig. 14. Glass Reactor, Dissassembled.

ultraviolet light,[10,14,22,25] alpha radiation,[1] and gamma radiation.[2,16,21] Studies using gamma radiation were made in this laboratory with cobalt-60.

The commercial importance of this addition product lies in the insecticidal properties of the gamma isomer. These properties have been described by Slade[31] and others.[9,10,18]

1. Laboratory Chlorination of Benzene in the Presence of Gamma Radiation

The procedure for chlorinating benzene under the influence of gamma radiation has been described previously.[2,16,21] In brief this procedure is as follows:

> A glass reactor containing benzene and carbon tetrachloride was lowered into the cobalt-60 gamma source and purged of air with nitrogen. A cooling medium was passed through an outer jacket of the reactor. Chlorine was bubbled into the reaction mixture until the first traces of HCl or Cl_2 appeared in the off gas from the reactor and then was regulated at a rate so that there was no HCl or Cl_2 in the off gas. The reaction was usually conducted until all the benzene was consumed.

It was reported[2] that the temperature could not be controlled during the reaction if pure benzene was used; therefore, carbon tetrachloride, an inert in the reaction, was added to give greater reaction volume for closer temperature control and also to act as a solvent for the solid reaction product.

Part of the data taken has been reported[2,16,21] and this is further supplemented by Table 7 and Fig. 15. Due to equipment breakage and recovery problems, the solid product in some cases was not obtained. Rather than discard these runs as useless, a weight percent completion of reaction has been assumed. In those runs where an obvious excess of chlorine was added, the average completion obtained was assumed; this was 95-percent completion based on the benzene. In those runs where the reaction was stopped before completion, the amount of chlorine added was recorded and 95 percent of the benzene that would react with the chlorine stoichiometrically was calculated. This amounted to 90 percent completion based on the total benzene concentration. The reaction velocity constant for the complete reaction was calculated. This constant k_{tr} is due to the combined temperature and radiation effects. A discussion of this combined constant is to be found in the theoretical work by this laboratory in Progress Report 5[3], pages 36-40. Figure 15 is a plot of this constant versus the reciprocal of the absolute temperature at a constant dose rate of 105 kilorep per hour. [G] values have also been calculated and are to be found in Table 7.

2. Rate of Reaction for the Addition Product of Chlorine and Benzene

The original investigation of this laboratory was concerned with the effect of gamma radiation on the yield of gamma isomer in the mixed isomer reaction products and it was determined that there was no apparent affect of gamma radiation on yield of gamma isomer. The rate of addition of chlorine to the reaction mixture was not constant nor uniform, and no data on the kinetics of the reaction based on chlorine concentration are available. This type of data is necessary to design a flow reactor for the system.

In the literature, Noyes and Leighton in Photochemistry of Gases[25] report the rate as approximately

$$r = -\frac{dP}{dt} = kI^{1/2} [Cl_2] [C_6H_6] .$$

In Chemical Action of Ultraviolet Rays,[10] the rate is given as

$$r \alpha (p_{Cl_2})^2$$

TABLE 7

IRRADIATION OF BENZENE AND CHLORINE BY COBALT-60 GAMMA RADIATION AT A DOSE RATE OF 105 KILOREP PER HOUR

Run Number	Volume C_6H_6, %	Yield $C_6H_6Cl_6$, gm	Average Reaction Temp, °R	Reaction Time, min	Weight Percent Completion on C_6H_6	Reaction Velocity Constant, k_{tr} $\frac{\text{gm-moles}}{(\text{hr})(\text{ft}^3)}$	G
129762	100						
129765	100	49.6	498	35	15.2	82.6	47,100
129766	30	*93	520	40	**95	136.0	51,600
129767	30	*93	525	35	**95	155.5	59,000
129768	30	102.8	526	25	105	241.0	91,400
129774	30	40.0	474	33	40.8	70.4	26,800
129861	25	*52.6	528	15	***90	205.0	76,000
129855	20	*62	528	14	**95	258.5	91,500
129769	20	*48	520	24	***90	116.5	42,500
129770	20	63.6	528	20	97.3	186.0	68,000
129771	10	32.0	528	25	98.0	74.7	26,100
129772	10	31.7	483	53	97.2	34.8	12,200
129773	10	40.8	477	46	125	51.8	18,100

*Calculated values using assumed percent completion.

** Weight percent completion on C_6H_6 assumed by average of other runs where excess chlorine was introduced.

***Weight percent completion on C_6H_6 assumed on runs where stoichiometric amounts of chlorine were added.

$$r \propto I^{1/2} \, p_{Cl_2} \, p_{C_6H_6}$$

$$r \propto I^{(1.6)} .$$

Luther and Guldberg report[22]

$$r \propto [Cl_2] ,$$

while Slator reports[32]

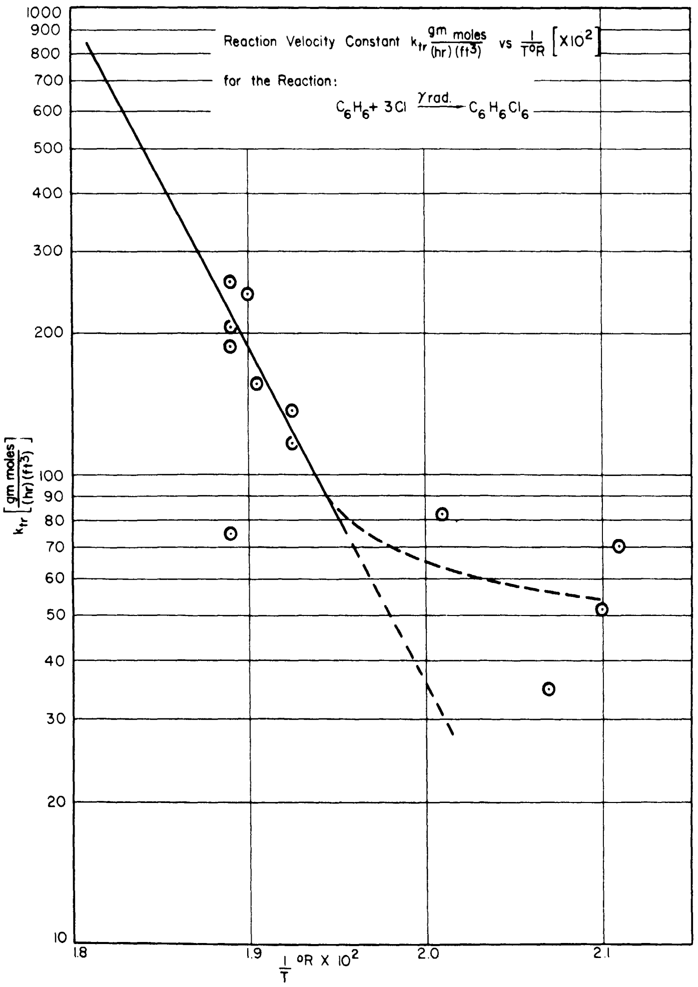


Fig. 15. Reaction Velocity Constant vs Reciprocal Temperature for the Chlorination of Benzene.

$$r \alpha T^{1.5}$$

and 10^6 molecules of reaction product per quantum of light, where

P = total pressure
p = partial pressure
I = ultravoilet intensity
T = temperature
r = rate
t = time
k = reaction velocity constant
[ ] = concentration.

3. Process Design Calculations for a Continuous Flow Reactor

For the purpose of this design calculation, it is necessary to make several assumptions regarding the data available. Enumerated with reasons for their selection, these assumptions are:

(1) The reaction is first order with respect to the chlorine concentration. An excess of benzene is to be used and recycled through the reactor. This excess will reduce the effect of small benzene concentration on the reaction rate and also provide a medium to slurry out the solid product. This also assumes a similar mechanism of reaction under gamma radiation as under ultraviolet rays. Luther and Goldberg, quoted above, give reaction as first order with respect to chlorine.

(2) The data obtained in this laboratory for different reaction temperatures give the reaction velocity constant. The plant is designed using the same radiation rate as was present in the laboratory. Only temperature is to be extrapolated.

(3) Gunther[14] and others[9] report a yield of 42% gamma isomer in the reaction of chlorine and benzene under ultraviolet light rays on the addition of NaOH to the reaction mixture. An addition of 2% NaOH in the mixture is assumed to give 32% gamma isomer by weight.

(4) The same reaction mechanism holds for the case of a mixture of liquid chlorine in benzene as for the case of bubbling gaseous chlorine through benzene. This permits the use of the reaction velocity constant of Table 7 and Fig. 15. The same percentage completion on the chlorine will be held. This allows use of these overall constants.

With these assumptions, the equation and material balance for the reaction capable of producing 1000 lb of gamma isomer per day on 95 percent completion of the chlorine are as follows:

$$C_6H_6 + 3Cl_2 + \text{gamma radiation} = C_6H_6Cl_6$$

Basis: 1000 lb gamma isomer per day

$$\text{Wt mixed isomers per day} = \frac{1000}{.32} = 3120 \text{ lb}$$

$$\text{Wt benzene per day} = \frac{(3120)(78)}{(.95)\ (291)} = 882 \text{ lb}$$

$$\text{Wt chlorine per day} = \frac{(3120)}{(.95)} \frac{(71)(3)}{(291)} = 2410 \text{ lb}$$

$$\text{Wt NaOH per day} = (3290)(.02) = 65 \text{ lb.}$$

If 100 percent excess benzene is used for the reasons in assumption 1, then the weight entering the reactor is

$$(882)(2) + (2410) + (65) = 4239 \text{ lb day or } 177 \text{ lb/hr} .$$

This corresponds to a feed rate of 2.38 ft^3/hr at 85°F. The equations for the rate of the reaction in the flow reactor (with the assumptions noted) of volume V_R are

$$r = k_{tr} [Cl_2]$$

$$\int dV_R = -\int \frac{dNCl_2}{r} = -\int \frac{dNCl_2}{k\ Cl_2}$$

$$= -F \int \frac{dNCl_2}{kN_{Cl_2}} \quad ,$$

where

$$F = ft^3/hr \text{ Feed Rate}$$

$$\left(N_{Cl_2}\right) = \frac{(\text{lb-moles } Cl_2)}{(hr)} \text{ Chlorine Rate}$$

$$k = \frac{1}{hr}$$

$$V_R = ft^3 \quad .$$

By selecting 85°F as the operating temperature, a liquid phase reaction may be obtained at not too high a pressure. With the range of data available, long range extrapolation of the data is not advisable due to the possibility of error. Assuming that 95 percent of the chlorine is reacted in a single pass through the reactor, the limits of integration are

$$\text{Inlet } Cl_2 \left(N_{Cl_2}\right) = \frac{2410}{(71)(24)} = 1.41 \frac{(\text{lb-moles } Cl_2)}{(hr)}$$

$$\text{Outlet } Cl_2 \left(N_{Cl_2}\right) = (1.41)(.05) = .07 \frac{(\text{lb-moles } Cl_2)}{(\text{hr})} .$$

The reaction velocity constant from Fig. 15 at 85°F is

$$445 \frac{(\text{gm-moles})}{(\text{hr})(\text{ft}^3)} .$$

Converted to units of 1/(hr) this is

$$k_{tr} = \frac{445 \text{ (gm-moles)} \quad (\text{ft}^3) \quad (\text{lb-mole}) \quad 78 \text{ lb}}{(\text{hr})(\text{ft}^3) \; (.88)(62.4)\text{lb} \; (454)(\text{gm-moles}) \; (\text{lb-mole})} = \frac{1.4}{(\text{hr})} .$$

The volume of the reactor under these conditions is then by integration of the above expression

$$V_R = \frac{(2.38)}{(1.4)} \text{ Log}_e \frac{1.41}{.07} = 5.1 \text{ cu ft.}$$

Using a nominal three-inch OD high alloy stainless-steel or nickel pipe for a reactor, approximately one hundred feet are necessary. This would correspond to ten passes ten feet long in the field of gamma radiation.

The heat of reaction for this system calculated by an approximate method[15,19] is 271,000 Btu/(lb mole) of benzene hexachloride. Thus, (271,000) ((3120)/(24)(291)) = 121,000 Btu/hr must be removed from the reaction mixture.

A five-inch OD steel pipe located concentric to the three-inch reactor pipe would make an effective heat-transfer jacket. The cooling medium would pass through the annuli between these pipes.

4. Description of the Proposed Plant Process

The flowsheet of the proposed process is to be found in Fig. 16. The liquid benzene and chlorine pass from the storage to a mixer, and then in the liquid phase at 15 atmospheres pressure through the nickel reactor in the radiation chamber. A plan view of this chamber is to be found in Fig. 17. Ten passes are necessary at the dose rate of 105 kilorep per hour.

From the reactor in the radiation cave the slurry of benzene hexachloride in benzene passes through a chamber where the pressure is

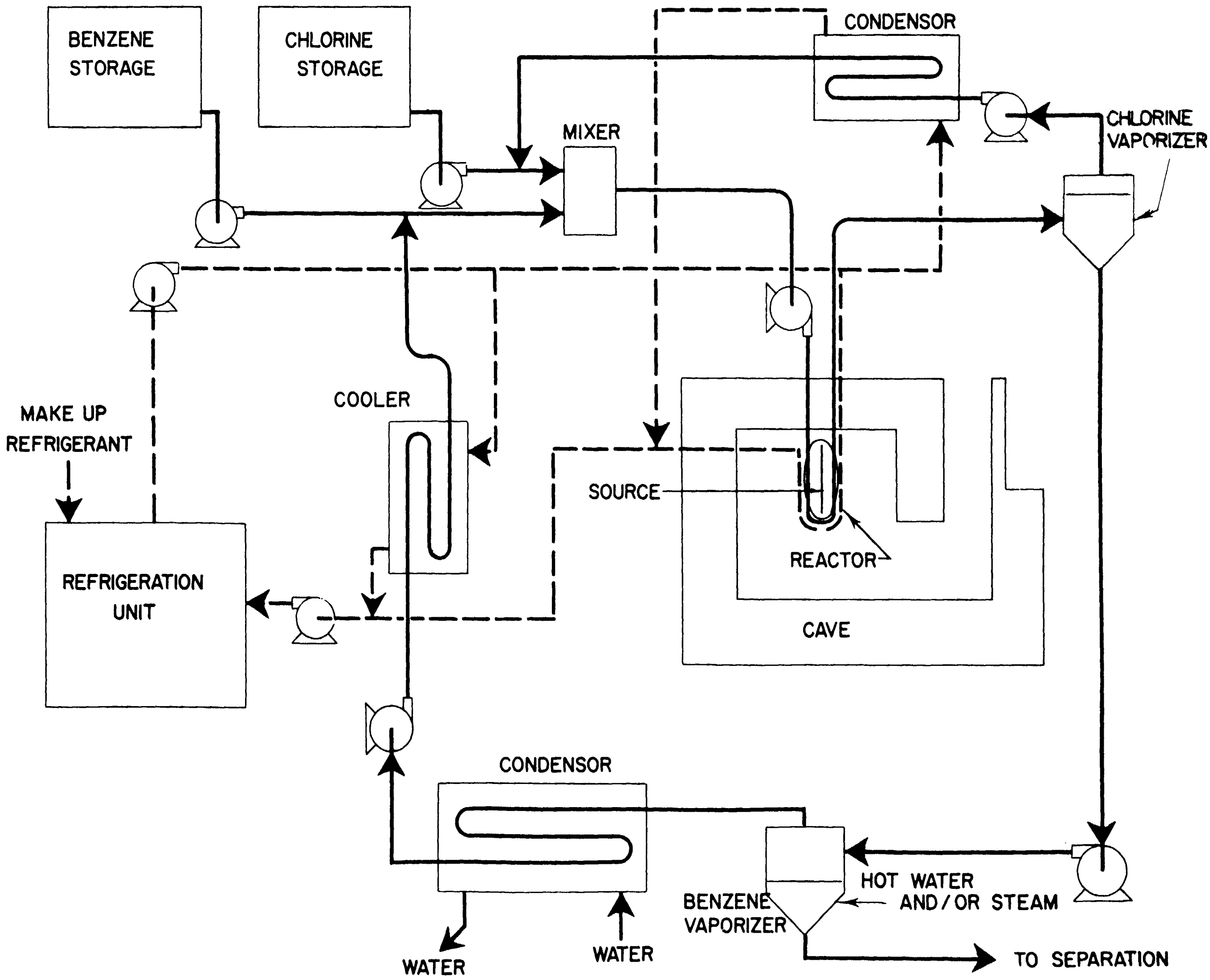


Fig. 16. Flowsheet of Proposed Plant Process for the Addition of Chlorine to Benzene under the Influence of Gamma Radiation.

reduced to 5 atmospheres. This will vaporize any remaining chlorine which may then be compressed and recycled back into the system. The benzene hexachloride stream from this chlorine vaporizer is pumped through a benzene vaporizer where it is sprayed on hot water or into steam. The pressure is reduced from 5 to 1 atmosphere. The excess benzene is vaporized and passed through a condenser and then recycled at 15 atmospheres into the reactor.

The vaporizing benzene leaves behind the benzene hexachloride in water which can be either dried or purified. The gamma isomer may be separated from the mixed isomers and sold as pure Lindane or as dusting powder in the unpurified form. This separation into the pure isomers is not of interest in this plant design. Methods of analysis and separation are reported in the literature.[5,20]

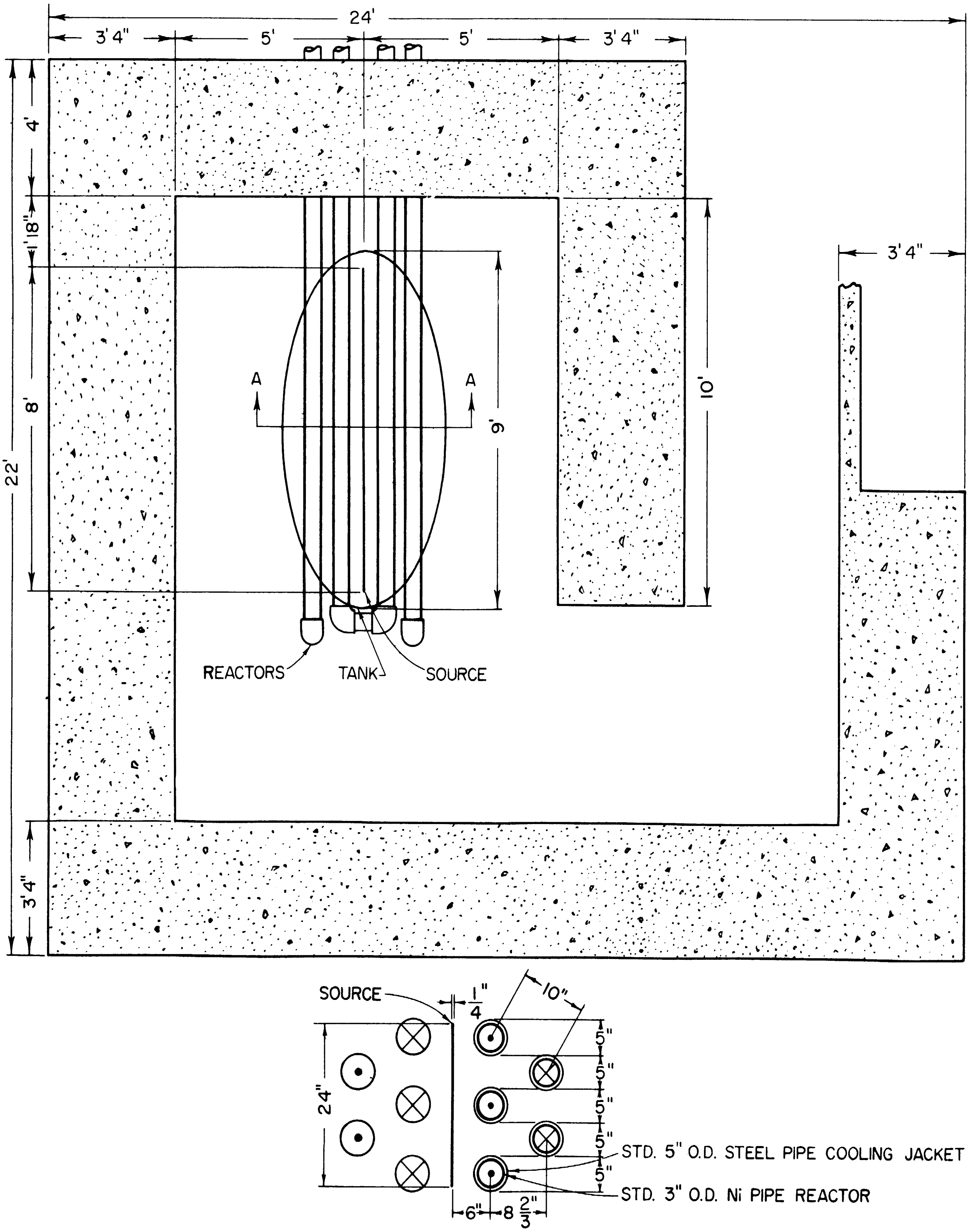


Fig. 17. Plan View of Proposed Reaction Cave Showing Reactors in Position.

Since the operating temperature was selected as 85°F for several reasons listed above, it is necessary to use a system of refrigeration to effect heat removal from the process. The cooling in the reactor, chlorine condenser, and benzene cooler must be accomplished by this refrigeration. This amounts to a removal of

Reactor		121,000 Btu/hr
Chlorine condensor	$\frac{(.07)(\text{lb moles})}{\text{hr}} (8800) \frac{\text{Btu}}{\text{lb mole}} =$	620 Btu/hr
Benzene cooler	$(1.05) \frac{(882)}{24} \frac{\text{lb}}{\text{hr}} (.46) \frac{\text{Btu}}{\text{lb °F}} (70)\ °\text{F} =$	1,250 Btu/hr
		122,870 Btu/hr

or approximately 123,000 Btu/hr from the three sources. The benzene condenser may have water as the cooling medium as it involves heat transfer at higher temperatures. At a higher operating temperature this refrigeration unit could be replaced by water as the cooling medium.

5. Calculation of Gamma Radiation Source Strength and Shielding

Two possible arrangements of the reactor tubes in relation to the source are considered. The gamma radiation source may be located inside the reactor or concentric to an annular flow reactor, or the source may be a finite rectangular plaque with the reactor located on either side.

This latter arrangement has been assumed for these calculations. The reactor size has been calculated to be ten passes each ten feet long. In order to utilize a rectangular radiation source most efficiently, the arrangement of reactor tubes is as pictured in cutaway AA of the plan view of Fig. 17. Five reactor tubes are placed on either side of the source with the first row six inches from the source. The second row is placed on an equilateral triangle pitch behind this first row.

Assuming a dose rate in the first row of tubes equal to that used in the laboratory work and correcting for any absorbing media between the source and this point the rate of gamma emmission on the surface of the source may be calculated. Gomberg, et al. [3,13] and Brownell, et al.[4] have done this. Using the equation developed by Lewis[21] for the intensity of radiation through an absorption medium which in turn utilizes the adsorption coefficients (μ) of Snyder and Powell,[33] the strength of the source is then calculated to be

$$I = I_o \, e^{-\sum \mu \rho x}$$

$$105 = I_o \, e^{-2.54 \frac{cm}{in.}\left[(.08)\frac{cm^2}{gm}(1.0)\frac{gm}{cm^3}(2.25)in. + (.075)\frac{cm^2}{gm}(8.38)\frac{gm}{cm^3}(.474)in.\right]}$$

where

$$\mu_{H_2O \text{ and } C_6H_6} = .08 \frac{cm^2}{gm} \; ; \qquad \mu_{Fe \text{ and } Ni} = .075 \frac{cm^2}{gm}$$

$$105 = I_o \, e^{-1.215}$$

$$I_o = (3.37)(105) \frac{krep}{hr} = 354 \frac{krep}{hr}$$

I = radiation intensity, krep/hr
μ = absorption coefficients, (cm^2/gm)
x = thickness absorber, (cm)
ρ = density of absorber, (gm/cm^3).

The specific activity of the gamma radiation source is assumed to be 3.8×10^3 (rep)/(hr)(curie).[3,13] A rectangular source plaque two feet by eight feet is assumed for the commercial design as compared to 16 cm^2 used in the laboratory experiment. Then the approximate total activity of the source plaque is

$$\frac{354\frac{(krep)}{(hr)} \times 1000 \frac{(rep)}{(krep)} \; 2(ft) \; 8(ft) \; 144\frac{(in.^2)}{(ft^2)} \; 6.45\frac{(cm^2)}{(in.^2)}}{3.8 \times 1000 \frac{(krep)}{(hr)(curie)} \qquad (16)(cm^2)} = 86{,}500 \text{ curies.}$$

The source-room size is selected and the concrete shielding necessary to protect the human operators can be calculated. Assuming that the dose rate in air varies immensely with the square of the distance from the source, the dose rate at a distance of five feet from the source would be

$$I = \left(\frac{x_o}{x}\right)^2 I_o$$

$$I = \left(\frac{1}{60}\right)^2 354{,}000 \frac{rep}{hr}$$

$$I = 985 \frac{rep}{hr} .$$

Assuming a .05 mr/hr to be a safe dose rate for the human operators,

$$I = I_o e^{-\mu x}$$

$$(.05) \frac{mr}{hr} = 985 \frac{rep}{hr} \times 1000 \frac{mr}{rep} e^{-.2x}$$

where

$$\mu = 0.2 \text{ cm}^{-1} \text{ for concrete}$$

$$x = \text{thickness of concrete in cm}$$

$$e^{.2x} = \frac{985{,}000}{.05} = 1.97\text{x}10^7$$

$$.2x = \ln 1.97\text{x}10^7$$

$$x = \frac{(7.294)(2.303)}{(.2)} = 84 \text{ cm}$$

$$x = 33 \text{ in.}$$

Assuming .05 mr/hr to be a safe dosage for these calculations, based on narrow-beam coefficients, a wall thickness of forty inches or three feet four inches is selected. (Based on broad-beam coefficients and forty inches of concrete, the radiation level is computed to be increased to about one mr/hr.) A well of twelve feet of water will permit lowering the source to a sufficient depth to provide adequate shielding. A plan view of the radiation chamber is shown in Fig. 17. A cutaway perspective view of this chamber showing the reactor in place is shown in Fig. 18. The reactor-end supports and the source elevator have been omitted from the figure to clarify the picture.

6. Estimation of Cost for the Proposed Equipment and Radiation Chamber

An itemized cost summary of the proposed radiation chamber is given in Table 8. The cost estimation for all equipment used in the process of manufacturing benzene hexachloride by the proposed method utilizing gamma radiation is to be found in Table 9. These estimates exclude the cost of the radiation source itself or of the separation unit needed to purify and/or separate the isomers.

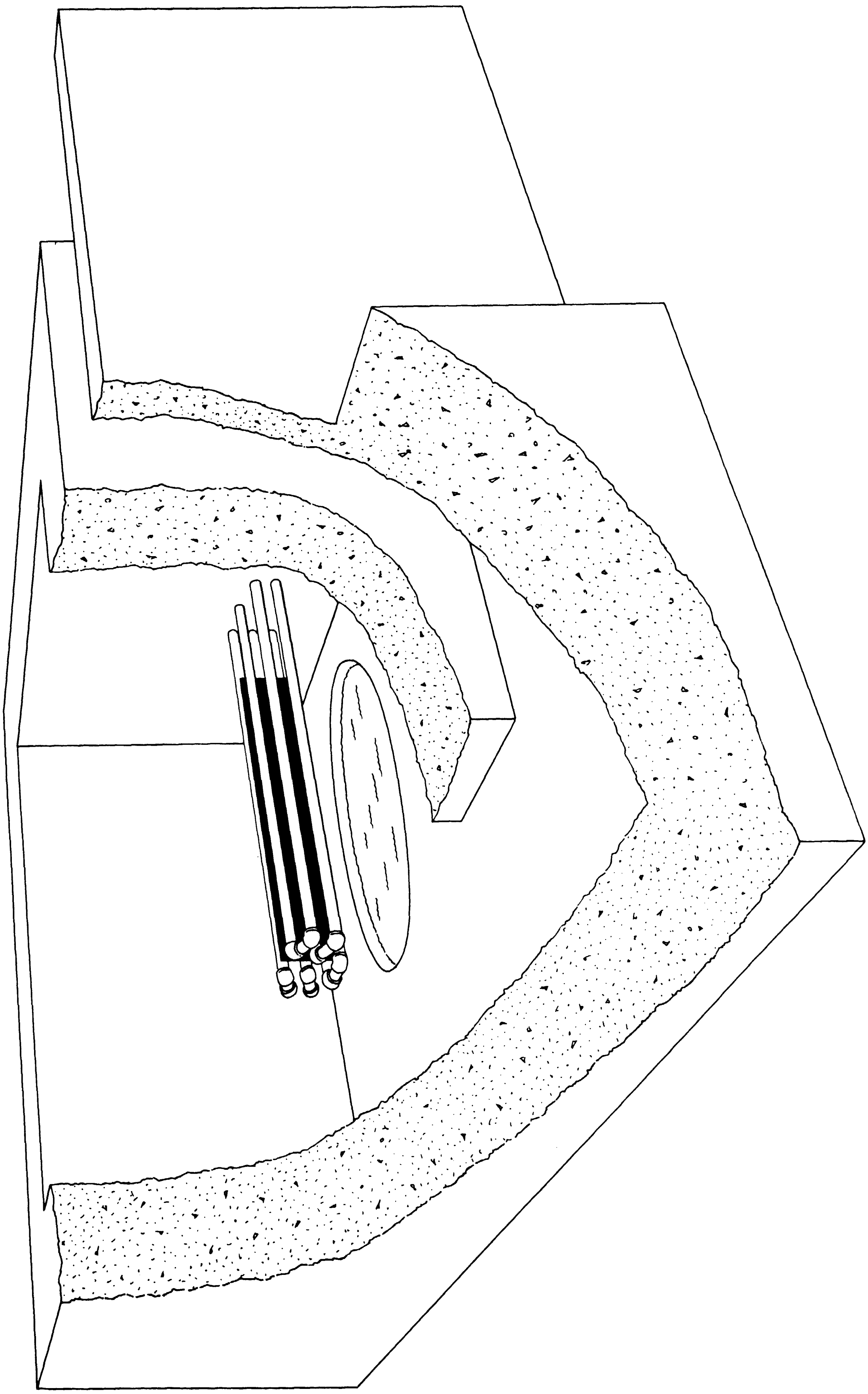


Fig. 18. Cutaway Drawing of Reactor Cave Showing Gamma Radiation Source and Reactor Placement.

TABLE 8

COST ESTIMATES FOR RADIATION CHAMBER

Concrete for walls, roof, floor, and well 130 yd at $20.00/yd	$ 2600
Reinforcing	400
Excavation for footings and well	800
Forms for concrete	800
Labor	1800
Stainless-steel well	1500
Elevator for source	1500
Ion-exchange equipment	2000
Monitoring equipment	1800
Painting, wiring, plumbing, and ventilation for cave including labor	3200
	$16,400
Miscellaneous contingencies (20%)	3300
Engineering (5%)	800
Contractors fee (10%)	1600
Total	$22,100

TABLE 9

COST ESTIMATES FOR EQUIPMENT EXCLUDING ISOMER SEPARATION

Nickel or stainless-steel reactor pipe and fittings into cave	$ 900
Heat-exchange jacket	400
Pumps and pipe from pumps	1900
Two-flash vaporizers	3000
Chlorine compressor and connections	8000
Heat exchanger for benzene	3000
Refrigeration unit	18000
Benzene and chlorine storage	4000
Total	$39,200
Installation (30% of cost)	11800
Building for housing pumps, control equipment, etc.	9000
Total	$60,000

7. Comparison of Gamma Radiation Sources to Give Constant Source Strength

The source plaque as described in calculating the total gamma-radiation strength could be one solid sheet of radioactive material or it could be composed of a number of individual sections. This would be essentially a rack which held the source made of several sections. These sections could be in the form of strips or cannisters and could be removed and/or replaced as it became necessary. This replacement time would depend on the permissable flux variation of the source and the half life of the material used for the source. For the following calculations, it is assumed that a flux variation of 20 percent will not affect the rate of reaction of benzene and chlorine appreciably. The time (t) necessary for a radioactive material of original intensity (I_o) to decay to an intensity (I) is

$$I = I_o e^{-kt}$$

$$t = \frac{1}{k} \ln \frac{I_o}{I} ,$$

where k is a constant of the material. Letting the material decay to half its original value, then

$$I = \frac{I_o}{2}$$

$$t_{.5} = \frac{1}{k} \ln 2 = \frac{.693}{k}$$

or

$$k = \frac{.693}{t_{.5}} ,$$

where $t_{.5}$ is the defined half life of the material. Then for any other time of decay

$$I = I_o e^{-\left(\frac{.693}{t_{.5}}\right) t} .$$

Letting $I = .80\ I_o$, then

$$.80 = e^{-\left(\frac{.693}{t_{.5}}\right) t_{.80}} ,$$

or

$$\frac{(.693)t_{.80}}{t_{.5}} = \ln \frac{1}{.80} = \ln 1.25 = .223$$

$$t_{.80} = \frac{.223}{.693} t_{.5} = .322\, t_{.5} .$$

This equation is the relation of the time for any material of half-life $t_{.5}$ to decay to 80 percent of its original value of intensity. Both $t_{.80}$ and $t_{.5}$ must be in the same units.

Assuming a finite number of sections of the source, then at the end of one .80-life, a quantity of gamma radiation equal to 20 percent of the rated source strength must be added to the source. If this means replacement of any section of the source, then the strength of the replaced section must also be present in the replacement section to keep the rated radiation value constant.

For example, assume that the rectangular plaque consists of twenty individual sections. At the beginning of any .80-life period, the plaque will be at the necessary rated strength calculated previously. The ratio of the section strength to the rated section strength will be $(.8)^n$, where n is the number of .8-lives since the section was replaced. Assuming one replacement per .8-life, the source will consist of twenty sections with strengths

Section	1	2	3	4	. . . .	19	20
	1	$(.8)^1$	$(.8)^2$	$(.8)^3$	. . . .	$(.8)^{18}$	$(.8)^{19}$.

At the end of any .8-life each section will have decayed by a .8 amount and the source will consist of twenty sections of strengths

Section	1	2	3	4	. . . .	19	20
	$(.8)^1$	$(.8)^2$	$(.8)^3$	$(.8)^4$	. . . .	$(.8)^{19}$	$(.8)^{20}$.

This shows that the addition of one section of rated section strength must replace a section of $(.8)^{20}$ of rated section strength. The replacement section strength S must then be equal to

$$\left[1.000 - (.8)^{20}\right] S = (86{,}500)(.20) \text{ curies,}$$

where 86,500 curies is the total rated source strength; then

$$\left[1.000 - .0115\right] S = 17{,}300 \text{ curies}$$

$$S = \frac{17{,}300}{.988} = 17{,}500 \, \frac{\text{curies}}{\text{gamma section}} \quad .$$

Table 10 is a comparison of four different gamma radiation sources of known half life. Three gross fission products, six-months old, one-year old, and two-years old, are compared with cesium-137. The approximate half lives of the fission products were taken from the literature.[38]

TABLE 10

COMPARISON OF FOUR GAMMA RADIATION SOURCES
TO GIVE CONSTANT SOURCE STRENGTH FOR FIVE YEARS

Fission-Product Material	Half life $t_{.5}$ yr	$t_{.8}$ yr	γ Sections to Start	γ Section Replacements in 5 yr	Total γ Sections in 5 yr	Total Radiation Necessary for 5 yr, curies
Gross 6-mo old	.20*	.064	5	78.1	83.1	1,455,000
Gross 1-yr old	.30*	.097	5	51.5	56.5	990,000
Gross 2-yr old	1.10*	.354	5	14.1	19.1	334,000
Pure Cs-137	33.	10.6	5	0	5	86,500

*Approximate half life for mixtures of elements of varying half life.

The calculations for the six-month-old fission-product material are illustrated here. The others follow similarly.

$$t_{.8} = .322 \, t_{.5} = (.322)(.20) \text{ year}$$

$$t_{.8} = .064 \text{ year} \quad .$$

The number of gamma radiation sections to start is five. The number of gamma replacements in five years is

$$\text{Replacements} = \frac{5.00}{t_{.80}} = \frac{5.00}{.064}$$

$$= 78.1$$

Then the total gamma sections necessary for five-years operation are

$$\text{Starting sections + Replacement sections} = \text{Total sections}$$

$$\text{Total sections} = 5 + 78.1 = 83.1 \quad .$$

Then the total rated radiation in curies necessary for five years operation is

$$(\text{Total sections})\left(\frac{\text{curies}}{\text{gamma section}}\right) = \text{Total radiation}$$

$$\text{Total radiation} = (83.1)(17{,}500) = 1{,}455{,}000 \text{ curies} \quad .$$

In the case of cesium-137 it is seen that the half life is long enough that no replacement sections are needed. The original five sections have not decayed to the permissable .80 level of radiation.

8. Cost Comparison of Benzene Hexachloride from Four Gamma Radiation Sources and from Present-Day Process

Little is known about the cost of either gross or concentrated fission products. There are some figures available for pure radioactive isotopes for specific purposes. A range of values expressed in dollars per curie is assumed in Table 11 for the four gamma radiation sources considered. The range is wide enough to take into consideration use of the fission product at the nuclear reactor site where no shipping is necessary and large quantities of hot fission products are presently in storage. These costs have been assumed on the basis of approximate equal cost per unit of energy of radiation delivered. Since the energy, E, delivered by a source is

TABLE 11

COMPARISON OF TOTAL PLANT COST FOR FOUR
GAMMA RADIATION SOURCES

Fission-Product Material	Range of Estimated Cost for Radiation, Dollars per Curie		Total Radiation Cost, Dollars		Total Plant Cost, Dollars	
	Low	High	Low	High	Low	High
Gross 6-mo old	.0005	.05	728	72,800	82,900	154,900
Gross 1-yr old	.001	.05	990	49,500	83,100	131,600
Gross 2-yr old	.02	.25	6,680	83,500	88,800	165,600
Pure Cs-137	.50	5.00	43,250	432,500	125,400	514,600

$$E = \int_0^t I_o \, e^{-\lambda t} = I_o \, (1-e^{-\lambda t})/\lambda \quad ,$$

it follows that the energy varies inversely as λ for large values of t. Since λ is the reciprocal of the half life, it follows that the energy delivered varies directly as the half life. Thus, a source with three times the half life of another can deliver three times as much radiation energy and is assumed to cost about three times as much.

The cost of long half-life sources is estimated to include packaging, shipping, and installation in a plant at some distance from where the source is prepared. The cost of very short half-life sources would be excessively high because of the large shipping cost. Therefore, it is assumed that the short half-life sources are to be used in the vicinity where they are produced. In this way it is believed that the cost per curie of the short half-life material (which might even be spent fuel elements) although low, is not unrealistic.

Table 11 is a comparison of the total plant cost including the radiation source for the radiation sources considered. The values and the method of calculation are presented for six-month-old fission-product material for the low cost estimated for the radiation.

(No. curies)(cost/curie) = Total cost for radiation

(1,455,000 curies)(.0005 $/curie) = $728

The total radiation cost plus the cost for equipment and for the radiation cave found in Tables 8 and 9 is the total plant cost.

Total cost for radiation	=	$ 728
Cost for radiation chamber	=	22,100
Cost for equipment	=	60,000
		$82,828 or $82,900

Table 12 is self explanatory. Carrying through the calculations for the low range value of six-month-old fission products and the annual cost of the proposed plant, amortizing the plant investment over a period of five years, and assuming 10 percent interest on the total plant, the investment amounts to

Annual plant cost, 82,900/5	$16,560
Interest, (.10)(82,900)	8,290
Working Capital	20,000
Operation and maintanence	30,000
Total Annual cost of plant operation =	$74,850 per year

Assuming operation and production during 290 days of the year, the daily cost of the operating plant would be

$$\frac{\$74,850}{290} = \$258 \text{ per day.}$$

The cost of raw material is[7,8,26]

Benzene at $.42 per gal	=	$50.50
Chlorine at $3.50 per 100 lb	=	84.50

or a total operating cost of $393 per day.

The cost of producing 3120 lb of mixed isomer is then

$$\frac{\$393 \text{ per day}}{3120 \text{ lb per day}} = \$.126 \text{ per lb mixed isomers.}$$

TABLE 12

COMPARISON OF TOTAL PRODUCTION PRICE OF THE GAMMA ISOMER OF BENZENE HEXACHLORIDE PRODUCED FROM FOUR GAMMA RADIATION SOURCES

Fission-Product Material, Dollars	Gross 6-mo old		Gross 1-yr old		Gross 2-yr old		Pure Cs-137	
	Low	High	Low	High	Low	High	Low	High
Annual plant cost on 5-yr amortization	16,560	31,000	16,610	26,300	17,720	33,100	25,100	103,000
Annual interest to stockholders, 10% on total plant cost	8,290	15,490	8,310	13,600	8,880	16,560	12,500	51,460
Working capital	20,000	20,000	20,000	20,000	20,000	20,000	20,000	20,000
Operation and maintenance of plant	30,000	30,000	30,000	30,000	30,000	30,000	30,000	30,000
Total annual cost, excluding raw materials	74,850	96,490	74,920	89,900	76,600	99,660	87,600	204,460
Daily cost assuming 290 operating days per year	258	333	258	310	264	344	302	705
Total daily operating cost, including raw materials	393	468	393	445	399	479	437	840
$/($\gamma$ unit)(lb isomers) on 32% γ isomer yield	.00393	.00468	.00393	.00445	.00399	.00479	.00437	.00840
$/($\gamma$ unit)(lb isomers), assuming 20% γ isomer yield	.0063	.0075	.0063	.00714	.0064	.00768	.0070	.01345

Defining the gamma unit as percent of gamma isomer per pound of mixed isomers, the cost per gamma unit assuming 32% gamma isomer produced is

$$\frac{.126}{32} = \$.00393 \text{ per gamma unit per pound.}$$

If the recovery was only 20 percent gamma isomer in the reaction of mixed isomers, as is true in many commercial ultraviolet units today, the production price for the same production of mixed isomers would be

$$\frac{.126}{20} = \$.0063 \text{ per gamma unit per pound .}$$

Benzene hexachloride is being sold at present market conditions[8,26] at \$.009 to \$.015 per gamma unit per pound in concentrations of 12 to 14% gamma isomer per pound. Using this lower figure for comparison, a savings of 50 to 225 percent can be realized by producing benzene hexachloride by gamma radiation activation as against conventional ultraviolet light activation. This is true for the whole range of costs of all the gross mixed fission products. Only the highest value estimated for cesium-137 falls above \$.009 per gamma unit per pound. This is, however, in the range of the price being asked today.

The separation cost of the isomers would be comparable to that method used today and has not been calculated. These calculations, however, reveal a more efficient means of obtaining the crude isomer mixture.

9. Conclusions and Summary

Several important improvements in existing plant operations have been overlooked in presenting this economic study. Some of the more important improvements present in this proposed plant design are

(1) Elimination of necessarily small costly equipment due to the necessity for ultraviolet radiation penetration. Gamma radiation will penetrate to the very center of the reaction mixture due to its very nature and wavelength.

(2) Elimination of glass or quartz equipment necessary to transmit ultraviolet radiation. Steel equipment may be used with gamma radiation.

(3) Availability of higher temperatures and pressures in carrying out the reaction for the reason listed in (2).

(4) The full capabilities in the use of gamma radiation have not been fully realized in this study due to the nature of the data used. There are several important regions where the use of gamma radiation could be investigated more fully. Enumerated they are:

(a) Higher rates of radiation should be investigated. The data presented are for a constant rate of radiation. The possibility of utilizing larger radiation sources to give larger radiation fluxes and increased rate of reaction should be studied.

(b) Running a larger chlorine recycle stream and/or running to a less complete reaction based on the chlorine would undoubtedly increase the overall rate of reaction. The rate of reaction is faster at higher concentrations of chlorine and running to 95 percent completion on the chlorine decreases the overall rate of reaction.

(c) Running at even higher pressure than presented here would allow running a higher temperature with the chlorine still in the liquid phase. The data available, although dependable and consistent, cover too small a temperature range to allow long extrapolation. At a higher temperature the reaction rate would be greater.

(d) For simplicity of design only one effective absorbing bank of tubes has been used (see Fig. 3, Sec. AA). Other rows of tubes could be placed behind the tubes shown to absorb radiation not used in this design. Brownell, et al.[3,4] show an increase in efficiency of 500 percent in using a multipass package plant as against a single pass dead carcass plant for irradiation of meat. By increasing the capacity of the plant and putting more reactor bundles in the radiation chamber, the cost of crude benzene hexachloride would be reduced. Only equipment costs would increase in the cost estimate. The major expense of the radiation source would be the same.

(e) An annular reactor could be used with the source of gamma radiation in the center. This would presumably utilize a higher percentage of the radiation emitted and decrease the rated strength necessary in the source.

(f) The plant cost was written off in five years, but it must be realized that in all cases there is gamma activity left at the end of five years. Less than 10 percent of the radiation of Cs-137 has been used at the end of this five-year write-off period.

For these reasons it is thought that the value of gamma radiation on the reaction has been underestimated. A more complete investigation would undoubtedly show a greater difference in the cost of benzene hexachloride as it is produced today by ultraviolet radiation and benzene hexachloride produced utilizing gamma radiation as presented here.

F. WORK CONTEMPLATED FOR THE NEAR FUTURE

Present plans are to make a few more runs on the polymerization of ethylene with gamma radiation to supplement the data already taken. Following that it is probable that no more experimental work will be undertaken on this reaction. However, a preliminary plant design, such as has been given for the chlorination of benzene, will probably be made to obtain an estimate of the cost of polyethylene produced by gamma radiation for purposes of comparison with the cost of polyethylene produced by the conventional high pressure process employed industrially today.

A few additional runs will be made on the rate of reaction of chlorine with toluene under gamma radiation utilizing the best technique for analysis of the off-gases of chlorine and hydrogen chloride. The proof that the addition compound is the principal product is now complete.

Chlorination of other compounds mentioned in previous reports will probably receive very high priority in the experimental work. The aromatics closely related to benzene and toluene, such as ethyl benzene, the xylenes, and naphthalene, will be chlorinated to determine whether gamma radiation favors the addition of chlorine to the double bonds. It is also expected to chlorinate some simple aliphatic alcohols, aldehydes, and acids. Some preliminary work along this line has shown that under gamma radiation chlorine probably converts a hydrated aldehydic group to an acidic group, a reaction which apparently does not occur when ultraviolet light is employed.

Some other high pressure polymerizations are scheduled to be tried soon. Also it is hoped to carry out a copolymerization under gamma radiation, with the combination of butadiene and styrene at low temperatures; this reaction will probably be tried first.

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PART III. SUBPROJECT M943-5, GAMMA IRRADIATION OF FOOD

A. ORGANOLEPTIC STUDIES

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1. INTRODUCTION

The problem of flavor in irradiated food was reviewed in Progress Report 6; also the design of experiments for flavor evaluation and the use of statistics in experimental flavor studies were described. In all tests either the "triangle" or "incomplete block ranking" procedure was used to evaluate taste panel results. Studies reported on fresh fruits included the use of "pasteurization" and "sterilization" doses of gamma radiation on dark sweet cherries, fresh peaches, plums, cantaloupe, apples, applesauce, and navel oranges. Brief studies of gamma-ray treatment of canned pork and canned beef were also reported. Studies of the refrigerator shelf-life of gamma-ray pasteurized raw ground pork, raw ground beef, chicken legs and changes in flavor during storage were made using the taste panel and the results have been described in Progress Report 6.

A continuation of the studies on ground raw pork and beef and navel oranges using incomplete block ranking tests to evaluate taste panel results are described in this report (Progress Report 7). Other experiments reported here include the use of low-dosage gamma irradiation to prevent sprouting of potatoes and onions with subsequent storage at 50°F and approximately 50 percent relative humidity. Packaging materials used for storage studies in these experiments were polyethylene, Kraft paper bags, burlap bags, and mesh bags. Taste panels were conducted periodically throughout the storage period. The shelf-life of gamma-ray pasteurized fresh asparagus, coleslaw, endive, and cabbage were observed and flavor changes recorded. Baking characteristics, eating qualities, and flavor changes of irradiated cake, all-purpose, and bread flour were observed when the flour was incorporated in cake, baking powder biscuits, and other bread formulas.

2. RESULTS OF THE TASTE PANEL EXPERIMENTS

a. Tests with Meat

(1) Ground Raw Pork: Ranking by Means of Paired Comparisons. In Progress Report 6 limited test results reported indicated that ground raw pork given a gamma-ray "pasteurization" dose between 60,000 and 80,000 rep would have a refrigerator (40°F) storage life of 10 days and possibly longer. Since the experimental material was exhausted after 10 days in the first study, a continuation of the storage life of gamma-ray pasteurized ground pork was observed in the experiment reported here.

Relatively lean ground loin of pork of the same quality as used in the previous experiments was obtained from the butcher shop of Food Service, University of Michigan. The meat was divided into four portions. One portion was stored in the frozen state (-15°F) to be used as a control. The other three portions were placed in closed glass containers and given the following dosages of irradiation: 50,000, 70,000, and 100,000 rep. Immediately after irradiation these samples were placed in the refrigerator for storage.

Taste panels were held after 9, 12, and 13 days of refrigerator storage. The meat was prepared following the same procedure as used in the experiments reported in Progress Report 6. Table 13 shows the statistical analysis of this extended shelf-life storage experiment of ground raw pork.

After 9 days storage at 40°F it was found that the sample given 100,000 rep had a slight off-flavor which some judges thought was rancidity; not characteristic of the "irradiation flavor" of irradiated lean meat. The samples given 50,000 and 70,000 rep were still acceptable to the taste panel

TABLE 13

RANKING OF PORK BY MEANS OF PAIRED COMPARISONS

Sitting No.	Storage Time, days	Dose and Total Scores for Each Dose*				P**	Significant Difference ?
		0	$5x10^4$	$7x10^4$	$1x10^5$		
1	9	24	25	19	22	.27	No
2	9	25	24	25	34	.01	Yes
		.30	.36	.30	.04		
3	12	17	22	26	25	.02	Yes
		.68	.17	.07	.08		
4	12	25	26	27	30	.58	No
5	13	21	32	25	30	-	Yes
		.61	.06	.25	.09		
6	13	25	21	26	36	-	Yes
		.22	.60	.18	-		

*Where significant differences exist estimates of the preference ratings are displayed below the total scores.

** - indicates less than 0.01.

at this time. After 12 days of storage the samples given 100,000 rep were definitely rancid and those given 50,000 and 70,000 rep had a very bad odor which did not disappear on cooking. The odors were considered to be more objectionable than the flavors. After 13 days of storage a greater preference for the control meat was shown. Based on these tests the maximum refrigerator storage life of raw ground pork given a radiation dose of between 60,000 and 80,000 rep is considered to be about 10 days. Greater radiation dosages probably cannot be used to further extend the refrigerator storage life of raw ground pork because of development of rancidity in the fats on storage.

(2) Ground Raw Beef: Ranking by Means of Paired Comparisons. Since it was found impossible to obtain beef of known age and without exposure to ultraviolet light from the Food Service of the University of

Michigan, it was decided to try obtaining beef directly from a small, local slaughterhouse. However, this decision resulted in some complications. After the first taste panel test it was the opinion of the judges that the beef was of poor quality and evidently obtained from an old animal as the meat was tough, contained much gristle, and had a poor flavor. Also, all the meat was observed to have a smoky odor and flavor. On checking with the slaughterhouse, it was discovered that after grinding the meat had been stored overnight in the same cold room with other meat. After the meat had been stored for 5 days in the refrigerator it was noticed that the beef lost some of this smoky odor and taste and tasted more like typical hamburger. The meat was ground round containing two ounces of fat per pound. As soon as this meat was obtained from the local slaughterhouse, it was divided into five portions. Two portions were set aside as controls; one of which was stored in the frozen condition for use after spoilage of the other control which was stored in the refrigerator (40°F). The three remaining portions of ground beef were given 50,000, 80,000, and 110,000 rep of gamma radiation, respectively. Enough beef was ordered for observation through 14 days of storage.

The results of this storage experiment are shown in Table 14. The control samples used for the taste panels held after 4 days of storage were those stored in the refrigerator and as these controls had spoiled at this time a preference was shown for the irradiated samples. The frozen control samples were used for the taste panels held after 5-days storage. At this time the irradiated beef given 80,000 and 110,000 rep seemed to have a slightly better flavor than the nonirradiated beef, but no preference was shown for the meat given 50,000 rep over the nonirradiated samples. After 7 and 8 days of storage the irradiated beef was still in very good condition, although the sample given 50,000 rep was not as well liked by the taste panel as the control. The last taste panel tests were held after 11 days of storage. At this time the judges were not able to make a preference, because neither the controls nor the irradiated samples had a good flavor. The smoky odor and flavor that was in the beef from the beginning of the experiment made it very difficult for the judges to make a decision as to the odor and flavor changes that had taken place in the beef in storage. These tests indicate that gamma-ray pasteurized raw ground beef can be stored at 40°F for 8 days without spoilage and very possibly for 10 days without noticeable flavor and odor changes. The color changes observed in the irradiated raw ground beef samples throughout the storage period were similar to those reported in Progress Report 6. Again, the stored beef given 1.1×10^5 rep retained more of its original red color than the other two portions given 5×10^4 and 8×10^4 rep.

More tests should be performed on gamma-ray pasteurized beef and arrangements should be made with a reliable meat packing house in order to obtain beef of prime quality.

TABLE 14

RANKING OF BEEF BY MEANS OF PAIRED COMPARISONS

Sitting No.	Storage Time, days	Dose and Total Scores for Each Dose* 0	$5x10^4$	$8x10^4$	$1.1x10^5$	P**	Significant Difference ?
1	1	25	21	22	22	.69	No
2	1	24	32	28	24	.06	?
3	4	30	20	21	19	-	Yes
		-	.32	.25	.43		
4	4	29	30	26	23	.19	No
***5	5	25	23	22	20	.49	No
6	5	29	29	28	22	.14	No
7	7	22	30	30	26	.06	?
8	7	28	32	24	24	.06	?
9	8	23	26	19	22	.20	No
10	8	22	24	23	21	.89	No

*Where significant differences exist estimates of the preference ratings are displayed below the total scores.

**- indicates less than 0.01.

***At this point fresh frozen control was substituted for the refrigerated control which was definitely spoiled.

b. Tests with Fresh Fruits

(1) Navel Oranges: Ranking by Means of Paired Comparisons. As mentioned in Progress Report 6, bitterness is produced in the juice extracted from early season navel oranges of root stock susceptible to bitterness if the juice is allowed to stand for about 1 hour. Juice from similar midseason fruit may require storage overnight before bitterness is detectable. Since the juice was not allowed to stand before being used in the previous taste panel experiments described in Progress Report 6, additional exploratory tests were made.

Whole oranges (Sunkist, Navel 150's) were given the following doses of irradiation: $3x10^4$, $6x10^4$, and $1.0x10^5$ rep. Immediately after irradiation the oranges were squeezed by hand using a Foley juicer. The juice from seven oranges was well mixed for each sample and two samples were used for each dose and for the nonirradiated samples. All samples of juice were then permitted to stand overnight at 40°F.

Two taste panels were held the following day. It was found that the juice from oranges given the lowest dose, $3x10^4$ rep, was less bitter than the juice from the nonirradiated oranges. The juice from oranges given $6x10^4$ and $1.0x10^5$ rep was more bitter than the juice from the nonirradiated oranges. Also the juice from oranges given the two highest doses had a typical irradiation odor which was described as an unpleasant cabbage-like taste and odor. Results of this test are shown in Table 15.

TABLE 15

RANKING OF ORANGES BY MEANS OF PAIRED COMPARISONS

First Test

Sitting No.	Storage Time, days	Dose and Total Scores for Each Dose*				P**	Significant Difference ?
		0	$3x10^4$	$6x10^4$	$1x10^5$		
1	1	25	27	25	31	.29	No
2	1	21 .58	23 .34	32 .04	32 .04	-	Yes

* Where significant differences exist estimates of the preference ratings are displayed below the total scores.

** - indicates less than 0.01.

More Navel Sunkist 150's were obtained for a second experiment. The juice from nine oranges was extracted for each sample and again two samples were used for each treatment and for nonirradiated samples. The samples of juice were held in sealed Mason jars and were given the following doses of irradiation: $3x10^4$, $6x10^4$, and $1.0x10^5$ rep. Again, all samples were stored overnight followed by two taste panel tests the next day. Table 16 gives the statistical analysis of the taste panel results. It was the opinion of the judges that there was no preference for either the irradiated or nonirradiated juice as both were acid and bitter.

Although the tests on the navel oranges and juice are inconclusive, the limited data indicate that a dose of irradiation of about 30,000 rep applied to whole oranges may aid in preventing the development of bitterness

TABLE 16

RANKING OF ORANGES BY MEANS OF PAIRED COMPARISONS

Second Test

Sitting No.	Storage Time, days	Dose and Total Scores for Each Dose				P	Significant Difference ?
		0	$3x10^4$	$6x10^4$	$1x10^5$		
1	1	25	20	22	23	.49	No
2	1	26	28	23	31	.15	No

on standing; whereas the irradiation of the juice rather than the whole orange indicated no improvement. The tests on whole oranges should be repeated and expanded to fully evaluate the possible merit of such an application of gamma radiation.

c. Tests with Vegetables

(1) Potatoes: Ranking by Means of Paired Comparisons. A. H. Sparrow and E. Christensen[1] found that low doses of gamma radiation will keep potatoes from sprouting over a 10-month storage period. At 10 months they reported that the nonirradiated potatoes were essentially spoiled, whereas those that had been exposed to 20,000 rep were still Grade A potatoes. Potatoes given 50,000 and 80,000 rep were Grade B potatoes when 10 months old. Taste tests showed acceptable flavor in all but the sprouted or nonirradiated potatoes. As the results on flavor reported by these authors were limited, long term storage experiments are being conducted at Michigan with enough potatoes to conduct four taste panel tests each month. In the first potato test begun on January 12, 1954, winter Minnesota-grown Irish-cobbler type potatoes were obtained from Food Service and were given the following doses of gamma radiation: 0, 7,000, 21,000, and 28,000 rep. Immediately after irradiation, the irradiated and nonirradiated potatoes were packed in perforated, unsealed, polyethylene bags; then placed in the Food Service cold room held at 50°F and approximately 50 percent relative humidity.

Four taste panel tests were held directly after irradiation to detect flavor differences, if any, in freshly irradiated potatoes. For each panel test six potatoes of each treatment were boiled in their jackets until tender. After cooking the potatoes were peeled and mashed. Milk, butter, and salt were added and the potatoes were whipped. The judges showed no preference for either the irradiated or nonirradiated potatoes. These Minnesota potatoes obtained from Food Service were not considered to be of the best quality. It was the general opinion of the judges that the irradiated potatoes, particularly those given 21,000 and 28,000 rep had a sweeter flavor than the controls. To a few judges the irradiated potatoes had a very slight burned taste with a faint similarity to the flavor of American fried potatoes.

After 3 weeks storage the control potatoes started to sprout as shown in Fig. 19; Fig. 20 shows 2-1/2 months storage. The irradiated potatoes were all in excellent conditions, but the controls had sprouted extensively and were soft and withered. After cooking and mashing it was noticed that the mashed controls contained many small lumps. The results of the taste panel tests given in Table 17 indicate that no preference was shown between the nonirradiated and irradiated potatoes after 2, 3, and 4-1/2 months of storage. Some judges observed the stored controls to be slightly sweeter than the stored irradiated potatoes; the converse of the results on freshly irradiated potatoes.

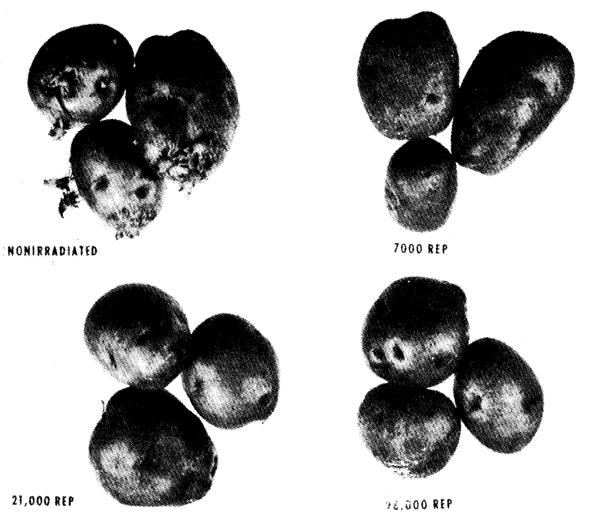


Fig. 19. Minnesota Potatoes Stored 3 Weeks after Irradiation. Control Potatoes (Upper Left) Have Begun to Sprout.

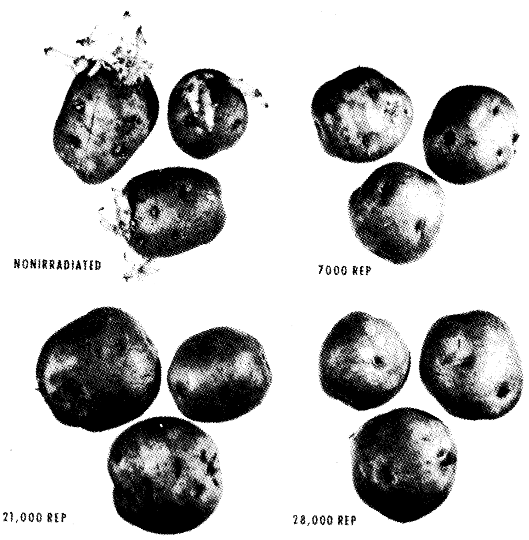


Fig. 20. Minnesota Potatoes Stored for 2-1/2 Months after Irradiation.

TABLE 17

RANKING OF POTATOES BY MEANS OF PAIRED COMPARISONS

Sitting No.	Storage Time, months	Dose and Total Scores for Each Dose* 0	$7x10^3$	$2.1x10^4$	$2.8x10^4$	P	Significant Difference ?
1	0	19	22	14	17	.04	Yes
		.12	.04	.61	.22		
2	0	28	24	28	28	.61	No
3	0	26	32	25	25	.14	No
4	0	25	26	31	26	.33	No
5	1	21	19	27	23	.07	No
6	1	22	24	23	21	.89	No
7	1	24	26	32	26	.12	No
8	1	23	27	27	31	.15	No
9	2	26	20	23	21	.27	No
10	2	25	29	25	29	.45	No
11	2	28	21	28	31	.03	Yes
		.15	.62	.15	.08		
12	2	25	30	25	28	.40	No
13	3	27	20	20	23	.09	No
14	3	29	26	29	24	.44	No
15	3	28	26	26	28	.91	No
16	3	24	26	30	28	.37	No
17	4.5	28	29	26	25	.70	No
18	4.5	26	24	28	30	.37	No
19	4.5	29	25	26	28	.70	No
20	4.5	31	26	27	24	.24	No

*Where significant differences exist estimates of the preference ratings are displayed below the total scores.

Potatoes stored 4-1/2 months are shown in Fig. 21. At this time the nonirradiated potatoes were sprouted to such an extent that little pulp was left in the potato and most potatoes of this group had developed black hearts. Some judges reported that the nonirradiated potato flavor had become sour and spoiled. It was not possible to continue the original experiment after 5-1/2 months since an insufficient quantity of controls were left as a result of excessive spoilage and sprouting. A few of the potatoes given 7000 rep had started to sprout after 4-1/2 months and those given 28,000 rep had a mottled appearance, but the potatoes given 21,000 rep were

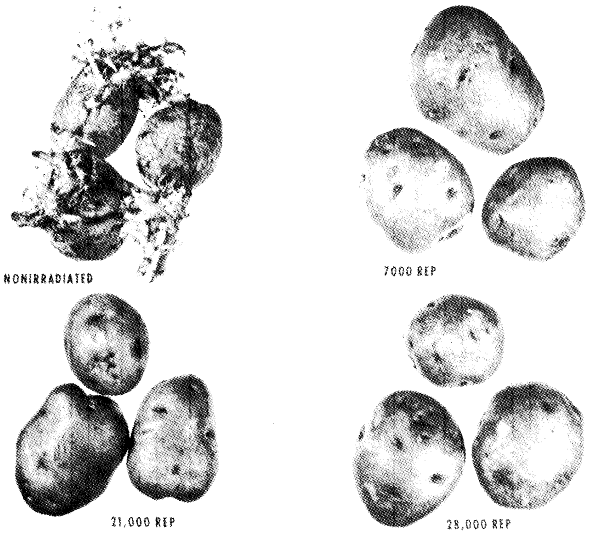


Fig. 21. Minnesota Potatoes Stored for 4-1/2 Months after Irradiation.

in excellent condition. The condition of the irradiated potatoes will be observed until further changes are noticed.

On May 12, 400 pounds of Idaho-grown Burbank russet potatoes were obtained for storage at 50°F and approximately 50 percent relative humidity. One hundred pounds each were exposed to 0, 7000, 21,000, and 28,000 rep of gamma radiation and then stored in the burlap bags in which they were received. After 3 weeks it was noticed that the nonirradiated potatoes had started to sprout as shown in the upper left of Fig. 22. Figure 23 shows the potatoes after 7 weeks. Sprouting of the controls was very irregular which led to the belief that these potatoes may have received a chemical pretreatment. Unfortunately, the history of these potatoes prior to May, 1954, was not known.

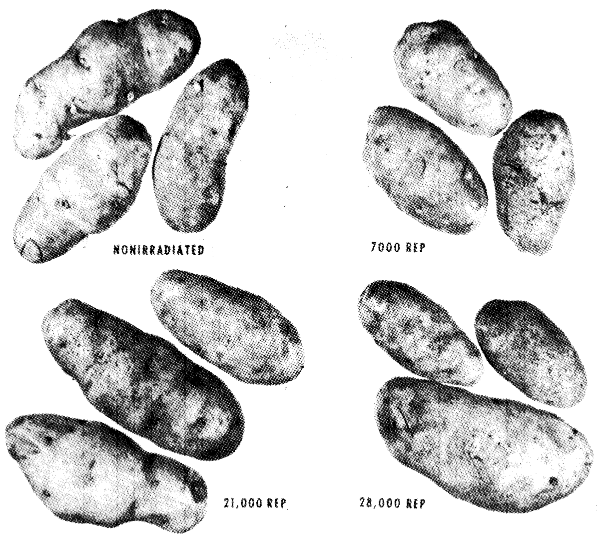


Fig. 22. Idaho Potatoes Stored for 3 Weeks after Irradiation. Nonirradiated Potatoes (Upper Left) Have Begun to Sprout.

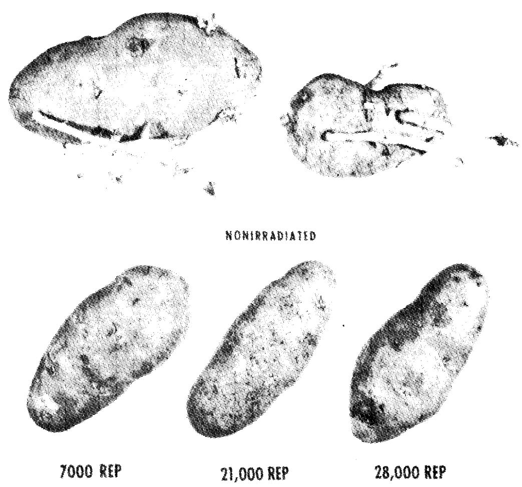


Fig. 23. Idaho Potatoes Stored for 7 Weeks after Irradiation.

(2) Onions: Ranking by Means of Paired Comparisons. Personnel of Brookhaven National Laboratory did not report experiments with irradiated onions and since onions are also a root vegetable susceptible to sprouting, some experiments were made in which onions were given the same low doses of gamma radiation as were given to the potatoes. Spanish onions were divided into four lots; exposed to 0, 7000, 21,000, and 28,000 rep; and packed in perforated, unsealed, polyethylene bags. The onions were stored in the same cold room in the Food Service building as were the potatoes at a temperature of 50°F and approximately 50 percent relative humidity.

Four taste panels were held immediately after irradiation. Since onions are served in a variety of ways, there was a question as to which method would be the best for the detection of flavor differences due to irradiation. It is difficult to fry onions to an even degree of brownness and fried onions are not too palatable when lukewarm as the fat solidifies. Raw onions were extremely strong in flavor and taste fatigue developed soon after tasting one or two samples. Boiling and seasoning with salt and butter was found to be the best method of preparation. Taste panel results are shown in Table 18.

TABLE 18

RANKING OF ONIONS BY MEANS OF PAIRED COMPARISONS

Sitting No.	Storage Time, months	Dose and Total Scores for Each Dose				P	Significant Difference ?
		0	$7x10^3$	$2.1x10^4$	$2.8x10^4$		
1	0	23	26	32	27	.07	No
2	0	23	27	31	27	.15	No
3	0	26	30	24	28	.37	No
4	0	26	21	24	19	.14	No
5	0	28	29	26	25	.70	No

No significant difference in preference was shown between the irradiated and nonirradiated onions immediately after irradiation. Irradiated onions lose some of their sharpness and are slightly sweeter. Even though it was not necessary to cook the irradiated onions as long as the controls for the same degree of tenderness, the texture of the irradiated onions is much softer. Judges who disliked irradiated onions did so because of loss of crispness, not because of the blander flavor.

After 1-1/2 and 2-1/2 months storage there was still no significant difference in flavor between the irradiated and nonirradiated onions as shown in Table 19. After 3-1/2 months storage there seemed to be a slight preference for the nonirradiated onions. Some of the nonirradiated onions did sprout as shown in Figs. 24 and 25. However, both the nonirradiated and irradiated onions were very susceptible to mold and after 2 months storage many of the onions had to be discarded since they would have been unedible. Although the polyethylene bags were perforated and unsealed, evidently the material was too effective as a moisture barrier for the storage of onions as onions require quite dry storage. It is believed that the extensive mold growth accounted for the poor flavor of all the onions after 3-1/2 months storage, therefore this experiment was discontinued.

TABLE 19

RANKING OF ONIONS BY MEANS OF PAIRED COMPARISONS

Sitting No.	Storage Time, months	Dose and Total Scores for Each Dose*				P	Significant Difference ?
		0	$7x10^3$	$2.1x10^4$	$2.8x10^4$		
1	1	24	27	33	24	.02	Yes
		.37	.21	.06	.37		
2	1	28	26	26	28	.91	No
3	1	15	19	18	20	.40	No
4	1	28	24	27	29	.58	No
5	2.5	29	23	28	28	.33	No
6	2.5	30	27	26	25	.59	No
7	2.5	18	24	23	25	.12	No
8	2.5	27	25	30	26	.58	No
9	3	21	23	22	24	.88	No
10	3	25	29	25	29	.45	No
11	3	17	18	18	19	.99	No
12	3	20	25	24	21	.41	No

*Where significant differences exist estimates of the preference ratings are displayed below the total scores.

Another storage experiment was attempted with small pearl onions (boiling onions). These onions are particularly susceptible to sprouting. Onions were divided into four lots and were exposed to 0, 7000, 21,000, and 28,000 rep of gamma radiation. The onions were packaged in medium size brown paper bags and taken to the cold room at Food Service for storage at

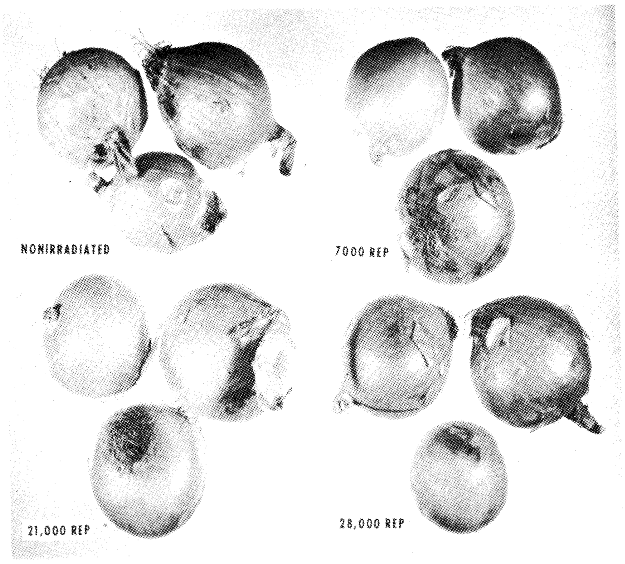


Fig. 24. Spanish Onions Stored for 2-1/2 Months after Irradiation.

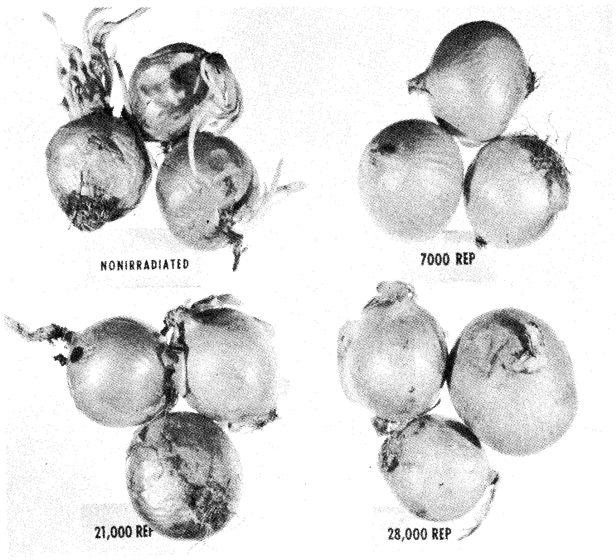


Fig. 25. Spanish Onions Stored for 3-1/2 Months after Irradiation.

50°F and approximately 50 percent relative humidity. Unexpectedly it was found that some of the irradiated onions sprouted more quickly than the non-irradiated onions. After 2 months of storage, twice as many irradiated onions of each dose had started to sprout. However, the early sprouts on the irradiated onions did not continue to grow. Figure 26 shows the pearls after 3 months storage.

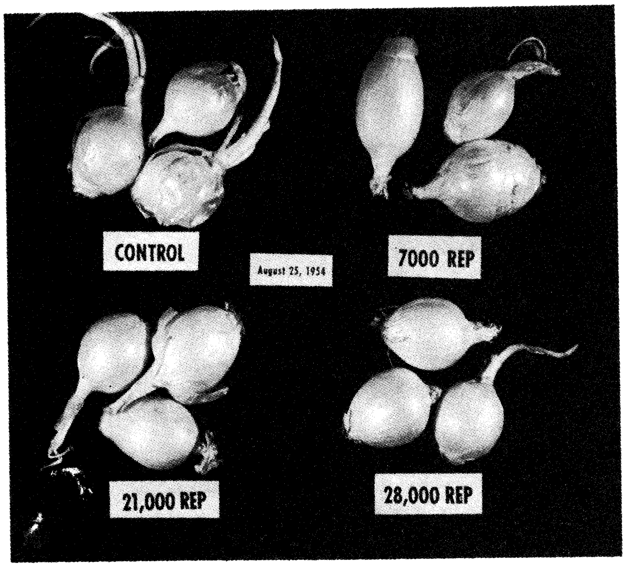


Fig. 26. Pearl Onions Stored for 3 Months after Irradiation.

To investigate further the effects of gamma radiation on onions, 400 pounds of yellow dry onions were obtained during the month of May for storage studies. The onions were irradiated and stored in the open mesh bags in which they were received. The onions were divided into four lots, then exposed to 0, 7000, 21,000, and 42,000 rep of gamma radiation. Immediately after irradiation the onions were moved to the cold room of Food Service for storage at 50°F and approximately 50 percent relative humidity. After 1-1/2 months storage, there was no noticeable change in the appearance of these onions. The changes in these onions will be observed regularly throughout the storage period.

(3) Coleslaw: Ranking by Means of Paired Comparisons. Mr. William Kuhn of the Aunt Mid Company of Detroit, Michigan, cooperated with personnel of the Fission Products Laboratory in experiments regarding the increase of the shelf-life of prepackaged salad vegetables by means of gamma radiation. The Aunt Mid Company packages three products in perforated cellophane bags; spinach, coleslaw, and a chef salad mixture of eight vegetables. Mr. Kuhn stated[2] that their problem was to prolong the life of the fresh packaged produce from the time it leaves the plant, to the supermarket's shelves, and ultimately to the consumer's table. On packaging the produce is immediately put into refrigerated rooms at the packaging plant. From the plant it is delivered in refrigerated trucks to either the large chain store's warehouse which is not refrigerated or to Aunt Mid's own distribution store which also is not refrigerated. Normally it is delivered to the retail store on the day packaged or the next day. At the retail store it may be put immediately into the retail refrigerated room and on refrigerated shelves, but more frequently it is not placed in the refrigerated rooms and many retail stores do not have properly refrigerated shelves.

It was decided to try prolonging the refrigerated and nonrefrigerated shelf-life of the packaged coleslaw by exposing it to low doses of gamma radiation. Low doses are effective in killing nonspore-forming bacteria, yeast, and molds; and undesirable flavor, texture, and color changes are usually not produced at these low levels.

The Aunt Mid Company supplied 72 cellophane packages and 20 polyethylene packages of coleslaw. The coleslaw was divided into four lots, then exposed to 0, 100,000, 500,000, and 1,000,000 rep of gamma radiation. An equal number of samples were stored at room and refrigerator temperature.

Coleslaw samples stored at room temperature started to deteriorate in 7 days. The control samples packaged in cellophane were in slightly better condition than the cellophane samples given 1.0×10^5 and 5×10^5 rep. The samples given 10^6 rep discolored extensively. All the samples packaged in polyethylene were in somewhat poorer condition than those in cellophane.

The coleslaw samples stored in the refrigerator were used for the taste panel tests. The four taste panel tests immediately after irradiation showed that with increasing doses of irradiation the coleslaw becomes sweeter, milder, and has less of the aftertaste and bitterness usually associated with fresh raw cabbage. At 100,000 rep there was no difference in color between the irradiated and nonirradiated samples and only a very slight flavor change. The samples given 5×10^5 and 10^6 rep were slightly discolored with a mottled appearance and lost some of their crispness.

Four additional taste panels were held with the refrigerated samples 1 week after the first four panels. At this time the control samples were found to be the least desirable as they had wilted and spoiled. The control samples had a brownish color due to wilting, whereas the samples given 5×10^5 and 10^6 rep had a gray mottled appearance. The sample given 100,000 rep was the only sample which retained its fresh white color and crispness. Those given 5×10^5 and 10^6 rep were more discolored and had less crispness than in the previous week. The irradiated samples showed no noticeable flavor change during this storage period.

Two more taste panel tests were held 17 days after irradiation. The controls had wilted and deteriorated to such an extent that the coleslaw was unpalatable and obnoxious to the panel. The coleslaw given 100,000 rep was still considered to be in good condition. Coleslaw given 5×10^5 and 10^6 rep showed extensive organic deterioration, which was most severe in the samples receiving the larger dose. The flavors of the samples receiving doses of 5×10^5 and 10^6 rep were of fair quality, but the odors and color changes were rather severe and were considered to be objectionable.

On the 18th day of storage the samples were examined again. At this time it was noticed that the samples given 100,000 rep were starting to wilt. Also it was found that the coleslaw packaged in cellophane was in better condition than that packaged in polyethylene at all the levels of gamma radiation used.

Statistical results of this experiment are shown in Table 20. Immediately after irradiation no significant difference in preference was shown between the irradiated and nonirradiated coleslaw. One week later the results of the taste panel tests show that the samples given 1×10^5 rep were definitely preferred. The samples of next order of preference were those given 5×10^5 rep. There was no significant preference between nonirradiated samples and those given 10^6 rep. No further changes in preference were found after 17 days of storage, except that the controls were slightly preferred over the samples given 10^6 rep.

In another coleslaw storage experiment 54 packages of coleslaw were divided into four portions, then exposed to 0, 50,000, 100,000, and 150,000 rep. These three levels of irradiation were selected for the purpose of learning whether a dose higher and lower than 100,000 rep would keep coleslaw fresh for a longer period of time.

All the samples were stored at refrigerator temperature at first. After 6 days some of the samples were removed to room storage. This schedule would resemble somewhat the treatment of the coleslaw from the time it is packaged until moved to retail stores and placed on produce stands without refrigeration facilities. The nonirradiated samples had already started to

TABLE 20

RANKING OF COLESLAW BY MEANS OF PAIRED COMPARISONS

First Experiment

Sitting No.	Storage Time, days	Dose and Total Scores for Each Dose*				P**	Significant Difference ?
		0	1×10^5	5×10^5	1×10^6		
1	0	26	25	29	28	.70	No
2	0	23	21	28	24	.87	No
3	0	23	27	24	16	-	Yes
		.08	.03	.06	.82		
4	0	25	28	23	20	.49	No
5	7	31	21	25	31	-	Yes
		.07	.61	.25	.07		
6	7	32	20	.27	29	-	Yes
		.05	.72	.14	.09		
7	7	24	22	19	25	.27	No
8	7	32	21	25	30	-	Yes
		.06	.61	.25	.09		
9	17	30	22	23	33	-	Yes
		.08	.49	.39	.04		
10	17	29	20	26	33	-	Yes
		.08	.72	.16	.03		

*Where significant differences exist estimates of the preference ratings are displayed below the total scores.

**- indicates less than 0.01.

wilt and mold growth was noticed 2 days after storage at room temperature. The irradiated samples were all considered to be in good condition after 2 days storage at room temperature. After 4 more days storage at room temperature small black spots of mold were noticed in all the irradiated samples. The controls were completely spoiled.

Three taste panels were held immediately after irradiation. Results are shown in Table 21. No significant difference in preference was shown between the irradiated and nonirradiated coleslaw. Also there were no differences in texture and color.

TABLE 21

RANKING OF COLESLAW BY MEANS OF PAIRED COMPARISIONS

Second Experiment

Sitting No.	Storage Time, days	Dose and Total Scores for Each Dose*				P**	Significant Difference ?
		0	$5x10^4$	$1x10^5$	$1.5x10^5$		
1	0	31	23	28	26	.15	No
2	0	21	24	24	21	.72	No
3	0	29	22	29	28	.13	No
4	7	29	18	20	23	-	Yes
		.02	.54	.31	.14		
5	7	22	16	19	16	.06	?
6	7	26	24	21	19	.14	No

* Where significant differences exist estimates of the preference ratings are displayed below the total scores.

** - indicates less than 0.01.

More taste panels were held 1 week later with the refrigerated samples. At this time the controls were in a very wilted condition. The results show a preference for the irradiated samples even though the judges thought there was no difference in flavor between the nonirradiated and irradiated samples. Since the controls were inedible 12 days after irradiation, no more taste panels were held. However, the irradiated samples were in excellent condition after 2 weeks storage in the refrigerator. Figure 27 shows the coleslaw samples after 20 days of storage. After 23 days of storage the samples given 50,000 rep were still in good condition, but those exposed to $1.0x10^5$ and $1.5x10^5$ rep were beginning to mold. In 4 weeks the samples given 50,000 rep were beginning to wilt, but no mold growth was noticed.

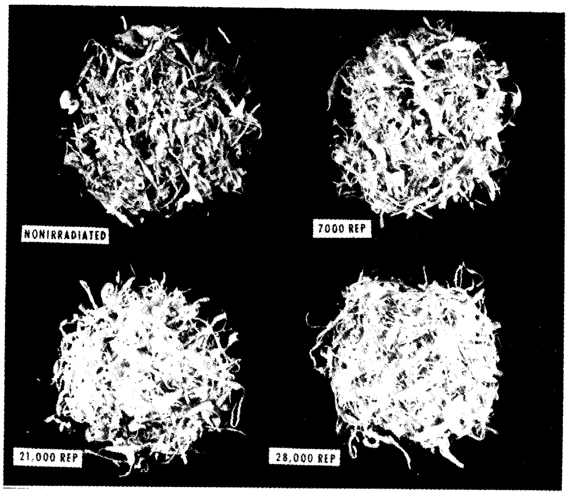


Fig. 27. Coleslaw Samples Stored for 20 Days after Irradiation.

These tests indicate that the refrigerator storage life of prepackaged coleslaw exposed to 50,000 rep can be prolonged to approximately 25 days. By comparison the refrigerator storage life of nonirradiated coleslaw is about 6 or 7 days. Neither the nonirradiated or irradiated coleslaw kept well at room temperature. No color, texture, or flavor changes were observed in coleslaw exposed to gamma radiation between 50,000 and 150,000 rep.

(4) Endive: Observation, No Taste Panel Data. The Aunt Mid Company uses a small amount of cut-up endive in their coleslaw salad. It was noticed in the first experiment on irradiated coleslaw that the endive spoiled rapidly. Two samples of endive packaged in cellophane were tested. One package was kept as a control and the other was given a gamma radiation dose of 50,000 rep. After 8 days storage in the refrigerator the control and irradiated samples were both fresh. After 12 days in the refrigerator the irradiated endive was in better condition than the control which was very wilted, but had turned slightly brown around the ends of the stalks and leaf edges. After 19 days the control endive had completely spoiled, but the irradiated samples had not undergone further change. The irradiated sample spoiled after 21 days.

(5) Cabbage: Observation, No Taste Panel Data. Three cabbages in which the cores had been removed were used in another experiment. These cabbages were packaged in unsealed cellophane bags. One cabbage was kept as a control and the other two were given gamma radiation doses of 50,000 and 100,000 rep, respectively. The irradiated cabbages were observed to be in better condition after 12 days of storage in the refrigerator. After 19 days storage the cavity of the control cabbage was completely black and the rest of the cabbage appeared wilted. Although the cavities of the irradiated cabbages were slightly brown, the rest of the cabbages were in much better condition than the nonirradiated cabbage.

(6) Fresh Asparagus: Ranking by Means of Paired Comparisons. Fresh asparagus obtained from Food Service was divided into eight lots. Two lots were set aside for controls and two lots each were given the following doses of gamma radiation: 200,000, 500,000, and 1,000,000 rep. One lot of each treatment was used for two taste panels the following day. The other samples were stored in perforated, unsealed, polyethylene bags at 40°F to be used for taste panels 1 week later.

The asparagus was cut into two-inch pieces and cooked until tender in a small amount of water with butter and salt added. Statistical results are shown in Table 22. A slight preference was shown for the nonirradiated asparagus. Asparagus was observed to lose its characteristic bouquet when irradiated. The samples exposed to 10^6 rep were slightly softer and somewhat bleached.

TABLE 22

RANKING OF ASPARAGUS BY MEANS OF PAIRED COMPARISONS

Sitting No.	Storage Time, days	Dose and Total Scores for Each Dose* 0	$2x10^5$	$5x10^5$	$1x10^6$	P**	Significant Difference ?
1	0	21	20	24	25	.41	No
2	0	14	20	17	21	.06	Yes
		.61	.09	.23	.07		
3	7	19	21	22	28	.02	Yes
		.45	.28	.22	.05		
4	7	13	17	18	24	-	Yes
		.78	.13	.09	-		

*Where significant differences exist estimates of the preference ratings are displayed below the total score.

**- indicates less than 0.01.

After 1 week storage in the refrigerator there was a decided preference for the nonirradiated asparagus. All the samples appeared in good condition except that the sample exposed to 10^6 rep was much softer than it was the week before. Also, the samples given $5x10^5$ and 10^6 rep had a very decided off-flavor.

d. Tests with Coffee: Ranking by Means of Paired Comparisons

One pound of roasted coffee was divided into four parts. One part was kept for a control and the other three parts were given 10^4, 10^5, and 10^6 rep of gamma radiation. The coffee was made in four dripolators using the standard amount of coffee (2 tablespoons per 3/4 cup water). The coffee was tasted while hot following regular taste panel procedure. Results for one panel are shown in Table 23. It was the general opinion of the judges that the irradiated coffee had lost its fresh coffee aroma and resembled stale coffee.

e. Tests with Irradiated Flour

(1) Cake: Ranking by Means of Paired Comparisons. In the first test with irradiated flour three types of flour, (1) bread flour,

TABLE 23

RANKING OF COFFEE BY MEANS OF PAIRED COMPARISONS

Sitting No.	Storage Time	Dose and Total Scores for Each Dose: 0	$1x10^4$	$1x10^5$	$1x10^6$	P	Significant Difference ?
1	0	25	26	29	28	.70	No

(2) all-purpose flour (General Mills), and (3) cake flour (Swansdown), were given 20,000 rep of gamma radiation. Baking tests were made the day following irradiation. A quick-mix cake flour recipe was used to make cakes with the irradiated and nonirradiated cake flour, whereas a quick-mix all-purpose flour recipe was used for the irradiated and nonirradiated all-purpose and bread flour (see Table 24 for formulas and method used).

TABLE 24

QUICK-MIX CAKE FORMULA DEVELOPED FOR USE
WITH CAKE FLOUR AND ALL-PURPOSE FLOUR

Quick-Mix Cake Flour Recipe		Quick-Mix All-Purpose Flour Recipe	
Ingredients		Ingredients	
Flour	2-1/4 c	Flour	2 c
Sugar	1-1/4 c	Sugar	1-1/4 c
Salt	1 tsp	Salt	1 tsp
Baking powder	3 tsp	Baking powder	3 tsp
Shortening	1/2 c	Shortening	1/2 c
Milk	7/8 c	Milk	1 c
Eggs	2	Eggs	2
Vanilla	1 tsp	Vanilla	1 tsp

Method

Sift together flour, sugar, salt, and baking powder. Place in mixing bowl with the shortening and approximately 3/4's of the milk. Beat for 2 minutes (150 strokes per minute). Scrape bowl often. Add eggs, vanilla, and remaining milk. Beat for 2 more minutes, scraping bowl often. Divide batter into two 8-inch layer pans. Bake at 350°F for 30 to 35 minutes. Let cool on racks for 10 minutes before removing from pans.

One cake was baked with each flour, nonirradiated and irradiated, and used for two taste panels. The data were not analyzed statistically since there were too many variables. However, the general conclusion made was that a low dose of gamma radiation of 20,000 rep does not affect the baking, eating qualities, and flavor of the three flours. The volumes of the cakes made with the irradiated flours were equal to those made with nonirradiated flours. The cakes made with the irradiated and nonirradiated bread flour were extremely heavy and soggy with poor symmetry and volume.

A second test with irradiated flour was made to explore whether or not the gluten of bread flour could be modified by radiation in such a way as to make a softer crumb. A quick-mix recipe for making cake with bread flour was first developed. Then cakes were made with irradiated bread flour that had been exposed to gamma radiation doses of 20,000, 50,000, 100,000, and 150,000 rep. Table 25 gives the formula and method used.

TABLE 25

QUICK-MIX CAKE FORMULA DEVELOPED FOR USE WITH BREAD FLOUR

Ingredients	
Flour	2 c
Sugar	1-1/3 c
Baking powder	3-1/2 tsp
Salt	1 tsp
Shortening	1/2 c
Milk	1 c
Eggs	2
Vanilla	1 tsp

Method

Sift together flour, sugar, salt, and baking powder. Place in mixing bowl with the shortening and approximately 3/4's of the milk. Beat for 2 minutes (150 strokes per minute). Scrape bowl often. Add eggs, vanilla, and remaining milk. Beat 2 more minutes, scraping bowl often. Divide batter into two 8-inch layer pans. Bake at 350°F for 30 to 35 minutes. Let cool on racks for 10 minutes before removing cakes from pans.

All the cakes browned evenly and had a golden brown crust. The cakes made with flour given 50,000, 100,000, and 150,000 rep were more flat topped (not as symmetrical) than the control and the cake made with 20,000 rep. There was no noticeable difference between the control cake and that made with 20,000 rep bread flour. The cakes made with 50,000 and 100,000 rep bread flour were considered to be of better quality than that made with 150,000 rep flour which had a wet soggy layer near the bottom of the cake. Taste panel results are shown in Table 26.

TABLE 26

RANKING OF BREAD FLOUR USED FOR CAKES BY MEANS OF PAIRED COMPARISONS

Second Test

Sitting No.	Storage Time	Dose and Total Score for Each Dose* 0	$5x10^4$	$1x10^5$	$1.5x10^5$	P	Significant Difference ?
1		26	26	28	28	.91	No
2		21	30	29	28	.04	Yes
		.62	.10	.12	.15		

*Where significant differences exist estimates of the preference ratings are displayed below the total scores.

At 50,000 rep flavor change was observed, but cakes made from flour given 100,000 and 150,000 rep were considered to have a different flavor. It is difficult to describe this flavor change other than to say that the sweetness appeared to have been increased. These tests indicate that cakes made with irradiated bread flour are not improved over those made with nonirradiated bread flour.

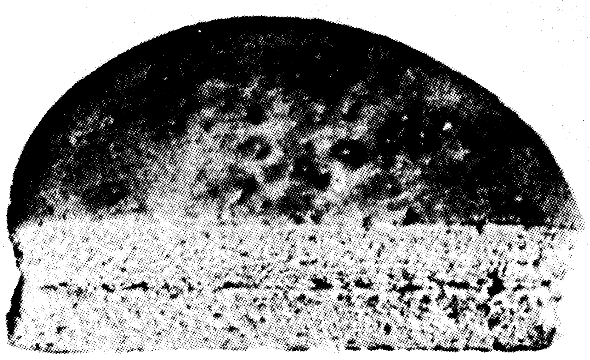
In a third test more bread flour was exposed to doses of 100,000, 150,000, 200,000, 500,000, and 1,000,000 rep. Cakes were made with all the irradiated flours and a nonirradiated control. For the taste panel tests cakes made with flours exposed to 150,000, 200,000, and 500,000 rep were used. Figure 28 shows the six cakes made with irradiated and non-irradiated bread flour. Table 27 gives the cake volumes. Characteristics of the cakes were as follows:

1. Control — This cake had the best symmetry, volume, texture, tenderness, and crumb color.

1

2

3



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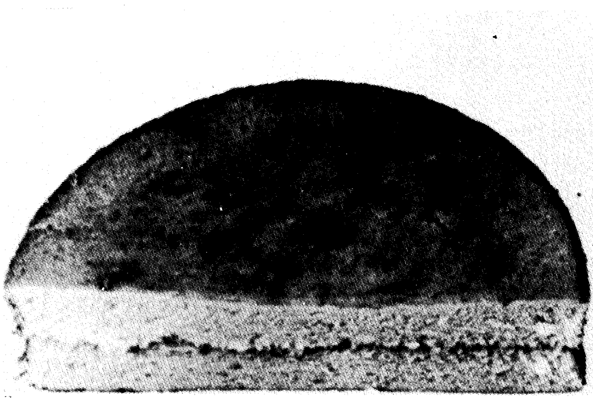

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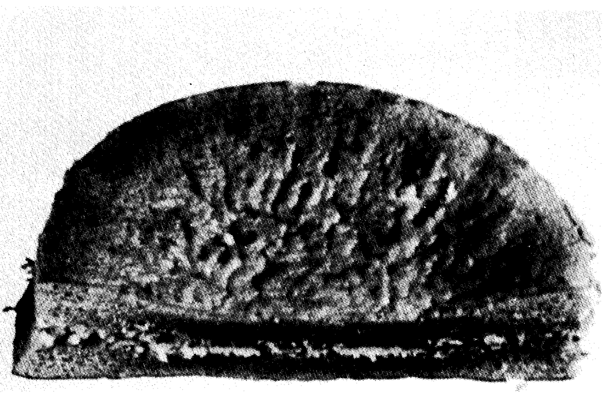

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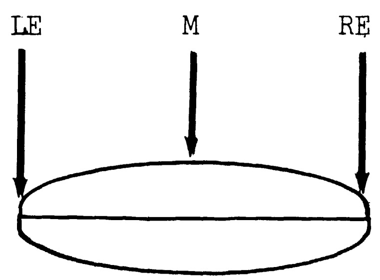
Fig. 28. Six Cakes Made with Nonirradiated and Irradiated Bread Flour.

TABLE 27

VOLUMES OF CAKES MADE WITH IRRADIATED BREAD FLOUR

	Layers	LE	M	RE
1. Control	1st	2-4/16*	2-13/16	2-6/16
	2nd	2-5/16	2-13/16	2-5/16
2. 1000,000 rep	1st	2-3/16	2-9/16	2-4/16
	2nd	2-3/16	2-10/16	2-2/16
3. 150,000 rep	1st	2-5/16	2-10/16	2
	2nd	2-4/16	2-11/16	2-2/16
4. 200,000 rep	1st	2-2/16	2-4/16	2-5/16
	2nd	2-2/16	2-8/16	2-4/16
5. 500,000 rep	1st	2-2/16	2	2-1/16
	2nd	2-1/16	1-14/16	1-15/16
6. 1,000,000 rep	1st	1-11/16	1-4/16	1-14/16
	2nd	1-14/16	1-6/16	2

*Measurements were made in 16th's of an inch at the points marked on this diagram. A layer has been cut in half and the two bottoms placed together at the middle.



2.	100,00 rep	A good cake, but not quite as good as the control. Texture was slightly coarse and compact. Cake was not quite as tender as control. There was no difference in crumb color; symmetry was good.
3.	150,000 rep	This cake was slightly poorer than the 100,000 rep cake. It had a heavy compact layer near the bottom and the crumb color was slightly darker than the control cake. The symmetry was good.
4.	200,000 rep	This cake had more of a heavy, compact layer near bottom. The texture was bready and coarse and the crumb was wet and sticky. Symmetry was poor; layers very flat topped.
5.	500,000 rep	The crust browned evenly but the cake had very poor symmetry; dipped in center. The texture was heavy, compact, bready, and coarse and was tough and gummy with a dark yellow crumb color.
6.	1,000,000 rep	This cake fell badly; had a very brown crust and dark crumb color. A wet, soggy, compact failure.

It was noticed that when mixing cakes with flour given 500,000 and 1,000,000 rep the batters tended to be drier and had a consistency like putty during the first 2 minutes of mixing. Taste panel results are shown in Table 28. The judges showed a slight preference for the control over the cakes made with flour given 150,000 and 200,000 rep. However, there was a decided preference for the control cake when paired with cake made with flour given 500,000 rep. Cakes made with irradiated flour tend to be sweeter, particularly the crust. Flour given $5x10^5$ and 10^6 rep had an odor resembling extreme mustiness.

(2) Baking Powder Biscuits: Ranking by Means of Paired Comparisons. Baking powder biscuits were baked with bread flour that had been exposed to 20,000, 50,000, 100,000, and 150,000 rep of gamma radiation. Table 29 gives the formula and method used. It was decided to bake biscuits because color changes, if any, in irradiated flour would show up better in biscuits. Two bakings were made with each sample of treated flour and the nonirradiated control. Two taste panels were held with these biscuits; results are shown in Table 30.

The control biscuits had the best volume. There was no difference in volume in the biscuits made with irradiated flour. All the biscuits browned an equal amount and there were no differences in outer appearances. Characteristics of the biscuits were as follows:

TABLE 28

RANKING OF BREAD FLOUR USED FOR CAKES
BY MEANS OF PAIRED COMPARISONS

Third Test

Sitting No.	Storage Time	Dose and Total Score for Each Dose*				P	Significant Difference?
		0	1.5×10^5	2.0×10^5	5.0×10^5		
1		18	23	22	27	.04	Yes
		.56	.17	.21	.06		
2		19	22	21	28	.02	Yes
		.45	.22	.28	.05		

*Where signigicant differences exist estimates of the preference ratings are displayed below the total scores.

TABLE 29

FORMULA DEVELOPED FOR USE IN MAKING
BISCUITS WITH IRRADIATED BREAD FLOUR

Flour	2 c
Baking powder	3 tsp
Salt	1/2 tsp
Shortening	1/4 c
Milk	180-190 cc

Method

Sift into mixing bowl the flour, baking powder, and salt. Cut in shortening. Add mikl and mix until dry ingredients are wet. Knead on pastry board 30 times before rolling out to 1/2 inch thickness. Cut biscuits with a 2-inch cutter. Bake at 450°F for 8 to 10 minutes on an ungreased baking sheet.

TABLE 30

RANKING OF BISCUITS BY MEANS OF PAIRED COMPARISONS

Sitting No.	Storage Time	Dose and Total Scores for Each Dose				P	Significant Difference ?
		0	$5x10^4$	$1x10^5$	$1.5x10^5$		
1		25	29	28	26	.70	No
2		19	18	17	18	.96	No

1. Control — These biscuits were even-grained and tender with light cell walls. They had the most tender outer crust.

2. 20,000 rep — These were slightly coarser and closer than the control; not as light a biscuit. There was no difference in crumb color between these biscuits and control biscuits.

3. 50,000 rep — These were lighter and more tender than 20,000 rep biscuits, not as close in texture, but still not quite equal to the control. Differences in crumb color between these biscuits and the controls were very slight.

4. 100,000 rep — These seemed to be the most tender of the irradiated biscuits, but not quite equal to the control. Also, they were more like the control in lightness and cell wall structure. They had a slightly grayer crumb color than the control biscuits.

5. 150,000 rep — These biscuits had a coarser texture, were more compact, and were not as tender as 100,000 rep bescuits and the control biscuits. The crumb color was slightly darker than 100,000 rep biscuits.

All the irradiated samples felt gummier in the mouth. Although there were no marked changes in the baking qualities of bread flour exposed to gamma radiation between 20,000 and 150,000 rep, there were some noticeable flavor changes. It seemed to some of the judges that the off-flavor of irradiated flour was accentuated by the presence of double-action baking powder. Not enough taste panel data are on hand to make any definite conclusions, but most of the panel members did not care for the flavor and eating qualities of the biscuits made with irradiated flour.

(3) Bread: Ranking by Means of Paired Comparisons. While studying the baking characteristics of irradiated bread flour, it was decided to make some tests with bread. The formula and method used are shown in Table 31.

TABLE 31

FORMULA DEVELOPED FOR USE IN MAKING BREAD WITH IRRADIATED FLOUR

Milk, scalded	1 c
Water	1/3 c
Shortening	2 T
Yeast	1/2 oz
Sugar	1 T
Salt	2 tsp
Flour, sifted	4 c, approximately

Method

Dissolve yeast in water (110°F). Pour scalded milk over shortening, sugar, and salt. When the milk mixture is lukewarm (80-85°F), add the yeast mixture. Add flour gradually. Knead dough 5 or 6 minutes until smooth, then place in a well-greased bowl and allow to rise from 1 hour 15 minutes to 1 hour 30 minutes. Knead dough again for 3 or 4 minutes; then allow to rise again for 45 to 60 minutes. Knead dough again for 1 minute; then shape it to fit the bread pan which is well greased. Allow dough to rise in the pan until slightly more than double in size. Bake for 30 minutes at 400°F and 20 minutes at 350°F. Yield: one (9x5x3) loaf.

Bread was baked with flour that had been exposed to 200,000, 500,000, and 1,000,000 rep gamma radiation and a nonirradiated control. Figure 29 shows sections of loaves of bread made with these flours. Characteristics of the bread were as follows:

Fig. 29. Sections of Four Loaves of Bread Made with Nonirradiated and Irradiated Bread Flour.

1. Control	This was the best loaf, tender cell walls, even grain, and the best volume.
2. 200,000 rep	This bread was still good, but volume not quite as good as the control loaf. There was not much difference in texture, tenderness, crumb color, and crust color between this loaf and control loaf.
3. 500,000 rep	The volume of this loaf was poor. There were more large gas holes and the texture was quite coarse near the bottom crust. The crumb was not as tender as the loaf made with 200,000 rep flour and crumb color was darker than loaf containing 200,000 rep flour.
4. 1,000,000 rep	The volume of this loaf was equal to that made with flour given 500,000 rep. There were many more gas holes than in 500,000 rep loaf and the crumb was not as tender. Some small hard lumps were noticed in the crumb and the color was a medium tan. Both the 500,000 and the 1,000,000 rep loaves did not have very thick crusts nor were the crusts as brown as the other loaves, even though all were baked for the same period of time.

Taste panel results are shown in Table 32. The judges had a slight preference for the nonirradiated loaf and those made from flour given 200,000 and 500,000 rep. However, the flavor of the control was definitely preferred to the loaf made with flour given a dose of 1,000,000 rep. Bread from the latter loaf was considered to have a definite off-flavor.

TABLE 32

RANKING OF BREAD FLOUR USED FOR BREAD BY MEANS OF PAIRED COMPARISONS

Sitting No.	Storage Time	Dose and Total Scores for Each Dose*				P**	Significant Difference ?
		0	$2x10^5$	$5x10^5$	$1x10^6$		
1		24	25	28	31	.19	No
2		19	20	22	29	-	Yes
		.44	.34	.20	.02		

*Where significant differences exist estimates of the preference ratings are displayed below the total scores.

**- indicates less than 0.01.

3. DISCUSSION AND CONCLUSIONS

Taste panel studies evaluated by ranking of paired comparisons continued to be an accurate and statistically reliable method of detecting flavor changes and determining flavor references in food subjected to gamma radiation.

The experiment on the refrigerator storage life of raw ground pork indicated that a radiation dose between 60,000 and 80,000 rep increases storage life to 10 or 11 days. This is the same conclusion reached from a previous experiment reported in Progress Report 6.[3] With doses higher than 80,000 rep, off-flavors occur on storage due to the development of rancidity.

The experiment on the keeping qualities of gamma-ray pasteurized raw ground beef stored at refrigerator temperature was not considered to be successful. The beef was obtained from a local slaughterhouse and was tough and of poor flavor with a marked smoky odor and taste. As a result, the judges of the taste panel were not able to distinguish off-flavors caused by irradiation and storage. Taste panel results indicate that ground raw beef exposed to a dose of gamma radiation between 80,000 and 110,000 rep will remain in good condition at refrigerator temperature for 8 days and possibly longer. This is in agreement with the experiment reported in Progress Report 6.[3] The beef given dosages of 50,000 rep did not keep as long as the portions given the higher doses. It is believed that beef can be given a higher radiation pasteurization dose than pork because the lower fat content and higher degree of saturation of the fatty acids in the fat makes it less subject to the development of rancidity.

Tests with navel oranges showed promising results when whole oranges were given 30,000 rep of gamma radiation. The juice extracted from these oranges did not develop a bitter flavor, whereas juice extracted from nonirradiated oranges and those given 60,000 and 100,000 rep did develop bitterness on standing. The direct irradiation of juice extracted from navel oranges did not prevent the development of bitterness when the juice was permitted to stand overnight.

From the results of the potato storage experiment to date, the statement can be made that potatoes exposed to 21,000 rep of gamma radiation may be expected to keep for at least 5-1/2-months storage at 50°F without sprouting, whereas the nonirradiated potatoes sprouted in 3 weeks. The potatoes given 7,000 rep and stored at 50°F thereafter showed a few small sprouts after 4-1/2 months. Potatoes given 28,000 had a mottled appearance after storage for 5-1/2 months. The optimum radiation dose is believed to lie between 7,000 and 21,000 rep.

Although the irradiated potatoes, particularly those given 21,000 and 28,000 rep, had a sweeter flavor and a slightly burnt taste, the judges showed no preference for either irradiated or nonirradiated potatoes. Flavor changes were not noticed in the irradiated potatoes during storage. The flavor of the nonirradiated potatoes eventually turned sour as a result of prolonged storage.

The experiment on the storage of pasteurized onions was not conclusive. This was partly the result of unsatisfactory storage conditons. Because onions need a dry atmosphere, perforated, unsealed polyethylene should not have been used for packaging and the humidity of the storage room should have been lower. However, it was not possible to regulate the humidity to suit the experiment because Food Service uses the cold storage room for other purposes.

The nonirradiated Spanish onions rooted in 1 month, whereas the irradiated onions did not root. Because of the poor storage conditions the onions were more susceptible to mold than to sprouting. Irradiated onions do not have as sharp a flavor, are slightly sweeter, and do not need to be cooked as long as nonirradiated onions.

Contrary to the results obtained with Spanish onions, gamma radiation promoted the initial sprouting of the small pearl or white boiling onions. Although the irradiated onions sprouted more rapidly, they did not continue to grow.

Although the shelf-life of pasteurized, prepackaged cut cabbage for coleslaw was increased when stored at refrigerator temperature, poor keeping qualities were noted when stored at room temperature. Nonirradiated cut cabbage for coleslaw remained in a fresh condition for only 6 days at 40°F, whereas that given 50,000 rep kept for 25 days and that given 100,000 rep kept for 17 days. Neither of these irradiated samples had noticeable off-flavors, textures, or color changes. It was also found that the coleslaw remained fresh longer when stored in perforated cellophane than when stored in perforated polyethylene.

Short observation experiments with endive and whole cabbages were promising. Such vegetables given gamma radiation doses of 50,000 and 100,000 rep remained fresh at refrigerator temperature for a longer period of time than the nonirradiated vegetables. No flavor changes were observed at these low doses.

The flavor of cooked irradiated fresh asparagus is considered to be flat when given 200,000 rep and a definite off-flavor is noticed when the dose is increased to 500,000 and 1,000,000 rep. The flavor changes in irradiated asparagus were accentuated on storage. The portion given 10^6 rep also became softer on storage.

Roasted coffee loses its fresh flavor and aroma when irradiated in air. More experiments should be conducted with coffee packed in vacuum.

Cake flour, all-purpose flour, and bread flour are not changed when given a dose of 20,000 rep gamma radiation. At 20,000 rep flour insects and weevils would be destroyed and insect eggs would be made sterile. Cakes made with these different flours were similar to the controls in all respects.

Bread flour given a dose higher than 50,000 rep is not improved. Cakes made with irradiated bread flour were progressively of poorer quality with increasing doses of gamma radiation. Cakes made with irradiated flour had low total volume, were heavier, more compact, gummier, and had a darker yellow crumb color. The gluten appears to lose its binding power and the flour has a drier or more starch-like texture in mixing. Flavor changes were noticeable in cakes made with bread flour given a dose of 100,000 rep. The cakes, particularly the crusts, have a sweeter flavor. Off-flavors were noticed with higher doses of irradiation.

When making biscuits with irradiated flour receiving 20,000 to 150,000 rep, it was necessary to add 10cc more milk to obtain a workable dough. This dryness of irradiated flour was also noticed when mixing cakes. Biscuits made with irradiated flour had a satisfactory appearance but the eating qualities and flavor were not equal to those of the controls. The biscuits made with irradiated flour were gummier and off-flavors were present in samples made with flour given dosages greater than 20,000 rep.

Aside from total volume, which was slightly smaller in the bread made with 20,000 rep bread flour, the loaves were equal to that made with the nonirradiated flour in all respects. Loaves made with flour given 5×10^5 and 10^6 rep were much poorer in volume and crumb color was comparable to honey bread.

4. REFERENCES

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B. STORAGE TESTS ON GAMMA IRRADIATED POTATOES AND ONIONS AND BIOCHEMICAL TESTS ON IRRADIATED POTATOES

Personnel:

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1. INTRODUCTION

Gamma irradiation of potatoes is believed to be one of the first applications of radiation that may be used by the food industry. For this reason this discussion was prepared for this report by Dr. Svend Pedersen.

2. REVIEW

a. Nature of the Problem

Relatively low dosages of gamma radiation have been observed by Sparrow and Christensen[1,2] and by this laboratory to delay greatly, if not actually prevent, the sprouting of potatoes. This observation indicates that the storage life of potatoes may be extended by gamma irradiation. In all probability late-harvested, mature, northern-grown potatoes could be kept throughout the following winter and spring until early harvested mature potatoes become available the following year. Cheaply applied gamma radiation could prevent the considerable financial losses hitherto taken for granted with regard to potatoes still in storage in the spring. Also this process can make quality potatoes such as Idaho russet, Maine or Long Island potatoes available throughout the year. Idaho russet potatoes are preferred by many cooks for baking and also for French fries. At present these potatoes and other high quality northern-grown mature potatoes are not available in the summer months.

The greatly reduced rate of sprouting can be expected to influence processes and reactions in the potato itself:

1. A reduced requirement for the formation of new tissues

2. An overall lower metabolic rate of potatoes in storage

3. A decreased rate of hydrolysis of stable storage starch molecules into more unstable and utilizable mono- and dis-acharides

4. A smaller loss in potential total energy per unit weight of potato assuming that the moisture content remains relatively unchanged

This low dosage of gamma radiation should not have any marked direct effect on existing enzyme systems in the potato, unless these differ from enzyme systems previously studied in other products or in aqueous solutions in resistance to gamma radiation. However, the low dosages of gamma radiation may act in some manner on the particular enzyme system which under storage conditions initiates the process of sprouting.

Of the carbohydrates, starch is the chief reserve compound stored in most plants with a few exceptions such as sugar cane, sugar beet, and Jerusalem artichoke. All plants in which water-insoluble compounds are stored require digestion of these by means of enzymes. For an example, starch changes by way of dextrin stages into water soluble sacchrides which can be transported or diffused from cell to cell whenever and wherever needed. These saccharides can then be further broken down by other enzyme systems within the cells. The resulting mono-saccharides can be used for energy purposes, such as respiration which takes place at all times as long as the plant tissue is living, and for catabolic changes necessary for the maintenance of life. The catabolism is especially high and the needs are correspondingly great under certain conditions such as when fruits are ripening; seeds are germinating; and tubers, such as potatoes, are sprouting. However, in the dormant storage stage the greatest need is for respiration. In this connection it may be pointed out that recent studies by Guzman-Barron[3] have shown a respiratory quotient of 1.0 for fresh, sliced potatoes indicating complete carbohydrate oxidation.

Ninety years ago it was shown conclusively that respiration was affected very little by light but that humidity and temperature did affect respiration to a marked degree. Later it was found that other factors such as accumulation of sugars, germination, and sprouting also played a role.

Respiration requires energy which is furnished by the oxidation of sugars. As respiration and general plant activities decrease with temperature to a minimum at or near the freezing point, it would be most economical from the point of view of energy loss to store potatoes at that temperature. However, potatoes cannot be satisfactorily stored at such temperatures for the complete storage period.

Storage near 32°F causes accumulation of sugar from the enzymatic breakdown of starch by phosphorylase and by α and β amylases. This is undesirable because it imparts a sweet taste to the potatoes. In addition, such potatoes are not suitable for frying purposes, especially for potato chips which take on a darker color with increasing amounts of sugars present. Up to 6 or 7 percent of total sugars is not uncommon under such storage conditions. This is roughly three times that normally found after storage at 50°F. Subsequently increasing the storage temperature to around 50°F causes most of these excess sugars to disappear in 3 to 4 weeks. By raising the temperature to about 70°F, an estimated 80 percent of the sugars is reconverted into starch and the remainder lost through respiration. This plethera of sugars available in the cells may be assumed to result in a general increase in the metabolism.

Storage of potatoes at 45 to 50°F is common. It is actually a compromise temperature at which the starch decomposition into sugars is satisfactorily controlled and a higher rate of respiration and metabolism accepted. At the same time sprouting, with the concomitant increased demand for sugars and subsequent loss, is kept under control.

During storage the relative quantities of different carbohydrates, such as starch, sucrose, and reducing sugars present at any one time, depend upon the prevailing conditions. During growth the reducing sugars and sucrose decrease and starch increases. Potatoes harvested before maturing ripen and mature during storage. While in storage loss of water takes place from respiration, depending upon the state of activity of the potato, and from transpiration, the rate of which is governed by the relative humidity of the air. In order to minimize the latter loss a relative humidity of 85 to 90 percent has been recommended.

Potatoes stored in large bins and at great depth for prolonged periods result in a high storage temperature because of poor ventilation and an increased respiratory rate. The latter is also increased when the potatoes are "wounded" by rough handling and by great pressure exerted on the lower layers in high stacking.

In large warehouses the use of ventilating air is necessary. In order to prevent water losses due to transpiration the ventilating air should be of approximately the same relative humidity as that desired in the storage bin.

Concerning the ventilation of large warehouses, it has been demonstrated that an atmosphere with normal oxygen content but high in carbon dioxide (up to 20 percent) depresses the accumulation of sugars at temperatures below 45°F over a period of 3 months.

Sprouting of potatoes is actually a proliferation of new tissue starting at the "eyes". It has been claimed that sprouting can be prevented when the content of auxins is high. If the content of auxin is increased by applying the synthetic auxin naphthalene acetic acid in the form of a volatile ester by dusting or spraying, sprouting can be delayed or prevented. This indicates an inhibiting action by the auxin when present in high concentrations.

The keeping qualities of potatoes are also influenced by the types of organisms found on the surface. At low storage temperatures the cryophilic type and at moderate temperatures the mesophilic type predominate.

During storage the potatoes very slowly decrease in potential nutritional value. This is due to the fact that respiration, essential for life, involves an expenditure of energy derived from the oxidation of sugars into carbon dioxide and water. Under favorable storage conditions this is a minor but unavoidable loss. This loss increases with storage temperature, which enhances respiration and other essential processes. This loss is also markedly increased whenever sprouting occurs because the new tissue also respires. As the sprouts themselves are inedible they represent a further decrease in the potential nutritional value of the parent potato.

Sprouting of potatoes in the past has been kept under limited control by providing favorable storage conditions. More recently spraying or dusting with a synthetic auxin has been used and within the last year Brookhaven National Laboratory has introduced gamma irradiation as a new method. In a more recent article Sparrow and Christensen[2] have reported inhibition of sprouting in potatoes (Kahtahdin variety) 18 months after 20,000 rep of gamma radiation. The results of those preliminary studies have been confirmed at the Fission Products Laboratory in a series of controlled investigations previously discussed under Part III, A, Organoleptic Studies, in this report.

The method of action of gamma radiation on potatoes is not known. It is postulated that it could be by

1. Retardation of the reaction of starch into sugars.

2. Promotion of the enzymatic reactions of reconversion of sugars back into starch.

3. Stimulations of auxin formation.

4. Possession in itself of an "auxin-like" property.

5. Destruction of the factor which "triggers" the initiation of sprouting.

6. Destruction of reproductive cells which may be more sensitive to radiation than storage cells.

b. Pertinent Literature on Potatoes

In a recent article by Pentzer and Heinze[4] the postharvest physiology of fruits and vegetables has been described. From the section on chilling injuries it is clear that factors other than temperature itself are involved such as the variety (even species), state of curing, and maturity. The governing mechanism is undoubtedly very complex and not definitely known. According to some it involves the accumulation and the removal of cell poison. Chemical analysis of sugars and organic acids known to accumulate at low storage temperatures have so far given no definite indication of an explanation. Finally, changes in the permeability of cell membranes must also be considered a factor at least in the early stages of chilling.

(1) Control of Sprouting with Chemicals. Sprouting of potatoes may be kept under control by chemical treatment prior to storage. Among the compounds used in such treatments are esters of naphthalene acetic acid and trichlorophenoxy-acetic acid, 3 chloroisoproplyl-N-phenyl carbamate, 2,3,5,6-tetrachlorobenzene, and maleic hydrazide. However, Ezell and Wilcox[5] have recently reported that preharvest spraying of the vines of sweet potatoes (Orange Little Stem) with maleic hydrazide caused numerous black pock marks on the tubers shortly after harvesting.

It has been reported in the literature that sprouting of stored potatoes can be retarded by the application of plant hormones. Whittenberger and Nutting,[6] however, could not confirm the claims that treating seed potato slices with indole acetic acid increased both the potato yield and the size of the potato starch granules. The latter is generally considered to enhance the commercial value.

On the basis of work performed in India, Mathur[7] studied losses in potatoes and found a minimum with undamaged potatoes stored at 41°F. When stored in high piles potatoes showed greater losses in the bottom layers than the upper layers. More losses occurred in smaller than in larger potatoes.

Sprouting was entirely prevented at 32°F with 31 percent sprouting at 41°F and 97.5 percent at 50°F. Duration of storage was not given in the abstract consulted.

According to a U.S. patent by Brown and Mellor[8] a dust, solution, emulsion, or dispersion of 1 to 20 percent of pure chlorinated mononitrobenzene, with at least three chorine atoms, dissolved in a nonphytocidal solvent will prevent sprouting of potatoes. Amount to be used is given as at least three ounces per ton of potatoes. In another patent by Zander[9] peeled, sliced potatoes are immersed in sulfur dioxide solution and subsequently washed with water to remove the sulfur dioxide. By this treatment it is claimed that potato chips of good flavor and color can be prepared regardless of variety of potato used and amount of reducing sugars present.

(2) Browning and Blackening. Browning of potatoes was shown by Thornton[10] to be due to the presence of reducing sugars, not to sucrose or the total sugars present. This browning is a problem of significant commercial importance. Approximately 10 percent of the total potato crop of 40,000,000 bushels in the middle forties was used for the manufacture of potato chips. Potatoes have a normal dormant stage of 9 to 12 weeks during which sprouting does not occur even at room temperature. The practice of potato chip manufactures in the past has been to keep potatoes at low storage temperature; this delays sprouting but causes an accumulation of sugar followed by storage at higher temperature, for a period prior to chipping, which reduces the sugar content.

Blackening of potatoes after cooking appears to be most common in potatoes grown in regions with low temperatures during the growing season. The actionof the enzyme polyphenolaxe is believed to be involved in this blackening. However, the black core at times found in the center of potatoes is believed to be due to a lack of available oxygen.

(3) Sugars. In 1924 Hopkins[11] found an accumulation of sugars in potatoes at low storage temperature, especially of sucrose, which accounts for the sweet taste. However, it is incorrect to assume that sweet tasting potatoes necessarily have been frozen because this accumulation proceeds rapidly at temperatures just above the freezing point.

Denny[12] observed significant increases in sucrose but not in reducing sugars in potatoes which had been treated with ethylene chlorhydrin, thiocyanates, and thiourea. Additional chemical compounds were found by Miller[13] to increase both respiration and the sucrose content of potatoes.

Arreguin-Lozano and Bonner[14] have reviewed the literature on sucrose in potatoes and performed many experiments on factors involved in the starch breakdown and sucrose formation by enzymatic reactions. In potatoes the weak beta amylase breaks starch down into maltose and residual dextrins, whereas the phosphorylase breaks starch down into glucose-1-phosphate. At low storage temperatures the amylase is almost inactive while the phosphorylase activity is very intense. The lower phosphorylase activity at higher storage temperatures is explained by the presence of an inhibitor, as yet unidentified, which apparently is inactive or absent at the lower temperatures.

These authors were not able to demonstrate and isolate from potatoes the enzyme phosphohexoisomerase which is considered to govern the equilibrium between glucose-6-phosphate and fructose-6-phosphate, both necessary for sucrose synthesis of sucrose in solutions of either glucose or fructose when incubated with potato slices. That fructose-6-phosphate and fructose-1,6-diphosphate are actually present in potatoes, indirect evidence of the presence of the above mentioned phosphohexoisomerase, can be seen in an article by Arreguin-Lozano and Bonner,[14] Table 1, page 722. Fructose-6-phosphate is present in greater quantities at lower storage temperature which favor sucrose synthesis while at higher storage temperatures with lesser sucrose synthesis glucose-6-phosphate predominates. Arreguin-Lozano and Bonner believe that starch breakdown and sucrose synthesis in the potato are influenced by three temperature dependent factors: regulation of the inhibitor of phosphorylase which is present in greater amounts at higher storage temperatures, regulation of the enzyme activity synthesizing sucrose which is favored at lower storage temperature, and regulation of the phosphohexoisomerase system which is most active at lower storage temperatures.

During wilting of plant leaves starch is changed into sugars. Such a change has apparently not been shown to occur when potato tubers wilt. Treadway, et al.[15] have shown that potatoes lose carbohydrates and water at approximately the same rate with the total solids remaining constant when stored for 37 weeks at between 1.1 to 15°C. Sprouting did not occur during this cited storage period below 3.3°C. Sprouts were found to contain only small amounts of starch while the concentration of sucrose was high.

(4) <u>Enzyme Systems</u>. In a recent review Schwimmer[16] discussed the literature pertaining to the enzyme systems present in the white potato on the basis of 110 appended references. The enzyme phosphorylase present in large amounts in potatoes is responsible for the reversible action of glucose-1-phosphate to amylase, one of the two chief components of starch. The second component amylopectin is chiefly synthesized by the Q-enzyme that has an action very similar to that of phosphorylase. Amylopectin can be changed into amylase by the action of the R-enzyme. Amylases, both α and β, are present but they have only a very limited function, splitting of maltose molecules.

Knowledge of the synthesis of sucrose from a solution of glucose and fructose in the presence of potato slices is still meager. This reaction is unique in that it proceeds more rapidly at lower temperatures. The synthesis is believed to be related to respiration and the buildup of high energy-level phosphate-bond compounds.

There are several phosphatases present in potatoes. One of them, apyrase, can liberate two out of the three phosphate groups in adenosine triphosphate. The function of aiding in stabilizing the resting state of the potato has been ascribed to this enzyme.

(5) Auxins. The observation that plants tend to grow toward the source of light is a very old one. That this phototropic action was related to definite chemical compounds was first clearly indicated in the classical investigations of Boysen-Jensen[17] on the coleoptile of Avena. Chemical compounds possessing the property of being able to promote and/or regulate plant growth were given the name hormone. At present, however, the term auxin is preferred. A large number of scientific articles have appeared relating to auxins. Gordon[18] has reviewed the occurrence, formation, and inactivation of auxins in a recent article.

The effects of x-rays on living matter have been studied since the nineties. Investigators studied the effects on plant growth in the twenties; but Skoog,[19] in 1935, was probably the first to study the effects on an auxin. Using a high voltage tube capable of being operated at 900 kilovolts and 3 to 4 milliamperes, Skoog demonstrated the inactivating effect of x-rays on an auxin (indoleacetic acid) as well as on auxin extracts from plants. He has shown this inactivation to be due to an oxidation on the basis of experiments in air and in a nitrogen atmosphere. Gordon and Weber[20] have since shown that the destructive effect in air probably may be ascribed to the opening of the ring structure based upon absorption changes in the visible and ultraviolet spectra.

The commonly heard statement "No auxin, no growth and too much auxin, prevention of growth" may be considered correct in a general sense. Only minimal quantities are required for plant growth and the application of an excess can delay sprouting. By the use of chemicals it is possible to prolong artifically the rest period of tubers. On the other hand the normal resting period can also be shortened by using ethylene chlorohydrin. In general, the total content of auxin can be divided into two types, free and combined. The belief prevails that the content of auxin not only varies in quantity during storage but that the concentration also varies in the different parts of the potato. Prior to sprouting of the potatoes in the spring an increase in auxin has been observed in the fleshy part of the potato while later the auxin, or its precursor, increases in the potato peel. The change of the precursors into auxin is generally assumed to be enzymatic in origin. One of the chief precursors may be presumed to be the amino acid tryptophane because auxin is a derivative thereof. In addition, it has been shown that the amount of biosynthesis of auxin in a medium depends upon the content of tryptophane in the medium.

(6) Respiration. In a series of four articles from the hygienic laboratory at the University of Michigan, Novy and coworkers[21,22,23,24] studied microbic respiration by means of the Novy respirometer. A study was made of the respiration in potatoes which showed a maximum gas production at or about 31°C.[24] The mechanism of respiration was destroyed after autoclaving at 40°C for 15 minutes.

The potato respiration utilizes oxygen in the Novy apparatus and carbon dioxide accumulates. Fresh slices of potatoes were transferred aseptically from tubers to the previously equilibrated Novy apparatus and at temperatures between 31 to 39°C a negative pressure developed. At 31°C the pressure dropped to -10 mm mercury after 19 hours; thereafter, pressure increased with time.

The effect of gamma radiation on fresh carrots, sealed raw in cans in a western-style closing machine in the Fission Products Laboratory, was studied in preliminary investigations begun in March, 1953.[25] Dosages applied varied from 1,200,000 to 1,900,000 rep. Subsequent to irradiation the cans were stored at room temperature and observed at intervals for excessive gas pressure over a period of 4 days. During the irradiation, which took 15 hours, a slight but noticeable pressure developed within the sealed can. This pressure increased with time until after 4 days the seams began to break and the experiment was discontinued. After the can was opened the irradiated carrots were observed to have darkened in color and softened in texture as compared with nonirradiated controls stored for the same period at room temperature. In a single experiment a No. 10 can of carrots was opened and the accumulated gas was carefully transferred to a gas analyzer in which there was carbon dioxide and nitrogen. This substantiated the early work of Novy who found the accumulated gas in the respirometer to be carbon dioxide. The evidence of a high gas pressure occurring within the cans containing irradiated carrots led to studies on other vegetables, especially potatoes, for the purpose of correlating the irradiation dosage with gas production. Preliminary work on potatoes canned, irradiated, and stored in the same manner as described for carrots, showed a developing gas pressure within the can of approximately the same rate and magnitude.

A series of investigations was next begun in which approximately 4 gm of aseptically obtained cylinders from a single, large, irradiated Idaho potato were transferred to the Novy apparatus for the determination of gas pressures at regularly scheduled intervals. In each single experiment sections of the potato cylinder came from an individual potato. Potatoes used in these experiments were irradiated with dosages varying from 200,000 to 2,000,000 rep. After the samples had been prepared and transferred to the Novy apparatus and the sample tube connected to the manometer, the apparatus was placed in an incubator at 37°C together with another apparatus containing the nonirradiated control. Gas pressures were determined after temperature equilibrium was obtained.

At the lower dosage levels gas pressures were highest decreasing with increasing dosages. To illustrate, the total pressure developed after a dosage of 250,000 rep was 180 mm mercury, while after a dosage of 2,000,000 rep the pressure was only 14 mm mercury. In the control a negative pressure (representing the oxygen uptake) was observed over the first 40 hours followed by a production of gas resulting in positive pressures. This characteristic lag observed in all controls was not noted with irradiated potatoes except for the very lowest dosages where a very slight negative pressure developed in the first few hours. The negative pressure obtained with incubated, nonirradiated control potatoes was approximately of the same magnitude as those obtained by Novy.[24] The greatest gas production was recorded with potatoes given a dosage of 250,000 rep. Plotting gas pressure versus time for this treatment gives almost a straight line up to a pressure of 180 mm after which the curve flattens out. After a dosage of 2,000,000 rep the curve reached a maximum of 13 mm mercury within 4 hours and then leveled off. This significant difference between gas pressures produced with nonirradiated control and low dosage irradiated Idaho potatoes requires further study. However, it appears evident that higher dosages of gamma radiation affect the activity of enzymes associated with respiration in potatoes.

Another important manifestation in potatoes exposed to gamma radiation was the continuous and progressively increasing darkening during incubation. The degree of darkening was most noticeable in potatoes exposed to the highest gamma radiation. Nonirradiated control potatoes showed no change in color over a similar period of time.

Sussman[26] recently reported some very significant data on the respiration of potatoes before and after exposure of 100 to 3,200,000 rep gamma irradiation. The data were obtained on freshly harvested, small, whole, Irish cobblers in a modified Warburg apparatus. An increase up to sixfold was observed in the oxygen uptake almost immediately after irradiation followed by a decrease after the first day. The increase in carbon dioxide reached a maximum a little later than for oxygen; but later the decrease was more or less parallel to that for oxygen. With the highest dosage, 3,200,000 rep, the potatoes darkened and softened after 3 days and the respirometric measurements indicated severe disturbances in metabolism. This attenuation was not due to destruction or inactivation of cytochrome oxidase whose activity was reduced at approximately the same rate. However, the activity of tyrosinase decreased after the potatoes had begun to blacken subsequent to the exposure to 3,200,000 rep.

(7) Decay. The chemical processes involved in the decay or rotting of potatoes are not definitely known. Most probably the decay is a combination of decomposition by bacteria and fungi in the exterior layers and an enzymatic breakdown of storage products in the interior. Pasteurization dosages of gamma rays appear to be capable of severely damaging the skin surface of tender fresh fruits, such as berries, thereby facilitating action by bacteria and fungi. However, the dosages applied to potatoes have apparently not significantly changed the structure of the potato surface. Moreover, from observation of the Minnesota-grown, Irish cobbler potatoes in the present storage experiments both processes evidently are involved either singly or in combination. Some of the heavily sprouted potatoes had decayed on the surface and slightly below, while the fleshy interior portion remained normal in both appearance and texture. Other control potatoes in a similar state of sprouting showed no evidence of decay on the surface and slightly below, but the central interior portion had decayed to an extent of 50 to 75 percent of the total area. These observations were made on July 26, 1954, when only three normal control potatoes were left after 6 months of local storage. At that time those potatoes with healthy interiors were used for sampling.

The chemical processes involved in sprouting, which frequently precedes decay, are not definitely known. As the sprouts are actually a vegetable proliferation originating in the region of the "eyes" of the tubers, the assumption may be made that the chemical compostition of tubers and their sprouts will differ. In contrast to the tuber the sprouts themselves, with a relatively high surface area, have a high rate of respiration and metabolism in addition to the actual growth. This creates a greater demand for transportable and utilizable sugars than is the case with tubers. The sprouts also have a lower starch content and a greater moisture content than the tubers. As mentioned previously Threadway[15] found that sprouts have a high sucrose content and a low starch content.

(8) Analysis. The solanine content of potato tubers has been studied by Wolf and Duggar[27] who found the concentration greatest in the peripheral zone including the periderm and cortical parenchyma. It appears that solanine is found in greatest concentration in the "eyes" and during storage there is an accumulation of solanine in the sprouts. Solanine, a glucoside containing the alkaloid solanidine and glucose, galactose, and rhamnose, has hemolytic properties and hence is of some importance to public health.

"The Chemistry and Technology of Food and Food Products" by A. F. Lee[28] gives the approximate composition of vegetables. For potatoes the following percentages are given: water 78.8, protein 2.0, fat 0.1, ash 0.99, total carbohydrate 19.1, fiber 0.4, sugar 0.9, and starch 14.7. These figures indicate that approximately sixteen percent of the carbohydrates are unaccounted for. There are great gaps in present knowledge concerning the distribution and concentration of individual carbohydrates present in the potato tuber.

3. EXPERIMENTAL STUDIES ON STORAGE OF IRRADIATED POTATOES AND ONIONS

a. Minnesota-Grown, Irish Cobbler Variety Potatoes

The investigations of Sparrow and Christensen[1] which provided the impetus for the taste panel studies on potatoes at the University of Michigan were not brought to the attention of the Fission Products Laboratory until January, 1954. By that time the normal potato storage season had already been underway for months. It was then impossible to inaugurate an extensive series of experiments with potatoes of known history. However, a small quantity of mature stored potatoes was obtained from the Food Service of the University of Michigan for use in taste panel studies. These potatoes were Minnesota-grown, Irish cobbler variety.

The potatoes were placed in storage at Food Service January 12,1954, at 50°F and 50 percent humidity. Small numbers of potatoes, estimated to be about 30, were placed in a series of polyethylene bags. Four groups, each consisting of several bags of potatoes, were given the following dosages of gamma radiation: (1) Control, no irradiation, (2) 7,000, (3) 21,000, and (4) 28,000 rep.

Observations on the keeping qualities were made periodically and potatoes were inspected at scheduled intervals during storage for taste-panel studies reported in a previous section of this progress report. Decaying potatoes were removed from the bags from time to time, but the first real total count and number of decayed potatoes were recorded July 14, 1954.

Sprouting occurred in nonirradiated control samples after 3 weeks of storage. After 4-1/2 months of storage, during which period many taste-panel tests were made (see Part III, A), the control potatoes showed sprouts up to 2 inches in length. After 5 1/2 months taste-panel studies were discontinued as a result of the scarcity of control potatoes. Those remaining were considered to be sour and spoiled by members of the taste panel.

The small number of control potatoes remaining in storage July 14, 1954, may be explained by the fact that more control potatoes had to be removed for comparison in taste panel work than any from each of the treated groups, and also because the rate of decay in the control group was greater than in the treated groups. It will be noted from Table 33 that on July 14, the percentage of decaying potatoes present in the control group was five times that in any of the treated groups and on July 26, the rate of decay was approximately twenty times as great. On this date the few remaining control potatoes were used for analytical determination, thus terminating the control group.

The physical appearance of the control potatoes was very poor and may be described as being mottled, wilted, shrunken, and decaying. A few spots of discoloration due to fungi were also noticed. The appearance of the treated potatoes was markedly better in that the texture was much firmer, the water loss less, and in general fewer blemishes on the skin of the potato. Very few of the treated potatoes have ever sprouted, whereas the greater percentage of the control potatoes sprouted, and then the sprouts died and withered. As could be expected, the physical appearance became progressively less desirable with time.

Figure 30 shows the exterior view of two controls with decayed sprouts and one potato from each of the three irradiated groups after storage for 6 months. In this case the controls first developed sprouts having a length of 2 or more inches. These sprouts then died and decayed. The fleshy part of the tubers then began to decay. The interior view, (Fig.31) showing the decay of the two control potatoes compared to the treated samples indicates clearly the decomposition of the nonirradiated potatoes.

b. Idaho-Grown, Burbank Russet Potatoes

Because treatment by gamma radiation had shown great promise by inhibiting sprouting and increasing the storage life of Minnesota potatoes, taste-panel studies and observations on keeping qualities were extended to include Idaho potatoes. The Idaho-grown potatoes were placed in storage in the same cold room under the same conditions as the Minnesota potatoes. Four groups of potatoes, each comprised of one bag of 100 lb, were treated in May, 1954, as follows: (1) Control, no irradiation, (2) 7,000, (3) 21,000, and (4) 28,000 rep.

TABLE 33

MINNESOTA-GROWN, IRISH COBBLER VARIETY POTATOES STORED IN PERFORATED POLYETHYLENE BAGS AT 50°F AND APPROXIMATELY 50 PERCENT HUMIDITY*

Date of Examination 1954	Treatment, rep	Total No. Potatoes	No. Decayed	% Decay Between Inspections	Accumulative % Decayed	No. Remaining in Storage	Remarks
14 July	Control	42	19	45	45	23	Controls in a poor condition
	7,000	128	10	8	8	118	
	21,000	76	7	9	9	69	
	28,000	90	7	8	8	83	
26 July	Control	23	19	83	91	4	These 4 used for sampling.
	7,000	111	3	3	11	108	
	21,000	69	3	5	14	66	
	28,000	76	3	4	11	73	
17 August	Control	0	0	0	100	0	
	7,000	108	4	4	15	104	
	21,000	66	2	3	16	64	
	28,000	73	3	4	16	70	
17 September	Control	0	0	0	100	0	
	7,000	104	7	7	21	97	
	21,000	51	1	2	18	50	
	28,000	70	3	4	19	67	
13 October	Control	0	0	0	100	0	
	7,000	97	9	9	27	88	
	21,000	50	12	24	38	38	
	28,000	35	3	8	26	32	

*Placed in storage in polyethylene bags at Food Service January, 1954. No record was kept of total number at the beginning or of the number decayed and removed before a complete examination and count was made on July 14, 1954.

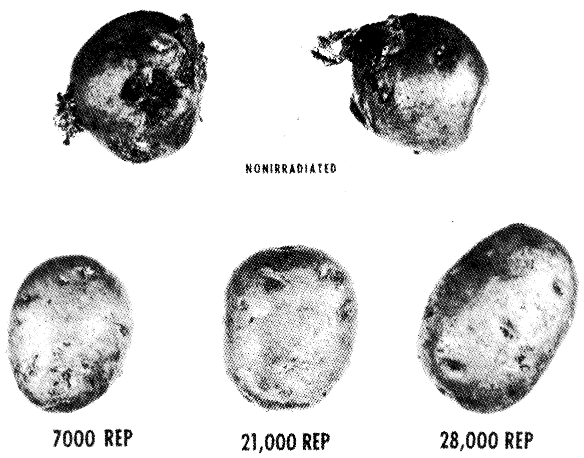

Fig. 30. Comparison of Exterior Appearance of Nonirradiated and Irradiated Minnesota-Grown, Irish Cobbler Variety Potatoes Showing the Decay of Sprouts on the Controls.

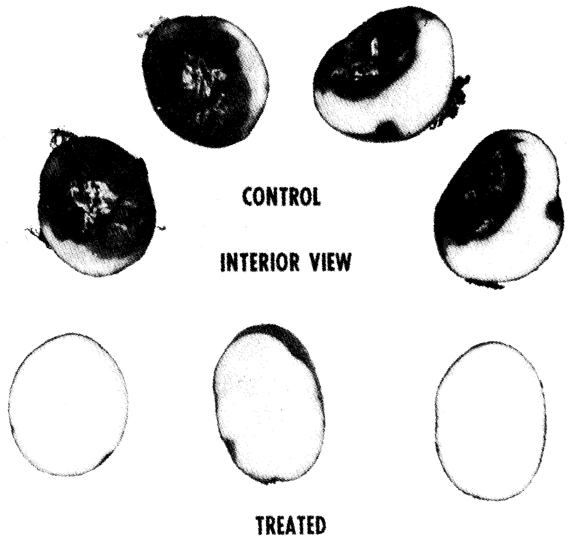

Fig. 31. Comparison of Interior Appearance of Nonirradiated and Irradiated Halved, Minnesota-Grown, Irish Cobbler Variety Potatoes Showing the Decay in Nonirradiated and Normal Appearance in Irradiated Specimens.

After three week's storage in the cold room the nonirradiated potatoes were beginning to sprout. On July 16, 1954, the potatoes in each of the four bags were counted and the decayed ones removed. The nonirradiated controls had all sprouted, some to a length of three or more inches, while the irradiated potatoes showed only the small proliferation at or around the "eyes" which existed in May, 1954, when these potatoes were irradiated. The sprouts on the control potatoes were exceedingly brittle and did not withstand rough handling. During the process of removal from the burlap bags, counting, sorting, and returning the potatoes to the bags, the sprouts broke off. These potatoes did not develop new sprouts of any noticeable length, although a limited proliferation in the region of the "eyes" was noted during subsequent storage.

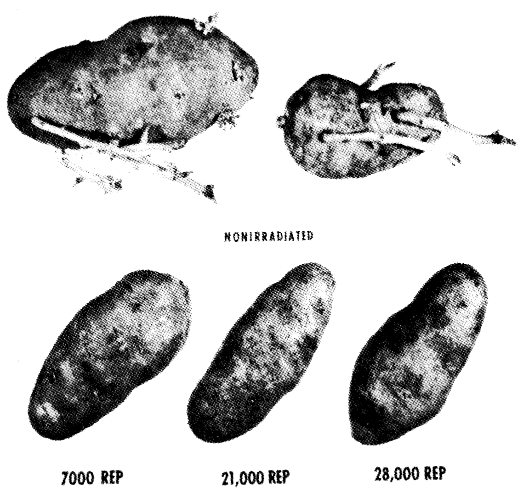

Fig. 32. Comparison of Exterior Appearance of Nonirradiated and Irradiated Idaho-Grown, Russet Variety Potatoes Showing Sprouts on the Controls.

In a few potatoes which were purposly handled with care the sprouts did not break off. The contrast in appearance of nonirradiated versus irradiated potatoes is shown in Fig. 32, taken July 16, 1954.

The long brittle sprouts which developed on the controls were, for the most part, broken off in handling. Table 34 gives a summary of the data on storage.

Figure 35 in the following section (Part III, C) shows controls which were carefully handled and kept in a refrigerator at about 45°F until September, 1954. In this case there was extensive sprouting of the controls, but no sprouting of the irradiated potatoes held under the same conditions. Table 34 indicates that the mechanical removal of sprouts in handling resulted in good storage life of the controls. This is contrary to the results observed with the Minnesota-grown potatoes. This may be a result of the difference in the properties of the two varieties or a result of difference in previous treatment during storage. Unfortunately, the history of these potatoes prior to purchase through the Food Service was unknown. It is possible that the Idaho-grown potatoes had received a chemical treatment.

In July, practically all of the untreated, control Idaho potatoes had sprouted, whereas the irradiated potatoes in all dosages,with the exception of a few in each group, did not sprout. Therefore, it is concluded that the desirable effect of gamma radiation preventing sprouting was demonstrated.

c. White Pearl Onions

The onion is another vegetable which deteriorates in storage due to sprouting and subsequent decay. Therefore, the decision was made to extend taste-panel studies and observations to include the rate of sprouting and decay in onions. Without knowing what the desirable dosages might be for onions, the decision was made to use the same as was previously used with success for potatoes.

Again, four approximately equal quantities of White Pearl Onions were selected for study, arranged into four separate groups, and given the gollowing treatment: (1) Control, no irradiation, (2) 7,000, (3) 21,000, and (4) 28,000 rep.

The onions in each group were distributed and placed in Kraft paper bags and placed in storage at the Food Service at 50°F and 50 percent humidity. These storage conditions were used because the space was available. However, the percentage of humidity was not optimal if judged upon the basis of molds which caused discoloration of the surface of many onions.

On July 1, 1954, after 3 months of storage, the rate of sprouting was approximately equal in all of the four groups. The sprouted onions were removed at that time. No record was made of the percentage of sprouting

TABLE 34

IDAHO-GROWN, BURBANK RUSSET VARIETY POTATOES
STORED IN BURLAP BAGS AT 50°F AND APPROXIMATELY 50 PERCENT HUMIDITY*

Date of Examination 1954	Treatment, rep	Total No. Potatoes	No. Decayed	% Decay Between Inspections	Accumulative % Decayed	No. Remaining in Storage
16 July	Control	169	2	1	1	167
	7,000	194	1	< 1	< 1	193
	21,000	197	1	< 1	< 1	196
	28,000	181	3	2	2	180
17 August	Control	167	2	1	2	165
	7,000	193	16	8	9	177
	21,000	196	14	7	8	182
	28,000	178	25	14	15	153
17 September	Control	156	1	< 1	3	155
	7,000	173	3	2	11	170
	21,000	161	3	2	10	158
	28,000	148	6	4	18	142
13 October	Control	130	0	0	3	130
	7,000	105	6	6	16	99
	21,000	158	0	0	10	158
	28,000	142	8	5	22	134

*Placed in storage in burlap bags (100 pounds each) at Food Service May, 1954. No record was kept of total number in each bag or the number decayed and removed. First complete counts were made July, 1954.

and decaying onions until August 17. On that date, the rate of sprouting was approximately equal in the nonirradiated control group and the group receiving 28,000 rep, but it was lower in the two other groups as is shown in Table 35. However, on that date the rate of decay was lowest in the control group. During the next 2 months the treated onions showed a marked lower rate of sprouting and decay as evidenced by the data recorded in September and October. The appearance of mildew seemed to be equally distributed in all of the groups. In October the sprouts on the onions showed signs of wilting and decay and only a very few had green living sprouts. Attention should also be given to the fact that onions on the surface may show a great deal of discoloration indicating decay; yet this, in many instances, is not the case as proven by observation of halved sections. The sprouted onions showed general deterioration in structure and firmness of texture as described previously under potatoes.

These preliminary studies on White Pearl Onions have shown that the optimal gamma radiation dosages for the prevention of sprouting and decay in storage may still have to be ascertained but under the conditions of these studies low doses of gamma radiation decreased the rate of sprouting.

d. Yellow Dry (Bermuda) Onions

Another variety of onions, Yellow Dry (Bermuda), was next studied because of its wide use. Four groups of onions were again selected, each group consisting of two bags of onions with 50 lb in each mesh bag. These onions were placed in storage in May, 1954, at Food Service at 50°F and 50 percent humidity after having been exposed to the following gamma radiation dosages: (1) Control, no irradiation, (2) 7,000, (3) 21,000, and (4) 42,000 rep.

A dosage of 42,000 rep in place of the usual 28,000 rep was used because the Bermuda variety apparently is of a coarser structure than the White Pearl and might, for that reason, withstand this higher dosage without any superficial deleterious changes in membrane permeability in its outer layers.

The rate of initial sprouting was found to be high in all four groups but by far the highest was in the control during the months of August and September as shown in Table 36. By October practically all of the controls had sprouted. There was a gradual increase in the percentage of sprouting in all four groups with time in storage. From the percentages of sprouted onions in the irradiated groups it appeared that the 7,000 rep

TABLE 35

WHITE PEARL ONIONS KEPT IN STORAGE IN KRAFT PAPER BAGS AT 50°F AND APPROXIMATELY 50 PERCENT HUMIDITY*

Date of Examination 1954	Treatment, rep	Total No. Onions	No. Sprouted	% Sprouted	No. Decayed	% Decayed	No. Remaining
17 August	Control	180	37	20	4	2	176
	7,000	161	22	14	9	6	152
	21,000	155	15	10	19	12	136
	28,000	149	32	21	13	9	136
22 September	Control	167	30	18	22	13	145
	7,000	152	16	11	9	6	143
	21,000	123	13	10	9	7	114
	28,000	134	15	11	11	8	123
14 October	Control	145	24	17	12	8	133
	7,000	141	10	7	5	4	136
	21,000	114	3	3	9	8	106
	28,000	115	15	13	8	7	107

*Placed in storage in Kraft paper bags at Food Service May, 1954. No record was made of total number at the beginning or of the number decayed or sprouted before August 17, when a complete examination and count were recorded.

TABLE 36

YELLOW DRY (BERMUDA) ONIONS STORED IN MESH BAGS
AT 50°F AND APPROXIMATELY 50 PERCENT HUMIDITY*

Date of Examination 1954	Treatment, rep	Total No. Onions	No. Sprouted	% Sprouted	No. Decayed	% Decayed	No. Remaining
18 August	Control	217	135	62	4	2	213
	Control	218	137	62	9	4	209
	7,000	221	28	13	7	3	214
	7,000	217	64	30	4	2	213
	21,000	178	85	48	2	1	176
	21,000	207	82	40	2	1	205
	42,000	221	117	53	1	< 1	220
	42,000	204	90	44	1	< 1	203
22 September	Control	207	172	83	21	10	186
	Control	193	151	80	28	15	165
	7,000	194	68	35	13	7	181
	7,000	210	34	16	12	6	198
	21,000	176	87	15	2	1	174
	21,000	198	130	66	13	7	185
	42,000	185	143	77	8	4	177
	42,000	200	132	66	8	4	192
25 October	Control	172			20	12	152
	Control	155			39	26	116
	7,000	192			39	20	153
	7,000	180			27	15	153
	21,000	174			20	11	154
	21,000	181			21	12	160
	42,000	192			12	6	180
	42,000	170			29	17	141

*Placed in storage May, 1954 in mesh bags at Food Service. No record was available of total number at the beginning or the number decayed or sprouted before August 18 when a complete examination and count were made.

dosage was the best dosage for this purpose. All irradiated onions remained firm and dry regardless of whether or not they had sprouted, whereas the control onions became moist and soft. Sprouts in the irradiated onions did not grow to a length exceeding 1-1/2 inches, while the sprouts in the controls grew to a length of 6 to 8 inches.

Another difference noted was the marked decrease in volume of the parent, control onion supporting the growth of the very long and numerous sprouts. On the other hand, sprouted, irradiated onions had decreased very little in volume and the texture remained firm and solid. The outer layers of the irradiated onions dried and fell off during storage but in the controls these leaves remained in place and became wet and clammy to the touch.

The contrast in appearance of the control and irradiated Bermuda onions is shown in Fig. 33, which was taken in the storage room at Food Service in October, 1954. It shows the length of the sprouts on the control onions which protruded several inches through the mesh bags.

Fig. 33. Comparison of Irradiated and Nonirradiated Bermuda Onions Showing the Sprouts on the Control Onions Protruding Several Inches Through the Mesh Bags.

In summarizing the comments on the storage experiments, the general conclusion can be drawn that a 7,000 rep dosage of gamma radiation for practical purposes inhibits sprouting and markedly increases the storage life of potatoes. The effect of gamma radiation on onions in the dosages administered is less precise and decisive but the inhibitive effect on the incidence and length of sprouts has been clearly demonstrated. Although low dosages of gamma radiation may not completely prevent the sprouting of onions it prevents the continued growth of sprouts, thereby keeping the onion more firm and increasing the storage life.

4. BIOCHEMICAL TESTS ON GAMMA IRRADIATED POTATO TUBERS

a. Analysis of Sugars in Stored Irradiated and Control Potato Tubers

The Fission Products Laboratory did not commence extensive studies on the gamma irradiation of potatoes until January, 1954. This precluded an immediate experiment to determine the quantities of and changes in the carbohydrates present in the potato over the entire storage season.

Without adequate and sufficient chemical data determined immediately after the potatoes had been harvested, it is impossible to establish any basal level of concentration, especially in view of the fact that the content of starch increases and sugars decrease immediately upon storage of potatoes harvested before maturity. That makes it mandatory either to know when the potatoes were harvested in the autumn and how they were stored prior to purchase by the laboratory, or to run chemical determinations extending several weeks prior to gamma irradiation. As a compromise, preliminary chemical determinations were made to ascertain the quantities of reducing sugars present in raw potatoes before and after incubation with invertase and, by the difference in the two, estimate the amount of sucrose present.

For completeness and accuracy, determinations of the existing quantities of moisture, reducing sugars before and after incubation with invertase, total carbohydrates, and starch should be performed simultaneously at least once every 2 weeks before irradiation until a reasonably accurate preirradiation level is apparent. Subsequent to irradiation the same determinations should be made daily for the first week and twice weekly for the first few weeks thereafter in order to detect early induced changes caused by gamma irradiation. After the initial effects from the irradiation seem to produce no further significant changes in concentration of the constituents to be determined, analytical determinations every two weeks during the storage season should suffice.

In the studies on potatoes in the Fission Products Laboratory the analytical work was limited to determinations of reducing sugars primarily because of lack of time and funds. The sugar concentrations are known to be relatively low and subject to fluctuations induced by the varying needs of the potato as well as by conditions prevailing during the period of storage. Further, the content of starch is high and changes, either by amylosis or resynthesis; these changes would be small percentagewise. In addition, it has been postulated that potatoes lose carbohydrates and water at approximately the same rate under favorable storage conditions.[15] For that reason moisture determinations were not included in these preliminary studies. There exists a dynamic, not static, equilibrium between carbohydrates, fats, and proteins in the potato, subject to fluctuations.

The analytical procedure used for the determination of reducing sugars was the "official" method[29] in a modified form[30] to permit working with a prepared potato slurry instead of dried samples. The modification is in part based on the directions of Somogyi.[31] The reducing sugars were determined by titration with approximately 0.005N thiosulfate made up fresh daily by dilution of a stock solution of known normality.

All of the reagents, prepared from the highest quality chemicals available, were first used for the determination of known solutions of highest quality d-glucose. A titration curve for the dilute sodium thiosulfate solution in terms of milligrams of glucose was constructed. Through the courtesy of General Foods[30] a dried potato sample of known sugar concentration was provided for checking on the analytical procedure.

Determinations of reducing sugars before and after incubation with invertase (Difco) were performed on nonirradiated and irradiated (7,000, 21,000, and 28,000 rep) Minnesota-grown potatoes which had been kept in local storage since January, 1954. Owing to the small quantity of potatoes, especially of controls, only four raw representative potatoes were taken for samploing. These potatoes were sliced into halves and carefully peeled in order to minimize losses of flesh layers directly beneath the peel and then cut into small pieces. These raw potato pieces were next randomly mixed on a large piece of paper. A known quantity, usually 300 gm, was placed with an equal weight of distilled water in a small size Waring Blendor. The potato pieces and water were then thoroughly mixed until no further subdivision of the potato particles could be observed. It was noted that the prepared slurry rapidly turned dark brown and that the potato particles settled out from the aqueous suspension almost immediately. By means of a 15-ml pipette, with the tip broken off to permit rapid filling, a quantity of slurry, usually about 15 gm, was immediately transferred to a previously weighed 100ml volumetric flask. The exact amount of slurry was determined by difference on an analytical balance. Because the slurry suspension settles out very rapidly, it is necessary to remove the samples immediately after a thorough swirling of the container. Duplicates were taken from all examined potato slurries and all sugar determinations on these duplicates were run in triplicate. Blanks were always run simultaneously with the sugars in all titations.

The analytical data presented in Table 37 were obtained during the month of July when the potatoes had been in local storage for 6 months after irradiation. All of the chemical determinations were performed in laboratory space furnished by the Department of Biological Chemistry, the University of Michigan Medical School.*

TABLE 37

COMPARATIVE ANALYTICAL DATA FOR REDUCING SUGARS BEFORE AND AFTER INCUBATION WITH INVERTASE FROM NONIRRADIATED AND IRRADIATED RAW MINNESOTA POTATOES STORED AT 50°F AND 50 PERCENT HUMIDITY SINCE JANUARY, 1954

Date 1954	Treatment, rep	Reducing Sugar,%		Sucrose %
		Before Incubation	After Incubation	By Difference
July 15	Control	1.89	2.93	0.99
July 17	7,000	1.22	1.82	0.57
July 21	21,000	1.30	1.55	0.23
July 12	28,000	1.39	1.87	0.45
July 26	Control	1.64	2.00	0.32
Average	2 controls	1.77	2.47	0.65
Average	3 irradiated	1.30	1.75	0.42
Difference	Numerical	0.47	0.72	0.23
	Percent	26.6	29.1	35.4

b. Change in Sugar Content of Irradiated Potato Tubers (10,000 rep) Directly Following Irradiation

(1) Introduction. In a recent highly significant article Sussman[26], using a modified Warburg apparatus, has definitely shown a five to sixfold increase in the oxygen uptake of Irish cobbler potato tubers exposed to gamma radiation. The potatoes used were tested within 60 days

*The Fission Products Laboratory gratefully acknowledges the use of these laboratory facilities.

after harvesting and were, therefore, most probably in a dormant state. The gamma radiation dosages used varied between 100 and 3,200,000 rep. The maximum effect on the oxygen uptake was observed in the range of 10,000 to 400,000 rep. The increased oxygen uptake was discernible within a few hours after irradiation reaching a maximum effect 24 hours after zero time for the 10,000 rep dosage followed by a steep decline until the fourth day and with a gradual return to the pretreatment level after 12 days.

The tremendous and rapid increase in oxygen uptake subsequent to gamma radiation is indicative of a great stimulus to the metabolism of potato tubers which would of necessity involve a similarly and simultaneously increased demand for transportable, utilizable high energy-containing compounds. In potatoes the chief storage product is starch which is in an equilibrium with the end product of amylosis, glucose. With the increased need for energy due to the gamma ray exposure the rate of amylosis must be increased by enzymatic breakdown of starch which results in more glucose being formed.

Inasmuch as it has been shown and reported in a previous section that the content of reducing sugars is lower in nonsprouted gamma irradiated potatoes than in sprouted nonirradiated potatoes, a state in which there also is a greater demand for utilizable sugars, consideration was given to an investigation in order to ascertain the following two points of interest.

1. Is there an increase in the content of reducing sugars in potato tubers after being exposed to small dosages of gamma radiation?

2. If there is an increase in the content of reducing sugars after subjecting potato tubers to small dosages of gamma radiation, is this increase almost immediate and parallel to the increased rate of respiration observed by Sussman[26].

(2) Experimental Procedure. A short-term experiment was designed in which one 100-lb bag of Idaho potatoes was kept as a control at Food Service at 50°F and 50 percent humidity since May, 1954. The bag was sampled (6 potatoes per sample) and the reducing sugar content was determined before and after incubation with invertase according to a modified Somogyi[30,31] method described in detail in a preceding section of this report. Determinations of the sugar content were first run on the nonirradiated potatoes in order to establish a base level. In other tests with individual potato samples the sugar content was determined at

a measured time interval after the completion of irradiation. A radiation dose of 10,000 rep was given in a period of 60 minutes by placing the potatoes at a distance of 11 inches from the periphery of the center well of the cobalt-60 source. Analyses were made of samples stored for 0, 1, 3 1/2, 5, 15, 25, 48, and 72 hours after irradiation.

Zero-hour standing was arbitrarily set as the exact time at which the gamma radiation was completed and analysis commences despite the fact that enzymatic activity presumably continued uninhibited in the potato tubers during the subsequent peeling, slicing, subdivision, and mixing in the Waring Blendor, and sampling and weighing out of slurry aliquots. Enzymatic activity was probably halted when these samples were diluted with ethyl alcohol and refluxed on the steambath approximately 2 hours later. However, the data obtained show that the first very minor increase in the quantity of reducing sugars did not occur until after 5 hours standing (on the basis of the above interpretation).

It should be kept in mind, however, that whereas Sussman[26] used freshly harvested potatoes in his experiment, the potatoes used in the experiments reported here had been stored for approximately 1 year and only under known conditions since May, 1954.

(3) Discussion. Owing to the fact that funds were not available to support this specific study only a very limited number of data could be made available. The data from this study, however, are believed to be of interest especially to those engaged in basic research because they indicate an effect, direct or indirect, of gamma radiation.

Regarding the sugar content of stored, irradiated, and control potatoes Table 37 shows that lower sugar values were obtained in the second control sample. These values possibly reflect the greater degree of decay in these potatoes which resulted in greater peeling losses. The potatoes used for the second control sample were in a state of greater decay and although the sprouts were still on the potatoes they appeared to be shrunken, nonliving tissue. With the flesh of the tuber itself barely in a living state, the need for a high concentration of sugars would be reduced, but the possibility that the decay involves decomposition of starch still exists.

Sugar concentration normally is increased when sprouting occurs, a process which necessitates a translocation of carbohydrate from tuber to sprouting tissue in a transportable or diffusible form, thus the higher values for reducing sugar in the controls than in gamma irradiated potatoes should be no surprise. The irradiated, nonsprouted potatoes have a lesser need for transportable sugar inasmuch as little or no new sprout tissue is being formed. While it may be incorrect to use average values for such limited data from the viewpoint of statistical analysis, the quantitative differences are of a magnitude which probably should not be overlooked. The trend is definitely indicated under the conditions of these experiments.

In the study on potatoes some members of the taste panel observed a sweeter taste in sprouted control samples than in the nonsprouted irradiated potatoes. If the sweetness in the potato is due to the content of sucrose, then the taste panel observation substantiates the analytical data which shows higher values for sucrose in the control after long storage.

Immediately after irradiation the members of the taste panel showed no unanimous preference for either nonirradiated of irradiated potatoes. However, some of the judges noted, besides a slight burned taste, a sweeter taste in potatoes exposed to the two highest dosages of gamma radiation when compared with the controls. If this sensory observation is correct, the gamma radiation at the two higher dosages could possibly have had an immediate but short lasting effect in favoring the formation of sugars by amylosis. Unfortunately, analytical data are not available for comparison.

The retarding effect of gamma radiation on sprouting in potatoes, first published by Sparrow,[1,2] has definitely been confirmed. In the preceding pages of discussion an attempt has been made to correlate analytical data and observations made by some members of the taste panel with the effect of gamma radiation. Chemical data confirm that sugar concentrations in sprouted controls after 5 to 6 months storage are greater than those sound in the irradiated potatoes. Gamma radiation may have had an immediate effect favoring the reaction starch to sugar. There can be little doubt on the basis of data here reported however, at least after 5 to 6 months storage, that one of the effects of gamma radiation has been to prevent the increase in reducing sugars and sucrose observed in the sprouted control. Since control and irradiated potatoes have been stored under identical conditions, it may be asserted that not only did gamma radiation retard sprouting in potatoes, but also the increase in sugar concentration in the control, generally associated with sprouting, was delayed if not inhibited in the irradiated potatoes.

Regarding the changes in sugar content directly following irradiation, attention is again directed to the fact that Sussman conducted his studies on freshly harvested Irish cobbler potatoes, whereas the data presented in this study were obtained on Idaho-grown potatoes (Burbank russet variety) which had been stored almost a full year.

Sussman plotted his oxygen uptake against time in days after irradiation without any intermediate points during the first 24 hours. For that reason sugar determinations in the present studies were run on separate, individual samples after 0, 1, 3 1/2, 5, 15, 25, 48, and 72 hours of standing after irradiation in order to ascertain, if possible, how immediate the effect of gamma radiation might be. Data in Table 38 and Fig. 34 show that sugar concentration actually fell slightly, when compared against the basal level, during the time lapse of irradiation and until all enzymatic action ceased on the steam bath. This in itself is not surprising on the basis that there ia an immediate and greatly increased stimulus to the metabolism of the potato because of the greatly increased oxygen uptake and with concomitant greater demand for sugars. Further, on the assumption that there is a certain period of time required before the enzymatic systems involved in amylosis can be stimulated to greater intensity and/or efficiency, an immediate decrease might be expected thus upsetting the normal equilibrium in the reversible reaction starch-glucose. If the effect of gamma radiation is not merely a phenomenon of a very short duration but extending over several hours or even days, then the prolonged increased need for extra energy, certainly in part furnished by sugar, might cause a temporary plethora in the gamma irradiated potato. If there is a definite reaction constant for the reversible reaction starch-glucose in the control potato under a given set of conditions, such as those under which the potatoes were stored at Food Service, there would appear to be no reason to expect that the reaction constant would remain unchanged after such a drastic treatment as that by intense, penetrating gamma rays.

TABLE 38

QUANTITATIVE DATA SHOWING CONTENT OF REDUCING SUGARS BEFORE AND AFTER INCUBATION WITH INVERTASE IN NONIRRADIATED AND IRRADIATED RAW IDAHO POTATOES STORED AT 50°F AND 50 PERCENT HUMIDITY SINCE MAY, 1954

Date 1954	Treatment, rep	Hours of Standing	Reducing Sugar % Before Incubation	Reducing Sugar % After Incubation	Sucrose % By Difference
September 30	Control	0	0.85	0.96	0.10
September 28	10,000	0	0.73	0.75	0.02
November 11	10,000	0	0.76	0.76	0.00
November 6	10,000	1	0.80	0.82	0.02
November 4	10,000	3.5	0.72	0.74	0.02
November 15	10,000	5	0.90	0.93	0.03
November 20	10,000	15	1.15	1.22	0.07
November 18	10,000	25	1.43	1.51	0.07
November 25	10,000	48	1.40	1.53	0.12
November 28	10,000	72	1.39	1.50	0.10

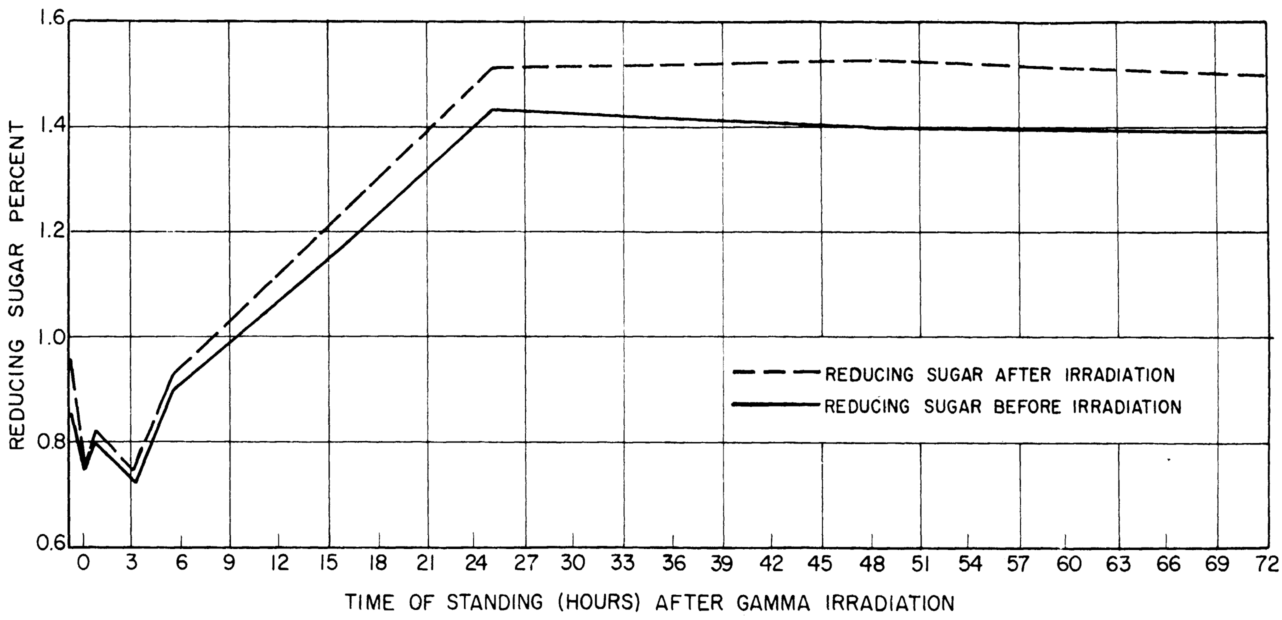

Fig. 34. The Influence of 10,000 rep Gamma Radiation on the Content of Reducing Sugar in Raw Idaho Potatoes

Percentagewise, the minor changes in sugar concentration shown after 0, 1, and 3 1/2 hours are very small and may not be significant. However, the increase after 5 hours most probably may be considered significant because it makes the first point on the curve above the basic level. In view of the still higher values after 15, 25, 48, and 72 hours, which when plotted give almost a straight line, it appears that under the conditions of this study the elevation in sugar content of the gamma radiated potatoes begins approximately after 5 hours of standing from zero time as previously defined. This marked increase after a few hours is of interest because Sussman[26] found an almost immediate effect on oxygen uptake amounting to an increase of sixfold within a few hours after irradiation.

On the basis of the values obtained after 5 hours standing subsequent to irradiation it would seem that a maximum concentration might have been reached somewhere between 15 to 48 hours standing, although this is not shown. Time did not permit the accumulation of additional data to complete the curve and to show the eventual complete return to basic pretreatment sugar concentration. The plotted data show a very even plateau extending for a period of at least 48 hours with perhaps a decline toward normal sugar concentrations beyond 72 hours.

The concentration of sucrose, indicated by the area between the two curves showing the concentration of reducing sugars before and after invertase, does not vary greatly. The higher values for sucrose are found where the reducing sugar concentrations were elevated above the normal level.

On the basis of the data presented in this short study and under the conditions of these experiments, the following conclusions may be drawn.

1. Gamma radiation dosages of 10,000 rep definitely cause a significant increase in reducing sugar concentration in Idaho potatoes.

2. Increased reducing sugar concentrations are discernible 5 hours after irradiation, reaching a possible maximum between 15 to 48 hours subsequent to irradiation.

3. The increase in reducing sugar concentrations are not as marked as the increase in oxygen uptake observed by Sussman; but in general the curve in this study follows the curve for oxygen uptake, although at a much lower level.

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C. DESIGNS FOR GAMMA IRRADIATION FACILITIES FOR POTATOES

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1. INTRODUCTION

It is believed that the gamma irradiation of potatoes may be the first commercial application of gamma radiation to foods. Therefore, consideration is given to the problems of design of gamma irradiation facilities for such use. This section covers the design of such facilities using three different sources of radiation: (1) cesium-137, (2) mixed fission products, and (3) cooling reactor-fuel elements. Cost studies are made for each design.

2. REVIEW OF THE PROBLEM

Potatoes are the most important vegetable food in the domestic diet. A greater tonnage of potatoes is handled than of any other vegetable produce. The volume sold is quite constant throughout the year and from year to year. Annual production during the 10-year period 1941 to 1951 averaged 415 million bushels with an average value of over $526,000,000.[1]

About 75 percent of potatoes are harvested in the fall and are known as late potatoes. The majority of these potatoes are grown in the northern states. The Snake River Valley of Idaho, the Red River Valley of Minnesota, Arvostook County, Maine, and Long Island, New York are well known for their production of potatoes. Dale County, Florida and Kern County, California also produce large quantities of potatoes, particularly for use in the early summer months when mature northern-grown potatoes are not available.

Home-grown northern potatoes cannot be stored on the typical farm for much more than 6 months. However, where better storage facilities are available northern late potatoes may be stored for 8 or 9 months.

Potatoes are perishable and the storage conditions are exacting if the potatoes are to be kept for many months. The Department of Agriculture recommends the following conditions.

"When potatoes are first placed in storage they should be held at a temperature of 60°F (taken at the top of the bin) and a humidity of 85 to 95 percent for about 10 days so that wounds will heal. Restricted ventilation will hold the humidity at this level. After the wound-healing period the storage area should be ventilated just enough to reduce the temperature at the top of the bin from 60 to 50°F in 45 days and for continued storage the top of the bin temperature should be reduced from 50 to 40°F during the next 45 days. More rapid cooling usually results in excessive shrinkage because of a lower storage humidity."[1] Potatoes to be held longer than 3 to 4 months should be stored at 40°F and a relative humidity of 90 percent after the initial cooling period but potatoes marketed within this time need not be stored at temperatures below 50°F.

Potatoes usually cannot be held in unrefrigerated storage later than early spring in even the northern states because of the seasonal temperature rise. This temperature rise following the period of dormancy results in rapid sprouting of the tubers. The use of refrigeration extends the storage period but even with refrigeration it is difficult to hold untreated potatoes later than May.

In addition to sprouting and rotting there are other problems in storing potatoes; such as maintaining a low sugar content, a problem which is primarily of importance to potato processors. About 10 percent of the potatoes sold for food are processed into products such as potato chips, dehydrated potatoes, canned potatoes, etc. In 1951 over 23 million bushels of potatoes were processed into potato chips.

Potato processors demand potatoes of medium to large size, high starch content, and low sugar content. The low sugar content is especially important to processors producing potato chips. Sugar in the potato carmelizes when fried producing dark or "burned" spots on the chips which are not desirable. Also, dehydrated potatoes having a high sugar content do not keep as well as dehydrated potatoes having a low sugar content.

If potatoes for processing are stored at too low a temperature, an accumulation of sugar in the potato results from the enzyme conversion of starch to sugar and from the low respiration rate at low temperatures.

Potatoes stored at a higher temperature will have a sufficiently rapid respiration to oxidize the sugar so as to maintain a low sugar content. As the low storage temperatures (32 to 40°F) result in excessive accumulation of sugar and the higher temperatures (50°F and above) result in excessive spoilage from sprouting and rotting, some compromise temperature such as 45 to 50°F may be used. Another method is the storage of the potatoes at the low temperatures followed by the raising of the temperature about 20°F above storage temperature for a period of 2 weeks or more prior to processing. Both of these methods result in increased spoilage from sprouting and rotting. Potatoes that have been held dormant at low temperature for a number of months sprout rapidly after the temperature is raised.

A variety of chemicals (Part III, Section B) have been found useful in treating potatoes to retard or prevent sprouting. However, the use of chemicals is associated with some disadvantages such as:

(1) difficulty in obtaining a uniform application
(2) nonuniform response of treated tubers
(3) need of special equipment to treat potatoes on a large scale
(4) plugging of ventilation ducts with dust
(5) possible toxicity of some of the chemicals
(6) inability to control sugar content of tubers by chemical treatment
(7) cost of some chemicals and some methods of application
(8) undesirable appearance of dust on potatoes in dusting process.

One large processor of potatoes in the Michigan area tried a chemical dusting treatment but then discontinued it. Chemical treatment cannot be considered to be completely satisfactory.

3. EFFECT OF GAMMA IRRADIATION ON POTATOES

Gamma radiation has been shown by Sparrow and Christensen to prevent the sprouting of small Katahdin variety potatoes for 18 months.[2,3] Tests in this laboratory with mature 1953-crop Minnesota-grown, Irish cobbler and Idaho-grown, Burbank russet varieties of potatoes have also shown the ability of low doses of gamma radiation to prevent sprouting. Several 100 lb and smaller bags of storage potatoes of these varieties were irradiated, respectively, in January, 1954, and May, 1954, and were stored at 50°F until September, 1954, at which time mature potatoes of the 1954 season had become available.

Figure 35 is a photograph taken in September, 1954, of Idaho-grown, Burbank russet variety potatoes of the 1953 crop. The irradiation of these potatoes was performed in May, 1954. Figures 30 and 31 (Part III, Section B) show external and internal appearance of Minnesota-grown, Irish cobbler

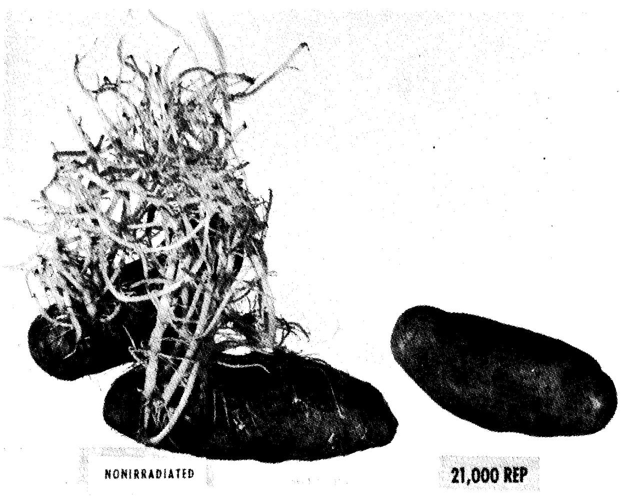


Fig. 35. Idaho-Grown, Burbank Russet Potatoes of the 1953 Crop. Irradiation Performed May, 1954. Stored at 50°F until Photographing September, 1954.

variety potatoes of the 1953 crop stored for 6 months at 50°F subsequent to irradiation in January, 1954. Note that the sprouts on the control tubers have died and withered and that they have undergone severe decomposition in the interior. By September these control tubers were completely rotten whereas the irradiated potatoes were still well preserved.

Over 2100 individual taste tests were made by a taste panel using irradiated potatoes and controls (see Part III, Section A). A statistical analysis of the results of these tests showed no preference for the controls over the irradiated potatoes. On the basis of these studies and some taste tests conducted at Brookhaven[3] there is no indication of a problem of off-flavor in potatoes as a result of irradiation in the dose range of 10,000 rep.

Tests by Sussman[4] on the respiration of irradiated potatoes have shown an increase up to sixfold in the respiration immediately following irradiation. This increase in respiration is observed without an increase in temperature. At present it is common practice to control the sugar content of potatoes stored at low temperature by increasing the respiration as a result of raising the temperature. However, this use of temperature to control respiration has the disadvantages of increasing the spoilage from sprouting and rotting and greatly limits the time available for processing. If irradiation were used to increase the respiration this might be accomplished without raising the temperature. Laboratory studies are needed to investigate this possibility. The use of irradiation to control the sugar content of stored potatoes should decrease the loss by spoilage, permit greater time for processing, and provide better control of the sugar content.

4. TYPES OF RADIATION SOURCES TO BE CONSIDERED

For purposes of comparison three different gamma sources have been selected: (1) refined cesium-137, (2) concentrated anhydrous mixed fission products, and (3) cooling reactor-fuel elements.

Since cooling reactor-fuel elements decay very rapidly the radiation facilities using this source of radiation will be designed to operate 6 months of the year during the late winter and spring. During the remainder of the year the plant will be shut down without a radiation source. Because of the slower decay of 2-year-old fission products and cesium-137 the radiation facilities utilizing these materials as sources of radiation will be designed to operate 260 days out of the year.

In order to simplify calculations and comparisons, the radiation source will be assigned the same overall geometrical shape as the source in the radiation facility for the processing of prepackaged fresh meat described in Progress Report 6.[5] This source was a rectangular plaque 6 feet high, 12 feet long, and 0.40 inch thick. The plaque was divided into 12 vertical elements with different specific activities so as to provide a uniform radiation field. The cesium-137 and the mixed-fission-product sources being considered for the irradiation of potatoes will be designed to provide a rectangular source of the same width and length, except that rod sources arranged in a vertical plane will be substituted for the vertical rectangular plaque.

The radiation dose for treatment of potatoes to prevent sprouting has been found to be about 10,000 rep which is about 1/10 the dose required for pasteurization of prepackaged meat. The capacity of the meat plant was estimated to be about 14 tons per hour. This is probably the correct order of magnitude for a typical potato irradiation facility. Therefore, the total activity of the source can be decreased by a factor of 10 to about 150,000 curies of cesium-137 or mixed fission products.

The decrease in the total activity required permits the use of a discontinuous radiation source without requiring an excessive specific activity. Thus, the rectangular plaque 6 feet by 12 feet might be replaced by a number of rods 6 feet long and suitably spaced over a width of 12 feet, so as to provide a uniform radiation field. This arrangement has the advantage of using the same specific activity in each rod which simplifies the fabrication of rod sources and the addition or replacement of source rods. These rod sources might consist initially of 20 rods of 2-inch stainless-steel pipe or tubing filled with either anhydrous cesium-137 or snhydrous mixed fission products and sealed at both ends. The cylindrical containers have the advantage of simplicity in shape and of mechanical strength.

An important point of economics to be considered is that the use of a discontinuous source permits the simple addition of rods to replace activity lost by decay. Using this procedure the number of rods eventually will be increased until some of the rods must be removed and replaced. However, for the case of cesium-137 only 1/10 of the original number of rods need to be added every 5 years to maintain the source activity (within 90 percent of the original activity) whereas 1/6 of the original number of rods would be needed if replacement were used. Thus, an original source consisting of 20 rods filled with cesium-137 could be kept at strength by adding 2 rods every 5 years for the next 30 years. Thereafter, the activity could be maintained by replacing an average of 3-2/3 of the older rods every 5 years. However, if replacement of rods had been used for the first 30 years rather than addition, the number of replacement rods required in the 30-year period would be increased by about 67 percent at a corresponding increase in expense. The same point of economics holds for rods filled with mixed fission products except that replacement would begin much earlier.

Cooling reactor-fuel elements should not be considered as suitable gamma sources for general use or for plants designed to operate continuously throughout the year. Reactor-fuel elements would be too expensive to use if the high inventory cost of fissionable material were charged against the fuel element when used as a source of radiation. However, for reasons relating to the processing techniques it is the practice in present processing plants to store these elements under water for several weeks before they are processed. During this period the intense radiation is dissipated in the water used to shield the cooling elements; except when used in some research experiments this radiation is presently being wasted. It is proposed that some of these elements be used as sources of radiation for a limited number of radiation facilities designed to process potatoes.

The potato irradiation facility using cooling reactor-fuel elements will be designed to operate only 6 months of the year, commencing operations about December 15 and ceasing operations about June 15. During the other 6 months the facility will be without a radiation source. A different design for the radiation facility using the fuel rods would be required since they would have a much higher activity than would the mixed fission products or the processed cesium-137; also the fuel elements have a different geometry.

A single reactor-fuel element would not provide a sufficiently uniform radiation field; therefore, two reactor-fuel elements will be considered. Proper spacing of the fuel elements from each other and from the axis of the conveyor handling the potatoes and limiting the width and length of the conveyor buckets makes it possible to give the potatoes a quite uniform dose of radiation with only two fuel elements.

5. DESIGN OF IRRADIATION CHAMBER USING CESIUM-137

The radiation chamber using cesium-137 as a source of gamma radiation will have the same basic design as the radiation facility designed to pasteurize prepackaged meat described in Progress Report 6.[5] However, certain changes will be required in order to adapt the previous design to the handling of potatoes. In this design the conveyor trays will be proportioned so that either bulk potatoes or 100-lb bags of potatoes can be handled. The tray width will be increased from 8 to 12 inches and the tray height will be increased to 24 inches. This will require an increase in the conveyor pitch from 4-1/2 to 6 inches. Because of the increase in width of the conveyor trays the spacing between each pass will be increased from 15 to 24 inches.

The increase in width of conveyor trays and the increase in spacing between conveyor passes increase the distance of the absorber from the source. The radiation field is decreased by the inverse square relationship and also because a much weaker source is being used. Therefore, only two passes of the conveyor will be used on either side of the radiation source rather than four passes as used in the previous design reported in Progress Report 6.[5]

Figure 36 shows a sectional elevation view of the modified radiation chamber designed to irradiate potatoes using either cesium-137 or mixed fission products as a source of radiation. One-hundred-lb bags of potatoes or bulk potatoes will be brought by belt conveyor A from the potato warehouse. The bags of potatoes or the bulk potatoes move down slide B into tray 1 of the irradiation conveyor while this conveyor is in the stationary phase of its intermittent operation. As the irradiation conveyor moves, the potatoes are carried down into the radiation chamber through opening C and past concrete shield D. Two vertical passes, E and F, are made on the right side of the row of source rods G and two passes, H and I, are made on the left side of the source. This arrangement permits irradiation of the potatoes from both sides so as to produce a more uniform radiation dose.

Well L is filled with water as in previous designs and is used to hold the source when the radiation must be shut off to permit entry to the radiation chamber for maintenance, routine inspection, or addition of source rods. If the radiation chamber is located above grade as shown in Fig. 36, a concrete wall, N, which is 3 feet by 0 inch thick, would be used for shielding. If the radiation chamber is placed below grade, the wall thickness may be reduced to that required for structural strength alone, as the earth will act as a radiation shield. A labyrinthine entrance to the radiation chamber is provided at the lower left as shown in Fig. 36.

A plan view of the radiation chamber is presented in Fig. 37. This view shows the simple laybrinth used as an access passage for routine inspection and maintenance. The conveyor in the radiation chamber may be driven by sprockets on stub shafts. Some of the sprockets may be "idlers", with one or more sprockets to be used as a "driver". Figure 38 shows the spacing between centers of the 2-inch source pipes filled with fission products. The variation in spacing with smaller pitch near the ends is used to produce a more uniform radiation field on either side of the source.

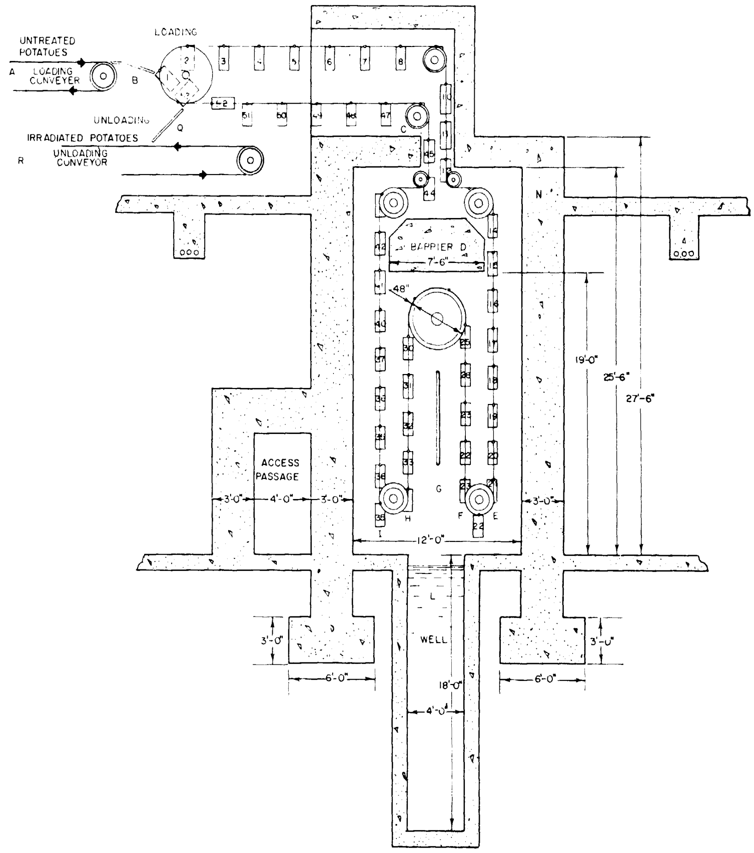


Fig. 36. Elevation View of Potato Irradiation Chamber Using Cesium-137 or Mixed Fission Products.

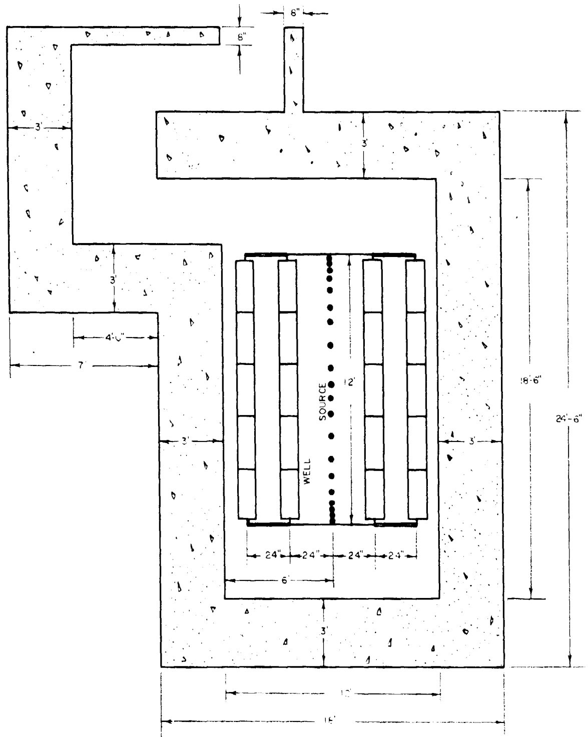
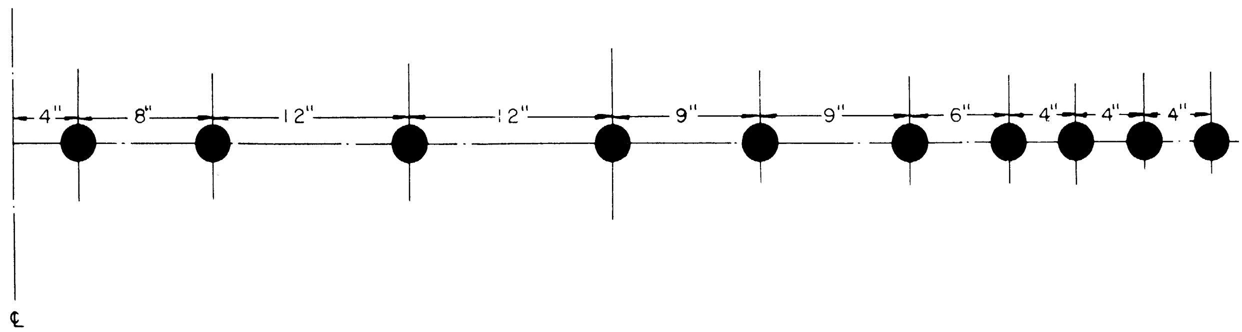


Fig. 37. Plan View of Irradiation Chamber Using Cesium-137 or Mixed Fission Products.



20 Rod Source Distribution, Symmetrical about ℄

Fig. 38. Plan View Showing Spacing of One-Half of Source Rods for Potato Irradiation Facility Using Either Cesium-137 or Mixed Fission Products.

6. CAPACITY CALCULATIONS FOR A MIXED FISSION PRODUCT OR CESIUM-137 SOURCE

The capacity of the radiation chamber shown in Figs. 36 and 37 will depend on the activity of the source and the dose required. The total gamma activity of the source has been set at 150,000 curies so as to provide a capacity considered typical. Fixing this activity fixes the magnitude of the radiation field. A specific gamma activity of 230 curies per pound is sufficient to provide 150,000 curies in the 20-source rods.

The radiation flux in a plane perpendicular to the face of the source at its center line was calculated for the lines 24 inches from the source and 48 inches from the source. The values determined are shown in Fig. 39. This figure shows that the radiation flux along the horizontal axis parallel to the source can be made approximately uniform by varying the activity of the source along the horizontal axis. This dimension may be eliminated from the calculations of the radiation field, permitting the plotting of the radiation field in two dimensions.

As the center line of the conveyor passes are spaced at 24 and 48 inches from the source, the dose rates (in air) were calculated at these distances as a function of the vertical distance above the center line of the source. These data were cros plotted to give the isodose curves shown in Fig. 40. The dose rates shown are for radiation in air and must be corrected for absorption in the potatoes. This correction was made using 8 inches as the half-value thickness for absorption in bulk potatoes for gamma radiation of 0.7 mev energy with an absorber efficiency of 80 percent. The isodose curves corrected for absorption are shown in Fig. 41.

Figure 42 shows a plot of the radiation flux in rep/hour at the center position of the potatoes being conveyed after corrections are made for absorption. The radiation flux is plotted as a function of the length

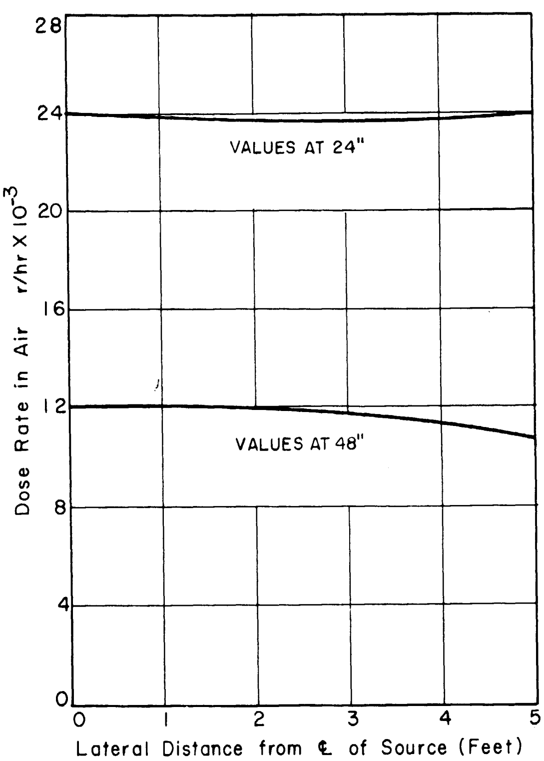


Fig. 39. Radiation Flux vs Lateral Distance for Source Consisting of Selectively Spaced Rods.

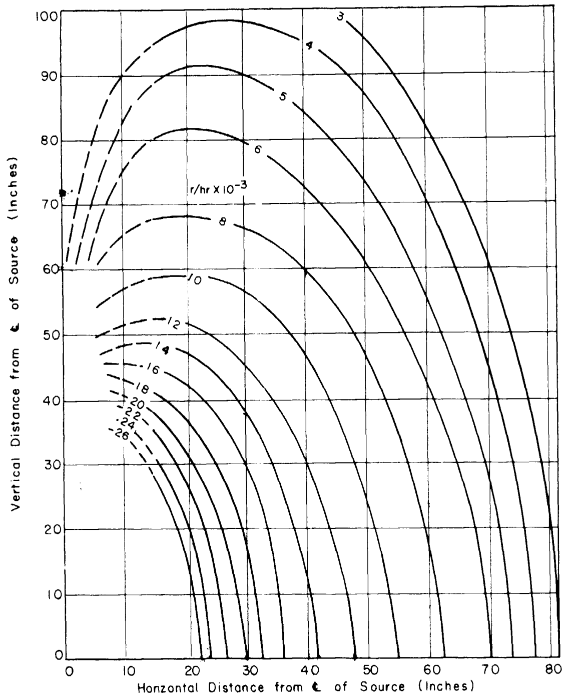


Fig. 40. Isodose Curves in Vertical Plane Perpendicular to Source at Center Line for One Quadrant of Radiation Chamber Using Cesium-137 or Mixed-Fission-Product Sources.

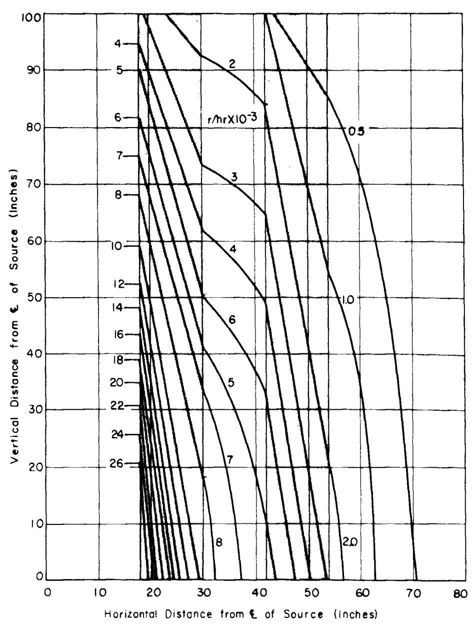


Fig. 41. Isodose Curves from Fig. 40 Corrected for Absorption.

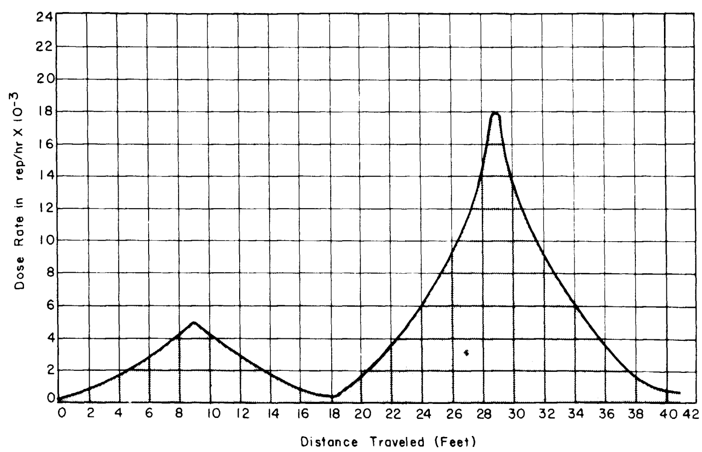


Fig. 42. Dosage Rate Received at Center (12-Inch Wide Conveyor Tray Filled with Potatoes) as a Function of Distance along Path in Radiation Chamber.

traveled through the first half of the radiation chamber. The plot of radiation flux for the second half of travel through the radiation chamber is a mirror image of this curve and is not shown. To determine the time required to absorb a given dose, the curve must be integrated graphically. Twice this integral gives the accumulated dose for the potatoes in the center of the conveyor tray as a function of the tray speed through the chamber. The accumulated dose may be expressed as

$$\text{Accumulated dose in center} = 2\,\frac{\text{rep ft/hr}}{\text{x ft/hr}},$$

where x = tray speed in feet per hour. From the plot of dose rate versus length of path traveled (Fig. 43), the accumulated dose at a tray speed of 1 foot per hour was determined to be 3.82 $(10)^5$ rep.

Dividing the specified dose by the accumulated dose as a function of tray speed yields a value which has the reciprocal units of tray speed. Multiplying this value times the length of path traveled in feet gives the time required per cycle for the potatoes to obtain a specified dose.

$$\text{Radiation time per cycle (for } 1\text{x}10^4 \text{ rep dose)} =$$

$$\frac{10^4 \text{ rep}}{3.82(10)^5 \text{ rep ft/hr}}\,(82)\text{ ft} = (2.14)\,\frac{\text{hr}}{\text{cycle}}\,.$$

There are 30 trays within the chamber each containing 2' x 1' x 12' = 24 cu ft of potatoes per tray; or 24 cu ft x 0.804 bu per cu ft = 19.3 bu of potatoes per tray. The capacity in bushels of potatoes per cycle would then be

$$\text{Capacity/cycle} = 30\,\frac{\text{trays in chamber}}{\text{cycle}} \times \frac{19.3\text{ bu}}{\text{tray}} = (579)\text{ bu/cycle.}$$

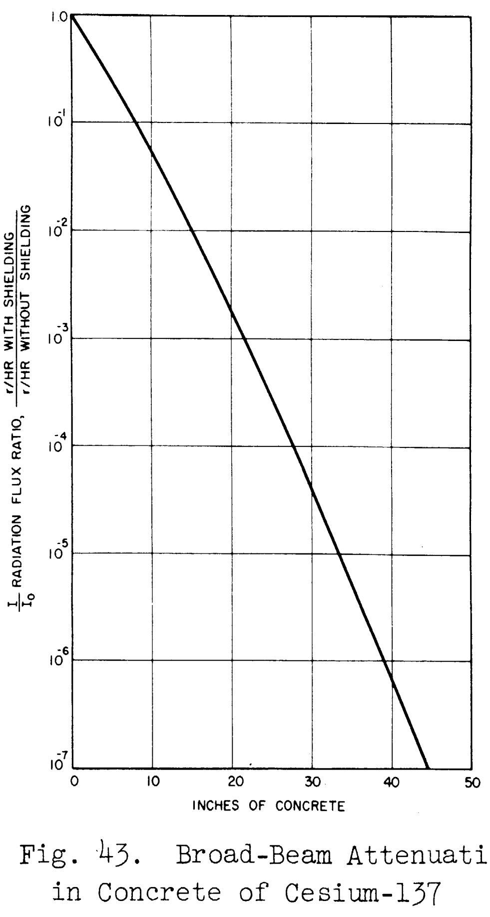


Fig. 43. Broad-Beam Attenuation in Concrete of Cesium-137 Gamma Radiation.

The capacity in bushels per hour may be obtained by dividing by the exposure time required per cycle.

$$\text{Capacity/hr} = (579)\ \frac{\text{bu}}{\text{cycle}}\ \frac{1}{(2.14)\ \text{hr/cycle}}$$

$$= (270)\ \text{bu/hr.}$$

7. TYPICAL SHIELDING CALCULATION

Using the latest broad-beam shielding data (National Bureau of Standards Handbook 54), dosage rates at points outside the radiation chamber were calculated from Fig. 43.

The dosage rate at a point S (see Fig. 36) on the center line of the source just outside the shielding wall (9 feet from the source) is calculated to be 350 rep per hour with no shielding. This value is obtained by extrapolation of the data in Fig. 40. Figure 43 shows that 3 feet of concrete will reduce this dosage rate by a factor of 3×10^{-6} to a level of approximately 1 mr/hr which is considered to be sufficiently below tolerance. See Progress Report 6[5] for calculations for water shielding.

8. COST ESTIMATES BASED ON THE USE OF CESIUM-137

a. Total Investment

The radiation chamber for treating potatoes using either cesium-137 or mixed fission products as a source of radiation is very similar to the radiation facility designed to pasteurize prepackaged meat as described in Progress Report 6.[5] An itemized cost estimate indicated that this latter radiation facility exclusive of the radiation source would cost $82,500. The facility to treat potatoes will have two passes on either side of the source rather than four, will have fewer trays, and will have slightly smaller overall dimensions. It is estimated that the radiation facility would cost 80 percent of the cost of the facility to pasteurize meats, or $66,000.

Using the 0.6 scaling factor for estimating the installed cost of the source of radiation[6] and using the figure of $6000 for a 1000-curie cesium-137 source, the installed cost of 150 kilocuries of cesium-137 is estimated to be

$$x = (\$6,000)\ \frac{(150,000)}{1000}^{0.6}$$

$$x = \$120,000$$

The cost of a steel-reinforced lead container for shipping 150 kilocuries is estimated to be $5000. Thus, the total investment becomes

Cost of radiation chamber and accessories	$ 66,000
Cost of shipping container	5,000
Cost of 150,000-curie cesium-137 source installed	120,000
Total installed cost	$191,000

b. Operation Costs

If the radiation facility were operated as a part of a large potato warehouse or a potato processing plant the annual operation costs might be estimated in a manner similar to the case of the pork-irradiation unit designed previously[7] and as shown in Table 39.

A cesium-137 source will lose 10 percent of its activity in 5 years. The cost of restoring this activity is calculated to be $30,000, once every 5 years. Straight-line amortization would make 20 percent chargeable each year or $6000 per year. Overhead on the payroll, general plant operation, and general administration is estimated according to recommended methods for chemical plants.[8]

The radiation chamber is considered taxable as property but the radiation source is a piece of equipment and not subject to property tax. Income tax should be calculated on the basis of profits, but since the value of irradiated potatoes has not yet been determined 2-1/2 percent of the total investment is used as an estimate of income tax considered chargeable to the irradiation facilities.

In estimating depreciation and obsolescence the radiation chamber and radiation source are considered to have a 10-year useful life. The total investment is amortized in a 10-year period by use of a sinking fund. The interest on cash paid into the sinking fund reduces the percentage of the initial investment chargeable each year from 10 to about 8 percent. This depreciation would provide for complete replacement of the chamber after 10 years. The source is assumed to have only 50 percent salvage value at the end of the 10-year amortization period even though it will be at full strength through activity replacement.

TABLE 39

ESTIMATED ANNUAL OPERATION COSTS FOR POTATO IRRADIATION FACILITY USING CESIUM-137

Salaries and Wages			
Two full-time operators with limited health-physics training	$10,000		
Supervisor and clerical labor (10% of operational labor)	1,000		
Salaries and wages not associated with operation of radiation chamber (50% of direct labor and supervising costs)	5,500		
		$16,500	
Other Operation Costs			
Maintaining source activity ($30,000 every 5 years)	6,000		
Utilities	1,000		
Repairs and maintenance on chamber and conveyor (5% of chambers and conveyor cost)	3,300		
Miscellaneous contingencies	1,000		
		11,300	
Overhead			
Payroll overhead (15% of cost of labor and supervision)	2,480		
General plant overhead (50% of cost of labor and operations)	13,900		
General administration overhead (10% of cost of labor and operation)	2,780		
		18,160	
Taxes, Interest, Insurance			
Property tax (2% of cost of radiation chamber)	1,320		
Income tax (2-1/2% of total investment)	4,780		
Interest (5% of total investment)	9,550		
Insurance (1% of total investment)	1,910		
		17,560	
Depreciation, Obsolesence			
Radiation chamber = (66,000 x 0.08)	5,280		
Radiation source = (initial cost-salvage value) x 8%			
= (120,000-60,000) x (0.08)	4,800		
		10,080	
			$74,600

c. Unit Costs

Based on a minimum dose of 10,000 rep and an operation schedule of two shifts (16 hr/day) and 260 operating days per year, the annual capacity is calculated to be

$$270 \text{ bu/hr } (16 \text{ hr/day}) (260 \text{ days/yr}) = 1.12(10)^6 \text{ bu/yr.}$$

Cost per bushel to be added to potatoes for a dose of 10,000 rep using a cesium-137 source will be

$$\frac{\$74,600}{1.12(10)^6} = \$0.066/\text{bu} .$$

Using a bulk density of 42 lb/cu ft the cost per ton will be

$$\frac{\$0.066(2000)}{0.804\ (42)} = \$3.91/\text{ton} .$$

9. COST ESTIMATES BASED ON THE USE OF MIXED FISSION PRODUCTS AS A RADIATION SOURCE

a. Some Factors Effecting the Cost of Mixed Fission Products

If processed, concentrated fission products are used as a source of radiation their cost might be determined in large part by the cost of concentrating and packaging. However, such information is not available because large quantities of the mixed fission products have not as yet been concentrated and a cost estimate for the processing, to be reliable, would require a rather thorough design of the process being considered. As there are many different types of fission-product wastes and also as many different types of processes that may be used in preparing these materials for industrial use, a direct approach to this cost estimate will be avoided. Another approach to the estimate of the value of mixed fission products might be made on the basis of the comparative value of such radioactive materials and cesium-137. Calculations described in Progress Report 6[5] indicate that 2-year-old fission products have gamma activity essentially from cesium-barium-137 and from cerium-praseodymium-144. The cerium-144 present in 2-year-old mixed fission products has 7-1/2 times the activity of cesium-137 but has a half life of only 290 days as compared to 33 years for the cesium.

The relative value of gamma sources having different half lives might be expressed in terms of some function of the ratio of the half lives.

The nature of this function will be discussed briefly with regard to (1) total dose delivered, (2) replacement costs, (3) total curie requirement, and (4) relative cost proportional to half lives.

(1) Total Dose Delivered. To estimate the relative values of two isotopes of approximately equal energy but different half lives, reference may be made to the total dose delivered.

The dosage rate, as a function of time, is

$$I = I_o e^{-\lambda t} ,$$

where

I_o = the dosage rate at time zero, rep/hr
λ = .693/T = the decay constant of the isotope in question
T = half life
t = elapsed time.

The total dose in rep delivered in any time (t) then is

$$\text{rep} = \int_0^t I\, dt = I_o \int_0^t e^{-\lambda t}\, dt$$

$$= \frac{I_o}{\lambda} \left[1-e^{-\lambda t}\right] .$$

When the elapsed time (t) is large compared to the half life (T), the term in the brackets $\left[1-e^{-\lambda t}\right]$ approaches unity and the dose delivered becomes proportional to the initial dose rate I_o and to the half life, T. When comparing isotopes of different half lives at a given time, t, the value of the long-lived isotope may be short of this "saturation value" while the short-lived isotope has reached its saturation value. Thus, the relative values would not be directly proportional to the half life, and the shorter-lived isotope would be given a relatively higher value. In practice, however, this apparent advantage in radiation value for the shorter-lived isotope would tend to be cancelled by the increased costs required for replacement. Therefore, it is probably a reasonable estimate that radiation value can be considered to be approximately proportional to half life based on total dose delivered.

(2) Replacement Costs. To analyze further the relative values of isotopes having different half lives replacement costs will be considered.

To maintain a radiation facility at design capacity a replacement of a part of the radiation source will be required wherever the radiation level falls below a predetermined minimum level. On permitting, for example a 10 percent drop in radiation intensity,

$$\frac{I}{I_o} = 0.9 = e^{-\frac{.693t}{T}}$$

$$.693\frac{t}{T} = 0.115$$

$$\frac{t}{T} = \frac{0.115}{0.693} = \text{a constant.}$$

As (t/T) is constant the time that will elapse between replacements is proportional to half life. Thus, on the basis of replacement cost alone, it is valid to assure a radiation value directly proportional to half life.

(3) Total Curie Requirement. Other things being equal, the number of curies required at each replacement will be the same, independent of half life. Thus, the total number of curies required to maintain operation at a given rate for a considerable period of time would be inversely proportional to half life. Once again, a reasonable estimate of the relative values of two isotopes of different half lives would appear to be in proportion to their half lives.

(4) Relative Cost Proportional to Half Lives. On the basis of these limited considerations, the relative value of gamma sources having different values of half life can be expressed in terms of the linear ratios of the half-life values, or in other words cerium-144 might be considered to have (290/(365x33)) or about 2.4 percent of the dollar value of cesium-137 as a gamma source over a long-term basis.

b. Cost Estimate for Mixed Fission Products

(1) Total Investment. The 150,000-curie gamma source used for the potato radiation facility if composed of 2-year-old mixed fission products would contain approximately 18,000 curies of cesium-137 and 132,000 curies of cerium-144 plus about 130,000 curies of promethium-147, ruthenium-106, and strontium-90, none of which have any gamma activity and are pure beta emitters. The daughter product of the ruthenium-106, rhodium-106, contributes slightly to the gamma radiation from the mixture.

If the 132,000 curies of cerium-144 are considered to have a dollar value of 2.4 percent of cerium-144, they would have a value equivalent to 3200 curies of cesium-137. Thus, the dollar value of the 150,000-curie gamma source of 2-year-old mixed fission products might be calculated as having a value equivalent to 18,000 + 3200 or 21,200 curies of cesium-137. Using the 0.6 scaling factor and the figure for installed cost of $6,000 for a 1,000-curie cesium-137 source, the estimate is made that the value of a 21,200-curie source would be

$$\text{Source cost} = x = \$6,000 \frac{(21,200)^{0.6}}{1000}$$

$$x = \$38,200.$$

$$\text{Total investment} = 38,200 + 5000 + 66,000 = \$109,200,$$

also

$$\frac{\$38,200}{21,200 \text{ curies}} = \$1.80/\text{equivalent curies, cesium-137}$$

$$\frac{\$38,200}{150,000 \text{ curies}} = \$0.25/\text{total curies of gamma activity.}$$

Using the same figure of $5,000 for shipping container and $66,000 for radiation chamber, the total installed cost is $109,200.

(2) Operation Costs. As a result of a more rapid decay of the mixed-fission-product source it will be necessary to replace 1/6 of the source every 4 months. This will amount to an operating expense estimated to be about $25,000 per year for replacement of mixed fission products plus installation charges. Using the same method of estimating annual operation cost as given in Table 39 but by substituting the different figures for cost of source installed, total installed cost, and cost of replacement, Table 40 was obtained.

TABLE 40

ESTIMATED ANNUAL OPERATION COSTS FOR POTATO IRRADIATION FACILITY USING MIXED FISSION PRODUCTS

1.	Wages, salaries	$16,500	
2.	Other operation costs	30,000	
3.	Overhead	30,380	
4.	Taxes, interest and insurance	10,570	
5.	Depreciation, obsolescence	6,410	
			$94,160

In Table 40 the estimated costs for wages and salaries remain the same for the mixed fission product facility as for the cesium-137 facility. The estimated costs for taxes, interest, and insurance, and depreciation and obsolescence will be somewhat lower for the mixed-fission-product facility than for the cesium-137 facility because of the lower investment in the source. However, the estimated other operation costs have increased about threefold because of the added costs for replacement for the shorter half-lived mixed-fission-product source. This results in a higher estimated annual operation cost using mixed fission products ($94,160 per year) than using cesium-137 ($74,700).

(3) Unit Costs. Based again on a minimum dose of 10,000 rep and an operation schedule of 2 shifts (16 hours per day) and 260 operating days per year, the annual irradiation capacity is calculated to be 1.12×10^6 bushels per year. Based on an annual operation cost of $82,440 per year the following costs are estimated.

Cost per bushel to be added to potatoes for a dose of 10,000 rep using a 2-year-old mixed fission product source will be

$$\frac{\$94,160}{1.12(10)^6} = \$0.084/\text{bu} \quad .$$

Using a bulk density of 42 lb/cu ft the cost per ton will be

$$\frac{\$0.084(2000)}{0.804(42)} = \$4.98/\text{ton} \quad .$$

Thus, although a very low value was placed on the cerium-144 the cost of replacement is sufficient to indicate that the use of mixed fission products would be more expensive than cesium-137.

10. DESIGN OF IRRADIATION FACILITY USING COOLING REACTOR-FUEL ELEMENTS

Cooling reactor-fuel elements have a very high gamma activity but decay at a very rapid rate. It is proposed that reactor-fuel elements be used for a period of 2 months and then be replaced. During each 2-month period, weekly adjustments can be made in the speed of the conveyor moving potatoes through the irradiation room to compensate for decay. If the radiation facility were operated for 6 months starting with freshly installed fuel elements about December 15 and operated until June 15, one replacement of fuel elements would be required about February 15 and the second replacement about April 15.

a. Design of Radiation Chamber

Figures 44, 45, 46, and 47 show the plan, elevation, end, and perspective views, respectively for a potato irradiation chamber using two cooling

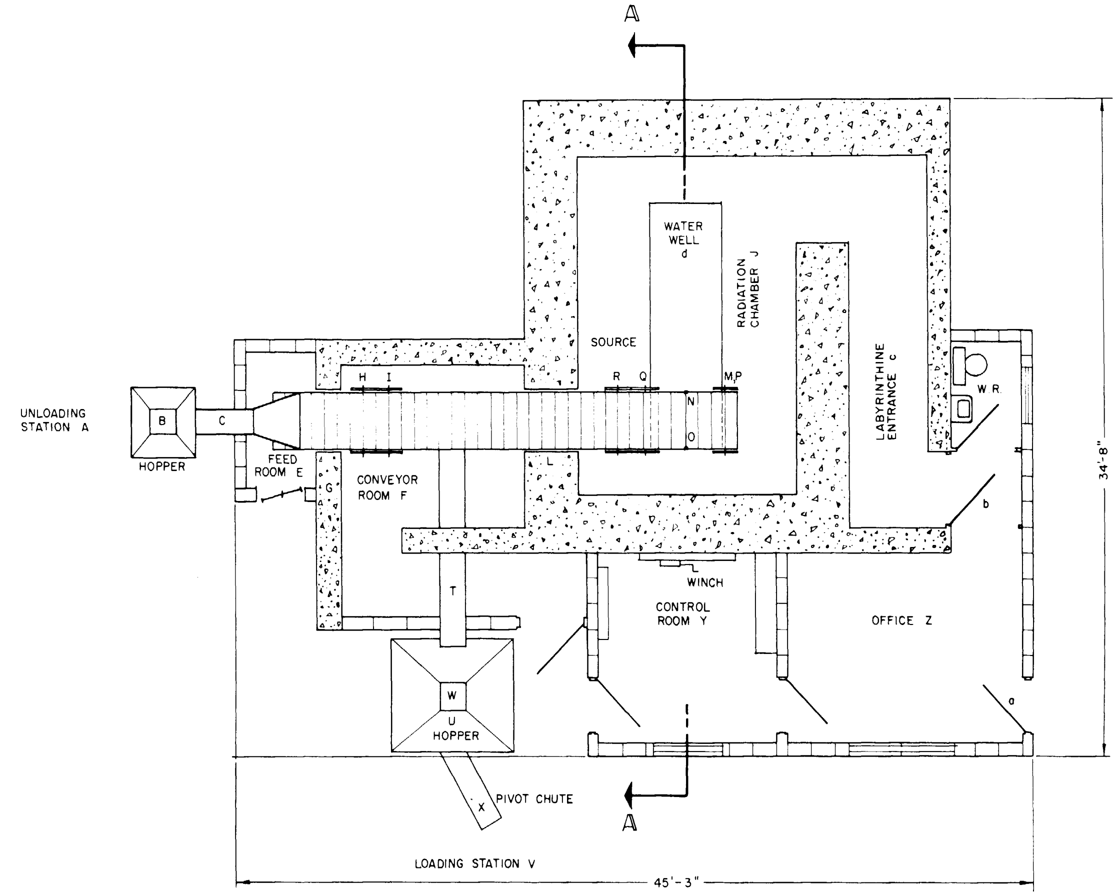


Fig. 44. Plan View of Potato Irradiation Facility Using Cooling Reactor-Fuel Elements.

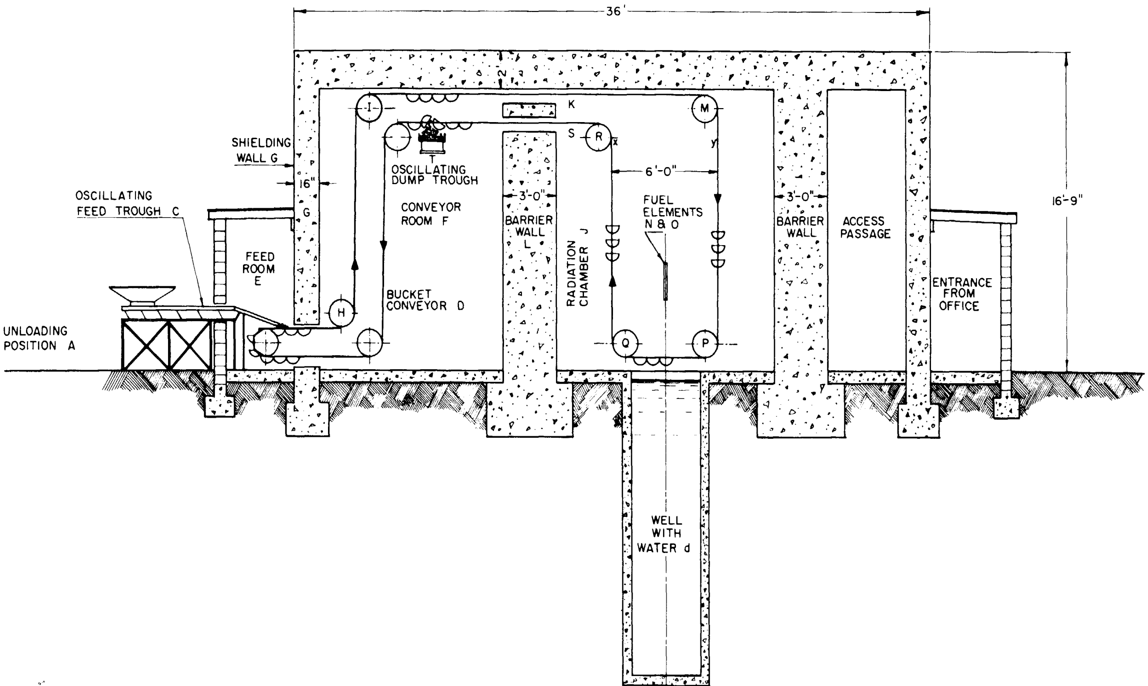


Fig. 45. Elevation View of Potato Irradiation Facility Using Cooling Reactor-Fuel Elements.

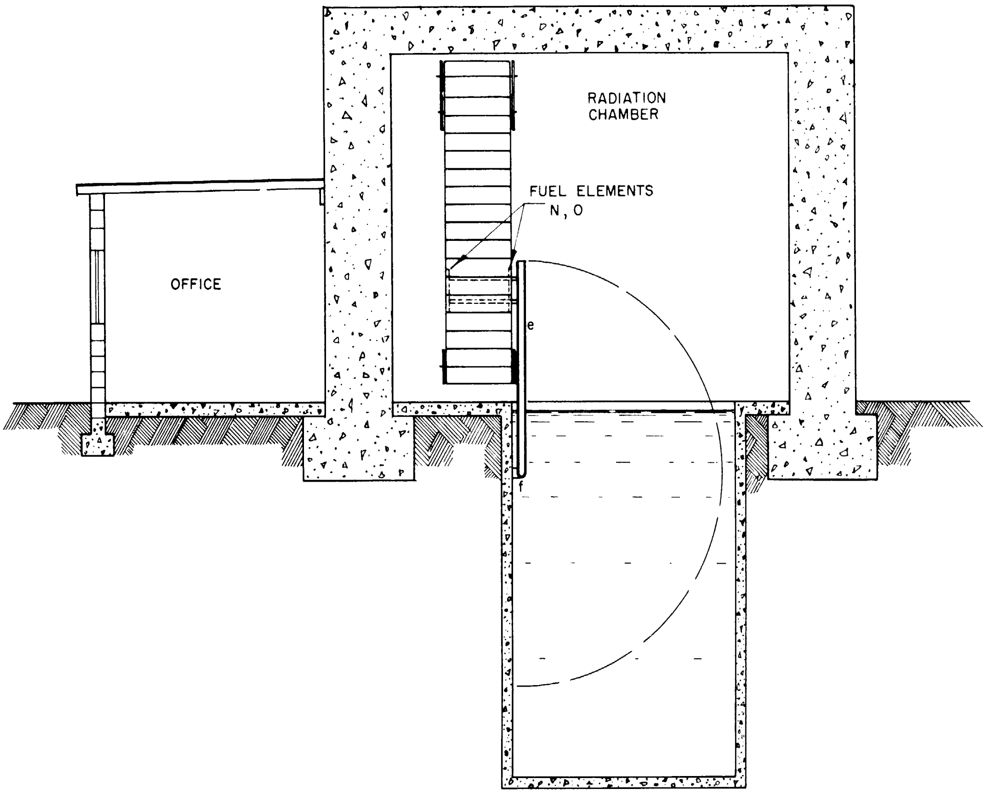


Fig. 46. End View of Potato Irradiation Facility
Using Cooling Reactor-Fuel Elements.

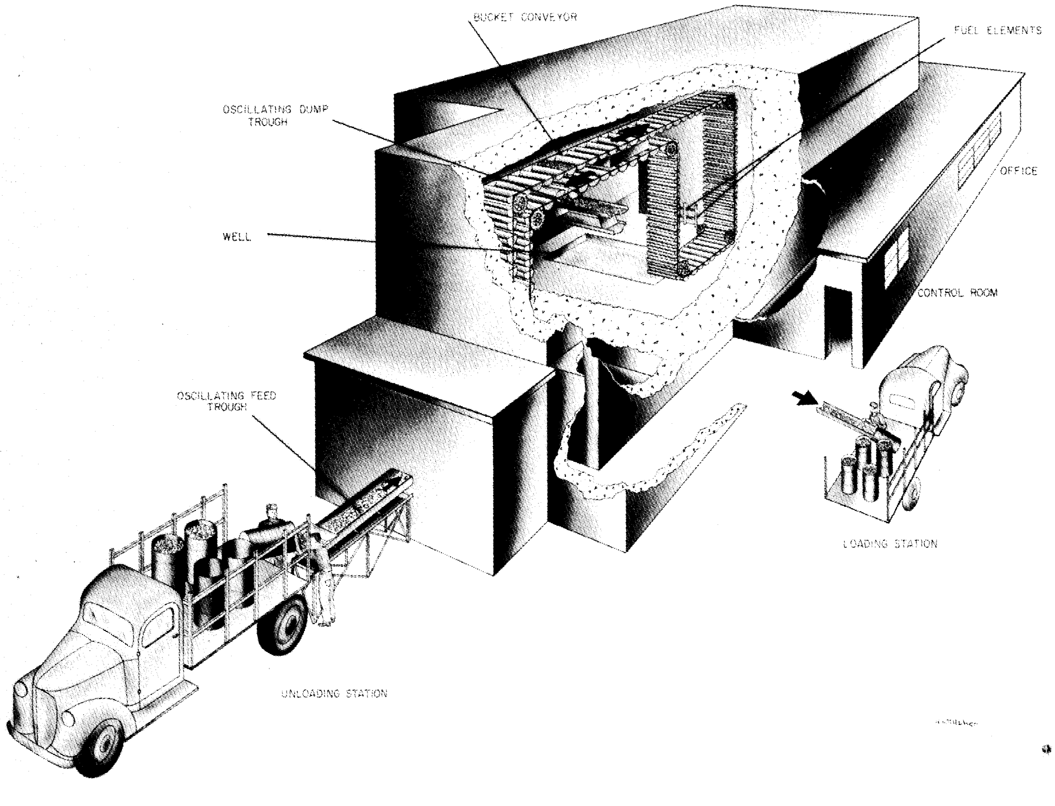


Fig. 47. Perspective Cutaway View of Potato Irradiation
Facility Using Cooling Reactor-Fuel Elements.

reactor-fuel elements as a source of radiation. This radiation facility would be operated only about 6 months of the year and shut down for 6 months. This design probably would be preferred for use by potato growers or a potato-grower cooperative association as the needs of potato growers for irradiation treatment of their stored potatoes would be seasonal. Therefore, an irradiation facility designed for seasonal operation would be preferred to a facility designed for operation the year around or for a major portion of the year.

Referring to Figs. 44 and 45, potatoes would be brought from storage in either sacks or barrels by the truckload. Trucks would unload potatoes at station A into hopper B. Hopper B would be lined with a cushioning material such as sponge rubber to prevent bruising in this operation. As the load of potatoes is being emptied into hopper B, oscillating conveyor C is put into operation. Potatoes are fed at a maximum rate of 14 tons per hour by oscillating feed-trough C in feed room E onto bucket conveyor D. The potatoes then move into the conveyor room F through an opening in the 16-inch shielding wall G. The bucket conveyor becomes a bucket elevator as the chain makes a 90° turn around the sprockets H and then another 90° turn around sprockets I. The potatoes enter the radiation chamber J through slot K in 3-foot 0-inch barrier wall L. The slots are small so as to minimize the radiation entering the conveyor room from the radiation chamber. The potatoes then pass around sprockets M and down past fuel elements N and O. The conveyor then turns a 90° angle around sprockets P and passes underneath the fuel elements. Another 90° bend brings the conveyor with potatoes around sprockets Q and up past the other side of the radiation source. Irradiating both sides of the buckets in this manner provides a more uniform dosage to the potato. Making another 90° bend around sprockets R the conveyor buckets pass out of the irradiation chamber through exit slot S. The buckets are tripped by a cam onto the oscillating dump trough T and are passed from the conveyor room into hopper U on the loading platform.

During this operation the truck which had unloaded at unloading station A moves down to loading station V and the irradiated potatoes which accumulate in hopper U are reloaded into the truck by opening gate W and use of pivot chute X.

The control room Y shown in the plan view contains a panel board and switches for operating the motors for the conveyors and the solenoids for the hopper gates. Also the winch used to raise and lower the cooling fuel elements is located in the control room. An office Z is shown for use in maintaining records of irradiation services, correspondence, etc.

Cooling reactor-fuel elements would be shipped in a lead cylindrical container 3 feet 6 inches in diameter which will be moved by a dolly through the office door a, the cave door b, and into the radiation chamber J by means of labyrinthine entrance c. The shipping container will be lowered into the water well d and the cooling reactor-fuel rods will be removed under water. The shipping container can then be removed from the well and the two fuel elements can be attached to the fuel-element holder shown in the end view. The fuel element holder is attached to a stainless-steel channel e which pivots about bushing f located 3 feet under water.

An interlock between the door to the irradiation cave and the winch operating the lift to the source prevents entrance to the radiation cave when the source is raised. Also a suitable monitor indicates the position of the source and radiation level in the cave. The barrier wall at the labyrinthine entrance and the three 90° bends in passing from the radiation cave to the cave door reduce the radiation field at the cave door to levels that can be safely tolerated from the standpoint of health physics. The hard gamma radiation passing through slots K and S will result in radiation levels above tolerance for routine operations in the conveyor room. Therefore, the conveyor room is considered to be a low-level radiation cave with 16 inches of concrete used in the outside wall for purposes of shielding. Again the use of a labyrinthine entrance reduces the radiation field at the outside entrance of the conveyor room to a safe level.

The water for the well and for the wash room will either be piped underground to the radiation facility or supplied by a well and a water pump located in the control room. A tool crib is also shown in the control room to provide for a small number of tools such as wrenches and grease guns, which may be required in the maintenance and servicing of the conveyors and controls.

b. Uniformity of Radiation Dose

The high gamma activity of the cooling fuel elements permits the use of a small number of fuel elements as the gamma source. The use of a single element would not be very satisfactory because of the space attenuation of the gamma field and the resulting difficulty in delivering a uniform radiation dose to potatoes conveyed through the radiation field. Proper spacing of two fuel elements results in the addition of the radiation field of one element to the radiation field of the other, which for a limited distance will give a zone of uniform radiation flux in one direction. If the conveyor moves perpendicular to this line of uniform radiation field the potatoes distributed across the conveyor will receive a uniform radiation dose.

The gamma flux from cooling fuel elements varies with time and with the original activity. Based on calculations for fuel elements cooled for 1 to 5 months it is estimated that a typical cooling element used for 2 months might have a radiation flux of 1×10^5 rep/hr at a radius of 3 feet from the midpoint of the element. Using this estimated flux, Fig. 48 was prepared in which the radiation field is shown along a plane perpendicular to the center lines

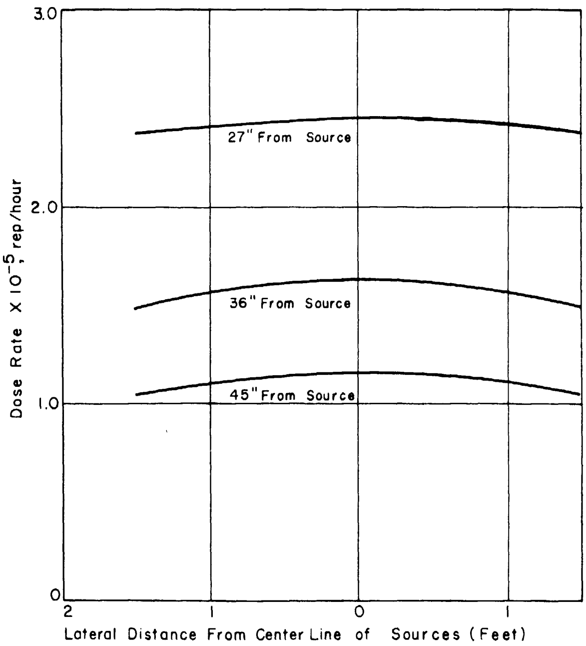


Fig. 48. Radiation Field in a Plane Perpendicular to the Center Lines of Two Parallel Fuel Elements Spaced 3 Feet Apart.

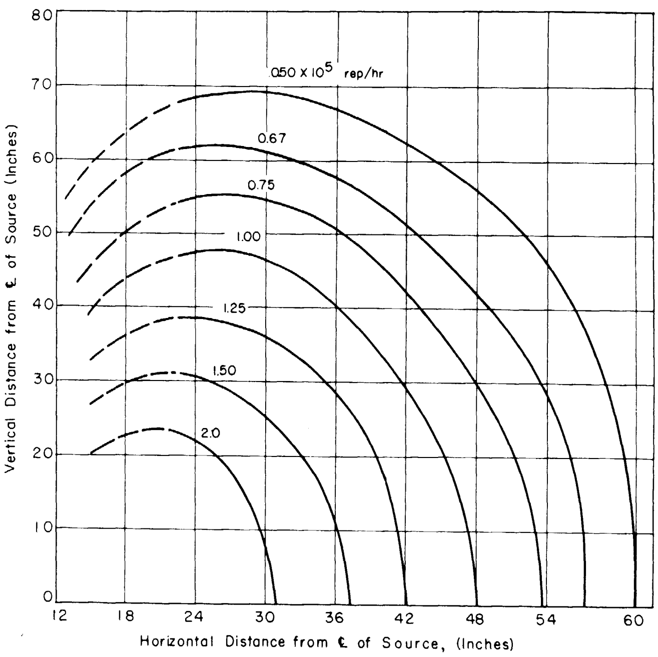


Fig. 49. Isodose Curves (rep/hr) in a Vertical Plane Equidistant from Two Parallel Vertical Fuel Elements Spaced 3 Feet Apart.

of two parallel fuel elements spaced 3 feet apart. Approximate uniformity of the radiation field at 27 inches from the axis of the source in the lateral direction is shown by the nearly flat curve. This insures a near-uniform dose to the potatoes independent of their lateral position. Isodose curves were plotted for the plane perpendicular to this direction. Figure 49 shows such isodose curves plotted for this plane which is parallel to the axis of the fuel element and equidistant from both elements.

The minimum radiation dose will be received by the potatoes located in the center of the conveyor bucket. Potatoes located on the edge of the bucket facing the fuel elements will receive a higher radiation dose than the potatoes in the center of the bucket as they pass down along the inner traverse because of the shorter distance between these potatoes and the source. However, this will be compensated for, to a large extent, by the lesser dose these same potatoes receive as they travel upwards in a position more distant from the radiation source. The sum of the doses in these two positions will be greater than the dose in the center of the bucket but this difference will not be great if the bucket is placed a sufficient distance from the source and if the bucket is not excessively wide.

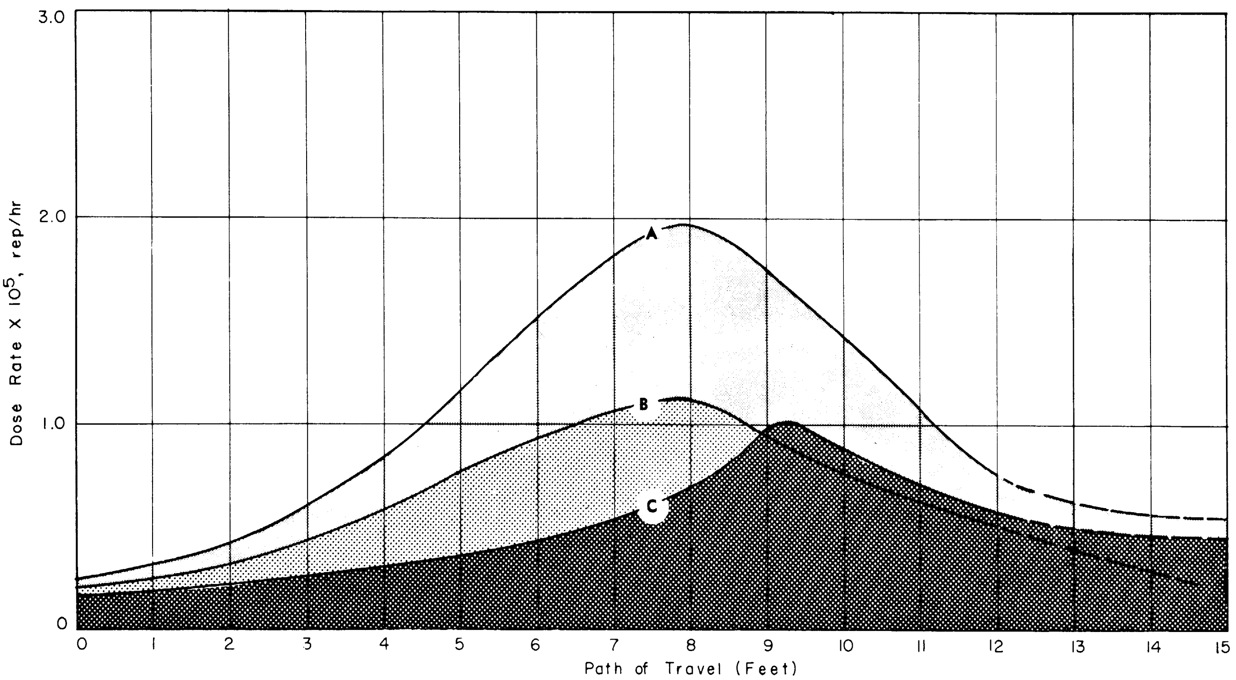


Fig. 50. Dose Rate Received by Potatoes in Three Positions (A, Nearest to Source; B, Center; and C, Farthest from Source) As a Function of Distance along Path in Radiation Chamber.

c. Capacity Calculations

Figure 50 is a plot of the dose rate as a function of distance traveled in the irradiation chamber for three potato positions. Position C depicts the dose received by a potato in the farthest position, B the central position, and A the nearest position of the bucket to the source. For any given conveyor speed this plot then becomes a plot of dose rate versus time and the integrated area under the curve becomes the accumulated dose as the potatoes travel through the radiation cave. For equal exposure from either side a potato located at position B would receive the minimum dose equal to twice the area under curve B. The maximum dose for a potato at the edge and exposed from either side is equal to the sum of the areas under curves A and C.

The area under the curve of Fig. 50 for position B was graphically determined to be 18.0×10^5 rep ft/hr. Dividing the value of this integral into the dose requirement of 10,000 rep and multiplying by the feet of travel yields the time required for the potatoes to accumulate the specified minimum dose. This time was calculated to be 10 minutes. It is expected that it will be necessary to change the setting of the conveyor speed regulator each week in proportion to the change in radiation flux as a result of decay.

A variable speed drive has been specified having a 3 to 1 speed variation to facilitate changing the conveyor speed as required.

The conveyor bucket of conveyor-elevator D will have a semicircular shape with a radius of 9 inches and a bucket length between chains of 3 feet. The chain will have a pitch of 18 inches. Each bucket will carry an estimated 2.65 cu ft of potatoes in bulk and using a conveyor speed of 3 ft per minute, a total of 2.65 cu ft buckets x 120 buckets/hr = 318 cu ft/hr will pass through the irradiation chamber. This capacity is equal to 318 x .804 bu/cu ft x 8 hr/shift = 2050 bu/8-hr shift. As potato growers probably would truck potatoes chiefly during the daylight hours, 2 daily shifts of 8 hours each would probably be desirable and the plant would operate on only a 5- or 6-day week. Thus, the maximum capacity using 2 shifts per day and a 6-day week over 26 weeks would be 2050 x 2 x 6 x 26 = $6.40(10)^5$ bu potatoes/year.

11. COST ESTIMATES BASED ON THE USE OF REACTOR-FUEL ELEMENTS 26 WEEKS PER YEAR

a. Total Investment

Table 41 itemizes the cost estimated for the construction of the irradiation facility, not including the radiation source. Since the source is rented, the cost of the irradiation facility will be the total investment.

b. Operation Costs

As this installation will be used only 6 months out of the year, it is estimated that the facility should be depreciated over a period no shorter than 10 years. If this radiation facility is operated by a group of potato growers on a cooperative basis, it is believed that three employees working 40 hours per week for 26 weeks of the year could operate the installation. One of the employees would be capable of handling a routine maintenance, operation, and control and should have sufficient health-physics training to follow approved health-physics procedures in routine operation and should know what to do in case of any unexpected emergency. The second employee would have the same training and duties and would act as relief for the first employee. The third employee would be expected to answer the telephone and handle the clerical work such as records, correspondence, etc.

Supervision and continuity of operation from year to year could be supplied on a part-time basis by an administrator from a potato-growers association or cooperative group. Table 42 shows an estimate for the annual costs of operation for a potato irradiation facility operated on the basis described.

TABLE 41

ESTIMATED COST OF POTATO IRRADIATION FACILITY USING COOLING FUEL ELEMENTS

Excavation and shoring for footings and well	$ 800	
Concrete for 4'x10'x16' well (20 yd at $20/yd)	400	
Reinforcing for well (1000 lb at $0.10/lb)	100	
Asphalt lining for well	150	
Forms for well (1000 bd ft at $100/M)	100	
Labor for forming and pouring well	600	
Concrete for walls and footings (225 yd at $20/yd)	4,500	
Forms for wall (4500 bd ft at $100/M)	450	
Labor for forming and pouring wall	2,500	
Concrete for floor (30 yd at $20/yd)	600	
Reinforcing for floor (500 lb at $0.10/lb)	50	
Labor for pouring floor	250	
Concrete for roof (50 yd at $20/yd)	1,000	
Forms for roof	800	
Reinforcing for roof (4000 lb at $0.10/lb)	400	
Labor for forming and pouring roof	800	
Lift mechanism for source	1,500	
Ion-exchange system for well water	3,000	
Monitoring equipment	4,000	
Unloading hopper	400	
Loading hopper	800	
Oscillating feed trough	1,000	
Oscillating dump trough	1,000	
Bucket conveyor L	6,000	
Bucket conveyor drive (3 P variable speed)	1,000	
Wash-room fixtures	200	
Office furniture	400	
Maintenance tools and supplies	200	
Heating and ventilating	1,200	
Access doors (with safety interlock)	1,400	
Road grading	600	
Wiring	400	
Water lines and labor for pipe fitting	800	
Backgrading	200	
Painting	600	
Subtotal for labor and materials	38,200	
Miscellaneous contingencies (10% of subtotal)	3,820	
Engineering costs (7% labor and materials)	2,670	
Contractors fee (10% of costs)	4,470	
Total		$49,160

TABLE 42

ESTIMATED ANNUAL OPERATION COSTS FOR POTATO IRRADIATION FACILITY USING COOLING REACTOR-FUEL ELEMENTS AND OPERATING 6 MONTHS PER YEAR

Salaries and Wages			
Two operators with limited health-physics training (Full time for 6 months/year, $10,000 year)	$5,000		
Supervision and clerical labor (10% of operational labor at full salary)	1,000		
Salaries and wages not associated with operation of radiation chamber (50% of direct labor and supervising costs)	3,000		
		$ 9,000	
Other Operation Costs			
Shipping costs for fuel elements 3 x 200 (every 2 months)	600		
Handling costs for fuel elements during transfer and installation 3 at $1000	3,000		
Rental of two fuel elements (Nominal charge assumed, $500/mo for 6 months)	3,000		
Repairs and maintenance on chamber and conveyor (5% of chamber and conveyor cost)	2,460		
Miscellaneous contingencies	1,000		
		10,060	
Overhead			
Payroll overhead (15% of cost of labor and supervision)	1,350		
General plant overhead (50% of cost of labor and operations)	9,530		
General administration overhead (10% of cost of labor and operation)	1,910		
		12,790	
Taxes, Interest, Insurance			
Property tax (2% cost of radiation chamber)	980		
Income tax (2-1/2% of total investment)	1,230		
Interest (5% of total investment)	2,460		
Insurance (1% of total investment)	490		
		5,160	
Depreciation, Obsolesence			
Radiation chamber ($49,160 x 0.08)	3,930		
		3,930	
			$40,940

c. Unit Costs

Based on an expected average operating capacity for a period of 26 weeks of the year this plant might be expected to irradiate 6.40×10^5 bushels of potatoes per season. With an annual estimated operating cost of $40,940 the cost for irradiation of potatoes is calculated.

Cost per bushel to be added to potatoes for a dose of 10,000 rep using two cooling reactor-fuel elements 26 weeks of the year.

$$\frac{\$40,940}{6.40(10)^5} = \$.064/\text{bu} \ .$$

Using a bulk density of 42 lb/cu ft the cost per ton will be

$$\frac{\$.064(2000)}{0.804(42)} = \$3.78/\text{ton} \ .$$

12. COST ESTIMATES BASED ON THE USE OF REACTOR-FUEL ELEMENTS 260 DAYS PER YEAR

These unit costs based on the use of fuel elements for 156 days per year are slightly less than for the case of using cesium-137 for 260 days per year. If, however, this facility were operated with fuel rods for 260 days per year the annual capacity would be increased to $1.06(10)^6$ bu/yr which is only slightly less than for the design using tray conveyors with two passes on either side of the source. The estimated annual operating cost is given in Table 43.

TABLE 43

ESTIMATED ANNUAL OPERATION COSTS FOR POTATO IRRADIATION FACILITY USING COOLING REACTOR-FUEL ELEMENTS AND OPERATING 260 DAYS PER YEAR

1.	Salaries and wages	$16,500	
2.	Other operation costs	16,880	
3.	Overhead	22,550	
4.	Taxes, interest, and insurance	5,160	
5.	Depreciation, obsolesence	3,930	
			$65,020

In Table 43 the estimated costs for wages and salaries are the same as for the mixed-fission-product and the cesium-137 facilities. The estimated costs for taxes, interest, and insurance, and depreciation and obsolescence are lower for the fuel-element facility than for both the cesium-137 facility and the mixed-fission-product facility because of the lower total investment. However, the estimated other operation costs for the fuel-element facility are greater than for the cesium-137 facility because of the added costs for replacement for the fuel elements. The net result is a slightly lower estimated annual operating cost using fuel elements ($65,020 per year) than using cesium-137 ($74,700 per year).

Based on a minimum dose of 10,000 rep, 260 operating days per year, an annual irradiation capacity of $1.06(10)^6$ bu/yr and an annual operation cost of $65,020 per year, the following costs are estimated.

Cost per bushel to be added to potatoes for a dose of 10,000 rep using two cooling reactor-fuel elements 260 days per year will be

$$\frac{\$65{,}020}{1.06(10)^6} = 0.061 \quad .$$

Using a bulk density of 42 lb cu ft the cost per ton will be

$$\frac{\$0.061(2000)}{0.804(42)} = \$3.62/\text{ton} \quad .$$

13. DISCUSSION

Based on a radiation dose of 10,000 rep, an operation schedule of 260 days per year and plant amortization over a 10-year period, the estimated minimum cost for irradiation of potatoes is $3.62 per ton when using two cooling reactor-fuel elements as a source of radiation. If cesium-137 were used as the source of radiation the estimated cost would be about 15 percent greater or $3.91 per ton. If 2-year-old mixed fission products were used the estimated cost would be about 36 percent greater or $4.98 per ton. However, if greater than 10 percent variation in radiation flux were permitted this cost might be reduced to less than that based on using cesium-137. For an irradiation season of 26 weeks per year rather than 260 days per year the minimum estimated irradiation cost (using fuel elements) is increased 14 percent to $3.78 per ton.

These costs are slightly higher than costs for chemical treatment to prevent sprouting of potatoes. The approximate cost of applying a dust of the methyl ester of naphthalene acetic acid is $2.50 per ton of potatoes treated. Maleic hydrazide can be applied as a spray to plants in the field at a cost of about $15 per acre.[9] For a moderate yield of 10 tons per acre this corresponds to a cost of $1.50 per ton. Irradiation of potatoes for the dose given would probably cost at least twice as much as chemical treatment. However, irradiation at this level would result in a very uniform treatment believed to be superior in uniformity to a chemical treatment. If cost is a controlling factor the dose might be reduced by as much as 50 percent resulting in an irradiation treatment anticipated as being competitive with chemical treatment both with regard to cost and probable uniformity of product.

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D. A STUDY OF THE POSSIBILITY OF NUCLEAR ACTIVATION OF ELEMENTS FOUND IN FOOD WITH COBALT-60 GAMMA RAYS

Personnel:

W. Wayne Meinke, Assistant Professor of Chemistry and Advisor of Fission Products Laboratory.

1. INTRODUCTION

It has been assumed by many who have studied the effects of x-rays and gamma rays on food sterilization, chemical reactions, etc. that these radiations induce no radioactivity. This assumption is based on the fact that the irradiated materials show no activity when tested with ordinary radiation monitors. In most cases, however, the background radiation level is such that small amounts of induced activity could be easily overlooked.

A search of the literature indicates that x-rays can indeed induce low levels of activity in certain elements. A 4.1-hour indium activity was reported[1] first in 1939 by x-ray excitation and was shown[2] to have a threshold for production of 1.2 ± 0.1 mev. Later it was found that lead[3] and silver[4] activities could also be produced by x-rays with energies up to 1.5 mev. Wiedenbeck[5,6,7] studied the excitation of these elements as well as other elements such as rhodium, cadmium, gold, and krypton. In each of these cases the ground state isotope was excited to an isomeric state by the x-radiation. The isomer then decayed by its characteristic half life and energy as shown in Table 44[8].

Guth[9] has shown that this excitation by x-rays is a "line absorption" in which x-rays of only a certain limited energy will induce the activity. It was found that the widths of the lowest nuclear levels of lead and indium are of the order of a few millivolts. Hence, in the continuous spectrum of 1.5 mev x-rays, only about one in 10^8 x-ray quanta has an energy suitable to induce activity.

TABLE 44[8]

NUCLEAR DATA FOR ISOMERS

Isomer	Half life, hr	min	sec	Radiation	Energy of Photons, (mev)	Gamma Rays Converted
Rh 103 m		57		γ	0.040	highly
Ag 107 m			44.3	γ	0.094	$e^-/\gamma = \sim 16$
Cd 111 m_2		48.6		γ	0.150	$e^-/\gamma = 2.3$
					0.246	$e^-/\gamma = 0.064$
In 115 m	4.50			γ (95%)	0.335	$e^-/\gamma = 0.98$
				β^-(5%)	0.83	
Au 197 m			7.4	γ	0.130	$e_k^-/\gamma \leq 2.0$
						K/L/M = 1/7.5/3.6
					0.279	$e_k/\gamma \sim 0.3$
Pb 204 m_2		68		γ	0.905	$e^-/\gamma \sim 0.1$
					0.374	$e^-/\gamma \sim 0.05$

A logical extension of this work was to determine whether indium activities could be produced by the gamma rays from radium and its daughters. Guth[9] reports that Goldhaber, Hill, and Szilard irradiated indium with the gamma rays from 1/2 gm of radium without producing any activity. Since it is known that the energy of these gamma rays is sufficient to excite the indium, the absence of activity must be attributed to the fact that none of its known gamma ray energies are equal to the energy of the activation level. The probability of excitation is therefore small and Guth concludes that only irradiation by a continuous x-ray spectrum is likely to lead to a line absorption.

With the advent of multikilocurie sources of cobalt-60 and eventual megacurie sources of fission products, gamma ray intensities many orders of magnitude greater than that from 1/2 gm of radium are available. Furthermore, the thickness of these sources tends to "smear" out the energy of the gamma rays emitted by the source until the gamma energy spectrum no longer shows sharp, nearly monoenergetic rays but instead a gamma ray distribution which approaches a continuous spectrum. This spectrum should then be nearly as effective as the continuous x-ray spectrum for activation. Harbottle[10] has recently published a report of the activation of indium and cadmium isomers with cobalt-60 gamma rays.

This section is a report of an initial study on an independent experimental program conceived as the result of discussions at the Symposium on Radiation Sterilization of Foods sponsored by the Committee on Foods of the Advisory Board on Quartermaster Research and Development. This symposium was held on June 26, 1953, at Massachusetts Institute of Technology under the general auspicies of the National Research Council. This program is not designed to evaluate the tolerance limits of various activities in foods. Its purpose is only to determine within the limits of detection available, the amount of activity (if any) induced in a number of elements by gamma rays from cobalt-60 and later from gross fission products. With such extensive effort being expended by a number of laboratories in trying to perfect the technique of food sterilization with gamma radiation, it was felt that some numerical limits for the amounts of activity induced by gamma radiation should be experimentally determined. Once determined these results could then be evaluated as to their hazard by the proper authorities.

This study was supported under University of Michigan, Engineering Research project M943-5 and the scintillation well counter was made available from an AEC Nuclear Chemistry grant under contract AT (11-1)-70.

2. EQUIPMENT

From the literature references it was apparent that if any activity were formed it would be low-level and would require a sensitive detector. In addition, Table 44 shows that, in general, the isomeric states formed by the activation decay by the emission of gamma rays. An ordinary Geiger tube, whose counting efficiency for gamma rays is only about 1 percent, is ineffective in detecting such radiation. A scintillation well counter had been obtained, however, from the Nuclear Instrument and Chemical Company for other low-level gamma-ray counting work on a nuclear chemistry grant from the Atomic Energy Commission. This counter, which was made available for the gamma activation work, enabled the sensitivity of the measurements to be increased.

The well counter consists of a 1-3/4-inch diameter and a 2-inch thick crystal of thallium activated sodium iodide. A 3/4-inch hole, 1-1/2inches deep, was drilled in the center of this crystal and the entire crystal is encased in 1/32-inch aluminum. A test tube containing a sample was inserted in the hold for measurement. Rays (beta and gamma) which penetrate the wall of the tube and the thin aluminum casing of the crystal were absorbed in the crystal giving a flash of light. This scintillation is then "seen" by a Dumont 6292 (K-1186) photomultiplier tube which feeds a scaler and register. The particular well counter used had a counting efficiency of about 43 percent for cobalt-60 rays and about 51 percent for those of iodine-131.

3. PROCEDURE

Since meat is one product for which radiation sterilization may be particularly well suited, the trace elements it contains were checked for possible activation by the cobalt-60 gamma-ray sources. The amount of various trace elements which occur in meat (beef) has been given in *American Scientist* based on a method developed by Arthur J. Mittledorf of the Armour Foundation[11]. He estimates that during a year the average American eats about 60 lb of beef. The amount in ounces of trace elements a person obtains in eating these 60 lb of beef is given in Table 45.

TABLE 45[11]

OUNCES OF TRACE ELEMENTS IN ONE YEAR'S CONSUMPTION (60 lb) OF BEEF

Aluminum	0.00001	Molybdenum	0.000014
Boron	0.00006	Nickel	0.000003
Calcium	0.0025	Phosphorus	1.6
Chromium	0.000003	Potassium	4.7
Cobalt	0.000002	Silcon	0.00006
Copper	0.00043	Silver	0.000013
Iron	0.0306	Sodium	0.40
Lead	0.00001	Tin	0.000002
Manganese	0.00002	Zinc	0.045
Magnesium	0.204		

Pure samples of each of these elements (as well as carbon, oxygen, sulfur and iodine) were placed in test tubes 1.6 cm in diameter and 15 cm long which just fit into the counting well. The test tubes were filled with at least 2 inches of sample so that the well would be completely filled with the sample for maximum detection sensitivity. These samples were irradiated by the 10-kilocurie cobalt-60 source for varying periods of time and then counted in the scintillation well counter. Because of a large detection area this counter has a high background counting rate. This background was reduced to about 300 counts per minute however by enclosing the counter in about 4 inches of lead. Counts of 1 hour duration both for background and sample gave reasonable statistical precision. A count of this length gives a "9/10 error" of about 1.2 percent for the determination.

4. RESULTS

a. Indium Activation

The method of activation by gamma radiation was first tested by the irradiation of indium in the cobalt-60 source. It was felt that if the source had enough intensity to produce activity in anything it would activate the indium. The tube was filled with about 85 gm of indium and irradiated in a flux position of about 220,000 rep per hour; nearly to saturation of the 4.5-hour activity. When counted, the sample showed activity about forty times the background of the well counter. The activity decayed with a half life of 4.41 hours with a 9/10 reliability factor of 0.05 hours. This value agrees with accepted literature values for In^{115m}. A rough attempt to evaluate the efficiency of the well counter gave a value of about 50 percent for the In^{115m} gamma ray. In addition a rough evaluation of self absorption of the radiations in the indium metal indicates that about 1000 γdis/min/gm of indium were formed in the irradiation or roughly 4.5 γdis/min/gm In per 1000 rep per hour saturation flux. Harbottle[10] has recently reported similar results in an independent investigation at Brookhaven National Laboratory. He reports an order of magnitude value for the cross section for formation of indium from cobalt-60 gamma rays of 10^{-9} barns.

b. Trace Element Activation

The work with indium was admittedly weighted heavily in favor of success but it established that the method was satisfactory to detect small amounts of activation induced by gamma radiation. Twenty-four samples of pure elements or compounds were then prepared and irradiated. The elements, their sources, and weights are listed in Table 46.

These elements were subjected to two series of experiments. In one the samples were irradiated simultaneously for a day or so and then rapidly removed from the source room and counted. It was possible by rapid manipulation of the source to have a sample in the well counter between 40 to 45 seconds after the end of irradiation. By raising the source immediately after removal of one sample, it was assured that any short-lived activity in the remaining samples would remain at saturation. Short-lived activities with half lives down to the order of 20 seconds could have been detected if formed in even 1/10 the yield of the indium. However, no activity greater than 5 percent of background was found in any of these samples in 20 minute-counts.

TABLE 46

FORM, SOURCE, AND WEIGHT OF ELEMENTS IRRADIATED IN GAMMA RAY SOURCE*

1. Aluminum powder - high purity - Alcoa - 2.7 gm
2. Elemental boron powder - Student prep - small amount of chemical contamination - 4.8 gm
3. Calcium metal turning - B and A reagent grade - 2.1 gm
4. Carbon - graphite flakes - Fisher - 2.9 gm
5. Chromium metal powder - Charles Hardy - 15.1 gm
6. Cobalt metal powder - Old Kahlbaum material - contains some Cl - 5.9 gm
7. Copper metal pellets - Standard Laboratory grade - 16.3 gm
8. Iron metal powder - Standard Laboratory grade - 12.2 gm
9. Elemental iodine crystals - Mallinckrodt analytical reagent - 11.0 gm
10. Lead metal filings - Mallinckrodt analytical reagent - 21.5 gm
11. Magnesium metal powder - Dow - high purity - 5.5 gm
12. Manganese metal powder - Charles Hardy - 15.1 gm
13. Molybdenum metal powder - Primus Chemical Company (1912) - 14.5 gm
14. Nickel metal shot - Mallinckrodt analytical reagent - 16.1 gm
15. Oxygen - (Al_2O_3) - Harshaw - 5.3 gm
16. Phosphorus - $(NH_4)_2HPO_4$ - Mallinckrodt analytical reagent - 3.7 gm
17. Potassium - KCl - General Chemical Company - reagent grade - 5.3 gm
18. Elemental silicon powder - Fairmount Chemical Company - 4.0 gm
19. Silver metal powder - G. Frederick Smith - reagent grade - 16.0 gm
20. Sodium chloride (NaCl) Merck analytical reagent - 5.9 gm
21. Sodium fluoride (NaF) - cp - 3.3 gm
22. Sulfur (roll) - cast - 4.38 gm
23. Tin metal granules - General Chemical Company - reagent grade - 15.1 gm
24. Zinc metal granules - cp - 10.6 gm

*The weight listed in the table is the amount of material contained in the bottom 1 cm of the test tube used in the irradiation and counting.

In a second set of experiments the samples were irradiated in the high flux position of the source at a flux level of 220,000 rep per hour for about 2 weeks. The samples were then removed individually and counted. They were compared with a background count of the same day and also with a blank sample count before irradiation. An hour background count fluctuated ± 1 percent over the 3 week period of the measurements. It was found that only two samples varied from this background region by more than ± 1 percent. One was potassium chloride whose natural radioactivity increased the blank count to 2/3 more than background. The irradiated sample gave the same count as the blank sample, however. The second sample was the chromium metal powder. An increase was found in this sample after irradiation but the activity did not decay over a period of several weeks and must be attributed to contamination.

5. DISCUSSION

It should be stressed that in the method used, the radiation must pass through the test tube, about 300 mg/cm^2 thick, and the aluminum casing of the crystal before it is detected. This then prevents the detection of gamma rays of less than about 30 kev energy or of most conversion electrons from highly converted gamma rays. Since early work in this field pointed out several elements whose isotopes on activation decayed with such weak radiations, the present work is somewhat limited. Nevertheless, it was thought worthwhile to perform the experiment with the equipment on hand and to recognize the fact that nuclides emitting certain very weak radiations would escape detection.

Activity found in a sample might also be accounted for by conditions other than gamma-ray activation. One of the elements tested, potassium, has a low abundance isotope that is naturally radioactive and was detectable in the scintillation crystal. Natural radium also often occurs to a small extent in many materials. Shandley[12], at the University of Rochester, has reported a highly sensitive method for determining the radium content of common foods by deemanation. He found that most of the foods tested fell within the range of 0.74 to 6.5×10^{-15} gm radium per gm of starting material. This amounts to about 10^{-9} microcurie of radium or about 2×10^{-3} disintegrations per minute per gm of food. The use of high purity samples of elements in the experiments reduced the possibility of such contamination. In addition as a check on the above items, careful background blank counts were taken on each sample and compared with the activity of the sample after irradiation.

6. SUMMARY

This work substantiates the belief that no radioactivity is formed when 24 elements common as trace elements or subsitutents in food are irradiated in a 10-kilocurie, cobalt-60 source. Two extensions of the work however should be made. For one a sensitive method should be utilized for detecting the weakest radiations without self-absorption and container-absorption effects. The use of a relatively large volume of a liquid scintillator such as used in low-level carbon-14 and tritium tracer work might give the required detection sensitivity in this weak radiation range[13,14,15]. Chemical problems of dissolving the element and scintillator in a mutual solvent would, however, add difficulties to this approach.

In addition, this work was further limited by the fact that it was done with cobalt-60, whose two gamma rays have an energy of 1.33 and 1.13 mev. Any commercial method of food sterilization would probably use gross fission products in some form. The gamma spectrum emitted by such a source depends on the age of the fission products and the energy of the neutrons producing the fission. The contribution of radiations from particular fission-product isotopes has been presented as a function of age for uranium-235 thermal neutron fission by Hunter and Ballou[16]. From their work it can be assumed that many of the gamma rays will have energies equal to or lower than those of the cobalt-60 radiations.

However, small percentages of higher energy gamma rays may be present in the fission products from an operating reactor after a few weeks or months of "cooling". The energy spectrum of "thick" sources of these products thus could start at a much higher energy than the cobalt spectrum. Since the early x-ray work showed that a number of elements were capable of excitation at energies between 1.5 and 3.0 mev, the experiments reported here should be repeated with kilocurie or larger sources of fission products. Suitable sources now exist in the gamma ray facility of the Materials Testing Reactor, Arco, Idaho, where spent fuel rods from the reactor are allowed to "cool down" before processing. Samples can be irradiated there in an area of high-flux fission-product radiation.

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PART IV. COOPERATIVE RESEARCH WITH MICHIGAN MEMORIAL-PHOENIX PROJECTS

A. ANIMAL-FEEDING EXPERIMENTS

Personnel:

Supervisors and Consultants: H. C. Eckstein, Professor of Biological Chemistry; L. E. Brownell, Supervisor of Fission Products Laboratory and Professor of Chemical and Metallurgical Engineering; L. L. Kempe, Assistant Professor of Bacteriology and Assistant Professor of Chemical and Metallurgical Engineering

Laboratory Personnel: H. O. France, Biochemist and Research Associate; R. J. Rose, Research Assistant; R. J. Welch, Research Assistant; R. C. Dennis, Statistician; E. Ambo, Laboratory Assistant

1. INTRODUCTION

A long-term, animal-feeding experiment was initiated January 31, 1954, to determine the effect of a diet in which the major nutrients had been irradiated with 4 megarep of cobalt-60 gamma rays on growth, reproduction, longevity, and the development of pathological conditions through four generations of rats. Data on growth and reproduction performance during the first and second breeding periods as well as the results of hematological studies are presented. Factors which may have a bearing on the suboptimal breeding record are discussed in some detail.

Pilot studies in which the canned meat constituent of the diet was irradiated with high doses and fed by stomach tube to a group of rats are also described, as well as studies involving the effect of the 4-megarep experimental diet on reproduction in mice.

2. PROCEDURE

a. Diet

Changes made in fat and vitamin E supplements at the end of the first breeding period are given in Table 47. Otherwise the composition of the diet is as given in Progress Report 6 (Table 29,p.171).[1] Composition of salt and vitamin mixtures and of the canned meat are given in Progress Report 5 (Tables 19-24, pages 113-117).[2]

b. Management of the Colony

A manual of procedure was completed for the guidance of the laboratory personnel which describes in detail the methods followed in the preparation of the diet and the care of the animals. H. O. France was responsible for the immediate supervison of the experiment until July 1, 1954, after which R. J. Rose assumed this responsibility.

c. Reproduction Studies

The parent-generation animals for the long-term experiment were first mated on April 21, 1954, when they were 121 days old. From each of the two groups of 62 rats, 20 males and 20 females were selected so they would be representative of the entire group in mean weight and in range of weights. The rats were subdivided into groups of 4 males and 4 females; each female in the subgroup was exposed in turn to each of the 4 males in the corresponding subgroup. For the first two pairings the animals were rotated at 2-week intervals and thereafter at weekly intervals. The principal reason for the longer interval between changes at the start of the breeding study, as well as for the delay beyond the scheduled age of 100 days for mating, was doubt concerning the condition of the vitamin premixes at the time originally planned for the start of the breeding period.

In the first breeding period each female was paired with up to 4 males; in the second breeding period pairing was continued with changes at weekly intervals either until pregnancy ensued or until the termination of the study.

The second breeding period was begun on June 28, 1954. In view of the poor record exhibited by the animals in the first breeding period, all the 124 animals in the colony were assigned to the second breeding experiment. Thus, for 11 pairs of animals in each group, the breeding period starting June 28 was the first time they had been mated. Each female that

TABLE 47

TIME SCHEDULE OF EVENTS IN THE LONG-TERM FEEDING AND BREEDING STUDIES

December 21, 1953	Date of birth of the 124 Holtzman rats which comprise the parent generation
January 13, 1954	Date of receipt of animals and start of feeding of experimental diet
January 16, 1954	Corn oil content of diet reduced from 10 to 5% on a dry basis; corn starch substituted
April 21, 1954	Start of first breeding period; 20 animals of each sex from each group mated at 121 days of age
April 27, 1954	Use of tocopherol corn oil premix discontinued
May 16, 1954	Extra tocopherol supplement of 10 mgm weekly fed to all rats directly in addition to that given in the diet
June 16, 1954	All tocopherol fed by direct oral administration to nonpregnant females at the rate of 60 mgm per week in three feedings, pregnant and lactating animals at the rate of 120 mgm per week and males at the rate of 10 mgm per week
June 22, 1954	Cellulose content increased from 4 to 5% on a solids basis in the diet in an attempt to counteract diarrhea in some of the lactating and suckling rats
June 28, 1954	Start of second breeding period for half the control and experimental rats (189 days old)
June 30, 1954	Cod liver oil withheld from diet; corn oil substituted
July 14, 1954	Cod liver oil added to diet at a 1% level; corn oil at a 9% level on a solids basis
July 15, 1954	Start of second breeding period for the remainder of the rats

had produced a litter during the first breeding period was allowed at least 10 days rest before being remated. Figure 82, Progress Report 5, outlines the plan being followed in the reproduction studies.

3. RESULTS

a. Long-Term Studies

(1) Growth. Figures 51 and 52 show the mean weight gain in the long-term experiment of the male and female rats that were fed from 22 days of age on the partially irradiated and nonirradiated semisynthetic diet. Ten weeks after weaning, the 31 males on the nonirradiated diet weighed an average of 384.2 gm and the 31 males on the partially irradiated diet weighed 370.6 gm. The 12 male rats in the first pilot study, of the same strain and from the same supplier but fed Rockland rat diet, had a mean body weight of 260 gm at this age (Progress Report 5, Figs. 83-85, page 122), approximately two-thirds of the weight of the long-term control males.

The growth rate obtained by Anderson and Smith[3] for the first 4 weeks after weaning with a natural diet supplemented with a paste high in fat, protein, and vitamin content was an average of 8 gm per day for the fastest growing male. This diet was stated by Dunn[4] to be near optimal for rat growth. The best growth rate attained by any individual parent-generation rat in the long-term experiment during the first 4 weeks after weaning was 8.2 gm per day observed for a male on the partially irradiated diet. The overall average daily weight gain for the male rats in the long-term experiment over this period was about 6.3 gm. The weight gain of the Flb animals for the first 4 weeks after weaning was slightly superior to that of the parent animals for the corresponding period.

(2) Reproduction. The results of the reproduction and lactation performance of control and experimental rats in the first and second breeding of the parent generation are given in Tables 48 and 49. The latter table also includes data on the performance of rats mated for the first time in the second breeding period. At the time the data were compiled, 18 of the 62 females had not given birth to young; consequently, the results for the second breeding period may be incomplete.

Male sterility was high; in the first breeding period 40 percent of the control and 45 percent of the experimental males were exposed without success to 1 or more females later proven fertile. The corresponding figures for the second breeding period were 45 percent for the control and 35 percent for the experimental males.

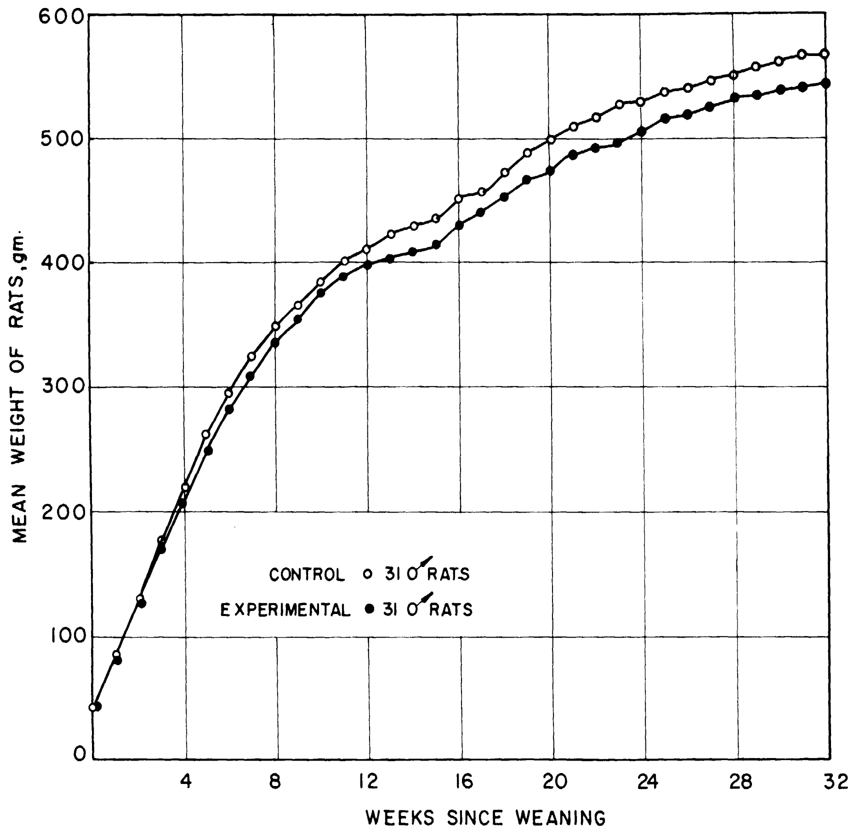

Fig. 51. Mean Weight Gain of Male Rats on Long-Term Experiment

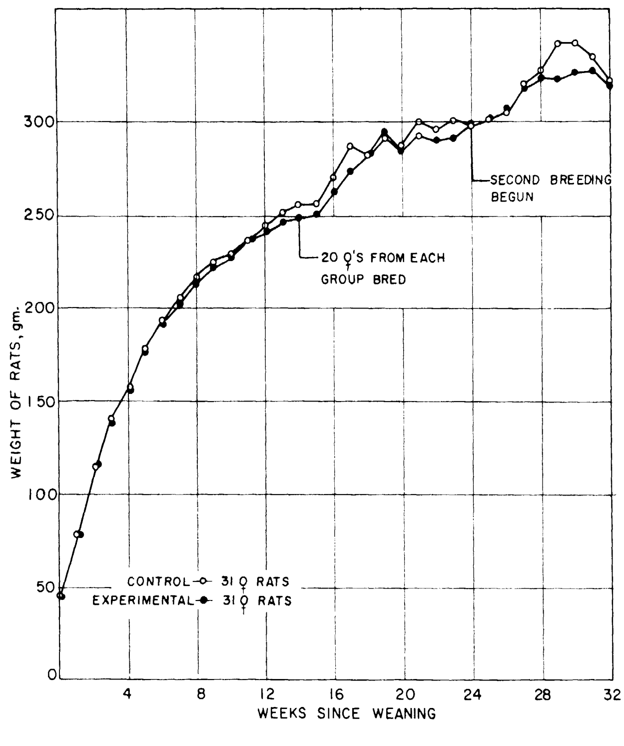

Fig. 52. Mean Weight of Female Rats on Long-Term Experiment

TABLE 48

REPRODUCTION AND LACTATION PERFORMANCE IN FIRST BREEDING (Fla) ANIMALS IN LONG-TERM EXPERIMENT

	Control Diet	Irradiated Diet
Number of females	20	20
Number of males	20	20
Number of females not conceiving after four matings	2	5
*Number of males probably sterile	8	9
**Number of males not proven	2	3
Number of females conceiving the first week	8	3
Number of females conceiving the second week	1	3
Number of females conceiving the third week	3	4
Number of females conceiving the fourth week	0	0
Number of females conceiving after the fourth week	4	5
Number of females resorbing fetuses	2	0
Number of litters born	16	15
Number of litters born dead	1	1
Number of litters born alive of which none survived until weaning	1	1
Number of pups born	157	119
Average number of pups born per litter	9.8	7.9
Average number of pups per litter at 5 days	8	6.6
Average number of pups per litter at 21 days	8	7.4
Number of pups reaching weaning	127	82
Number of pups born dead	5	3
Number of pups born alive not surviving until weaning	25	21
Average weight of pups at 21 days (gm)	46.8	45.5
Average number of pups weaned per female bred	6.4	4.1

*Mated unsuccessfully with at least one female which later became pregnant by another male.

**Mated only with animals which later proved sterile or which were pregnant at time of mating.

TABLE 49

REPRODUCTION AND LACTATION PERFORMANCE IN SECOND BREEDING OF PARENT ANIMALS OF LONG-TERM EXPERIMENT

(Incomplete)

	Animals Bred for the Second Time at 189 Days of Age		Animals Bred for the First Time at 189 Days of Age		Combined Results of the Second Breeding Period	
	Control	Experimental	Control	Experimental	Control	Experimental
Number of females	20	20	11	11	31	31
Number of males	20	20	11	11	31	31
Number of females not conceiving after at least six matings	2	6	4	6	6	12
Number of males probably sterile	10	5	4	6	14	11
Number of males not proven	2	2	3	7	5	9
Number of females conceiving the first week	5	4	0	1	5	5
Number of females conceiving the second week	3	4	1	1	4	5
Number of females conceiving the third week	4	3	0	1	4	4
Number of females conceiving the fourth week	3	2	1	0	4	2
Number of females conceiving after the fourth week	1	1	3	1	4	2
Number of females resorbing fetuses	2	0	1	1	3	1
Number of litters born	16	14	5	4	21	18
Number of litters born entirely dead at birth	1	0	0	0	1	0
Total number of litters born alive of which none survived until weaning	1	3	3	2	4	5
Total number of pups born	145	115	37	37	182	152
Average number of pups born per litter	9.1	8.2	7.4	9.2	8.7	8.5
Average number of pups per litter at 5 days	6.1	5.6	4.4	2.3	5.4	5.1
Average number of pups per litter at 21 days	6.1	5.6	4.4	2.3	5.4	5.1
Number of pups reaching weaning	97	81	22	9	119	90
Number of pups born dead	17	11	2	4	19	15
Number of pups born alive not surviving until weaning	30	23	16	28	46	51
Average weight of pups at 21 days (gm)	57.8	52.7	55.0	56.1	58.0	55.4
Average number of pups weaned per female bred	4.9	4.1	2.0	0.8	3.6	2.8

The incidence of sterility in females was lower than in males; 10 percent of the control females and 25 percent of the experimental females apparently did not conceive after exposure to 4 males in the first breeding period. Data for the second breeding period are incomplete. At the time of writing a combined total of 76 pregnancies of a possible 102 occurred. The females of each group had 51 opportunities to produce litters; the control females succeeded 37 times, and the experimentals 33 times. Resorption gestation was responsible for a total of 6 reproduction failures.

The combined total of young produced by the control females was 339 and for the experimental females, 271. Early mortality was high with practically all the deaths occurring within 24 hours of birth. In the first breeding period about 18 percent of the young of the control females died before 21 days of age; in the second breeding period 32 percent of the young of multiparas and 43 percent of the young of primaparas died before weaning. The corresponding figures for the experimental animals were about 21, 29, and 76 percent, respectively. Combined early mortality was about 28 percent for the control group and 33 percent for the experimental group.

The total weight at weaning of the offspring of the 20 control females bred twice was 5.9 kg for first litters and 5.6 kg for second litters. For the offspring of the corresponding experimental females the total first litter weight was 3.7 kg; total second litter weight was 4.3 kg.

Reproduction records of individual animals reveal that only 3 control females showed greater success in ability to bear and raise second litters, whereas 12 of the 20 experimental females showed improvement. Of the 87 females that did not conceive during the first breeding period, 6 (2 that were controls) did so during the second but only 1 control and 1 experimental litter survived to weaning.

The results, as yet incomplete, indicate high incidences of male fertility and early deaths in the young. Overall performance of experimental animals in bearing and raising second litter young was improved in most respects over the first breeding-period performance. Control animals were less successful in all categories except weight of young at weaning. If the change made in the vitamin E and cod liver oil supplementation at the end of the first breeding period had any beneficial influence, this was reflected principally in the increased weights of the young at weaning. Animals fed partially irradiated diets were somewhat inferior in reproduction and lactation performance to those fed nonirradiated diets in almost all categories.

Figure 53 shows photographs of a typical male, female, and litter from both the control and experimental groups of animals. The corresponding animals in both groups have nearly identical appearance as is shown by comparison of the photographs.

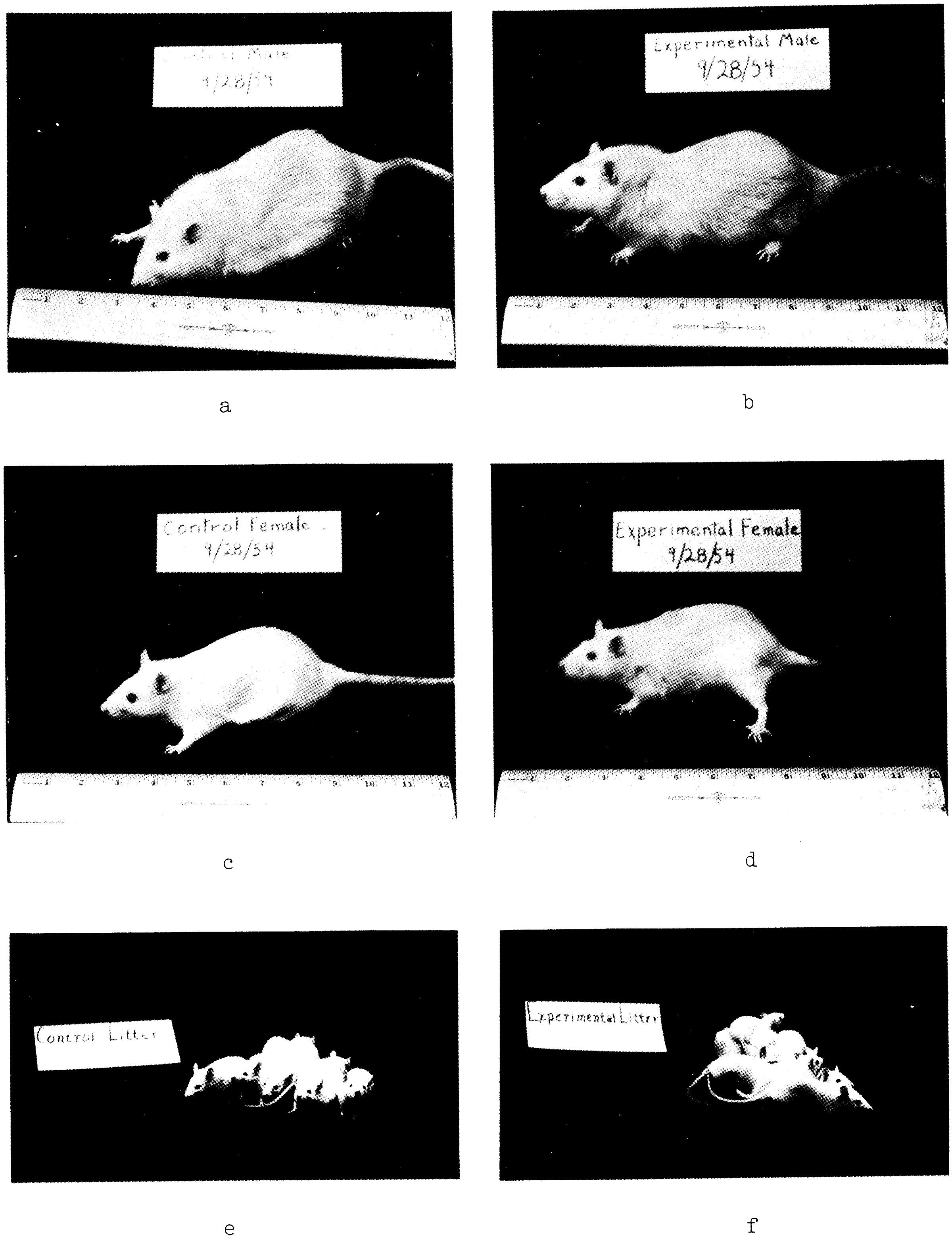

Fig. 53. Typical Animals from Long-Term Feeding Experiment Using Experimental Diet Receiving Radiation of 4 Megarep.

(3) Hematology. Table 50 gives the results from the hematological studies on parent rats in the long-term experiment. The table lists the arithmetic average of tests on 27 individual animals. The table indicates no significant difference between control animals and animals on the experimental diet. In addition to the studies reported in Table 50 brilliant cresyl blue films were examined for platelets and no marked differences were observed.

(4) Histopathology. Figure 54 shows a rat which developed a lateral tumor at approximately 8 months of age. This growth first became apparent August 16, 1954. The picture was taken 9 days later. The period of most rapid growth for this particular tumor seemed to be during the first 10 days after its detection. The tumor continued to grow steadily and 40 days after detection the infected rat appeared sickly and emaciated. The animal was sacrificed and an autopsy made. Table 51 shows the autopsy report.

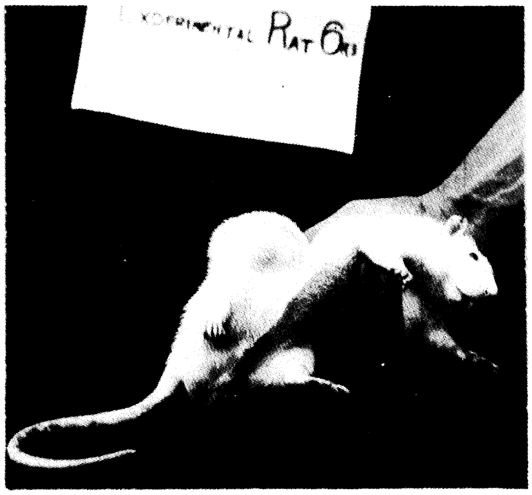

Fig. 54. Male Rat on Irradiated Diet Showing Development of Lateral Tumor at the Age of 8 Months.

Another rat developed a scrotal tumor which was detected for the first time August 23, 1954. This tumor developed so rapidly that on September 10, 1954, it seemed advisable to sacrifice the animal in order that the tumor might be removed before the rat died. Because of the rapidity with which this particular tumor grew, and because of the haste with which the sacrifice had to be performed, no pictures of the animal were obtained. Figure 55 shows a photograph of the tumor after pathological examination. At the time of sacrifice the rat weighed 682 gm. The tumor when removed weighed 140 gm or approximately 21 percent of the total body weight. The heart, lungs, liver, spleen, kidney, adrenal gland, a section of the small intestine, and the testes were removed from the animal and histopathological examinations made of these organs. The complete report of the pathologist appears in Table 52.

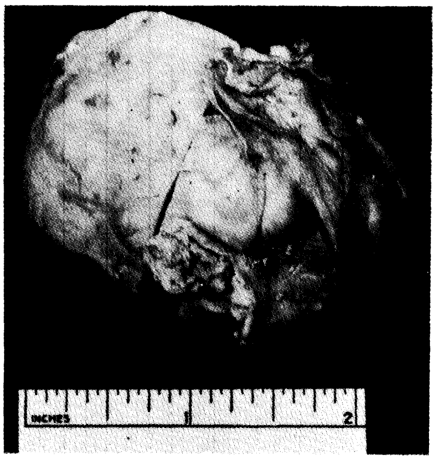

Fig. 55. Scrotal Tumor from Rat on Irradiated Diet (after Pathological Examination).

Both of the animals developing tumors were of the experimental group. These two are the only animals which have developed tumors and the number is, of course, too small to be of statistical significance. However, Mr. Holtzman, of the Holtzman Company which supplied the

TABLE 50

AVERAGE OF RESULTS OF HEMATOLOGICAL STUDIES ON INDIVIDUAL PARENT RATS IN LONG-TERM EXPERIMENT*

Group	Hemoglobin, gm/100 cc	Hematocrit, %	Reticulocyte, %	White Blood Count, No./cu mm	Eosinophils, No./cu mm	Differential Count: Lymphocytes, %	Differential Count: Polymorphonuclear Leukocytes, %
Control Males	14.9	47.4	2.9	10,760	119	80.0	18.1
Control Females	14.5	45.5	3.0	9,550	120.0	81.0	17.3
Experimental Males	15.0	47.7	2.5	13,390	113.0	83.7	14.7
Experimental Females	14.8	45.9	1.7	11,260	128.0	81.7	16.3

*Five to seven animals were used for each test reported.

TABLE 51

HISTOPATHOLOGICAL REPORT $D2R_3$, 2425-LBG ON
EXPERIMENTAL MALE RAT WITH LATERAL TUMOR

The neoplasm is an adenofibroma of the mammary gland. There is proliferation, in an orderly fashion, of ductal epithelium and an extensive periductal proliferation about the numerous ducts. The fibrous tissue is actively proliferating and is cellular. Neither element is pleomorphic enough to be considered malignant. The tumor may not represent a true neoplasm.

Tissue	Observation
Heart	Moderate acute passive congestion. No lipidosis.
Lung	Emphysema. Fat stain negative.
Spleen	Small foci of erythropoiesis.
Liver	Moderate lipidosis in hepatic cells and Kupffer cells. Rather diffuse distribution, but the center of lobules not involved.
Small Intestine	Negative.
Adrenal	Abundant cortical lipids.
Kidney	Thickening of basement membrane of tufts with ischemia. Occasional gray casts in tubules. Glomerulonephrosis. No lipidosis.
Vertebral Column	After decalcification, no evidence of avitaminosis-A, -C, or -D. Marrow and visible nerves normal.
Testes	Marked degenerative changes in tubular epithelium. Aspermatogenesis. No mitotic figures present. Epithelium completely lost in many. Some tubules contain necrotic cells; others are calcified.

Summary: There is no satisfactory evidence that the lipidosis of the liver represents liver damage. The lipid is probably dietary lipid and its appearance is in response to the high fat diet. The renal changes indicate early reversible damage. The degenerative changes in the testes are definite and extensive. Adenofibroma of the mammary gland.

TABLE 52

HISTOPATHOLOGICAL REPORT $D4R_1$, 1832-LBG ON EXPERIMENTAL MALE RAT WITH SCROTAL TUMOR

Tissue	Observation
Heart	No significant abnormality of the myocardium. No lipidosis.
Lung	Slight peribronchial lymphocytic infiltration, fat stain negative.
Spleen	A few lymphoid follicles remain but most of the spleen is replaced by tissue like bone marrow. Erythropoietic and granulopoietic cells and numerous megakaryocytes present. Myeloid metaplasia.
Small Intestine	Negative.
Liver	Acute passive congestion. Very slight lipidosis.
Kidney	Negative. No lipidosis.
Testes	No spermatozoa present. The seminiferous epithelium presents various stages of atrophy and necrosis. The pattern is variable in different areas. In the more nearly normal tubules spermatogenesis is abnormal. No mitotic figures present. There are bizarre cellular forms, some with large dark nuclei, and others with many nuclei. Interstitial cells are not increased in number.
Scrotal Tumor	A moderately well-differentiated spindle cell supporting tissue neoplasm with many minute blood vessels. Angiofibrosarcoma. Tissue growing by expansion and invasion; no capsule can be identified. No metastases have been found. This is compatible with the lack of anaplasia of the neoplasm. Areas of necrosis are present within the tissue.

Summary: Well-differentiated angiofibrosarcoma of the scrotum. Atrophy and necrosis of the seminiferous tubules with the production of bizarre multinucleated forms. Marked myeloid metaplasia of the spleen.

parent animals, commented that he had never previously observed such tumors in this strain of rats. A point of interest, in view of the high incidence of male sterility in the long-term experiment, was finding that both of these rats showed extensive degenerative changes of the testes.

b. Pilot Studies

(1) Second Litter Breeding Performance of Rats Fed 2-Megarep Diets for 3 Months. Table 53 gives the final second litter data for the rats in the first pilot study. A comparison with the results of first litter performance (Table 26, page 124, Progress Report 5) shows a somewhat decreased reproductive capacity after 3 months on the experimental diet as compared with 10 days on the diet prior to the first mating.

TABLE 53

REPRODUCTION DATA ON SECOND LITTERS OF RATS
IN 2-MEGAREP PILOT EXPERIMENT

	Nonirradiated Diet	Partially Irradiated Diet	Completely Irradiated Diet
Number of females bred	4	5	5
Numbers of litters	2	4	5
Average number per litter at birth	9.0	9.8	10.3
Average number per litter at weaning	8.0	8.3	8.2
Average weight at weaning (gm)	50.0	41.5	38.6
Average number young weaned per female bred	4.0	8.3	8.2

In the first breeding period (starting June 1, 1953) all females produced young; in the second (starting August 10, 1953) 3 did not. The total number of young weaned were 128 and 90, respectively; mean weights at weaning were 54.4 gm and 41.5 gm, respectively; number of young weaned per female bred was 9.0 and 6.4, respectively. These are the results of pooling data for the control and experimental groups. The numbers of animals in each group are too small to permit valid conclusions to be drawn about differences between groups. The only category in which control animals were inferior, based on the limited data, was in number of females not producing young.

The diet fed in the 2- and 20-megarep pilot studies differed from that fed in the long-term study (Table 29, page 171, Progress Report 6) in that it contained 5 percent more corn oil. In these groups, as in the long-term group, the experimental diet appeared to have an adverse effect on reproduction apart from irradiation-induced changes.

(2) Reproduction Data on Offspring of Rats Fed 20-Megarep Completely Irradiated Diet. Animals fed a completely irradiated 20-megarep diet developed characteristic symptoms of vitamin deficiencies as described in Progress Report 6. When a nonirradiated vitamin mixture including tocopherol was added in the original amount to the 20-megarep completely irradiated diet, the reproductive performance was on a par with the animals in the long-term experiment. Figure 56 shows an original pair of animals on the XC diet (completely irradiated at 20 megarep) after receiving the nonirradiated vitamin supplements. Figure 56 should be compared with Figs. 70 through 72, pages 188 and 189, of Progress Report 6, which shows animals with characteristic symptoms of vitamin deficiencies. The offspring of the animals shown in Fig. 56 were placed on stock rations (Rockland rat diet) and produced normal litters averaging 10 young per litter for six litters. Figure 57 shows one such litter.

Fig. 56. A Pair of Animals after Recovery from Vitamin Deficiency Produced by XC Diet (Completely Irradiated at 20 Megarep).

Fig. 57. Litter from Animals Shown in Fig. 56.

(3) Histopathology of Rats on 20-Megarep Diet. Twenty-eight of the animals used in the 20-megarep pilot study were sacrificed for histopathological examination. Organs were identified only by code number at the time of examination. The summary of results after decoding is given in Table 54. Pathologists,R. C. Hendrix, M.D. and R. C. Wanstrom, M.D., expressed the opinion that no significant tissue changes induced by the diet are indicated by the results shown in Table 54.

(4) Forced Feeding Studies. The effect of forced feeding of diets containing meat irradiated at 23 to 45 megarep was studied by observing

TABLE 54

HISTOPATHOLOGICAL CHANGES IN PILOT STUDY RATS ON 20-MEGAREP DIET

Tissue	Observation	Nonirradiated Diet	20-Megarep Partial Irradiation	20-Megarep Complete Irradiation
Heart	No change	6	12	5
	Congestion	1	1	0
	Pale staining	1	0	0
Lung	Emphysema	6	15	6
	Early lobular pneumonia	1	1	0
	Occasional lipophages	1	3	0
Liver	No change	0	0	1
	Lipidosis, slight to moderate	2	11	3
	Lipidosis, well marked	4	4	1
	Lipid in Kupffer cells	4	3	0
Spleen	No change	1	4	2
	Congestion	2	8	1
	Hemophages	1	3	0
	Small foci of hematopoiesis	2	1	2
Small Intestine	No change	3	10	5
	Mucin, moderate to abundant	2	3	0
	Chronic catarrhal enteritis	1	0	0
Kidney	No change	2	5	2
	Congestion	1	4	0
	Cloudy swelling	2	2	3
	Lipidosis	1	5	0
	Dilated tubules containing inspissated protein	1	0	0
Adrenal	Cortical steroid abundant	5	7	3
	Patchy loss of cortical lipids	1	1	2
NUMBER OF ANIMALS		7	15	6
COMMENT	Depletion of stored fat	2	3	2

the changes in body weights of adult male rats. The purpose of this experiment was to determine the possible threshold for the development of acute toxicity in irradiated canned meat. A previous exploratory study in which the rats were allowed to eat stock ration ad-lib between the forced feedings, and in which part of the total food intake was given by stomach tube had indicated that tube-fed meat irradiated at 25 megarep did not produce any different effect on weight change than did nonirradiated meat. The tube feeding in itself was, however, sufficiently stressful to cause a marked reduction in weight gain when compared with controls not tube fed.

A group of 50 male Holtzman rats averaging 355 gm at the start of the experiment were tube fed the nonirradiated diet for 7 days. Ten animals were eliminated because they showed signs of respiratory infection, or responded unfavorably to the feeding procedure. The remaining 40 rats were divided into two equal groups of similar weight (about 312 gm). One group was used as a tube-fed control and the other group was tube fed a diet containing canned meat irradiated at 23 megarep. The composition of the diet was canned meat (Swift's beef for babies), 1000 gm; sucrose, 70 gm; corn oil, 15 gm; HMW salt mixture, 10 gm; cellulose, 20 gm; tocopherol, 0.3 gm; choline chloride, 1.2 gm; and vitamin mixture, 15 gm. The composition of the various mixtures was given in Progress Report 5.

Water was added initially to give a solid content of about 20 percent to make the diet more fluid as it was observed that the canned meat exposed to this level of irradiation developed a rubbery consistency. After mixing in a Waring blender the diet containing the irradiated meat adhered tenaciously to the walls of the containers; large volumes of water were required to wash down the adhering meat. Even with this precaution the quantitative recovery of all the solids was difficult, therefore, the dry weight of each batch was determined and the solid content of both diets adjusted to 16 to 18 percent. Sedimentation occurred more rapidly in the control diet, necessitating stirring between each sample taken for feeding. The fluid diet was carefully measured from a 30 ml hypodermic syringe and the rats were fed twice daily, except on Sundays when they were fed once. The amount given per feeding was 20 ml, a quantity that maintained body weight at a fairly constant level of about 275 gm from the third to the sixth week of tube feeding. Weight curves are shown in Fig. 58.

The irradiation exposure of the meat was increased gradually to 45 megarep over the first 2 weeks. Inability to irradiate sufficient material to supply the needs of the original 20 animals in the initial experimental group at the higher level led to a reduction in the size of control and experimental groups to 12 each on the 19th day of the experiment. The average weights of the animals in each of the groups of reduced size was the same as in the corresponding group prior to reduction of the group size.

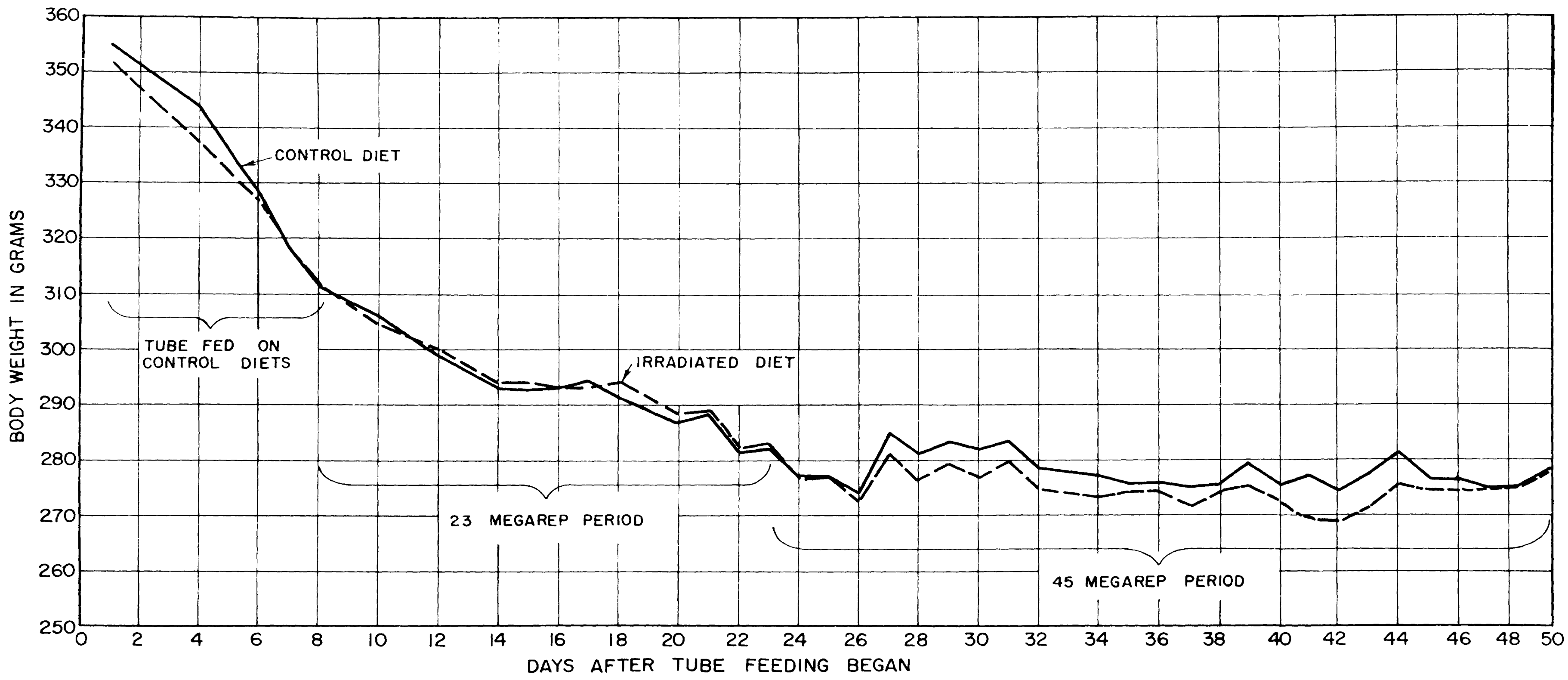

Fig. 58. Weight Curves in Tube-Feeding Pilot Experiment Using 23- to 45-Megarep Irradiated Meat

The animals removed from the experimental diets were kept under observation for several weeks after their return to ad-lib feeding on the stock ration. Weight changes were followed for at least 2 weeks, at which time the original weight of both groups had been regained. No differences were observed between the two groups during the period of initial weight loss or during the recovery period. Diarrhea occurred in some of the animals fed the irradiated meat but an increase in the roughage content abolished this. Otherwise, there were no grossly observable symptoms of any deleterious action of forced feeding of a diet consisting largely of meat irradiated with doses up to 45 megarep.

(5) Studies on Mice Fed 4-Megarep Diet. Reproduction studies and other observations were made on two generations of mice fed the same diets that the rats received in the initial phases of the long-term experiment. Two groups of mice, one composed of highly inbred animals of the Bagg albino strain and the other of mixed-strain, pigmented animals, were transferred from Purina Laboratory chow to the experimental diets at the time they were bred. Six Bagg albino and 16 mixed-strain female offspring of the animals on irradiated and like numbers of the offspring of the animals on the nonirradiated diet were continued on the diets that their parents received. An additional control group was set up by transferring 6 albino and 16 mixed-strain females from the experimental rations to Purina Laboratory chow at the time they were mated for the production of the second filial generation. Parent-generation mice were not removed from the breeding cages for rearing their young, whereas pregnant females of the first filial generation were put in separate cages when they became pregnant. This change in procedure was accompanied by an apparent overall improvement in reproduction performance for second generation animals.

Results of the reproduction studies on the first two generations, not complete at the time of writing, are presented in Table 55. The breeding potential of the albino mice was too low to permit setting up groups of adequate size and a high incidence of sterility among the albino males of the first filial generation necessitated the replacement of the albino males with mixed-strain males of proven fertility. None of the albino females on the irradiated ration had produced young by the time they were 6 months old and both the male and female animals on this diet developed skin lesions with loss of hair and inflammation of the eyes. Reproduction performance of the albino females fed the nonirradiated rations appeared to be normal. The data are included despite the small numbers of animals because the results suggest that the Bagg albino mice may be highly sensitive to dietary changes produced by irradiation, at least on the experimental ration fed.

The mice of the first filial generation which were transferred to Purina Laboratory chow when they were mature lagged behind the animals

TABLE 55

DATA ON REPRODUCTION OF TWO STRAINS OF MICE FED 4-MEGAREP PARTIALLY IRRADIATED DIETS

	Parent Generation				First Filial Generation					
	Nonirradiated Ration		Irradiated Ration		Nonirradiated Ration		Irradiated Ration		Purina Lab Chow	
	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)
Number of females	7	8	5	8	6	16	6	16	6	16
Number of litters born alive	5	6	3	8	5	15	0	16	6	12*
Total number of young born	21	37	14	46	45	121	0	113	43	89*
Average number of young born per litter	4.2	6.2	4.7	5.8	9.0	8.1	0	7.1	7.3	7.4*
Total number of young weaned	18	24	7	46	*	105	0	97	*	78*
Average number of young weaned per litter	3.6	4.0	2.0	5.8	*	7.0	0	6.1	*	6.5*
Average weight of young at 21 days (gm)	*	*	*	*	*	9.9	0	9.8	*	9.6*
Percent of weaned young having equilibrium distrubance	0	0	0	0	*	12.7	0	24.7	0	0*

(1) Bagg albino mice
(2) Hybrid pigmented mice
* Records incomplete

allowed to remain on the experimental ration in producing young, an occurrence probably attributable to the stress that the dietary change entailed.

Reproduction and lactation performance of the mice on the nonirradiated experimental ration compared favorably with that expected of animals raised on stock ration, in contrast with the performance of the rats in the long-term experiment. The mixed-strain animals on the irradiated diet showed appreciable impairment of ability to bear and raise their young. Some of the second filial mixed-strain generation young, however, showed evidence of a neurological disorder characterized by an impaired ability to maintain equilibrium. The disorder was apparently attributable to the experimental diet and was twice as frequent in the offspring of mice fed irradiated ration as in those fed nonirradiated ration.

The weights of the young of the mixed-strain mice at the time of weaning were approximately the same for the animals on irradiated and nonirradiated experimental ration and on Purina Laboratory chow. Subsequent weight gains were also similar at least for the first two or three postweaning weeks, a result again in contrast with that observed for rats in the long-term experiment.

4. DISCUSSION

The results of the first breeding study of the long-term experiment, expressed in terms of average number of young weaned per female bred, indicate that the reproductive capacity of the animals on the irradiated diet was about two-thirds that of the animals on the nonirradiated diet. The latter animals in turn had a reproductive capacity of about two-thirds that of similar rats fed the stock ration under the same conditions. Thus, despite the superior growth of the animals fed the semisynthetic experimental ration, the diet did not appear to be optimal for reproduction and lactation.

These observations led to a review of the work of other investigators who fed diets containing ingredients similar to those incorporated in the ration fed in the present experiments. The factors which have been examined in relation to their possible influence on the suboptimal reproduction and lactation performance of the animals, including those concerned with the management of the colony, as well as those related to the composition of the diet, are discussed. Mention is also made of factors that might have a role in such differences as were observed between the animals on the nonirradiated and irradiated diets.

a. Observations Regarding the Conduct of the Experiment

(1) Infection. Despite the efforts to maintain clean, vermin-free, and constant environmental conditions as described in the previous reports, an appreciable incidence of mild respiratory infection was noted in the long-term experiment animals shortly after their arrival. Inasmuch as the colony had been free from respiratory disease prior to the receipt of the shipment of the 124 weanling Holtzman rats which comprise the parent generation, the animals apparently either had the infection when shipped or contracted the infection as a result of shipment in the middle of winter (they were in transit overnight via railway express). In support of this conclusion is the fact that young rats of the first group of approximately the same age as the new group remained free of infection. Overt evidence of infection was slight and at the time of writing only 2 of the 124 animals have displayed symptoms severe enough to warrant their isolation. The highest incidence of infection occurred in the portion of the colony in closest proximity to the air intake of the air-conditioning unit. Many of the animals infected were from the same litter of 15 offspring. Thus, either exposure to periodic episodes of overly rapid air movement or conditions during the first 3 weeks of life, or both, may have had a role in the development of the respiratory infection.

Several cases of diarrhea were noted in the first litters of the F1 generation when they were about 18 days of age. Roughage was increased from a 4 to a 5 percent level and more rigorous sanitation measures were undertaken. These measures did not appear to eliminate the diarrhea entirely, but as only one of the more than 200 weaned young died in the first 1 to 2 months of their post-weaning life or have shown evidence of infection other than of the respiratory tract on autopsy, the diarrhea is tentatively ascribed to some peculiarity of the diet rather than to paratyphoid or other infection. Several of the mothers had transient diarrhea when the young were near weaning age and one, a control, had diarrhea and respiratory infection with marked emaciation which disappeared after weaning. The infections were about equally distributed between the groups on the nonirradiated and on the partially irradiated diets.

(2) Feeding Schedules and Diet Composition. No deficiencies appear to have occurred in feeding or watering schedules. Food and water vessels were washed daily and drinking tubes were not interchanged except after sterilization.

A transitory leveling off of the growth curve was observed during the 11th and 12th experimental weeks. The vitamin premixes were believed to be at fault in that the water-soluble vitamin mixture had been allowed to remain at room temperatures for several days and the tocopherol-corn oil premix contained a brown sediment at the time of the growth plateau. The tocopherol used in this mix was found to be discolored and malodorous. The premixes and the tocopherol were discarded. The water-soluble vitamin premix is regularly stored up to a week or 10 days at a -25°F temperature. As indicated in Table 47, the use of a tocopherol-corn oil premix was discontinued.

The diet on hand at the time of the decline of weight gain, which diet was made up from the questionable premixes, was fed to groups of rats and mice not in the long-term experiment to compare the effects on growth with diet made up from fresh vitamin mixtures. A slight decrease of weight gain occurred in the groups on the questionable diet. However, later examination of the growth data obtained by Rice[5] of Swift and Company in a similar study in which raw irradiated or nonirradiated hamburger was a large constituent of the diet disclosed a growth plateau in their male rats at approximately 10 weeks of age. Perhaps, therefore, the slight transient decrease in growth rate was largely due to a characteristic of the animals or of the diet rather than to a temporary vitamin deficiency.

The conditions under which the diet and the diet ingredients are stored may influence their nutritional value. For example, rancid fats interfere with growth (Greenberg and Frazer[6]), promote liver, lung, and other damage (Abell, et al.[7] and Luttrell and Mason[8]), and impair reproductive capacity in the rat (Kennelly and Quackenbush[9]). Rancid fats in the diet also destroy vitamin E (Gallego[10]). The injury caused by rancid fats is ameliorated but not prevented completely by vitamin E administration (Burr and Barnes[11] and Fitzhugh, et al.[12]).Storage at temperatures of 20°C are reported to inhibit the development of rancidity (Fitzhugh, et al.[12] and Morris, et al.[13]).

The mixed diets, except for the 44-hour period during which the irradiated portion is in the source room, are kept under refrigeration as are all the ingredients except the salt mix and corn starch. During mixing and feeding, however, the diets are at room temperature which at times exceeds 30°C. The diets are kept for no longer than a week before supplementation with vitamins, oils, and minerals and only 3 days (over week ends) after supplementation.

(3) Sanitation. A temporary shortage of caretaking assistance during the first breeding period led for a time to less than optimum conditions of breeding-cage cleanliness. The animals were at first allowed to raise their litters in 7"x9" Hoeltge cages fitted with metal floors on which wood shavings were placed. These small cages quickly became wet and unsanitary; loose food, feces, and water from bottles which drained into the shavings caused these conditions. The correction of the unsanitary conditions did not improve the performance of the mothers in raising their young to weaning age to any substantial degree, nor did it eliminate the diarrhea in young and mothers that occurred in some litters approaching weaning age.

The diarrhea disappeared without treatment after weaning. The wood shavings and shredded paper currently used in the breeding cages are sterilized by exposure to gamma radiation from the cobalt source. As stated in Progress Report 6, a manual of procedure has been prepared to inform the personnel of proper management of the colony.

b. Dietary Factors

Of significance with respect to the parallel occurrences of excellent growth and inferior reproduction and lactation performance in the present experiment is the statement of Osborne and Mendel[14] that "in accelerated growth there may be actual 'distortions' that are detrimental rather than advantageous for certain physiological functions". The observation of Mayer[15] that "a diet promoting good economical growth in youth tends to promote fat deposition in adulthood, and possibly the development of degenerative diseases" supports this assumption. Callison, et al.[16] reported findings indicating that human diets which promoted greatest initial weight gain in rats (diets rich in fat) were associated with a high incidence of lung and skin lesions.

These and similar observations which link rapid weight gain with disturbances of body function may be interpreted as indicating that excessive caloric intake may sometimes be harmful in itself. Attention is given in this section, however, to the possibilities that deficiencies or toxic levels of essential micronutrients or the incorporation in the diet of certain of the vitamin-rich supplements as well as the fat content may have influenced the animals adversely, especially insofar as reproduction is concerned.

(1) Vitamin A. During the period when the cod liver oil constituted 5 percent of the diet, the average mature male rat assimilated over 1300 units of vitamin A daily, assuming that no destruction of the vitamin occurred after it was mixed into the diet. At the level of cod liver oil fed after July 14, 1954, the vitamin A intake was reduced to 170 and 260 units daily, respectively.

Requirements for normal growth and reproduction of the rat are given by McCoy[17] as 30 to 50 units per day. Vitamin A is susceptible to oxidative destruction and according to Lease, et al.[18] loses its activity when mixed together with rancid or ozonized fats. But, in view of the large excess of vitamin A over minimal requirements, the possibility that sufficient destruction occurred through oxidative changes in the irradiated portion of the diet to affect the reproduction of the rats appears remote.

The amount of vitamin A ingested seems to be sufficiently low to be nontoxic and below the dose of 50,000 units which has been reported to result in hemorrhagic tendencies, termination of pregnancy in the rat (Moore and Wang[19]), and hypoprothrombinemia (Light[20]). No evidence of thrombocyte deficiency or of impaired clotting time was noted during the hematological examinations of the 6-month-old rats. Vedder and Rosenberg[21] found that feeding jewfish liver oil, a rich source of vitamin A, in quantities which supplied up to 100,000 units per day caused bone fragility, growth failure, and hemorrhagic tendencies in the rat. They showed, however, that the toxicity was not related to the vitamin A content but rather to the highly unsaturated fatty-acid fractions of the oil.

(2) Vitamin D. Minimal requirements of vitamin D occur at calcium phosphorus ratios of 1:1 to 2:1 when phosphorus levels are adequate (McCoy[17]). The Ca:P ration of the diet fed in the current experiment is about 4:1 but the level at which the vitamin D is supplied (about 100 units per day at a 5-percent level of cod liver oil and 20 units per day at a 1-percent level) should compensate adequately for this imbalance. Dixon and Hoyle[22] reported the daily dose for the appearance of urinary calculi to be many times higher than the antirachitic level and found the fatally toxic dose to be much above this.

(3) Vitamin E. McCoy's summary of the vitamin E requirements of the rat is as follows: about 1 mg of alpha tocopherol per day is sufficient for normal growth; 3 mgm per day permits gestation; and a single dose of 10 mgm to females on the day of littering will prevent paralysis of the suckling young.

As indicated in Table 47, tocopherol was incorporated in the diet at a level of 50 mgm per 100 gm of diet on a dry basis until June 16, 1954. Thereafter, it was fed orally in corn oil three times a week so that males received 10 mgm, nonpregnant females 60 mgm, and pregnant and lactating females 120 mgm weekly. Between May 16 and June 16, weekly oral supplements of 20 mgm per rat were scheduled in order to take care of any increased needs of pregnant and lactating females for vitamin E over those provided by the tocopherol incorporated in the diet.

The tocopherol intake at the time of the first mating, assuming no destruction on addition to the diet, was about 10 mgm per day for the average female and about 15 mgm per day for the average male. Assuming a 50 percent loss on admixture with the diet and a further 50 percent loss in the digestive tract, the vitamin E levels should have been adequate for gestation. Dr. E. E. Rice[5] of Swift and Company Research Laboratories reported a recovery of two-thirds of the tocopherol mixed into their ration. Nevertheless, the reproduction data show a number of cases of either resorption gestation or pseudopregnancy and of failure to conceive. The 20 mgm tocopherol weekly supplements fed directly near or during the time of gestation (mating had taken place about 3 weeks before the first additional supplement was given) failed to prevent deaths of the newly-born young. These young did not die with the typical symptoms of avitaminosis E, which according to Evans and Burr[22] are manifested by paralysis and death of the young after they are more than 18 days old.

The large number of cases of male sterility likewise cannot be explained by a vitamin E deficiency, for while their requirements are less than those of the females they presumably received, as a result of their higher food consumption, amounts of tocopherol in excess of those ingested by the females.

A comparison of the results of the reproductive performance of the rats in the pilot studies with those of the first mating of the animals in the long-term experiment indicates that the experimental diet, rather than the strain of animals or the conditions under which they are kept, may have been responsible for the suboptimal breeding record. The 14 females in the first pilot study (see Progress Report 5) which was designed to determine the effects of diet partially or completely irradiated with 2 megarep of cobalt-60 gamma rays weaned an average of 9.1 young per female bred. The animals which had been in the laboratory since March 27, 1953, were changed from stock ration (Rockland rat diet) to the experimental diet on May 22 and were mated on June 1. They were mated again early in August and the first young were born September 3 after the parent animals had been on the experimental diet somewhat over 3 months. This mating resulted in only 6.4 young per female bred. The best reproductive performance was among the 5 females and 4 males on the completely irradiated ration; their average was 8.1 young weaned per female bred as compared with an average of 5.4 young weaned for each of the 9 females on the partially and nonirradiated diets. The growth curves show that slightly better weight gain in the males was achieved on the completely irradiated ration, although the difference was not statistically significant (Progress Report 5).

The rats used in the 20-megarep-diet pilot study were the offspring of one animal on the partially irradiated diet and of two animals on the completely irradiated diet of the first pilot study. They were divided into four groups and placed on nonirradiated, partially, or completely irradiated diets as described in Progress Report 6. The 4 females on the completely irradiated diet did not breed; their vaginas had not opened by the time they were 100 days old. The combined record of the 9 females on the partially and nonirradiated diet was 6.7 young per female bred. The 3 control animals were not superior in breeding performance to the 6 females on partially irradiated diets, although their young were larger at 21 days of age.

Addition of nonirradiated, water-soluble vitamin mixture (powdered yeast, liver extract, and pure vitamins) to the diet completely irradiated at the 20-megarep level allowed reproduction to take place as previously reported. Further supplementation with tocopherol after irradiation, in the same amount as that added to the basic experimental ration (0.05 percent on a solids basis), resulted in a larger second litter. The offspring obtained from rats fed completely irradiated 20-megarep diet supplemented with nonirradiated vitamins were put on stock ration at about 30 days of age. These animals produced and weaned normal litters of the expected size of about 10 young.

With the possible exception of the animals fed diets completely irradiated at the 20-megarep level, no symptoms definitely attributable to vitamin E deficiency have been observed either in the pilot studies or the long-term experiment. However, in order to minimize the destruction of vitamin E in the diet and in the digestive tract, the tocopherol was omitted from the diet and fed as an oral supplement before the second breeding of the rats in the long-term experiment was started. Since Rice[5] found that 50 mgm of tocopherol per rat per week were adequate for reproduction in several generations of rats fed a diet high in meat and animal fat, a supplement of approximately this amount was decided on for the females. A report (Escudero and Herraiz[24]) of testicular damage caused by a dose of tocopherol considerably below that fed in the present experiment, as well as the smaller requirements of the male (Evans and Burr[23] and Mason[25]) seem to justify a reduced tocopherol dosage schedule for the males.

The schedule of tocopherol supplementation currently in effect calls for 60 mgm weekly for nonpregnant females and for 120 mgm weekly for pregnant and lactating females. The larger amount is given to the latter animals because of the increased requirements for gestation and because lactating females are reported to excrete 50 percent of exogenous tocopherol in their milk (Pappenheimer, et al.[26]). Males are given 10 mgm in one weekly feeding.

The alpha tocopherol is fed dropwise as a 20% solution in corn oil. Nonpregnant females receive six drops and pregnant females twelve drops three times weekly. The supplement is administered early in the afternoon several hours before the peak of nocturnal feeding occurs.

(4) Salt Content. Hubbell, Mendel, and Wakeman[27] originally fed their salt mixture at a 2 percent level, but noted that this amount might be marginal. Most workers feed the HMW salt mixture at the 4 percent level which would bring the potassium and phosphorus closer to the amounts recommended by McCoy[17] for the albino rat. Also additional supplementation with manganese, which was the procedure in the first pilot studies (Progress Report 5), is rendered unnecessary by feeding the salt mix at the higher level. Consequently the HMW salt mixture is currently incorporated in the diet at the 4 percent level. As was noted in the section on vitamin D, the Ca:P ratio

of the HMW salt mixture is about 4:1. The diet fed in the pilot studies contained about 3 percent of HMW salt mix on a dry basis.

(5) Dietary Fat. As stated in Progress Report 6, the fat content of the diet fed in the long-term experiment was reduced to 11.5-percent from the 16.5-percent level fed to the animals in the pilot studies by reducing the corn oil content and substituting corn starch. Thus, the diet fed in the current experiment for the first 24 weeks contained 5 percent each of corn oil and of cod liver oil. Starting July 14, 1954, corn oil content was increased to 9 percent and cod liver oil decreased to 1 percent on a dry basis. The fat contained in the canned meat accounts for about 1.5 percent of the fat content of the final mix.

The changes were made as a result of a review of the experiences of other investigators and after the reduced reproductive capacity of the animals in the pilot and long-term experiments became evident. Some of the divergent views concerning influence of the kind and amount of fat on reproduction, lactation, and other aspects of rat physiology are presented in connection with possible relevance to the present experiment.

Deuel, et al.[28] reported that diets containing 30-percent levels of six different vegetable fats and butter in mineralized skim milk fed to rats for 12 to 16 weeks were adequate for male and female fertility. Dam and Granados[29] found no impairment of reproductive capacity (lactation was not studied) of rats fed 20% lard with 0.02% vitamin E from the time of weaning. On the other hand, French, et al.[30] found a statistically significant decrement in reproductive performance of rats fed a diet containing 23% corn oil as compared with a commercial ration containing 4.5% fat.

Emerson and Evans[31] in a well-controlled experiment found that resorption gestation occurred in rats fed a diet containing 22% lard and 2% cod liver oil, but not in rats fed these fats at lower levels. Mason and Harris[32] reported that rats on a moderate intake of fat gave a higher fertility response to vitamin E supplementation than rats on a high fat intake. Callison, et al.[16], who fed rats five different diets with fat contents varying from 4 to 17 percent, observed that the higher fat intakes were consistently correlated with the development of excess body fat, bronchiectasis, and skin lesions.

No improvement was noted in reproduction performance of the rats in the present experiment fed a diet containing 11.5 percent fat as compared with pilot-study animals fed fat at a 16.5-percent level. The high incidence of lung lesions in pilot-study animals, of skin lesions in second generation mice on the diet containing 11.5% fat, and the absence of these effects in littermates put on Purina Laboratory chow at maturity are, however, interesting in view of observations of Callison, et al.

Regarding the influence of the kind of fat on reproduction, reports of studies on effects of hydrogenation, rancidity, deficiency of essential fatty acids, and presence of large amounts of highly unsaturated fatty acids have been examined. Of these factors only the last appears to be germane to the present experiment, in view of the high unsaturation of some of the fatty acids in cod liver oil.

Hartwell[33] has presented data which indicates that cod liver oil at a 4-percent level in a synthetic diet increased the growth of rats, but that when this amount was incorporated into the diet of pregnant rats death ensued. Nonpregnant rats fed a diet containing 14% cod liver oil were sterile; uteri, mammary glands, and testes were abnormal. Agduhr[34] found that mature albino rats fed cod liver oil in a dose of 5 ml per kg of body weight by stomach tube for 30 days lost weight and exhibited a decrease in red blood-cell count, a lymphopenia, myocardial lesions, and other pathological changes. Mice, calves, and pigs were adversely affected by lower doses. Dogs and cats that had been administered cod liver oil between the ages of 1 month and 1 year showed no signs of estrus and failed to produce young when paired. The procreative abilities of mice were impaired. Animals on an incomplete diet and those fed cod liver oil over a long period of time suffered greater injury than those fed a complete diet or when cod liver oil was administered over a limited period. Levels of 0.1 ml per kg of body weight were not observed to have any injurious effect. Rather, levels of cod liver oil between 0.1 and 1.0 ml per kg of body weight promoted greater weight gain than the same diet devoid of the oil. By way of comparison, the parent-generation rats in the present experiment were ingesting up to 3 ml of cod liver oil per kg of body weight daily at maturity. Agduhr's findings have been confirmed by several other investigators.

As the importance of vitamin E became recognized, evidence accumulated that at least part of the injury caused by cod liver oil was due to the destruction of vitamin E in the diet, the gastrointestinal tract, or the tissues.[23] But, as noted above, vitamin E levels considerably in excess of those regarded as adequate for the rat failed to prevent poor reproduction and lactation performance, an occurrence also reported by other workers.

Whether cod liver oil exerts deleterious effects as a result of destruction of essential micronutrients or because of a direct toxic action is still a subject for debate. Emmerie, et al.[35] found that a fraction acutely toxic to guinea pigs could be obtained from the nonsaponifiable portion of cod liver oil by chromatographic adsorption techniques. Animals that died from several weeks of daily oral administration of extracts from about 33 gm of cod liver oil did not show symptoms of vitamin E deficiency; mesenteric lymph nodes and connective tissues appeared to be particularly involved. Highly unsaturated alpha glyceryl ethers, including butyl and chimyl alcohols, were present in the most toxic fractions. Toxicity appeared to be correlated with degree of unsaturation, a finding consistent with that of a number of other investigators. Since the amount of cod liver oil represented by the fractions administered by Emmerie, et al.[35] was in excess of 20 times that fed in the earlier stages of the present experiment, and in the absence of conclusive evidence that no changes took place in the course of the isolation of the highly reactive fractions that would influence their toxicity, no conclusions can be drawn regarding the pertinence of their findings to the data presented here.

Of interest in relation to the results given in Tables 48 and 49 which show high early mortality, a high incidence of male sterility, and an early loss of fertility in the long-term rats are results of other rat-feeding studies involving dietary stress presumably resulting from the unsaturated fatty-acid content of lard or cod liver oil or both. Goettsch and Pappenheimer[36] found that 30 percent of the young of females fed large doses of vitamin E at conception died within 10 days of birth with no signs of vitamin E deficiency,an occurrence which parallels closely the results of the present experiment. Gyorgy[37] found male rats to be more susceptible to the injurious effects of the stressful diets than female rats. Even in the presence of normally adequate amounts of vitamin E, such diets resulted in early loss of fertility (Evans and Burr[23]) and is a reversal of the usual occurrence of greater breeding success with second litters than with first (Farmer, et al.[38]). Massive doses of vitamin E administered to rats fed a diet rich in highly unsaturated fats resulted in only partial repair of the injury, according to Himsworth[39] and Gyorgy.[40]

(6) Other Dietary Factors. The dietary regime of the rats in the present experiment does not include any allowance of fresh vegetables or preparations derived from them. Commercial rations which permit good reproduction and lactation contain alfalfa powder or similar constituents. Several investigators have found improved reproductive performance of animals of various species on either natural or synthetic diets or combinations thereof when given supplements of green vegetables or preparations made from them. Farmer, et al.[38] with a diet containing adequate sources of vitamins A and E found green food supplements to improve the reproductive performance of guinea pigs. Other evidence for the presence of unidentified essential nutritional factors in green food is reviewed by Phillips and Constant.[41]

Reports that liver or liver-concentrate supplements may have adverse effects on rats are of interest in view of the fact that the experimental diet contains about 1.5 percent 1:20 liver concentrate (Wilson and Company) on a dry basis. Hubbel and Krehl[42] found that a 3-percent supplement of whole liver substance resulted in a slight decrease in female fertility. Survival and growth of the young were, however, favorably influenced. Bosshardt and Huff[43] found that the stress occasioned by the incorporation of 2% succinylsulfathiazole in a low fat diet fed to mice was enhanced by supplementation with 2% liver concentrate, as evidenced by failure to gain in weight. Liver concentrate fed at a 5-percent level in the absence of the sulfa drug had no adverse influence; a liver concentrate level of 10 percent was stressful. The weight-gain retarding effect of the liver concentrate was attributed by these investigators to a water-insoluble metabolite antagonist which interferes with fatty-acid synthesis.

(7) Irradiation-Induced Changes in Fat. Since the diet fed in the present experiment contained some irradiated fat, it seems appropriate to call attention to a recent publication by Hannan and Shepard[44] in which effects of ionizing radiations on lipids are reviewed. These authors point out that by loss of a hydrogen atom from a reactive methylene group adjacent to a double bond, unsaturated fatty acids are able to form resonance-stabilized free radicals. These react with oxygen to form peroxide radicals, which in turn form hydroperoxides by capturing a hydrogen atom from a second fatty-acid chain. A new free radical is thus formed and a chain reaction ensues. Breakdown of fat continues despite cessation of the irradiation and despite storage at low temperatures, occurrences which may be of significance in the present experiment.

5. CONCLUSIONS

Neither the pilot studies nor the results on the long-term experiment have as yet indicated any acute toxicity to rats fed irradiated diets receiving a dose of up to 45 megarep. In the long-term experiment (4-megarep dose) the animals on the control diet have shown a slightly superior growth rate as compared to the animals on the irradiated diet. In the long-term feeding experiment at the end of 224 days, 31 males had an average weight of 560 gm as compared to 530 gm for the same number of males on the irradiated diet. There was little difference in weight between the females in the two groups, but variation in weights due to pregnancies make these data more difficult to interpret.

Pilot studies using a radiation dose of 20 megarep have indicated an appreciable loss of vitamins when the complete diet was irradiated. Feeding a nonirradiated vitamin supplement was shown to correct the symptoms of apparent vitamin deficiency.

With regard to reproduction and lactation performance, the albino rats on the control diet appear to be slightly superior to the animals on the irradiated diet. Additional data are required to determine whether or not these results are significant.

The second generation of Bagg-strain albino mice on the 4-megarep experimental diet failed to reproduce, whereas the mixed-pigmented strain of mice on the same diet reproduced as did control groups for both strains. The number of animals involved was too small to be of statistical value; however, this pilot study indicates that Bagg-strain albino mice may be more sensitive to the effects of radiation on the experimental diet than are other animals used to date.

The experimental diet may be considered optimal, or nearly so, for growth, as evidenced by a growth rate of as high as 8 gm per day over the first 4 weeks after weaning in the animals on both the control and irradiated diet. The experimental diet is not as satisfactory as a commercial-ration diet with regard to reproduction and lactation. Greater numbers of offspring born per litter and offspring weaned per litter have been observed with this strain of animals fed stock ration.

The experimental diet appears to introduce a certain degree of stress with regard to reproduction and lactation. However, the differences in performance observed between the control animals and the animals on the irradiated diet are comparatively small.

Histopathological examinations of animals used in the pilot study indicated no significant difference between those on the control and irradiated diets. In the long-term studies 2 out of 62 animals on the irradiated diet have developed rapidly growing tumors. Further data are required to indicate whether or not the irradiation of the diet has a significant influence in the development of such tumors.

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B. STUDIES ON THE GAMMA IRRADIATION OF VARIOUS SPECIES OF PLANTS

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1. INTRODUCTION

This study of the effects of gamma irradiation on various species of plants has been supported by Michigan Memorial-Phoenix Project 71. The irradiations were conducted in the radiation cave of the large cobalt-60 gamma source of the Fission Products Laboratory and the evaluation of the results was conducted in the laboratories of the Department of Botany, University of Michigan.

2. RESULTS

A considerable number of species of plants has been irradiated and these experiments have provided some limited information. The death of irradiated plants may be delayed for months. This led to the belief that low dosages were without effect and resulted in the use of excessive dosages all of which have been lethal in their final effect. Further tests are needed to determine what dosages can be withstood by the various plants and plant parts. Unfortunately, dosages which are not finally lethal may be without any morphogenetic effect.

a. Seed Plants

(1) Urginea Maritima. Bulblets were treated with dosages of 50,000 to 500,000 rep. All were killed.

(2) Kalanchoe Tubiflorae. Young plants were treated with dosages of 50,000 to 500,000 rep. In all these the mature leaves were killed. The young leaf primordia persisted for 3 months but finally all growing parts died.

(3) Kalanchoe Tubiflorae with Crown Gall Tumors. Young plants with crown gall tumors on them were irradiated with dosages of 50,000 to 500,000 rep. All the leaves were killed but apparently all those with tumors lived longer than those without tumors. The difference in length of life was small. This trial shows that crown gallabacteria cannot be killed out in leaves by irradiation since the galls are more resistant to radiation than other parts of the leaves.

(4) Zebrina and Pellionia. Cuttings were irradiated with dosages of 500 to 5,000 rep. They were then kept in damp chambers to observe root formation. One lot of pellionia showed a stimulus, but development of all, including controls, was abnormal.

(5) Zamia. Zamia ovules which were unfertilized were irradiated with the hope of inducing parthogenesis. No results of any kind could be observed.

(6) Ginkgo. Pollen was irradiated with dosages from 100 to 80,000 rep. The pollen was then cultured on agar to observe consequences. Normal growth of pollen tubes was observed in all with dosages of 500 rep or less. With dosages above 500 rep pollen tubes grew but the nuclei did not divide. With higher radiation the growth of pollen tubes was reduced. At dosages of 80,000 rep the tubes were very short but were still alive.

b. Seedlings

Seedlings of some 20 species were treated with some result, but the work needs repeating because of irregular behavior in some sets.

c. Ferns

(1) Cyrtomium, Young Sporophytes. In some plants given 1,000 to 8,000 rep juvenile leaves were induced. Dosages of 50,000 to 200,000 rep were lethal. Speed of killing was proportional to dosages but the plants remained alive and practically without any change for 3 to 4 months and then died.

(2) Pteris Longifolia. Young plants were irradiated without noticeable effect. A dose of 800,000 rep caused death in a week or so. Lower radiations showed no effect at all for weeks; then all plants perished except those exposed to dosages below 10,000 rep.

d. Gametophytes of Ferns

Pteris longifolia spores were germinated and grown for a week, then irradiated at dosages from 250 to 128,000 rep.

250 rep - Normal.
500 rep - Normal.
1,000 rep - Normal.
2,000 rep - Normal.
4,000 rep - Some cells had long, threadlike chloroplasts, and in some the plastids were clumped into masses.
8,000 rep - Phizoids were very abundant, and in many the chloroplasts were in radial arrangement.
16,000 rep - Radial arrangement of plastids; some rhizoids were dark and contorted.
32,000 rep - Rhizoids had thickened tips, and some rhizoids had chloroplasts; radial arrangement of plastids was common.
64,000 rep - Smaller than for 32,000 rep and also than for 128,000 rep.
128,000 rep - Giant rhizoids were as wide as spores, fairly short, and some had swollen buds; growth of thalli was retarded; and plastids of many showed radial arrangement.

Algae are very abundant in these cultures and seem to be favored by radiation.

e. Liverworts

Lunularia gemmae were irradiated with dosages from 1,000 to 160,000 rep. All treatments caused some delay in growth.

1,000 rep - Delayed growth, thickening of thalli, and splitting of tips shown.
2,000 rep - The same as for 1,000 rep but effects were increased.
3,000 rep - Growth was more delayed, lobes thicker.
4,000 rep - Lobes were more irregular than for 3,000 rep.
5,000 rep - These were much like 4,000 rep.
10,000 rep - Thallus was much thickened but not much grown. There were 1 to 6 cylindrical upright lobes per thallus. Much splitting of tips occurred and some lobes expanded at top.
20,000 rep - These were the same as for 10,000 rep.
40,000 rep - Very slow growth and spots of dead cells shown, but still were alive and green.
80,000 rep - These were not growing. They were full of spots of dead cells but still green between after 4 weeks.
160,000 rep - All were completely dead within 1 week.

These morphogenetic effects on the liverwort lunularia are the most striking of any that have been secured on any plant. They are being compared with those secured by ultraviolet radiation, by chemical treatment, and by subjection to various physical factors.

C. EFFECTS OF GAMMA RADIATION ON ELASTOMERS

Personnel:

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1. INTRODUCTION

A program of fundamental research on rubber and elastomers is being conducted at the University of Michigan with support from Phoenix Project 43. Emphasis in the work is directed toward gaining a better understanding of the molecular, structural, and physical properties of elastomeric materials. Funds for fellowships and equipment were provided by a gift from the Goodyear Tire and Rubber Company to the Michigan Memorial-Phoenix Project.*

2. REVIEW

The effects on plastics and elastomers of large doses of radiation from radioactive sources have been the subject of a number of published investigations. Davidson and Geib[1] reported results of the exposure of elastomers, mostly uncured, to pile radiations. Lawton, Bueche, and Balwit[2] employed an artificially produced beam of 800-kv electrons. They listed two classes of polymers, those which were cross-linked by the radiation and those which were degraded.

The most thorough study of the effects for any one polymer is presented by the investigations of Charlesby[3,4,5] on polyethylene. This work

*The authors wish to express their thanks to the Goodyear Tire and Rubber Company, H. J. Osterhof for permission to publish this work, and to G. K. Higgins who prepared the mechanical goods compounds.

also gives an excellent appraisal of the general nature of the reactions and changes which radiation may be expected to induce in hydrocarbon polymers.

Ryan[6] has published the results of exposure of a variety of elastomers to intense gamma radiation. Wall and Magat[7] and Berstein, Farmer, Rothschild, and Spalding[8] studied the effects of radiation on polymerization reactions with results which are illuminating in a general way in regard to the reactions of radiation on polymers themselves.

There is still a great deal of background information required on radiation-induced changes in elastomers to understand these changes scientifically and to use the elastomers most advantageously. The present work is a survey of the effects of a large dose of gamma radiation on a variety of elastomers and compounds. It adds to work already published by the greater variety of elastomers and elastomer compounds studied by the more exact specification of the type of radiation and the temperature during the exposure, and by the exclusion of possible effects due to the presence of oxygen.

3. EXPERIMENTAL PROCEDURE

The elastomer of plastic compounds were mounted in a position near the rods of the high-level, cobalt-60 source in the radiation cave of the Fission Products Laboratory where the intensity was about 100,000 rep per hour. The dose used in this work was 10^8 rep, representing a totalized exposure to the source of 500 to 1000 hours.

The presence of ozone in the cave was noticeable so that the samples were sealed in a nitrogen atmosphere to eliminate the possibility of ozone attack. In the first run the samples were sealed under nitrogen in a package of polyethylene film. In the second experiment duplicate samples as well as additional elastomers were sealed in a dessicator which had been flushed out with nitrogen. Although the polyethylene sheets employed in the first experiment deteriorated, the results for the similar samples in the two experiments were essentially identical; therefore, it was concluded that oxygen effects had been eliminated in both experiments. Hence the results given for many of the compounds represent the average of measurements of the properties of the samples from the two experiments.

The vulcanized elastomers were prepared as test sheets nominally 0.080 inch in thickness. The uncured elastomers were pressed out into sheets about 0.1-inch gage at 275°F, placed between heavy aluminum foil, and clamped flat at room temperature for 24 hours. They were exposed in the aluminum foil.

Stress-strain tests were made with an Instron tester; the control test pieces were measured at the same time as the exposed test pieces in order to eliminate normal aging effects. For the vulcanized samples small dumbbells were used; the neck was 1/10 inch wide and about 3/4 inch long. Crosshead speed was 10 inches a minute and chart speed was 20 inches a minute. The raw polymers were tested in the form of strips 1 inch wide. All the elastomers used were commercial materials. The temperature during irradiation was in range from 60 to 70°F. The testing temperature was 77°F.

4. DISCUSSION OF RESULTS

Results from the stress-strain records are given in Table 56. It must be realized that the effects of the radiation are complex and such comparisons may be affected by the dose, the dose rate, and the type of radiation. Charlesby[3] however has pointed out that qualitatively the effects from various types of high-energy radiation should be very similar. This similarity is due to the fact that only a very small proportion of the molecules is affected directly by the primary radiation. The molecules are activated principally with relatively small energies available from the degradation of the incident high-energy radiation by successive interactions with the electrons and nuclei.[9]

Thus, the classification of elastomers, those cross-linked by irradiation and those broken down, arrived at by experiments using 800-kv electrons, in general is also evident in this work with a gamma-ray source.[2] Ryan[6] on the other hand used gamma rays from the Hanford "cooling off" pit and found stiffening only. The probable reason for this difference in the results lies in the size of the radiation dose. The gamma flux used by Ryan was 10^{11} photons/cm^2 sec for a period of 6 months, about 10 times the dose used in this work.

Charlesby[3] recognized three stages in the effect of the dose on polyethylene. In the first stage, there was slight cross-linking with little change in properties; in the second stage, the cross-linking was extensive but the properties were still similar to ordinary polyethylene at room temperature; in the third stage, the polyethylene became glasslike, discolored, and brittle.

It is quite possible, as is indicated by a comparison of the results, that some of the elastomers such as Chemigum SL and Thiokol should actually show some degradation or softening before the final stage of embrittlement sets in. Both the butyl gum stock and raw butyl rubber were reduced to a sticky "goo" by the exposure. This effect is a characteristic of free radical agents on butyl rubber, as compared to their cross-linking action with most elastomers. Thus the experiments with butyl rubber are a striking confirmation that the effects of radiation on elastomers are brought about by free radical mechanisms.

TABLE 56

EFFECT OF 100-MEGAREP DOSES OF GAMMA RADIATION ON STRESS-STRAIN PROPERTIES OF ELASTOMERS

Compound	Material	Cure Min/°F	100% Modulus, psi Original	Exposed	% Change	Tensile Strength, psi Original	Exposed	% Change	Ultimate Elongation, % Original	Exposed	% Change
Mech Gds	Natural rubber	30/275	107	197	+84	3400	2600	-24	715	645	-10
Mech Gds	Natural rubber	30/275	387	652	+68	4000	2835	-29	562	367	-35
F	Natural rubber	20/290	372	748	+101	2800	2040	-27	403	210	-47
H	Natural rubber	10/311	67	135	+101	1750	2080	+19	660	590	-11
H	Natural rubber	20/311	67	130	+94	1650	1820	+10	700	570	-19
H	Natural rubber	30/311	95	131	+38	1700	1500	-12	740	540	-27
	Milled Smoked Sheet	—	24	98	+308	25	225	+800	157	303	+93
Mech Gds	Cold rubber	30/275	297	595	+100	3035	2525	-17	520	310	-40
G	Cold rubber	40/290	321	982	+206	3365	3300	-2	505	230	-54
D	Cold rubber	30/290	116	194	+67	264	265	0	313	160	-49
D	Cold rubber	60/290	113	190	+68	226	258	+14	245	168	-31
E	Cold rubber	30/290	48	150	+212	361	238	-34	1000	212	-79
E	Cold rubber	60/290	77	192	+151	225	338	+50	499	230	-54
	Cold rubber	—	22			16	80	+400	918	52	-94
A	Chemigum N	45/290	390	2790	+615	2820	2790	-2	445	100	-78
A	Chemigum N	90/290	398			2820	2880	+2	440	80	-82
Mech Gds	Chemigum N	30/290	256	1170	+356	1670	1410	-16	562	122	-78
Mech Gds	Chemigum N	35/295	580			1980	1750	-12	300	85	-72
Mech Gds	{Chemigum N / GR-S}	30/295	370	2160	+480	2260	2240	-1	425	105	-75
Mech Gds	{Chemigum N / Vinylite*}	40/305	500	22		2240	2320	+36	465	60	-87
Mech Gds	Vinyl chloride copolymer	5/295	1000	910	-9	1870	2460	+32	215	272	+27
C	Neoprene	30/290	140			4115	400	-90	983	67	-93
C	Neoprene	50/290	153			4695	372	-92	980	68	-93
Mech Gds	Neoprene	30/295	124			2285	1358	-41	885	88	-90
Mech Gds	Neoprene-GR-S	30/295	540			2410	1600	-34	325	55	-83
Mech Gds	Thiokol**	30/295	540	435	-20	1080	610	-44	293	177	-40
Mech Gds	Silastic***	10/260	71			1300	450	-65	725	55	-92
	Chemigum SL	15/280	298	150	-50	6350	2260	-65	880	590	-33
I	Hypalon	25/290	455			1655	1430	-14	164	19	-88
	Polyethylene		2200			2300	1080	-53	115	6	-95

*Bakelite Corporation, New York
**Thiokol Corporation, Trenton, New Jersey
***Dow-Corning Corporation, Midland, Michigan

Charlesby[3] recognized the occurrence of surface oxidation of polyethylene during exposure to radiation. No evidence of oxidation could be seen in the infrared absorption spectrum of the irradiated butyl rubber.

The sample of polyethylene listed in Table 56 was not sealed in nitrogen, as were the other samples, but was taken from the container bag used in the first run; it was commercial film, 0.0015-inch gage. After the exposure, a strong band occurred in the infrared spectrum at about 5.8μ, showing that oxidation did contribute to the change in properties which occurred. The 5.8μ band is characteristic for the carbonyl group in organic compounds.

A predominately cross-linking character for the radiation-induced reactions is indicated in most of these tests by an increase in modulus and a decrease in ultimate elongation. The tensile strength usually dropped. The most striking exception occurred in the case of raw natural rubber for which beneficial cross-linking reactions actually brought about the rudimentary cure. The effects were similar with raw GR-S but were not so pronounced.

All things considered, natural rubber appeared to withstand this radiation dose better than any other of the vulcanized elastomers. Since the free radical character of the changes induced by radiation is well recognized, it is reasonable to expect that the type of compounding can affect the results.

5. SUMMARY AND CONCLUSIONS

A survey has been made of the effects of intense gamma radiation on the physical properties of elastomers and plastics. Such information is required to determine the types of elastomers which are most resistant to radiation, to estimate the service life of elastomers exposed to radiation, and to show favorable trends in compounding for radiation resistance.

Intense radiation may affect the physical properties of elastomers beneficially or adversely, depending upon the type of elastomer and the exposure dose, although hardening and embrittlement are most commonly observed. The possibility is apparent of developing elastomer compounds with improved radiation resistance, especially by the use of chemical agents which affect free radical reactions. The nature of the elastomer however will probably severely limit what can be accomplished by compounding.

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PART V. COOPERATIVE STUDIES PERFORMED WITH PERSONNEL OF THE UNIVERSITY OF MICHIGAN HOSPITAL

A. THE USE OF COBALT-60 AS A STERILIZING AGENT FOR AORTIC HOMOGRAFTS

This is a preprint of a paper presented November 16, 1954, at the Annual Meeting of the American College of Surgeons. No part of this paper is to be reproduced without permission of the authors. The paper will be published in the medical literature at a later date.

Personnel:

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1. The Effect of Gamma-Ray Irradiation on the Structural Integrity of the Graft

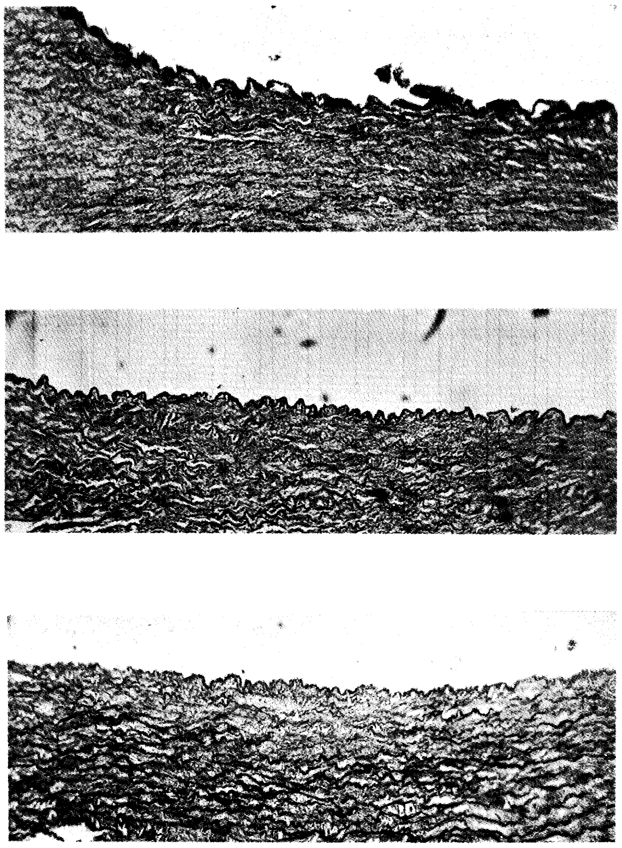
The increasing demand for vessel grafts has made it difficult for most hospitals to maintain an adequate supply. Many otherwise suitable grafts have had to be discarded because they were contaminated either by infection existing in the donor or by improper handling during removal and preparation of the grafts for storage. A simple method of sterilizing contaminated grafts would greatly increase the number of usable grafts. However, any technique of sterilization must preserve the structural integrity of the graft as well as destroy the contaminants. Ethylene oxide[1] and beta-propriolactone[2,3] have been employed successfully to sterilize vessel grafts, as has irradiation from the Van de Graaff[4] and Capacitron machines.[5]

Extensive bacteriologic studies carried out in the Fission Products Laboratory at the University of Michigan with a 10,000-curie source of gamma radiation from cobalt-60 have demonstrated the bactericidal effect of gamma radiation.[6,7] It is effective against organisms ranging in size from viruses to yeasts. The bactericidal dose for each organism depends on individual susceptibility, size, morphology, and physical state. A review of these studies suggested that a dose of 2,000,000 rep would destroy organisms normally encountered as contaminants of blood-vessel homografts. Some of the organisms frequently encountered as contaminants were Staphylococcus albus and aureus, Bacillus subtilis, Streptoccus fetalis, and organisms of the Actinomyces groups. However, since the same deleterious changes produced within the bacterial cells by gamma radiation are also produced within tissue cells, it was necessary to determine whether or not the changes produced by gamma radiation would alter the structural integrity of arteries to a degree that would preclude their use as grafts. In an attempt to answer this question, segments of dog aorta which had been exposed to gamma radiation were examined for any microscopic changes and were also given a clinical trial by being inserted as homografts in the thoracic aorta of dogs.*

2. Methods

Graded doses of gamma radiation ranging from 15,000 to 4,000,000 rep were administered to fresh segments of a dog aorta which were (1) placed in dry, sterile test tubes, (2) placed in sterile 0.9% sodium chloride solution, and (3) lyophilized. Since gamma radiation produces ionization, certain rather strong oxidative reactions occur in the presence of water. Because of this it was felt that the presence of water at the time of irradiation might result in greater damage to the grafts. Therefore, the effects of gamma radiation on the segments of aorta were compared when administered (1) in the presence of water of the tissue itself, (2) in the presence of a water and an electrolyte medium, and (3) in the absence of water (lyophilization). Four groups of controls were used: (1) segments fixed in 10% formalin immediately after removal, (2) segments refrigerated at 1°C for a time equal to that required for irradiation, (3) segments stored at room temperature for a similar length of time, and (4) lyophilized segments reconstituted in 0.9% sodium chloride solution. All segments were fixed in 10% formalin and microscopic sections prepared and stained Because the media with its elastic fibers is the structurally important layer of the vessel graft, Verhoeff elastic stain was used in addition to hematoxylin and eosin stains. Consistent microscopic changes attributable to gamma radiation could not be detected in any of the irradiated segments of aorta (Fig. 59). There was some patchy fragmentation of the elastic tissue fibers but this could not be correlated with the amount of irradiation given. Since microscopic examination did not reveal any differences between the irradiated and control segments, it alone was not considered an adequate method for evaluating the effects of gamma radiation.

*The authors wish to express their appreciation to Professor Ruth C. Wanstrom of the Department of Pathology, University of Michigan Medical School, for her cooperation in the microscopic examinations.



(1)

(2)

(3)

Fig. 59. Photomicrograph of Three Segments of Dog Aorta: (1) Control Segment, (2) Segment Irradiated with 15,000 rep, (3) Segment Irradiated with 4,000,000 rep.

Irradiated segments of dog aorta were placed in the thoracic aortas of 16 dogs. The grafts had been obtained without aseptic precautions within 1 hour after death of the donor dog. Cultures from the segments of aorta showed Streptococcus viridana, Staphylococcus aureus, paracolon bacilli of the aerogenes type, and numerous gram-negative bacilli. The segments of aorta were preserved in Tyrode solution or were lyophilized and then exposed to 2,000,000 rep. The irradiated segments were stored in Tyrode solution for periods ranging from 7 to 19 days. The lyophilized segments were stored from 13 days to 7 months. Culture specimens were taken from the storage solution or reconstituting solution at the time of insertion of the aortic grafts. Blood flow to the distal aorta was maintained with a polyethylene shunt while the grafts were sutured in place with continuous everting mattress sutures of No. 00000 silk. No anticoagulants were employed at any time. Each day for 5 days postoperatively, 400,000 units of penicillin (S-R) were administered intramuscularly.

3. Results

Cultures were made following irradiation of 7 contaminated grafts stored in Tyrode solution and no organisms were recovered. Thirty-one cultures were made of the reconstituting solution (0.9% sodium chloride) of lyophilized, contaminated grafts which had been irradiated. One culture contained a few gram-negative anaerobic bacilli. Because of the very small number of organisms found and because the reconstituting solution was exposed to the air for a period of at least 1 hour, this single positive culture may well have been due to contamination at the time of reconstitution rather than to survival of organisms following irradiation.

Four of the 16 dogs operated died within 3 days from complications which included renal and spinal cord damage, operative hemorrhage, and atelectasis. Gross examination of the grafts in these animals revealed no changes except for the formation of a wedge-shaped thrombus at the suture lines after the second day. Microscopically, there was loss of the intima of the graft on the first day and also patchy loss of staining reaction of the nuclei of the lyophilized grafts. It is interesting to note that this patchy loss of nuclear staining, which was interpreted as evidence of cellular necrosis, was not evident at the time of reconstitution of the lyophilized grafts but occurred only after implantation of the grafts. This probably indicates that even though the cells do not survive the process of lyophilization, they are so perfectly preserved morphologically as to undergo the same staining reactions as do living cells.

Seven of the surviving 12 dogs are alive and well, after periods ranging from 1 to 4 months since their operation. All of these dogs have excellent pulses in the hind legs. Five dogs were sacrificed at intervals ranging from 3 weeks to 45 weeks following insertion of the graft. At 3 weeks the graft on the first dog showed a red, finely granular, intimal surface; about 1 cm of the length of the graft had been covered with endothelium. There were no thrombi and the graft was patent. The second dog was sacrificed at the end of 8-1/2 weeks. Externally there was no disparity in size between the host aorta and the graft and the adventitial surface was smooth and continuous with the host vessel. The intimal surface was gray and glistening and in some areas was finely wrinkled. Microscopically, the graft was completely covered by intima. In the third dog, sacrificed at the end of 9 weeks, the graft was patent but the intimal surface displayed two red, shaggy areas of erosion measuring 4 and 6 mm in diameter. There was no thrombosis or calcification at these sites and the remainder of the intimal surface was smooth and glistening. The fourth dog was sacrificed at the end of 20-1/2 weeks (Fig. 60). There was a slight narrowing at the suture lines which was attributed to the small size of the graft originally. The intimal surface was smooth, glistening, and in no way different from the host in gross appearance. The fifth dog was sacrificed at

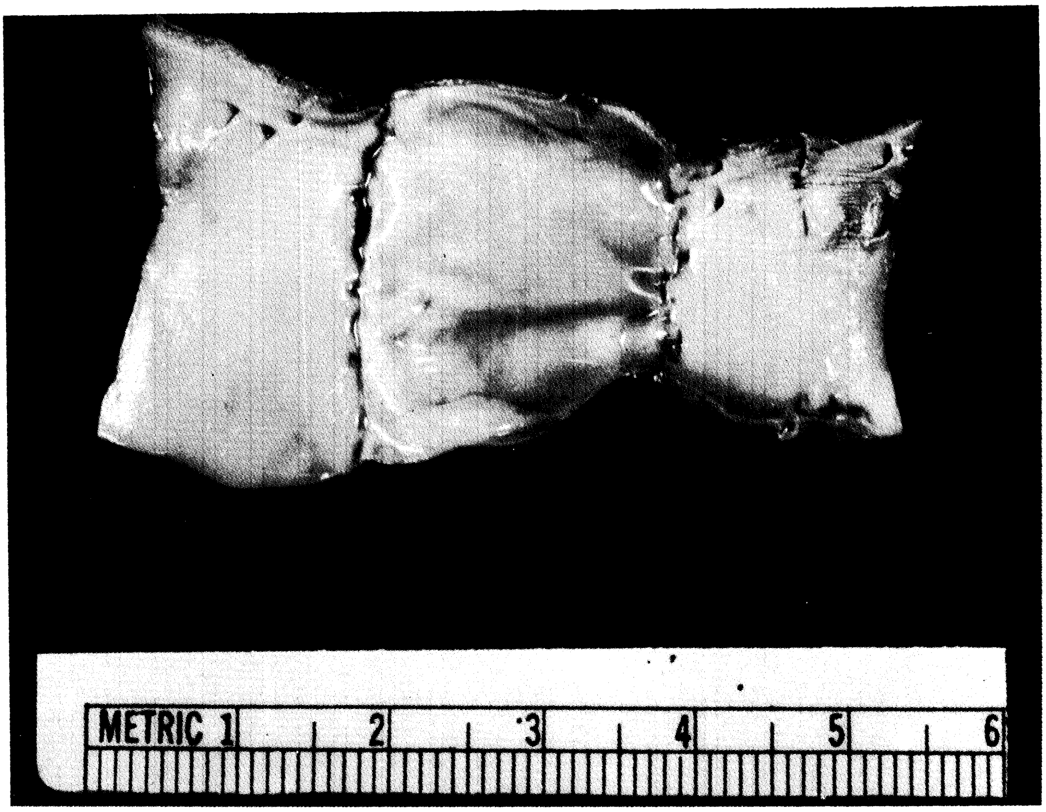


Fig. 60. An Irradiated, Lyophilized Aortic Homograft 145 Days after Insertion.

the end of 45 weeks. Externally, the contour of the graft matched that of the host vessel perfectly. The intimal surface was smooth and glistening. In one portion of the graft there were two pale yellow areas measuring 3 mm in diameter which could not be identified microscopically. Grossly, there was no evidence of sclerosis or degenerative change in any of the grafts. Microscopically, the changes observed were loss of intima on the first day of implantation, loss of nuclear stain, or cellular necrosis, and thinning of the graft wall to about 60 percent its original size. In none of the vessels was there any evidence of foreign body reaction to the graft. Neither was there evidence of any delay due to irradiation in intimal proliferation along the inner surface of the graft or in fibroplastic proliferation around the adventitial surface of the graft. The elastic fibers in the grafts remained intact and appeared unchanged. Mucoid degeneration in isolated areas was noted in several of the grafts beginning as early as 2 months after insertion. The 10-month-old graft showed one small area of calcification near the distal anastomosis.

From these results, it was felt that no adverse changes in the aortic segments could be attributed to gamma radiation and that the irradiated aortic segments functioned well as homografts when inserted into the thoracic aorta of dogs. Further evaluation of the method is of course necessary and should include a longer period of observation of the implanted grafts. Tensile strength studies of irradiated grafts are now in progress. Also, more extensive bacteriological studies with regard to viruses are under way and may indicate the necessity for a higher dose or irradiation.

4. Conclusion

Preliminary studies suggest that gamma radiation from a cobalt-60 source does not destroy the structural integrity of aortic homografts in dogs, and that such radiation provides a simple and effective method of sterilization.

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B. RADIATION STERILIZATION OF BONE-BANK BONE

This material will be reported in the medical literature at a later date; therefore, no part of this section of the report is to be reproduced without the permission of the authors.

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1. Introduction

Two major problems confront the surgeon who makes use of bone-bank bone. One is the difficulty encountered in obtaining the bone and the other is controlling its sterility.

A simple method of obtaining and sterilizing bone-bank bone without greatly altering its ability to stimulate new bone formation would certainly improve the usefulness of bone-bank bone and increase its popularity.

Bone-bank bone, subjected to storage in antiseptics and antibiotics, or to boiling, freezing, or freeze-drying, may still harbor living bacteria and viruses or be so altered in its chemical and physical properties that its usefulness as a transplant is impaired. Kreuz[1] has reported that 16.3 percent of their bone-bank bone, obtained from fresh cadavers, was rejected because of bacterial contamination, and Shutkin[2] reported a case of homologous serum hepatitis following the use of refrigerated bone-bank bone.

If the surgeon were to assume that all bone obtained for the bone bank were contaminated, he would be forced to sterilize it by one means or another prior to transplantation. The purpose of this section of the progress report is to describe the experimental sterilization of bone-bank bone by subjecting it to gamma radiation from cobalt-60.

2. Procedure

The following experiments were conducted to determine how much radiation is necessary to sterilize bone and to test the effectiveness of irradiated bone in stimulating new bone formation.

Segments of adult dog humerus, obtained without using sterile precautions, were intentionally contaminated with a 4-day-old culture of B. tetanus and sealed in screw-top glass jars.

Segments of adult dog rib were also contaminated with a mixed culture of B. tetanus, Clostridium botulinum 62A, Clostridium 213B, and putrefactive anaerobe NCA3679. The contaminated bone was exposed to varying levels of high-intensity gamma radiation and subcultured at periodic intervals in Brewer's thioglycolate media.

Following radiation the bone was examined grossly and microscopically for structural changes and transplanted into dogs to test its effectiveness as a homogeneous bone transplant.

Segments of ribs were used for transplants and were obtained from adult mongrel dogs without using sterile precautions. Test ribs were sealed in plastic or glass tubes, subjected to a sterilizing dose of gamma radiation, and stored at room temperature pending transplantation. Control ribs were obtained and handled in the same manner but received no irradiation. Approximately 1/2 to 1 inch of the midulnar shaft was excised subperiosteally and split pieces of rib were used to fill the gap. No attempt was made to reapproximate the periosteum. Aseptic operative techniques were used in all test and control procedures. None of the animals received antibiotics pre- or post-operatively and no external fixative devices were used to support the extremity.

3. Results

It was found that 2,000,000 rep and over were required to consistently obtain sterile cultures. Most of the transplanted bone received approximately 3,000,000 rep.

No changes grossly or microscopically could be detected in the bone immediately following irradiation, but irradiated bone stored at room temperature for a period of 6 months demonstrated complete necrosis of the marrow and focal areas of calcium crystals in the marrow spaces. Deep-freezing irradiated bone served only to retard and not prevent this gradual autolysis of the marrow. The same process of gradual deterioration was observed in nonirradiated, frozen bone. (No change was detected in the trabeculae other than gradual disappearance of cells from their lacunae.)

Twenty ulnae in 18 dogs received irradiated rib. All wounds healed primarily with no evidence of wound infection. Periodic x-ray examinations revealed gradual absorption of the transplant and new bone formations within the gap. No unusual systemic reactions were noted post-operatively and the first few dogs operated on in May, 1954, had microscopic and radiographic evidence of bony union between the transplant and one or the other ulnar fragments at approximately 3 months. Microscopic examination of the transplant sites consistently demonstrated absorption of the transplant and new bone formation, with no evidence of infection or foreign body reaction.

Five ulnae in 5 dogs received control rib. Two of the control animals developed a severe wound infection and a generalized toxic reaction 10 days post-operatively which necessitated their sacrifice. Two developed a mild wound infection with abscess formation but recovered following an incision and drainage of the abscess. One healed primarily with no evidence of wound infection and progressed to early union.

4. Conclusions

Preliminary experimental work indicates that dog bone can be sterilized by gamma radiation and transplanted without immediate, harmful effects. Bone so sterilized retains its ability to stimulate new bone formation and produces no foreign body reaction in the host.

Preliminary work on the radiation sterilization of lyophilized human bone is now under way. If successful, procurement of sterile bone-bank bone will be made less difficult, and the surgeon will be assured of a safe and effective homogeneous transplant.

5. References

1. Kreuz, F. P., Hyatt, P. W., Turner, T. P., and Bassett, C. A. L., "Use of Preserved Tissue in Orthopedic Surgery", AMA Arch Surg, 64, 148-153 (February, 1952).

2. Shutkin, Ned M., "Homologous Serum Hepatitis Following the Use of Refrigerated Bone-Bank Bone", J Bone and Joint Surg, 36-A, No. 1, 160 (1954).

PART VI. COOPERATIVE RESEARCH WITH INDUSTRIAL LABORATORIES

A. IRRADIATION OF GREEN COFFEE BEANS AND ROASTED GROUND COFFEE

1. Introduction

In order to determine the effect of gamma radiation on the extraction of coffee, green coffee beans and roasted ground coffee were irradiated by the Fission Products Laboratory. The coffee was supplied and the results analyzed by J. A. Folger and Company.

2. Procedure

Coffee, 20 lb of roasted, ground coffee and 25 lb of green coffee beans, was irradiated. Each lot was divided into five equal parts and vacuum-packed in 4-lb coffee cans. Both the green coffee beans and the roasted, ground coffee were irradiated in the cans at the following levels: 10,000; 50,000; 100,000; 500,000; and 1,000,000 rep.

Each lot was roasted and ground before testing for extraction and then given the following tests: (1) extraction yield of water solubles (the method used here is the same as Official Method A 4-38 of the American Oil Chemists Society, except that water was used instead of ether) and (2) comparative taste testing.

3. Results

A summary of the results of the examination made in the J. A. Folger and Company laboratories is given in Table 57.

TABLE 57

RESULTS OF TESTS ON COFFEE EXPOSED TO GAMMA RADIATION

Irradiation Level Roentgen Units	Extraction Yield, %	Taste Differences from Control
	Green Coffee	
10,000	26.2	None
50,000	26.0	None
100,000	25.5	None
500,000	25.8	None
1,000,000	25.5	None
Control	25.2	—
	Cut, Ground Coffee	
10,000	25.0	None
50,000	24.8	None
100,000	25.2	None
500,000	24.9	None
1,000,000	25.4	Slightly undesirable flavor
Control	24.9	—

4. Conclusions

Irradiation of green coffee beans has no noticeable effect on the extraction or flavor of the finished product after roasting and grinding. Irradiation of roasted, ground coffee has no effect on the extraction and only a very slight effect on the taste of the coffee.

B. IRRADIATION OF SEED WALNUTS

1. Introduction

In order to explore the use of gamma radiation to produce new mutant strains, irradiation studies were made with Thomas-variety black walnuts. The nuts were supplied by the Northern Nut Growers Association through A. L. Barlow, a member of this organization. The nuts were irradiated in the Fission Products Laboratory and the results are being evaluated by the Northern Nut Growers Association.

2. Procedure

Since data on the nonlethal limit for gamma irradiation of walnut seed were not available, a preliminary study was conducted with several lots of 50 nuts each using the following amounts of radiation: 1,000; 5,000; 10,000; 20,000; 30,000; 40,000; and 50,000 rep.

After irradiation the walnuts were labeled and stratified by placing on them a mat of sawdust and covering them with several layers of damp burlap to hasten germination. A biweekly inspection was made to check germination progress and to dampen the walnuts and burlap.

Table 58 indicates germination and growing results as of August 22, 1954.

TABLE 58

OBSERVATIONS ON THE GERMINATION OF IRRADIATED WALNUTS

Number Stratified 3-27-54	Treatment	Germinations 5-24-54	Germinations Total 7-3-54	Number Growing 8-8-54	Number Growing 8-22-54
50	None	3	7	5	5
50	1,000	2	4	3	4
50	5,000	3	6	6	6
50	10,000	2	8	7	7
50	20,000	2	10	9	9
50	30,000	2	16	10	12
50	40,000	3	16	1	3
50	50,000	3	18	0	1

During the biweekly inspections when the walnuts were noted to be germinating they were planted in the growing plots (Fig. 62). When growth was first observed above the ground many of the untreated seedlings were of greenish hues. Several of the second and third leaves were considerably distorted from the normal shape; however, subsequent leaves were the customary shape (Fig. 61).

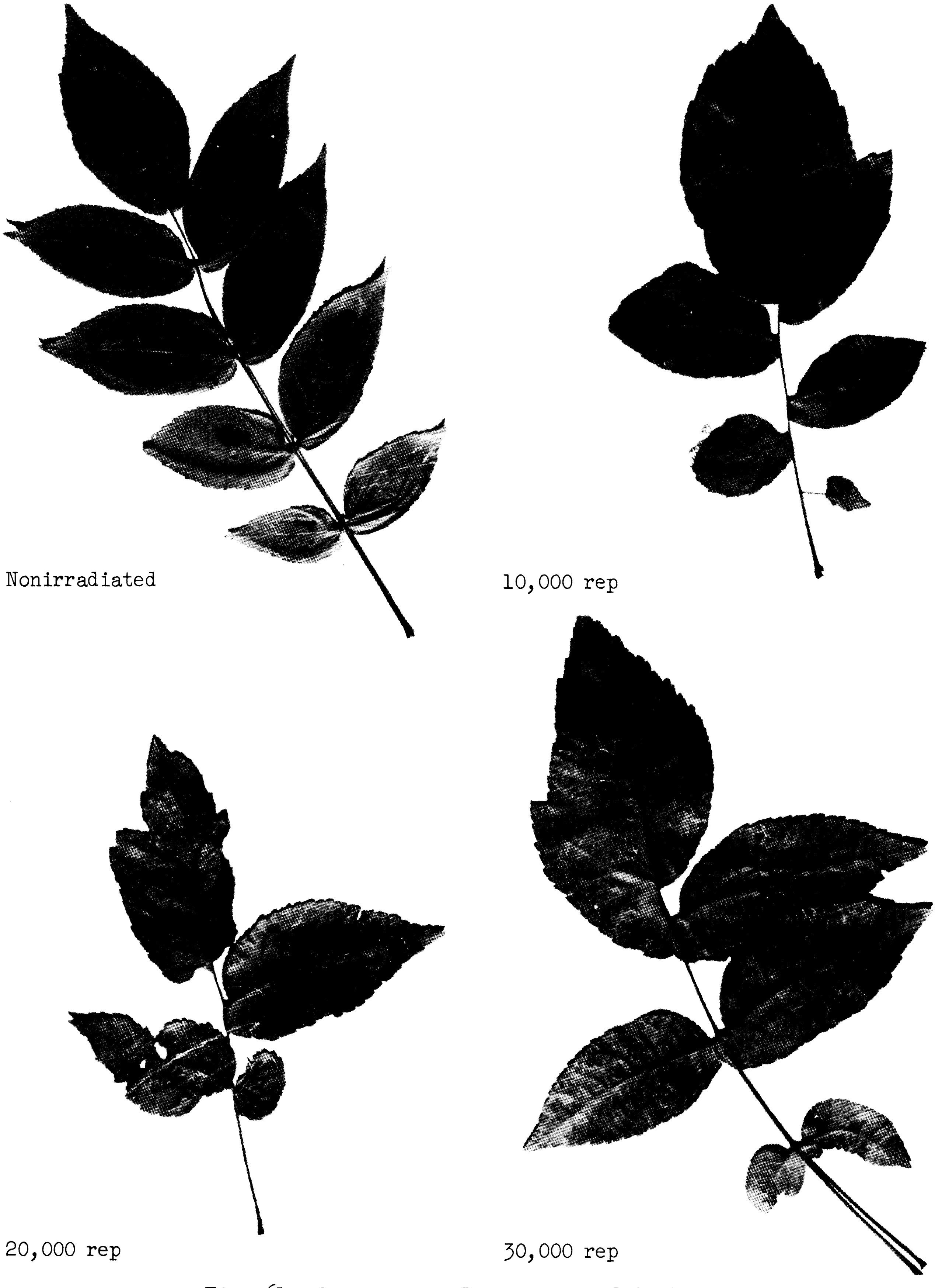

Fig. 61. Leaves from Irradiated and Control Thomas-Variety Black Walnut Seedlings.

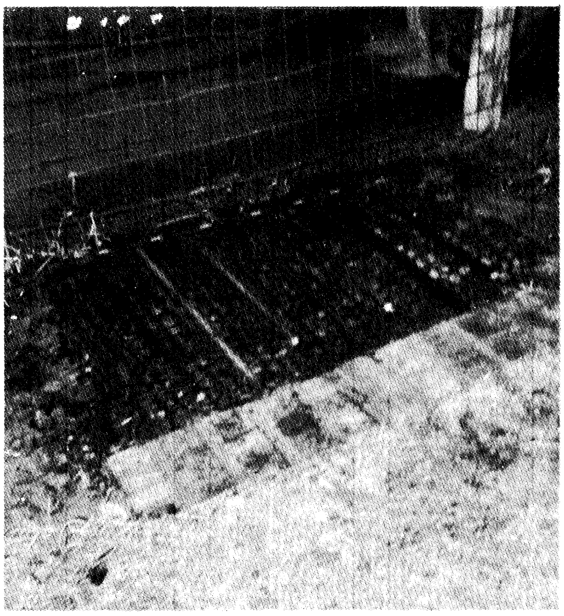

Fig. 62. Growing Plot for Germinated Walnuts.

Sections from the irradiated seedlings will be grafted to hasten bearing in order to check any mutations which might occur, and a report on this phase of the experiment will be submitted in the future.

C. IRRADIATION OF RAW FRESH CRABMEAT

1. Introduction

There are three distribution problems in the crabmeat industry: (1) because of its extremely perishable nature, the radius of the crabmeat market is limited by length of time in transit and icing facilities; (2) for the same reason, both retail and wholesale shelf-life is very short; and (3) since the heaviest production of crabmeat occurs during the late summer months when prices are lowest with the highest prices being reached in late winter or early spring when production is limited, there is the problem of trying to even out the market by satisfactorily holding the low-cost meat of late summer for the higher winter price.

Quick freezing as now practiced seems to impair both texture and taste. The Fish and Wildlife Service has developed a not too satisfactory method of pasteurization by sealing meat in No. 2 enameled cans or 1/2 lb flats, using hot water 160°F for 10 minutes to 170°F for 1 minute. By this method, which is used to a certain extent in this area, there is a slight discoloration of the processed product, a slight change in the delicate flavor, and a rather high percentage of spoilage (about 10 percent). However, some of the meat sealed in these cans has kept well for as long as 6 months when stored at 35 to 43°F. Some packers repack this pasteurized product into conventional crabmeat cans with perforations in the bottom and sell it as fresh crabmeat on the high winter market.

2. Procedure

The Crisfield Company supplied by air express 5 lb of iced, raw, fresh backfin or "hump" crabmeat of the Atlantic Blue crab in perforated crabmeat cans. This crabmeat was transferred to ten No. 2 cans and two cans each were given the following dosages at room temperature: 0; 40,000; 80,000; 120,000; and 1,000,000 rep. After irradiation the iced samples were returned air express.

3. Results

One week after irradiation one set of the cans was opened at the Crisfield Company. The results are shown in Table 59. In view of these preliminary tests it appears that the most promising radiation dosage is in the range of 120,000 rep.

TABLE 59

RESULTS OF TESTS ON IRRADIATED FRESH CRABMEAT

Sample Number	Dose	Condition	Odor	Flavor	Bacteria Count*
1	Control	Spoiled	Off	No taste test	$3x10^6$
2	40,000	Slightly spoiled	Slightly off	No taste test	$3x10^6$
3	80,000	Very slightly spoiled	Slightly off	Slightly off	$30x10^6$
4	120,000	Excellent	Excellent	Excellent	$.78x10^6$
5	1,000,000	Dry, cooked appearance	Musty	Musty	$3x10^3$

*None of the samples, including the control, showed any evidence of E. coli, the most troublesome organism commonly found in fresh crabmeat.

PART VII. SUBPROJECT M943-7, OPERATION OF THE FISSION PRODUCTS LABORATORY

Personnel:

L. E. Brownell, Supervisor and Professor of Chemical and Metallurgical Engineering; J. V. Nehemias, Assistant Supervisor and Health Physicist; D. E. Harmer, Chemist; and E. Ambo, Laboratory Assistant

Advisors:

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A. ROUTINE USE OF THE HIGH-LEVEL GAMMA RADIATION SOURCES

The high-level gamma source has been in routine operation throughout the period of this report. Unnecessary down-time has been minimized and near-capacity operation maintained.

During August, 1954, the wire mesh cap for the source was found to interfere with normal raising and lowering operations. The source was then removed from its platform and placed at the bottom of the well. With the duplicate (dummy) source in place, detailed observations were made of the cap misalignments responsible for the interference. The necessary changes were made in the wire mesh cap and the source put back in use without incident.

The clarity and pH of the well water have been maintained at satisfactory levels. Periodic radiological checks of the filters indicate no radiocobalt in the well water. A comprehensive manual of operations has been prepared describing in detail the operation of the ion-exchange system.

B. OPERATIONAL TECHNIQUES

1. Canned Raw Frozen Foods

An annular insulated container was designed and constructed by the personnel of the American Can Company for the purpose of irradiating canned, uncooked food in the frozen state. Figure 63 illustrates this "irradiation ice box" in use. In an irradiation experiment involving a large number of containers such as those shown, 26 commercial No. 2 cans (3-3/8 inches in diameter and 4-1/2 inches long) have been simultaneously treated at a dosage rate of 100,000 rep per hour. The cans were periodically rotated during the irradiation and the unit was kept closed to maintain thermal insulation. Experiments have been performed at ice temperature and at dry ice temperature. The temperature was controlled by placing containers of ice or dry ice in the box around the outside of the samples.

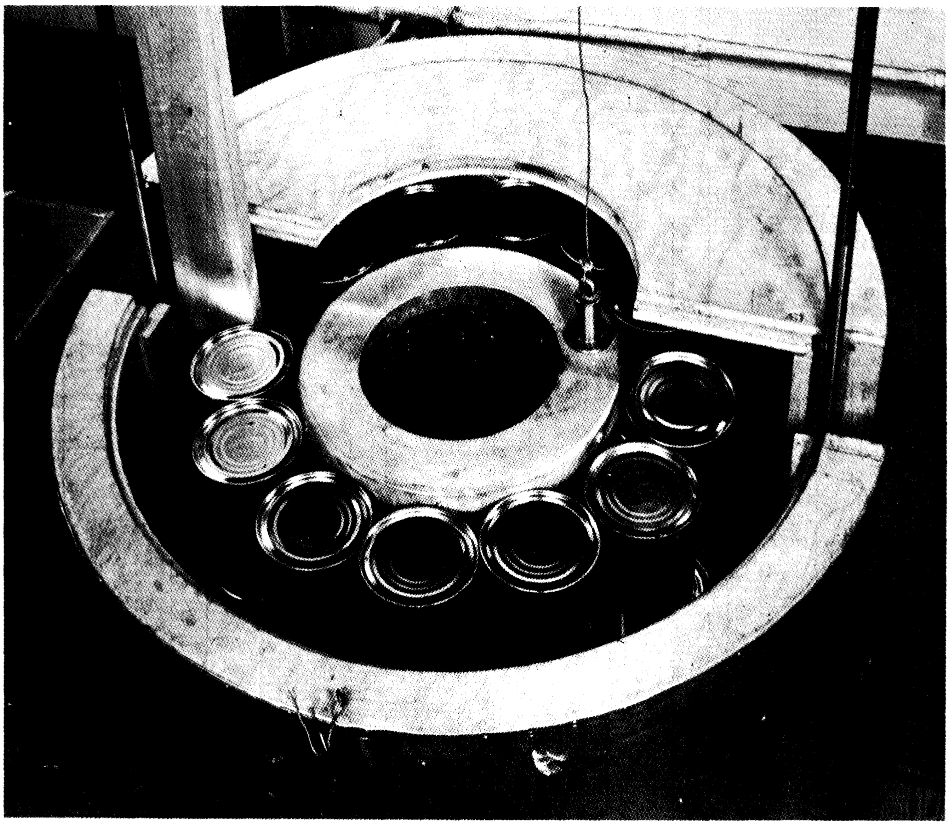

Fig. 63. "Radiation Ice Box" Designed To Permit Irradiation of Canned Fresh Food in the Frozen State. During an Experiment the Spaces are Filled with Ice or Dry Ice and the Box Is Thermally Sealed.

2. Canned Fresh Foods

Several experiments have been performed using commercial cans at various positions. Varying dosage rate in this manner makes it possible to provide a wide range of dosage values with a single exposure. In the case of the experiment illustrated in Fig. 64 an exposure time of 8 hours would yield:

Can	Distance from Center of Can to Wire Basket, in.	Dosage Rate, rep/hour	Total Dose, rep
A	—	250,000	2,000,000
B	3	87,500	700,000
C	4.25	62,500	500,000
D	5.5	50,000	400,000
E	6.75	37,500	300,000
F	9.25	25,000	200,000
G	15	12,500	100,000
H	24	6,250	50,000
I	42	2,500	20,000

In such an experiment, those cans located outside the source cylinder must be rotated, either continually or periodically, to improve the degree of uniformity of dosage throughout the can.

3. Vegetables and Grains

A relatively low level of treatment, 20,000 rep, has been found to prevent reproduction of the insects which infest wheat and other grains and to prevent the sprouting of potatoes and onions under storage. Sacks containing 100 lb of such produce can be conveniently irradiated. Figure 65 shows some potatoes, onions, and wheat in commercial containers in position to be irradiated.

C. FUTURE HIGH-LEVEL GAMMA RADIATION SOURCE

Atomic Energy of Canada, Limited has offered to supply more radiocobalt to make up part of the calculated deficiency in the "10-kilocurie" source. The cobalt rods and aluminum tubes (for jacketing) have been purchased and sent to Chalk River, Canada, and arrangements for the neutron bombardments have been completed. Delivery of the new source is anticipated in the summer of 1955.

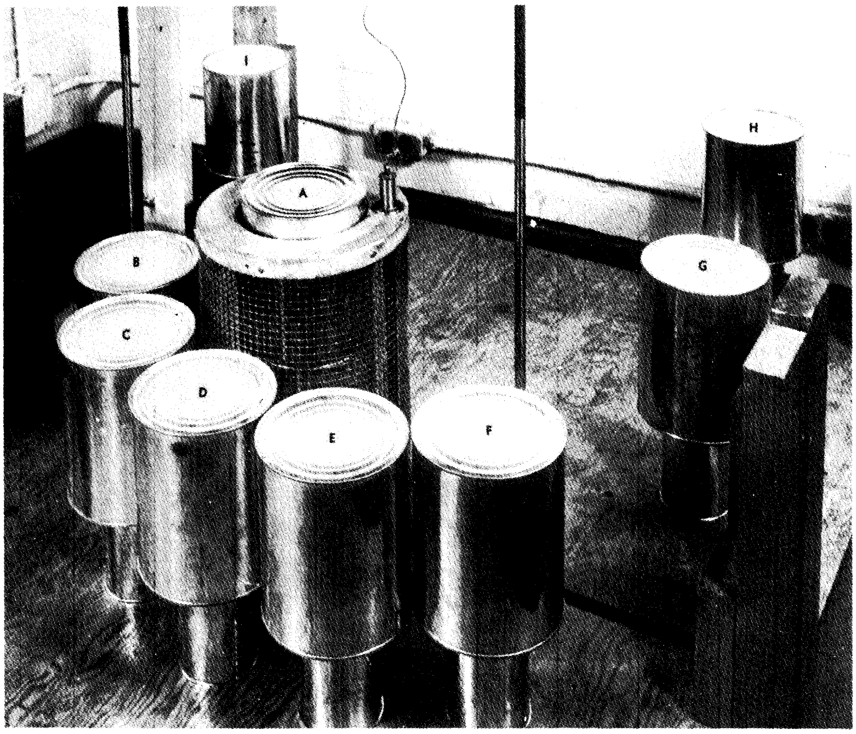

Fig. 64. Commercial No. 10 Cans around the Source in Position To Be Irradiated.

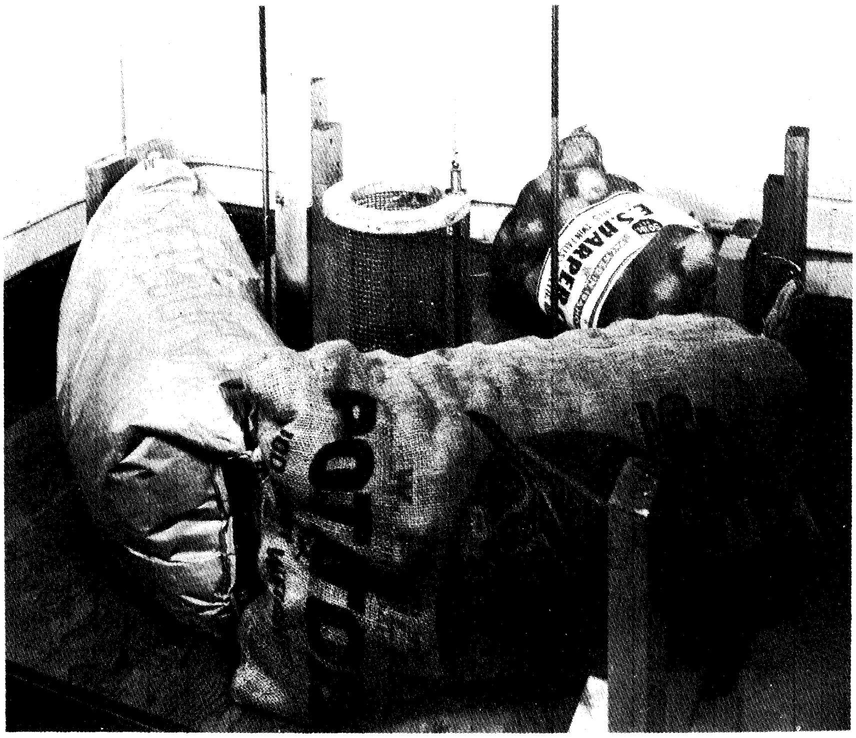

Fig. 65. Potatoes, Onions, and Wheat in Commercial Containers in Position To Be Irradiated.

At about the same time, the new Phoenix Research Laboratory will be completed on the North Campus of the University of Michigan. It is proposed to put the new source into immediate use in the gamma-radiation facility on the ground floor of the Phoenix Laboratory.

The new source will consist of 41 rods physically identical with those used in the present source (see Progress Report 4) with approximately twice the specific activity. These rods will be arranged in a hollow cylinder of somewhat smaller radius than the present source. It is anticipated that the central high-flux position will contain a commercial No. 2 can. The reduction in source diameter will make higher flux levels possible. The estimate is about $1x10^6$ rep per hour.