A STUDY OF THE EFFECT OF STERILIZING IRRADIATION ON THE PERMEABILITY OF CELLULOSICS AND PETROLEUM WAXES

Thesis for the Degree of M. S.
MICHIGAN STATE UNIVERSITY
Hugh Elvin Lockhart
1960

ABSTRACT

The effect of two levels of sterilizing radiation on the permeability of cellulosics and petroleum wax was studied. Non-radiated samples were included as a control. The particular radiation levels under consideration, one and six megarads, fall into a borderline area between weak and strong radiation. The findings of this study indicate that the effect on water vapor permeability is negligible.

The experimental work involved included selection of an effective method for casting 50 mil films of methyl cellulose and petroleum wax, performance and evaluation of water vapor transmission tests, and construction of gas permeability apparatus.

A STUDY OF THE EFFECT OF STERILIZING IRRADIATION ON THE PERMEABILITY OF CELLULOSICS AND PETROLEUM WAXES

By

HUGH ELVIN LOCKHART

A THESIS

Submitted to the College of Agriculture
Michigan State University of Agriculture and
Applied Science in partial fulfillment of
the requirements for the degree of

MASTER OF SCIENCE

School of Packaging

1960

ACKNOWLEDGEMENT

This study was pursued under the sponsorship of the Quartermaster Food and Container Laboratory, Chicago, Illinois.

Special thanks are extended to Dr. H. J. Raphael for his assistance and encouragement during the course of this effort. Beverly Lockhart shouldered cheerfully the usual burden which befalls the wife of a graduate student in undertaking the task of typing.

TABLE OF CONTENTS

INTRODUCTION

BACKGROUND

EXPERIMENTAL PROCEDURE

RESULTS

CONCLUSIONS AND RECOMMENDATIONS

APPENDICES

INTRODUCTION

Purpose of Study

This thesis covers approximately one half of a project undertaken by the writer for the United States Army Quartermaster Corps Food and Container Research Laboratories, Chicago, Illinois. It is anticipated that in the future most if not all of military food packaging will involve sterilization by irradiation as opposed to such methods as sterilization by heating, or arrest of bacterial action by freezing. The reasons for this are readily apparent. Once irradiated, foodstuffs can be stored indefinitely under a wide variety of conditions, ranging from below zero cold to tropical heat and humidity. The only requirement to be met is that of providing a barrier against contamination during storage.

For meats, one possible barrier (or package) is a dip coating of some kind. This kind of package is comparatively inexpensive, is easily and quickly applied, and is generally easy to open, since, ideally the dip coating material needs only to be slit once, then stripped off the product. In addition, a properly applied dip coating should provide a "perfect" seal with no joints or breaks to give discontinuities in the package.

Since irradiated foods need no special storage conditions, the ideal package for these foods would be one which can follow the product. That is, one which itself will withstand all conditions from arctic subzero cold to tropical heat and humidity. It is over this entire range that military food must be protected. Ideally, any given package should give protection over this range, thus eliminating many materials and many steps from the handling of military foodstuffs.

With these considerations in mind, this phase of the study concerns itself with the water vapor and oxygen permeability characteristics of dip coating materials before and after sterilizing irradiation doses when subjected to varying conditions of temperature and humidity.

Scope of Study

Basically, the Quartermaster Corps contract calls for investigation of two commercial cellulosic materials and of waxes available from one manufacturer. The cellulosic materials are Ethocell, manufactured by Dow Chemical Company and Wascopak manufactured by Wasco Chemical Company. Ethocell is an ethyl cellulose and the Wascopak is believed to be the same.

The difference between the two is most likely in the plasticizers used. These, of course, are not known. The wax selected is a blend of 70% microcrystalline, 20% paraffine and 10% butyl plasticizer, manufactured by Kalamazoo Paraffine Company. In addition, it was requested that such other materials as were discovered be investigated. In addition to the tests mentioned above, blocking tests and impact tests were to be conducted on all materials. In the interest of brevity, coherence, and continuity, only the permeability tests on the cellulosics and the waxes have been selected for discussion in this report.

Definitions and Explanations

Dip coating is a definitive term. The product to be coated is dipped in the coating material. Normally this is a hot melt process, with cellulosics heated to 300°F - 325°F and waxes - those used in this study - heated to 200°F - 220°F. The coating material adheres to the product at a thickness determined by viscosity, melt temperature, temperature of the dipped product and time of immersion. Generally, coating thickness increases with higher viscosity, lower temperature of melt and lower temperature of product. Longer immersion time also results in thicker coating.

To say that the coating adheres to the product is not entirely true, at least not in high degree. Actually, an ideal dip coating would not adhere to the product at all, but would solidify around it and by cohesive force alone seal it off from the environment. The better dip coating materials approach this ideal closely, hence their ease of removal. It has been noted that in cases where spots of good adhesion have occurred, small pieces of meat have been torn out of the block of meat upon removal of the coating. This effect was ordinarily associated with localized thawing of frozen meat before and during dipping. This indicates the importance of dipping frozen meats.

Water vapor transmission rate or water vapor permeability is defined as the weight of water transmitted per unit time per unit area of test specimen. This is easily determined as the slope of the plot of weight of water gained by a dessicant versus time. The transmission of water vapor is usually not constant over an entire determination, but displays considerable variability during the first few readings and again at the end of the test when the dessicant approaches saturation. For most purposes the slope of the curve is computed from a curve drawn on coordinate paper.

However, for this study the slopes were determined statistically in order to obtain a quantitative estimate of the variation occurring.

Water vapor permeability data are, of course, a direct measure of the rate at which a material transmits water vapor to or from the item it encloses. These data are adequate for the comparison of different materials, but for a good estimate of package performance more information must be known. Of prime importance is the amount of moisture gain or loss the product can tolerate. Also of prime importance is information regarding such discontinuities in the materials as seals and seams. In the particular case of dip coatings, discontinuities may occur at the point of suspension of the product or at points of contact if the package should be handled prior to complete setting of the film.

The cellulosic materials studied are subject to the accumulation of air bubbles within the melt during melting and dipping. These, if included in a coating, would contribute discontinuities to the coating. Because of the unpredictability of all of these sources of discontinuity, every effort was made to avoid their inclusion in the study. The results reported are for clear films without detectable discontinuity.

Gas permeability is a concept similar to water vapor permeability. By definition, gas permeability is volume of gas transmitted per unit area per unit time at standard temperature and pressure. In this study, the test gas is oxygen. The interest in oxygen arises from the fact that this gas has a deleterious effect on meat, affecting color and palatability. The same arguments and restrictions apply to use of this data as were stated in the paragraphs referring to water vapor permeability.

It has been determined that a proper sterilizing radiation dose for foods is on the order of six megarads radiation.[1] Dunn, Campbell, Fram and Hutchins(4)[2] report that spore forming micro-organisms are difficult to eliminate and that these are completely destroyed by 2 mega roentgens radiation. They report also that all of the micro-organisms in raw milk and soil have been destroyed by 1 mega roentgen. The mechanism of radiation sterilization has been stated nicely by Trump and Van de Graff(13): "In the case of living tissue, injury - and in the limit death - of a cell is the result of physicochemical changes brought about by direct absorption of ionizing energy or indirectly by the reaction of surrounding irradiated tissue."

---

(1) Private conversation with Professor Lyman J. Bratzler, Department of Animal Husbandry, Michigan State University.

(2) Numbers in parenthesis refer to references appended.

We will see later that this mechanism is really a modification of the general effect of radiation upon materials in general.

Radiation dosage is computed on the basis of radiation energy absorbed. Thus, any source of energy may be used to produce a given dose. The amount of energy required to produce a particular dose will depend on the dosage unit used, the material being irradiated and the particular radiation source involved. Three units of dosage are in common use, the rad, the roentgen and the rep. The rad is defined as the absorption of 100 ergs radiation energy per gram of material irradiated. The roentgen (r) is defined as 83.8 ergs per gram of air. The amount of radiation required for this will produce 93 ergs per gram of water or tissue. The rep (roentgen equivalent physical) springs from the difference noted above for the roentgen. This unit is equivalent to 83.8 ergs radiation energy absorbed per gram of tissue or water(12). For purposes of comparison, the following radiations will produce a one roentgen dose of radiation absorbed (12):

1.1×10^9 2 mev γ rays / cm^2
7.2×10^8 thermal neutrons / cm^2
7.2×10^7 2 mev neutrons / cm^2
4.3×10^7 1 mev β particles / cm^2

A one rad dose would then be produced by $\frac{100}{83.8}$ times any of the above source radiations (e.g. 1.3×10^9 2 mev γ rays / cm^2).

The general effects of radiation on plastics are quite extensive when they occur. Among these effects can be listed(12):

> Breaking of chemical bonds and formation of free radicals
>
> Dissociation, or degradation of the molecule, and
>
> Rearrangement of the molecule.

These effects may occur at different levels for different molecules; or, at low radiation levels, they may not occur at all.

BACKGROUND

Irradiation

The effect under investigation is that of irradiation upon three specific materials, two cellulosics and a microcrystalline wax formulation. The literature reveals some disagreement among researchers as to the extent of the effects of radiation on cellulosics as used in packaging. One thing is clear, however. The one megarad dosage included in this study is well below the dose level which causes change in cellulosics, while the six megarad dosage falls in a rather ill-defined area where, apparently, changes may or may not occur. Saeman, Lawton and Millett(11) report the depolymerization of cellulose at 10^7 roentgens, but little effect at 10^5roentgens. Karel and Proctor(6) report a deleterious effect on cellophane at doses above 3.5 megareps, but attribute this to the effect of radiation on coatings and plasticizers. For example, they found the WVTR of coated cellophane to be as much as 2 - 2.5 times greater for dosages of 7 megareps than for non-irradiated film.

The essence of the theory of the damaging effects of irradiation has been stated by Slater(10); "Radiation damage is the result of displacement of atoms in a solid by particles passing through it, of the particles which remain embedded in the solid as impurities, and of the ionization produced by the particles." He modifies this statement with the explanation that the effect of radiation on covalent compounds is one of electronic excitation rather than atomic. In the covalent bond, electrons are displaced from one configuration to another and are there trapped for a relatively long time. This effect can occur at relatively low energy levels. It is not necessarily damaging, since the shift in configuration could bring about an improvement in properties as is the case with crosslinking(12). Charlesby(3) points out that in order to be greatly affected, a plastic must consist of linear or branched chains. Here, then, is an indication of what might be expected. The cellulose molecule is of course not linear or branched, its configuration being a three dimensional ring structure. This would indicate that a cellulosic material ought not to be greatly affected. However, the plasticizers used in cellophane and other cellulosics may be of a linear or branched chain arrangement,(6) and here is where the effect may occur, as was found experimentally by Karel and Proctor as mentioned above.

With reference to the waxes, the wax itself is a linear molecule(5) so it seems reasonable to expect that there may be some effect on the material as a result of irradiation.

Permeability

The mechanism of permeation through a film for water vapor and the permanent gases (O_2, N_2, H_2) is essentially the same. Obviously, in the presence of holes or cracks, the movement of gas or vapor is a mass movement, or diffusion governed by laws of flow through tubes and orifices (1,8). This, however, is considered to be outside the area of interest in the investigation of gas barriers for packaging. In the absence of these faults in the barrier, the mechanism is one of activated diffusion(9). That is, the gas or vapor dissolves in one surface of the film, diffuses through the film under a concentration gradient and evaporates from the film at the low concentration, or low-pressure side.

Under steady state (equilibrium) conditions, Fick's law has been found to apply and the transmission proceeds according to:

$$q = -D \frac{dc}{dx}$$

where q is the amount of gas diffusing through unit area in unit time, D is the diffusion constant and dc/dx is the concentration gradient across thickness dx.

Assuming D to be independant of concentration, this expression can be integrated across the total thickness of the film ℓ to obtain:

$$q = \frac{D\ (c_1 - c_2)}{\ell}$$

where c_1 and c_2 are the concentrations of the gas in the two surfaces.

Normally gas concentrations are measured in terms of the pressure, p, of the gas which is at equilibrium with the film, and c can thus be expressed by means of Henry's law $c = S \cdot p$ where S is the solubility coefficient of the gas in the film. Thus,

$$q = \frac{D \cdot S\ (p_1 - p_2)}{\ell}$$

The product D·S is referred to as the permeability constant P, and we have

$$P = D \cdot S\ \frac{\ell \cdot q}{(p_1 - p_2)}$$

This treatment assumes that D and S are independent of concentration. This is generally true for permanent gases, but deviations occur for water and organic vapors. P is always found to be independent of thickness.(9)[3]

Rogers and associates(9) and Riddell(8) have shown that the usual expression for P is:

$$P = \frac{Q\ell}{Atdp}$$

---

(3) This development of permeability theory has been taken from Rogers, et al, "Studies in the Gas and Vapor Permeability of Plastic Films and Coated Papers," Reference 9.

where Q is the total volume of gas passing through the film, ℓ is the thickness of the film, A is the area under test, t is the time, and dp is the pressure differential between the two surfaces of the film. The units for P are usually expressed as cm^3 of gas at S.T.P. per second per cm Hg per mm thickness per square centimeter surface area.

Physically, diffusion can be thought of as occurring through inter atomic "holes" which inevitably occur in the tangle of polymer chains in the essentially amorphous film. Then, to an extent, the diffusion is dependent on the looseness of the polymer structure. Anything which affects the mobility of the structure affects the diffusion. Mobility refers to the freedom of the atomic structure to move; lengthen, contract, entangle or disentangle. During all of this, a greater or lesser number of holes is appearing, disappearing and reappearing. In a sense, greater mobility implies greater effective looseness. Thus, addition of plasticizers, or increase in temperature will increase the mobility of the polymer structure and so increase the diffusion through the film. Diffusion necessarily depends on the size relationship between the interatomic spaces and the size of the diffusing molecule. Thus, Rogers and associates report a regular decrease in diffusion for the series oxygen, nitrogen, carbon dioxide.

Solubility is highly dependent on the principle of "like dissolves like." In the case of gases, condensation is of even greater importance, the more easily condensed gas being the more soluble. Barrer(1) reports a linear relationship between the logarithm of solubility and the boiling point of a gas dissolved in rubber.

Permeation by water vapor follows the pattern outlined above, but certain special effects must be considered. Many films (e.g. cellulosics, nylon) are hydrophilic substances. These films are plasticized by the water vapor, and diffusion increases with increased plasticizing. This explains the observed phenomenon of increased water vapor permeability with increased relative humidity.

The behavior of waxes when exposed to gas or vapor conditions conducive to permeation is characteristic of crystalline substances. In a crystalline substance, "holes" do not exist within the crystal, that is, there is no free space for the unhindered passage of a molecule of vapor or gas. In addition, the solubility of gas or vapor is decreased by increasing crystallinity. Fox(5) describes waxes as platy, crystalline substances. He reports the failure of waxes to follow the permeability statement of Fick's law, and attributes this to the platy structure.

He proposes that the only practical path for gases and vapors to follow through a wax film is one which requires the molecule to seek a route through the microscopic cracks which exist between the crystalline plates existing in waxes. This argument seems to explain the extremely low permeabilities reported for waxes. Brabender(2) reports that a 100% microcrystalline wax passed only 0.01 grams of water vapor through ten square inches surface area in twenty-seven days.

EXPERIMENTAL

PROCEDURE

Preliminary Work

The first step in the investigation was that of determining the thickness of coat of each material when dipped at the manufacturer's recommended temperature. Here a direct approach was used. Frozen meat was cut into blocks approximately 2" x 3" x 1", the size being dictated by the size of the melting pot available. The meat blocks were then dipped into the molten material and air cooled.

After cooling, one-half inch strips were cut out of the coating, the length of the strip running from top to bottom of the meat chunk. The thickness of the strips was then determined by means of a T M I deadweight micrometer. It was found that there was a regular increase in thickness from top to bottom for all samples. This is attributed to the considerable run down of molten material which was noticed during the cooling period. For the purposes of this study an average was taken of all readings top to bottom. Here the tacit assumption was made that the variation in permeabilities would be linear with variation in thickness, thus allowing the taking of averages on a simple numerical basis. The averages determined were 48 mils for Ethocell; 42 mils for Wascopak; and 65 mils for microcrystalline wax.

Having obtained an average coating thickness, it was necessary to cast sheets of the materials at the average thickness determined for each. It was desirable to cast fairly large sheets, since one of the tests to be used requires a sample of five inches diameter. After some experimentation it was found that the most effective casting plate for the cellulosics was one of aluminum, ½ inch thick. This thickness was found necessary to support the weight of the material without bending. The large mass of metal also speeded cooling of the caxt material. The sides of the plate were built up to the required thickness with masking tape, this being rapidly applied and easily varied as to height. The most easily handled sheet size was found to be 6" x 12", two sheets being cast on a 13½" square plate. Once the material was poured on the plate, it was necessary to remove the excess with a doctor blade.

To avoid too rapid surface cooling and consequent scuffing of the cast sheet, the doctor blade was heated in a 100°C oven for several hours and then returned to the oven immediately after each use. While it was essential not to chill the air-sheet interface, it was found just as important to provide a quick chill at the plate-sheet interface. With anything less than a quick chill, adhesive forces came into play making removal of the sheet difficult or impossible. Proper use of the doctor blade proved to be an art. Variations in pressure and speed of travel across the sheet surface could cause variations in sheet thickness of as much as 20-30 mils. Practice, however, yielded results, and samples were obtained with extreme variation of 5 mils on either side of the desired thickness. Casting of the wax involved yet another problem. The adherent properties of the wax are marked. A 1" aluminum plate was used, but this did not provide a sufficiently rapid chill to prevent adherence. It was decided that a safe release agent would be petrolatum. This proved to be effective. Petrolatum was selected because of its similarity to the wax itself. Excess petrolatum was removed from the sample by wiping first with a dry cloth and then with a cloth saturated in acetone.

No special procedure was used to obtain representative samples, since in most cases only small lots were received from the manufacturer. In addition, it was assumed that the melt process of manufacture would provide uniformity within each batch, at least.

Water Vapor Transmission Rate

The test used for water vapor transmission was the Technical Association of the Pulp and Paper Industry (TAPPI) Standard T 448m-49, Water Vapor Permeability of Paper and Paperboard. This test was specified by the Quartermaster Corps for water vapor permeability tests. Briefly, the method specifies that a membrane of the material to be tested shall separate an atmosphere below 5% relative humidity at 73°F from one of 50% relative humidity and 73°F. The low R.H. atmosphere is attained by means of a dessicant contained in a test cell, the mouth of which is covered with the test material. The edges of the specimen are sealed into the cell by means of a wax mixture of 60% microcrystalline wax and 40% crystalline paraffine wax. The test area is defined by a template placed on the specimen around which the molten wax. is poured.

Shrinkage of the wax during cooling is negligable, so the test area is defined as being equal to the area of the template in contact with the specimen. This area must be accurately known for computation of the water vapor transmission rate. The area was found to be 53.28 cm^2 and 54.20 cm^2 for the two templates used. The test cell is a small dish about 3 inches in diameter, with a grooved lip to accept the edge sealing wax. (See Figure I) The dessicant used was silica gel in a quantity of about 30 grams occupying approximately 40 cubic centimeters volume. The same method was used for evaluation of water vapor transmission rate at $100^{O}F$ and 90% relative humidity, which conditions were additionally specified by the Quartermaster Corps.

Preliminary tests showed that for the Wascopak at 50% and $73^{O}F$ an inflection point was reached at about the 130th hour. From 0-60 hours a straight line was obtained, and again from 170-280 hours a straight line was obtained. It should be noted that this second straight line had approximately the same slope as the first segment. For this reason, and because the first 60 hours is probably the most significant in applications to food, it was decided

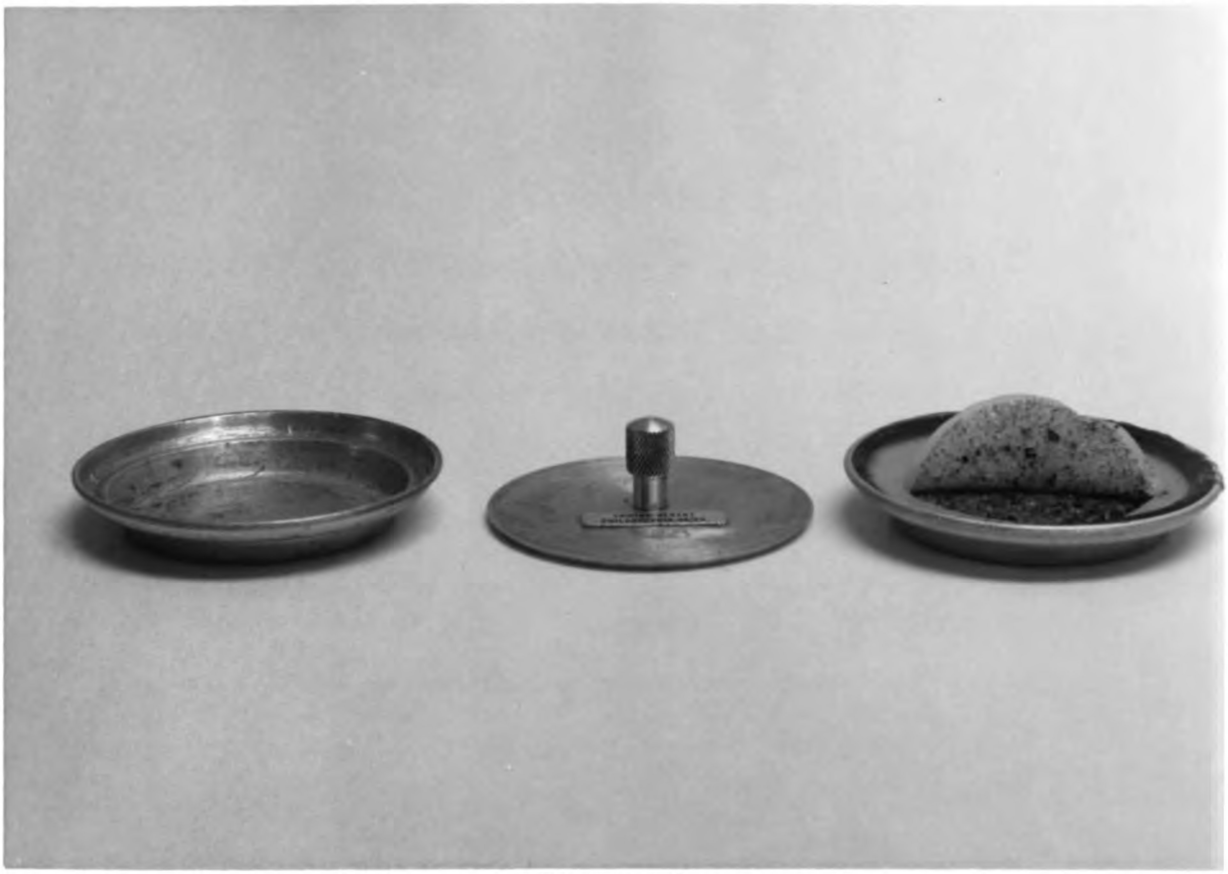

Figure 1
WVTR Test Dish and Sample

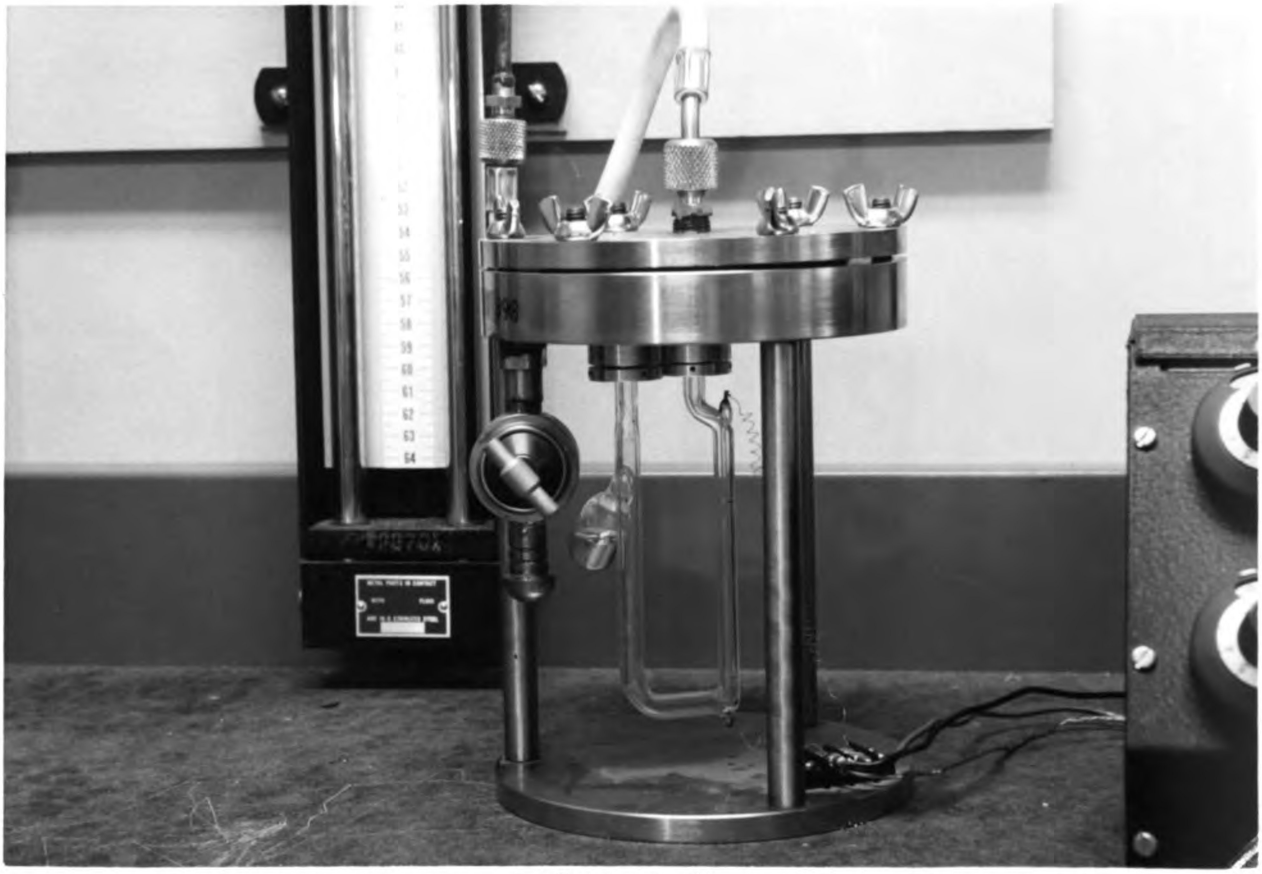

Figure 2
Permeability Cell

to continue the tests for sixty hours then stop. Five specimens of each radiation level (0,1,6 megarads) were tested for each sample of dip coating material. In most cases nine readings of weight were obtained on each specimen before the sixty hour period elapsed.

Oxygen Permeability

The Quartermaster Corps specification for this test was only that a currently approved standard test be used. Investigation revealed that a test developed at Dow Chemical Company was winning wide favor. In fact, a standard test recently published by the American Society for Testing Materials (ASTM Designation D1434-56T Tentative Method of Test for Gas Transmission Rate of Plastic Sheeting) is based on this test. The ASTM test was used for this determination.

Basically, the method of conducting the test is as follows:

A sheet of the material under test is exposed to pure oxygen at a pressure differential through the sheet of one atmosphere. The flow of oxygen through the sheet is measured by means of its effect on a capillary mercury column, the rate of depression of the column being proportional to the rate of flow through

the sheet.

The apparatus used consists of a vacuum tight cell and manometer (See Figure 2) in which the specimen is tightly clamped. The gas is introduced at the top of the cell by means of appropriate tubing from a pressurized cylinder of gas. (See Figure 3) The system is closed at a vacuum pump which serves the purpose of evacuating the system prior to introduction of the test gas (See Figure 3). The entire system is evacuated and flushed with the test gas several times to insure that the permeability being measured is that for the test gas only. In the final step of preparation, the system is evacuated, the cell valve closed, and the mercury dumped into the cell manometer from the storage bulb. Then the gas is introduced into the system to a pressure differential of one atmosphere as measured on the U-tube manometer (See Figure 3).

One of the cells used in the apparatus was purchased from Custom Scientific Instrument Company Model CS-89. The other three were manufactured locally. The cell manometers for the locally manufactured cells were purchased from H. S. Martin and Company, Chicago and were calibrated in the School of Packaging laboratory.

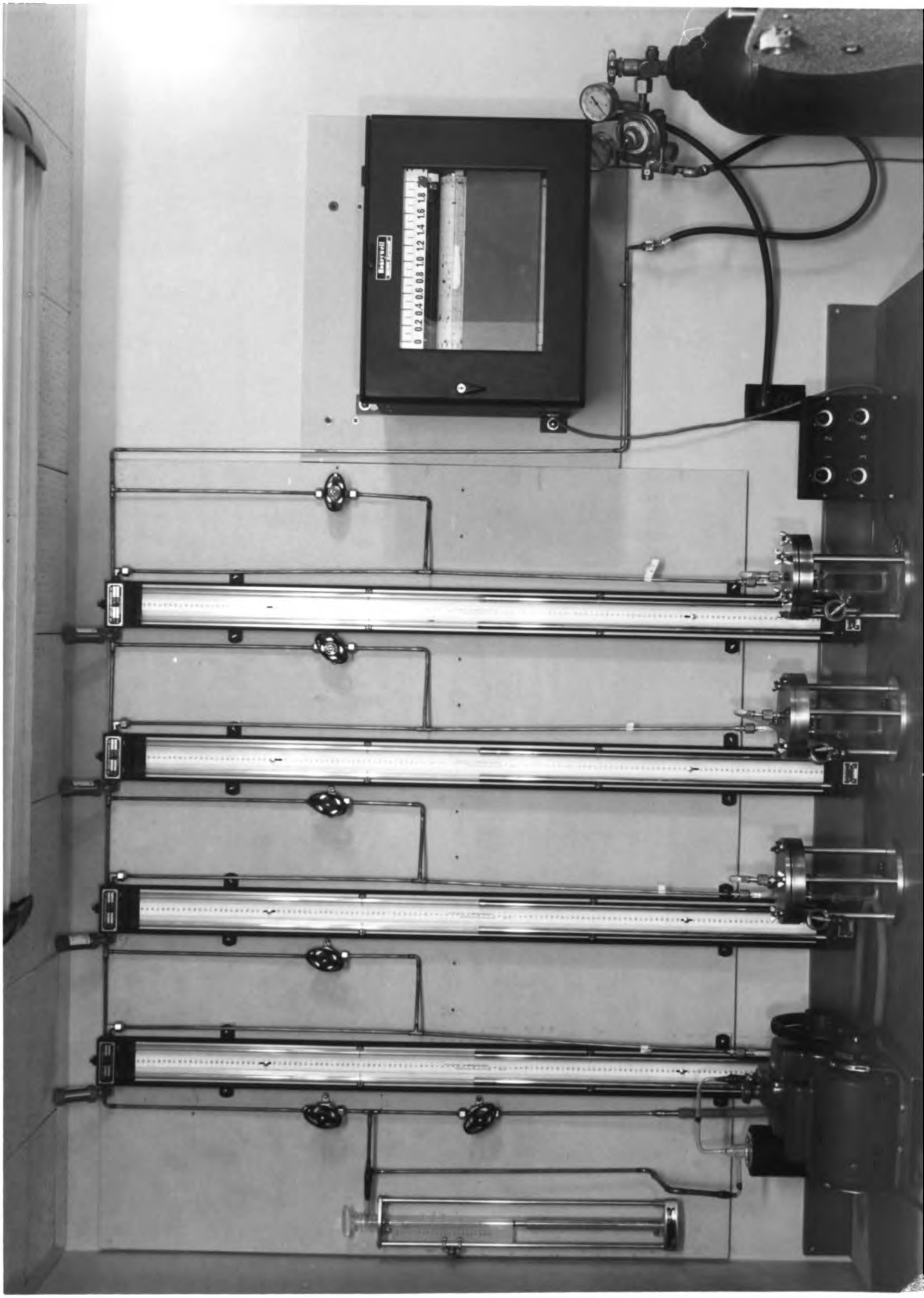

Figure 3
Complete Gas Permeability Apparatus

The first step of the calibration required the determination of the volume of the capillary in cubic millimeters to three significant figures. This was done by mercury emplacement. A mercury pressure system was devised using a Yale hypodermic syringe of 50 cc capacity. This was mounted in a locally manufactured screw operated closing device (See Figure 5).

The entire system was filled with mercury and attached with a spring clamp to the inverted cell manometer (See Figure 4). A glass tubing connector was flanged and ground in the packaging laboratory. The face of each flange was lightly greased to aid in sealing. Between the two flanges was placed a three component slide system. The slide was copper shim stock 0.012" thick, about two inches long by three eighths of an inch wide with a one eighth inch hole drilled in the center (See Figure 4). This was placed across the middle of the lower flange area. Two smaller pieces of the same material were placed on each side of the slide to provide support. The manometer was then lowered into contact with the slide assembly and the whole assembly clamped together. By means of the screw operated hypodermic plunger, the mercury was forced into the capillary.

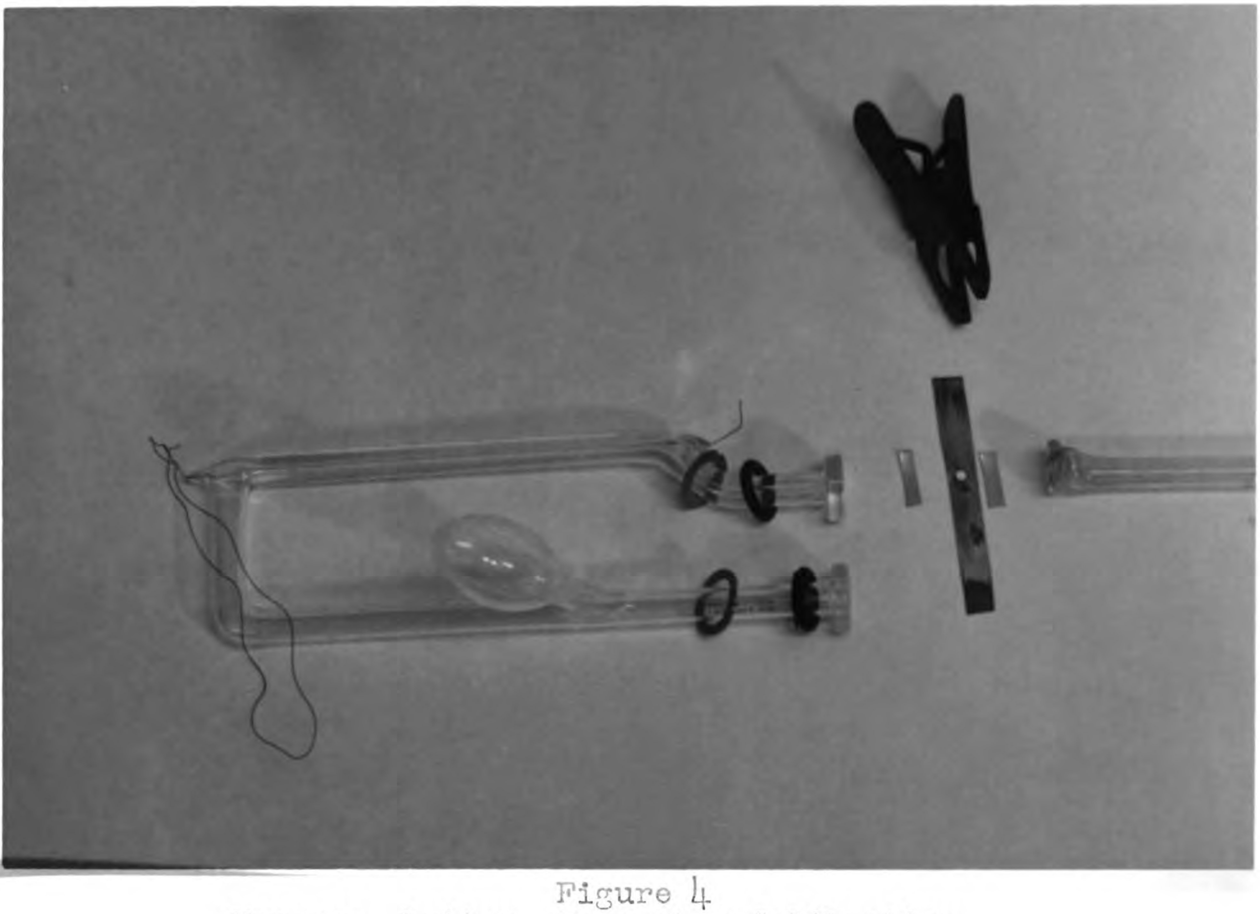

Figure 4
Mercury Shutter, Manometer Calibration

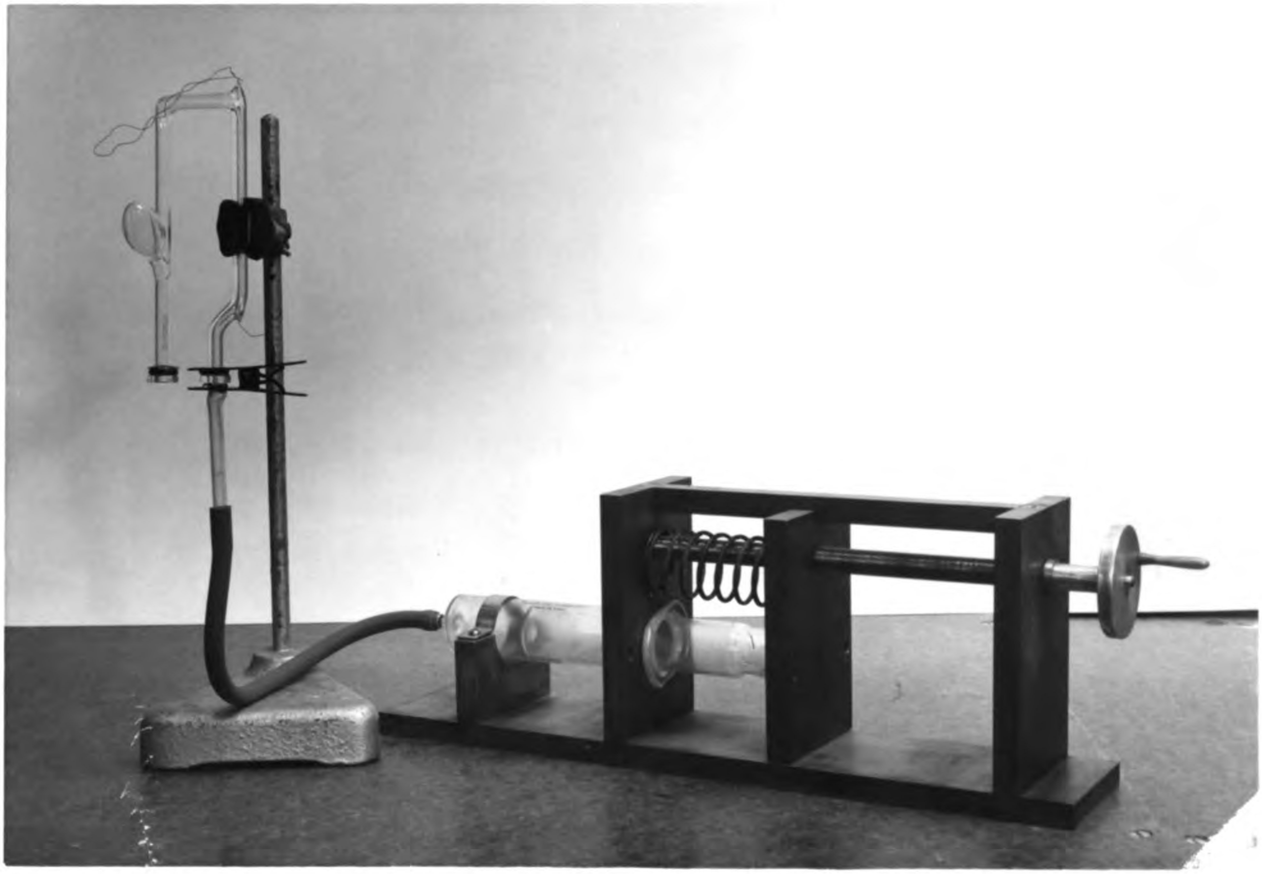

Figure 5
Manometer Calibration Apparatus

Care was taken to obtain as flat a meniscus as possible. This meniscus was then aligned with one of two marks scribed on the tube. The slide was then pushed through, sealing the mercury into the capillary. Then by weight difference the amount of mercury and consequently the volume of the capillary were determined. This volume was determined for two sections of the capillary - one from the opening to the first scribed line, the other from the first line to the second, a distance of 65 mm ± 0.2mm. These volumes were determined by the above method with a variation between highest and lowest values which was of the order of 1% of the average volume. At least three determinations were made for each volume of each tube. From the volume and the distance between the scribed lines, the area of the capillary was determined. These values will all be used directly or indirectly in later calculations. The remaining volume - in the body of the cell - is determinable from the direct measurement of the openings in question. These measurements were made with a Starrett No. 122 Vernier Caliper graduated in 0.001" on the vernier.

The problem of constructing an adequate vacuum system consumed a large portion of the research time available. The method used specifies that a vacuum of 0.2 mm mercury or better shall be attained. This is not considered to be a high vacuum and so this range is not discussed in high vacuum literature. Experience proved, however, that this degree of vacuum is neither easily attained nor easily held once reached. Since the vacuum was originally felt to be rather easily attained, a plastic tubing vacuum system was tried, using polyethylene tubing with brass compression fittings manufactured by Dekoron Products Company. The highest vacuum obtained with this system was 5 mm, probably as a result of the multiplicity of threaded fittings in the system. The system was then converted to copper tubing, using solder joints and flare fittings treated with glyptal. The flare fittings were included for easy take-down of the system when necessary for cleaning purposes. This system attains a vacuum of 0.8 mm mercury using a Cenco Hyvac 2 vacuum pump and cold trap to remove oil and water vapors. Cooling is accomplished with acetone and dry ice.

For the sake of efficiency, and in order to have a continuous record, electrical recording of the mercury depression in the cell manometer was selected. A platinum wire was incorporated in the capillary leg of the manometer by the manufacturer. This is connected in parallel with a 50 ohm variable resistor to a Minneapolis-Honeywell Model Y153 x 64 - W8 Brown 8 point recorder with range 0-2 ohms plus an adjustable padding resistor for range 0-8.7 ohms. This arrangement provides for range spreading as well as for zero adjustment. The changing resistance as the mercury column rises or falls is in this way recorded automatically for each cell.

This system, of course, required calibration and this was done as follows: mercury was admitted to the manometer and the level adjusted to zero reading, the level reading being made with a cathetometer. The mercury column was then lowered by steps until several different levels had been read with the cathetometer. For each mercury level reading, the recorder position was recorded, and a graph prepared of mercury level versus recorder pen position (See Figure 6).

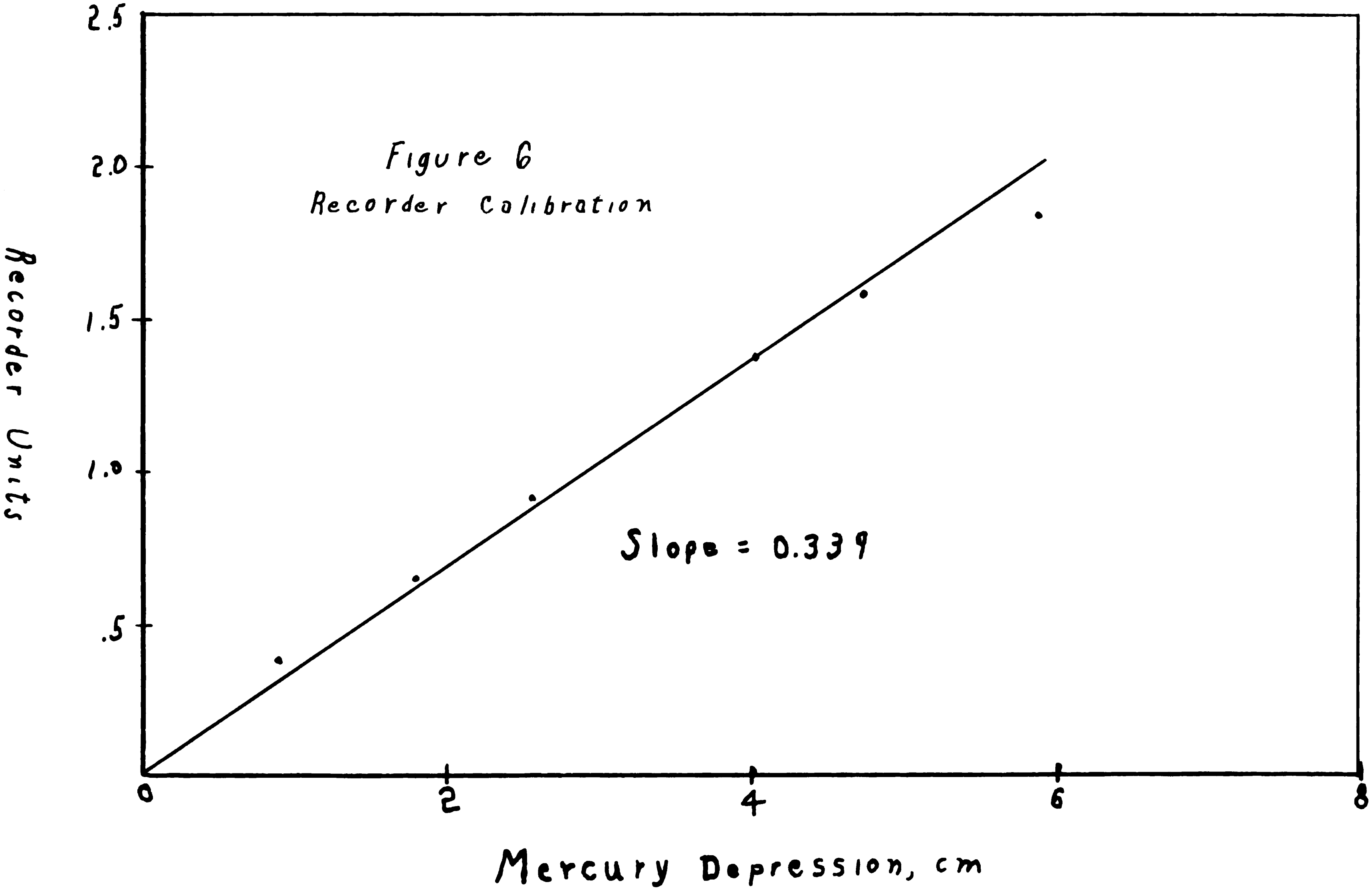

Figure 6
Recorder Calibration

The slope of the line so determined is taken as the recorder travel constant to be used in later calculations as specified in ASTM test D1434-56T.

RESULTS

Data Treatment - Water Vapor Transmission Rate

Raw data (cumulative weights) were converted to weight gained, and this data was treated statistically. The regression of weight gain on time was calculated using a standard regression analysis (see sample calculation, Appendix II, Table VI). Where lack of significance of the regression was indicated, the regression coefficient was not calculated. The coefficient has the usual statistical notation, "b".

For the Wasco material, two cases were found, by means of the gap test or the test for stragglers, in which the values were "wild". In order to complete the variance table, these were treated as missing values and an appropriate value was found by using a series of approximations.

In determining the effect of radiation levels, a two way analysis of variance was employed using the "F" statistic. The variation, σ, was obtained as a result of the calculations in this analysis table. The 5% level of significance was used in determining effect of radiation.

Water Vapor Transmission Rate Results

The results of the WVTR tests are summarized in Tables 1 and 2, Appendix I, while the analysis tables will be found in Appendix II.

It was found that for both cellulosic materials, the radiation had no effect which could be shown with the statistic used. The fact that some samples show an increase in WVTR with increased radiation while others show a decrease can be accepted as evidence that all values represent normal variation from the true mean, which mean is not affected by radiation doseage. Of greater interest is the tremendous variation observed in some WVTR results. A part of this variation may occur as a result of undetected discontinuities in the specimens tested. Another source of this error could be thickness variation. A brief attempt was made to determine if this were the case. A plot was made of WVTR versus thickness for two samples of Wascopak (See Figure 7). This proved inconclusive, however, since in both cases the regression degenerated into a hodge-podge of apparently unrelated points. Many more than five samples would be necessary to make a sensible estimate of any correlation between these two variables.

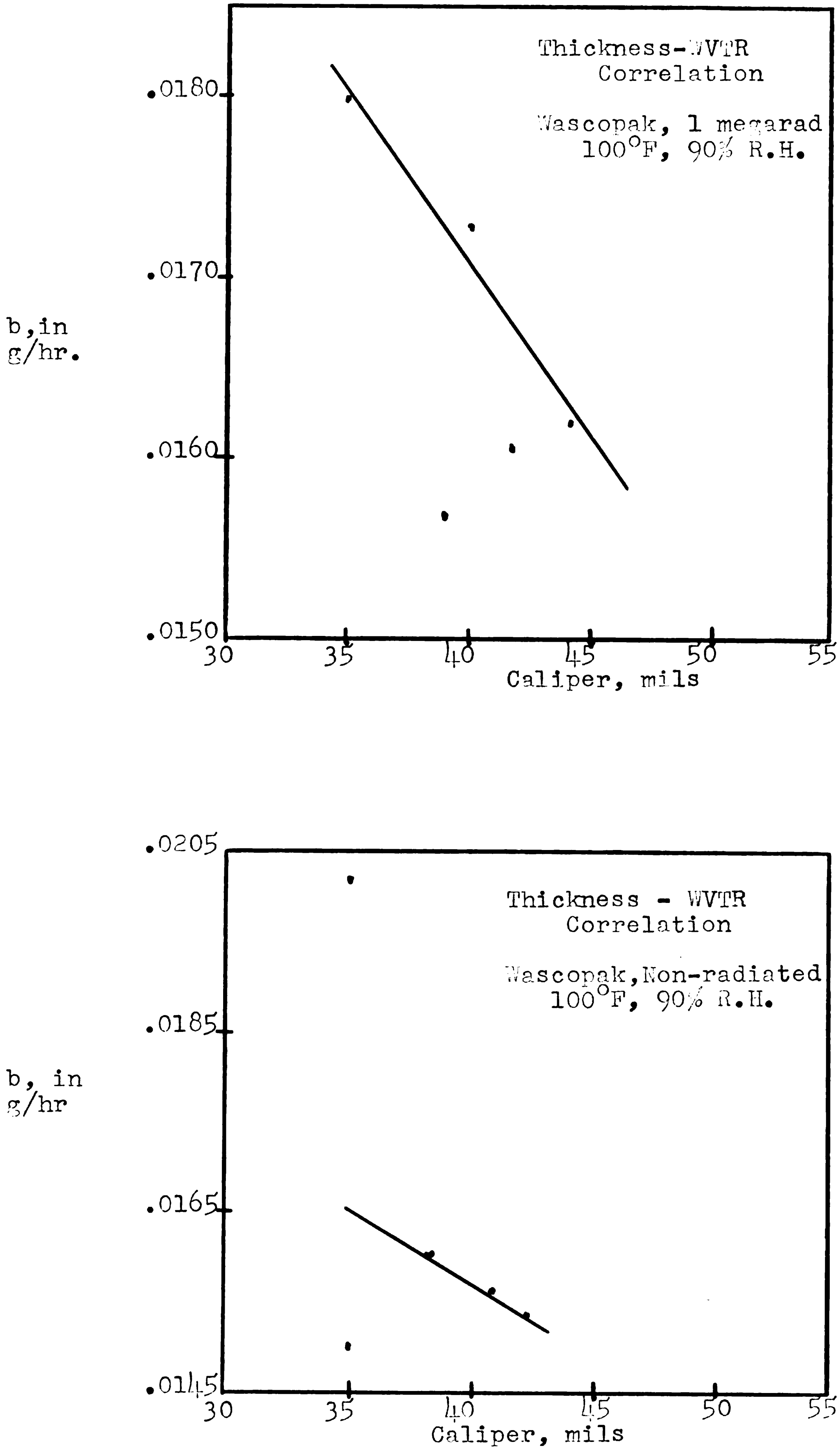
Thickness-WVTR Correlation
Wascopak, 1 megarad
100°F, 90% R.H.
b, in g/hr.
.0180
.0170
.0160
.0150
30
35
40
45
50
55
Caliper, mils
Thickness - WVTR Correlation
Wascopak, Non-radiated
100°F, 90% R.H.
b, in g/hr
.0205
.0185
.0165
.0145
30
35
40
45
50
55
Caliper, mils

Figure 7

In addition, although it is not much discussed in the literature, there is evidence that the relationship between thickness and permeability is not linear. This, of course, complicates the problem of finding the relationship with any degree of accuracy.

The lack of effect of radiation on the cellulosic materials is not surprising, in view of the statements made earlier. It would be expected that there would be little or no effect if the plasticizers were not affected. This seems to have been the case.

Water Vapor Transmission Rate Results with Microcrystalline Wax. The microcrystalline wax behaved much as expected. In the tests run at 73°F and 50% R.H., the wax showed a regular decrease in WVTR with increased radiation doseage (Table 2, Appendix I). This behavior was predicted earlier in the discussion of the effect of radiation on linear and branched chain materials. The fact that this effect begins to appear at 1 megarad doseage is somewhat surprising.

The behavior of the wax at high temperature and humidity was not expected. If anything, the results noted above should have been more pronounced.

Under these conditions, only two of fifteen specimens showed any significant regression at all - where lack of significance is shown, the WVTR is taken as zero. This result, coupled with the fact that weight loss predominates seems to indicate that some volatilization is occurring which covers any WVTR which does exist. An explanation of this may be found in the method of sample preparation discussed earlier. It may be that entrapped acetone, or even petrolatum is volatilized at a temperature of 100°F. Several attempts were made to take data (Tables VII and VIII, Appendix II) which would provide an explanation of this. These showed that specimens treated to eliminate entrapped acetone or petrolatum generally showed a weight increase, but no significant WVTR.

Oxygen Permeability

Because of continuing equipment difficulties, no data have yet been taken on oxygen permeability. However, the test equipment is functioning and project testing will begin immediately.

CONCLUSIONS AND RECOMMENDATIONS

Water Vapor Transmission Rate

It is evident that from the water vapor permeability standpoint, the best of the materials is the microcrystalline wax. However, other tests have shown this material to be lacking in low temperature flexibility. In this case, then, the preferred material is the Ethocell, which does remain flexible at low temperatures and which displays no effect from radiation.

Further investigation could profitably be made. These materials should suit well the investigation of the effect of thickness upon permeability, since it is relatively easy to obtain any desired thickness. This constitutes an area of basic research in the packaging field. One problem which needs to be solved is that of positively excluding air bubbles and surface discontinuities in the case of the cellulosics.

The behavior of the wax samples would provide an interesting study.

Oxygen Permeability

The gas transmission testing apparatus is already under study with an eye to improving it with regard to flexibility and mobility.

A redesign effort will attempt to put it on a rolling cart for transport to various conditioned atmospheres. A variation of the test being planned is that of providing for controlled humidification of the gas introduced. This will provide a means of studying the effect of moisture on various hydrophilic films, as discussed earlier.

The wisdom of using a one atmosphere differential can be questioned, since this is not the normal condition for a packaging material. A solution to this problem is the use of a nitrogen sweep gas on one side of the film with no pressure differential. If this sweep gas is fed to a gas chromatograph, permeability determinations should be quick and accurate.

It was recently pointed out by McAvoy (17) that a serious drawback exists in present methods of calibrating the transmission cell. The present method of calibration, according to McAvoy, does not account for the pressure effect on the filter paper used as a pad for the transmission sample. He reports volume changes under these conditions of as much as 14% of the total volume of the cell, and recommends a calibration method utilizing compressive loads which simulate actual test conditions. This new information will be investigated immediately.

APPENDIX I

Table I
Results of Water Vapor Transmission Rate Tests on Cellulosic Materials
(100°F, 90% R.H.)

Material	Radiation level Megarads	b g/hr/50cm^2	WVTR g/m^2/24hrs	σ g/m^2/24hrs
Wascopak	0	.0156	74.9	
	1	.0167	80.2	89.3
	6	.0178	85.4	
Dow	0	.00711	34.1	
Ethocell	1	.00827	39.7	5.62
	6	.00815	39.1	
Micro-	0	Not computed		
Crystalline	1	1.22×10^{-4}	.586(1 spec.)	
Wax	6	9.02×10^{-5}	.433(1 spec.)	

Table 2
Results of Water Vapor Transmission Rate Tests
73°F, 50% R.H.

Material	Radiation level megarads	b g/hr/50cm²	WVTR g/m²/24hrs	σ g/m²/24hrs
Wascopak	0	.00247	11.9	26.0
	1	.00263	12.6	
	6	.00256	12.3	
Dow Ethocell	0	.00115	5.52	.432
	1	.00107	5.14	
	6	.00109	5.23	
Micro-Crystalline Wax	0	3.66×10^{-5}	.176 (5spec.)	.0816
	1	2.44×10^{-5}	.117 (4spec.)	
	6	-7.1×10^{-5}	-.341 (1spec.)	

Table 3

Thickness of Water Vapor Transmission Specimens

(All Tests)

Specimens tested at 100°F, 90% R.H.

Material	Dose Level	Specimen Thickness, mils 1	2	3	4	5
Ethocell	0	45	42	50	43	55
	1	37	43	41	37	42
	6	--	--	--	--	--
Wascopak	0	42	35	35	41	38
	1	44	40	35	41	39
	6	41	41	45	41	41
Microcrystalline Wax	0	65	66	66		63
	1	66	62	65	61	64
	6	70	65	65	60	65

Specimens tested at 73°F, 50% R.H.

Ethocell	0	48	54	45	47	48
	1	51	50	53	49	51
	6	47	48	54	55	51
Wascopak	0	44	40	41	45	41
	1	38	38	40	40	39
	6	42	40	39	41	43
Microcrystalline Wax	0	65	66	68	62	64
	1	68	70	68	65	70
	6	71	66	60	71	69

Table 4
Weight Gain of Water Vapor Transmission Samples
Mascopak 39-94A
100°F 90% R.H.

Non irradiated

Elapsed Time, hrs.	Weight Gain, grams				
	1	2	3	4	5
4.08	0.0597	0.1086	0.0665	0.0786	0.0687
8.08	0.1184	0.1948	0.1239	0.1473	0.1345
12.16	0.1832	0.2803	0.1855	0.2198	0.1985
14.16	0.2150	0.3207	0.2111	0.2491	0.2276
20.83	0.3134	0.4784	0.3086	0.3565	0.3336
27.83	0.4183	0.6127	0.4137	0.4706	0.4501
35.66	0.5448	0.7626	0.5363	0.5865	0.5763
45.00	0.6936	0.9329	0.6790	0.7161	0.7243

1 Megarad Radiation Dose

Elapsed Time, hrs.	Weight Gain, grams				
	1	2	3	4	5
4.08	0.0795	0.0695	0.0737	0.0736	0.0801
8.08	0.1503	0.1373	0.1476	0.1503	0.2102
12.16	0.2188	0.2105	0.2205	0.2333	0.2303
14.16	0.2496	0.2423	0.2539	0.2623	0.2621
20.83	0.3603	0.3563	0.3713	0.3652	0.3693
27.83	0.4757	0.4841	0.4991	0.4793	0.4805
35.58	0.6014	0.6193	0.6420	0.6051	0.6058
45.08	0.7450	0.7761	0.8078	0.7472	0.7475
59.42	0.9783	1.0314	1.0721	0.9753	0.9780

6 Megarads Radiation Dose

Elapsed Time, hrs.	Weight Gain, grams				
	1	2	3	4	5
4.42	0.0899	0.0736	0.0581	0.0695	0.0663
12.50	0.2560	0.2135	0.1852	0.2120	0.1971
22.00	0.4397	0.3722	0.2958	0.3773	0.4632
27.92	0.5693	0.4864	0.3896	0.4831	0.4799
36.33	0.7420	0.6322	0.5174	0.6269	0.5999
47.08	0.9570	0.8282	0.6655	0.8217	0.7645
52.83	1.0705	0.9297	0.7463	0.9215	0.8514
61.50	1.2276	1.0733	0.8602	1.0639	0.9745

Table 5
Weight Gain of Water Vapor Transmission Samples
Wascopak 39-94A
73°F, 50% R.H.

Non irradiated

Elapsed	Weight Gain, grams				
Time, hrs.	1	2	3	4	5
4.08	0.017	0.018	0.020	0.016	0.018
8.42	0.030	0.031	0.036	0.028	0.033
12.08	0.039	0.040	0.047	0.038	0.043
21.08	0.061	0.063	0.075	0.058	0.070
28.58	0.079	0.082	0.097	0.076	0.091
35.08	0.095	0.098	0.114	0.092	0.109
44.58	0.114	0.117	0.136	0.110	0.131
53.42	0.134	0.137	0.159	0.129	0.153
58.92	0.146	0.149	0.173	0.140	0.166

1 Megarad Radiation

Elapsed	Weight Gain, grams				
Time, hrs.	1	2	3	4	5
4.08	0.017	0.016	0.017	0.016	0.015
8.42	0.032	0.030	0.032	0.029	0.024
12.08	0.043	0.040	0.042	0.038	0.039
21.08	0.069	0.066	0.068	0.063	0.064
28.58	0.090	0.086	0.090	0.082	0.084
35.08	0.108	0.104	0.108	0.099	0.101
44.58	0.130	0.126	0.130	0.119	0.122
53.42	0.153	0.148	0.152	0.140	0.144
58.92	0.167	0.162	0.166	0.153	0.158

6 Megarads Radiation

Elapsed	Weight Gain, grams				
Time, hrs.	1	2	3	4	5
4.08	0.015	0.019	0.017	0.014	0.012
8.42	0.029	0.024	0.033	0.028	0.025
12.08	0.039	0.033	0.045	0.036	0.034
21.08	0.065	0.055	0.073	0.060	0.056
28.58	0.086	0.075	0.097	0.080	0.075
35.08	0.105	0.091	0.116	0.097	0.091
44.58	0.126	0.110	0.140	0.117	0.110
53.42	0.149	0.130	0.165	0.139	0.129
58.92	0.163	0.143	0.180	0.152	0.141

Table 6
Weight Gain of Water Vapor
Transmission Samples
Ethocell
100°F, 90% R.H.

Non irradiated

Elapsed Time, hrs.	Weight Gain, grams 1	2	3	4	5
4.33	0.0235	0.0292	0.0229	0.0280	0.0199
12.75	0.0762	0.0869	0.0698	0.0830	0.0659
22.25	0.1509	0.1898	0.1246	0.1626	0.1175
28.08	0.2072	0.2052	0.1650	0.1936	0.1567
36.50	0.2592	0.2668	0.2140	0.2513	0.2036
47.16	0.3393	0.4212	0.3081	0.3560	0.2700
53.00	0.3821	0.3910*	0.3180	0.3807	0.3050
61.58	0.4460	0.4544	0.3691	0.4344	0.3529

*Apparent failure after 47th hour

1 Megarad Radiation Dose

Elapsed Time, hrs.	Weight Gain, grams 1	2	3	4	5
4.25	0.0107	0.0298	0.0319	0.0412	0.0542
12.58	0.0760	0.0822	0.0949	0.1145	0.0943
22.08	0.1590	0.1430	0.1661	0.1990	0.1635
28.00	0.2081	0.1813	0.2162	0.2581	0.2126
36.42	0.2722	0.2414	0.2807	0.3345	0.2747
46.92	0.3638	0.3196	0.3695	0.4387	0.3600
52.75	0.4123	0.3596	0.4168	0.4957	0.4053
61.42	0.4809	0.4162	0.6241	0.5741	0.4693

6 Megarads Radiation Dose

Elapsed Time, hrs.	Weight Gain, grams 1	2	3	4	5
4.58	0.0392	0.0329	0.0367	0.0778*	0.0347
8.33	0.0725	0.0601	0.0665	0.0610	0.0654
16.91	0.1379	0.1170	0.1321	0.1296	0.1329
22.91	0.1905	0.1522	0.1819	0.1734	0.1830
28.91	0.2396	0.1981	0.2284	0.2286	0.2304
40.25	0.3299	0.2743	0.3159	0.3189	0.3195
46.11	0.3858	0.3209	0.3693	0.3740	0.3782
52.50	0.4404	0.3645	0.4213	0.4274	0.4260
62.50	0.5278	0.4321	0.5051	0.5142	0.5927

*Liquid water on surface of sample

Table 7
Weight Gain of Water Vapor Transmission Samples
Ethocell
73°F, 50% R.H.

Non irradiated

Elapsed	Weight Gain, grams				
Time, hrs.	1	2	3	4	5
4.75	0.006	0.005	0.007	0.006	0.005
8.08	.010	.010	.012	.011	.011
10.75	.014	.013	.017	.016	.015
21.25	.027	.025	.030	.028	.027
28.33	.036	.033	.040	.038	.037
46.08	.054	.048	.058	.056	.054
51.92	.062	.055	.066	.063	.061
60.08	.071	.063	.076	.072	.071

1 Megarads Radiation

Elapsed	Weight Gain, grams				
Time, hrs.	1	2	3	4	5
4.75	0.005	0.004	0.005	0.005	0.004
8.08	.010	.009	.010	.010	.009
10.75	.014	.013	.014	.015	.013
21.25	.026	.025	.025	.027	.025
28.33	.034	.034	.030	.036	.034
46.08	.050	.050	.050	.051	.050
51.92	.057	.057	.056	.057	.057
60.08	.066	.065	.065	.066	.066

6 Megarads Radiation

Elapsed	Weight Gain, grams				
Time, hrs.	1	2	3	4	5
4.75	0.004	0.004	0.004	0.003	0.004
8.08	.010	.010	.008	.007	-.005
10.75	.014	.014	.012	.011	-.004
21.25	.027	.027	.022	.021	.008
28.33	.036	.036	.031	.029	.017
46.08	.055	.055	.046	.043	.034
51.92	.062	.062	.053	.049	.041
60.08	.072	.072	.060	.057	.048

Table 8
Weight Gain of Water Vapor Transmission Samples
Microcrystalline Wax
100°F, 90% R.H.

Non irradiated

Elapsed Time, hrs.	Weight Gain, grams 1	2	3	4	5
4.42	-0.0107	-0.0065	-0.0178	-0.0065	-0.0056
10.58	- .0095	- .0069	- .0160	- .0064	- .0064
21.50	- .0105	- .0069	- .0177	- .0059	- .0064
27.25	- .0061	- .0063	- .0070	- .0056	- .0052
30.08	- .0081	- .0058	- .0163	- .0044	- .0046
45.25	- .0106	- .0089	- .0194	- .0068	- .0066
52.25	- .0071	- .0063	- .0157	- .0039	- .0036

1 Megarad Radiation

Elapsed Time, hrs.	Weight Gain, grams 1	2	3	4	5
4.33	-0.0077	-0.0018	-0.0043	-0.0040	-0.0041
10.50	- .0086	- .0020	- .0050	- .0019	- .0037
21.42	- .0090	- .0012	- .0056	- .0014	- .0042
27.17	- .0081	+ .0056	- .0065	- .0017	- .0021
30.00	- .0076	+ .0001	- .0061	- .0009	- .0013
45.17	- .0097	- .0043	- .0082	+ .0008	- .0041
52.17	- .0075	- .0012	- .0055	+ .0030	- .0005

6 Megarads Radiation

Elapsed Time, hrs.	Weight Gain, grams 1	2	3	4	5
4.33	-0.0043	-0.0023	-0.0022	-0.0036	+0.0021
10.50	- .0062	- .0010	- .0018	- .0030	+ .0026
21.42	- .0058	+ .0102	- .0008	- .0029	- .0089
27.17	- .0049	+ .0100	- .0007	- .0041	- .0081
30.00	- .0050	+ .0103	- .0003	+ .0070	- .0090
45.17	- .0091	+ .0020	+ .0011	+ .0059	- .0084
52.17	- .0063	+ .0032	+ .0023	- .0006	- .0073

Table 9
Weight Gain of Water Vapor
Transmission Samples
Microcrystalline Wax
73°F, 50% R.H.

Non irradiated

Elapsed			Weight Gain, grams		
Time, hrs.	1	2	3	4	5
5.33	0.0025	0.0022	0.0028	0.0023	0.0024
12.00	.0032	.0033	.0036	.0027	.0031
22.33	.0039	.0037	.0043	.0031	.0045
29.67	.0048	.0048	.0053	.0042	.0056
45.58	.0042	.0040	.0047	.0034	.0051
54.42	.0049	.0046	.0053	.0044	.0055
60.42	.0046	.0043	.0050	.0039	.0053

1 Megarad Radiation

Elapsed			Weight Gain, grams		
Time, hrs.	1	2	3	4	5
5.33	0.0021	0.0023	0.0018	.0022	.0026
12.00	.0026	.0026	.0019	.0027	.0035
22.33	.0028	.0028	.0021	.0030	.0041
29.67	.0038	.0042	.0023	.0037	.0047
45.58	.0031	.0034	.0016	.0033	.0048
54.42	.0042	.0050	.0016	.0037	.0047

6 Megarads Radiation

Elapsed			Weight Gain, grams		
Time, hrs.	1	2	3	4	5
5.33	0.0008	0.0018	0.0003	0.0001	0.0066
12.00	.0014	.0026	.0002	-.0004	.0073
22.33	.0023	.0027	-.0003	-.0012	.0075
29.67	.0015	.0037	-.0011	-.0011	.0077
45.58	.0015	.0027	-.0023	-.0028	.0075
54.42	.0005	.0036	-.0032	-.0031	.0069
60.42	.0007	.0031	-.0035	-.0036	.0068

APPENDIX II

Table I
Analysis of Variance
Water Vapor Transmission Rate
Wascopak
100°F, 90% R.H.

		Specimen					
Radiation Levels	0	154*	203	150	156	160	823
	1	162	173	180	161	157	833
	6	201	177	141	175	156	850
	T	517	553	471	492	473	2506

* These values are b x 10^4

Source of Variance	Sum of Squares	df	Mean Square	σ	F
Total	441,000	14			
Replications	7,000	4	1,750		
Radiation Levels	157,000	2	78,500		2.25 n.s.
Error	277,000	8	34,600	186	

True σ_e = 0.0186 g_{H_2O}/hr

Table II
Analysis of Variance
Water Vapor Transmission Rate
Wascopak
73°F, 50% R.H.

		Specimen					
		1	2	3	4	5	T
	0	232*	237	275	225	268	1237
Radiation	1	270	264	270	248	261	1313
Levels	6	268	232	295	250	233	1278
	T	770	733	840	723	762	3828

* These values are $b \times 10^5$

Source of Variance	Sum of Squares	df	Mean Square	σ	F
Total	5744×10^3	14			
Replications	2815×10^3	4	704×10^3		
Radiation Levels	578×10^3	2	289×10^3		.98 n.s.
Error	2351×10^3	8	294×10^3	5.42×10^2	

True $\sigma_e = .00542\ g_{H_2O}/hr$

Table III
Analysis of Variance
Water Vapor Transmission Rate
Dow Ethocell
100°F, 90% R.H.

Specimen

		1	2	3	4	5	T
	0	744*	873	624	727	588	3556
Radiation Levels	1	826	685	939	940	747	4137
	6	840	691	806	839	900	4076
	T	2410	2249	2369	2506	2235	11769

* These values are b x 10^5

Source of Variance	Sum of Squares	df	Mean Square	σ	F
Total	166,826	14			
Replications	17,217	4			
Radiation Levels	40,779	2	20,389		1.50 n.s.
Error	108,830	8	13,603	116.8	

True σ_e = .00117 g_{H_2O}/hr

Table IV
Analysis of Variance
Water Vapor Transmission Rate
Dow Ethocell
73°F, 50% R.H.

		Specimen					
		1	2	3	4	5	T
	0	117*	102	122	118	115	547
Radiation	1	107	108	106	107	109	537
Levels	6	120	120	110	95	95	540
	T	344	330	338	320	319	1651

* These values are $b \times 10^5$

Source of Variance	Sum of Squares	df	Mean Square	σ	F
Total	1035	14			
Replications	169	4	42.25		
Radiation Levels	160	2	80.00		n.s.
Error	706	8	88.25	9.4	

True $\sigma_e = .00009\ g_{H_2O}/hr$

Table V_A
Analysis of Variance
Water Vapor Transmission Rate
Microcrystalline Wax
73°F, 50% R.H.

		Specimens					
		1	2	3	4	5	T
Radiation Levels	0	3.5*	3.2	3.6	3.0	5.0	18.3
	1	3.0	4.6	0	2.4	3.2	13.2
	6	0†	0	-7.1	0	0	-7.1
	T	6.5	7.8	-3.5	5.4	8.2	24.4

* These values are b x 10^5

† Where regression proved not significant, the value is entered as 0.

Source of Variance	Sum of Squares	df	Mean Square	σ	F
Total	126.4	14			
Replications	31.0	4			
Radiation Levels	72.3	2	36.1		12.4**
Error	23.1	8	2.9	1.7×10^{-5}	

σ_e = .000017 g/hr

Table V_B
Analysis of Variance
Water Vapor Transmission Rate
Microcrystalline Wax
73°F, 50% R.H.

Specimens

		1	2	3	4	5	T
Radiation Levels	0	3.5*	3.2	3.6	3.0	5.0	18.3
	1	3.0	4.6	-1.0	2.4	3.2	12.2
	6	-1.1	2.0	-7.1	-4.7	-0.1	-11.0
	T	5.4	9.8	-4.5	.7	8.1	19.5

* These values are b x 10^5

Source of Variance	Sum of Squares	df	Mean Square	F	σ
Total	169.00	14			
Replications	45.15	4	11.29		
Radiation Levels	95.60	2	47.80	13.5**	
Error	28.25	8	3.53		1.88

True $\sigma_e = 1.88 \times 10^{-5} = 0.00002\,g_{H_2O}/hr$

Table VI

Regression Analysis Sample Calculation

We need to obtain the sums of squares of deviations. These are obtained from:

$$\Sigma x^2 = \Sigma X^2 - \frac{(\Sigma X)^2}{N}$$

$$\Sigma y^2 = \Sigma Y^2 - \frac{(\Sigma Y)^2}{N}$$

and $$\Sigma xy = \Sigma XY - \frac{(\Sigma X)(\Sigma Y)}{N}$$

and $$b = \frac{\Sigma xy}{\Sigma x^2}$$

Using W - 0 - 1 as an example:

$$\Sigma X = 266.24 \qquad N = 9$$

$$\Sigma Y = .716$$

$$\Sigma XY = 28.516$$

$$\frac{\Sigma X \Sigma Y}{N} = \underline{21.181}$$
$$\Sigma xy = 7.335$$

$$\Sigma Y^2 = .074000$$

$$\frac{(\Sigma Y)^2}{N} = \underline{.056962}$$
$$\Sigma y^2 = .017038$$

$$\Sigma X^2 = 11{,}037.898$$

$$\frac{(\Sigma X)^2}{N} = \underline{7{,}875.970}$$
$$\Sigma x^2 = 3{,}161.928$$

Sum of Squares for Linear Regression

$$\frac{(\Sigma xy)^2}{\Sigma x} = \frac{53.802}{3161.928} = .017016$$

In the analysis table,

Source of Variance	SS	df	MS	F
Total	.017038	8		
Regression	.017016	1	$17,016\text{x}10^{-6}$	**
Error	.000022	7	$3\text{x}10^{-6}$	

and finally,

$$b = \frac{\Sigma xy}{\Sigma x^2} = \frac{7.335}{3161} = .00232$$

Table VII

Weight Gain
Microcrystalline Wax

Wax placed on glass plate, exposed for two days to 73°F, 50% R.H. then exposed at 100°F and 90% R.H. Petrolatum removed with cloth and acetone

	Wt. Gain, grams			
Elapsed time, hrs	1	2	3	4
2.33	-.0010	-.0006	-.0005	-.0005
4.49	+.0002	+.0006	+.0004	+.0004
7.42	+.0008	+.0013	+.0012	+.0012
11.42	+.0010	+.0015	+.0014	+.0010
23.67	+.0006	+.0018	+.0009	+.0003
27.17	+.0010	-.0070	+.0018	+.0016
29.33	+.0012	-.0066	+.0022	+.0014
46.08	+.0004	-.0074	+.0016	+.0009
51.50	+.0015	-.0098	+.0015	+.0011

Table VIII

Water Vapor Transmission

Test

Microcrystalline Wax

No release agent was used with this wax.

Elapsed Time, hrs.	Weight Gain, grams 1	2
3.42	.0039	.0067
9.16	.0030	.0018
19.42	.0028	.0001
21.42	.0072	.0024
23.42	.0065	.0023
25.67	.0066	.0031
27.42	.0060	.0027
42.92	.0038	.0008
50.42	.0063	.0018

Bibliography

1. Barrer, Richard M., *Diffusion In and Through Solids*, Cambridge University Press London,1941

2. Brabender, George J., *Water Vapor Permeability of Moisture Sensitive Materials* Paper Trade J Oct. 19, 1944

3. Charlesby, Arthur, *Beneficial Effects of Radiation on Plastics and Elastomers*, Nucleonics Vol 14, No. 9, 1956

4. Dunn, Cecil G.; Campbell, William L.; Fram, Harvey; Hutchins, Ardelia: *Biological and Photo-Chemical Effects of High Energy, Electrostatically Produced Roentgen Rays and Cathode Rays*, J. Applied Physics 19 pp 605-616

5. Fox, R.C., *Wax Crystal Structure vs WVTR* Modern Packaging October , 1958

6. Karel, M. and Proctor, B. E. *Cathode Ray Irradiation of Cellophanes*, Modern Packaging January 1957

7. McDermott, P. F., *Moisture Migration* Refrig Eng 42 (Aug. 1941)

8. Riddell, G.L., *Science and Packaging* Chemistry and Industry 66 Jan 18, 1947

9. Rogers, C.; Meyer, J.A.; Stannett, V.; Szwarc, M. *Studies in the Gas and Vapor Permeability of Plastic Films and Coated Papers*, Parts I & II Tappi, Vol 39, No. 11 (1958)

10. Slater, J.C. *Effects of Radiation on Materials* J. Applied Physics 22,237 Mar 51

11. Saeman, Jerome F.; Lawton, Elliot J.; Millet, Merrill A. *Effect of High Energy Cathode Rays on Cellulose* Ind. And Eng Chemistry 44 (12) pp 2048-2052

12. Sun, K.H. *Effects of Atomic Radiations on High* Polymers, Modern Plastics (327(1) September 1954

13. Trump, J.G. and Van de Graff, R.J. *Irradiation of Biological Materials by High Energy Roentgen Rays and Cathode Rays.* J. Applied Physics 19 pp 599-604

14. Weiser, Harry B. *Colloid Chemistry* John Wiley and Sons, New York 2nd Edition, 1958

15. Yarwood, J. *High Vacuum Technique* John Wiley and Sons, New York, 1955

16. Goulden, Cyril H . *Methods of Statistical Analysis* John Wiley and Sons, 2nd Ed. 1952

17. McAvoy, Thomas, *A New Calibration Technique for Gas Transmission Measuring Apparatus* ASTM Bulletin, Jan. 1960