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POLYMERIZATION OF ETHYLENE BY GAMMA RADIATION

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The production of waste fission products from the operation of nuclear reactors has created a difficult disposal problem because of the highly radioactive character of these wastes. A temporary solution of the problem has been found by constructing expensive storage facilities to isolate the wastes. In order to reduce the cost of this program to the taxpayer, the Atomic Energy Commission decided to institute at different laboratories and universities a series of investigations to discover and develop useful applications for the fission products that are now regarded as costly wastes. This and the succeeding paper deal with the use of radiation from fission products to catalyze or promote chemical reactions.

The gross fission products are powerful emitters of gamma and beta radiation. It has long been known that both these types of radiation can cause some chemical reactions to occur. Beta radiation has a very low penetrating power; therefore, application of it in promoting chemical reaction requires almost direct contact between the reacting chemicals and the source of radiation. Since the fission products might contaminate any chemical products if used in direct contact, most of the effort in this study was devoted to the promotion of chemical reactions by the highly penetrating gamma radiation emanating from a source that can be kept isolated from the reacting chemicals.

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At the time this work was started fission product sources of gamma radiation had not been made available by the Atomic Energy Commission, because of difficulties in concentrating and packaging the fission products. However, several government laboratories were producing artificial cobalt-60 in nuclear piles. Cobalt-60 emits a gamma radiation which is comparable, though not identical, with the spectrum of gamma emissions from gross fission products. Two cobalt-60 sources were used to supply the radiation for these studies. One of these sources was nominally rated at one kilocurie and the other at ten kilocuries. (3)(14)

A great many compounds and mixtures of different compounds were irradiated with the cobalt-60 sources and a number of chemical reactions were detected. Among these reactions is the polymerization of ethylene gas both when pure and when mixed with other compounds. This reaction is considered in detail in this paper.

PREVIOUS WORK

Ethylene was polymerized at 77°F and about 1 atmosphere by Lind, Bardwell, and Perry (13) using alpha radiation from radon to initiate the reaction. The products were liquids, reported to be olefins of higher molecular weight, and gases, reported to be hydrogen and methane.

Ethylene was observed to polymerize when irradiated with ultraviolet light. LeRoy and Steacie (10) noted that the metal-sensitizers, cadmium, zinc, and mercury, accelerated the polymerization of ethylene irradiated with ultraviolet light and simultaneously caused some dehydrogenation to acetylene. In this work temperatures ranged from 77 to 572°F and pressures from 15 mm to 400 mm of mercury absolute.

Danby and Hinshelwood (4) investigated the polymerization of ethylene by ultraviolet light at 572°F and pressures to 400 mm of mercury absolute, as sensitized by acetone, by diethyl ketone, and by acetaldehyde. Taylor and Emeleus (14) found that the polymerization of ethylene was photosensitized by ammonia at 68 to 212°F and 5 to 15 cm of mercury, though no solid polymer was produced. The same authors also found (20) that methyl and ethyl amines at 2 to 25 cm of mercury and temperatures of 392°F showed similar behavior. These reactions were studied chiefly in the gas phase.

Ethylene has also been polymerized by gamma radiation. The reaction was studied by Bretton et al. (7) at 1 atmosphere and at temperatures ranging from room temperature to 372°F. The products were solids, usually yellowish or brown in color; other properties were not reported.

APPARATUS

A special reactor bomb (Figure 1) designed to work at 2000 psi and 650°F was constructed and used for the polymerization of ethylene by means of gamma irradiation. It was necessary for the reactor to be inserted into an access hole 1 1/2 inches in diameter in the kilocurie cobalt-60 source, (~~Figure~~ (14)). The thickness of the walls of the vessel was kept to a minimum in order to make a maximum amount of working space available within the bomb and also to allow a maximum amount of gamma radiation to pass through to the contents. AISI 304 stainless steel was chosen as the material of construction for the head, body, and flanges of the bomb. The parts were welded together with AISI 347 rod, except for the body flange, which was screwed on. A tongue-and-groove joint was used between the body and the head. The tongue was machined on the body, the walls of which

were too thin to receive the groove. The head-flanges were of slightly smaller diameter and thickness than normally recommended in order to fit inside the kilocurie source. In spite of the ^{small} size the head of the bomb accommodated two electrical power leads, four thermocouple leads, and entrance line, an exit line, a pressure guage, and a rupture disc. There was barely room to accommodate two 1/4 inch tubing lines inside the gasket circle of the head, and consequently most of the connections were made through the side of the bar which was used for the head. The same set of holes was used to admit process materials and electrical leads, and the pressure gauge and rupture disc were place on the external tubing lines. The bomb was hydrostatically tested at 2800 psi, hammer tested at 2000 psi and all welds were radiographed before the vessel was placed in service.

EXPERIMENTAL PROCEDURE

In this work ethylene was irradiated while at room temperature and at pressures of 250 to 1600 pounds per square inch. ~~pressures~~. Some tests were made in which ethylene was reacted alone and some in which the ethylene was mixed with other reactants. Before making a run the reaction bomb was evacuated to a pressure of less than 1 mm of mercury, after which ethylene was added from a supply cylinder. The bomb was then placed in either the 1 kilocurie source or the 10 kilocurie source until the proper dose had been accumulated. After irradiation, the bomb was removed from the source, the unreacted ethylene was vented and analyzed by an Orsat apparatus, and the accumulated polymer was removed mechanically and weighed. The mass of ethylene charged to the bomb was determined from the measured temperature, pressure, and volume, by means of the thermodynamic

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properties given by York and White (21). The conditions prevailing during reaction were noted and average values were assumed to have prevailed throughout the course of the reaction.

RESULTS OF IRRADIATING ETHYLENE

A white solid polyethylene resulted from the irradiation of ethylene with cobalt-60 gamma rays. A quantitative measure of the reaction was determined by calculating the radiation yield in terms of the "G" value; i.e., the number of molecules of ethylene undergoing polymerization per 100 electron volts of energy absorbed from the radiation. The roentgen equivalent physical, or rep, was adopted as the unit of measure of the absorption of gamma radiation by ethylene. The rep was taken to correspond to the absorption of 93 ergs per gram of absorber as is customary in this field. The dose rate in rep per hour in the absorber was determined by a combination of methods. Chemical dosimetry was conducted by Harmer (1) using the method of Weiss (22), and the results were correlated and extended by the methods of Lewis, Nehemias, Harmer, and Martin (11). The dose rates so determined were applied to the calculations of the radiation yields.

In place of G a quantity A was generally calculated in this work. A was defined as the gram moles reacted per metric ton of ethylene charged per megarep of radiation. By a comparison of units the following relation can be established:

$$A \left(\frac{\text{gram moles reacted}}{\text{metric ton} \times \text{megarep}} \right) = \frac{G}{1.04} \left(\frac{\text{molecules reacted}}{100 \text{ electron volts}} \right)$$

Consequently, A can be seen to be almost equal numerically to G, and the dimensions of the two quantities can be seen to be equivalent. With some simplification the following relation was developed:

$$A \left(\frac{\text{gram moles reacted}}{\text{metric ton} \times \text{megarep}} \right) = \frac{\text{weight fraction ethylene reacted} \times 10^6}{\text{molecular weight of ethylene} \times \text{dose, megarep}}$$

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The simplified relation was used in calculating A values in the following work. These A values are plotted versus the radiation dose in Figure 2.

The yield of polymer was found to be quite small until the system had received a dose of about $2\frac{1}{2}$ megarep. The yield then increased rapidly to a value of about 2500 ~~grammoles~~ reacted/(metric ton)(megarep) at about 3 megarep, and remained nearly constant up to doses of 7 megarep, the highest dose studied (See Fig. ~~2B~~²). It should be noted that 1 gram-mole reacted/(metric ton)(megarep) is equivalent to 0.97 molecules reacted per 100 electron-volts. About $\frac{1}{3}$ of the monomer was polymerized in three days in the center of the 10 kilocurie source.

DISCUSSION OF POLYMERIZATION OF ETHYLENE

In ^{many of} the early runs the time of radiation was approximately the same as what was later found to be the induction period. The amounts of product from these runs varied from practically nothing to very appreciable quantities. Before the length of time of the induction period was ascertained, it was thought that impurities in the ethylene might be responsible for the inconsistent results. ^{See Table I} The ethylene was analyzed immediately upon removal from the storage cylinder, after charging to the reactor but before irradiation, and on removing from the reactor after irradiating (See Table ^{II}). Only ~~trace~~^{small} amounts of impurities, such as carbon monoxide, carbon dioxide, oxygen, paraffin hydrocarbons, and nitrogen were found. There was no apparent correlation between these impurities and the variation in the amount of polymer produced by radiation.

The influence of solvents or other materials which might have been present accidentally in those runs in which appreciable polymerization occurred ~~was checked~~ by adding the following materials successively to separate batches of reacting ethylene: acetone, acetaldehyde, air and acetone, air and water, carbon dioxide, sulfur dioxide. ^{None} of these only sulfur dioxide had any appreciable effect ^{on the polymerization of ethylene}. The addition ~~of large amounts~~ of sulfur dioxide ^{did} result in the production of a white powder which proved to have a sulfur content close to that of the equimolar addition product of sulfur dioxide and ethylene. (See Figure 2). Matthew and Elder (15), and Snow and Frey (17) have reported similar reactions between sulfur dioxide and olefins under ultraviolet light. ~~Small amounts of sulfur dioxide had little effect on the yield.~~ In some runs in which noncondensable gases were vented from liquid ethylene before charging to the reactor the initial rate of reaction was increased to some extent though not enough to explain the discrepancy in yields.

The presence of the long induction period and the explanation of the wide variation in yield of polymer was found when several runs were made with long periods of radiation. What was earlier an apparent inconsistency in results is now readily understandable from an examination of Figure 2, for up to about ^{1/2} megarep little or no reaction occurs; and runs made in that range cannot give consistent yields of product.

Average values of dose rates used in these studies varied from about 30 kilorep/hour to about 90 kilorep/hour. It should be noted however, that errors exist in the method of calculating the dose rates used in estimating the G values. A Victoreen ratemeter was used to measure the dose rates on the axis of the bomb. This instrument detects the secondary photons produced by scattering from the wall of the bomb, but probably does not detect any electrons

liberated from the metal by gamma radiation. Consequently, more ionization probably occurred than was taken into account by the calculations, in which the effect of the bomb wall was neglected; omission of this

effect gives G values which are too high. On the other hand the primary beam was assumed to undergo no appreciable absorption within the ethylene in the bomb. If account were taken of absorption of primaries within the ethylene, ~~there would be~~ ^{then} less radiation ^(would be given credit for) initiating the reaction. Omission of this effect, therefore, gives G values that are too low.

Thus, neglect of nonequilibrium secondaries and neglect of the absorption gradient of primary gamma intensity within the ethylene may be somewhat compensating. The importance of accounting for the above errors in dosimetry is recognized, and the complexity of the problems of measurement would indicate the desirability of pursuing this work further in future studies. The values given for G in Figure ² ~~1~~ should be regarded as relative rather than absolute, since all determinations were made in the same equipment and using similar procedure.

No consistent effect of pressure on G value could be noted. Elevated temperatures were investigated only briefly, but preliminary results indicated that considerably increased rates of polymerization would result in irradiated systems at temperatures of 200-400°F, as compared with those obtained at room temperature.

EVALUATION OF THE POLYETHYLENE PRODUCT

General: The polyethylene obtained as a result of gamma irradiation of ethylene was subjected to a brief program of evaluation. The properties considered most basic to an understanding of the material

were investigated. Most experimental work was concerned with determinations of solution viscosity, melt viscosity, density, and tensile strength. Melting points of some samples were also determined. Molecular weights were estimated from the determinations of viscosities of solutions and of melts. Crystallinity was estimated from determinations of density.

These measurements and derived quantities probably need no further explanation with the exception of the concept of crystallinity of a polymer. The degree of crystallinity of a polymer is measured by the degree to which the molecules of polymer are arranged parallel to each other. An arrangement of parallel molecules results in a repetitive structural pattern such as that found among the molecule of a crystal. A random orientation of molecules similar to a pile of jackstraws might be expected to be less dense than a parallel arrangement such as that just described, and it has been found that percentage crystallinity maybe correlated with the density of polyethylene, (See Kirk-Othmer⁽⁹⁾).

Some explanation is given to the manner of presenting the data obtained from the experiments on the polymer. The properties of the polyethylene are presented as functions of the radiation yield of the polymerization reaction because of the reasons given below. In addition molecular weight and crystallinity are presented as functions of dose.

Lind¹² (~~32~~) has shown that in many gaseous systems, approximately one molecule reacts per ion pair formed in the system. In the irradiation of ethylene a variable number of molecules, usually much greater than one, react for each ion pair formed; evidently, the

polymerization of ethylene is a chain reaction. For this calculation it is assumed that one chain is initiated for every ion pair formed, that all chains are of equal length, and further that the formation of each ion pair requires 32.5 electron-volts of energy; a value (16) approximately correct for gases at one atmosphere. The densities of ethylene under the conditions of reaction were greater than at one atmosphere and therefore the energy required per ion pair may be different from the value given. The G value which is based on 100 electron-volts may therefore be divided by three to give the approximate number of molecules reacted for each ion pair formed, and this result may then be multiplied by the molecular weight of the monomer in order to estimate the average molecular weight of the polymer.

Consequently; since the molecular² weight is directly proportional to the G value, it is advantageous to consider the properties of the polyethylene as functions of the G value. The results of most determinations could be correlated against the G value better than they could against dose, although the G value has been shown to be a ~~function~~ ^{function} of dose. The data in Figure 2 indicate that the G value was about 0.1 to 1.0 until about 0.5 megarep had been received. The G value then increased rapidly with increasing dose until it reached a nearly constant value of about 2000 for doses of about 3 to 7 megarep.

Experimental: All the samples of polyethylene which were produced were white. Some were fluffy powders and others were tough, coherent masses.

Portions of each of the samples of polyethylene which occurred in yields of 4 grams or more were molded into sheets as an operation preliminary to further examination. A two-compartment mold was used, one compartment at a time. Samples were placed between aluminum foil

in the mold, preheated to 300°F, pressed to 1000 psi, and cooled to about 125°F under pressure. The resulting sheets were 2.5 by 4 by 0.025 inch. All such sheets proved to have the characteristic milky, translucent appearance of polyethylene. The sheets molded from the powders were brittle while those from the tough reaction products were also tough.

Molecular weights were estimated from viscosities of solutions, measured as follows: Solutions were prepared in concentrations of 0.01 per cent and of 0.125 per cent by weight in tetralin. Viscosities of these solutions and of the tetralin were measured in modified Ostwald pipettes at 212°F. Specific viscosities were calculated and divided by the respective concentrations. The resulting ratios were plotted as a function of the concentration of polymer, and the curves were extrapolated to zero concentration to give intrinsic viscosity. Intrinsic viscosity was assumed to be directly proportional to molecular weight. The concentration was computed in units of gram moles of monomeric ethylene per liter of solution. The constant of proportionality was computed on the basis that the molecular weight of Bakelite DYNH^{W95} 20,000 as given by Dienes and Klemm (6). Tani (¹⁸~~21~~) also reported on intrinsic viscosities of polyethylene in tetralin.

The method of Dienes and Klemm (⁶~~21~~) was used to estimate molecular weights from melt viscosities. Viscosities were measured in a parallel-plate plastometer with an attached dial ^ggage reading to 0.01 millimeter, and the entire assembly was placed in an oven. Temperatures of 248°F and 266°F were used. The samples were placed between sheets of aluminum foil about 1 1/2 mils thick. The thickness of the sheets of foil was measured in the plastometer before each determination.

Crystallinity was estimated from the density (See Kirk-Othmer⁽⁹⁾). Density was determined by weighing directly in water after degassing the immersed sample under reduced pressure.

Tensile properties of the polyethylenes were examined by the following procedure: Specimens for testing were cut from the molded sheets by means of a die. The resulting specimens were 0.079 by 0.025 inch in the smallest cross section. The narrowest section was 1 1/2 inches long. Tension was applied in a Gardner-Parks testing machine (7).

Melting points were determined on a melting-point bar of the design of Dennis⁵ (~~2~~).

Discussion: The results of evaluation of the properties of the radiation polymerized polyethylene are summarized in Table ~~II~~^{III}. The molecular weight is plotted as a function of radiation yield in Figure ~~2~~³. The molecular weights and crystallinities are plotted as functions of dose in Figure ~~2~~⁴. The values obtained were assumed to be weight average molecular weights. (See Table ~~II~~^{III}) Although differences in crystallinity and cross linking between these samples and thermally polymerized samples may make the relationship less valid.

Determinations of molecular weight by melt viscosity may be subject to similar criticism. As shown in Figure ~~2~~³ and ~~2~~⁴ the molecular weights determined by solution viscosity do not agree well with those determined by melt viscosity. Neither do the molecular weights from solution viscosity appear to display any regular variations with dose or with G value, in contrast to the regular behavior of molecular weights from melt viscosities.

Values of crystallinity are plotted as a function of radiation yield in Figure ~~2~~⁵. The crystallinities varied from about 127 per
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cent for samples of low radiation yield to about 71 per cent for samples of high radiation yield. All these samples were of considerably higher crystallinity than was the Bakelite DYNH, which had a crystallinity of about 61 per cent. It is possible that radiation-polymerized samples were of higher crystallinity than the thermally polymerized sample of DYNH because the temperature of polymerization was lower for the radiation-polymerized samples. The samples of low radiation yield would be expected to be more highly crystalline than those of high radiation yield, since radiation yield has been shown to increase with dose (Figure ~~2~~²). Cross-linking and branching would probably also increase with dose, and both cross-linking and branching decrease crystallinity.

Tensile properties are reported in terms of stress as a function of strain in Figure ~~6~~⁶. The irradiated samples all have properties similar to those of a brittle material. The samples subjected to higher doses have higher tensile strengths and are more ductile than those subjected to lower doses of radiation. Such behavior would be likely if the irradiation increased cross-linking and branching. The Bakelite DYNH shows the characteristic elongation of several hundred percent before rupture [See Kirk-Othmer (~~2~~⁹), p. 942.] Ultimate tensile stress as a function of radiation yield is plotted in Figure ~~5~~⁵. The ultimate tensile stress increased markedly with radiation yield and consequently with dose. A set of structural properties such as those just described for the radiation-polymerized polyethylene might be desirable for certain applications, but the properties differ from those of most polyethylene currently marketed.

Melting points are plotted as a function of radiation yield in Figure 7, ~~being~~ ^{being} curves/given for the upper and the lower ends of

the melting point range. The results show that there is a small increase in the temperature of initial softening with increase in radiation yield, and that there is a large increase in the temperature of complete melting. The higher melting points indicate higher degrees of cross-linking as a result of the higher doses of radiation, and are thus in conformity with the results of the other determinations mentioned above.

SUMMARY

(1) Ethylene was polymerized by exposure to gamma radiation from cobalt-60.

(2) [Polyethylene formed by ^Ygamma irradiation was subjected to a preliminary evaluation. The polymer was found to be denser, less ductile, and of a higher ultimate strength than Bakelite DYNH polyethylene. Molecular weights of the radiation-polymerized materials increased with dose of radiation to a value of about 40,000 when estimated from melt viscosities. Most samples were insoluble in tetralin, and estimates of molecular weights from solution viscosities were not conclusive. Crystallinities estimated from densities varied from 71 to 77 per²-cent.] ↓
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(3) [Mixtures of ethylene and sulfur dioxide were found to polymerize under gamma radiation, giving appreciable yields of a white powder whose composition represented approximately the equimolar addition of ethylene and sulfur dioxide. No studies were made on the properties of this polymer.] end

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Fig. 50. Radiation Yield as Function of Dose of Radiation in Polymerization of Ethylene.

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Fig. 60. Molecular Weight as Function of Radiation Yield of Polyethylene.

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Fig. 4. Molecular Weight and Crystallinity as Functions of Radiation Dose for Polymerization.

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Fig. ⁵~~63~~. Crystallinity and Tensile Strength as Functions of Radiation Yield for Polyethylene.

Fig. ⁶ ~~62~~. Stress-Strain Plots for Test Specimens of Polyethylene.

Fig. 7. Melting Points as Functions of Radiation Yield of Polyethylene.

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TABLE I

IRRADIATION OF ETHYLENE

Order No.	Page No.	Starting Date	Psig		Avg. Psia	Avg. Temp., °F	Grams Polymer	Hours Irrad.	Grams Polymer per Hour	A = (Metric Ton)(Megarep)	G-Moles Reacted		Averaged over Reactor		Other Reactants	Source	Position of Reactor	Mfr.	Cylinder No.	Remarks
			Initial	Final							(Metric Ton)(Megarep)	(Metric Ton)(Megarep)	Dose Rate Kilorep/Hr	Total Dose Megarep						
2	132113	19 Dec 52	830	785	822	69	1.64	69.7	0.024	608.	29.	2.02	1 MI acetone	1 kc	--	--	Matheson	FF 737	--	
3	132118	12 Jan 53	820	795	822	70	0.666	61.	0.011	291.	28.9	1.76	--	1 kc	--	--	"	"	--	
4	132147	5 Mar 53	1430	1350	1395	230	0.292	17.2	0.017	485.	29.5	0.507	--	10 kc	8" NE 9" up	--	"	"	--	
5	132149-II	7 Mar 53	820	700	775	50	0.004	0.9	0.0003	12.7	42.	0.666	--	10 kc	8" NE 9" up	--	"	"	--	
1	132159	26 Nov 52	790	780	800	68	0.394	42.	0.009	258	29.5	1.24	1 MI acetone	1 kc	--	--	"	"	--	
6	132250	10 Mar 53	810	715	777	50	0.167	16.2	0.010	94.5	96.	1.55	--	10 kc	Q on Q, Base at Base	--	"	"	--	
7	132252	11 Mar 53	1430	1330	1395	200	1.04	16.1	0.06	513.	82.5	1.33	--	10 kc	Do.	--	"	"	--	
8	132253	12 Mar 53	698	692	710	45	1.48	87.7	0.017	423.	42.0	3.69	--	10 kc	8" NE 9" up	--	"	"	--	
9	132254	17 Mar 53	655	655	660	45	--	23.3	--	--	42.0	0.979	650 psi N ₂ and 15 psi air vent, chg. C ₂ H ₄	10 kc	8" NE 9" up	--	"	"	--	
10	132255	18 Mar 53	800	710	770	45	0.088	21.5	0.004	865.	42.0	0.904	--	10 kc	8" NE 9" up	--	"	"	--	
11	132256	19 Mar 53	840	760	815	420	0.381	15.3	0.025	1800.	29.3	0.448	--	10 kc	8" NE 9" up	--	"	"	--	
12	132258	23 Mar 53	810	70	455	45	0.010	39.	0.0003	12.5	41.9	1.63	10 MI acetone	10 kc	--	--	"	"	Leak	
13	132259	26 Mar 53	788	183	485	61.	--	15.3	--	--	95.5	1.46	--	10 kc	Q on Q, Base at Base	--	"	"	--	
14	132263-I	4 May 53	1600	1425	1528	72	--	21.9	--	--	27.7	0.608	--	1 kc	--	--	"	"	--	
15	132263-II	5 May 53	620	595	623	72	0.022	23.	0.001	45.8	27.7	0.638	--	1 kc	--	--	"	"	--	
16	132264	7 May 53	615	600	622	69	<0.0001	20.7	<0.000005	<0.25	27.6	0.572	100 MI O ₂	1 kc	--	--	"	"	--	
17	132265	8 May 53	620	608	631	67	<0.0001	21.4	<0.000005	<0.24	27.6	0.591	25 MI O ₂	1 kc	--	--	"	"	--	
18	132266	9 May 53	594	577	601	72	0.034	25.6	0.0013	67.5	27.5	0.703	5.0 MI O ₂	1 kc	--	--	"	"	--	
19	132267	10 May 53	600	575	603	75	<0.001	17.0	<0.00005	3.1	27.5	0.468	2.0 MI O ₂	1 kc	--	--	"	"	--	
20	132268	11 May 53	617	597	622	70	0.017	22.3	0.0008	36.6	27.5	0.613	10.0 MI O ₂	1 kc	--	--	"	"	--	
21	132269	12 May 53	605	590	612	71	0.030	21.1	0.0014	71.	27.5	0.701	10.0 MI O ₂	1 kc	--	--	"	"	--	
22	132270	13 May 53	637	629	638	66	0.016	14.8	0.0011	50.	27.5	0.407	5.0 MI O ₂	1 kc	--	--	"	"	--	
23	132275	4 Jun 53	550	525	553	70	0.031	16.8	0.002	94.	31.3	0.526	--	10 kc	8" SW 13-7/8" up	--	"	"	--	
24	132276	7 Jul 53	490	490	505	76	0.021	22.1	0.001	65.	27.0	0.597	23 mm air 1 MI acetone	1 kc	--	--	"	"	--	
25	132277	8 Jul 53	460	460	475	78	--	20.1	--	0.0	27.0	0.543	95 MI air 1 MI water	1 kc	--	--	"	"	--	
26	132278	9 Jul 53	460	455	473	73	0.011	18.3	0.0006	43.1	27.0	0.494	215 mm CO ₂	1 kc	--	--	"	"	--	
27	132279	10 Jul 53	445	440	458	~75	0.136	69.8	0.002	147.	27.0	1.88	33 mm SO ₂	1 kc	--	--	"	"	--	
28	132280	13 Jul 53	426	395	425	87	Lost, severe grams tar	16.5	--	--	27.0	0.446	4,598 gm AlCl ₃	1 kc	--	--	"	"	--	
29	132281	14 Jul 53	430	420	440	81	0.013	20.0	0.001	52.2	27.0	0.540	--	1 kc	--	--	"	"	--	
30	132282	15 Jul 53	420	410	430	79	0.657	23.2	0.028	184.	27.0	0.627	1 atm SO ₂	1 kc	--	--	"	"	A ₈₀₂ = 575	
31	132284	16 Jul 53	420	385	417	83	2.692	16.1	0.167	241.	27.0	0.435	55 psia SO ₂	1 kc	--	--	"	"	A ₈₀₂ = 3380	
32	132285	17 Jul 53	393	70	246	83	30.9	64.0	0.483	2450.	27.0	1.73	25.96 gm SO ₂	1 kc	--	--	"	"	A ₈₀₂ = 4800	
33	132287	20 Jul 53	400	390	410	--	--	--	--	--	0	0	27.9 gm SO ₂	--	--	--	"	"	--	
34	132293	3 Aug 53	810	790	815	80	0.208	17.0	0.012	357.	26.8	0.456	--	1 kc	--	--	Ohio Chem.	G28087	--	
35	132294	4 Aug 53	930	910	935	76	0.472	21.8	0.022	443.	26.8	0.584	--	1 kc	--	--	"	"	--	
36	132295	5 Aug 53	615	610	628	76	0.010	16.0	0.0006	30.	26.8	0.428	Alk. pyrogallol	1 kc	--	--	Matheson	FF 737	--	
37	132297	6 Aug 53	955	950	968	75	4.45	71.5	0.0623	750.	26.8	1.91	--	1 kc	--	--	"	"	Powdery, soft lumps, white	
38	132300	10 Aug 53	320	320	335	75	0.005	15.7	0.0003	35.6	26.8	0.421	--	1 kc	--	--	"	"	--	
39	132301	11 Aug 53	985	965	990	80	0.084	16.3	0.005	67.	26.8	0.437	--	1 kc	--	--	"	"	--	
40	132303	12 Aug 53	945	930	950	75	0.085	14.6	0.006	112.	26.8	0.392	--	1 kc	--	--	"	"	--	
41	132304	13 Aug 53	860	840	865	80	0.150	16.3	0.009	234.	26.8	0.437	--	1 kc	--	--	"	"	--	
42	132305	14 Aug 53	870	815	857	74	1.70	63.6	0.0268	638.	26.8	1.70	--	1 kc	--	--	"	"	--	
43	132306	17 Aug 53	935	900	932	70	0.309	11.8	0.0262	509.	26.7	0.315	--	1 kc	--	--	Ohio Chem.	G28087	--	
44	132307	18 Aug 53	875	870	888	72	0.428	18.5	0.0231	551.	26.7	0.494	--	1 kc	--	--	"	"	--	
45	132309	20 Aug 53	1015	970	1002	75	<0.005	17.4	<0.0002	7.3	26.7	0.465	--	1 kc	--	--	Matheson	5772	--	
46	132360	21 Aug 53	920	865	920	74	2.6	66.8	0.040	802.	26.7	1.76	--	1 kc	--	--	"	"	--	
47	132361	24 Aug 53	825	785	820	79	--	15.7	--	0.0	26.5	0.416	--	1 kc	--	--	"	"	--	
48	132362	25 Aug 53	975	946	975	79	0.029	14.0	0.0021	28.0	26.5	0.372	--	1 kc	--	--	"	"	--	
49	132363	26 Aug 53	1200	1000	1115	90	21.5	116.5	0.185	2380.	26.5	3.09	--	1 kc	--	--	Carbide and Carbon	JK 370331	In small to medium lumps, slightly off white, moderately coherent	
50	132366	5 Sept 53	1010	924	980	77	4.9	90.5	0.0541	730.	26.5	2.40	--	1 kc	--	--	Matheson	5772	Powdery, in small, soft lumps, white	
51	132369	9 Sept 53	1005	925	980	73	--	17.0	--	0.0	26.5	0.451	--	1 kc	--	--	"	"	--	
52	132370	10 Sept 53	1000	970	1000	70	1.5	21.3	0.0705	790.	26.5	0.565	--	1 kc	--	--	Carbide and Carbon	JK 370331	--	
53	132372	11 Sept 53	513	475	510	60	0.16	34.5	0.00464	298.	26.5	0.912	Acetaldehyde	1 kc	--	--	Matheson	5772	--	
54	132373	14 Sept 53	1500	960	1245	~70	51.3	262.7	0.195	2200.	26.5	6.97	--	1 kc	--	--	"	"	Had to be cut out of bomb. Some fines, all coherent, white	
55	132375	25 Sept 53	1570	1080	1315	~65	43.6	71.8	0.607	1915. avg	89.6	6.43	--	10 kc	On Q	--	"	"	Had to be cut out of bomb. Some fines, all coherent, white	
56	132376	30 Sept 53	1780	1110	1450	79	12.5	17.0 23.7 28.8	0.18 avg	745. avg	89.5 89.5 98.0	0.50 2.12 1.67	--	10 kc	On Q 6-5/8 out	--	U.S. Ind. Chem.	I 1065	White, soft powder, very small lumps	

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TABLE ~~XIV~~

ANALYSES OF ETHYLENE FROM STORAGE CYLINDERS AND FROM REACTOR

Material No.	Mfg.	%CO ₂	%O ₂	%CO	%N ₂	% Combustible as marked	Number of Determinations
1	Math FF737	0.06	0.02	0.02	0.15	0.1 propane?	duplicate
2	Math 5772	0.06	0.02+	0.00	0.07	0.29 propane	duplicate
3	OC G28087	0.10 _±	0.02 _±	0.00+	0.47 total	ethane?	single sample
4	C and C JK370331	0.37	0.02	0.05	1.7	0.8 pentane	triplicate
5	USI IC-1065	0.08	0.05	0.005(?)	0.21	0.45 methane	duplicate

Code: Math- The Matheson Co., Inc.

OC - Ohio Chemical and Surgical Equipment Co.

C and C - Carbide and Carbon Chemicals Corp.

USI - United States Industrial Chemicals, Inc.

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TABLE XIV (cont)

ANALYSES OF ETHYLENE FROM STORAGE CYLINDERS AND FROM REACTOR

Material No.	Dose	G	Page No.	%CO ₂ *	%O ₂ *	%CO*	%N ₂	%Combustible as marked		Number of Determinations
4	3.09	2380	132363	0.37 0.62	0.02 0.07+	0.05 0.07	0.00	3.8	ethane	duplicate; first sample of 1 l. or more
2	2.40	730	132366	0.06 0.05	0.02 0.008	0.00 0.002	0.12	0.14	ethane(?)	duplicate
2	0.45	0	132369	0.06 0.02	0.02 0.05	0.00 0.01	0.22 total		-?-	duplicate
4	0.57	790	132370	0.37 0.34	0.02 0.01	0.05 0.01+	2.49 total		-?-	duplicate
2	0.91	298	132372	0.06 0.05+	0.02 0.005	0.00 0.00	0.23 total		-?-	duplicate; acetaldehyde added
2	6.97	2200	132373	0.06 0.05	0.02 ≤0.01	0.00 0.00	0.09	0.18	methane	single
3	6.43	1915	132375	0.10 0.11	0.02 0.02	0.00 0.009	0.16	0.32	ethane (?)	single
5	4.29	745	132376	0.08 0.07	0.05 0.009	0.005(?) 0.00	0.10	0.63	methane	single

*Top, before irradiation
Bottom, after irradiation

(no removal by distillation and no addition to ethylene unless noted)

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III
TABLE ~~XV~~

PROPERTIES OF POLYETHYLENE PRODUCED

Page Number	Dose, Megarep	Radiation Yield, A	Melting Point, °F Lower/Upper	Density g/cm ³	Ultimate Tensile, psi	Elongation, Percent at rupture	Crystallinity Percent	Molecular Weight by Melt Viscosity	Weight by Solution Viscosity
132250	1.55	95	219/226						
132268	0.61	37	216/225						
132269	0.58	71	205/217						
132276	0.60	63	207/214						
132281	0.54	52	196/205						
132297	1.91	750	241/244	0.951	450	4	77	26,300	insol.
132362	0.37	28	234/235						
132363	3.09	2400	248/689	0.941	2200	42	71	34,400	insol.
132366	2.40	730	210/248	0.951	770	2	77	28,100	4200
132369	0.45	0.1							
132370	0.57	790	203/252						8800
132372	0.91	298	199/207						insol.
132373	6.97	2200	234/720	0.943	2100	29	72	40,500	insol.
132375	6.43	1900	241/610	0.941	2300	79	71	37,300	insol.
132376	4.29	745	241/244	0.951	630	3	77	11,900	3700
Bakelite DYNH				0.921	1500	550	61	21,800	20,000 assumed

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