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**HOT-FLOW CHARACTERISTICS
OF
IRRADIATED POLYETHYLENE**

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GENERAL DYNAMICS | FORT WORTH

ABSTRACT

Polyethylene disks, 0.55 in. diameter by 0.137 in. thick, were irradiated in air in the Ground Test Reactor to five different gamma-ray dose levels: 2.2×10^8 , 4.72×10^8 , 1.64×10^9 , 4.99×10^9 , and 1.33×10^{10} ergs/gm(C) plus the dose imparted by the associated neutron flux. After irradiation, the samples were compressed in a parallel-plate plastometer under constant load of 260°F for a period of five minutes. The thickness and the diameter of the extruded samples were measured at 75°F and found to be correlated with the absorbed gamma-ray dose in a manner described by two empirically derived equations.

An evaluation of the radiation-induced change in viscosity, η , of the disks showed that the dose region where η departs from linearity is coincident with the most sensitive region of response in the hot-flow characteristics [$10^8 - 1.5 \times 10^9$ erg/gm(C)].

The results suggest that in this dose region, the described experimental procedure may be usefully employed for dosimetric purposes.

REPORT SUMMARY

Small disks of low-density (0.01 gm/cm³) polyethylene, 0.55 in. diameter by 0.137 in. thick, were irradiated in air at 75°F in the Ground Test Reactor to five different gamma-ray dose levels: $D_1 = 2.2 \times 10^8$, $D_2 = 4.72 \times 10^8$, $D_3 = 1.64 \times 10^9$, $D_4 = 4.99 \times 10^9$, and $D_5 = 1.33 \times 10^{10}$ ergs/gm(C) plus the dose imparted by the associated neutron flux. After irradiation, the samples were compressed in a parallel-plate plastomer by a constant load of 13,124 gm at 260°F for a period of five minutes. The thus extruded samples were cooled to 75°F and their thickness and diameter were then determined. The following average values of thickness were measured for the controls and the five sets of irradiated (D_1, D_2, D_3, D_4, D_5) samples, respectively: 0.026, 0.041, 0.053, 0.131, 0.138, and 0.136 in. The corresponding values for the diameter of the extruded samples were 1.32, 1.063, 0.894, 0.583, 0.564, and 0.562 in. It was found that these experimental data could be well fitted to the two equations:

$$t_D = \frac{t_u t_o \exp \{2.625 D\}}{t_u + t_o [\exp \{2.625 D\} - 1]}$$

$$d_D = \left[\exp \{-2.625 D\} (d_o^2 - d_u^2) + d_u^2 \right]^{0.5}$$

where t_D, d_D = the thickness and the diameter, respectively, of disk irradiated to dose D gigaergs/gm(C),

t_o, d_o = thickness and diameter, respectively, of extruded unirradiated disk, and

t_u, d_u = original thickness and diameter, respectively, of extruded unirradiated disk.

Accordingly, the most sensitive response of the hot-flow behavior to radiation takes place with a dose range of from 1×10^8 to 1.5×10^9 ergs/gm(C).

Analysis of these data on the basis of the Dienes-Klemm relationship yielded the following viscosity values for the unirradiated and irradiated polyethylene disks:

<u>Dose Level</u> <u>[ergs/gm(C)]</u>	<u>Viscosity</u> <u>(poise)</u>
0	1.925×10^6
D ₁ (2.2×10^8)	1.194×10^7
D ₂ (4.72×10^8)	3.396×10^7
D ₃ (1.64×10^9)	7.595×10^9
D ₄ (4.99×10^9)	∞

The dose region where the viscosity increase is exponential is found to be coincident with the most sensitive region of the flow behavior.

The results suggest that this experimental procedure may be suitable for dosimetric purposes.

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I. INTRODUCTION

The effects of nuclear radiation on polyethylene have been rather extensively studied by a large number of investigators. The principal radiation-induced changes that have been observed in this material are intermolecular crosslink formation, evolution of hydrogen and low-molecular-weight hydrocarbon gases, unsaturation, progressive reduction in crystallinity, discoloration, and, in the presence of air, oxidation reactions.

The formation of intermolecular crosslinks and the disruption of the crystalline regions result in a considerable modification of the physical properties. In the initial phase of irradiation, the polymer, while still fusible, exhibits a markedly increased melt viscosity. This is attributable to the presence, within the otherwise still unaltered system, of high molecular-weight species that have been formed by crosslinking reactions. Concurrently, however, the crystalline areas are progressively destroyed, which is reflected in a decrease in specific density and in heightened room-temperature flexibility.

Upon further irradiation, the polymer is converted into a two-phase gel-sol system. At this stage, the crosslink density has increased to such an extent that a significant fraction of the molecules have been tied into an infusible, insoluble, three dimensional network (gel-phase) interpenetrated by a soluble fraction of "free" molecules still unconstrained by intermolecular bonds. The room-temperature mechanical behavior

of the polymer, at this point, is still mainly a function of the residual degree of crystallinity.

However, above the transition region corresponding to the melting of the crystallites, the material does not flow any longer but exhibits elastomeric behavior typical of loosely crosslinked polymer networks. As crosslinking proceeds, the segmental motions become progressively more restricted. Crystallinity disappears altogether and the mechanical properties are determined predominantly by the crosslink density. The polymer becomes increasingly more brittle and, eventually, is transformed into a glass-like material having a high modulus of elasticity, a low ultimate elongation, and increased specific density.

In their effect on the room-temperature behavior, the two radiation-induced processes of crosslinking and reduction in crystallinity are to some extent compensatory. By examining the polymer above the transition region where melting of the crystallites takes place, one can separate the various effects of crosslinking on several physical properties.

For example, Charlesby in his pioneering studies on irradiated polyethylene (Ref. 1) investigated the extrusion rate of irradiated polyethylene as a function of dose. He used a Davenport grader in which the samples were extruded under standard conditions of pressure at a temperature of 190°C . He found that, over a limited range of dose (0 - 0.4 megarad), the flow rate was a linear function of dose.

The present investigation had as its objective the determination of the residual deformation in small irradiated polyethylene disks after they were allowed to creep for a certain period of time under a static compressive load at a temperature beyond the normal melting range. The expectation was that this residual deformation could, in some manner, be correlated with the dose absorbed by the samples.

II. EXPERIMENTAL METHOD

2.1 Sample Preparation

The samples consisted of small disks of polyethylene, 0.550 (± 0.0024) in. diameter by 0.137 (± 0.0032) in. thick. The disks were cut with a circular die from polyethylene sheet stock having a specific density of 0.91 cm/gm³ and a Shore D hardness of 45. Altogether, 60 such samples were prepared and mounted in groups of 10 on six perforated, rectangular, aluminum trays, each about 2.5 x 5 in. On each tray, the samples were arranged in two parallel rows of five sample each, the distance between to nearest neighbors being about 0.5 in. Each tray was then completely wrapped with aluminum-foil tape. One of the trays was stored in the laboratory at 75°F as a control group, while the remaining five trays were mounted in the center of five rectangular (22 x 26 in.), perforated, aluminum panels.

2.2 Dosimetry

On each of the five panels, four dosimetry packets were arranged, 90° apart, in a circular configuration around the polyethylene samples. The average distance from the center of the panel to the dosimeter was eight inches. Along with the dosimeters, additional organic specimens (elastomers) were mounted symmetrically within the four quadrants of this circular configuration. The dosimeters contained in each of the four packets on the five panels are given in Table I.

Table I
Dosimeter Description

Dosimeter		Contents of Dosimetry Packets on Panel				
		1	2	3	4	5
Type	Radiation Detected					
Aluminum foil	Neutrons ($E > 8$ Mev)	X	X	X	X	X
Sulfur pellet	Neutrons ($E > 2.9$ Mev)	X	X	X		
Sulfur-epoxy disk	Neutrons ($E > 2.9$ Mev)				X	X
Pair of bare and cadmium-covered copper foils	Thermal neutrons	X	X	X	X	X
Nitrous-oxide ampoule	Gamma rays	X		X	X	X
Tetrachloro-ethylene ampoule	Gamma rays	X	X			

Panel 2 was intended to be positioned in the location of minimum total dose [on the order of 10^8 ergs/gm(C)], whereas Panel 1 was supposed to receive the next highest dose [on the order of 5×10^8 ergs/gm(C)]. However, the planned location of these two panels was inadvertently interchanged. This is the reason for the special gamma-ray dosimeter arrangement on the two panels: the tetrachloroethylene dosimeters on Panel 2 were expected to record the lowest dose, while the employment of two types of gamma-ray dosimeters on Panel 1 was designed for a transition dose close to the upper limit of usefulness of the tetrachloroethylene dosimeter and close to the lower limit of the nitrous-oxide system.

2.3 Sample Irradiation

The sample irradiation was carried out in the Radiation Effects Testing System at GD/FW's Nuclear Aerospace Research Facility (NARF). In the system, the Ground Test Reactor (GTR) is used as the radiation source. It is located in one side (the wet side) of a pool divided by a dam wall into a wet and dry side. The dry side of the pool is the irradiation cell. The GTR is positioned in a closet-like structure that is built into the center of the dam and protrudes into the irradiation cell. Thus, three faces of the closet (the GTR) are available for irradiation testing.

The materials, components, or systems that are to be tested are placed in environmental chambers that are transported on pallets down into the irradiation cell by a remotely controlled three-track shuttle system. Temperatures inside the chambers are controlled (-65°F to 450°F) from an air-duct system that terminates beneath the pallets at the three testing positions.

Two environmental chambers were used during the test described here. Panels 1 and 2 were irradiated in one chamber for 2.5 hours at 0.6 Mw reactor power, while Panels 3, 4, and 5 were exposed in a second chamber for 5 hours at 3 Mw. The locations were selected in such a manner that the five panels would receive total gamma-ray doses in the range of from 10^8 to 10^{10} ergs/gm(C). During the irradiation the samples were immersed in an air environment, and the temperature was monitored by thermocouples embedded within standard ASTM compression-set buttons of silicone rubber mounted on the various panels. An

attempt was made to maintain the temperature within these monitored samples as close to 75°F as possible by circulating a refrigerated-air current in the environmental chambers. This was achieved for the samples mounted on Panels 1, 2, and 3. However, the temperature as monitored in the samples on Panels 4 and 5 could not be held constant despite the fact that the ambient air had been cooled to 40°F. Radiation-induced heating produced an appreciable temperature rise in the monitored samples of these two panels. Within a period of about 75 minutes after the reactor had been brought to peak power, the temperature recorded for Panels 4 and 5 had risen to 120°F and 200°F, respectively, and remained at these levels, with short fluctuations of $\pm 10^\circ\text{F}$, until reactor shutdown. It must be kept in mind, however, that the mass of the monitored compression button was about 22 times greater and its surface-to-volume ratio about 30 times smaller than the respective values for the polyethylene disks. For this reason, it may safely be assumed that the latter never reached the high temperatures of the compression-set buttons.

2.4 Sample Testing

The samples were tested 10 days after removal from the reactor. The test apparatus consisted of a simple plastometer in which the sample could be compressed under constant load between two parallel plates. During the test, the plastometer remained in an oven thermostatically maintained at 260°F.

Each sample was sandwiched between two circular tinned platens and this assembly was introduced between the plates of the plastometer. Only one sample was compressed at a time. After application of a static load of 13,124 gm, the oven was closed and allowed to attain the test temperature of 260°F. The average time required for the oven to attain this temperature after insertion of the sample was 50 seconds. The sample was then allowed to creep at this temperature for a period of 5 minutes. Thereafter, the platens, with the sample, were removed from the oven and allowed to cool to about 75°F. Cooling of the assembly was completed approximately 5 minutes after removal from the oven. The sample was then separated from the platens and measured for its physical dimensions. The thickness was measured at five different points, i.e., at the four quadrants near the periphery and in the center. The diameter of each sample was measured between three different pairs of points spaced about equally apart along the circumference of the disk. The hardness of the samples was measured with a Shore D durometer.

III. EXPERIMENTAL RESULTS

Prior to the irradiation, the samples were of cylindrical shape. Their average diameter was 0.550 in. and their average thickness, 0.137 in. On each of the five irradiation panels, 10 polyethylene disks were mounted on aluminum trays. Ten polyethylene disks were kept in the laboratory at 75°F as control specimens. The dosimeters mounted on the panels in circular arrangement yielded the dose values listed in Table II.

As described above, the irradiated as well as the control samples were each subjected singly to a constant load of 13,124 gm for a period of 5 minutes at 260°F. After removal from the oven and a cooling period of about 5 minutes, the thickness and the diameter of the extruded samples were gaged at 75°F. The dimensions thus measured are listed in Table III. Figure 1 is a photographic top view of the extruded samples arranged in groups of 5 per dose level (panel number).

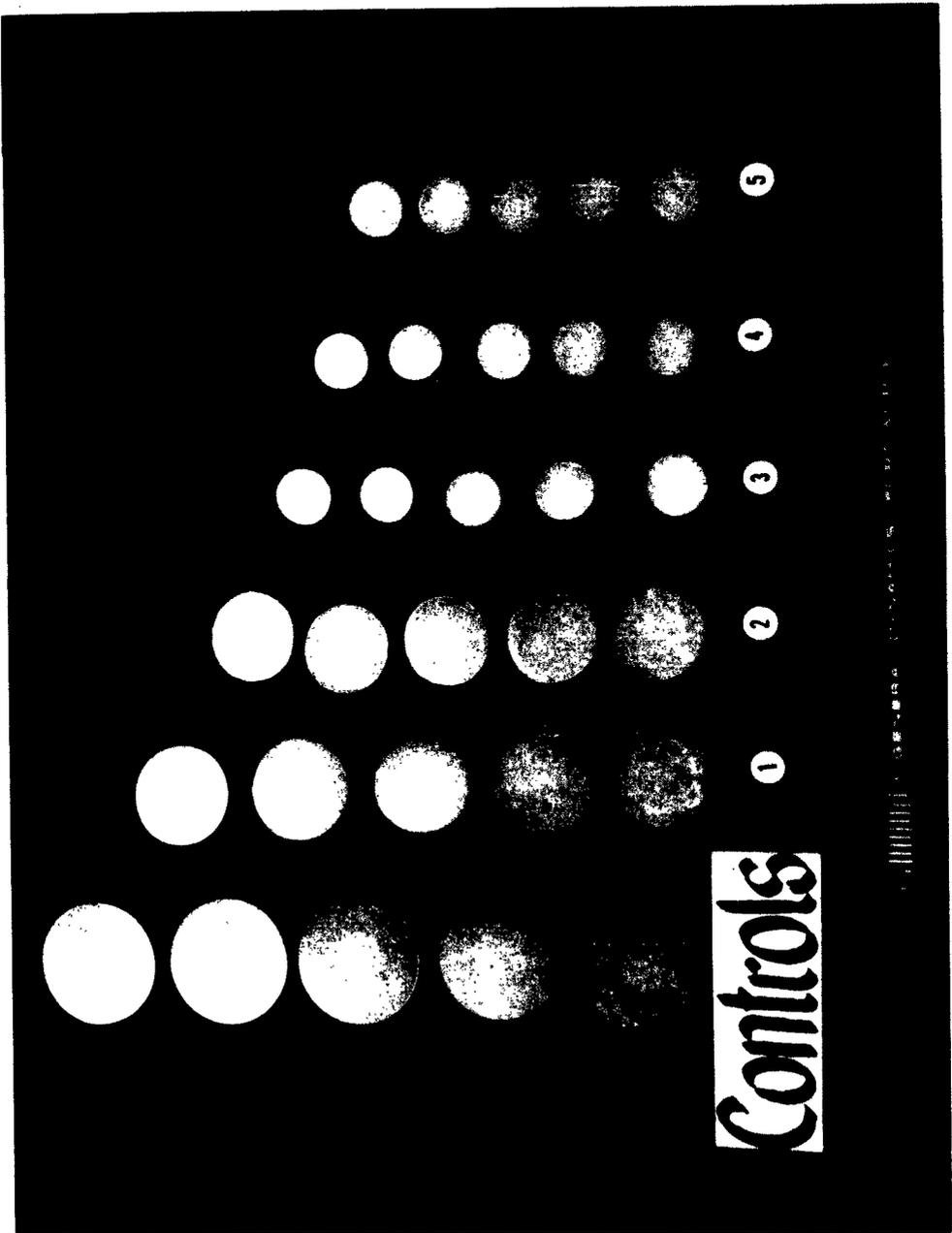


Figure 1. Irradiated Polyethylene Disks after Extrusion

Table II
Dosimetric Data of Polyethylene Irradiation

Integrated Neutron Flux [$n/cm^2 \times 10^{-12}$]			Absorbed Gamma-Ray Dose [$ergs/gm(C) \times 10^{-9}$]	
Aluminum Foil ($E > 8$ Mev)	Sulfur ($E = 2.9$ Mev)	Copper Foils bare, Cd-cov. (thermal neut)	Nitrous oxide	Tetrachloro- ethylene
Panel 1				
1.38	33.3	11.4	0.168	0.219
1.38	31.3	9.07	0.244	0.219
1.35	32.6	11.9	0.261	0.210
1.34	32.1	11.1	0.244	0.219
Average *	Average	Average	Combined Average	
1.36 ± 0.02	32.5 ± 0.66	10.7 ± 1.22	0.222 ± 0.030	

Panel 2

3.03	71.8	9.88	-----	0.455
3.20	76.7	18.3	-----	0.465
3.59	88.1	11.2	-----	0.517
3.21	73.4	8.75	-----	0.455
Average	Average	Average		Average
3.26 ± 0.237	77.6 ± 7.35	12.0 ± 4.30		0.473 ± 0.033

Panel 3

9.40	251	90.2	1.51	-----
9.29	262	97.7	1.89	-----
9.40	257	-----	1.58	-----
10.8	252	-----	1.60	-----
Average	Average	Average	Average	
9.72 ± 0.72	256 ± 5.5	94.0 ± 5.3	1.65 ± 0.17	

Panel 4

41.0	1230	82.1	5.08	-----
41.5	1230	61.6	4.91	-----
42.6	1380	62.6	5.08	-----
38.6	1410	109	4.91	-----
Average	Average	Average	Average	
40.9 ± 1.7	1310 ± 95	78.8 ± 22.2	5.00 ± 0.095	

Panel 5

106	3320	-----	13.8	-----
105	3470	-----	14.0	-----
106	3270	-----	12.0	-----
-----	3120	-----	-----	-----
Average	Average		Average	
106	3300 ± 145		13.2 ± 1.14	

* Average value and standard deviation, s

Table III
Dimensions of Extruded Polyethylene Samples*
(inches)

Measurement	Controls (D = 0)		D ₁ = 2.2 x 10 ⁸ erg/gm(C)		D ₂ = 4.72 x 10 ⁸ erg/gm(C)		D ₃ = 1.64 x 10 ⁹ erg/gm(C)		D ₄ = 4.99 x 10 ⁹ erg/gm(C)		D ₅ = 1.33 x 10 ¹⁰ erg/gm(C)	
	Thick- ness	Diam- eter	Thick- ness	Diam- eter	Thick- ness	Diam- eter	Thick- ness	Diam- eter	Thick- ness	Diam- eter	Thick- ness	Diam- eter
1	0.025	1.33	0.039	1.052	0.051	0.921	0.129	0.581	0.135	0.570	0.135	0.569
2	0.025	1.30	0.050	0.949	0.053	0.895	0.111	0.621	0.138	0.555	0.137	0.569
3	0.026	1.31	0.038	1.094	0.053	0.888	0.126	0.593	0.145	0.563	0.139	0.561
4	0.026	1.36	0.037	1.141	0.052	0.905	0.137	0.565	0.139	0.562	0.135	0.558
5	0.024	1.33	0.041	1.078	0.057	0.869	0.137	0.578	0.140	0.562	0.139	0.559
6	0.026	1.28	0.041	1.068	0.051	0.923	0.134	0.569	0.139	0.566	0.135	0.561
7	0.027	1.33	0.036	1.084	0.053	0.888	0.133	0.564	0.137	0.574	0.134	0.561
8	0.026	1.31	0.035	1.104	0.053	0.898	0.136	0.575	0.138	0.551	0.138	0.558
9	0.026	1.31	0.042	1.058	0.051	0.902	0.131	0.592	0.136	0.571	0.136	0.563
10	0.026	1.30	0.045	1.000	0.058	0.847	0.131	0.580	0.137	0.568	0.134	0.561
Average Values	0.026	1.32	0.041	1.063	0.053	0.894	0.131	0.583	0.138	0.564	0.136	0.562
Standard Deviation of the Samples	0.0026	0.0226	0.0046	0.0543	0.0025	0.023	0.0077	0.0161	0.0028	0.0073	0.0019	0.0040

* The ten individual values of thickness and diameter listed constitute the arithmetic mean of five different measurements of thickness and three different measurements of diameter on one and the same sample.

IV. DISCUSSION OF RESULTS

4.1 Dose-Thickness Correlation

If the extruded thickness of the polyethylene disks is plotted as a function of gamma-ray dose, it becomes evident that the most sensitive response of the hot-flow behavior to radiation takes place within a dose range of from 1×10^8 to 1.5×10^9 ergs/gm(C). It was found that the experimental data could be well fitted to the function

$$t_D = \frac{t_u t_o \exp \{v D\}}{t_u + t_o [\exp \{v D\} - 1]} \quad (1)$$

where t_D = final extruded thickness of disk irradiated to dose D gigaergs/gm(C) ,

t_o = final extruded thickness of unirradiated disk,

t_u = unextruded original thickness of disk, and

v = material parameter (in our case v was found to be 2.625)..

The experimental data and a plot of equation 1 are represented in Figure 2. Within the most sensitive region of response (0.1 to 1.5 gigaergs/gm(C)), the behavior is very excellently predicted by the simpler function

$$\Delta t = \frac{t_u - t_o}{1 + \alpha \exp \{-\beta D\}} \quad \text{for } D > 0 \quad (2)$$

where $\Delta t - t_D = t_o$ and,

α, β = constants (in the case $\alpha = 14$ and $\beta = 3.34$)

This equation, together with the appropriate experimental points, is plotted in Figure 3.

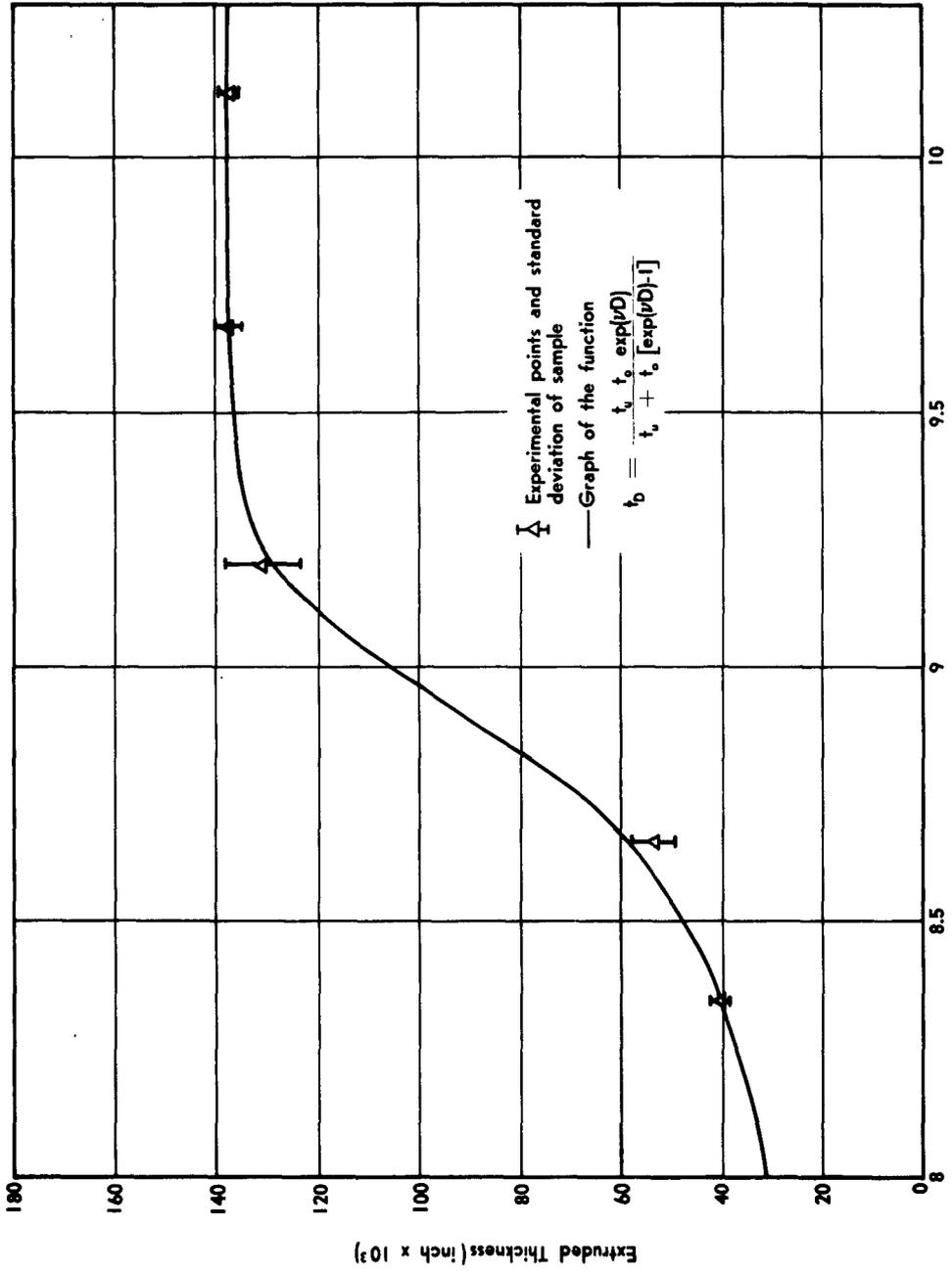


Figure 2. Extruded Thickness of Polyethylene Disks as a Function of Gamma-Ray Dose

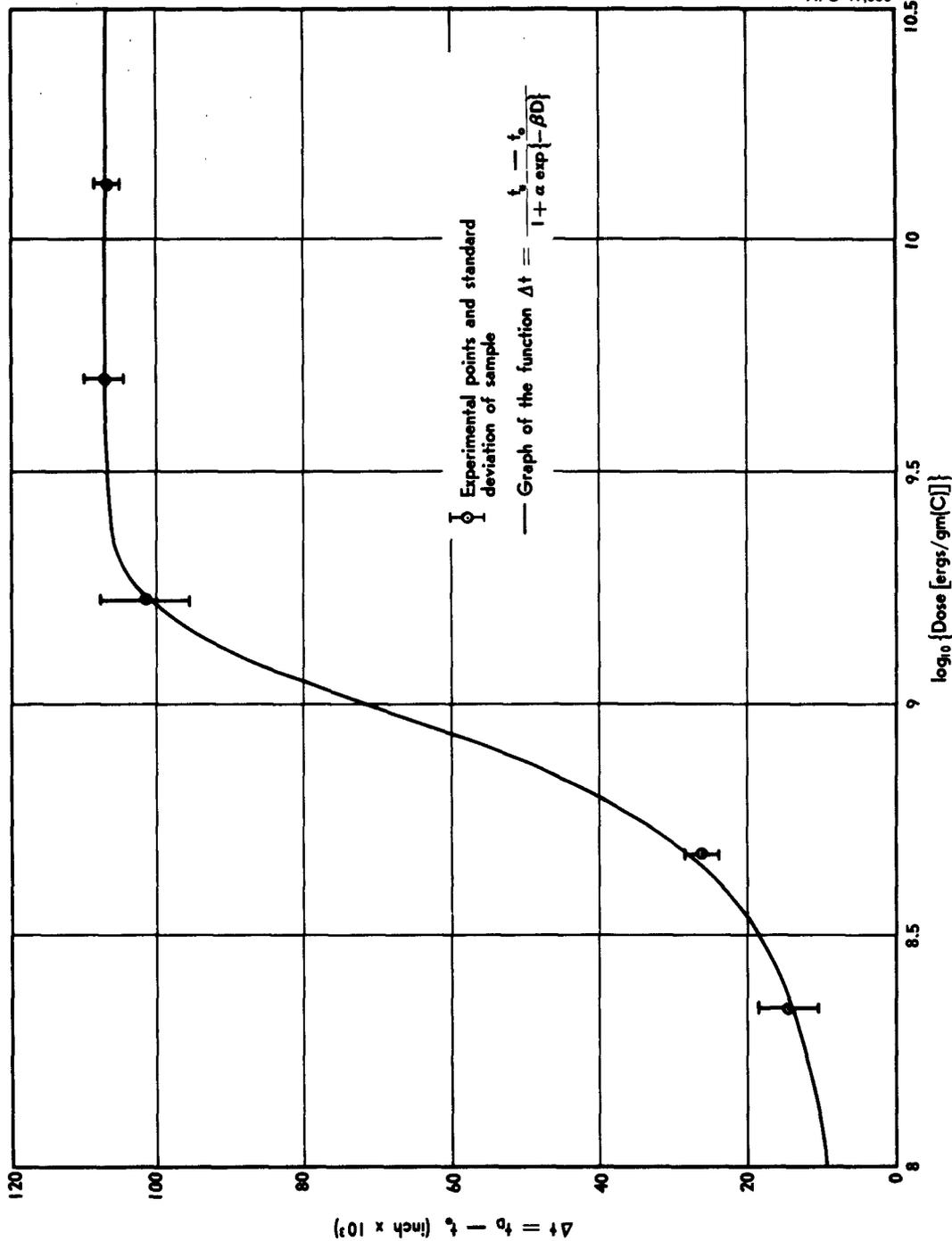


Figure 3. Increase in Extruded Thickness of Polyethylene Disks as a Function of Gamma-Ray Dose

4.2 Dose-Diameter Correlation

The average volume of the average sample disk as determined from the dimensional measurements was 0.0345 (± 0.0015) in³. If it is assumed then, on the basis of the standard deviation, that the volume of the sample disks remained the same, within the accuracy of the dimensional measurements, Equation 1 can be modified to express the extruded-disk diameter as a function of gamma-ray dose. Accordingly,

$$d_D = \left[\exp \left\{ -vD \right\} (d_o^2 - d_u^2) + d_u^2 \right]^{1/2} \quad (3)$$

where d_D = average diameter of the extruded disk after irradiation to dose D [gigaergs/gm(C)],

d_o = final average diameter of the extruded un-irradiated disk,

d_u = unextruded original diameter of disk, and

v = material parameter ($v = 2.625$).

Figure 4 is a plot of Equation 3 together with the observed diameter values of the extruded disks. However, it must be mentioned that, in our own case, the thickness measurements are more accurate than the diameter measurements. The lesser accuracy of the latter can be ascribed principally to two circumstances: (1) the extruded disks lost some of their circularity in the process of compression and (2) during the measurement of the diameter in the automatic gage, the thinner disks, particularly, tended to buckle slightly. Therefore, a precision ruler was used to determine the diametrical dimensions.

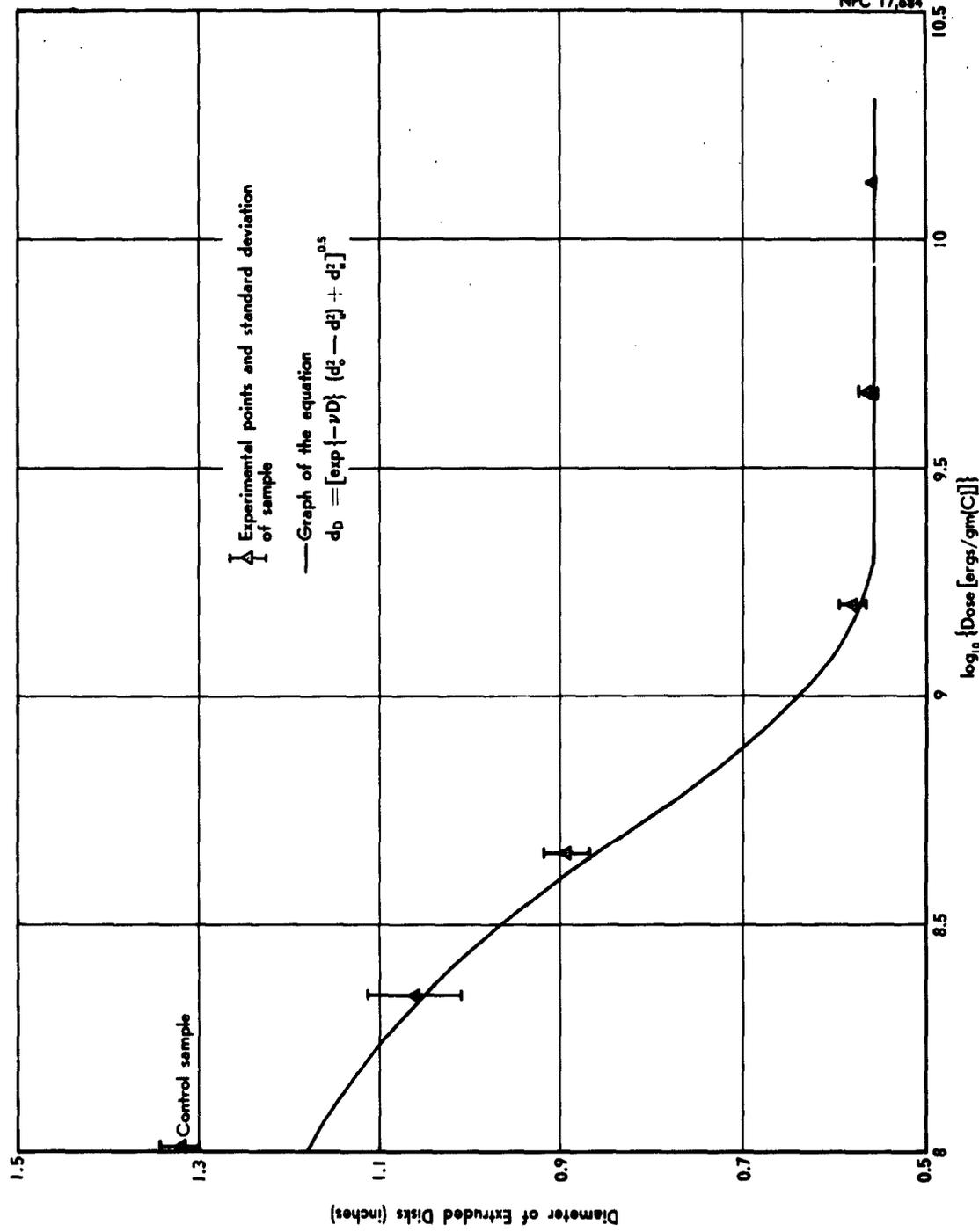


Figure 4. Diameter of Extruded Polyethylene Disks as a Function of Gamma-Ray Dose

4.3 Neutron Dose Calculations

In the foregoing calculations, only the dose recorded by the gamma-ray dosimeters was taken into account, while any contribution from the neutrons was neglected. Unfortunately, the problem of measuring the energy deposited by neutrons in materials is formidably complex and has not yet been resolved in any satisfactory sense. Therefore, any estimation, on the basis of the available flux data, of the neutron contribution to the chemically effective dose is quite conjectural.

However, for the purpose of relative evaluation, let us assume in our case that only the neutrons of energy $E > 2.9$ Mev make a significant contribution to the effective dose. Minder's calculations (Ref. 3) of neutron dose absorbed by H_2O as a function of neutron energy suggest that for neutrons of $E > 3$ Mev a value of 3.5×10^{-7} ergs/gm per n/cm^2 is reasonable. If we regard this value as an average approximate conversion factor for polyethylene [(and realizing that it is not based on carbon (C))] and apply it to our sulfur dosimetry data (see Table II), the quantities shown in Table IV are calculated.

According to these calculations, the average neutron contribution amounts only to about 6% of the total dose. However, on the assumption of normally distributed random errors in the gamma-ray dose measurements, the probable error in the present gamma-ray data is calculated to be 6.22%. Under these conditions, it would be expected to be quite difficult to even substantiate unambiguously the reality of a neutron contribution.

Table IV
Calculated Doses

Parameter	Panel 1	Panel 2	Panel 3	Panel 4	Panel 5
Measured integrated flux [(n/cm ² x 10 ¹² E 2.9 Mev)]	32.5	77.6	256	1310	3300
Calculated neutron dose D _n [ergs/gm x 10 ⁻⁷]	1.1375	2.716	8.96	45.85	115.5
Measured gamma dose D _γ [ergs/gm(C) x 10 ⁻⁷]	22.2	47.3	165	500	1320
D _γ /D _n ratio	19.51	17.4	18.14	10.9	11.42
Calculated total absorbed dose [ergs/gm x 10 ⁻⁹]	0.233	0.499	1.739	5.456	14.345
Percent contribution from neutrons to total dose	4.9	5.44	5.15	8.40	8.05

4.4 Dose-Viscosity Correlation

Dienes and Klemm (Ref. 2) developed a method for the measurement of the viscosity of high polymers in the range of from 10⁴ to 10⁹ poises by means of a parallel-plate viscosimeter. Accordingly,

$$h^{-4} = \frac{8\pi F}{3\eta V^2} \tau + K \quad (4)$$

where h = plate separation

F = applied force,

τ = time of application of force,

η = viscosity,

V = volume of sample, and

K = constant.

If we rearrange Equation 4 and assume that in our case $h = t_D$, we have

$$\eta = \frac{8\pi F\tau}{3V^2 (t_D^{-4} - K)} \quad (4a)$$

Since K must have the same dimensions as t_D^{-4} , it is not unreasonable for the purpose of a relative evaluation to set $K = t_u^{-4}$. Equation 4a then becomes

$$\eta = \frac{8\pi F\tau}{3V^2 (t_D^{-4} - t_u^{-4})} \quad \frac{\text{dyne-sec}}{\text{cm}^2}$$

In the present case, $F = 1.287 \times 10^7$ dynes, $\tau = 300$ sec, $V = 0.5653 \text{ cm}^3$ and $t_u = 0.3479$ cm. Hence, by substituting the values of t_D in the proper dose categories given in Table III we obtain the viscosity values shown in Table V for the various polyethylene samples.

Table V
Viscosity Values
(poise)

Dose Level [ergs/gm(C)]				
$D_0 (0)$	$D_1(2.2 \times 10^8)$	$D_2(4.7 \times 10^8)$	$D_3(1.64 \times 10^9)$	$D_4(4.99 \times 10^9)$
1.925×10^6	1.194×10^7	3.396×10^7	7.595×10^9	∞

In Figure 5, these values of viscosity are plotted as a function of gamma-ray dose. On this basis, one can easily see that the region of drastic change in viscosity is coincident with the most sensitive region of response in the hot-flow characteristics $[10^8 - 1.5 \times 10^9 \text{ ergs/gm(C)}]$.

4.5 Dose-Hardness Correlation

The post-extrusion Shore D hardness values of the samples do not markedly vary with dose. The values given in Table VI represent the arithmetic mean of 20 measurements for each set of samples.

Table VI
Shore D Hardness Values

Control Unextruded	After Extrusion					
	Control	Dose 1	Dose 2	Dose 3	Dose 4	Dose 5
44.6	37.8	43.1	45.3	46.1	46.3	45.7

The greatest difference exists between the hardness values of the unextruded and extruded control samples. This is doubtless due to the rapid cooling of the extruded disks with more amorphous regions being frozen in than are present in the original material.

4.6 Adhesion and Discoloration

Finally, it may be remarked as a matter of parenthetical interest that the post-extrusion adhesion of the samples to the platens seemed to increase with dose up to $4.72 \times 10^8 \text{ ergs/gm(C)}$

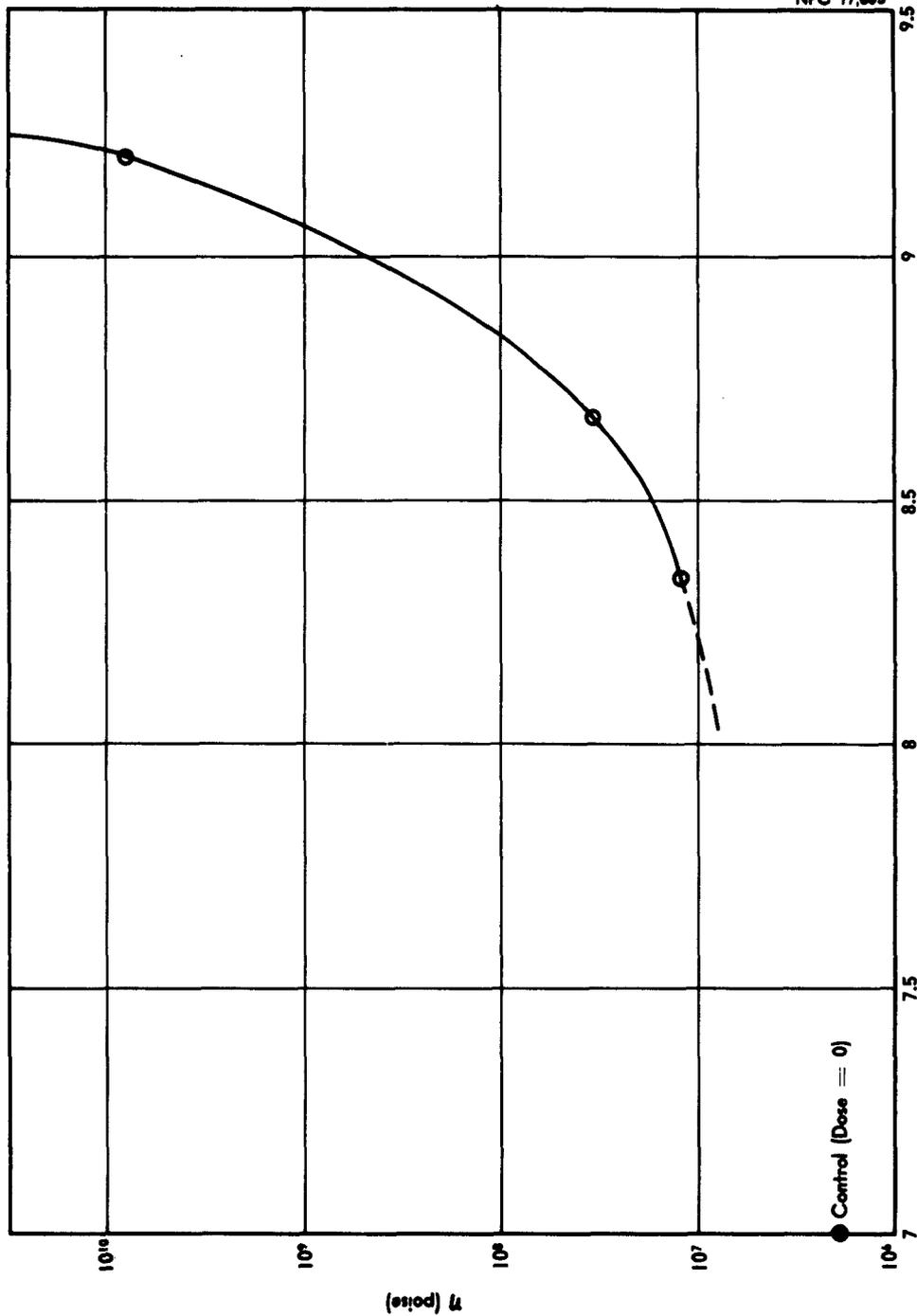


Figure 5. Viscosity of Irradiated Polyethylene Disks as a Function of Gamma-Ray Dose

but, beyond that, it fell off notably and almost disappeared completely at 1.33×10^{10} ergs/gm(C). The radiation-induced yellowish discoloration reported by many previous investigators was observed faintly in the samples exposed to 4.99×10^9 ergs/gm(C) and markedly so in the samples exposed to 1.33×10^{10} ergs/gm(C).

V. CONCLUSIONS

The data indicate that the hot-flow characteristics of irradiated polyethylene are a sensitive function of dose within the range of from 1×10^8 to 1.5×10^9 ergs/gm(C). It was shown that this range also corresponds to the region of greatest viscosity change. The results suggest that, within this sensitive range, the dimensional changes of irradiated polyethylene samples after hot compressive extrusion can be quite accurately correlated with the absorbed gamma-ray dose and that, therefore, the described experimental procedure may conceivably be used as a dosimetric method.

REFERENCES

1. Charlesby, A., Atomic Radiation and Polymers. New York: Pergamon Press (1960), p. 206.
2. Raff, R. A. V., and Allison, J. B., Polyethylene. New York: Interscience Publishers, Incorporated (1956) p. 314.
3. Minder, W., "Dosimetrie." Chemische Reaktionen ionisierender Strahlen, edited by H. Mohler. Aarau (Switzerland): Sauerländer and Company (1958), pp. 108-166.

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