

Utilization of the Changes in Mechanical Properties as Dosimeter Due to Gamma Irradiation of High-Density Polyethylene (HDPE)

Abdelnabi Ali Elamin^{1,*}, Elfatih Ahmed Hassan², Abdelrahman Elhassan Mohamed–Osman³, Mubarak Dirar Abd-Alla³, Mohamed Habib Ahmed Elkanzi⁴

¹Department of Physics, Faculty of Science and Technology, Omdurman Islamic University, Omdurman, Sudan

²Department of Chemistry, College of Science., Sudan University of Science & Technology, Khartoum, Sudan

³Department of Physics, College of Science., Sudan University of Science & Technology, Khartoum, Sudan

⁴Department of Astronomy and Meteorology, Faculty of Science and Technology Omdurman Islamic University, Omdurman, Sudan

Email address:

aelamain@yahoo.com (A. A. Elamin)

*Corresponding author

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Abstract: Polymer materials are required to be used in nuclear power stations as insulation materials of electrical wire and cable. For the use of polymer materials in such regions, the electrical performance under different environments of radiation is different from that without radiation has been found through previous research. The aim of this paper is to use the change in mechanical properties with radiation dose to design polymer dosimeter. The experimental results show an improvement of Black PE mechanical properties such as distension, tensile strength and elongation at break as dose increases, indicating the predominance of cross-linking over oxidative degradation. The predominance of cross-linking results show linear relations between distension, tensile strength and elongation at break versus absorbed dose of gamma rays source. These relationships between these properties and dose level can be utilized as measuring tool (Dosimeter) for radiation exposure that incident on the HDPE samples.

Keywords: Dosimeter, Cross-Linking, Distension

1. Introduction

Among the most important and versatile of the hundreds of commercial plastics is polyethylene (PE). Polyethylene contains the chemical elements carbon and hydrogen, and produced through polymerization of ethylene. Polyethylene is used in a wide variety of applications because, based on its structure, it can be produced in many different forms. The first type to be commercially exploited was called low density polyethylene (LDPE) or branched polyethylene. This polymer is characterized by a large degree of branching, forcing the molecules to be packed rather loosely forming a low density material. LDPE is soft and pliable and has applications ranging from plastic bags, containers, textiles, and electrical insulation, to coatings for

packaging materials, Chanda et al (2006). It contains 2.25% well dispersed carbon black in order to ensure excellent weathering resistance. Antioxidants and processing aids are added to obtain excellent long term properties and a better process ability.

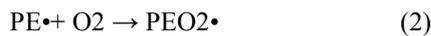
Radiation effects on polymer materials may be cross-linking or scission of polymer chains. The interaction radiations with polymer materials depend on the type of radiation, radiation dose and types of polymeric materials. The cross-linking is the intermolecular bond formation between polymer chains. The degree of cross-linking is proportional to the radiation dose. However, radiation cross-linking is only limited to few applications such as cross – linking of rubber for tyres, cables, pipes and heat shrinkable rubber (improvement of polymer). In contrast, scission

chains is opposite of cross-linking action as, in which the rupturing of C-C bond takes place (degradation of polymer), (Phil Hyun Kang†, et al. 2006). A discussion of the effect of radiation on mechanical strength as a consequence of the balance between cross-linking and oxidative degradation is presented in many publications.

Wüdrich (1985) presents a review of the literature on values for mechanical resistance parameters for several plastics and elastomers submitted to radiation under different conditions. Instead of directly presenting the parameter values, the radiation effects were compared using the half-dose value concept, defined as the absorbed dose necessary to reduce the parameter value to 50% of the initial value. According to the author, increases in parameter values were observed, mainly in casesn involving high doses, but these effects were not relevant in practical terms. An analysis of the above-mentioned data shows that, when irradiation is carried out in the absence of oxygen (vacuum or inert atmosphere), the effect of radiation on the mechanical resistance parameters is independent of dose rate. On the other hand, in the presence of air, the relationship between effect and dose rate is evident. For example, for high-density polyethylene, the half-dose value for ultimate tensile stress is reduced gradually with the reduction in dose rate. In the absence of air, this dose is larger than the values for irradiation in the presence of air. Wüdrich attributes this behavior to the influence of oxygen in the degradation of polymers (oxidative degradation). This effect increases with the reduction in dose rate because it is a time-related process due to two mechanisms: the diffusion of oxygen in the polymer and the disintegration reaction of the peroxides formed. Singh (1999) presents mechanisms that corroborate these comments. According to the author, two phenomena occur as a consequence of the HDPE gamma irradiation process: the formation of cross-linking and oxidative degradation. In general, Eq. (1) shows the reaction between free radicals (PE •) of HDPE, produced by the loss of a hydrogen atom of HDPE (represented as PE), forming cross-linking between polymer molecules. This recombination reaction is predominant in cases of irradiation in vacuum or inert atmospheres.



During irradiation in the presence of air, the formation of peroxide radical (Eq. (2)) and the reaction of the PE free radical and the peroxide radical (Eq. (3)) predominates.



Singh (1999) also discusses that at very high dose rates (10³ Gy/s, generated in electron accelerators), the formation of cross-linking (Eq. (1)) is the predominant reaction. This happens because the oxygen is quickly consumed and the formation of peroxide radical (Eq. (2)) becomes limited to the oxygen diffusion rate.

The complexity of the phenomena resulting from HDPE

irradiation is also discussed by Premnath et al. (1999). According to the authors, specific effects of polyethylene irradiation may differ depending on factors such as polymer molecular weight; the presence of additives; temperature; storage under atmospheric conditions before, during and after irradiation; and size of the samples, among others Based on these literature review conclusions, experiments were designed in order to study the influence of gamma irradiation dose and dose on mechanical strength parameters. This paper presents experimental results of the irradiation in air with a Cobalt-60 gamma source of high-density polyethylene (HDPE) samples to address the radiation effect on compression and tensile parameters and also to find the relationship between doses of gamma source and mechanical properties which utilized as dosimeter.

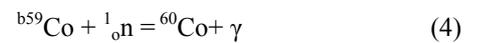
2. Experimental Work

2.1. Samples Preparing

The black PE samples were prepared as sheets with thickness ranging between 2 to 4 mm. The black PE samples contain 2.25% well dispersed carbon black in order to ensure excellent weathering resistance. Antioxidants and processing aids are added to obtain excellent long term properties and a better process ability, with a density 951 Kg/m³, Melt Flow Rate 190°C\2.16 Kg in range at g/10min due to ISO1133 and thermal Stability is 210°C at ≥ 15 min due to EN 728.

2.2. Samples Irradiation

The samples above which were irradiated by different doses (10,30,50,75,108 KGY) of gamma rays emitted from⁶⁰Co with an activity of (37GBq) in room temperature, the dosimetry calibration is using reference standard dosimeter Fricke (Fricke is chemical dosimeter consists of ferrous ammonium sulphate, sulphuric acid and sodium chloride.) and lifetime of (2.27years) it is produced by the following reaction:



⁶⁰Co is primarily a β⁻ emitter (0.311Mev), which decays into an excited state of ⁶⁰Ni. The excited nucleus of ⁶⁰Ni gives up its excess energy by emitting two gamma photons in cascade with energies of (1.17) and (1.33) Mev as shown in fig 1.; the medium energy being (1.25Mev). The source of ⁶⁰Co is housed in protective container called the treatment heat, which reduces the exposure rate of the radiation to a low level everywhere outside the useful beam. The treatment heat also contains a shutter for "shutting off" the useful beam and an adjustable collimator to control the size and shape of the beam. The importance of ⁶⁰Co source is obtain gamma rays with energy of (1.25Mev).

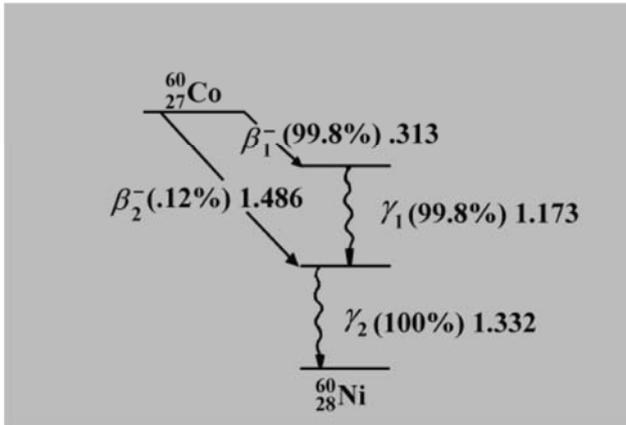


Figure 1. Decay scheme of radionuclide cobalt-60.

2.3. Determination of Distension

2.3.1. Equipment Used

Lastometer - I G\ Lst\ D - Giuliani Company, Torino, Italy. Sr. no 13812- (2003), in National Leather Development Center (Khartoum - Sudan).

2.3.2. Theory

The determination of distension and strength of grains of any polymer sample may conveniently be carried out with the help of the ball burst test. In this test, a hemispherical hardened metallic tip is pressed against one face of the polymer disc held in a suitable non-slipping grip and the force needed to make the tip pierce through the test specimen need to be determined to evaluate the grain strength. The distension of at which the tip pierces through should also be determined. The apparatus used for this evaluation is called Lastometer.

The distension shall be taken as the distance through which the clamp moves relative to the ball in a direction normal to the plane occupied by polymer when the disc is clamped and is under zero loads

2.3.3. Distension Test Procedure

1. The conditioned specimen was clamped in the instrument with its "flesh" surface adjacent to the ball, and its "grain" surface flat.
2. The distension was increased at a rate of approximately one-fifth of a millimeter per second, and the "grain" surface was watched for the occurrence of a crack in the "grain". When grain crack occurs note the load and distension and continue the loading with as little as possible. If the disc bursts before the maximum load of the instrument was reached, note the load and distension at burst.
3. The report of any test should state the load and distension at grain crack, and the corresponding values at burst if the polymer disc burst before the maximum load was reached. If a number of tests were made, report the results of each and not merely their arithmetic means. If the sample is known to be other than full grain polymer, state this.

2.4. Determination of Tensile Strength

2.4.1. Equipment Use

1. Universal Tester Machine - India, model Te Vs 200 – Sr no 1378.1.1.
2. IG/DE/S. Digital Dynamometer, ser No 14546 / (2007), Giuliani/ Torino- Italy.

2.4.2. Theory

Tensile strength is important for a material that is going to be stretched or under tension. Then, using the test machine, continue to increase the amount of force, and stress naturally, on the sample until it breaks. The stress needed to break the sample is the tensile strength of the material.

Likewise, one can imagine similar tests for compression or flexural strength. In all cases, the strength is the stress needed to break the sample.

Since tensile stress is the force placed on the sample divided by the cross-sectional area of the sample, tensile stress, and tensile strength as well, are both measured in units of force divided by units of area, usually N/cm^2 . Stress and strength can also be measured in mega Pascal (MPa) or giga pascals (GPa). It's easy to convert between the different units, because $1 \text{ MPa} = 100 \text{ N}/\text{cm}^2$, $1 \text{ GPa} = 100,000 \text{ N}/\text{cm}^2$, and of course $1 \text{ GPa} = 1,000 \text{ MPa}$.

Other times, stress and strength are measured in the old English units of pounds per square inch, or psi. If you ever have to convert psi to N/cm^2 , the conversion factor is $1 \text{ N}/\text{cm}^2 = 1.45 \text{ psi}$.

2.4.3. Tensile Strength Test Procedure

The samples were prepared according to standard ASTM D638 The most common specimen for ISO 527 is the ISO 3167 Type1. Multipurpose specimen. ASTM D882 uses strips cut from thin sheet or film. Specimens are placed in the grips of the universal tester at a specified grip separation and pulled until failure. For ASTM D638 the test speed is determined by the material specification. For ISO 527 the test speed is typically 5 or 50mm/min for measuring strength and elongation and 1mm/min for measuring modulus.

2.5. Determination of Elongation at Break

2.5.1. Equipment Use

1. Universal Tester Machine - India, model Te Vs 200 – Sr. no 1378.1.1.
2. IG/DE/S. Digital Dynamometer, ser No 14546 /(2007), Giuliani/ Torino- Italy.

2.5.2. Theory

It is important to study polymer's mechanical properties than merely knowing how strong it is. All strength tells us is how much stress is needed to break something. It doesn't tell us anything about what happens to our sample while we're trying to break it. That's where it pays to study the elongation behavior of a polymer sample. Elongation is a type of deformation. Deformation is simply a change in shape that anything undergoes under stress. When we're talking about tensile stress, the sample deforms by stretching, becoming

longer. We call this elongation, of course.

Usually we talk about percent elongation, which is just the length the polymer sample is after it is stretched (L), divided by the original length of the sample (L_0), and then multiplied by 100. There are a number of things we measure related to elongation. Which is most important depends on the type of material one is studying. Two important things we measure are ultimate elongation and elastic elongation.

Ultimate elongation is important for any kind of material. It is nothing more than the amount you can stretch the sample before it breaks. Elastic elongation is the percent elongation you can reach without permanently deforming your sample. That is, how much can you stretch it, and still have the sample snap back to its original length once you release the stress on it. This is important if your material is an elastomeric. Elastomers have to be able to stretch a long distance and still bounce back. Most of them can stretch from 500 to 1000 % elongation and return to their original lengths without any trouble.

2.5.3. The Elongation at Break Test Procedure

The most common specimen for ASTM D638 is a Type I

Table 1. The mechanical properties of the samples before and after irradiated by using different doses of gamma rays of 1.25MeV emitted from ⁶⁰CO source.

Total dose (KGy)	0	5	10	15	20	25	35	40	50	60	65	70	75	90	108
The Distension	9	?	9.5	?	?	9.8	?	?	12	?	?	?	13	?	14
The Tensile Strength (Kg/cm ²)	176	?	176.8	?	?	197.6	?	?	199	?	?	?	199.8	?	205
The Elongation at break %	10	?	11	?	?	15	?	?	19	?	?	?	22	?	27

Suppose $F(x)$ is a function whose value at certain points x_0, x_1, \dots, x_n such as (0,10,25,50,75 and 108)KGy (total dose) are known. The values are $f(x_0), f(x_1), \dots, f(x_n)$. Consider a point x different from x_0, x_1, \dots, x_n . $F(5), F(15), F(20), F(35), F(40), F(45), F(60), F(65), F(70)$ and $F(90)$ are not known from for the values of total dose irradiation. We can find an approximate value of $F(x)$ from the known values. This method of finding $F(x)$ from these known values is called interpolation. We say that we interpolate $F(x)$ from $f(0), f(10), f(25), f(50), f(75)$ and $f(108)$. The Linear Interpolation use two points x_0, x_1 and f_0, f_1 be the function values at these two points respectively. Let x be a point between x_0 and x_1 . We are interested in interpolating $F(x)$ from the values $F(x_0)$ and $F(x_1)$. Generally, the linear interpolation formula may be written as:

$$F(x) = (1 - p)f_0 + pf_1 \tag{5}$$

This is called the linear interpolation formula. Since x is a

Table 2. Changes in mechanical properties due to gamma irradiation of black PE before interpolation.

Total dose (KGy)	0	10	25	50	75	108
The Distension	9	9.5	9.8	12	13	14
The Tensile Strength (Kg/cm ²)	176	176.8	197.6	199	199.8	205
The Elongation at break %	10	11	15	19	22	27

Table 3. Interpolating mechanical properties as function of total irradiation dose.

Total dose (KGy)	5	15	20	30	35	40	60	70	90	100
The Distension	9.25	9.6	9.7	10.24	10.68	11.12	12.4	12.8	13.45	13-76
The Tensile Strength (Kg/cm ²)	176.4	183.6	187.2	197.88	198.16	198.44	199.32	199.64	202.14	203.75
The Elongation at break %	10.5	12.32	13	15.8	16.6	17.4	20.2	21.4	24.25	25.8

tensile bar. The most common specimen for ISO 527 is the ISO 3167 Type1 A multipurpose specimen. ASTM D882 uses strips cut from thin sheet or film. Specimens are placed in the grips of the universal tester at a specified grip separation and pulled until failure. For ASTM D638 the test speed is determined by the material specification. For ISO 527 the test speed is typically 5 or 50mm/min for measuring strength and elongation and 1mm/min for measuring modulus. An extensometer is used to determine elongation at break in National Leather Development Center (Khartoum - Sudan).

2.6. Interpolating the Mechanical Properties as Function of Total Doses

The change in mechanical properties of black PE samples due to irradiation by different doses (0, 10, 25, 50, 75 and 108 KGY) at room temperature, which was shown fig 2 are not enough points to find the relationship between mechanical properties and total doses, which that to be utilized as dosimeter.

point between x_0 and x_1 , p is a nonnegative fractional value is $x-x_0/x_1-x_0$, ANCONA, M. G. (2002).

3. Results and Discussion

Different mechanical properties of a given insulator or dielectric material can respond differently to radiation. In general, the influence of radiation on these properties of a particular dielectric constant depends greatly on the chemical transformations that take place within it or its surroundings. For polymeric materials, in particular, cross linking and chain scission are believed to play an influential role on the rate of deterioration of these dielectrics.

Mechanical properties of Black PE which change by radiation are most useful in the mechanical aspect of application of dielectric polymer. These properties include distension, tensile strength and elongation at break as shown table 2 before interpolation and table 3 after interpolation.

In view of Fig (2) it is clear that distension for black HDPE samples increases when the total irradiation dose of gamma rays increases, for HDPE samples which are exposed gamma rays, this increase may be attributed to the cross linking process which is described by equation (1). In this process the gamma radiation causes free radicals in the CH chain which causes chemical reaction which leads to

formation of new large CH bond. Thus gamma rays change the bond and molecular weight from light to heavy molecular weight. The fact that the distension increases when the molecular weight increases is in conformity with common sense. This is since the molecular weight increase increases the density which increases the distension.

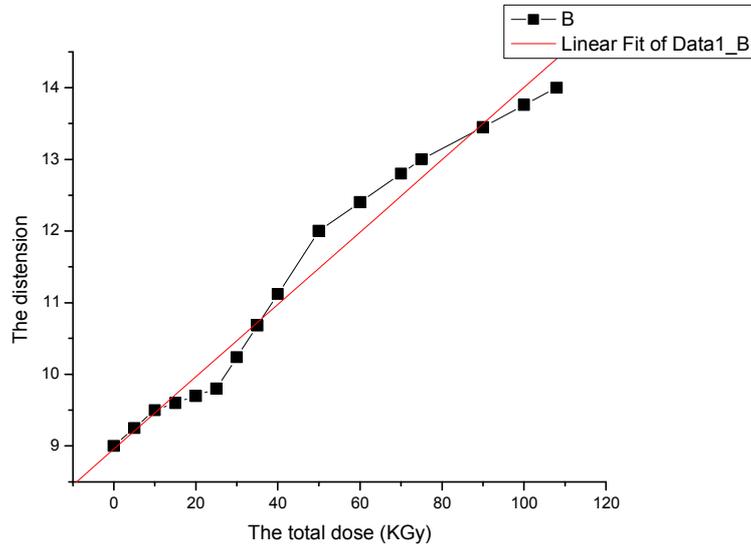


Figure. 2. The relationship between Distension and total dose irradiation of gamma rays for black PE.

The relation between distension and dose take the form

$$Y_1 = 8.957 + 0.0505x \tag{6}$$

Where y_1 is the Distension for Black PE and x is the total dose of irradiation in KG. Thus it is clear that the effect of radiation on the Distension property depends on the type of

radiation, type of interaction and chemical composition of the material.

In view of Figure. (3) it is clear that tensile strength increases when the total irradiation dose of gamma rays increases for black PE samples. This behavior has been observed also by Patel et al (2006).

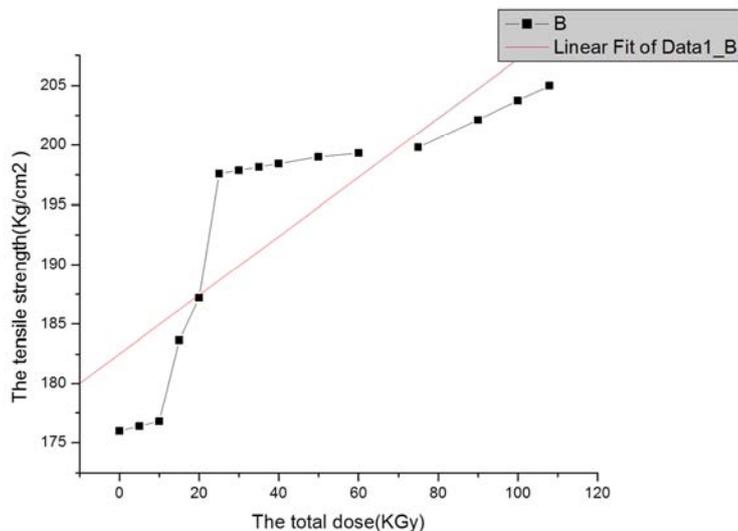


Figure. 3. The relationship between tensile strength and total dose irradiation of gamma rays for black PE.

By figure 3 shows relation between tensile strength and absorbed dose of gamma rays source as follows: -

$$Y_2 = 182.492 + 0.247x \tag{7}$$

where y_2 the tensile strength in Kg. cm^{-2} for Black PE and x is the total dose of irradiation in KGy.

In view of Fig. (4) It is clear that the elongation at break increase for HDPE samples when the dose of irradiation of

gamma rays increases. In this study, the elongation at break increases by >10 for this samples and more than 50% increment after 108 KGY.

For black HDPE samples which exposed to gamma rays, these increases may be attributed to the cross-linking process.

This result has been obtained in opposite direction by Mark W. et al 2005, because they were used difference polymer and difference dose (exposure silicone rubber by gamma rays).

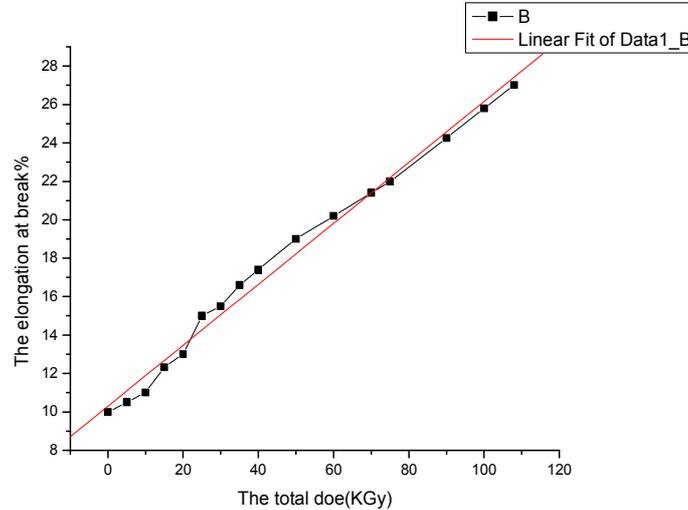


Figure 4. The relationship between elongation at break and total dose irradiation of gamma rays for black PE.

Figure 4, (using the Origin6.1 program), shows the relation between the elongation at break and absorbed dose of gamma rays source follows: -

$$Y_3 = 10.29327 + 0.15863x \quad (8)$$

Where Y_3 is the elongation at break for, Black PE, and x is the total dose of irradiation for Gamma rays.

4. Conclusions

Analysis of the changes in the mechanical properties of the HDPE submitted to gamma irradiation shows that:-

- (i) The Distension, tensile strength and elongation at break of Black PE samples increases when the absorbed dose of Gamma radiation increases depend on the types of interaction cross linking and chain scission.
- (ii) The depending of mechanical properties on gamma dose shown on equations (6.), (7), and (8) for distension, tensile strength and Elongation at break respectively can be used to measure the absorbed dose of Gamma radiation emitted by sources.

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