Engineering Journal of University of Qatar , Vol. 10, 1997, p. 133 - 142

ULTRASONIC CHARACTERIZATION, TENSILE TESTING AND THERMAL PROPERTIES OF γ IRRADIATED LOW DENSITY POLYETHYLENE, FB 5005

Zuhair Mohamed El-Qassem* and Mohamed El-Said Kassem**
* Associate Professor, Mechanical Eng. Dept., Faculty of Engineering
** Professor, Physics Department, Faculty of Science
University of Qatar, Doha, Qatar.

ABSTRACT

Ultrasonic pulse-echo technique, tensile testing and thermal gravimetric analysis have been utilised to characterise and monitor the effect of γ irradiation on the physicomechanical properties of low density polyethylene LDPE, grade FB 5005. The sonic modulus, elastic modulus, rupture stress and decomposition temperature exhibit a minimum value at 2 Mrad irradiated samples, whereas, toughness and rupture strain exhibit a maximum at the same irradiation dose. The changes in mechanical properties (rupture stress and toughness) correlate well with decomposition temperature and the change in the sonic modulus correlates as well with elastic modulus for the tested material. The results are discussed in terms of cross-linking. Below 2 Mrad the loss of crystallinity overrides the effect of cross-linking, whereas above 2 Mrad the cross-linking dominates.

Keywords: Sonic modulus, Cross-linking, γ-Irradiated LDPE, Decomposition temperature, Crystallinity, Toughness

INTRODUCTION

It is well known that radiation interacts with polymer material to cause ionisation, leaving some molecules in a highly excited state. The excitation energy may become localised in a particular chemical bond causing it to break. This could produce unsaturated polymer chain as in the case of polyethylene (PE) when hydrogen gas is formed(1). Breaking the bond may occur on adjacent molecules with the important effect of cross-linking the chains (2).

Cross-linking of PE is sometimes commercially desired when it permits higher thermal stability and higher tensile strength (3). However, cross-linking is not always beneficial to the material and may result in deterioration of some properties through destruction and degradation of the macro molecules (4,5). In this work the effect of γ radiation on the acoustical, mechanical and thermal properties of the low density polyethylene (LDPE) has been investigated. In addition, the results may be used to find some correlation between the parameters obtained.

This work would help in understanding how to improve stability of the material both during processing and subsequently during environmental exposure. Beside this, the results may be directed toward decomposition of PE following exposure to ionising radiation which would facilitate its disposal as the accumulation of waste polymeric material is a major component of environmental pollution (6) and the PE represents the largest volume of the plastic used (7).

It should be mentioned that the attenuation of elastic waves at the ultrasonic frequencies has been used in this work for the determination of sonic modulus (G) which is indicative for mechanical properties of crystalline polymers (8). It is known also that sonic pulse in the MHz range causes real displacements of the molecules from their equilibrium positions as it passes through the specimens (9). Thus, the velocity of the wave in the material will depend on its density and stiffness (10), which may be expressed as

$$C_{ii} = \rho \, V_{ii}^2 \qquad (1)$$

Where C_{ii} is the stiffness constant in a particular direction; ρ is the density of the material; and V_{ii} is the velocity of a particular wave propagating in a particular direction.

For an isotropic sample, the sonic modulus may be given by the following equation (10):

$$E = [C_{11}(C_{11} + C_{12}) - 2C_{12}^2] / (C_{11} + C_{12}) \qquad (2)$$

The unknown C_{12} for isotropic bodies may be obtained from the following relation (10):

$$\upsilon_{12} = C_{12}(C_{11} - C_{12}) / (C_{11}^2 - C_{12}^2) \qquad (3)$$

where υ_{12} is the in-plane Poisson's ratio, which for the investigated LDPE may be taken as 0.315 (11,12).

EXPERIMENTAL

The material used in this work was LDPE locally produced by Qatar Petrochemical Company (QAPCO), FB005 with the properties as listed in Table 1.

Table 1. Properties of Lotrene FB 5005 LDPE as Supplied by QAPCO

Polymer Properties	Value	Unit
Melt flow index	0.5 - 0.7	g/10 min.
Density	0.919 - 0.921	g/cm^3
Crystalline melting point	111	°C
Tensile strength at break	24	MPa

LDPE pellets were compression-moulded into plate with approximately 3 mm thickness in a compression moulding machine at 30 MPa and 165°C for 20 minutes. Square samples (25 mm x 25 mm) were cut from the plates for ultrasonic measurements to obtain the sonic modulus. In addition, tensile samples were also cut from the plates with a gauge length of 25 mm and a width of 10 mm.

Both sets of prepared samples were γ irradiated in air to different doses upto 100 Mrad at room temperature using a CO gamma cell.

Ultrasonic measurements were performed on the square, γ irradiated samples using Kraut Kramer equipment model USL-32. All the measurements were done at a frequency of 4 MHz. The velocity of sound propagation, V_S (mm/s) through the specimen can be determined from the equation:

$$V_s = 2D / T \tag{4}$$

where D is the length of test specimen in the direction of measurement in mm and T is the transit time in sec. It is possible now to calculate the stiffness constant mentioned in equation (1) from the relation:

$$C_{11} = \rho \ (V_s)^2 \tag{5}$$

The density, ρ. was assumed to be constant (= 0.92 g/cm^3) and C_{12} was obtained using equation (3).

Tensile testing was carried out using Lloyd Instruments materials testing machine linked to a remote microcomputer for data acquisition and analysis. The load was measured by a load cell 5 kN capacity, while the displacement was measured using an internal extensiometer. The speed of testing was 100 mm/min. Lloyd Data Analysis Package (DAP) was used to analyse the tensile properties from the load-extensions diagrams. The following properties were determined: rupture stress, rupture strain, modulus of elasticity and toughness which has been taken as the area under the stress-strain curve.

The thermal properties of the fabricated, γ irradiated and tensile tested samples were measured in N_2 atmosphere at heating rate of 5 K/min. The method of measurements have been described elsewhere (12).

RESULTS AND DISCUSSION

The sonic modulus (G) as obtained from equation (1). The decomposition temperature T_d as obtained from the thermal tests versus the irradiation dose are shown in Fig. 1. It is clear from the figure that both variables G and T_d are sensitive to the irradiation dose. They both decrease rapidly with increasing dose until a minimum value is reached at 2 Mrad, which may be referred to as "first stage". This is followed by a "second stage" where both variables increase until approximately 100 Mrad.

Such sensitivity of the material is also detected for the mechanical properties measurements in the present work, as can be seen in Figs. 2 and 3. Fig. 2 shows the typical stress-strain curves for the tested material under different γ radiation conditions and Fig. 3 shows the rupture stress, rupture strain, elastic modulus and toughness (area under stress-strain curve) versus the irradiation dose. It is visible from Fig. 3 that increasing the irradiation dose would lead to a rapid decrease in elastic modulus and rupture stress (stress at break) during the first stage, then both properties increased steadily until 100 Mrad in the second stage. Similarly, toughness and rupture strain (ductility or elongation at break) showed rapid increase upto 2 Mrad followed by steadily decrease in the second stage.

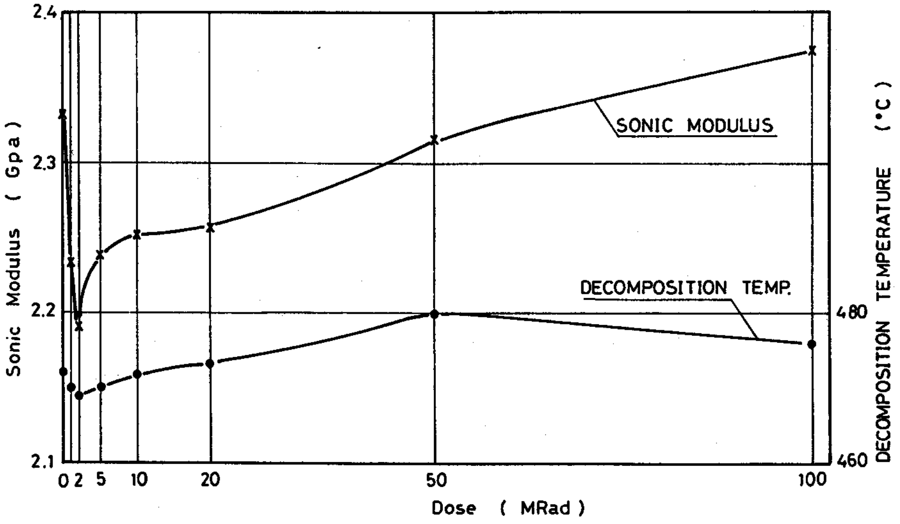

Fig. 1. **Effect of γ radiation doses on the sonic modulus and decomposition temperature for the tested material**

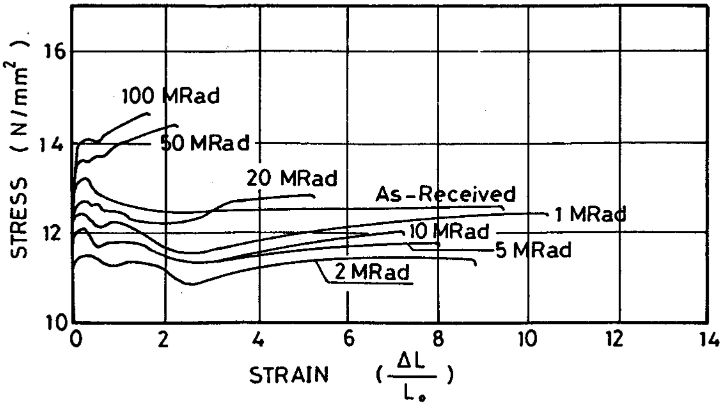

Fig. 2. **Stress-strain curves of LDPE, grade FB5005 at different γ radiation exposure dose**

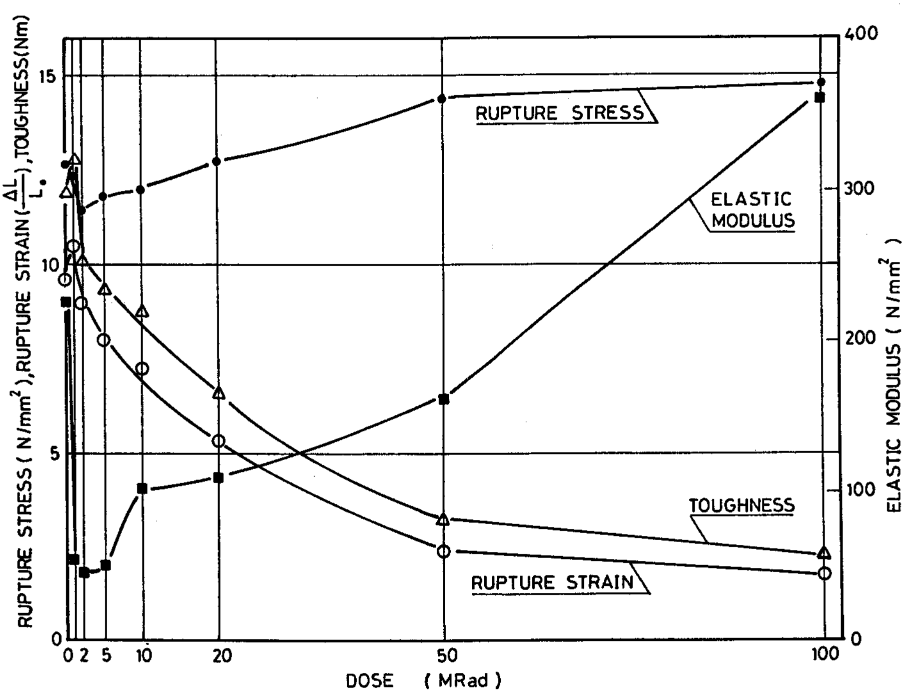

Fig. 3. Effect of γ radiation doses on the rupture stress, elastic modulus, rupture strain and toughness for the tested material

The net reduction in the ductility and the net increase in the strength of the material may occur due to a decrease in the average chain length with increase in irradiation dose which leads to cross-linking. It seems to be that properties of the material depend on the cross-links. If the number is small, the material will be ductile and tough, else the material will be hard and brittle. This means that cross-linking in the present study starts to be effective in controlling the properties of the γ radiation LDPE in the second stage only, where the γ radiation doses are larger than 2 Mrad, which is in accord with Zahran *et al.* (8) and the work recorded by Ungar and Keller (13).

However, the general (net) behaviour of the material may not be surprising for radiation cross-linking in the LDPE, grade FB 5005 but the peculiar behaviour of the material in the first stage is interesting. It is suggested that two opposing processes are taking place in the first stage simultaneously: a decrease in the crystallinity of the material and an increase in the degree of cross-linking, where the former process dominates. This is supported by the information given in Fig. 4 which shows that the velocity of the sonic wave decreases with irradiation dose until a minimum value is reached at 2 Mrad which is a good indicator that perturbation of the crystalline phase and loss of crystallinity are taking place (12). Upon further increase in the irradiation dose, (second stage), the velocity of the sonic wave increases which indicates that perturbation of the crystalline lattice diminishes and crystallinity increases where cross-linking becomes effective.

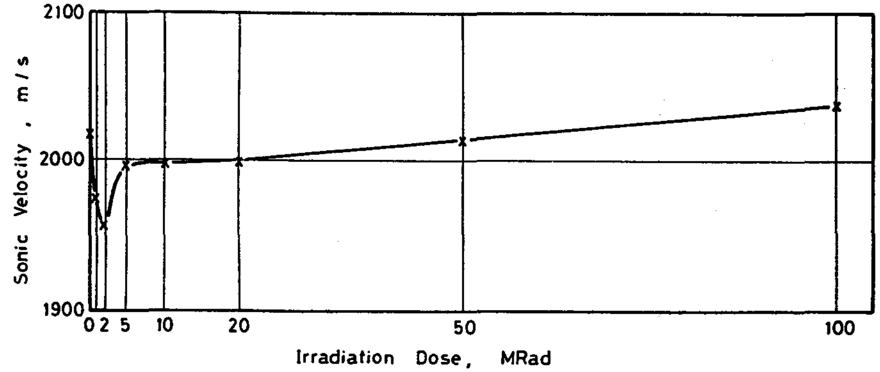

Fig. 4. Effect of irradiation dose on the velocity of sonic-wave propagation in LDPE, grade FB5005

It should be noted that the rupture stress and the toughness for the tested material correlated well with its decomposition temperature to give a linear relationship as can be seen in Fig. 5. In addition, a linear relation between the sonic and the elastic modulus was also obtained as can be seen in Fig. 6.

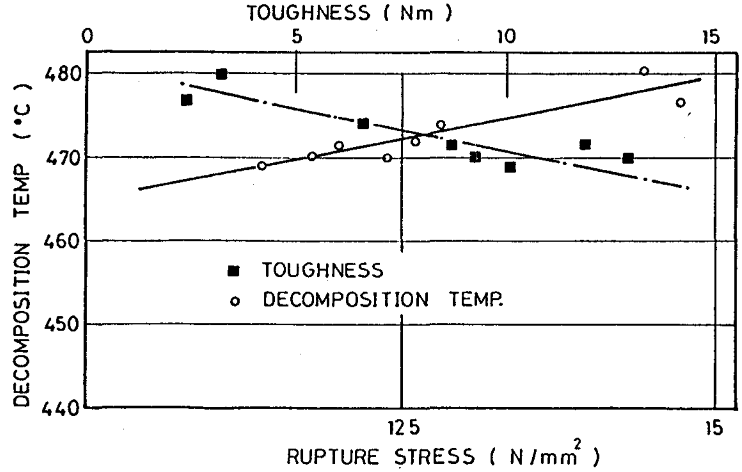

Fig. 5. Relationship between rupture stress and toughness with decomposition temperature at different Υ radiation dose

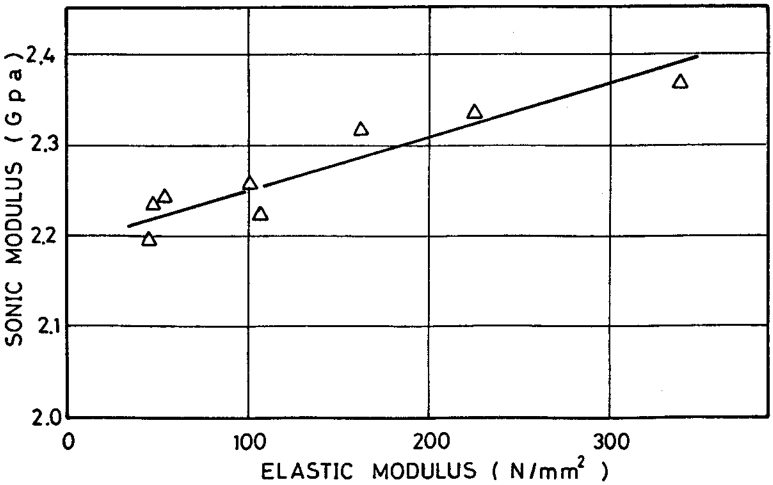

Fig. 6. Relationship between sonic modulus and elastic modulus for the LDPE, grade FB5005

CONCLUSION

1. The γ radiation of LDPE with doses greater than 20 Mrad is beneficial to some mechanical and thermal properties of LDPE, grade FB 5005 (maximum strength, rupture strength, elastic modulus, sonic modulus and decomposition temperature).

2. Such radiation causes deterioration of some other mechanical properties namely, toughness and ductility which are more important for processing and services of the material in the practical life.

3. The effect of irradiation cross-linking starts to be effective for γ radiation doses greater than 2 Mrad.

4. A linear correlation between the following pairs have been noticed: toughness/decomposition temperature, rupture stress/decomposition temperature and sonic modulus/elastic modulus.

ACKNOWLEDGEMENT

The authors gratefully acknowledge support received from the Scientific and Applied Research Centre, University of Qatar in the funding of this research through project SARC03/6-1996.

REFERENCES

1. **Tager, A., 1978**. Physical Chemistry of Polymers, Mir, Moscow.
2. **Christopher Hall, 1985**. Polymer Materials, MacMillan Publishers Ltd.
3. **Billmeyer, Jr., F.W., 1984**. Textbook of Polymer Science, Third ed., Wiley, New York.
4. **Godjevargev, G., Kostov, F. and Kostoski, D., 1993**. Mechanical and Thermal Properties of γ Irradiated LDPE - App. Blend. Rad. Effects and Defects in Solids, 127 (2), p. 259.
5. **Ray, I. and Khastgir, D., 1993**. Correlation between Morphology with Mechanical, Thermal, Physicomechanical Properties and Electrical Conductivity for EVA-LDPE Blends, Polymers, 34(10), 2030-2037.
6. **Purcell, T.C., 1971**. Polymer Preprints, 12, No. 2, p. 81.
7. **Bourgeois, J.R. and Blackett, P.W., 1988**. Engineering Plastics, ASM International, Volume 2, p. 163.
8. **Zahran, A.R.R., Kandeil, A.Y., Higazi A.A. and Kassem, M.E., 1993**. Ultrasonic and Thermal Properties of γ Irradiated Low Density Polyethylene, Journal of Applied Science, Vol. 49, pp. 1291-1297.
9. **Samuel, R.J., 1974**. Structured Polymer Properties, Wiley, New York.
10. **Read, B. and Dean, G., 1978**. The Determination of Dynamic Properties of Polymers and Composites, Wiley, New York.

11. **Chivers, R.C., Parry, R.J. and Rider, J.G., 1980**. A Spectroscopic Method for Measurements of the Ultrasonic Properties of Polymers, Proceedings of the third Eur. Conf. Manchester, pp. 18-20.

12. **Kassem, M.E., Al-Kubaisi, A.H. and Amin, R.R., 1996**. Thermogravimetric and Dielectric Relaxation Spectroscopy of Thio Semicarbazide Schiff Base and its Metal Complex. J. Thermal Analysis. Accepted.

13. **Ungar, G. and Keller, A., 1990**. Radiation and Crystals of Polyethylene and Paraffins, Symposium on Radiation Effects on Polymers, Washington D.C., pp. 101-118.