

EFFECTS OF THE ELECTRIC FIELD AND GAMMA RADIATION ON THE OPTICAL PROPERTIES OF MODIFIED LOW DENSITY POLYETHYLENE

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تأثير كل من المجال الكهربائي وأشعة جاما على الخواص الضوئية
لمادة البولي ايثيلين منخفض الكثافة

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درست الخواص الضوئية لعينات من مادة البولي ايثيلين منخفض الكثافة من النوع CD 0230 والمستعمل في عمل العزل الخارجي لأسلاك التوصيل الكهربائي. عرضت العينات لجرعات اشعاعية في مدى صفر - ١٠٠ ميغاراد ومجال كهربائي صفر - ٤١٠ فولت / سم وتم قياس طيف الامتصاص في مدى طول موجي ٢٠٠ - ١١٠٠ نانومتر. أظهرت النتائج أن كل من معامل الامتصاص واتساع الفجوة يكون معتمدا على جرعة الأشعاع كما أن البارامترات الضوئية تتغير أسياً مع شدة المجال الكهربائي. فسرت هذه النتائج على أساس كل من التفسير وإعادة التشابك والتوزيع الذري بفعل المجال المؤثر.

KEY WORDS : Low density polyethylene, γ irradiation, D.C. electric field, optical absorption.

ABSTRACT

The effect of electric field and gamma radiation on the optical properties of low density polyethylene LDPE grade CD 0230 have been studied. The samples were irradiated at gamma doses of 0-100 Mrad and varying electric field of $0-10^4$ V/cm. The optical absorption spectra were measured in the wavelength range of 200-1100 nm. The optical absorption coefficient α and the energy gap ΔE values were found to be gamma dose dependent. Also these optical parameters show a dependence on the electric field. The change in the polymer crystallinity due to irradiation can be attributed to the induced bond scission and subsequent crosslinking.

INTRODUCTION

The Optical studies are almost unique tools for reaching a detailed knowledge of molecular properties. Structure - property relationship is the possibility of handing in detail the electron phonon coupling i.e. coupling between electrons and vibrations (phonons) in such a low band gap materials. These properties are of relevant in electro-optical and all optical switching which are the basis of modern technologies in telecommunications. Polymers exhibit useful non-linear optics and electric properties [1-3] which qualify them as active components in electronic devices. The particular advantage of organic materials is the possibility of tailoring physical properties in some cases the electrically attractive behaviour can even combine with a controlled photoreactivity and fabrication of microstructure. Polyethylene, PE, is widely used as an electrical insulation material because of its low dielectric loss and superior breakdown strength [4].

The dielectric loss of PE is low due to its depolarisation orientation polarisation is low. In super clean material, super clean environment and dry method of crosslinking technology, the field concentration caused by impurity and microvoid in extra high power cable of LDPE insulation has been decreased greatly. In order to improve the stabilisation of LDPE to thermal and environmental to be used as insulating cables of high power in scientific instrument, the effect of impurities and voids are of interest.

The aim of the present work is to study the effect of both radiation and applied DC field on the optical properties of polyethylene grade CD0230 used in the insulating cables.

EXPERIMENTAL

(a) Samples:

The material used in this work was low density polyethylene Lotrene (CD0230) produced by Qatar

Petrochemical Company (QAPCO). The polymer has a melt flow index 1.8 - 2.2, crystalline melting temperature 111°C and its density at 23°C is 0.922 - 0.924 gm/cm³. LDPE was fabricated into disc like specimens 2.5 cm in diameter and thickness 0.1 - 0.2 cm.

EXTERNAL TREATMENT

The samples were subjected to different applied D.C. field varying from 0-10⁴ V/cm for half an hour [5]. The disc like samples were irradiated at room temperature in air using a gamma cell 220 manufactured by Atomic Energy Canada. The operating dose rate was 0.6 Mrad/h and a set of samples spanning an integrated dose from 0 to 100 Mrad.

OPTICAL ABSORPTION SPECTRA

The absorption spectra of the virgin as well as treated samples were measured by U.V.-160 Shmadzu spectrophotometer in the wave length range 200 - 1100 nm. A perfectly flat sample of LDPE thin sheet has been placed vertically in the path of the sample beam while the reference beam reach directly to the detector. So, the spectral data obtained are absolute values.

RESULTS AND DISCUSSION

Effect of electric field

Fig. 1 shows the spectral distribution of absorbency for LDPE (CD0230) films in the visible and UV range at different applied DC field. It is clear that the absorbency decreased to minimum value at electric field 10² V/cm and increased upon the increase of field. This behaviour can be attributed to preturbation followed by degradation [6,7] of LDPE as results of electric field which displace charge and lead it to field induced elastic strain in materials. The absorption coefficient (α) at angular frequency (ω) is given by:

$$\alpha(\omega) = \frac{1}{t} \ell n \frac{I_0}{I} \quad (1)$$

Where I_0 and I are the intensity of incident and transmitted light respectively and t is the film thickness.

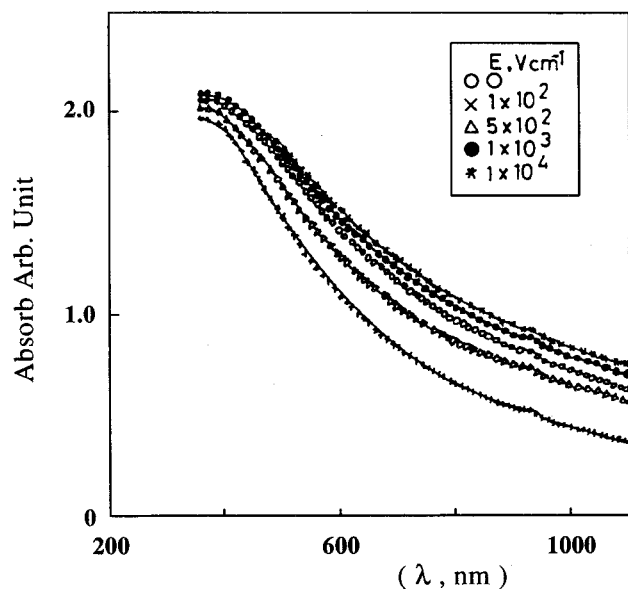


Fig. (1) Spectral distribution of optical absorption at different applied DC electric Field for LDPE grade CD0230.

Fig. (2) shows the variation of the logarithmic of the absorption coefficient as a function of photon energy. the optical absorption and particularly the absorption band edge is good method for studying optically induced transition and gives information about the structure and optical energy gap of CD0230, LDPE.

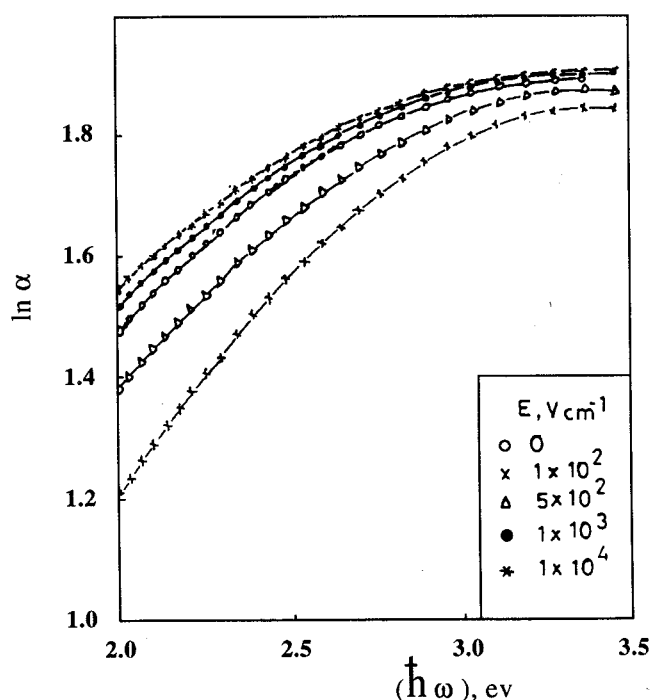


Fig. (2) Variation of the logarithmic of absorption coefficient with photon energy at different applied electric field.

The absorption coefficient $\alpha(\omega)$ for many amorphous or crystalline materials behave exponentially as a function of photon energy $h\omega$ in the form:

$$\alpha(\omega) = \alpha_0 \exp(\hbar\omega / \Delta E) \quad (2)$$

Where (α_0) is constant \hbar is the reduced plank constant and ΔE is an energy which is interpreted as the width of band tail of the localized state in the normally forbidden band gap that is associated with the amorphous nature of the materials.

The estimated energy band tail showed an increase with the field applied in logarithmic relation

$$\Delta E (ev) = - 0.67 + 0.59 \ln (\text{field}) \quad (3)$$

This behaviour can be attributed due to as the externally applied field induces the electric stress at the interface and enhance polymer morphology and it has an advantage of easy and exact manipulation in developing polymer morphology oriented to desired direction [8, 9]. More study, the absorption edge of in LDPE spectra was analysed using the formula quoted by Davis and Mott [10]

$$\alpha(\omega) = \beta (\hbar\omega - E_{opt})^n \quad (4)$$

Where E_{opt} is the optical energy gap of the system, β is the constant and n is an index determined by the nature of the transition during the absorption process. As shown in Fig. (3) the linearity of $\alpha(\omega)^{1/n} = f(\hbar\omega)$ indicates the existence of direct transition. The extrapolation of the linear part of the curves towards lower energies yields a value for the forbidden energy band E_{opt} at different applied electric field can be obtained. The estimated values of E_{opt} were found to increase with increases field upto 10^2 v/cm followed by a decrease as applied field increase. This behaviour can be explained by surface electric stress creation at low applied electric field, in which the degree of deformation is increased in the meanwhile the morphology of polymer is easily controlled by the direction and strength of the applied

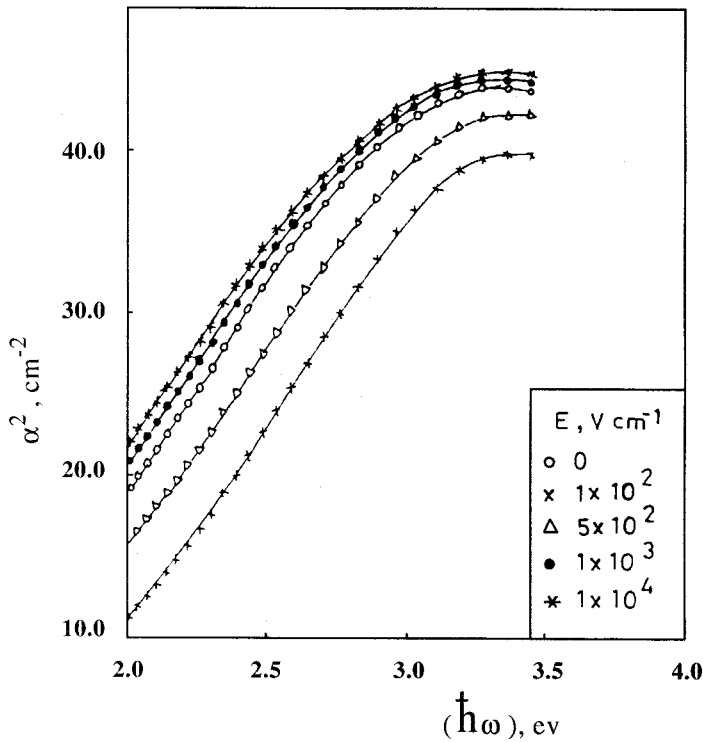


Fig. (3) Variation of α^2 with $\hbar\omega$ for LDPE exposed with different electric field.

electric field.

Fig. (4) represents the experimental data of the optical absorption spectral distribution for virgin and radiated polyethylene samples. It is clear from this figure that for the irradiated samples of 0.3 cm thickness, there is no sharp absorption band edge wave length. For exposed samples to various dose of γ radiation the change of the absorption edge wave length shift to longer wave length i.e. to lower energy. However, for an irradiation dose up to 10 Mrad, the absorption band edge wavelength show a shift to shorter wave length i.e. to higher energies. As the dose increases from 0 to 100 Mrad, both the absorption band tail width of the localised state, ΔE , optical energies of transition E_{opt}^d , E_{opt}^i are obtained from equations (2,4) and figures 5, 6, 7. The data obtained are given in Table (1). It is clear that from 0 up to 10 Mrad, the change in the optical energy of transition E_{opt}^d , E_{opt}^i for the γ irradiated samples ΔE_{opt}^d and E_{opt}^i decreases with increase of the dose. On further increase of the radiation dose there is a slight change in E_{opt}^d and no significant change in E_{opt}^i . The optical energy band tail showed opposite behaviour of the E_{opt}^i . This behaviour

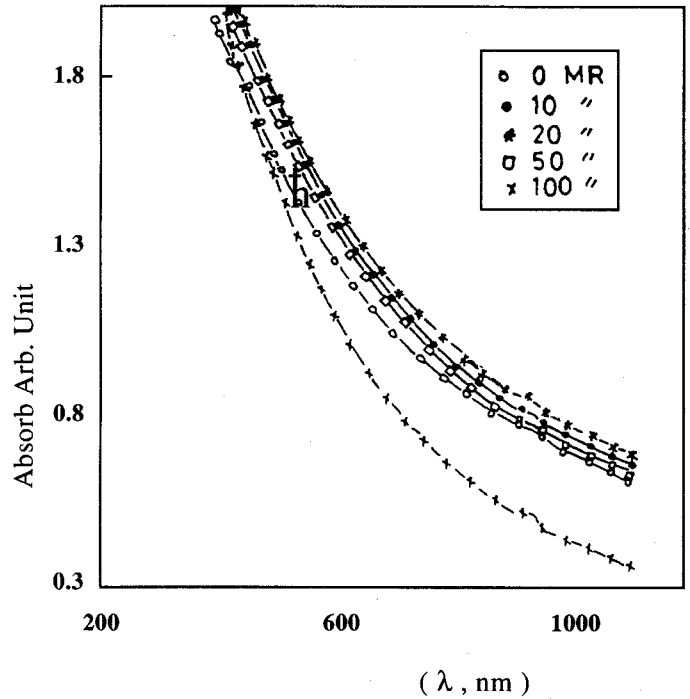


Fig. (4) Optical absorption spectra versus wave length for unirradiated and γ radiated CD0230 LDPE.

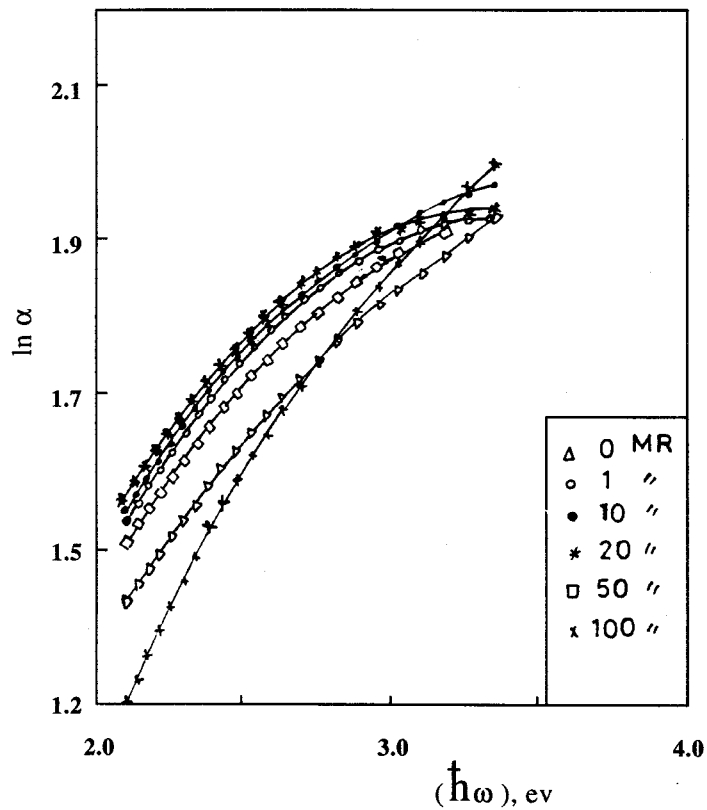


Fig. (5) The photoenergy dependence of absorption coefficient of LDPE, CD0230 as $\ln \alpha = f(\hbar\omega)$.

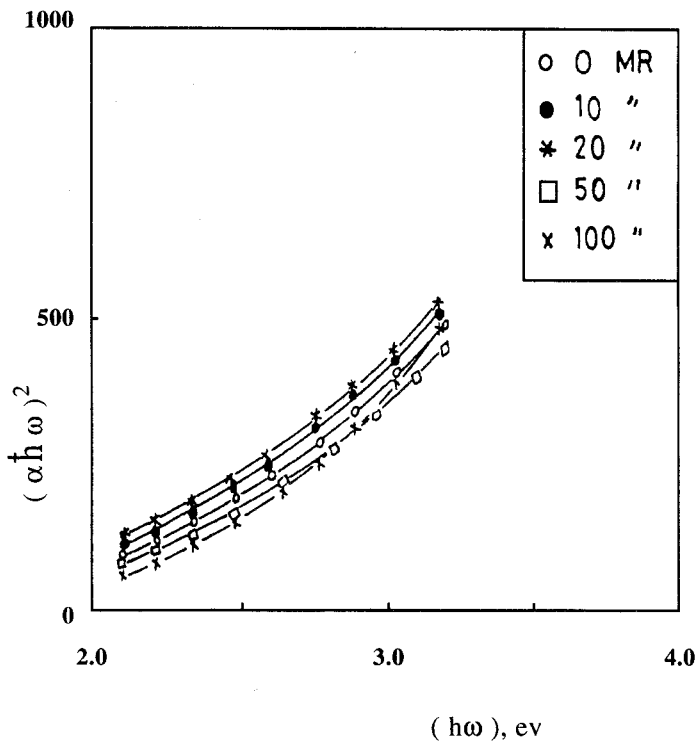


Fig. (6) The dependence of $(\alpha \hbar \omega)^2$ on the photon energy $h\omega$ at different γ radiation doses of CD0230

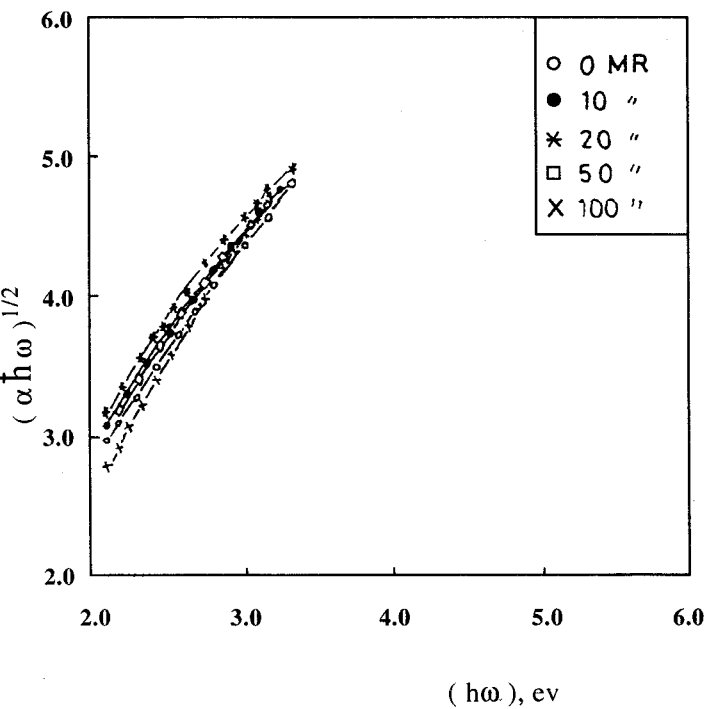


Fig. (7) The dependence of $(\alpha \hbar \omega)^{1/2}$ on the photon energy $h\omega$ at different γ radiation doses of CD0230

can be explained as in the low γ doses up to 10 Mrad, the crystalline structure is assumed to be perturbed and leading to an increase in the degree of disorder [11]. From the density of states model, it is known that E_{opt} decrease with increasing the degree of disorder [12]. Furthermore, the band tailing shifts to higher energies and extends into forbidden band [13].

Conclusions:

1. In the low density polyethylene, the two mechanisms of chain session and cross-linking compete.
2. The effect of high applied electric field showed no significant effect the polymer structure.
3. Degradation is promoted during long term of γ -irradiation.

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TABLE (1)

Dose	ΔE (ev)	E_{opt}^d (ev)	E_{opt}^i (ev)
0	2.01	1.56	2.42
1	2.12	1.33	1.95
5	2.25	1.45	1.9
10	2.35	1.56	2.0
20	1.58	1.56	2.07
100	1.45	1.56	2.5

REFERENCES

1. **Gilulis, M.D., G. Micocci, Rella, P., Sicalia and Tepora, A. (1993).** Optical Absorption of Tellurium Suboxide thin films. *Phys. Status Solidi* Vol. 136, pp. K.101
2. **Enomoto, Y. and T., Murakani, (1985)** Optical Detectors using superconducting thin film *Japanese J. of Appl. Phys.*, Vol. 24 p. 471.
3. **Khan M.S.R. and A., Reza, (1992)** Growth of discontinuous gold films and study of their electrical and optical properties *Phys. Status solidi* Vol. 129 pp. K11.
4. **Tanimoto, G., M., Okashita, Aida, F. and Y., Fujiwara (1991).** Temperature dependence of $\tan \delta$ in polyethylene proceeding of 3rd International Conf. On properties. and application on dielectric materials, pp. 1068 Tokyo, Japan.
5. **Kassem, M.E., M., El. Muraikhi, Al-Houty, L. and A.A., Mohammed, (1992).** Field induced phase in IC-C Phase of Li Rb SO₄ Crystals. *Phase transition*, Vol. 38, pp. 55
6. **Kassem, M.E., A.Y., Kandeil, and Al-Houty, L. (1992).** Effect of γ irradiation on electrical and mechanical properties of phenolic compacts. *J. Polym. Mater.* Vol. 9, 113
7. **Mahmoud, S. A., N. K., Madi, Kassem, M. E., and A. , El-Khatib , (1996).** Changes in the electrical properties of pure and doped polymers under the influence of small doses of x-rays. *Rad. Effects and def. in solids* Vol. p.
8. **Ankel, B., G. Leising, and Kahlert, H. (1987).** Optical properties of conjugated segments embedded in polyvinylidene chloride, *Solid State Commu.* Vol. 62 p. 245.
9. **Jones, R.A., G.A. Salmon, and Ward, I.M. (1993).** Radiation-induced crosslinking of polyethylene in the presence of acetylene: A. Gel. Fraction, UV-visible and ESR spectroscopy study. *J. Polym. Sci, Part B Polymerphy* Vol. 31, p. 807.
10. **Mott, N.F. and E. A. Davis (1979)** "Electronic process in noncrystalline Materials" 2nd ed. (Clarendon Press. Oxford).
11. **Zahran, R.R., A.Y., Kandeil, Higazy, A.A., and M.E. Kassem, (1993).** Ultrasonic and Thermal properties of γ irradiated low density polyethylene. *J. Applied Polymer, science* Vol. 49, p. 1297.
12. **Navratil, J., P., Lostak, Drasar, C. and T. Blachut, (1996).** Optical properties of (Bi_{1-x} Sb x)₂ Se₃ single crystals *Phys. Stat. Sol. B.*, Vol. 194, p. 783.
13. **Al-Ani, S.k., C.A. Hogaith and El-Mallawany (1985).** A study of optical absorption in tellurite and tungsten-tellurite glasses. *J. Mater., Sci.* Vol. 20, p 661.