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Study of the effect of radiation and frequency on the electrical properties and ultrasonic properties of polyethylene

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ABSTRACT

The authors presented this manuscript to study the effect of γ-radiation and frequency on dissipation factor, conductivity, dielectric constant, density, longitudinal ultrasonic velocity, and longitudinal modulus of polyethylene that is frequently used in many industrial polymeric applications, using γ-radiation doses from 0 to 30 Gy. The frequency range is from $2*10^{-6}$ to $10 * 10^{-6}$ Hz. It was observed that the conductivity of polyethylene increase with increasing frequency from $4 *10^{-6}$ to $10*10^{-6}$ Hz also it is slightly increased with increasing radiation dose from 10 Gy up to 30 Gy. The longitudinal ultrasonic velocity decreases with radiation dose from 10 Gy to 30 Gy.

Key words: polyethylene, Gamma radiation, electrical properties, ultrasonic properties

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INTRODUCTION

Radiation interacts with polymers in two ways: chain scission, which results in reduced tensile strength and elongation, and crosslinking, which increases tensile strength but reduces elongation. Both reactions occur simultaneously, but one is usually predominant, depending on the specific polymer and additives involved. Chain scission classically affects stressed polymers to a greater extent than non stressed polymers. The radiation interaction was mainly due to an oxidation process of the main polymer chain yielding a conductivity alteration **[1&2].**

A dielectric material is both electrically and mechanically isotropic before applying electrical field. When an electrical field is applied to this material, it is subjected to internal stress and thus electrets extend along the electrical field direction as the result of these electrical stresses. This effect is termed electro restriction. The deformation involves density changes, with corresponding changes in dielectric constant or the refractive index. The magnitude of the deformation depends not only on the elastic modulus of the medium, but also on dielectric constant and density. Mechanical deformation does give rise to an internal electric polarization proportional to the strain **[3].**

While an electric field cannot accelerate electrons and ions in an insulator, it can nevertheless displace these charges by small distances. Since positive and negative charges are displaced in opposite directions, the electric field generates an electric polarization that creates an internal electric field opposite to the one that is applied. The total electric field inside the body is then decreased by the factor κ, called the dielectric constant. This polarization can be due to the distortion of electronic orbitals, by the displacement of ions or by the reorientation of existing dipoles in the solid **[4]**.

Much of the work on radiation effects is concerned with the use of radiation as a tool to modify the properties of a particular polymer. For example, UV light has been used to create refractive index patterns in polyimide films doped with UV-sensitive benzoin type photosensitizers. Other recently reported examples of radiation-induced modification processes include crosslinking of poly(tetrafluoroethylene) by x-rays and electrons , radiation-induced formation of diene, triene and tetraene from polyethylene in the presence of acetylene, electron irradiation-induced gelation of blends of polystyrene - polyvinyl methyl ether), and gamma ray irradiation induced grafting of styrene in poly(vinylidene fluoride) films. Other studies have shown that radiation can also alter the electrical conductivity of a polymer **[5-9].**

Yu Chao Li et al. [10] studied the effect of conductive nanofiller, expanded graphite (EG), on polyvinylidene fluoride (PVDF) by direct melt blending process. The electrical conductivity and dielectric properties of resulting PVDF/EG composites were investigated in a wide range of frequencies from 102 to 108 Hz. It evidence that, the conductivity and permittivity of percolating PVDF/EG composites were observed to be temperature dependent.

The frequency dependent dielectric and conductivity behavior of a plasticized polymer nanocomposite electrolytes (PPNCEs) based on poly(ethylene oxide) + NaClO4 with dodecyl amine modified montmorillonite (DMMT) as the filler and poly(ethylene glycol) as the plasticizer have been studied by *Dillip K. Pradhan* et al. [11]. The formation of nanocomposites and changes in the structural and microstructural properties of the materials were investigated by x-ray diffraction (XRD) and optical microscopy techniques. Studies of dielectric properties at lower frequencies show that the relaxation contribution is superimposed by electrode polarization effect. The appearance of peak for each concentration in the loss tangent suggests the presence of relaxing dipoles in the polymer nanocomposite electrolyte (PNCE) films. On addition of plasticizer, the peak shifts towards higher frequency side suggesting the speed up the relaxation time. The dc conductivity increases with increase in plasticizer concentration. Analysis of frequency dependence of dielectric and modulus formalism suggests that the ionic and polymer segmental motions are strongly coupled. S. Darwish et al. [12] studied the effect of The temperature on dielectric properties of polyethylene in the frequency range from 10^1 to 10^5Hz. The frequency dependence of the complex impedance in the complex plane was fitted by semicircles. An equivalent circuit of a bulk resistance in series with parallel surface resistance - capacitance combination, could represent the system.

Poly ethylene terephthalate films with semiconducting and interface layers were investigated.The electrical properties, such as volume resistivity, tanδ (dissipation factor), and breakdown strength were measured by D. S. Kim et al. [13]. It is found that the volume resistivity and electrical properties of PET films are changed. Breakdown strength and dissipation factor of PET film with semiconducting layer are decreased more greatly than PET and PET/PET films.

MATERIALS AND METHODS

1.1. Electrical properties

Electrical sample holder supplied by the general Radio company, Hartshorn type 1690-A, was used to accommodate the polyethylene specimen, diameter 4.2 cm, and of about 0.2 cm thickness (Polyethylene was supplied by alsweedy company , Cairo - Egypt). This holder can measure the thickness accurately up to 0.00254 cm (0.001 inch). The capacitance and dissipation factor of the various samples were measured using the General Radio Capacitance Measuring System, Type 1615-A. This bridge is a transformer ratio arms designed to measure the capacitance and dissipation factor with high accuracy up to 0.01%.

For a dielectric sample placed between two parallel plates capacitor, the measured values of the capacitance and dissipation factor were used to calculate the conductivity and the dielectric constant using the following equations [14]:

$$\sigma = \frac{1}{\rho}$$

Where σ is the conductivity

ρ is the resistivity

$$\varepsilon = \frac{C * D}{\varepsilon_0 * A}$$

Where ε is the dielectric constant
ε_0 is the dielectric constant of free space ($8.85*10^{-12}$ Fm^{-1})
A is the cross section area (m^2)

1.2. Ultrasonic measurement

Ultrasonic pulse echo technique introduces beams of high frequency sound waves into the material. The sound waves, at carefully optimized frequencies, travel through the material with some attendant loss of energy (attenuation) and are reflected at interfaces. The reflected beam is displayed and then analyzed to define the physical state of the materials.

The ultrasound measurements in this study were performed using an ultrasound flaw detector (USIP 20) to display echo, an oscilloscope (54615B) to obtain the time traveling through specimen, a longitudinal transducer (central frequency of 0.50MHz and band width of 1 MHz), and a shear transducer (central frequency of 0.45MHz and band width of 0.9MHz). The incident wave is generated from the transducer directly contacting the specimen. This is achieved by applying a coupling agent of glycerin. Thus, the incident waves are transmitted in a sample of thickness X and reflected back and forth at the two surfaces. When the reflected wave reaches the upper surface and is received by the transducer, an echo signal containing several oscillations will be gained. A series of echo signals can be obtained as ultrasound waves are reflected between the two surfaces. However, the amplitudes of the echo signals are gradually decreased with time due to ultrasonic attenuation, the first two echo signals, of amplitudes (A_1, A_2), and corresponding time (t_1, t_2) can be read and chosen to calculate the velocity v and the attenuation α,

$$V = 2 X / (t_2 - t_1)$$

The measurements of ultrasonic velocities were performed several times and the accuracy is about 0.02 %.

RESULTS AND DISCUSSION

1.3. Effect of frequency on electrical properties of polyethylene sample

Figures 1– 3 display the frequency dependence of electrical properties. The variation of conductivity with frequency for polyethylene sample is shown in Figure 1 at room temperature. At low frequency, the electrical conductivity of the polyethylene is decreasing with frequency up to 4 µHz then it increasing with increasing frequency up to 10 µHz. The polyethylene sample have higher dissipation factor at 4 µHz frequency as shown if Figure 2. Figure 3 show the relation between dielectric constant and frequency at each radiation dose. It indicates that the dielectric constant is slightly increased with increasing frequency. The slightly increasing of dielectric constant is mainly attributed to the match of interfacial polarization of polyethylene sample to external electric field at applied frequencies [12&15]. Dielectrics may be broadly divided into, non-polar materials, and polar materials. In non-polar materials the molecules, which are usually diatomic and composed of two atoms of the same type, may be represented as positive nuclei of charge "q" surrounded by a symmetrically distributed negative electron cloud of charge "-q" . In the absence of an applied field the centers of gravity of the positive and negative charge distribution coincide. When the molecules are placed in an external electric field, the positive and negative charges experience electric forces tending to move them apart in the direction of the external field. In polar dielectrics the molecules, which are normally composed of two or more different atoms, have dipole moments even in the absence of an electric field, which is the centers of their positive and negative charges do not coincide. In the presence of an externally applied field the molecules tend to themselves in the direction of the field [16].

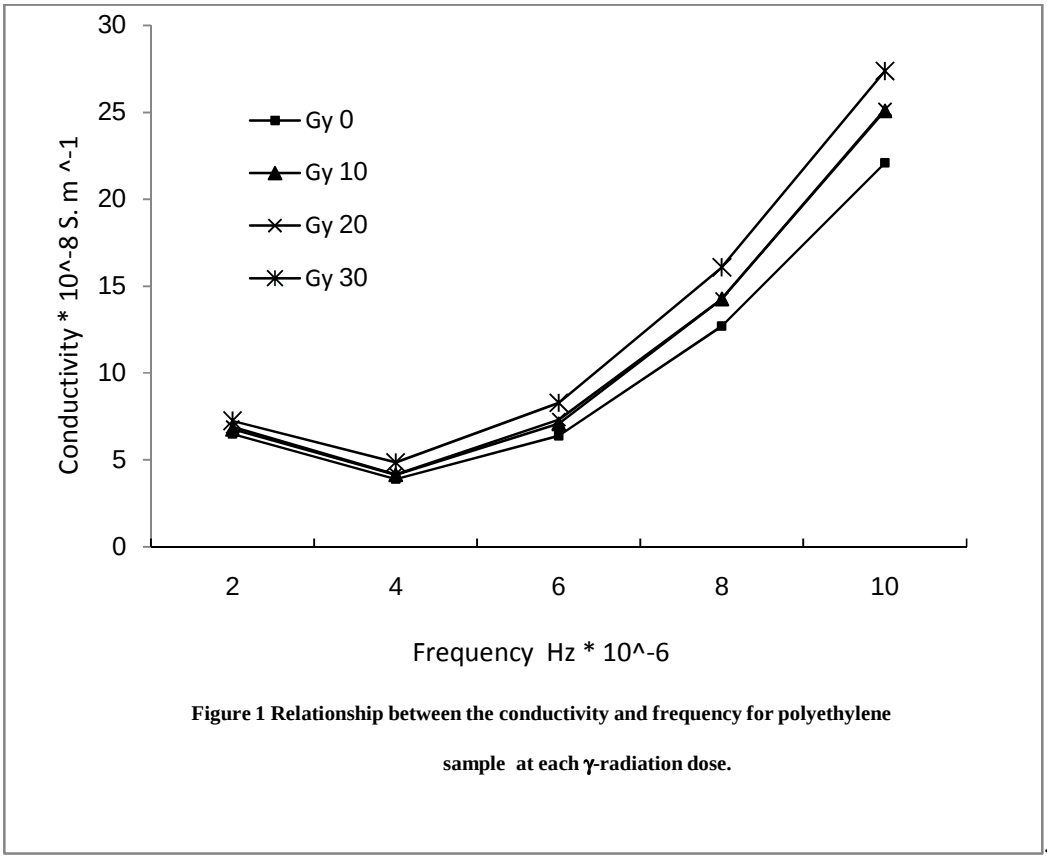

Figure 1 Relationship between the conductivity and frequency for polyethylene sample at each γ-radiation dose.

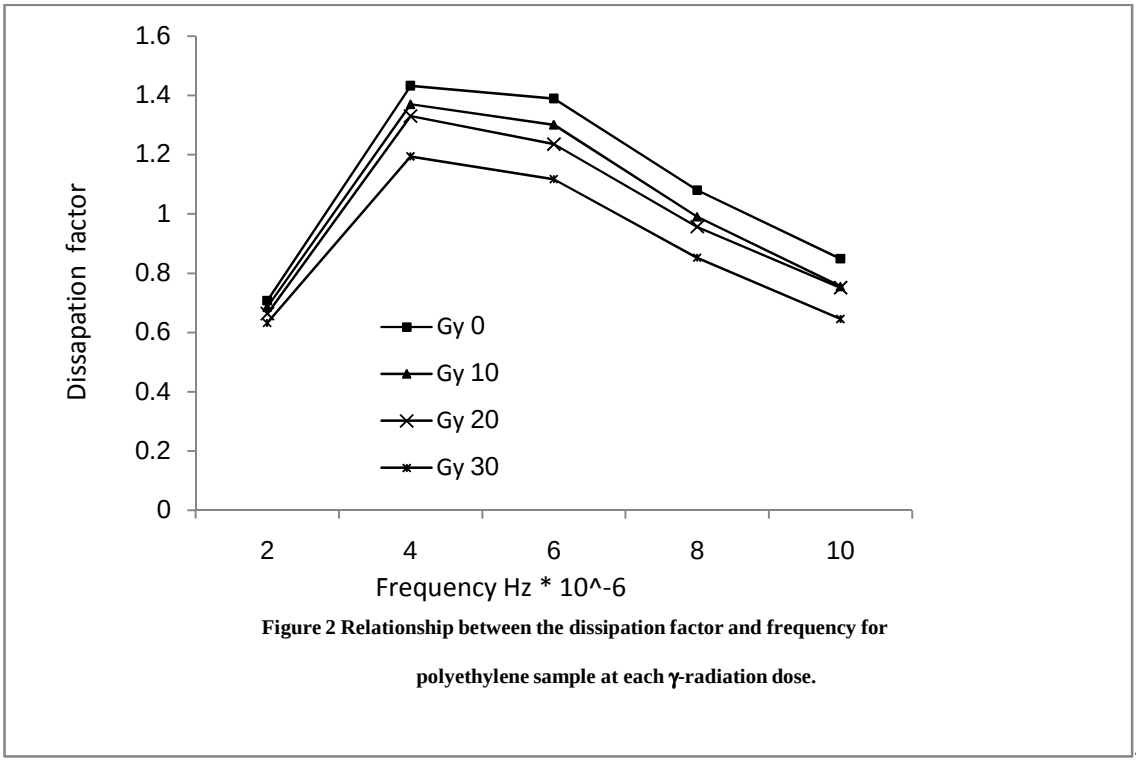

Figure 2 Relationship between the dissipation factor and frequency for polyethylene sample at each γ-radiation dose.

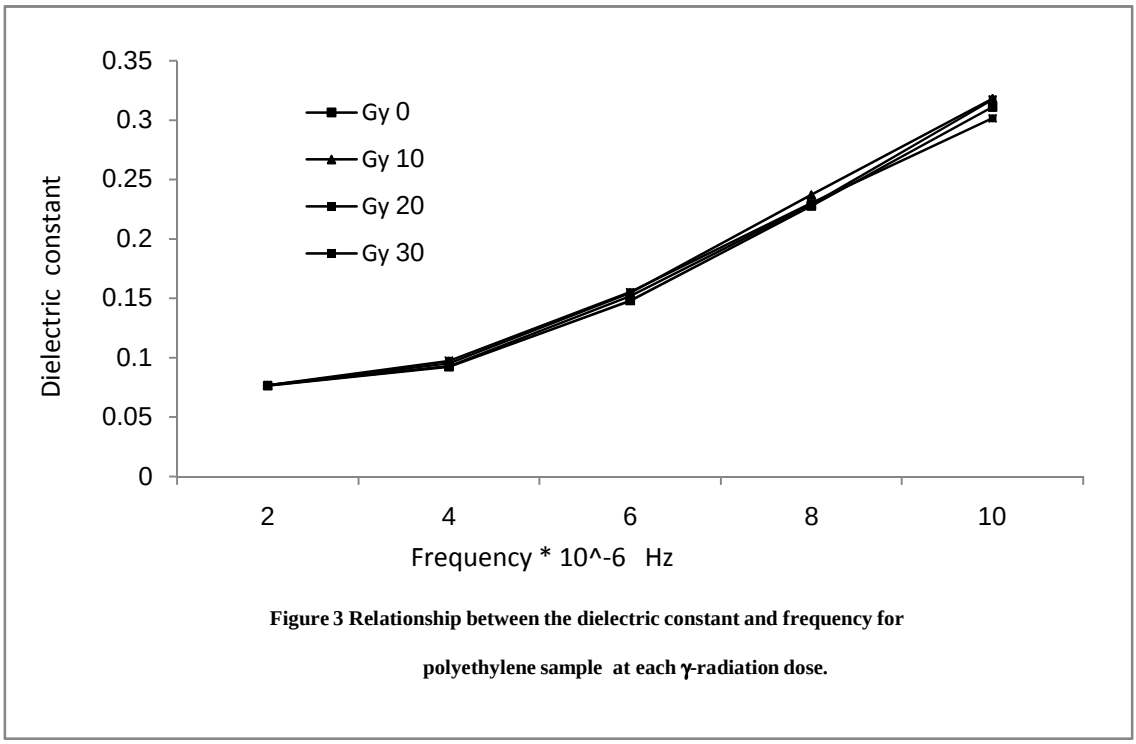

Figure 3 Relationship between the dielectric constant and frequency for polyethylene sample at each γ-radiation dose.

1.4. Effect of γ-radiation on electrical properties of polyethylene sample

Figures 4-5 illustrates the variation of conductivity and dielectric constant of polyethylene sample with increasing of radiation dose. Figure 4 indicate that the conductivity of polyethylene sample is slightly increased with increasing radiation dose from 10 Gy up to 30 Gy. It may be presumed that the action of gamma rays results in excitations of its molecules and creation of free electrons and ions that migrate through the polymer network till they are trapped somewhere, leaving deficient regions. These electronic and ionic configurations cause the changes in the electric conductivity of PE samples. Generally, the major effects in polymers arise from the dissociation of primary valence bonds into radicals [17].

Figure 5 show the relation between dielectric constant and radiation dose at each frequency applied. It indicates that the dielectric constant remains nearly unchanged with increasing Radiation dose from 10 Gy up to 30 Gy. Because, radiation interacts with polymers in two ways: chain scission, which results in reduced tensile strength and elongation, and crosslinking. Both reactions occur simultaneously, but in this case first way is predominant because in polymers under irradiation, a chain of reactions, in which oxygen and moisture from the environment may be incorporated, starts with rapid electronic phenomena. This is followed by the generation of reactive, short-lived intermediate compounds. The main result of ionization is the breaking of chemical bonds and the creation of new ones, which causes increase in conductivity and leads to long-lived forms of physical breakdown **[18-20]** and Chain scissions, which follow a free radical mechanism, the additional thermal oxidation degradation takes place in the irradiated polymer and its blends during their application at high temperature conditions **[21]**. Hence, most polymeric systems require the addition of stabilizers to provide protection during radiation processing and end use.

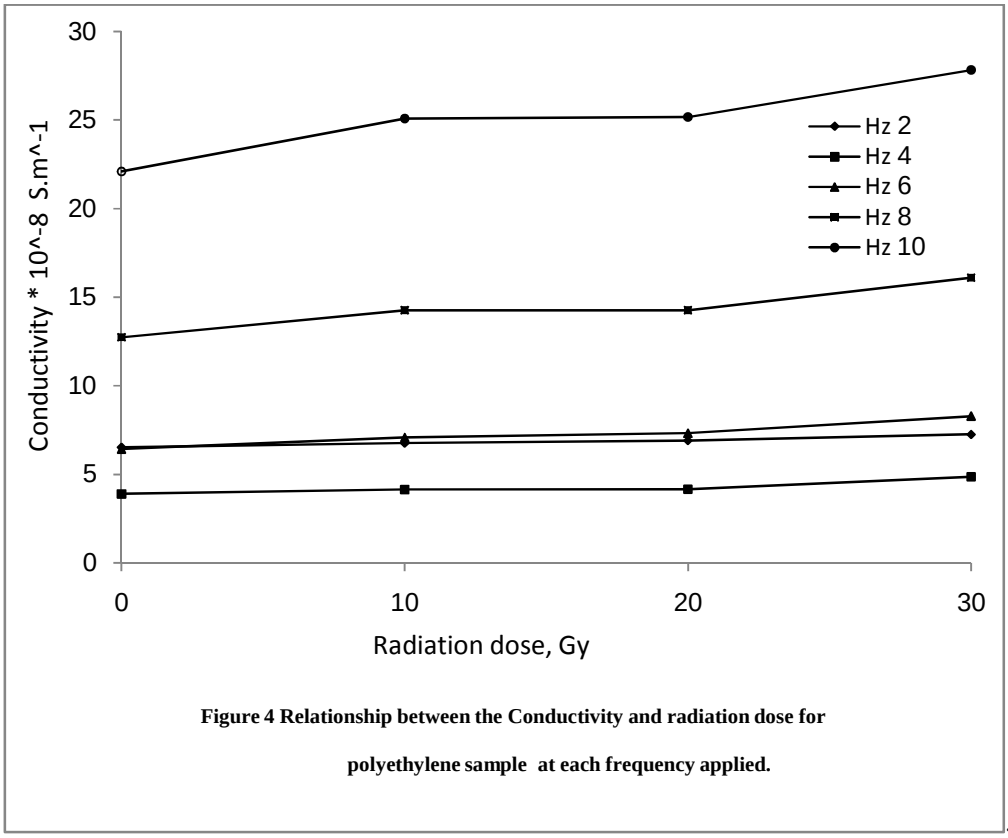

Figure 4 Relationship between the Conductivity and radiation dose for polyethylene sample at each frequency applied.

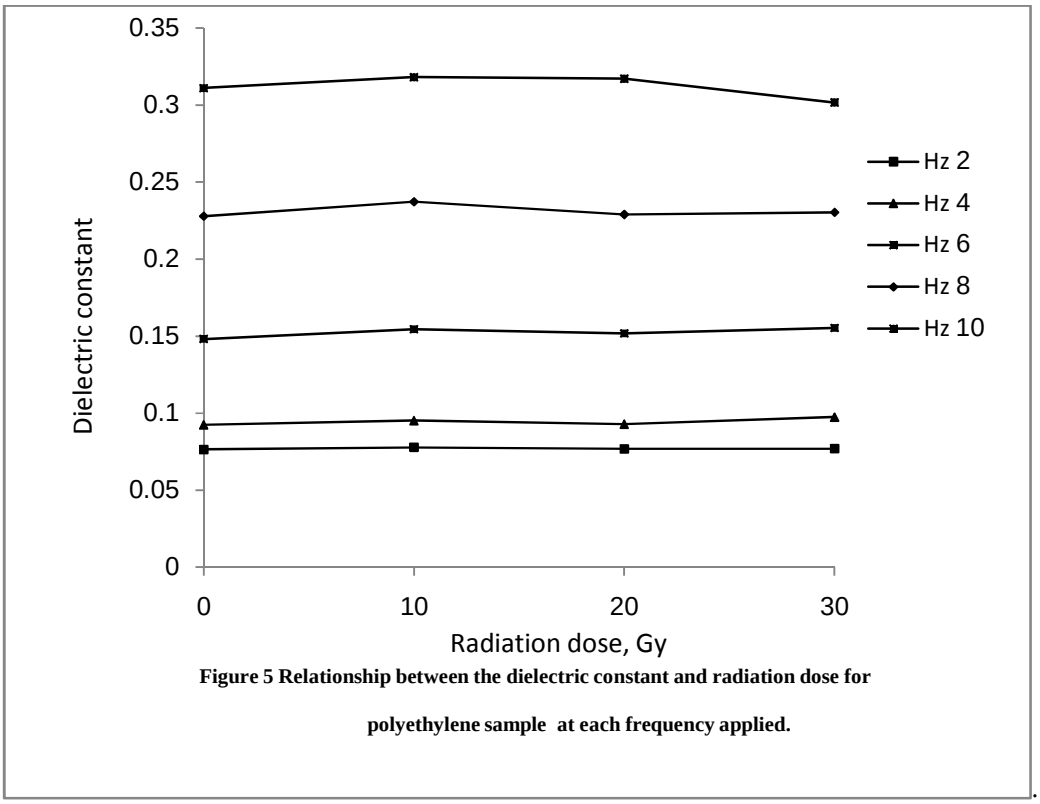

Figure 5 Relationship between the dielectric constant and radiation dose for polyethylene sample at each frequency applied.

1.5. Effect of γ-radiation on ultrasonic properties of polyethylene sample

The density and molar volume of solid materials depend upon many factors such as structure, cross-link density, and dimensionality of interstitial spaces [22].

Figure 6 shows the relation between the density of polyethylene and radiation dose. The density values were found to increase with the increase of radiation dose from 0 Gy to 10 Gy. Moreover, it showed that the radiation dose in range 10 Gy up to 30 Gy has no evidence effect on the density of polyethylene sample. The increase in density values with the increase of radiation dose from 0 Gy to 10 Gy was expected as due to cross linking at lower doses [23]. This behavior was found to correlate with those shown in Figures 4 and 5 indicating that the radiation dose has no evidence effect on the conductivity and dielectric constant of polyethylene sample.

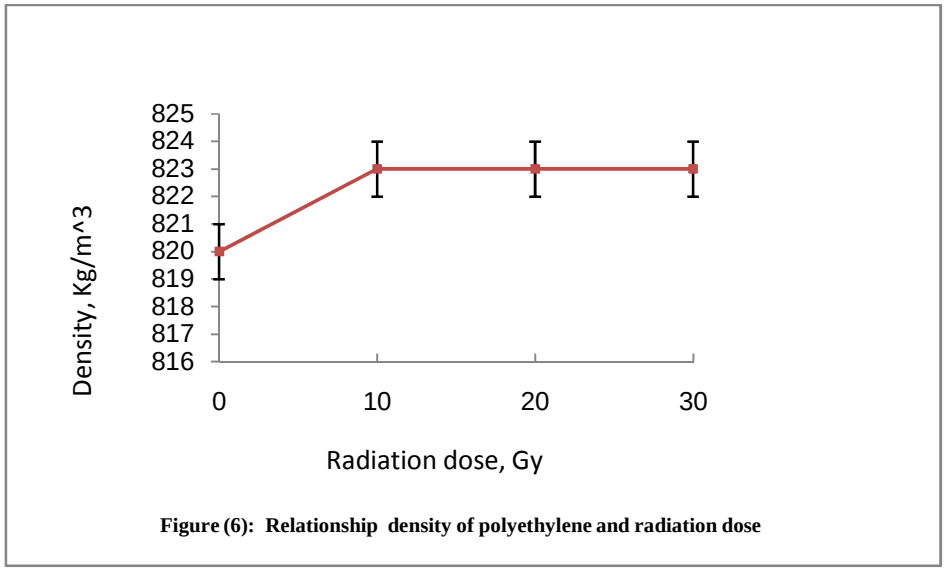

Figure (6): Relationship density of polyethylene and radiation dose

Figure (7), shows the relation between the longitudinal ultrasonic velocity with γ-radiation dose from 0 Gy to 30 Gy. It showed the increase in longitudinal ultrasonic velocity with radiation dose from 0 Gy to 10 Gy, and then decreased with the increase in radiation dose from 10 Gy to 30 Gy.

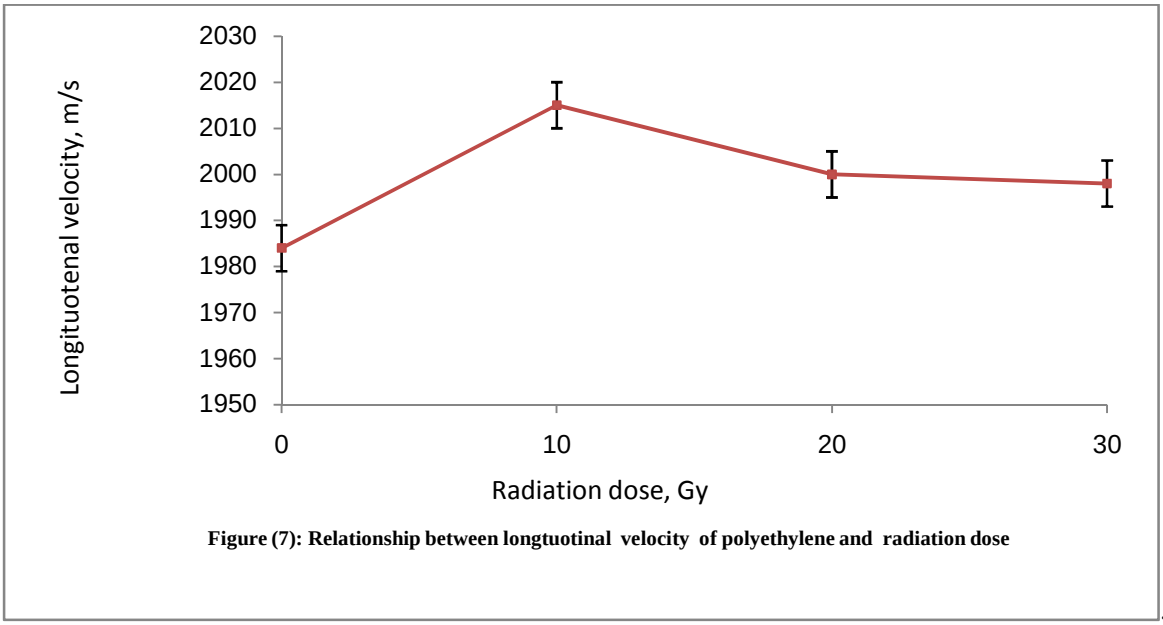

Figure (7): Relationship between longtuotinal velocity of polyethylene and radiation dose

In general, the increase of ultrasonic wave velocity is related to the decrease in inter-molecular spacing of the material. Also, Suljovrujic [24] has reported that irradiation of polymers results in the creating of free active molecules with electron deficiency (radicals), in the breaking bonds of and the making of new once. Two main effects result when PEs is subjected to ionizing radiation in air: crosslinking and oxidative degradation. Moreover, J. Suarez et al. [23] have reported that PE gamma irradiation process involves crosslinking at lower doses and chain scission at higher doses. Therefore, the increase in longitudinal ultrasonic velocity at lower doses from 0 Gy to 10 Gy was expected as due to the crosslinking and then its decrease at higher doses from 10 Gy to 30 Gy was expected as due to the chain scission.

Figure (8), shows the relation between the longitudinal modulus and radiation dose. It showed the increase in longitudinal modulus at lower doses from 0 Gy to 10 Gy and then decreased at higher doses from 10 Gy to 30 Gy. In fact, the increase in longitudinal modulus at lower doses was expected as due to the increase in crosslinking which means the decrease in crystallinity of PE as reported before [23-25] confirming the increase in rigidity of polymer. Then, the decrease in longitudinal modulus at higher doses was expected as due to the chain scission which means the decrease in rigidity of PE. Therefore, the behavior of longitudinal modulus with radiation doses correlates well with those of conductivity, dielectric constant and density.

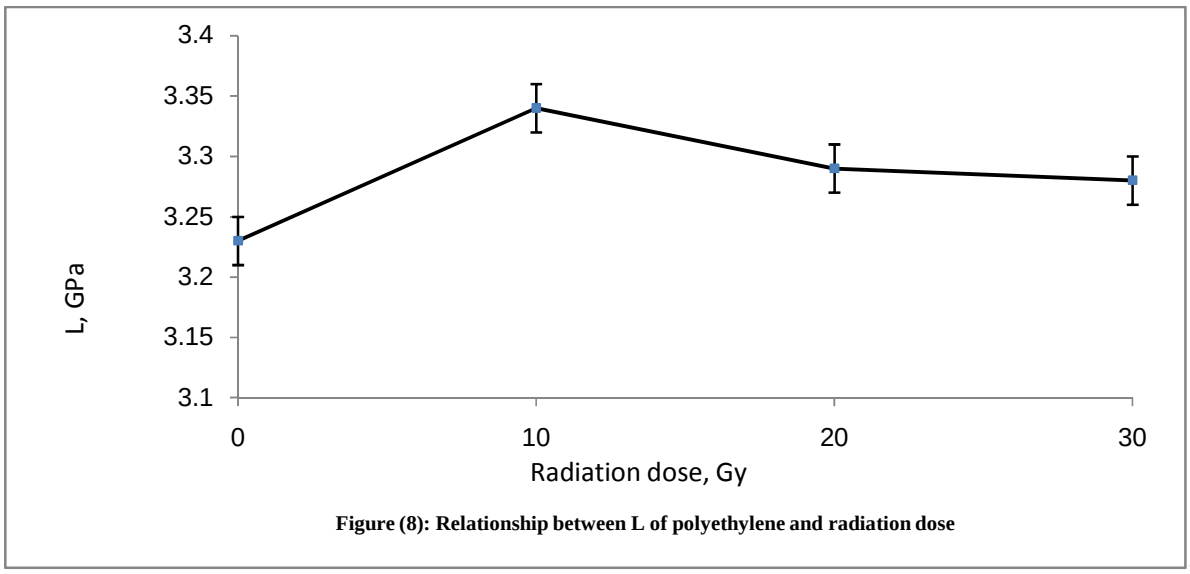

Figure (8): Relationship between L of polyethylene and radiation dose

CONCLUSION

- At low frequency, the electrical conductivity of the polyethylene is decreasing with frequency up to 4 µHz then it increasing with increasing frequency up to 10 µHz.
- The conductivity of polyethylene sample is slightly increased with increasing radiation dose from 10 Gy up to 30 Gy.
- The dielectric constant remains nearly unchanged with increasing Radiation dose from 10 Gy up to 30 Gy.
- It showed the increase in longitudinal ultrasonic velocity with radiation dose from 0 Gy to 10 Gy, and then decreased with the increase in radiation dose from 10 Gy to 30 Gy.
- The longitudinal modulus at lower doses from 0 Gy to 10 Gy and then decreased at higher doses from 10 Gy to 30 Gy

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