Study on electrical property relaxations of 10 MeV electron beam irradiated HDPE

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Abstract: Cross linking of the HDPE was performed with high energy electron beam. EPR spectroscopy was used to study the life time of unpaired electron in the irradiated samples. Dielectric constant and dielectric strength almost remained unchanged but, volume resistivity and dissipation factor increased and decreased respectively, after irradiation. The results shows that the dissipation factor is going to be recovered slowly with the storing time at room temperature. It is believed that variation of some electrical properties during time after irradiation are due to the effects of free radical and charge trapping remained in the bulk of material.

Key words: Electrical properties, breakdown voltage, volume resistance, EPR measurements, dielectric loss tangent, dielectric constant, electron beam, high density polyethylene.

Introduction

The largest volume of polymeric materials used for wire and cable insulation is thermoplastics, namely polyethylene and polyvinyl chloride and to lesser degree, elastomeric compounds. The main reason for prevalence of PE and PVC in wire and cable insulation is their easy processing and relative low cost. However their main disadvantage is that their physical properties such as plastic flow at elevated temperatures, environmental stress cracking, poor solvent resistance and low softening temperatures, cannot always meet demands imposed on them by modern applications. Cross linking of these materials improves their toughness flexibility and impact resistance, resistance to solvents and chemicals, and their service temperature. Essentially three cross linking process are used in the wire and cable industry. Two types of chemical crosslinking, those employing organic peroxides or silanes, respectively, are still frequently used for the improvement of polyethylene and other polymer based insulations for wire and cable. Due to the fact that radiation cross linking occurs in PE at room temperature, make it distinguished with the case of traditional chemical cross linking. On the other hand, many merits of radiation cross linking such as low energy consumption, high line speed and so on; have made it more advantageous over traditional chemical means [1-4]. Polymeric materials used for electrical insulation not only have to exhibit acceptable physical properties but good electrical properties. In the present study attempt has been made to compare electrical properties of high density polyethylene, before and following irradiation.

Results and discussion

Electrical properties of the HDPE samples were studied dynamically. Fig. 1 demonstrates that volume resistance of the samples increase compared to the original value, with standing time. Dielectric constant (permittivity) of an insulating material is defined as the ratio of the charge stored in an insulating material placed between two metallic plates to the charge that can be stored when the insulating material is replaced by air. Simply stated, the dielectric constant indicates the ability of an insulator to store electrical energy, so it is desirable to have the capacitance of the insulating material to be as minimum as possible.

Fig. 1. Volume resistance variations of the samples vs. time.

Fig. 2 shows that after irradiation the dielectric constant reduce very slowly against the time. This may be due to radiation crosslinking induced restrictions, which makes charge storage capability low and consequently dielectric constants of the samples were modified after irradiation.

Fig. 2. Dielectric constant variations of the samples vs. time.

In all electrical applications, it is desirable to keep the electrical losses at a minimum value. The dissipation factor ($\tan\delta$) is a measure of such electrical inefficiency of insulating material. On the other hand, $\tan\delta$ is a means of measuring dipole moment in a dielectric material. Permanent dipoles of a molecule are aligned by an applied electric field. $\tan\delta$ is an electrical characteristic that is related to the chemical structure and degree of crosslinking of polymers. For most polymers $\tan\delta$ decrease as the degree of crosslinking increase, because reactive polar groups are either neutralized or hindered by crosslinking. From the Fig. 3 it can be concluded that $\tan\delta$ of the sample varied greatly with standing time at room temperature.

Fig. 3. Dissipation factor ($\tan\delta$) variations of the samples vs. time.

Fig. 4. Dielectric strength (breakdown voltage) variations of the samples vs. time.

On the other hand, wire and cable are required to withstand over voltages under different conditions, for example, during aging (heat treatment under moist conditions). To evaluate the over voltage performance of the insulation, dielectric break down tests were made. Dielectric breakdown is caused when electrons detached from the molecules acquire sufficient energy in the electric field to yield secondary ionization and avalanche. The dielectric strength of an insulating material is defined, as the maximum voltage required producing a dielectric breakdown. Fig. 4

indicates that the dielectric strength of the samples did not change standing over time. In other words, that the trapped free radicals and also charge trapping induced by EB, did not have considerable influence on dielectric strength.

To interpret the obtained results it can be said that when a semi crystalline polymer like polyethylene is irradiated with high energy EB radiations, the electrons which enter to amorphous regions start to react to the medium with familiar cross linking mechanisms and others enter to the crystalline parts. It is believed that those radicals that are trapped in crystal regions have a relatively long, half life [5]. These radicals are formed in the core of crystalline region and slowly diffuse to the interface with the amorphous region. Consequently, radiation induced conductivity may change along the time. It is well known that electrical conductivity of some polymers increases following irradiation. Directly after irradiation the conductivity does not decrease immediately to the initial level but decays usually exponentially.

The results of EPR spectroscopy analysis is illustrated in Fig. 5. Peak to peak value which is a measure of most intensive free radicals concentration is plotted against time. From the diagram it can be concluded that radical concentration has a time dependent function and its population reduces standing over the time. Some authors found that the dielectric loss tangent is correlated with relaxation of polar groups induced by radiolytic oxidation [6, 7]. Paramagnetic species induced by ionizing radiation in PE are well known to be alkyl, allyl and peroxy radicals [5]. However there is no direct correlation between radicals generated in polyethylene and charge decay. On the other hand charge concentration was not measured by EPR method. Alkyl radicals observed by EPR spectroscopy have insignificant contribution to the distribution of electron density in the material, as well. There are several investigations concerning so called charge trapping event in dielectric and insulating material [8-12]. According to the obtained results concerning the electrical properties, the effects are attributed to charge trapping in different structural defects.

Fig. 5. EPR spectroscopy analysis (peak-to-peak signal value) of the HDPE samples as the free radicals concentration variations vs. time.

Conclusions

In semi crystalline polymers like polyethylene, existence of two parts of crystalline and amorphous regions makes irradiation process more complex with respect to post

irradiation events. From the EPR spectroscopic results, it can be concluded that after irradiation some radicals remained in the polymer and specially trapped in crystalline parts. Some electrical properties of the polymer were affected as a function of time and the others did not. In other words, dielectric constant and dielectric strength remained unchanged but volume resistivity and dissipation factor decreased after irradiation due to effect of remained free radicals, and slowly recovered with the storage time at room temperature. These effects are attributed to charge trapping in different structural defects than the radicals observed by EPR spectroscopy due to its insignificant contribution.

Experimental

Materials

High density polyethylene (HDPE) supplied by Bandar Imam Petrochemical Company HDPE 44005 with density of 0.944 g/cm³ and MFI of 0.55 was used in this investigation. Samples of granular shape were compression molded using Dr. Collin warm press instrument at 165 °C to prepare sheets with 2±0.1 mm thickness.

Irradiation

The samples were irradiated with dose of 300 kGy and at the constant dose rate. The irradiation process was performed using the Rhodotron type electron accelerator machine, TT200 model, using 10 MeV electron beam with a maximum of 8 mA beam current. The maximum penetration depth of 10 MeV electron beam in HDPE is ~5 cm (practical range) therefore, the dose uniformity ratio along the aforementioned sheet thickness can be assumed very close to one.

EPR measurements

The sheet sample was ground using a laboratory scale grinder (Brabender, Germany) in order to obtain powdered form of polyethylene. Resulted sample was placed in EPR tube and inserted into cavity of EPR spectrometer **Bruker** EMS-104. Concentration of free radicals was estimated and compared in relative and measured at room temperature.

Volume Resistance

Volume resistance was measured at room temperature by TERA OHMMETER CEAST Company, made by Italy [13].

Dielectric loss tangent and dielectric constant

A Dielectric Loss measurement system of ANDO ELECTRIC Company, model TRS-10T, made by Japan, was used for determine the dielectric constant, Dielectric loss tangent of the samples. Frequency was tuned at 1 MHz in the experiments [14].

Breakdown voltage

A Dielectric Rigidity system, P/N 6135.053, CEAST Company, made by Italy was used for determining the breakdown voltage of the samples [15].

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