

Radiation Effects on Thermoluminescence Characteristics of HDPE Containing Additives

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1. Introduction

Polymeric materials are widely used for electrical insulation in a broad range of applications that cover the power supply industry to inner and outer space. However, the electrical performance of these materials could be compromised by their working environment and one of the most deleterious is that where a nuclear radiation is present.

Radiation effects on polymers can be interpreted by two main reactions, a cross-linking reaction and degradation reactions or a main-chain scission process. There are no absolute rules for determining whether or not any given polymer will cross-link or degrade upon an irradiation. But, the polymers can be divided empirically into two groups; polymers which are cross-linked by radiation (especially by the incorporation of chemical cross-linking promoters) and polymers which degrade by radiation into a product of lower molecular weight due to random main-chain scission process. These polymers become very hard and brittle with a high dose of radiation [1]. Most polymeric materials contain some stabilizers such as flame retardant and antioxidant to prevent combustion and oxidation. Because of these additives, degradation mechanism of the polymer became complicated.

Many of the novel properties of the insulating materials used in nuclear power plants are important for radiation degradation. Therefore we have used the thermal methods such as thermoluminescence (TL) detection for irradiated high density polyethylene containing flame retardant and antioxidant.

2. Experimental

2.1 Sample preparation

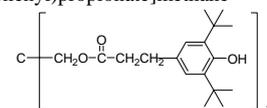
The sample was a high density polyethylene (HDPE), and mixed with various flame retardants and antioxidants. The formulations of the specimen are listed in Table 1. HDPE was blended with flame retardant of 30 phr and antioxidants of 5 phr. Sample was mixed by a Banbury mixer (Moriyama) at 180° C and was molded into a slab of 2 mm thickness by a hot-pressing power at 180° C. Samples were irradiated using a ⁶⁰Co γ -ray and a ray up to 1,000 kGy at a dose rate of 5 kGy per hour in the presence of an air atmosphere.

2.2 Thermoluminescence analyses

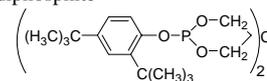
Table 1. Formulations of HDPE specimen containing flame retardant and antioxidant.

Composition Sample	HDPE	Al(OH) ₃	Mg(OH) ₂	Sb ₂ O ₃	1010*	6260**
HDPE		-	-	-	-	-
PA-1		30	-	-	3	-
PA-2		30	-	-	-	3
PM-1	100	-	30	-	3	-
PM-2		-	30	-	-	3
PS-1		-	-	30	3	-
PS-2		-	-	30	-	3

* Songnox 1010: Tetrakis[methylene-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate]methane



** Songnox 6260: bis(2,4-di-tert-butylphenyl)pentaerythritol diphosphite



Thermoluminescence (TL) curves in the presence of air were recorded on a TL Detector (Saint-Gobain Industrial Ceramics Co. Model 2210). The initial temperature was equilibrated to 50°C, and a light emission between 50°C and 300°C was measured with a heating rate of 6°C/sec. Each signal was recorded per 1°C from the light emission which was converted into an electrical discharge. In order to maintain a constant area for the emission surface, each sample was cut to 5 mm diameter.

3. Results

Luminescence detection from external energy has been a useful tool for evaluation of defect and diagnosis of materials. It has been shown [2] that TL in polyethylene held above ambient temperature is due to the de-trapping of the trapped charge carriers and their subsequent recombination with luminescence centers.

TL spectra of irradiated pure HDPE are shown in Figure 1. The TL spectra of γ -ray irradiated HDPE were increased overall, compared with that of the non-irradiated HDPE. For an increasing irradiation dose, the values of the luminescence intensity show an increasing tendency, it is considered that increased conductive ions, gas and carriers due to the chain scissions of HDPE by γ -ray irradiation. It can be explained that radiation cross-linking byproducts are the source of luminescence as in the previous research [3]. The formation of radicals in irradiated polyethylene is the sequence of a

series of events initiated by the absorption of energy from the incident high energy photon or electron. Not always is the electron ejected from the parent molecule recaptured by the polymer molecule to give a radical; it may become a mobile electron, away from its parent molecule, or it may be trapped and released far later. The physical effects include a radiation induced conductivity and TL [4].

Figure 2 shows the results of the integrated TL response as a function of the radiation doses. The values of integrated TL response increased as the irradiation dose increased, especially at 200 kGy. It is well known that cross-linking dominates at low irradiation dose, below 50 kGy, while scission becomes the major process at higher doses [5]. In the figure 2, there is an obvious tendency. Below irradiation doses of 800 kGy, the integrated TL response of all samples increased slowly with irradiation doses. It showed a sharply increase in the specimens containing Songnox 1010 at high irradiation doses. It is considered that the changes of integrated TL value for all specimens are attributed to the modification of the additives, Songnox 1010, by irradiation. As regards to the sample containing additives, flame retardants influenced to the TL characteristics more than antioxidants.

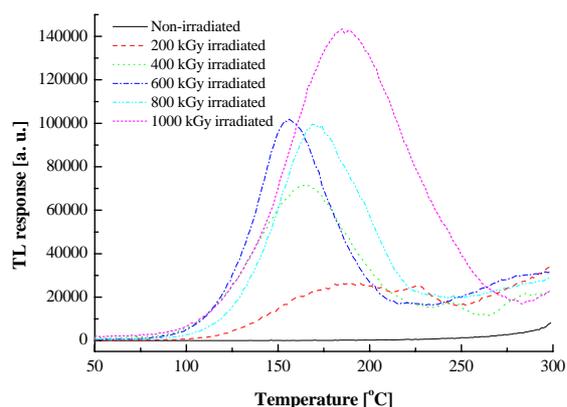


Figure 1. TL spectra of irradiated HDPE sample.

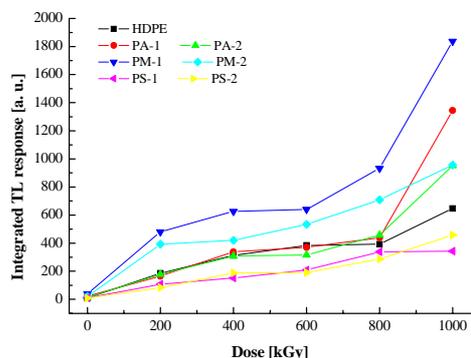


Figure 2. Integrated TL response of irradiated sample between 50°C and 300°C.

4. Conclusion

A thermo-luminescence detection of γ -ray irradiated HDPE containing flame retardant and antioxidant were

carried out for an evaluation of the radiation degradation.

The most effective flame retardant in HDPE for radiation degradation is Antimony trioxide. Songnox 6260 would not decompose at high dose irradiation. Also, the integrated TL response of HDPE without additive showed a linear tendency for an irradiation doses, it could be a useful tool for an evaluation of the radiation degradation of HDPE.

Acknowledgments

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