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# Radiation effects on dielectric properties of ethylene propylene rubber

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**Abstract**

In order to evaluate the radiation degradation of ethylene propylene rubber (EPR), radiation effects on EPR was investigated by using dielectric analysis and thermal-gravimetric analysis. Permittivity, loss factor, tan $\delta$ and thermal decomposition temperature were observed for γ-ray irradiated EPR. As radiation dose increased, the peak temperature of loss factor and tan $\delta$ of EPR increased, and loss factor and tan $\delta$ at peak temperature decreased. Activation energies were calculated using loss factor and thermal decomposition for γ-ray irradiated EPR as well. The trends of both calculated activation energies showed same tendencies as radiation dose was increased.
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*Keywords:* EPR; Radiation; Dielectric analysis; Activation energy

## 1. Introduction

The electrical properties of polymeric insulating materials could be compromised by their working environment, and one of the most deleterious is that maybe exposed to nuclear radiation [1]. Particularly, the electrical failure induced from the breakdown of insulation materials is important in nuclear power generating stations. Therefore, excellent radiation resistance characteristics as well as electrical and mechanical properties must be considered for these materials so that serious accidents can be avoided.

Radiation and thermal degradation under the normal and accident operation of a reactor in the nuclear power generating stations can be the deleterious degradation factors. IEEE quality standards regulate that class 1E electric cable, field splices and connections can endure $^{60}$Co to a dosage of 0.5 MGy under normal conditions, even 1.5 MGy for loss of coolant accident (LOCA) condition [2].

Dielectric tan $\delta$ is widely used for the evaluation of insulation degradation with electrical insulating resistance [3]. In case of an increasing leakage current through the insulation materials, increased dielectric tan $\delta$ would be accompanied, then the temperature inside the materials would increase as increasing applied voltage, so that breakdown strength would be degraded consequently. Because of these phenomena, dielectric tan $\delta$ can cause breakdown in the insulating materials for the long term and increase abruptly by absorption of the moisture. Therefore, diagnosis of dielectric properties could be one of the evaluation methods for degradation of the insulating materials [4]. At the moment, the diagnosis of dielectric properties for degradation complies with IEC standard method 62067-2002, which is measuring only tan $\delta$ values under room temperature and specific frequency. However, these values are sensitive to the measuring temperature and humidity, it is usual to take the conservative values.

In order to evaluate and normalize the degradation level of irradiated EPR, dielectric factors are measured under various temperatures and frequencies, and the temperature showing the variation of the molecular mobility remarkably is determined. Thereafter, degradation level of irradiated EPR was evaluated using dielectric relaxation intensities by means of drawing Cole–Cole's circular arcs. Because dielectric characteristics are related to molecular interaction, and these affect its mechanical properties. The thermal gravimetric analysis and mechanical properties were measured as well.

## 2. Experimental

### 2.1. Sample preparation

In this study, EPR was used as a control specimen. EPR was 50:50 [mol%] composition ratio of ethylene to propylene and provided by JINRO Industries (Korea).

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For the purpose of comparing the dielectric, thermal analyses and mechanical properties as radiation degradation, the samples were irradiated with γ-rays in the presence of air at room temperature, in a $^{60}$Co facility at the Korea Atomic Energy Research Institute. The total doses were 400, 800, 1200, 1600, and 2000 kGy at a dose rate of 5 kGy/h.

### 2.2. Dielectric properties measurement

In the dielectric analysis, the sample is placed in direct contact with the sensor. The electrodes transmit an applied oscillating voltage to the sample and sense the response of the sample from the applied voltage. The optimum sensor arrangement for monitoring the bulk properties of the EPR sheet is the ceramic parallel plate sensor. The parallel plate sensor consists of lower and upper electrodes. The lower electrode is the excitation electrode and contains resistance temperature detector (RTD) to accurately monitor sample temperature. The upper electrode is the response electrode and contains a guard ring to prevent fringing effects. Fig. 1 shows a measurement system to measure dielectric permittivity, loss factor, and tan $\delta$.

Dielectric properties were investigated using Dielectric Analyzer (TA instrument, Model 2970). A sample of EPR sheet (0.2 mm thick) was placed on the ceramic parallel plate sensor, and after purging for 3 min with dry nitrogen gas, the upper ram was lowered to exert 300 N of force on the sample. Data was acquired while heating at a rate of 3 °C/min from −150 to 50 °C and multiplexing frequency (1, 3, 10, 30, 100, 300 Hz and 1, 3, 10, 30, 100 kHz) at a constant voltage of 1 V.

Dielectric analysis offers high sensitivity, a wide frequency range, and the ability to accommodate a wide variety of samples. These qualities permit the characterization of subtle molecular transitions which are not easily distinguishable by other thermal analysis techniques. The energy of activation can be obtain by performing a linear least squares analysis on the plot of ln(frequency) versus $1/T_{max}$, where $T_{max}$ is the temperature which corresponds to the loss factor peak maximum at a certain test frequency. The slop of the resultant plot when multiplied by the gas constant $R$ (=8.314 J/mol) will reveal the energy of activation.

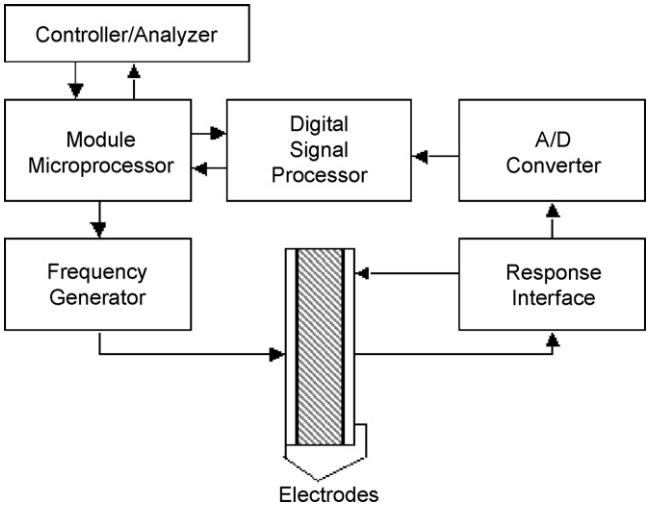


Fig. 1. Block diagram of dielectric parameters measurement system.

### 2.3. Thermogravimetric analysis

Thermogravimetric analysis was performed to confirm the co-relationship between dielectric properties and mechanical properties. Thermogravimetric analysis was carried out with 10 °C/min increased rate for measuring the temperature at 5% weight loss measurement and 10, 25, 50, 75, and 100 °C/min increased rates for thermal decomposition activation energy calculation, in a nitrogen atmosphere using thermogravimetric analyzer (TA instrument, Model 2950).

The Kissinger method has been used in this study to determine the activation energy from plots of the logarithm of the heating rate versus the inverse of the temperature at the maximum reaction rate in constant heating rate experiments [5]. The activation energy can be determined by the Kissinger method without a precise knowledge of the reaction mechanism, using the following equation:

$$\ln\left(\frac{\beta}{T_{inf}^2}\right) = \left\{\ln\frac{AR}{E} + \ln[n(1-\alpha_{inf})^{n-1}]\right\} - \frac{E}{RT_{inf}} \quad (1)$$

where $\beta$ is the heating rate, $T_{inf}$ is the temperature corresponding to the inflection point of the thermal oxidative degradation curves which corresponding to the maximum reaction rate, $A$ is the pre-exponential factor, $\alpha_{inf}$ is the extent of conversion at $T_{inf}$, and $n$ is the reaction order. From a plot of $\ln(\beta/T_{inf}^2)$ versus $1/T_{inf}$ and the fitting to a straight line, the activation energy $E$ can be calculated from the slop of this line. $T_{inf}$ was measured using differential thermogravimetry (DTG) curves at various heating rates.

### 2.4. Mechanical properties

The control and the irradiated EPR samples were subject to mechanical tensile testing following ASTM standard D638 [6]. The tensile properties of the sample at room temperature were evaluated using an Instron universal mechanical tester (Model 1130), after γ-irradiation. A crosshead speed of 100 mm/min and a guage length of 50 mm were used. The specimen load was sensed by a 500 kg capacity Instron type-A load cell. This cell was mechanically calibrated by precision standard weights prior to testing each set of samples. From these experiments, elongation at break of all samples was obtained. Five specimens were tested and the average values were obtained. All tensile tests were run under time mode.

$$\varepsilon = \frac{L - L_0}{L_0} \times 100 \quad (2)$$

where $\varepsilon$ is elongation at break, $L$ is the length at break, and $L_0$ is initial length of the specimen.

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