A Study on the Properties of CSPE According to Accelerated Thermal Aging Years

Jung-Hoon Lee*, Myeong-Kyun Kang*, Jun-Soo Jeon*, Seung-Hoon Lee*, In-Yong Kim*, Hyun-Shin Park* and Yong-Deok Shin[†]

Abstract – The accelerated thermal aging of CSPE (chlorosulfonated polyethylene) was carried out for 40.41, 121.22, and 202.04 days at 100°C, which are equivalent to 20, 60, and 100years of aging at 50°C, respectively. The volume electrical resistivities of the accelerated thermally aged CSPE samples for 0, 40.41, 121.22, and 202.04 days were 1.107×10¹⁴–2.097×10¹⁴, 7.752×10¹³–1.556×10¹⁴, 7.693× 10¹³–1.521×10¹⁴, and 7.380×10¹³–1.304×10¹⁴Ω·cm, respectively, at room temperature. The permittivities of the accelerated thermally aged CSPE samples for 0, 40.41, 121.22, and 202.04 days were 2.89×10⁻¹¹ – 3.65×10⁻¹¹, 3.40×10⁻¹¹ – 3.70×10⁻¹¹, 3.50×10⁻¹¹ – 3.82×10⁻¹¹, and 3.76×10⁻¹¹ – 4.13×10⁻¹¹ F/m, respectively, at room temperature. The EAB (elongation at break) of the accelerated thermally aged CSPE samples for 0, 40.41, 121.22, and 202.04 days were 98.8–101.3, 59.5–60.3, 37.8–39.2, and 41.8–44.3%, respectively, at room temperature. The apparent densities of the accelerated thermally aged CSPE samples for 0, 40.41, 121.22, and 202.04 days were 1.603–1.614, 1.611–1.613, 1.622–1.628, and 1.618–1.620g/cm³, respectively, at room temperature. The measured currents of the accelerated thermally aged CSPE and the standard sample were almost constant after 5 min of applying a 300-V/mm electric field to the CSPE. The V-I slope of the accelerated thermally aged CSPE sample was increased if the applied electric field was increased at room temperature, and the V-I slope of the accelerated thermally aged CSPE was higher than that of standard CSPE.

Keywords: Accelerated thermally aged CSPE, Volume electrical resistivity, Permittivity, EAB, Apparent density, V-I slope

1. Introduction

Recently, we have been reminded that power supplies are very important for NPPs(nuclear power plants). The radiation leaks in the Fukushima NPPs resulted from a blackout caused by an unexpected, large tsunami [1].

Based on data from 2012, nine units among Korea's NPPs (23 units total) have been operating for more than 20 years. Kori unit 1 has operated for longer than the design life (30 years), and Wolsung unit 1 has marked the end of its predicted design life(30 years). Additionally, approximately 80% of the world's NPPs have operated for more than 20 years.

Among aging and degraded apparatus in NPPs, electric power cables are very important for the conveyance of electric power and signals and for the safe operation of equipment. The technological development of the optimum degradation evaluation is constantly improving to predict the correct residual life via suitable condition monitoring. The replacement of NPP cables requires

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considerable time and money and involves great difficulty [2, 3].

Condition monitoring of NPP cables uses an evaluation tool to accurately determine the degree of deterioration of the insulation material or jacket of a cable and to find a suitable replacement time [4-5].

CSPE is obtained via the simultaneous chlorination and chlorosulfonation of polyethylene. Further, CSPE is a polymer that consists of a modified polyethylene backbone with chloro- and sulfonyl chloride side groups. Crosslinking can be achieved with different curing methods (e.g., sulfur, peroxides, and maleimide) to produce a commercial, generic Hypalon rubber [6-7].

CSPE is an important and widely used rubber. Further, CSPE is commonly used as a sheath material in electrical cables that are employed in nuclear power facilities and also used in auto supplies, life-saving equipment, and building materials [8-9].

This study was performed to determine the degree of deterioration of a standard CSPE sample according to accelerated thermal aging years. The physical and electrical properties of the accelerated thermally aged CSPE and a standard sample were evaluated by conducting EAB (elongation at break), apparent density, volume electrical resistivity, permittivity, and V-I measurements.

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2. Experimental Procedure

2.1 Sample preparation

A flat-type CSPE with a thickness of 1mm (Taihan Electric Wire Co., Ltd.) was used as a standard CSPE. The accelerated thermal aging of CSPE was carried out for 0, 40.41, 121.22, and 202.04 days at 100°C, which are equivalent to 0, 20, 60, and 100years of aging at 50°C, and then were designated as standard, 100-1, 100-3, and 100-5, respectively.

2.2 Measurement of volume electrical resistivity

The resistance of a CSPE cable changes according to various parameters, such as the shape and size of the insulating material. However, the volume electrical resistivity does not change and is not affected by these parameters. The volume electrical resistivity of CSPE is measured through a three-terminal guard-ring electrode of a KSM 3015. The three-terminal guard-ring electrode is designed as shown in Fig. 1, consisting of two parallel-plate electrodes in order to apply 500-VDC to CSPE and an added guard-ring electrode to absorb the leakage current. Volume electrical resistivity ρ (Ω ·cm) is expressed as follows:

$$\rho = R \frac{S}{t} = \frac{V}{I} \cdot \frac{S}{t},\tag{1}$$

Where V is the voltage of the digital voltmeter, I is the current of the electrometer, S is the upper electrode area, and t is the thickness of the CSPE.

2.3 Measurement of permittivity

A system used to measure the time constant of CSPE is designed as shown in Fig. 2. The electric capacity, C_s , of CSPE is calculated by using Eq. (2), and then, the permittivity of CSPE is calculated through a parallel-plate capacitor.

$$v = V_0 e^{-t/RC_s} \tag{2}$$

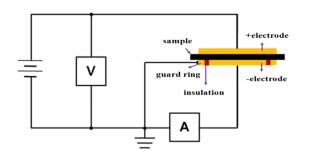


Fig. 1. Three-terminal system for measuring volume electrical resistivity

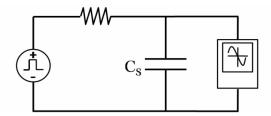


Fig. 2. System used to measure time constant of a specimen

In Eq. (2), V_o is the applied voltage at t=0, v is the discharged voltage of CSPE after t s, and R is 180 k Ω . The electrode diameter used in these experiments is 80mm, and the charging and discharging pulse times are 20ms.

2.4 Measurement of current-time curve

The electrical conductivity of solid-state insulators generates a charging current due to capacitance and absorptive and leakage current due to insulating resistance in DC high-voltage. The charging current disappears when a high DC voltage is applied to solidstate insulators, the absorptive current ends gradually after several minutes, and the leakage current occurs continuously in solid-state insulators [7, 10]. To handle the leakage current consisting of surface leakage and volume electrical conduction, a three-terminal guard-ring electrode is designed as shown in Fig. 1, consisting of two parallel-plate electrodes in order to apply 500- VDC to the CSPE and an added guard-ring electrode to absorb the surface leakage current. A measurement system for the current-time curve of the CSPE is designed in order to apply 300-VDC/mm to the CSPE. The current was measured for 15 min after the 300-VDC/mm electric field was applied to the CSPE.

2.5 Measurement of V-I curve

The volume electrical conductive current of CSPE was measured in the range from 50 to 500-VDC, and a measurement was conducted after 90s in order to remove the charging and absorptive current when the voltage applied to the CSPE increased incrementally from 50 to 500-VDC.

2.6 Measurement of EAB

The EAB of each CSPE was measured according to the ASTM (American Society for Testing and Materials) standard D412.

2.7 Measurement of apparent density

The apparent density of each CSPE specimen was measured 10 times by employing the Archimedes method.

3. Experimental Results and Discussion

3.1 Volume electrical resistivity

As shown in Fig. 3, the volume electrical resistivities of standard, 100-1, 100-3, and 100-5 were 1.503×10^{14} , 1.154×10^{14} , 1.088×10^{14} , $1.005\times10^{14}\Omega$ cm at room tem-perature, respectively. The volume electrical resistivities of the accelerated thermally aged CSPEs were lower than that of standard CSPE and decreased with the accelerated thermally aging time. It is certain that the ionic (electron or hole) leakage current was increased by the separation of the branch chain of CSPE polymer from the main chain of polyethylene as a result of the thermal stress of accelerated thermal aging.

3.2 Permittivity

As shown in Fig. 4, the permittivities of standard, 100-1, 100-3, and 100-5 were 3.41×10⁻¹¹, 3.56×10⁻¹¹, 3.71×10⁻¹¹, and 3.91×10⁻¹¹ F/m, respectively, at room temperature. The permittivities of the accelerated thermally aged CSPE samples were higher than that of standard CSPE and increased with the accelerated thermally aging time. It is understood that the orientation polarization of the accelerated thermally aged CSPE was generated by permanent dipole moments because of the separation of the polyethylene branch chain. Branch chains of CSPE

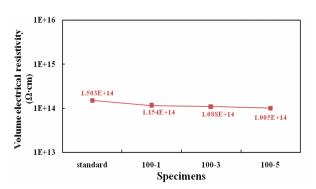


Fig. 3. Volume electrical resistivity of CSPE according to accelerated thermal aging time

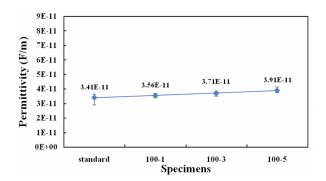


Fig. 4. Permittivity of CSPE according to accelerated thermal aging time

polymer were separated from main chains of polyethylene or destroyed by radiation exposure, and the capacitance of the CSPE polymer was increased according to the amount of radiation exposure in reference [11].

3.3 Current-time curve

As shown in Fig. 5, the measured current of the 100-5 sample was higher than that of the standard CSPE. The measured current of the 100-5 sample and standard CSPE ranged from 0.971 to 0.853nA and from 0.636 to 0.576nA, respectively, which were almost constant, after 5 min when a 300-VDC/mm electric field was applied to the CSPE. It is understood that the measured current of the 100-5 sample and the standard CSPE mean volume electrical conductive current of CSPE, respectively, in the range from 5 min to 15 min after a 300-VDC/mm electric field was applied to CSPE. It is certain that the volume electrical conductive current of CSPE is dependent on the ionic (electron or hole) leakage current caused by the thermal stress of accelerated thermal aging. The measured current of the 100-5 sample and standard CSPE included the absorptive current due to insulating resistance from the start to 5 min after the 300-VDC/mm electric field was applied to the CSPE.

As shown in Fig. 6, the absorptive current versus time of the 100-5sample and standard CSPE is similar to the measured current versus time, and the absorptive current of the 100-5sample was higher than that of the standard CSPE.

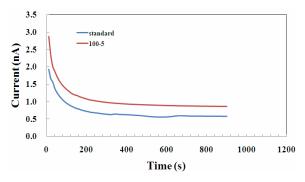


Fig. 5. Measured current of CSPE versus time after application of 300-VDC/mm electric field

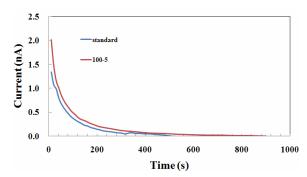


Fig. 6. Absorptive current of CSPE versus time after application of 300-VDC/mm electric field

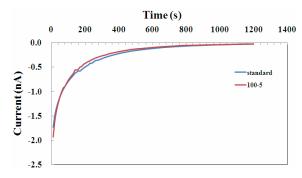


Fig. 7. Discharged current of CSPE versus time after removal of electric field

The ionic (electron or hole) leakage current was increased by the thermal stress of accelerated thermal aging. The absorptive currents of the 100-5 sample and standard CSPE were almost constant, respectively, after 5 min when a 300-VDC/mm electric field was applied to the CSPE.

As shown in Fig. 7, the discharged current versus time of the 100-5sample and standard CSPE were measured when electric field was cut off after a 300-VDC/mm electric field was applied to the CSPE for 15 min. It is understood that the discharged current of the 100-5 sample(1.938-1.119nA) was higher than that of the standard CSPE(1.733-1.113nA) from the start to 50s because the permittivity of the 100-5 sample was higher than that of the standard CSPE. However, it is understood that the discharged current of the 100-5 sample(1.018-0.053nA) was lower than that of the standard CSPE (1.029-0.054nA) in the range from 60 to 900s because the volume electrical resistivity of the 100-5 sample was lower than that of the standard CSPE.

3.4 V-I curve

Fig. 8 shows the measured current versus the electric field of the 100-5sample and the standard CSPE, and the measured current increased with the electric field. The measured current of the 100-5 sample was higher than that of the standard CSPE from the 100-VDC/mm electric field to the end of the test. As shown in Fig. 9, the V-I slope of the 100-5 sample and the standard CSPE is calculated by using Eq. (3), and the V-I slope of those samples increased with the electric field. The V-I slope of the 100-5 sample was higher than that of the standard CSPE from a 150-VDC/mm electric field to the end of the test. The volume electrical conductive current of the CSPE was dependent on the ionic (electron or hole) leakage current caused by the thermal stress of accelerated thermal aging.

$$V-I \ slope = \frac{increased \ current-initial \ current}{increased \ electric \ field-initial \ electric \ field}$$

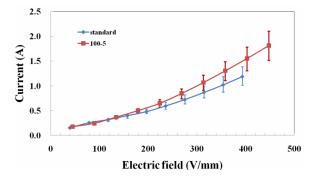


Fig. 8. Measured current of CSPE versus electric field

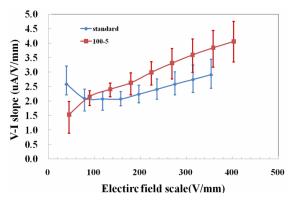


Fig. 9. V-I slope of CSPE versus electric field scale

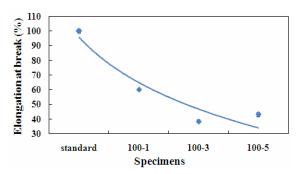


Fig. 10. EAB of CSPE according to accelerated thermal aging time

3.5 Elongation at break

Fig. 10 shows the EAB of the 100-1, 100-3, and 100-5samples and standard CSPE, which decreased until 60 years of accelerated thermal aging. The EAB of CSPE decreased as the accelerated thermal aging years increased because the percent elongation of the CSPE was dependent on the thermosetting degree. Thermosetting polymers become permanently hard when heat is applied to CSPE and do not soften upon subsequent heating.

3.6 Apparent density

Fig. 11 shows the apparent density of the 100-1, 100-3, and 100-5 samples and the standard CSPE, which slightly

(3)

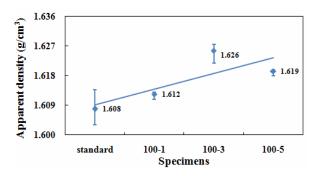


Fig. 11. Apparent density of CSPE according to accelerated thermal aging time

increased over accelerated thermal aging of up to 60 years. The apparent density of CSPE increased with accelerated thermal aging because the apparent density of CSPE was dependent on the thermosetting degree.

4. Conclusions

The physical and electrical properties used as evaluation tools to determine the degree of deterioration of the accelerated thermally aged CSPE were as follows:

- The volume electrical resistivity and permittivity of the accelerated thermally aged CSPE decreased and increased according to the accelerated thermally aging time.
- The measured current of the accelerated thermally aged CSPE and standard sample were almost constant, respectively, after 5 min when a 300-VDC/mm electric field was applied to the CSPE.
- 3. The discharged current of the accelerated thermally aged CSPE was higher than that of the standard sample in the range from the start to 50 s but lower than that of the standard sample in the range from 60 to 900 s.
- 4. The measured current versus the electric field of the accelerated thermally aged CSPE and standard sample increased with the electric field. The V-I slope of the accelerated thermally aged CSPE was higher than that of the standard sample.
- 5. The EAB and apparent density of the accelerated thermally aged CSPE decreased and increased according to the number of accelerated thermal aging years.

The volume electrical resistivity, permittivity, and V-I slope of CSPE suggest evaluating the possibility of the aging state of accelerated thermally aged CSPE based on the EAB and apparent density. The volume electrical resistivity, permittivity, and V-I slope could be used as evaluative tools to determine the degree of deterioration of the insulation material or jacket of a cable and find a suitable replacement time.

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