

# ANALYSIS OF STRUCTURAL CHANGES AND DIELECTRIC PROPERTIES UNDER THERMAL AGING OF CROSS-LINKED POLYETHYLENE USED AS POWER CABLES INSULATION

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## ABSTRACT

Cross-linked polyethylene polymer insulation is widely used as a raw material for insulation of cables. Polyethylene with added peroxide and silane is mostly used. Here, the mechanism of the change of the dielectric loss angle tangent was studied in the case of applying different voltages (in the range of 2-12 kV) to polyethylene samples with added silane and peroxide subjected to heat aging. Combined, i.e., the effect of both thermal aging and applied voltage on the tangent of the dielectric loss angle was investigated during measurements in the frequency range of 50-400 Hz. In the obtained results, it was observed that the value of the dielectric loss angle exceeds the effect of voltage, and the reasons for this increase are mentioned in the article. When we increase the aging period, peaks in the dielectric losses also appeared in the last periods. Also, in the article, the structural change of the insulation was studied by means of Fourier transform infrared spectra (FTIR).

**Keywords:** dielectric loss angle, polymer insulation, XLPE, cross-linking, thermal aging, Fourier transform infrared spectra

## INTRODUCTION

Reliable and durable insulation materials are required for power transmission in electrical networks. Polyethylene, polyvinyl chloride, polystyrene, etc. materials included in the group of thermoplastic polymers meet this demand, and for this reason, they are widely used as insulating materials in various fields of industry. The fact that they are mainly used as insulating materials in high voltage installations is associated with their high electrical properties as well as mechanical, thermal and chemical properties. Polymers and polymer composites also play an indispensable role as insulating materials in the cable industry. Polyethylene (PE), Polyvinyl Chloride (PVC) is mainly used in low and medium voltages, while polyethylene (XLPE) is used in high voltages.(1)

It is well known that XLPE insulation has excellent electrical characteristics as an insulation material in the cable industry. However, these properties may change due to various factors. This leads to a decrease in the efficiency of the energy system and the occurrence of accidents. Therefore, the investigation of a number of insulation and coating materials, electrophysical properties, especially with accelerated aging tests ( $\epsilon$ ,  $\rho$ ,  $\text{tg}\delta$ ,  $\sigma$ , IZI), and determination of their application, which are widely used in the cable industry such as XLPE insulation, are very relevant issues. Extensive research is being done in this field.

XLPE cable condition assessment has always been an important research point in the field of high voltages. For this, great efforts have been made by scientists from all over the world and they have proposed various methods of condition assessment, such as temperature measurement, partial discharge detection, dielectric response method and dielectric loss angle ( $\tan \delta$ ) [2-6].

In a study conducted by M.G Niasar and his team, oil-impregnated insulation paper material aged up to 31 days at 160 °C. Dielectric properties after aging of the insulating material were evaluated by partial discharge measurements and instantaneous breakdown measurements taken at a constant voltage applied until the material was collapsed. The results showed that the number and size of partial discharges increased in thermally aged paper. However, during partial discharge measurements, it was found that the material's ability to withstand partial discharges was longer after thermal aging. This is associated with the expulsion of moisture in the structure of the material during thermal aging. There was no significant difference in electrical strength values [7].

In another study, oil-impregnated craft papers were divided into 3 groups, unaged, aged at 90°C, 750 hours and 1500 hours, and their electrical strength and dielectric parameters (dielectric loss factor and dielectric permittivity) were evaluated. Although the electrical strength tended to increase during the first 750-hour aging periods, it remained higher than its initial value, although it decreased as the aging process continued. The values of the dielectric loss angle ( $\tan\delta$ ) showed a decrease until the 750h hour of aging and remained stable thereafter [8].

In a study by Yudi Saputra and his team, 10% and 20% micro boron nitride particles were added to the epoxy material. Pure epoxy, 10% micro BN added epoxy and 20% micro BN added epoxy insulation were subjected to thermal aging at 150°C for 500 hours. During thermal aging,  $\tan\delta$ , electrical strength and relative dielectric permittivity of 3 types of materials were measured and the results were compared. When the  $\tan\delta$  results during thermal aging were compared, pure epoxide was found to have the highest values. However, while the loss angle of pure epoxy tended to decrease with thermal aging, the dielectric loss angles of doped epoxides remained almost constant during thermal aging [9].

In their study, Wang et al. aged kraft paper and polycarbonate (PC) material at 90°C, 110°C, and 130°C for different durations at a maximum of 300 days for each temperature, while simultaneously applying an electric field of 3 kV/mm. Dielectric spectroscopy method (frequency and temperature dependent  $\tan\delta$  measurement) was applied to investigate the effects of applied electro-thermal wear. The obtained results showed that  $\tan\delta$  values of unaged and aged polycarbonate samples at 3 different temperatures were lower than kraft paper under the same conditions. Furthermore, increasing the aging temperature led to higher  $\tan\delta$  values for both materials [10].

In the research of H.Abdallah et al., along with thermal aging, electric aging was also performed and the cross-linked polyethylene was aged for up to 25 hours under an electric field of 5 kV/mm. First, changes in dielectric loss angle values at 5 and 10 kHz were observed against aging. While no significant change was observed up to 20 h, a large increase occurred at 20 h, and in the next 5 h, the loss angle decreased again to a value close to the initial value due to aging. In this process, it was observed that the loss factor is higher at low frequency. With aging, the surface resistance decreased from  $10^{16} \Omega$  to  $4.10^{12} \Omega$ . There was no significant change in volume resistance [11].

The conductivity of XLPE insulation materials is highly dependent on the presence of polar molecules to promote charge transfer. Pure PE is non-polar, and polar molecules are often formed from additives and peroxide degradation products. The presence, content, and distribution of polar molecules, as well as changes in the structure and chemical composition of the XLPE insulation during thermal aging, can be detected by infrared Fourier spectra (FTIR) [12].

In this article, it is shown how the dielectric losses and the tangent of the dielectric loss angle change after thermal aging of polyethylene insulating materials with added silane and peroxide.

## DATA AND METHODOLOGY

First of all, it should be noted that all the performed works were carried out in the laboratories located at Yıldız Technical University.

### *A. Research object*

As a research object, round samples obtained by hot pressing method from granular polyethylene with organic peroxide and silane compounds were taken. XLPE 0.6/1 kV (Kalpana India KI XL-03) and XLPE 6-35 kV (Borealis 0592) raw materials are used in the production of low and medium voltage cables at the GokNur Baku cable factory. These additives are non-ionic in nature and do not affect the electrical conductivity of the base polymer.

### *B. Preparation of samples*

Sampling was achieved in a temperature-controlled, 80-ton hydraulic press (Figure 1). Taking samples from granular material was carried out by keeping them at a temperature of 130<sup>0</sup> C under a pressure of 15 MPa for 10 minutes (hot pressing method) and cooling them sharply in an ice-water mixture (0<sup>0</sup> C). The samples taken for the experiment have a thickness of (1.5 ± 0.01) mm and a diameter of 25 mm.

### *C. Measurement methodology*

The presented insulation materials were thermally aged for a total of 450 hours, with 6 periods of 75 hours at 120 °C using the air circulation industrial oven shown in Figure 2.

Dielectric loss measurements of insulating materials under thermal aging were performed using an Omicron CP100/CPTD1 measuring device (Figure 1). The CPC100/CPTD1 device performs dielectric loss, tan $\delta$ , quality factor, inductance, capacitance, impedance and phase angle, current measurements at frequencies between 50-400 Hz and voltage 2-12 kV. Only tan $\delta$  values are considered here.

The structural change of the samples was determined by Fourier transform infrared (FTIR) spectroscopy device (Figure 4). With this device, it is possible to test solid, liquid and gaseous substances with high resolution (up to 0.5 cm<sup>-1</sup>). The measuring range of the device is between (4000 cm<sup>-1</sup>- 650 cm<sup>-1</sup>).



**Figure 1. 80-ton hydraulic press: 4 kW power, 400mm\*400mm plate, heatable up to 200 degrees, time control, PLC control panel.**



**Figure 2. Industrial oven used for thermal aging**



**Figure 3. Omicron CPC 100/CPTD1**



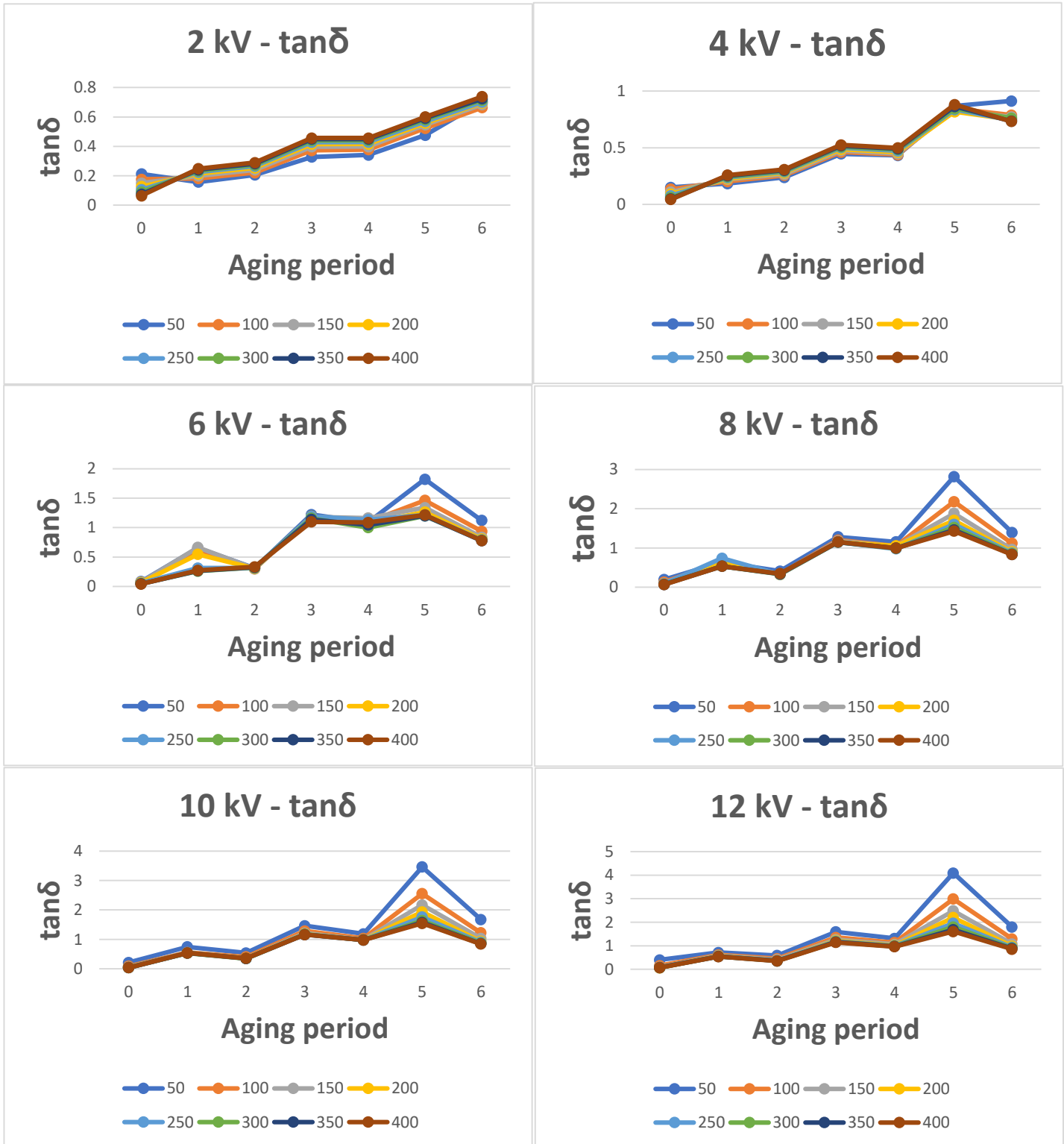
**Figure 4. Perkin Elmer FTIR device**

## **RESULTS**

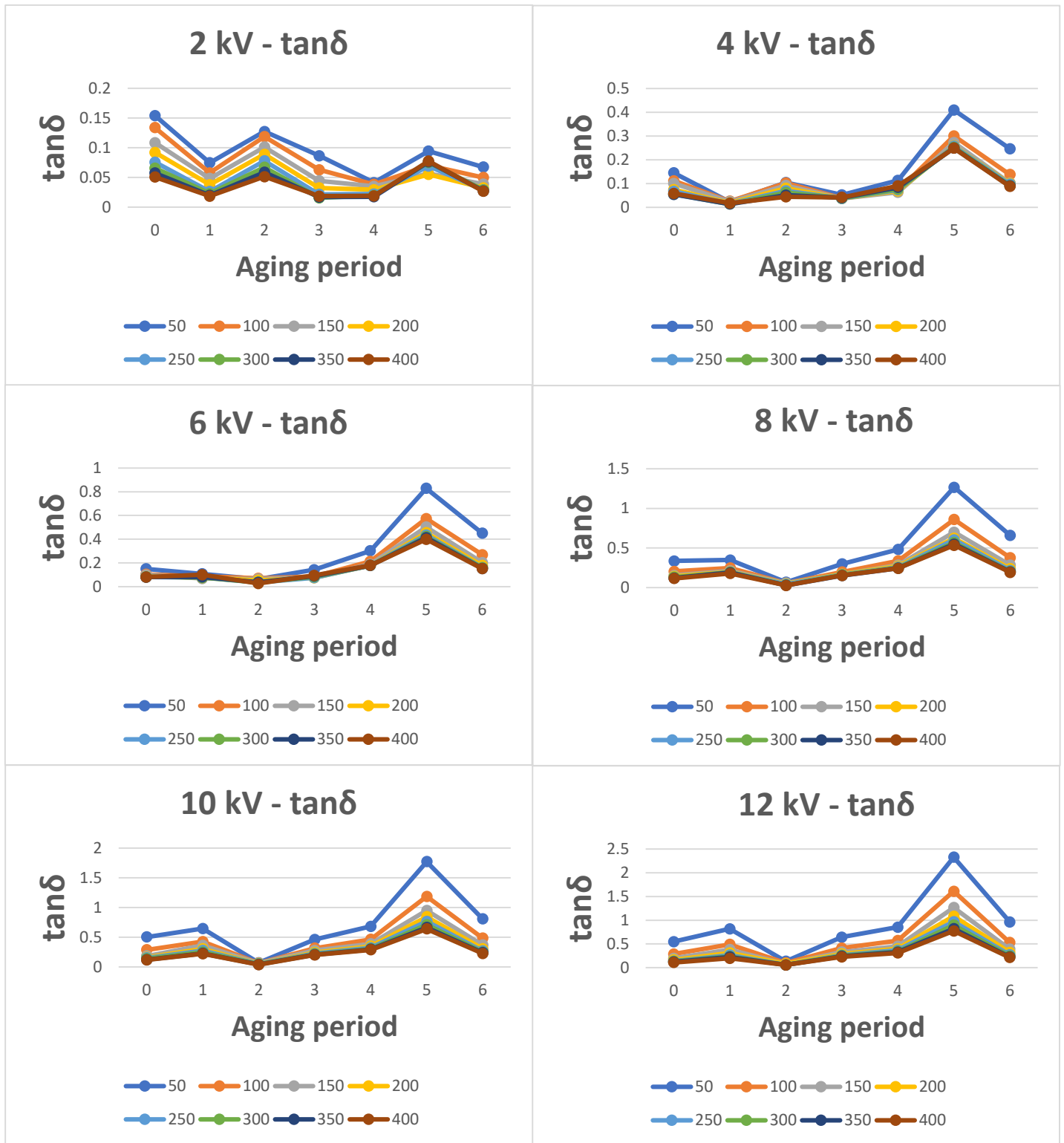
After each thermal period, dielectric loss measurements were made using the same cylindrical electrode with a diameter of 80 mm. Loss measurements of the samples were made between 2 and 12 kV at 2 kV intervals. At each applied voltage, the frequency was increased from 50 Hz to 400 Hz in 50 Hz intervals. Thus, a total of 48 measurements were obtained. This process was applied to 3 samples with the same characteristics and the average value of the 3 samples was taken as the test result.

Figures 5 and 6 show the results obtained from measurements for polyethylene materials with peroxide and silane addition, respectively XLPE 35 kV and XLPE 0.6 kV.

a) *tanδ analysis of samples*



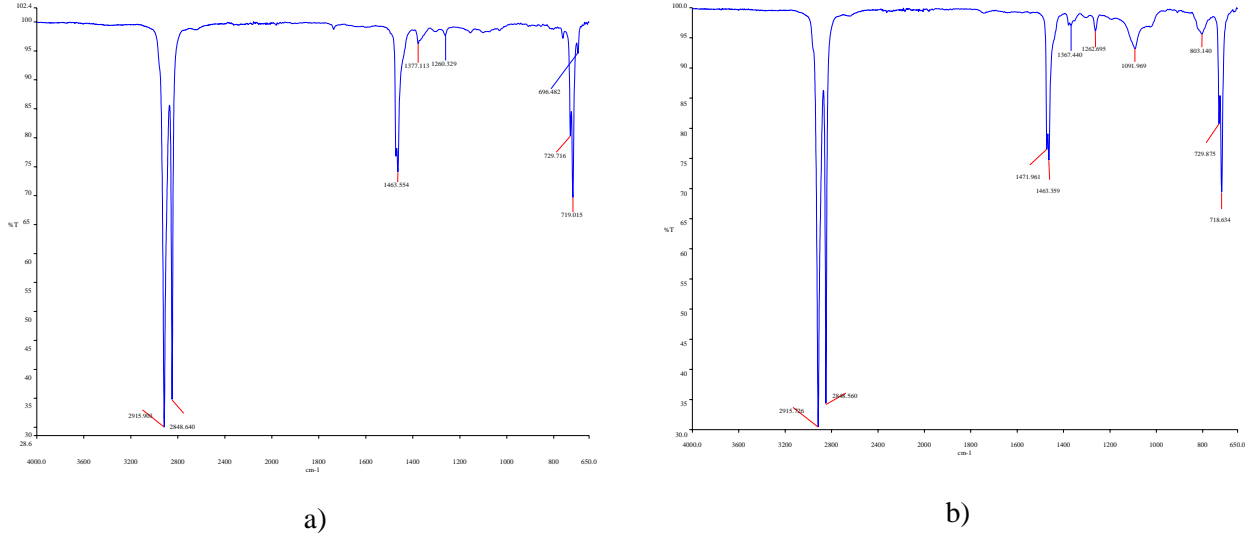
**Figure 5. Dependences of the dielectric loss angle of the XLPE 35 kV peroxide-added polyethylene sample on aging time at different voltages**



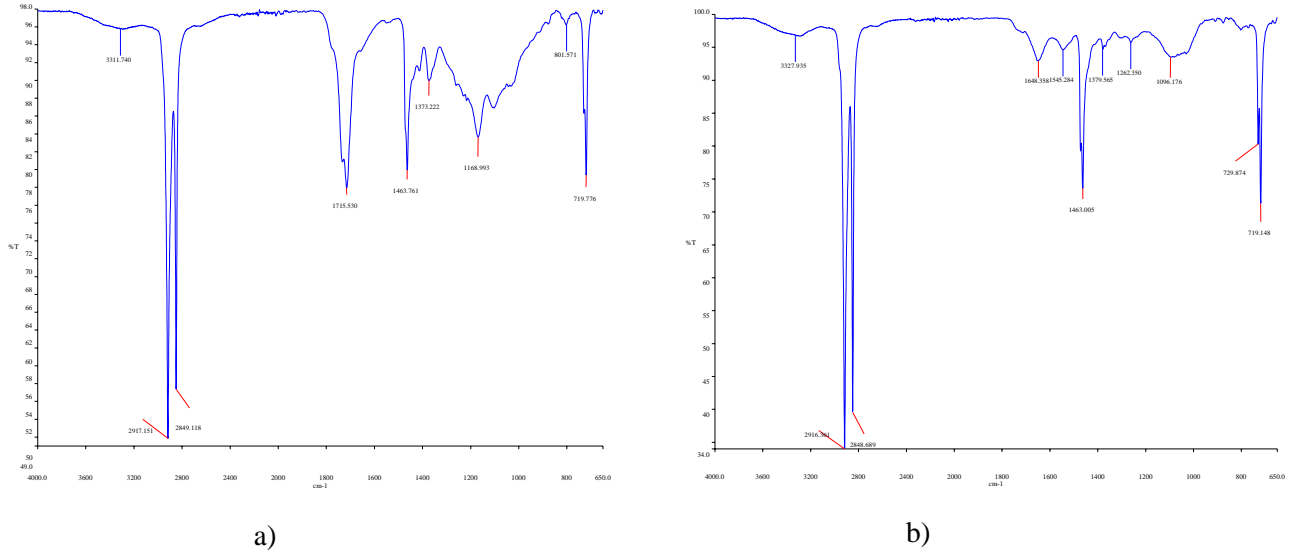
**Figure 6. Dependences of the dielectric loss angle of the XLPE 0.6 kV silane-added polyethylene sample on aging time at different voltages**

b) *FTIR analysis of samples*

The FTIR curves of the samples before and after aging are shown in Figures 7 and 8 for polyethylene insulating materials with peroxide and silane addition, respectively.



**Figure 7 FTIR spectrum of polyethylene insulation added with peroxide (a) and silane (b) before aging**



**Figure 8 FTIR spectrum of polyethylene insulation added with peroxide (a) and silane (b) after aging**

## DISCUSSION

For both materials, the values of the dielectric loss angle appear to increase over the thermal aging period. The main reason for this is the chemical reactions that occur in the dielectric during the thermal aging period. The value of the tangent of the dielectric loss angle was  $4.1 \times 10^{-2}$  for peroxide-added polyethylene and  $2.5 \times 10^{-2}$  for silane-added polyethylene. Although the polarity of the Si-O bond is higher than the polarity of the C-C bond, since the temperature of the samples was  $120^{\circ}\text{C}$ , the destruction of peroxide occurred inside the material. For this reason, dielectric losses were higher in peroxide polyethylene. As can be seen from the graphs, the value of the dielectric loss angle is larger at 50 Hz for both materials.

From Figures 5 and 6, it can be seen that the dielectric loss angle increases as the value of the applied voltage increases with thermal aging. Almost at the end of the aging period, sharp differences are observed at different frequencies after about 6 kV. Peaks at higher voltages were observed in the 5th period of aging in both materials. These peaks were caused by relaxation phenomena as a result of the movement of macromolecular chains of the polymer.

FTIR results - In general, no significant changes were observed in the materials after aging. The peaks at  $2915\text{ cm}^{-1}$  and  $2848\text{ cm}^{-1}$  are due to asymmetric and symmetric stretching vibrations of  $\text{CH}_2$  methylene groups.  $719$  and  $729\text{ cm}^{-1}$  are simultaneously due to the rocking vibrations of the  $\text{CH}_2$  bond [13,14]. Peaks of  $1471$  and  $1463\text{ cm}^{-1}$  were observed as a result of wag vibration of the  $\text{CH}_2$  functional group [15].  $1377\text{ cm}^{-1}$  reflects the symmetric deformation of  $\text{CH}_3$  methyl group [16]. The first one at  $1715\text{ cm}^{-1}$  shows the absorption of carboxylic acid and ketones [ $\text{RC}(=\text{O})\text{OH}$ ,  $\text{RC}(=\text{O})\text{R}'$ ], where R and R' represent the chain of the aged polymer and its fragment [17,18]. The second band at  $1737\text{ cm}^{-1}$  characterizes the absorption of aldehyde or ether [ $\text{RC}(=\text{O})\text{OR}'$ ]. Moreover, the absorption at  $1168\text{ cm}^{-1}$  is suggested to be related to vibrations of the -C-O-C- bond. Finally, in the region between  $3000$  and  $3500\text{ cm}^{-1}$ , a slight increase in the absorption of the -OH function of hydroxyl groups can be observed after aging [17,18]. The silane group has absorption peaks at  $1091$  and  $1096\text{ cm}^{-1}$  due to the stretching vibrations of the Si-O-C bond. There is another peak at  $1262\text{ cm}^{-1}$  associated with the Si- $\text{CH}_2$  functional group.

## CONCLUSIONS

Dielectric losses show an increase in thermally aged samples. The reasons for this are the chemical reactions occurring inside the dielectric. The value of the tangent of the dielectric loss angle at the frequency of 50 Hz was higher. When we increase the value of the applied voltage, it can be seen that the value of the tangent of the dielectric loss angle increases at higher voltages. It was observed that the dielectric losses are higher due to the combined effect of thermal wear and applied voltage.

The main changes in FTIR spectra obtained as a result of thermooxidative reactions after thermal aging are the formation of C=O carbonyl groups. At the same time, it was observed that O-H originates from hydroxyl groups.

## REFERENCES

1. Yao Zhou, Simin Peng, Jun Hu and Jinliang He. Polymeric Insulation Materials for HVDC Cables: Development, Challenges and Future Perspective. IEEE Transactions on Dielectrics and Electrical Insulation Vol. 24, No. 3; June 2017. DOI:10.1109/TDEI.2017.006205
2. A.K. Pradhan, B. Chatterjee, D. Dey, S. Chakravorti, Time growing frequency sweep signal based insulation condition monitoring in frequency domain spectroscopy, IEEE Trans. Dielectrics Electrical Insulation 23 (2016) 1898–1906.



3. S.L. Zhao, K. Zhou, M. He, M. Xie, F.Z. Zhang, Dielectric response characteristics and insulation condition evaluation under impulse voltage for cables, *High Voltage Eng.* 45 (2019) 1297–1304.
4. B. Pang, B. Zhu, X.L. Wei, S. Wang, R.H. Li, On-line Monitoring Method for Long Distance Power Cable Insulation, *IEEE Trans. Dielectrics Electrical Insulation* 23 (2016) 70–76.
5. D. Kim, Y. Cho, S.M. Kim, A Study on Three Dimensional assessment of the aging condition of polymeric medium voltage cables applying Very Low Frequency (VLF) tan delta Diagnostic, *IEEE Trans. Dielectrics Electrical Insulation* 21 (2014) 940–947.
6. Y. Yang, D.M. Hepburn, C.K. Zhou, W.J. Zhou, W. Jiang, Z. Tian, On-line monitoring and analysis of the dielectric loss in cross-bonded HV cable system, *Electr. Power Syst Res.* 149 (2017) 89–101.
7. M. G. Niasar, N. Taylor, P. Janus, X. Wang, H. Edin, and R. C. Kiiza, “Partial discharges in a cavity embedded in oil-impregnated paper: Effect of electrical and thermal aging,” *IEEE Trans. Dielectr. Electr. Insul.*, vol. 22, no.2, pp. 1071–1079, 2015
8. I. Kolcunova, M. Pavlik, and L. Liso, “Assessment of long thermal ageing on the oil-paper insulation,” *Adv. Electr. Electron. Eng.*, vol. 14, no. 5, pp. 506–511, 2016,
9. Y. Saputra, M. Kim, S. Suwarno, Y. Jeon, and Y. Byeon, “The Effect of Thermal Aging on Dielectric Properties and Tracking Erosion Test of Micro Bn Composites,” *Proc. 2nd Int. Conf. High Volt. Eng. Power Syst. Towar.Sustain. Reliab. Power Deliv. ICHVEPS 2019*, 2019.
10. W. Wang, K. Yang, C. Yue, S. Chen, and D. He, “Study on aging characteristics of oil-immersed paper and polymer material based on dielectric loss,” *Annu. Rep. - Conf. Electr. Insul. Dielectr. Phenomena, CEIDP*, pp. 882–885, 2012
11. A. Hedir, M. Moudoud, O. Jbara, S. Rondot, and F. Slimani, “Degradation Mechanisms of Cross-linked Polyethylene Insulation by Thermal and Electrical Aging,” *11 Conférence la Société Français d’Electrostatique*, no. June 2019, 2018.
12. J. V. Gulmine and L. Akcelrud, “FTIR characterization of aged XLPE,” *Polym. Test.*, vol. 25, pp. 932-942, 2006.
13. A. M. Nobrega, M. L. B. Martinez et A. A. A. de Queiroz, “Investigation and analysis of electrical aging of XLPE insulation for medium voltage covered conductors manufactured in Brazil,” *IEEE Trans. Dielectr. Electr. Insul.*, vol. 20, no. 2, pp. 628–640, Apr. 2013.
14. Boudou L, Guastavino J, Physico-chemical observations on polyethylene base resin under the effect of thermal ageing, *J Phys D Appl Phys* , 35, 1-4, 2002.
15. Dalal SB, Gorur RS, Aging of distribution cables in service and its simulation in the laboratory, *IEEE Trans Dielec Elec Insu*, 12, 139-146, 2005
16. Aljoumaa, K.; Ajjji, Z. J. *Radioanal. Nucl. Chem.* 2015,311, 355.
17. C. Stancu, P. V. Notingher, P. Notingher et M. Lungulescu, “Space charge and electric field in thermally aged multilayer joints model,” *IEEE Trans. Dielectr. Electr. Insul.*, vol. 23, no. 2, pp. 633–644, Apr. 2016.
18. B. Ouyang, H. Li, X. Zhang, S. Wang and J. Li, “The role of microstructure changes on space charge distribution of XLPE during thermooxidative aging,” *IEEE Trans. Dielectr. Electr. Insul.*, vol. 24, no. 6, pp. 3849–3859, Dec. 2017.