The Influence of Octaphenyl POSS Addition on the Electro-aging Characteristics of Polypropylene

Xiaosi Lin[1, 2], Chuanjie Lin[5], Wah Hoon Siew[2], John Liggat[3], Martin Given[2], Jinliang He[4]

[1] State Grid Fujian Technical Training Centre, State Grid Fujian Electric Power Co., Ltd., 362013, China (Since Jul. 2022)

[2]Department of Electronic and Electrical Engineering, University of Strathclyde, G1 1XW, UK

[3]Department of Pure and Applied Chemistry, University of Strathclyde, G1 1XL, UK

[4]State Key Laboratory of Power System, Department of Electrical Engineering, Tsinghua University, 362013, China

[5] State Grid Fuzhou Power Supply Company, State Grid Fujian Electric Power Co., Ltd., 350004, China

Abstract: Due to the good electrical performance and high thermal stability, there is a tendency to develop the new generation of polymeric insulation materials based on polypropylene (PP). Compared with XLPE, PP is a thermoplastic polymer which can be recycled after use. However, the tertiary carbon structure on the side chain makes PP easier to be oxidized and degraded under high temperatures and high electric field. Nanocomposite technology can improve the electrical properties of polypropylene and reduce the damage of polypropylene under high electric fields. In this paper, the effects of octaphenyl polyhedral oligomeric silsesquioxane (OpPOSS) nanofillers on the electro-aging characteristics of PP are reported. Measurement techniques including thermally stimulated depolarization current (TSDC); scanning electron microscope (SEM); breakdown strength tests and conductivity tests, were used to understand the mechanisms by which the addition of OpPOSS influences the aging characteristics of PP. The lifespan of PP and OpPOSS/PP nanocomposites have been estimated by use of the Weibull distribution. As a results of this research work, the addition of OpPOSS nanofiller could improve the long-term performance of PP for HVDC applications.

Keywords: electrical performance, thermal stability, polypropylene, HVDC insulation, electro-thermal aging, lifespan.

I. INTRODUCTION

As one of the commercial thermoplastic polymers, PP has become one of the favourite choices for polymeric insulation materials, because of its advantages of low cost, excellent electrical properties, good thermal stability and low dielectric loss [1-4]. During last few decades, many scientific reports demonstrated the nanocomposites technology can be utilized to enhance the electrical performance of PP's nanocomposites, including DC breakdown strength, space charge characteristics, dielectrics properties, etc [1-4].

From research papers [5-8], many kinds of nanofillers can be used to improve the electrical performance of polymers, such as MgO, SiO_2, Al_2O_3, et al. However, Q. Zhong, et al. indicated that the improvement of electrical properties of polymers is inversely proportional to the particle size of nanofillers [9], and that as a consequence agglomeration problem would be a challenge in the manufacture of polymeric nanocomposites.

Compared with inorganic nanofillers, POSS has attracted attention due to its special structure. The chemical bonds of Si-O-Si build the main cage of POSS and each corner is connected with a side-group. As shown in Fig. 1, the main cage of OpPOSS can perform as a nano-silica to improve the electrical properties of polymers [10] while the eight phenyl groups connected to the corners introduce the polar group which could improve the trapping characteristics of polymers.

In this paper, OpPOSS/PP nanocomposites were subjected to electrothermal ageing under an electric field of 30 kV/mm and a temperature of 110°C. The electrical properties of aged samples were tested after 5, 10, 15, 20, 25 and 30 days. It was found that the addition of OpPOSS/PP could enhance the aging characteristics and operational lifespan of PP.

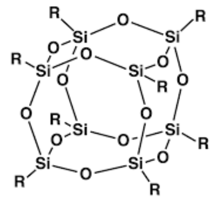

Figure 1. The chemical structure of OpPOSS molecule

II. MATERIALS AND EXPERIMENTS

A. Materials

The matrix material, isotactic polypropylene (iPP) and the nanofiller, OpPOSS were purchased from Aladdin Industrial Inc., China. The density of iPP and OpPOSS nanofillers are 0.92 g/cm^3 and 1.25 g/cm^3 respectively. Xylene used was also provided from Aladdin Industrial Inc., China

B. Nanocomposites preparation

The pellets of PP and 2.0 phr OpPOSS nanofiller were dispersed and blended in xylene at a temperature of 120 °C. After even mixing was achieved, the solution of

OpPOSS/PP/xylene were dried at 100°C in the drying box. The resulting 2.0 phr OpPOSS/PP nanocomposites powder was then placed in moulds and pressed under 15 MPa and 200°C for 15 min, then cooled down to room temperature under the pressure of 10 MPa within 5 min. The samples were then placed in a vacuum oven at 60 °C for 2 hours to remove the moisture on the surface of samples This allowed the manufacture of nanocomposite samples with thickness of 100 and 200 μm.

C. Electro-thermal aging process design

The system used to electrothermally stress the 100 and 200 μm. samples of PP and its nanocomposites is shown in Fig. 2. The ageing was carried out in a vacuum oven. In a practical DC cable, the maximum operational temperature and DC electric field would be limited t0 70 °C and 15 kV/mm respectively. In this research, the temperature of electro-thermal aging process was set to 110°C and the electric field was adjusted to 30 kV/mm in order to accelerate the aging process of PP and nanocomposites. In addition, there were no antioxidants in the manufactured samples while in a practical cable antioxidants are added into insulation layer to delay the aging process of the insulation materials.

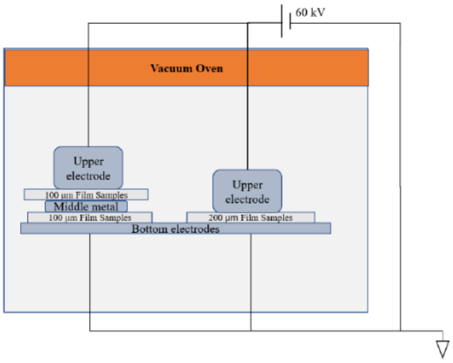

Figure 2. The experimental design of electro-thermal aging process for PP and its nanocomposites

The upper electrode block, bottom electrode board and middle metal sheet are all made of 304 stainless steel. There are arc chamfers on the upper electrode and the middle metal sheet so that the electric field is distributed uniformly in the samples. The total thickness of the samples being aged is around 200 μm. Therefore, an electric field of 30 kV/mm was applied when the power source provided 60 kV DC voltage to the samples. The vacuum was set to 0.01 Pa in the vacuum oven. Samples were taken out for electrical tests every 5 days.

D. The experimental design for lifespan estimation

The DC system shown in Fig. 3 was designed to measure the time to failure of aged PP and OpPOSS/PP nanocomposites under a field of 60 kV/mm, a temperature of 110 °C a pressure of 0.01 Pa. The duration time until the breakdown occurred was recorded to estimate the lifespan of samples.

III. Experimental Results and Analysis

A. Thermally stimulated depolarized current (TSDC) result

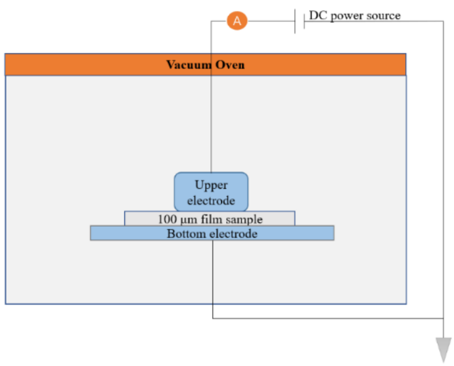

Figure 3. The experimental design for the lifespan estimation of PP and its nanocomposites.

The TSDC curves of PP and its nanocomposites have been given in Fig. 4. In PP and its nanocomposites, the TSDC curves have a thermal stimulation current peak at - 5 °C, which is due to the glass transition point of PP. The TSDC curve of PP shows a peak of thermally stimulated current at 64 °C which is corresponding to the trapping level of 0.8 eV. Compared with PP, the TSDC curve of OpPOSS/PP nanocomposites demonstrated two peak values of 1.79 and 0.95 pA at 98 °C and 128 °C respectively, which means the addition of OpPOSS introduced many deep traps with the trapping levels of 1.02 and 1.1 eV to improve the trapping characteristics of PP [12].

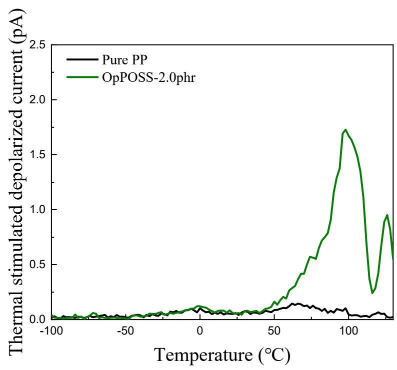

Figure 4. The TSDC results of PP and OpPOSS/PP nanocomposites

B. SEM observation

The surface microstructure of PP and 2.0 phr OpPOSS/PP nanocomposites shown in Fig. 5 and Fig. 6. after being subjected to electro-thermal aging process for differing times days were observed using SEM techniques. Cracks of different severity appeared on the surface of the aged samples after electro-thermal aging process. The surface roughness of PP increased with the increase of aging time. After 15 days of electro-thermal aging obvious cavities began to appear, and the deterioration of PP surface began to become more marked with the increase of time. Although the cracks on the surface of OpPOSS/PP nanocomposites became more serious with the increase of electro-thermal aging time, the surface roughness and deterioration degree were lower than that of PP. Because PP and OpPOSS/PP nanocomposites were aged in a vacuum oven, and the upper and lower surfaces of the samples are covered by electrodes, the damage of the samples mainly came from electrical stress rather than through reactions the oxidation. Therefore, the results showed that the electrical damage of PP in the aging process is decreased by the addition of OpPOSS.

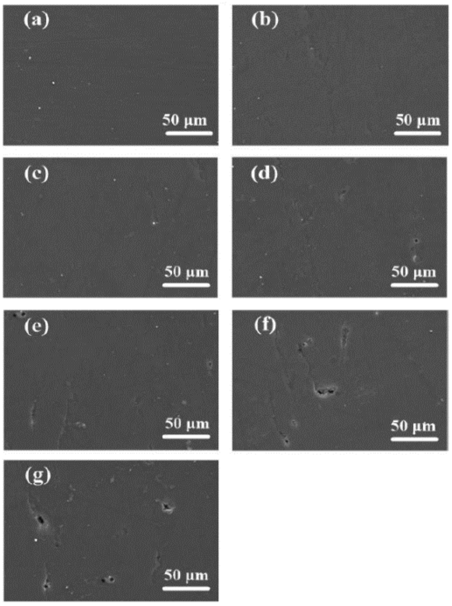

Figure 5. The aged PP films by the aging process after 0 (a), 5 (b), 10 (c), 15 (d), 20 (e), 25 (f) and 30 (g).

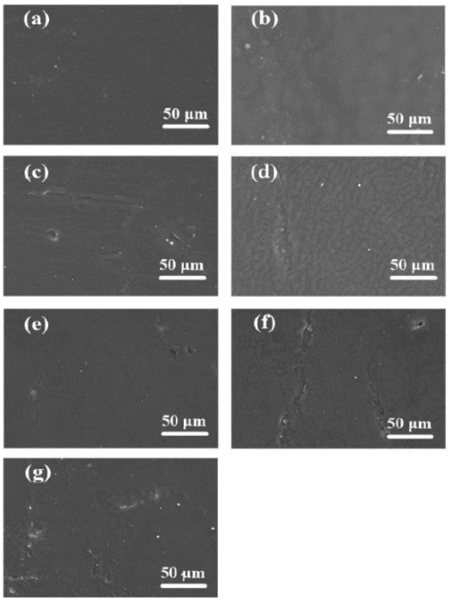

Figure 6. The aged OpPOSS/PP nanocomposites films by the aging process after 0 (a), 5 (b), 10 (c), 15 (d), 20 (e), 25 (f) and 30 (g)

C. DC breakdown strength

The DC breakdown strength of aged PP and OpPOSS/PP nanocomposites after the electro-thermal aging process are shown in Fig. 7 by fitting to a 2-parameter Weibull distribution and the Weibull parameters are shown in Fig. 8 [13]. The critical value E_0 and the shaping factor β of breakdown strength were used to describe the breakdown strength of samples with the breakdown probability of 63.2% and the dispersion of breakdown strength respectively.

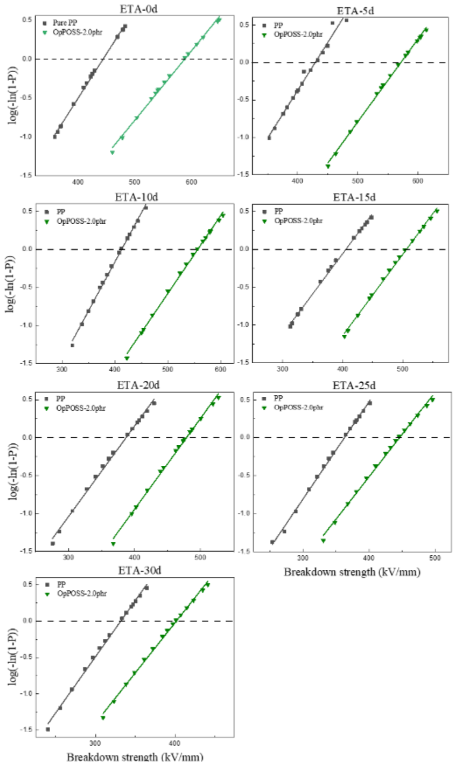

Figure 7. The DC breakdown strength of aged PP and OpPOSS/PP nanocomposites

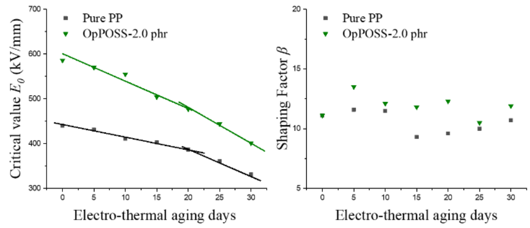

Figure 8. The parameters of DC breakdown strength of aged PP and OpPOSS/PP nanocomposites

From Fig. 7 and Fig. 8, the DC breakdown strength of PP was decreased continuously with the increase of electro-thermal aging time. When the electro-thermal aging time was 30 days, the DC breakdown strength of PP decreased from 440 kV/mm to 331 kV/mm, with a decrease of 24.8 %. While the critical value of DC breakdown field strength of OpPOSS/PP nanocomposites decreased from 586 kV/mm to 401 kV/mm, with a decrease of 31.6 %. With the electro-thermal aging process, the effect of OpPOSS on the DC breakdown strength of PP decreased. The OpPOSS/PP can still improve the breakdown strength of PP from 331 kV/mm to 401 kV/mm by the increasing rate of 21.1 %.

D. DC Conductivity

The DC conductivity measurement was described in [11]. Fig. 9 shows the DC conductive current of PP and OpPOSS/PP nanocomposites as a function of electric field increases during the electro-thermal aging process, which means that the resistivity is decreased with the increase of aging time. When the aging time was 30 days, the conductivity of OpPOSS/PP was still to 22.6 % of that of pure PP. Although OpPOSS nanocomposites would be degraded by the electro-thermal aging, the deep trap introduced by OpPOSS could capture the homocharges injection from the electrode and further inhibit the charge transport in PP. Therefore, the electrical damage of PP in the aging process was reduced and the coupling between OpPOSS and PP was protected.

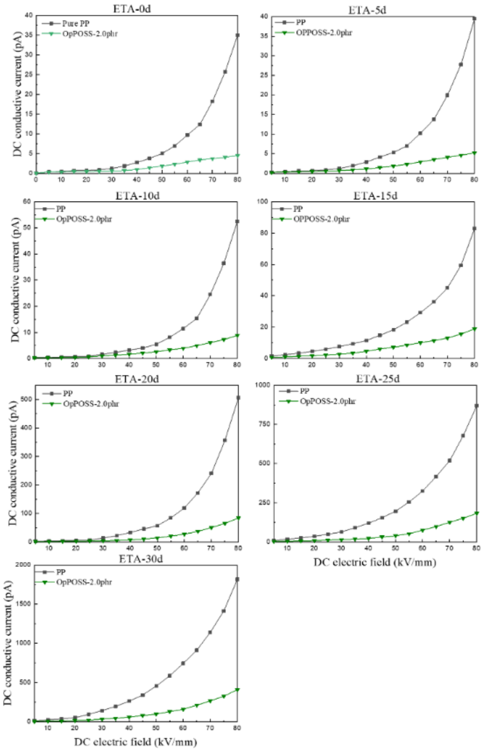

Figure 9. The DC conductivity of aged PP and OpPOSS/PP nanocomposites

E. *The lifespan estimation of samples*

The lifespan calculation method is described as follows [13]. The Weibull distribution is generally used to describe the breakdown strength of materials. Under the condition of time t and the electric field E, the breakdown probability is

$$P(t,E) = 1 - \exp\left(-C\int_0^t \left(kt^{k-1}E^{\lambda}\right)dt\right) \quad (1)$$

Where k is a shaping factor, λ is the distribution factor and C is the constant about the temperature.

When the applied electric field is constant,

$$P(t,E) = 1 - \exp\left(-Ct^kE^{\lambda}\right) \quad (2)$$

$$tE^n = c \quad (3)$$

Where c is a constant and $n = \lambda/k$

Assume The lifespan of PP and OpPOSS/PP to be t_0.

$$\frac{t}{t_0} = (\frac{E_0}{E})^n \quad (4)$$

Table 1. The breakdown time of samples under 60 kV/mm and 110°C

Samples	PP	2.0 phr OpPOSS/PP
Breakdown time (h)	4.21	7.70

In this part, the value of n is set to 9. Therefore, according to Tab. 1, the lifespans of PP and 2.0 phr OpPOSS/PP nanocomposites under15 kV/mm and 110°C are estimated to 126.0 and 230.4 years respectively.

IV. CONCLUSION

The addition of OpPOSS nanofillers is aimed to improve the long-term performance of the base PP insulation material. The improvements to electro-thermal aging characteristics have been investigated. The results indicated the introduction of deep traps could capture the mobile charges then build the potential barrier so that the charge injection and charge transportation could be suppressed. Therefore, the electrical damage would be reduced during the operation, finally the lifespan of PP could be enhanced.

V. ACKNOWLEDGE

This work is supported in part by the National Key R&D Program of China (grant 2018YFE0200100) and National Natural Science Foundation of China under Grant 51921005 and the funding from University of Strathclyde.

VI. REFERENCE

[1] T. Tanaka, "Dielectric nanocomposites with insulating properties", *IEEE Trans. Dielectr. Electr. Insul.*, 12(5), pp. 914-928, October. 2005.
[2] Y. Zhou, B. Dang, H. Wang, J. Liu, Q. Li et al., "Polypropylene-based ternary nanocomposites for recyclable high-voltage direct-current cable insulation," *Composites Science and Technology*, 165, pp. 168-174, 2018.
[3] Y. Cao, P. C. Irwin and K. Younsi, "The future of nanodielectrics in the electrical power industry," *IEEE Trans. Dielectr. Electr. Insul*., 11(5), pp. 797-807, October 2004.
[4] K. Lau, A. Vaughan, G. Chen, I.. Hosier and A. Holt, "On interfaces and the DC breakdown performance of polyethylene/silica nanocomposites," *2014 IEEE Conference on Electrical Insulation and Dielectric Phenomena (CEIDP)*, Des Moines, IA, 2014, pp. 679-682.
[5] S. Hu, Y. Zhou, C. Yuan, W. Wang, J. Hu, et al, "Surface-modification effect of MgO nanoparticles on the electrical properties of polypropylene nanocomposite", *IET High Volt*, 5(3), pp. 249-255, June, 2020.
[6] M. Gao, J. Yang, H. Zhao, M. Hu and S. Xie, "Study on Electric Tree of Nano-SiO2Polypropylene Composite Insulation Material," *2019 2nd International Conference on Electrical Materials and Power Equipment (ICEMPE)*, pp. 301-304, 2019.
[7] X. Duan, WH. Siew, M. Given, J. Liggat and J. He, "Effect of different surface treatment agents on the physical chemistry and electrical properties of polyethylene nano-alumina nanocomposites," *IET High Volt*, 5(4), pp. 397-402, Aug. 2020.
[8] Y. Zhou, J. Hu, B. Dang and J. He, "Titanium oxide nanoparticle increases shallow traps to suppress space charge accumulation in polypropylene dielectrics," *RSC adv.*, 6(54), pp. 48720-48727, May. 2016.
[9] Q. Zhong, L. Lan, J. Wu, et al., "Effect of nano-filler grain size on space charge behavior in LDPE /MgO nanocomposite", *Proceedings of 2014 International Symposium on Electrical Insulating Materials*, pp. 429-432, 2014
[10] S. Peng, B. Dang, Y. Zhou, J. Hu and J. He, "Functionalized TiO2 nanoparticles tune the aggregation structure and trapping property of polyethylene nanocomposites," *J. Phys. Chem. C*, 120(43), pp. 24754–24761, 2016
[11] X. Lin, WH. Siew, J. Liggat, M. Martin and J. He, "Octavinyl polyhedral oligomeric silsesquioxane on tailoring the DC electrical characteristics of polypropylene," *IET High Volt*, 7(1), pp. 137-146, Sept. 2021.
[12] F. Tian, W. Bu, L. Shi, C. Yang, Y. Wang et al., "Theory of modified thermally stimulated current and direct determination of trap level distribution," *J. Electrostat*., 69(1), pp. 7-10, Feb. 2011.
[13] Electric strength of insulating materials - Test methods - Part 2: Additional requirements for tests using direct voltage, IEC Standard 60243-2, 2013-11-26.
[14] G. Montanari, G. Mazzanti and L. Simoni, "Progress in electrothermal life modeling of electrical insulation during the last decades," in *IEEE Trans Dielectr Electr Insul,* 9(5), pp. 730-745, Oct. 2002.