# Space Charge and Breakdown Strength Behaviour of PP/POE/MgO Nanocomposites

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***Abstract*- Polypropylene (PP) is considered a competitive candidate to replace widely used cross-linked polyethene (XLPE) in cable material application, as it can potentially withstand higher operating temperature and has excellent electrical properties. Besides, it is a more environment-friendly material, since PP does not require crosslinking and, therefore, can be easily recycled at the end of its life. This paper investigated the influence on the tensile strength, space charge and breakdown behaviours of the PP by adding polyolefin elastomer (POE) and Magnesium Oxide (MgO) nanoparticles. 10 or 20 wt. % of POE and 5 or 10 wt. % of nano-MgO were introduced into PP. Results show that the PP with 10 wt. % of PE02 and 5 wt. % of surface treated-MgO has the highest breakdown strength among all systems. Also, the introduction of POE and nano-MgO can significantly improve the mechanical flexibility. In combination with the observed suppression of space charge accumulation, which shows that PP/POE/MgO nanocomposites are a viable option for high voltage (HV) cable insulation material in the future**.

***Keywords- nanocomposites, polypropylene, magnesium oxide, silane coupling agent, space charge, breakdown strength, tensile strength***

## I. INTRODUCTION

As an important developing direction of smart grids, HV technology lays the groundwork for the ongoing trend of long distance, ultra-high voltage direct current transmission [1]. Cables play an important role in HV transmission, and electrical insulation is a crucial factor in limiting cable development [2]. Cross-linked polyethene (XLPE), the current state of the art in cable insulation material, has some limitation in terms of dielectric and thermal characteristics, as well as the lack of recyclability. Even in “ultra-pure” XLPE, impurities produced during the industrial production process is inevitable [2, 3]. Hence, PP as a recyclable and eco-friendly material was coming into the view of researchers. Compared to PE, PP has comparable electrical properties such as electrical breakdown strength, but better performance on inhibiting space charge and a higher melting temperature, which is more than 160 ℃ [2]. Due to its thermoplastic nature, PP can be recycled and reshaped [3]. However, poor mechanical performance and low thermal conductivity limit its development in HV cable applications [4].

Previous studies have shown that PP/POE blends can enhance the mechanical flexibility, enabling PP to become a potential insulation material for HV cables. However, the decrease in electrical breakdown performance and the issue of space charge accumulation still limit its development [4].

Based on [4], a small number of nano-fillers such as nano-MgO in PP may suppress the space charge accumulation. It is assumed that the accumulation of space charge may be influenced at the polymer-particle interphase via surface treatment of nanoparticles. In this paper, different concentrations of POE and MgO were introduced into PP. Breakdown strength, mechanical properties as well as the potential suppression of space charge accumulation were investigated, especially the role of surface treatment.

## II. EXPERIMENTAL

### *A. Materials*

Isotactic polypropylene (iPP, average $M_w$~340,000), Poly(propylene-co-ethylene) (PE02, 2 % of ethylene content) are obtained from Sigma-Aldrich. Nano-MgO (4810NH, 20 nm) was purchased from SkySpring Nanomaterials Inc. Triethoxy (octyl) silane (C8-E, 440213) from Sigma-Aldrich was used for surface functionalisation of nano-MgO.

### *B. Surface Treatment of Nano-MgO*

A method used for surface functionalisation from previous work [5] was applied and a brief procedure is listed below. The amount of silane coupling agent (SCA) was selected based on the ratio of nanoparticles to SCA used in [6] and 1.57 ml of C8-E for 2 g of nano-MgO was applied (details are listed in Table I). Nano-MgO/C8-E/toluene mixture was stirred at 110.6 ℃ for 4 hours in a dry nitrogen environment. The functionalised nano-MgO was extracted by a washing procedure for three times where 300 ml of toluene in total was used. During washing, a pallet formed by nano-MgO will form at the bottom of the centrifuging tube after a 10 min centrifuging at 8000 rpm. A sonication bath was used to help to break the nano-MgO pallet for next washing. IPA was added after last washing, a surface functionalised nano-MgO/IPA suspension was obtained at the end.

### *C. Preparation of PP/PE02/MgO Nanocomposites*

An improved blending procedure based on previous work [5, 7] was used. Samples were prepared with around 5 g weight for each dielectric system. The required amount of PP and PE02 were weighted by a digital balance and placed in a 250 ml breaker, then 50 ml of xylene was added. The mixture was heated and stirred at 140 ℃ until all polymer dissolved. For untreated nano-MgO, the required amount of nano-MgO was added into 10 ml IPA and then sonicated for 30 min with a probe sonicator to form suspensions, while surface functionalised nano-MgO/IPA suspensions were prepared during surface functionalisation procedure.

When the polymer/xylene mixture was removed from heat and cooled to about 100 ℃, the nano-MgO/IPA mixture was added. The mixture was then mixed by a stirring rod until it cannot be stirred. The resulting blends were cut into pieces and placed in a fume cupboard for 48 h. After the solvent was dried out, the blends were pressed into sheets and placed in a

TABLE I

Details of SCA in Reference and SCA Used in This work

| Silane Coupling Agent | Physical structure | Molar mass (g/mol) | Density (g/ml) | Amount used (ml) |
|---|---|---|---|---|
| Trimethoxyl(octyl)silane (reference) [6] | $CH_3(CH_2)_7Si(OCH_3)_3$ | 234.4 | 0.907 | 7 (10g $SiO_2$) |
| Triethoxy(octyl) silane | $CH_3(CH_2)_7Si(OC_2H_5)_3$ | 276.49 | 1.382 | 1.57 (2g MgO) |

vacuum oven at 70 □ for 3 days to evaporate the solvent residual. In the end, the blends were pressed into different samples for testing and stored under vacuum.

Considering pure PP as a reference, a total of 9 dielectric systems were prepared. Systems without surface treatment were marked as *e.g.* 'PP10-5', which is representative of the nanocomposites containing 10 wt. % of PE02 and 5 wt. % of untreated nano-MgO, while 'PPS20-10' is representative of the nanocomposites containing 20 wt. % of PE02 and 10 wt. % of surface treated nano-MgO.

### *D. Characterisations*

AC and DC breakdown tests were conducted on the AC/DC breakdown kit according to ASTM standard D149-87 at room temperature. The thickness of samples is 85 ± 5 µm for all tested systems. For both AC and DC tests, the ramp-up rate was set to 200 V/s. All samples were inserted between two ball electrodes (6.3 mm in diameter) and immersed in silicone oil. In total, 20 valid test results were used per sample type for Weibull analysis.

Pulsed electro-acoustic (PEA) method was employed in this experiment, which transforms the acoustic signal to electrical signals and then represents the digital signal in the scope. In this paper, disc-shaped samples of 3 cm diameter and 120 ± 10 µm thickness were used. For the measurement, a voltage of 600 V with a pulse width of 10 ns was applied to generate the acoustic wave and a minimum of 20 kV/mm electrical DC field was applied. The space charge distribution curve was recorded during the 1-hour charging and 1-hour discharging period.

For tensile tests, Tinius Olsen H25KS was used. Rectangle samples in the size of 3 cm*1 cm and 250 ± 20 µm thickness were made. They were nipped between the upper and lower grip with a gap of 25 mm. Then samples were stretched at a constant rate of 10 mm/min until the break.

## III. Results and Discussion

### *A. AC/DC Breakdown Behaviours*

The breakdown strength data of each sample was analysed by the 2-Parameter Weibull distribution and the results are shown in Fig.1 for AC and Fig.2 for DC. The scale parameters ($E_0$) and the shape parameters (β) can be also found in Fig. 1 and Fig. 2.

Although the PP filled with 10 % of PE02 and 5 wt. % of C8-treated MgO shows a slight increase in DC breakdown

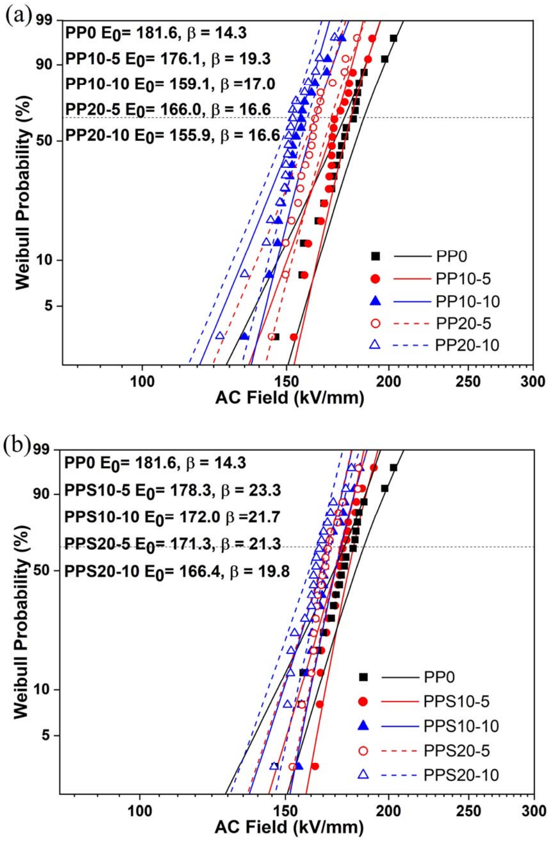

Fig. 1. AC breakdown strength of (a) PP/PE02/untreated nano-MgO systems, (b) PP/PE02/surface treated nano-MgO systems

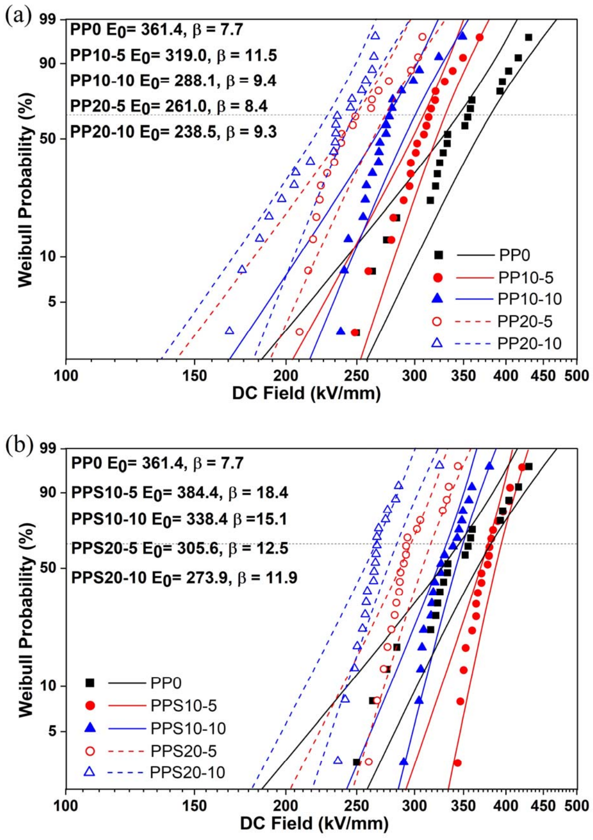

Fig. 2. DC breakdown strength of (a) PP/PE02/untreated nano-MgO systems, (b) PP/PE02/surface treated nano-MgO systems

strength when compared with the unfilled PP, the results suggested that the introduction of PE02 and nano-MgO will lead to the overall reduction of AC and DC breakdown strength. This reduction can be related to the introduction of PE02, the phase separation between the different polymers leads to decreased breakdown strength [8]. Also, the comparatively high filler loading and resulting particle agglomeration can lead to the deteriorated breakdown strength. Many works have shown that the highest breakdown strength of nanocomposites will usually occur at lower filler loading levels, typically about 1 to 3 wt. % [4, 9]. Zhou *et al.* suggest the increased DC breakdown strength is closely related to the suppressed space charge accumulation under DC voltage [4]. This is consistent with the space charge results shown in this paper, the homo charge near the cathode can be effectively suppressed with the help of the surface functionalisation. It is obvious that all samples filled with C8-treated MgO have higher AC and DC breakdown strength than their counterparts filled with non-treated MgO. This can be attributed to the increased compatibility between filler and matrix with the help of the grafted organic layer on the MgO surface [5].

### B. *Space Charge Behaviour*

Fig. 3 shows space charge distributions for PP, PP10-5 and PPS10-5. For pure PP, an obvious hetero charge accumulation can be observed near the cathode. With the increase in the charging time, the peak decreases continuously. This could result from the charge injection near the cathode. However, this phenomenon cannot be observed on PP filled with PE02 and nano-MgO shows in Fig. 3 (b) and (c). Many researchers suggested that the MgO nanoparticles can act as 'traps' and suppress the charge injection and the accumulation of space charge [9-11]. The bandgap of nano-MgO is 7.8 eV, which can be considered as 'deep traps' embedded in polymeric insulation material [9, 11]. The introduction of nano-MgO can increase the number and depth of these 'traps', consequently suppressing the space charge accumulation near the electrodes. Results of nanocomposites with higher content of nano-MgO and PE02 were omitted where this inhibitory effect on space charge accumulation is decreased.

As can be observed in Fig. 3 (b) and (c), the nanoparticles with surface treatment have a better inhibition effect on the space charge accumulation during the charging process. The charge injection rate in samples with C8-E treated nano-MgO is lower than in specimens with untreated nano-MgO. This result indicated the grafted organic layer may lead to the increased trap depth of nanoparticles or larger interfacial region that related to the change of the particle surface chemistry [12, 13].

### C. *Tensile Test*

Stress-strain curves were used to analyse the mechanical properties of each system and the results are shown in Fig. 4. With the PE02 content increasing, a significant enhancement of sample strain and decreased yield strength can be observed in most cases. As shown in Table II, a reduction of Young's modulus after introducing PE02 from 0 wt. % to 10 wt. % and 20 wt. % can be observed. This can be attributed to the addition PE02 which normally shows a better tensile strength than PP.

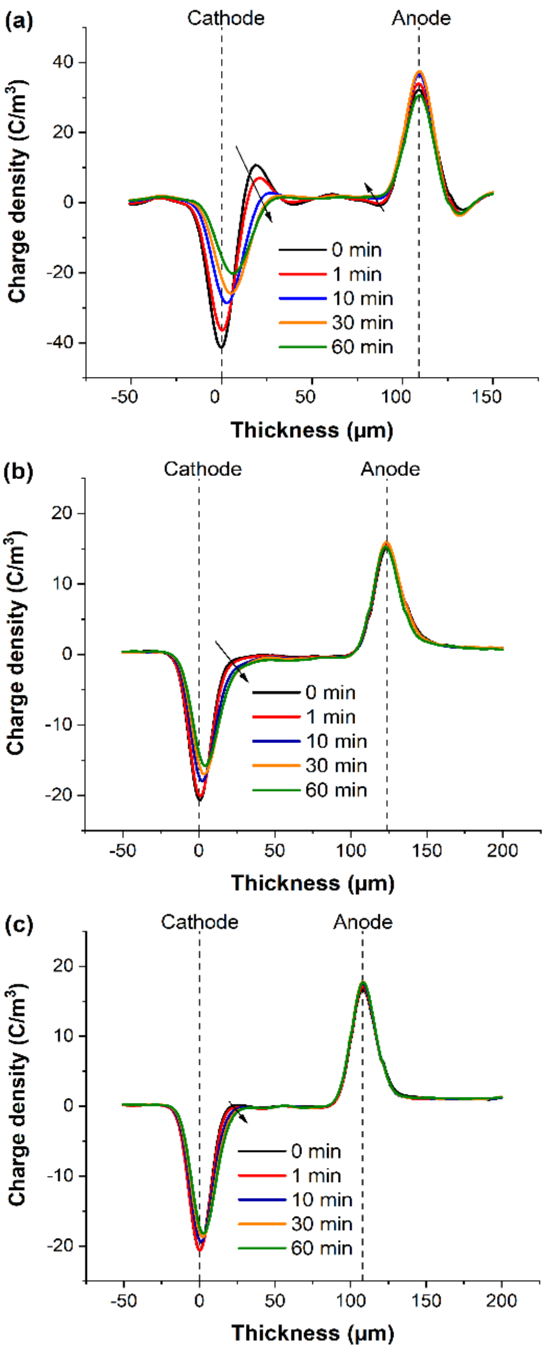

Fig. 3. Space charge behaviour of (a) Pure PP, (b) PP10-5, (c) PPS10-5 system

When focusing on the effect of surface treatment, it seems that the introduction of nano-MgO with or without surface treatment didn't have a significant effect on the stain, yield point and Young's modulus of each system.

It worth to mention that some samples, such as PP10-10 and PPS20-10, show inconsistent results. This might be related to the cooling process during the sample preparation procedure cannot be controlled precisely and result in the inconsistent thermal history of different systems. Further work related to differential scanning calorimetry measurement will be conducted in the future.

Among all systems, it can be observed that the PP/PE02/MgO nanocomposites with 20 % of PE02 content can effectively enhance the flexibility of PP, making it more suitable as cable insulation material.

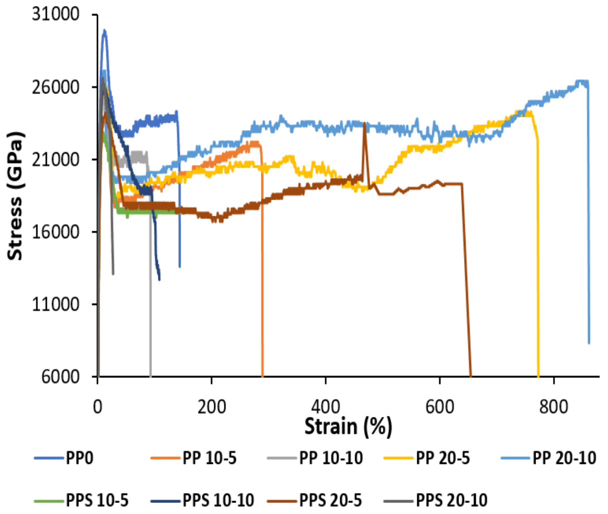

Fig. 4. Stress-Strain curves for all systems

TABLE II
YOUNG'S MODULUS VALUES FOR ALL SYSTEMS

| Materials | Young's Modulus (MPa) |
|---|---|
| PP0 | 287.6 ± 8.2 |
| PP 10-5 | 243.2± 10.6 |
| PP 10-10 | 248.9 ± 4.7 |
| PP 20-5 | 211.7 ± 7.2 |
| PP 20-10 | 206.3 ± 6 |
| PPS 10-5 | 252.7 ± 4.8 |
| PPS 10-10 | 239.9 ± 9.5 |
| PPS 20-5 | 204.7 ± 8.8 |
| PPS 20-10 | 215.1 ± 8.8 |

## IV. CONCLUSIONS

The addition of nano-MgO and PE02 result in a decrease of both the AC and DC breakdown strength in most cases. However, increased DC breakdown strength and suppressed space charge accumulation can be observed by adding a small amount of C8-E treated nano-MgO. All the surface treated systems exhibit a better AC and DC breakdown as well as space charge profiles, which can be related to the increased compatibility and nano-MgO dispersion.

In terms of tensile strength, no significant increase can be observed with the help of C8-E treatment. Although the best tensile strength can be obtained by adding 20 wt. % of PE02, a significant decrease in breakdown strength can be observed. A standout result is a specimen with 10 wt. % PE02 and 5 wt. % C8-E treated-MgO, striking a good balance of breakdown strength, space charge suppression and enhanced the tensile strength. Hence, this polymer nanocomposite is considered to be a viable option for HV cable insulation material in the future.

## ACKNOWLEDGEMENT

The authors would like to thank Professor George Chen and Dr. Miao Hao for their support. This work is based on the final project to fulfil Min Wei's degree of Master of Science in Energy and Sustainability with Electrical Power Engineering at University of Southampton.

## REFERENCES

[1] M. Jeroense, "HVDC, the next generation of transmission: Highlights with focus on extruded cable systems," *IEEJ Trans. Electr. Electron. Eng.*, vol. 5, no. 4, pp. 400–404, 2010.

[2] T. Andritsch, A. S. Vaughan, and G. C. Stevens, "Novel insulation materials for high voltage cable systems," *IEEE Electr. Insul. Mag.*, 2017.

[3] K. Kurahashi, Y. Matsuda, Y. Miyashita, T. Demura, A. Ueda, and K. Yoshino, "The application of novel polypropylene to the insulation of electric power cable (3)," *Electr. Eng. Japan (English Transl. Denki Gakkai Ronbunshi)*, vol. 155, no. 3, pp. 1–8, 2003.

[4] Y. Zhou, J. He, J. Hu, and B. Dang, "Surface-modified MgO nanoparticle enhances the mechanical and direct-current electrical characteristics of polypropylene/polyolefin elastomer nanodielectrics," *J. Appl. Polym. Sci.*, vol. 133, no. 1, 2016.

[5] X. Wang, T. Andritsch, and G. Chen, "Effect of surface functionalization on the dielectric properties of Polypropylene Aluminium Nitride Nanocomposites," in *IEEE Int. Conf. on Dielectr. (ICD)*, 2018.

[6] I. L. Hosier, M. Praeger, A. F. Holt, A. S. Vaughan, and S. G. Swingler, "On the effect of functionalizer chain length and water content in polyethylene/silica nanocomposites: Part I—Dielectric properties and breakdown strength," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 24, no. 3, pp. 1698–1707, 2017.

[7] I. L. Hosier, M. Praeger, A. S. Vaughan, and S. G. Swingler, "The effects of water on the dielectric properties of aluminum based nanocomposites," *IEEE Trans. Nanotechnol.*, vol. PP, no. 99, pp. 1–10, 2016.

[8] I. L. Hosier, A. S. Vaughan, and S. G. Swingler, "An investigation of the potential of polypropylene and its blends for use in recyclable high voltage cable insulation systems," *J. Mater. Sci.*, vol. 46, no. 11, pp. 4058–4070, 2011.

[9] T. Andritsch, "Epoxy Based Nanodielectrics for High Voltage DC-Applications: Synthesis, Dielectric Properties and Space Charge Dynamics," TU Delft, 2010.

[10] J. W. Zha, Y. Wang, W. K. Li, S. J. Wang, and Z. M. Dang, "Electrical properties of polypropylene/styrene-ethylene-butylene-styrene block copolymer/MgO nanocomposites," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 24, no. 3, pp. 1457–1464, 2017.

[11] T. Takada, Y. Hayase, Y. Tanaka, and T. Okamoto, "Space charge trapping in electrical potential well caused by permanent and induced dipoles," in *IEEE Conference Electr. Insul. Dielectr. Phenom. (CEIDP)*, 2007, pp. 417–420.

[12] K. Y. Lau, A. S. Vaughan, G. Chen, and I. L. Hosier, "Space charge dynamics in silica-based polyethylene nanocomposites," in *IEEE Int. Conf. Solid Dielectr. (ICSD)*, 2013, pp. 880–883.

[13] L. Zhang, Y. Zhou, X. Cui, Y. Sha, T. H. Le, Q. Ye, and J. Tian, "Effect of nanoparticle surface modification on breakdown and space charge behavior of XLPE/SiO 2 nanocomposites," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 21, no. 4, pp. 1554–1564, 2014.