

Electrical breakdown and chemical properties of polypropylene/calcium carbonate nanocomposites

A. Azmi¹, K. Y. Lau², N. A. Ahmad³, Z. Abdul-Malek⁴, C. W. Tan⁵

^{1,2,3,4}Institute of High Voltage and High Current, School of Electrical Engineering, Universiti Teknologi Malaysia, Malaysia

⁵Division of Electrical Power Engineering, School of Electrical Engineering, Universiti Teknologi Malaysia, Malaysia

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ABSTRACT

Calcium carbonate (CaCO₃) is an inorganic material which has been introduced in polymeric insulation to improve the thermal and electrical properties of insulation. In this research, a comparative study was performed between unfilled polypropylene (PP) and PP nanocomposites containing 5 wt% of CaCO₃ nanofiller. The electrical breakdown performances of unfilled PP and nanocomposites were investigated. The chemical content of the material was carried out using Fourier transform infrared (FTIR) spectroscopy. The findings showed that the introduction of CaCO₃ to the PP altered the chemical bonding of the material, where two additional characteristic absorption bands appeared in the FTIR spectra of PP/CaCO₃ nanocomposites. In addition, PP/CaCO₃ nanocomposites had slight changes on the AC breakdown strength compared to unfilled PP. Meanwhile, the DC breakdown performance of the nanocomposites was apparently lower than that of unfilled PP. These were attributed to the agglomeration of CaCO₃.

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Corresponding Author:

Aizat Bin Azmi,
Institute of High Voltage and High Current,
School of Electrical Engineering,
Universiti Teknologi Malaysia,
81310 Johor Bahru, Malaysia.
Email: aizat.azmi.1987@gmail.com

1. INTRODUCTION

Nowadays, the demand for electrical power supply around the world has increased tremendously as all sectors in every country depend strongly on power supply in order to fulfill their needs. Hence, manufacturers related to electrical power industry, especially producers of high voltage cables, have an important responsibility to ensure that their cables can address the increased power demand. This leads to the need of more advanced electrical insulation, which is expected to have better endurance and reliability than conventional insulation systems [1-3].

Polymer nanocomposites' use in electrical insulation was introduced in 1990s by Lewis [4]. The idea behind this is to combine polymers with a few weight percent (wt%) of nanometer-sized fillers under homogeneous mixing. In this, the physical, chemical and electrical properties of nanocomposites were found to be better than the unfilled polymers.

Cross-linked polyethylene (XLPE) has been widely used as high voltage AC (HVAC) and high voltage DC (HVDC) cable insulation [5-7]. Nevertheless, many studies reported the drawbacks of XLPE, such as its difficulty to recycle, increased space charge accumulation, and poor long-term deterioration properties [1]. Polypropylene (PP) material has a potential to replace XLPE cables. PP has been reported to have higher melting temperature than XLPE (commonly above 150 °C) [8-9]. Besides that, PP exhibits high mechanical strength [10-11] and low dielectric constant [12]. Furthermore, PP possesses good characteristics as high voltage cable insulation as it has reduced space charge accumulation [13], high volume resistivity [14], and high breakdown strength [15-17] compared with XLPE.

Previous research on nanocomposites in electrical insulation mostly focused on single-metal oxide nanofillers such as magnesium oxide (MgO), titanium dioxide (TiO₂), zinc oxide (ZnO), and aluminum oxide (Al₂O₃) [18-20] as nanofillers. Meanwhile, the use of multi-element oxide nanofillers, from the perspective of dielectric, has been less explored. Hence, it is important to investigate the effect of different types of multi-element oxide nanofillers on the dielectric properties of PP nanocomposites. The results are useful to guide the development of potentially recyclable HVDC and HVAC cable insulation materials.

Therefore, this research aimed to investigate the breakdown characteristics (i.e., AC and DC breakdown) and chemical structure of PP containing CaCO₃ nanofiller (also known as PP/CaCO₃ nanocomposites). The use of CaCO₃ as an alternative inorganic filler in PP will contribute to further understanding of the effects of using multi-element oxide nanofillers for cable insulation applications.

2. RESEARCH METHOD

2.1. Sample preparation

PP used in this study composed of 50% of PP homopolymer and 50% of PP impact copolymer, which were obtained from Lotte Chemical Titan, with manufacturer codes of TITANPRO 6531M and TITANPRO SM340, respectively. Besides that, CaCO₃ nanofiller, which was obtained from the SkySpring Nanomaterials, was used. The nanofiller had a particle size ranging from 15 to 40 nm. Nanofiller concentration level of 5 wt% was chosen. Brabender melt mixer was used for sample preparation. The rotational speed was set to 50 rpm. Meanwhile, a temperature of 180°C was used throughout the mixing process. The complete mixing duration was set to 10 min. Next, a hydraulic laboratory press was employed to produce disc-shaped sample films. In this, the processing temperature and pressure were set to 180°C and 3 tons, respectively. Next, the samples were cool down naturally under laboratory ambient environment. Figure 1 and Figure 2 demonstrate an example of a PP compound produced from the Brabender melt mixer and a disc-shaped PP sample produced from the hydraulic laboratory press, respectively.



Figure 1. Polypropylene sample prepared by means of Brabender melt mixer



Figure 2. Polypropylene sample prepared by means of hydraulic laboratory press

2.2. Breakdown testing

AC and DC breakdown tests were carried out by means of a dielectric strength tester. The tests were done in accordance with the guidelines laid out in the American Society for Testing and Materials (ASTM) D149. Each sample, with 100 μm of thickness, was used for these tests. The sample was placed between two sphere electrodes. Besides that, the sample was immersed in mineral oil in order to prevent surface discharges. The breakdown strength test was conducted by applying a step voltage of 1 kV every 20 s for AC breakdown tests. The voltage was increased until the sample experienced breakdown. Meanwhile, a step voltage of 2 kV every 20 s was used for DC breakdown tests. Similarly, the voltage was increased until the occurrence of breakdown. The breakdown readings were recorded and analyzed by using the two-parameter Weibull distribution analysis. As shown in (1) and (2) represent the dielectric breakdown strength and Bernard's approximation, respectively. Meanwhile, As shown in (3) shows the probability of breakdown strength by means of cumulative distribution function (CDF) through Weibull distribution analysis.

$$\text{BreakdownStrength} \left(\frac{\text{kV}}{\text{mm}} \right) = \frac{\text{BreakdownVoltage}(\text{kV})}{\text{SpecimenThickness}(\text{mm})} \quad (1)$$

$$F(i, N) = \frac{i-0.3}{N+0.4} \tag{2}$$

$$F(E_{bd}; \alpha, \beta) = 1 - \exp \left[- \left(\frac{E_{bd}}{\alpha} \right)^\beta \right] \tag{3}$$

Based on (2), i represents the sequence of dielectric breakdown strength. Meanwhile, N is the total number of breakdown tests. Based on (3), the probability of dielectric breakdown strength of the specimen is represented as $F(E_{bd}; \alpha, \beta)$, where α is the scale parameter and β is the shape parameter based on the Weibull analysis.

2.3. Fourier transform infrared analysis

Fourier transform infrared (FTIR) spectroscopy was used to obtain the chemical information pertaining to the prepared nanocomposites. The FTIR machine model IRTracer-100 from SHIMADZU was used. The spectral data were collected from 600 cm^{-1} to 4000 cm^{-1} over 8 scans. For this analysis, sample with 100 μm of thickness was used.

2.4. Morphological characterization

Morphological characterization of PP nanocomposites containing CaCO_3 was carried out to analyse the dispersion state of CaCO_3 nanoparticles in PP using Hitachi TM3000. Generally, a 15 kV voltage at 38 mm distance was applied during the characterization. Prior to SEM, the samples were immersed in liquid nitrogen and fractured. The fracture surfaces of the samples were then sputter coated with platinum to minimize charge accumulation and poor resolution during morphological inspections.

3. RESULTS AND DISCUSSION

3.1. Breakdown results

Figures 3(a) and 3(b) demonstrate the AC and DC breakdown strength of unfilled PP and PP nanocomposites, respectively; the Weibull data are listed in Table 1. It should be noted that the scale parameter α represents the breakdown strength while the shape parameter β represents the reliability of the Weibull distribution.

Clearly, these unfilled PP and PP nanocomposite samples behaved differently under AC and DC applied fields. It can be noted that the unfilled PP had higher AC and DC breakdown strength than that of PP/ CaCO_3 nanocomposites, with 112 kV mm^{-1} and 323 kV mm^{-1} , respectively. Meanwhile, PP/ CaCO_3 nanocomposites had the AC and DC breakdown strength of 96 kV mm^{-1} and 257 kV mm^{-1} , respectively. These results show that the breakdown strength of PP/ CaCO_3 nanocomposites under AC and DC applied fields was lower compared with unfilled PP. These results are in line with the research in [21-24].

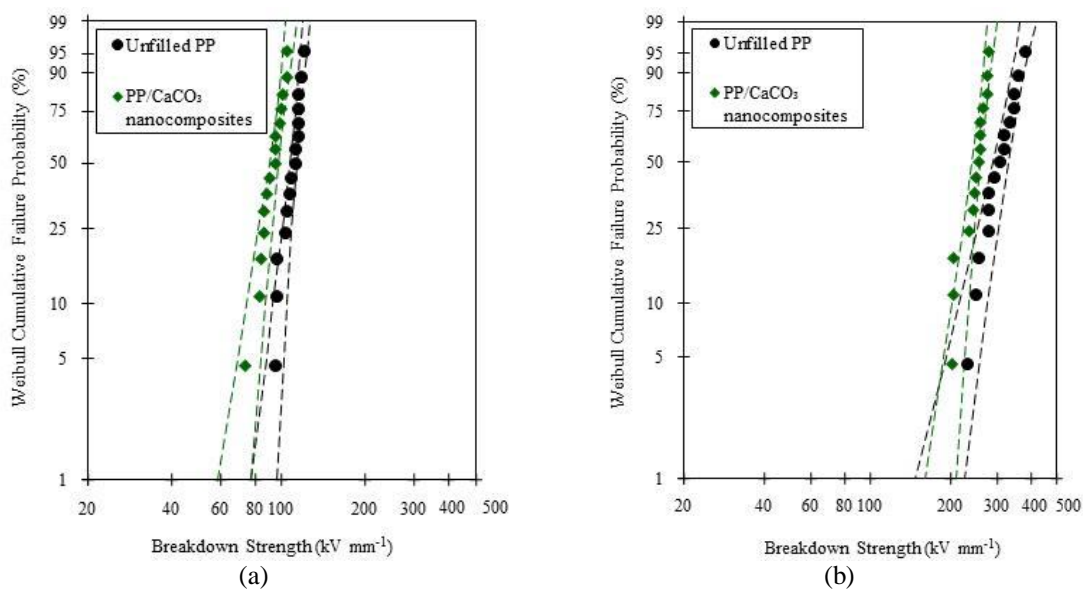


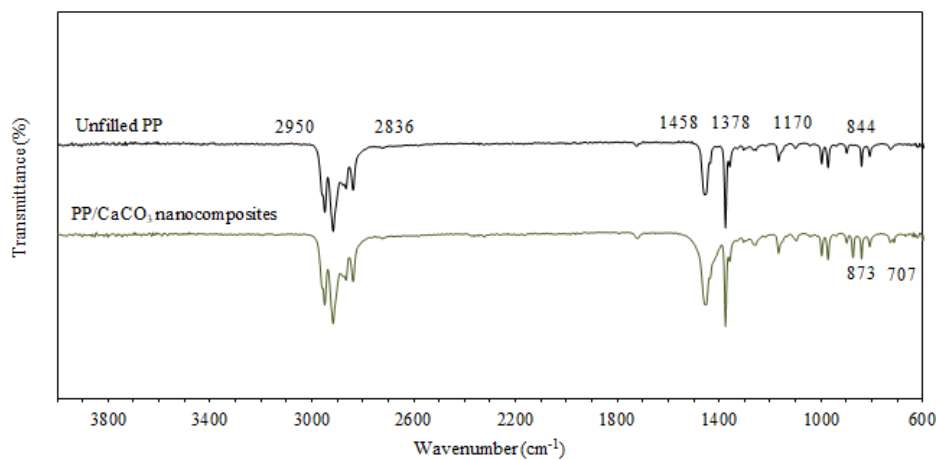
Figure 3. Weibull plots for comparing (a) AC and (b) DC breakdown strength of unfilled PP and PP/ CaCO_3 nanocomposites

Table 1. Weibull distribution parameters

Breakdown	Sample	α (kVmm ⁻¹)	β
AC	Unfilled PP	112 ± 3	17 ± 8
AC	PP/CaCO ₃	96 ± 4	13 ± 5
DC	Unfilled PP	323 ± 18	8 ± 3
DC	PP/CaCO ₃	257 ± 9	14 ± 5

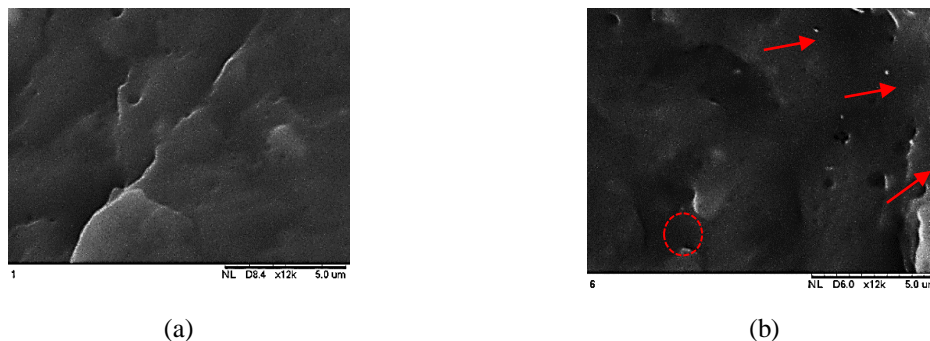
3.2. Fourier transform infrared spectra

Figure 4 illustrates the FTIR spectra of unfilled PP and PP nanocomposites. As a reference, the characteristic absorption bands for the unfilled PP are 2950 cm⁻¹, 2836 cm⁻¹, 1458 cm⁻¹, 1378 cm⁻¹, 1170 cm⁻¹ and 844 cm⁻¹. These FTIR values of PP are in line with the values reported in the literature [25]. Besides that, the characteristic absorption bands at 873 cm⁻¹ and 707 cm⁻¹ of PP/CaCO₃ nanocomposites are indicative of fundamental band of calcite and asymmetrical stretching vibration peaks of Ca-O [26]. It can be concluded that the CaCO₃ nanofiller was successfully attached in PP polymer as demonstrated in the FTIR analysis.

Figure 4. FTIR spectra comparing unfilled PP and PP/CaCO₃ nanocomposites

3.3. Morphology

Figures 5(a) and 5(b) show the SEM micrograph obtained from unfilled PP and PP/CaCO₃ nanocomposites. The SEM micrograph of unfilled PP reveals some degree of the structure of PP blends, but the detailed structure and morphology of the unfilled PP is not well revealed in this fracture surface. Meanwhile, there are several CaCO₃ nanoparticles of about 100 nm in size that could be observed as indicated by the arrows in Figure 5(b). However, the agglomeration of CaCO₃ nanoparticles could also be observed as indicated by the circles. It was expected that, agglomeration would lead to reduced breakdown strength. Nevertheless, an alternative CaCO₃ nanofiller, such as surface modified CaCO₃ nanofiller, could be explored to improve the dispersion the nanoparticles across the samples in an attempt to improve the breakdown performance.

Figure 5. SEM micrograph of (a) unfilled PP and (b) PP/CaCO₃ nanocomposites

4. CONCLUSIONS

This research was conducted to investigate the breakdown characteristics (i.e., AC and DC breakdown) and chemical structure of unfilled PP and PP/CaCO₃ nanocomposites. It can be concluded that, the CaCO₃ nanofiller was successfully attached to PP polymer as demonstrated in the FTIR analysis. Besides that, the breakdown strength of PP/CaCO₃ nanocomposites under AC and DC applied fields was lower compared with unfilled PP. This was due to the agglomeration of CaCO₃, which in turns reduced the breakdown performances.

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BIOGRAPHIES OF AUTHORS



Aizat Azmi received the B.Eng. degree in Electrical (Control and Instrumentation) and the M.Eng. degree in Electrical Engineering from Universiti Teknologi Malaysia (UTM), Johor Bahru, Malaysia, in 2011 and 2014, respectively. He is currently a PhD candidate at High Voltage and High Current (IVAT) Universiti Teknologi Malaysia. His research interests are in field of high voltage cable insulation and sensor technology.



Kwan Yiew Lau received his B.Eng. degree in Electrical Engineering with First Class Honors from Universiti Teknologi Malaysia in 2007. After working as an Electronics Development Engineer at Dyson Malaysia, he joined Universiti Teknologi Malaysia as a Tutor in 2008. He then completed his MEng degree in Electrical Power Engineering at the same university in 2010. Later in 2013, he received his Ph.D. degree in Electronics and Electrical Engineering from the University of Southampton, UK. He is now a Senior Lecturer at the Institute of High Voltage and High Current, Universiti Teknologi Malaysia. He is also a Chartered Engineer of the Engineering Council UK, a Professional Engineer of the Board of Engineers Malaysia, and the Past Chair of the IEEE Dielectrics and Electrical Insulation Society (DEIS) Malaysia.



Noor Azlinda Ahmad received her B.Eng. (Electrical) and M. Eng. (Electrical) Degree from Universiti Teknologi Malaysia in 2001 and 2003, respectively. She completed her PhD in Atmospheric Discharge from Uppsala University, Sweden in 2011. At present she is a senior lecturer at Institute of High Voltage and High Current (IVAT) Universiti Teknologi Malaysia. Her major research interests are lightning protection, measurement, detection and characterization.



Zulkurnain Abdul-Malek (M'03) received the B.E. degree in electrical and computer systems from Monash University, Melbourne, Australia, in 1989, the M.Sc. degree in electrical and electromagnetic engineering with industrial applications from the University of Wales Cardiff, U.K., in 1995 and the Ph.D. degree in high voltage engineering from Cardiff University, U.K., in 1999. He has been with Universiti Teknologi Malaysia (UTM) for 30 years, and he is currently a Professor of High Voltage Engineering with the Faculty of Engineering, and the Director of the Institute of High Voltage and High Current (IVAT), UTM.



Chee Wei Tan (M'11–SM'17) received the B.Eng. degree in electrical engineering (First Class Honors) from Universiti Teknologi Malaysia (UTM), Johor, Malaysia, in 2003, and the Ph.D. degree in electrical engineering from Imperial College London, London, U.K., in 2008. He is an Associate Professor with UTM and a Member of the Power Electronics and Drives Research Group, School of Electrical Engineering, Faculty of Engineering, UTM. He is also a Chartered Engineer registered with the Engineering Council, U.K. and a Professional Engineer registered with the Board of Engineers Malaysia. He is an active participant of IEEE activities and conferences. His research interests include the application of power electronics in renewable/alternative energy systems, control of power electronics and energy management systems in microgrids.