STRUCTURE AND DIELECTRIC PROPERTIES OF POLYPROPYLENE CONTAINING MULTI-ELEMENT OXIDE NANOFILLERS

AIZAT BIN AZMI

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School of Electrical Engineering
Faculty of Engineering
Universiti Teknologi Malaysia

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DEDICATION

This thesis is dedicated to my father, mother, wife, siblings, and kids

Thank you for your support

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ABSTRACT

Polymer nanocomposites have attracted significant research attention especially in the field of high voltage insulation. The enhancement in the dielectric properties of polymer nanocomposites is led by the unique interphase interactions between nanoparticles and base polymers. However, common single metal oxide nanofillers, which are supposed to improve the dielectric properties of nanocomposites, often led to reduced electrical breakdown strength. Recently, multi-element oxide nanofillers have been shown to possess favorable properties compared to single metal oxide nanofillers. Nevertheless, very few systematic studies have been carried out to determine the structure-dielectric property relationship of multi-element oxide nanofillers, especially when added to polypropylene (PP). In the current work, different types of multi-element oxide nanofillers, namely, untreated magnesium aluminate ($MgAl_2O_4$), untreated calcium carbonate ($CaCO_3$), and surface-modified calcium carbonate (*t*-$CaCO_3$), were added to PP to determine their effects on thermal, chemical, structural, and dielectric properties of PP, before and after aging. As such, thermogravimetric analysis, differential scanning calorimetry, Fourier transforms infrared, scanning electron microscopy, dielectric response, AC breakdown, and DC breakdown measurements were performed. The results demonstrated that PP nanocomposites containing $MgAl_2O_4$ possessed up to 58% lowered breakdown strength than unfilled PP. Adding $CaCO_3$ to PP resulted in up to 43% higher breakdown strength of the nanocomposites compared to PP/$MgAl_2O_4$ nanocomposites. Notably, PP/*t*-$CaCO_3$ nanocomposites possessed the highest breakdown strength (up to 45%) among the nanocomposite systems considered. While unfilled PP showed much reduced breakdown strength (up to 27%) after aging, all the nanocomposites demonstrated less detrimental effects on their breakdown strength (up to only 21%) compared to unfilled PP, and that the breakdown strength of PP nanocomposites was generally more predictable after aging. The structure-property relationship governing these dielectric changes is subsequently discussed. This paves the way for the development of future power cable insulation systems based on nanostructured PP technology.

ABSTRAK

Polimer nanokomposit telah menarik minat penyelidikan yang ketara khususnya dalam bidang penebatan voltan tinggi. Peningkatan ciri-ciri dielektrik polimer nanokomposit dipengaruhi oleh interaksi interfasa yang unik antara partikel nano dan polimer. Walau bagaimanapun, pengisi nano oksida logam tunggal biasa, yang sepatutnya meningkatkan sifat dielektrik nanokomposit, mengakibatkan pengurangan kekuatan pecah tebat. Baru-baru ini, pengisi nano oksida berbilang unsur telah tertunjuk mempunyai sifat yang baik berbanding pengisi nano oksida logam tunggal. Namun begitu, sangat sedikit kajian sistematik yang dijalankan untuk menentukan hubungan sifat struktur-dielektrik pengisi nano oksida berbilang unsur, terutamanya apabila ditambah kepada polipropilena (PP). Dalam kajian ini, pelbagai jenis pengisi nano oksida berbilang unsur, iaitu, magnesium aluminat ($MgAl_2O_4$), kalsium karbonat ($CaCO_3$) tidak dirawat dan kalsium karbonat dirawat (*t*-$CaCO_3$), telah ditambah kepada PP untuk menentukan kesannya pada sifat terma, kimia, struktur dan dielektrik PP, sebelum dan selepas penuaan. Oleh itu, analisis termogravimetrik, kalorimetri pengimbasan pembezaan, transformasi Fourier inframerah, mikroskop elektron pengimbasan, tindak balas dielektrik, kekuatan pecah tebat AC dan kekuatan pecah tebat DC telah dilakukan. Keputusan menunjukkan bahawa PP nanokomposit yang mengandungi $MgAl_2O_4$ mempunyai kekuatan pecah tebat yang lebih rendah sehingga 58% daripada PP tidak terisi. Penambahan $CaCO_3$ kepada PP menghasilkan kekuatan pecah tebat sehingga 43% lebih tinggi berbanding PP/$MgAl_2O_4$ nanokomposit. Khususnya, PP/*t*-$CaCO_3$ nanokomposit mempunyai kekuatan pecah tebat tertinggi antara sistem nanokomposit yang dipertimbangkan (sehingga 45%). Walaupun PP tidak terisi menunjukkan kekuatan pecah tebat yang jauh berkurangan (sehingga 27%) selepas penuaan, semua nanokomposit menunjukkan kesan yang kurang memudaratkan pada kekuatan pecah tebat (sehingga hanya 21%) berbanding PP tidak terisi, dan bahawa kekuatan pecah tebat PP nanokomposit secara amnya adalah lebih mudah diramal selepas penuaan. Hubungan struktur-sifat yang mengawal perubahan dielektrik ini kemudiannya dibincangkan. Ini memberi laluan kepada pembangunan sistem penebat kabel kuasa pada masa hadapan berdasarkan teknologi PP berstruktur nano.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

AC	-	Alternating current
Al_2O_3	-	Aluminum oxide
AIN	-	Aluminum nitride
ASTM	-	American Society for Testing and Materials
BN	-	Boron nitride
$CaCO_3$		Calcium carbonate
CH_2	-	Methylene
DC	-	Direct current
DSC	-	Different scanning calorimetry
FTIR	-	Fourier transform infrared spectroscopy
HDPE	-	High-density polyethylene
iPP	-	Isotactic polypropylene
LDPE	-	Low-density polyethylene
MgO	-	Magnesium oxide
$MgAl_2O_4$	-	Magnesium aluminate
MDPE	-	Medium-density polyethylene
PE	-	Polyethylene
PP	-	Polypropylene
PPh	-	Polypropylene homopolymer
PPi	-	Polypropylene ethylene impact copolymer
PPr	-	Polypropylene random copolymer
rPP	-	Ethylene-propylene random copolymer
SEM	-	Scanning electron microscopy
SiO_2	-	Silicon dioxide
TEPCO	-	Tokyo Electric Power Company
TiO_2	-	Titanium dioxide
TGA	-	Thermogravimetric analysis
XLPE	-	Cross-linked polyethylene
ZnO	-	Zinc oxide

LIST OF SYMBOLS

α	-	Scale parameter
β	-	Shape parameter
°C	-	Degree celsius
ΔH	-	Melting enthalpy
ε'	-	Real permittivity
h	-	Hour
Hz	-	Hertz
kV	-	Kilovolt
$P(x)$	-	Cumulative failure probability
rpm	-	Rotation per minutes
wt%	-	Weight percentage
χ	-	Crystallinity

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

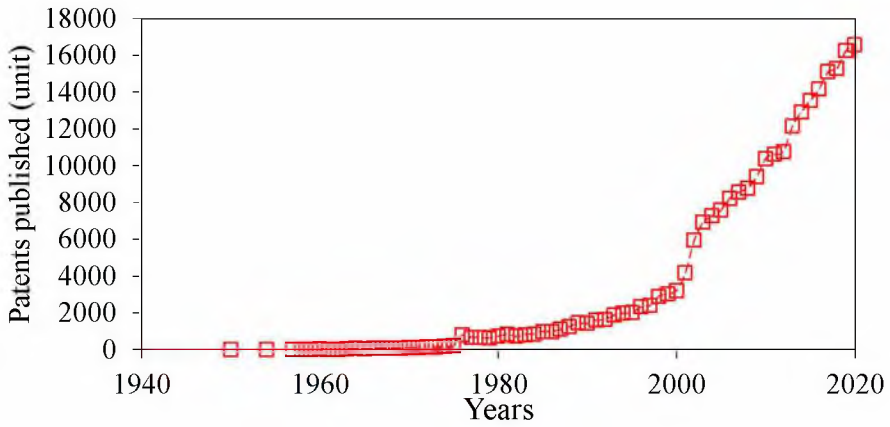
1.1 Research Background

Nowadays, electrical power has become an essential part of modern life. Almost all work activities depend on the supply power to fulfill their demands. As a result, the manufacturers of power equipment are keen to develop better and more reliable components for the generation, transmission, and distribution of power. This leads to the need for more advanced electrical insulation systems, which are expected to have better endurance and reliability than conventional insulation systems in the generation, transmission, and distribution of electricity. To date, polymeric insulation materials have been extensively used as high voltage insulation materials due to their low dielectric constant and dielectric loss, excellent mechanical flexibility, ease of processing, and low cost. Even though the development of polymers as high voltage insulators is widespread due to their benefits to the industry, research on new polymeric materials has been ongoing and is essential to further improve the performance of the current insulation materials.

Currently, the most common polymeric materials used for commercial high voltage alternating current (HVAC) [1] and high voltage direct current (HVDC) cables are low-density polyethylene (LDPE), high-density polyethylene (HDPE), combinations of LDPE and HDPE with different compositions, and cross-linked polyethylene (XLPE) [2-8]. Of note, XLPE has been introduced specifically to improve the thermo-mechanical issues of LDPE; the cross-linked macromolecular network characteristic of XLPE has been employed for this improvement. Besides that, XLPE has been developed to compensate for the poor mechanical properties exhibited by LDPE at high temperatures. However, XLPE is difficult to be recycled at the end of its lifetime due to its thermoset nature resulted from the crosslinking process. This is indeed not consistent with the concept of environmental protection and sustainable

development. In addition, by-products generated during the crosslinking process could enhance the formation of space charge accumulation within the XLPE insulation which in turn deteriorated the electrical property of XLPE. Previous researches that were conducted by Nuriziani et al. [9] and Hirai et al. [10] showed that the cross-linking byproducts introduced more home charges and hetero charges in the XLPE system. The presence of byproducts has led to deteriorated electrical properties in XLPE systems. A similar finding was also reported by Maeno et al. [11]. In the research, Maeno et al. [4] concluded that the space charge formation can only be reduced if a proper method is developed to eliminate all the crosslinking by-products. Besides that, Xiaoguang et al. [12] experimentally demonstrated the thermal expansion drawback of XLPE insulation when operated at high temperatures. Similarly, Qi et al. [12] found that the typical XLPE cables expanded up to 12% due to thermal expansion when exposed to 120 ^{o}C of temperature. The poor thermal performance of XLPE is indeed due to its relatively low melting points. Another drawback of XLPE is the need for the degassing process, which is time-consuming and costly.

Recently, polypropylene (PP) materials have been extensively explored in high voltage insulation [13-20]. This material has a huge potential to replace the conventional XLPE cable system [21-25]. Previous researches show that PP is thermally stable and could be operated at high-temperature conditions. This is mainly due to PP's characteristics such as having a high melting temperature (commonly above 150 °C) [26], low dielectric constant [27], high mechanical strength [28], high volume resistivity [29], and reduced space charge accumulation [30]. Besides that, PP is classified as a thermoplastic material that can easily be recycled compared to the conventional XLPE cable system [31]. Figure 1.1 demonstrates the number of patents that have been patented based on PP insulation in general [32]. To date, the total number of PP patents across various fields, including electrical insulation is 253,429. Significantly, the number of patents increases over the years. Besides that, United States contributes the highest number of patents registered, followed by, the European countries as shown in Figure 1.1b. Meanwhile, Semiconductor Energy Lab, Canon KK, and Fujifilm Corp are the top companies that published the highest patents of PP insulation (see Figure 1.1c).



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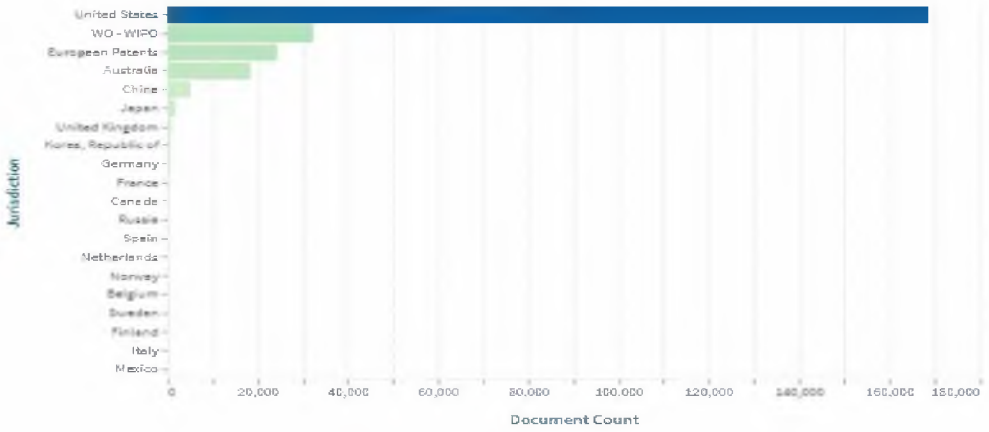

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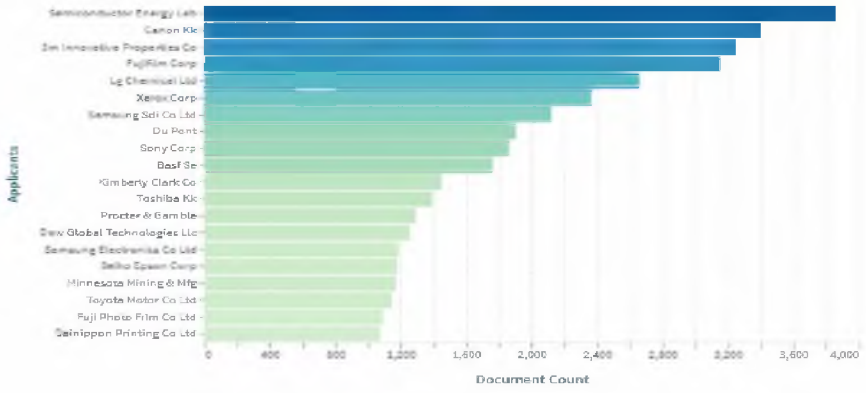

(c)

Figure 1.1: PP patents filed across various field, including electrical insulation (a) number of patents published, (b) jurisdiction at the patents registered, and (c) applicants obtained from lens.org [32]

Nevertheless, isotactic polypropylenes (iPP) have the problem of being too brittle for inclusion into practical cable designs [33]. Hence, various researches have focused on modifying the elasticity of PP and one popular method is by blending PP

with softer materials [34-41]. Hosier et al. [42] and Green et al. [43] discovered that PP blends have improved mechanical and electrical properties compared to their respective standalone iPP. Green et al. [43] suggested that the blended PP could potentially improve the effective tie molecule density and enhance the balance of properties required for successful applications in power cables.

Meanwhile, nanocomposites materials have gained research interests since 2003 especially in the field of dielectrics and electrical insulation. Polymer nanocomposites provide unique properties, where with only a few weight percentages, the mechanical, electrical, and physical properties enhancements can be achieved. Indeed, those improvements were not possible using conventional microscale fillers. Generally, there are three major features in polymer nanocomposites compared to conventional microcomposites. Firstly, the amount of nanofiller used in the nanocomposites was lesser than that of microcomposites. Secondly, the size of the nanofiller was less than 100 nm. Thirdly, the nanofiller has a greater specific surface area. These features reflect the nanocomposites' properties. Hence, the mixture of polymer with nanofiller will result in significant change as compared to the unfilled base polymer [44-47].

Currently, single metal oxide nanofillers have been added to PP to improve the dielectric properties of the resulting systems. For example, Cao et al. [48] discovered that the addition of 1 wt% of magnesium oxide (MgO) into PP has restricted the electric field distortion and space charge accumulation within the PP nanocomposites. As a result, the DC breakdown strength of PP nanocomposites has improved. A similar observation on dielectric properties enhancement using single metal oxide nanofillers was also reported by Zhou et al. [29]. In the research work, the addition of zinc oxide (ZnO), aluminum oxide (Al_2O_3), and titanium dioxide (TiO_2) to PP enhanced the DC breakdown strength of PP nanocomposites. Besides that, those nanoparticles have also enhanced the DC volume resistivity, permittivity, and space charge behavior of PP nanocomposites. Zha et al. [49] reported that the addition of 0.5 wt% of ZnO led to the improvement in dielectric properties of PP nanocomposites such as DC breakdown strength, reduced space charge, and mechanical properties of PP nanocomposites. Meanwhile, Mirjalili et al. [50] discovered the enhancement in the mechanical

properties of PP when PP was added with 1 wt% of Al_2O_3 nanofiller. Of note, all of these reports show that the respective nanofillers have been well dispersed in PP, thus improving the dielectric properties of PP.

Nevertheless, agglomeration and water absorption are the main problems for nanocomposites containing single metal oxide nanofillers. According to previous researches, the agglomeration of single metal oxide nanofillers was led by the high surface energy possesses by the single metal oxide nanofillers. Nanofiller agglomeration deteriorates the electrical properties of the nanocomposites [51-53]. Meanwhile, Lau et al. [54] discovered that the presence of water in nanocomposites is critical and required significant attention especially for those containing single metal oxide nanofillers. Generally, single metal oxide nanofillers contain surface hydroxyl groups which may promote water absorption [55]. The water absorption phenomenon often leads to much reduced dielectric performances especially in the area of high voltage insulation [56-60]. Of note, water absorption usually occurred at the interphase of the nanocomposites. As a result, water absorption will increase the overall electrical conductivity of the nanocomposites and consequently reduce the electrical breakdown strength of the nanocomposites.

Recently, multi-element oxide nanofillers have shown unique characteristics over single metal oxide nanofillers, such as having a compact structure with excellent thermal, mechanical, and electrical properties compared to single-metal oxide nanofillers [61-63]. Samad et al. [64] found that polyethylene (PE) nanocomposites containing magnesium aluminate ($MgAl_2O_4$) (multi-element oxide nanofiller) have greater breakdown strength than that of PE containing Al_2O_3 (single metal oxide nanofiller). Meanwhile, Virtanen et al. [61] reported that calcium carbonate ($CaCO_3$) with 150 nm in size and homogeneously dispersed has increased the DC breakdown strength of PP. Besides that, the addition of $CaCO_3$ has improved the thermal stability of PP [65, 66] and poly(vinylidene fluoride) (PVDF) [67]. Lin et al. [26] demonstrated that the crystallization temperature of PP nanocomposites increased with the $CaCO_3$ nanofiller loading level. A similar observation was reported by Fuad et al. [68]. Generally, the improvement in thermal, mechanical, and electrical properties was led by a high specific surface area possessed by the $CaCO_3$ nanofiller.

Although the use of multi-element oxide nanofillers in nanocomposites seems promising, the application of such nanofillers is less well explored from the perspective of nanocomposite dielectrics. In addition, insulations are commonly used for a long period. Due to prolonged exposure at service temperature, the physicochemical characteristics of the insulation will be compromised. Hence, their resistance to aging needs to be considered. To date, thermal aging of PE and XLPE materials has been well studied [69-73]. Nevertheless, there was little literature related to thermal aging on PP nanocomposites. Furthermore, the effects of thermal aging on PP nanocomposites are still not well understood from the perspective of dielectrics. Specifically, the degradation mechanism under service stress (high-temperature exposure) was insufficiently understood for the evaluation of the life expectancy of PP nanocomposites, which is very important from the practical point of view.

In short, very few systematic investigations have been conducted on the dielectric effects of multi-element oxide nanofillers, especially when added to PP, albeit that the benefits of using multi-element oxide nanofillers in improving the breakdown strength of nanocomposites have been reported elsewhere [61]. Besides that, available literature on the aging of PP and its nanocomposites under different aging temperatures and conditions (i.e., under vacuum and air circulation) is scarce. Therefore, in the current work, the effects of multi-element oxide nanofillers, i.e., $MgAl_2O_4$, $CaCO_3$, and surface treated $CaCO_3$ (*t*-$CaCO_3$) nanofillers, on the structure and dielectric properties of PP were investigated. In addition, the effects of different aging temperatures on the structure and dielectric properties of unfilled PP and PP nanocomposites containing $MgAl_2O_4$, $CaCO_3$, and *t*-$CaCO_3$ nanofillers were also investigated. The aging temperatures of 110 °C and 140 °C were specifically chosen to be below the peak melting temperature of PP such that aging was conducted in the solid-state, under conditions relevant to practical cable insulation. Furthermore, the effects of aging under different environmental conditions, i.e., aging under vacuum (which considered the effect of heat in the absence of air) and aging under air circulation (also known as thermo-oxidative aging, which considered the effect of heat in the presence of air) were carried out.

1.2 Problem Statement

The demand for electrical power supply has increased tremendously as more power is required for the operation of electrical and electronic apparatuses. Consequently, thermal and electrical stresses on electrical insulating materials also increase rapidly with increasing voltage, which necessitates the development of insulating materials with excellent dielectric properties. Nevertheless, the conventional XLPE material, which is widely used for HVAC and HVDC cables, demonstrates several limitations which complicate its development as future power cables. Specifically, XLPE has a low melting temperature, generates byproducts during cable manufacturing, is time consuming for the crosslinking process, and is difficulty to recycle at the end of its lifecycle. Therefore, PP material has recently been proposed to address the aforementioned issues of the XLPE material. Lately, single-metal oxide nanofillers have been added to polymers, including PP, to improve the electrical properties of the materials. However, the addition of such nanofillers can also otherwise degrade the breakdown strength of the nanocomposites, if issues involving agglomeration and water absorption are not properly addressed. In contrast, multi-element oxide nanofillers, such as $MgAl_2O_4$, $CaCO_3$, and *t*-$CaCO_3$, exhibit different characteristics from single-metal oxide nanofillers in terms of chemical, thermal, and electrical properties. The promising results of using multi-element oxide nanofillers in improving the breakdown strength of nanocomposites can be attributed to good dispersion of the nanofillers in polymers and the nanofillers possessing less pronounced water adsorption effects. Nevertheless, the use of multi-element oxide nanofillers, from the perspective of dielectrics, has been less explored. Very few systematic investigations have been conducted on the dielectric effects of multi-element oxide nanofillers, especially when added to PP. In addition, the effects of thermal aging on unfilled PP and PP nanocomposites have not been well studied from the perspective of dielectrics.

1.3 Objectives of the Research

The main objectives of the research were:

1. To formulate and characterize reproducible unfilled PP and PP nanocomposites containing untreated magnesium aluminate ($MgAl_2O_4$), untreated calcium carbonate ($CaCO_3$), and treated calcium carbonate (t-$CaCO_3$) nanofillers.

2. To determine the dielectric response and breakdown characteristics of unfilled PP and PP nanocomposites containing $MgAl_2O_4$, $CaCO_3$, and t-$CaCO_3$ nanofillers.

3. To investigate the effects of thermal aging on the structure and dielectric properties of unfilled PP and PP nanocomposites containing $MgAl_2O_4$, $CaCO_3$, and t-$CaCO_3$ nanofillers.

1.4 Scope of the Study

The scope of this research was as follows:

1. This study is conducted to formulate and characterize reproducible unfilled PP and PP nanocomposites for future HVAC and HVDC power cable insulation systems. Hence, the base polymer used in this research was a PP blend composed of 50% PP homopolymer (PPh) and 50% PP impact copolymer (PPi). Of note, PPh has a high flexural modulus of 17000 kg cm^{-2} which is considered brittle. Meanwhile, PPi has a lower flexural modulus (13500 kg cm^{-2}) than that of PPh, which is desirable to improve the mechanical flexibility and toughness characteristics of the final PP material. Nevertheless, the mechanical tests were not considered in this research.

2. Three different types of multi-element oxide nanofillers were chosen, i.e., $MgAl_2O_4$, $CaCO_3$, and *t*-$CaCO_3$. The amounts of the nanofillers were chosen as 1 wt%, 2 wt%, and 5 wt%. Such amounts of nanofillers were chosen to avoid severe agglomeration issues. According to literature, if the nanofiller amounts added in the polymer were greater than 5 wt%, the tendency of nanofiller to agglomerate was high and this could jeopardize the dielectric properties.

3. Aging of samples was carried out under two different conditions, namely, vacuum and air circulation. Specifically, under vacuum, the samples were aged at 110 °C and 140 °C for 360 h under atmospheric pressure of 133 Pa. This led to three sample types for comparative assessments. They were unaged samples (20 °C), samples aged at 110 °C, and samples aged at 140 °C. The aging temperatures were chosen to be below the peak melting temperature of PP such that aging was conducted in the solid-state, under conditions relevant to practical cable insulation. Meanwhile, under air circulation, another batch of samples was aged at 110 °C for 360 h with 40% of the air circulated in the oven to consider the effect of aging in the presence of air.

4. The samples were characterized using laboratory analytical techniques. For instance, the thermal behavior of the materials was measured using differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA). The temperatures for DSC measurements ranged from 60 °C to 180 °C. Meanwhile, TGA data were collected in the temperatures range from 30 °C to 900 °C. The chemical content of the material was obtained using Fourier transform infrared spectroscopy (FTIR) and the spectral data were collected from 500 cm^{-1} to 4000 cm^{-1}. Meanwhile, the structure and morphology of the samples were characterized using scanning electron microscopy (SEM).

5. The dielectric response of the samples was examined based on the guidelines in the American Society for Testing and Materials (ASTM) D150 standard, namely, Standard Test Methods for AC Loss Characteristics and Permittivity (Dielectric Constant) of Solid Electrical Insulation. The frequency range of the measured dielectric permittivity was from 100 Hz to 100 kHz. Below 100 Hz,

the measured frequency values were unstable and therefore the measurement was not pursued. Besides that, the imaginary permittivity of the materials was not discussed in the thesis as the measured imaginary permittivity of the material was very low due to the limitation of the instrument.

6. Electrical breakdown tests (AC and DC) were conducted based on the guidelines in the ASTM D149 [74] and D3755 standards [75]. The rated AC voltage for the breakdown equipment was 80 kV while the rated DC voltage for the breakdown equipment was 110 kV [76]. Two-parameter Weibull distribution was used for analyzing the breakdown data. The AC and DC breakdown strength of the material was correlated with the thermal, structure, chemical, and dielectric permittivity.

1.5 Contributions of Research

The present study has successfully formulated and characterized unfilled PP and PP nanocomposites containing $MgAl_2O_4$, $CaCO_3$, and *t*-$CaCO_3$. The selection of PP-based materials was generally inspired to replace conventional XLPE materials for HVAC and HVDC cable insulation. The thermal melting behavior of unfilled PP and PP nanocomposites demonstrated a high melting temperature of approximately 162 °C, which was greater than that of XLPE (105 °C). Practically, XLPE possesses thermal expansion when operated under high-temperature conditions. As such, having a high melting temperature exhibited by unfilled PP and PP nanocomposites as shown in the current work can prevent thermal expansion issues and minimize the cost of operation and cable replacement.

The addition of $MgAl_2O_4$, $CaCO_3$, and *t*-$CaCO_3$ to PP affected the breakdown strength of the resulting PP nanocomposites. Specifically, PP/*t*-$CaCO_3$ nanocomposites with better interfacial effects demonstrated an improved breakdown strength. This was attributed to the consequence of electrical conduction effects becoming less dominant over the favorable nanofiller/polymer interactions at large separations between nanoparticles. Meanwhile, PP/$MgAl_2O_4$ had lowered breakdown

strength due to an increase in the local electric field as a consequence of permittivity mismatches between the PP and $MgAl_2O_4$, which was exacerbated by nanofiller agglomeration. Overall, the use of different multi-element oxide nanofillers ($MgAl_2O_4$, $CaCO_3$, and *t*-$CaCO_3$) in the current PP system demonstrates the importance of engineering the local interactions between nanoparticles and polymer to achieve desirable dielectric properties.

The findings on the effects of aging on the materials under vacuum and air circulation conditions revealed a different degree of effects. For instance, aging the materials under vacuum conditions caused the samples to experience structural changes with unchanged chemical properties. Meanwhile, aging the materials under air circulation conditions caused the materials to experience both structural changes and chemical degradation, where new absorption bands at 1752 cm^{-1} were produced, indicating the carbonyl bands. Significantly, the intensity of carbonyl bands reduced with increasing $MgAl_2O_4$, $CaCO_3$, and *t*-$CaCO_3$ amounts. These results showed that the addition of high amounts of $MgAl_2O_4$, $CaCO_3$, and *t*-$CaCO_3$ in PP helped to delay the appearance of thermo-oxidative reactions after aging under air circulation.

Besides that, aging unfilled PP and PP nanocomposites under vacuum and air circulation conditions reduced their DC breakdown strength. Specifically, unfilled PP experienced much reduction in the DC breakdown strength as compared to PP nanocomposites at the same elevated temperatures and conditions. A contributory factor to the huge reduction in the DC breakdown strength of unfilled PP was the significant increase in the real relative permittivity experienced by unfilled PP after aging. In contrast, PP nanocomposites' lesser variations in the real relative permittivity after aging suggested that the addition of $MgAl_2O_4$, $CaCO_3$, and *t*-$CaCO_3$ to PP changed the polymer network through interactions between $MgAl_2O_4$ to PP. As such, the DC breakdown performance of PP nanocomposites was less susceptible to aging compared to unfilled PP, and that the DC breakdown properties of PP nanocomposites were generally more predictable after aging. These findings show that PP nanocomposites are more practical for use in power cable insulation when considering for the long-term operation. Overall, the current study of structure-dielectric property relationships of unfilled PP and PP nanocomposites before and after aging paves the

way for the development of future HVAC and HVDC cable insulation systems based on nanostructured PP technology.

1.6 Thesis Organisation

The thesis is organized as follows:

Chapter 2 discusses the theory and fundamental concepts related to polymers and nanocomposites. These include the structure and characteristics of the materials. Research related to the development of polymeric insulation materials is reviewed in this chapter too. Besides that, an overview of aging effects on the dielectric properties, chemical content, structure, and morphology of polymeric insulation materials is presented.

Chapter 3 presents a detailed description of the methodology of the research. The sample preparation method used in the current study is presented. The characterization and testing methods used, i.e., DSC, TGA, FTIR, SEM, dielectric response, and electrical breakdown tests are discussed. The methods for the thermal aging process are also presented.

Chapter 4 focuses on the experimental results and discussion. Specifically, the findings from DSC, TGA, FTIR, SEM, dielectric response and electrical breakdown tests of the evaluated samples are presented and discussed.

Finally, Chapter 5 concludes the findings of the current work. Recommendations for future work are also included.

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LIST OF PUBLICATIONS

[1] **A. Azmi**, K. Y. Lau, N. A. Ahmad, Z. Abdul-Malek, C. W. Tan, K. Y. Ching, and A. S. Vaughan, "Structure-Dielectric Property Relationship in Polypropylene/Multi-element Oxide Nanocomposites," *IEEE Trans. Nanotechnol.*, vol. 20, pp. 377–385, 2021.

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