EFFECT OF SILICA AND POFA ON ELECTRICAL, PHYSICAL AND MECHANICAL PROPERTIES OF LINEAR LOW-DENSITY POLYETHYLENE-NATURAL RUBBER COMPOSITE

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THESIS SUBMITTED IN FULFILLMENT FOR THE
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2017

DECLARATION

I hereby declare that the material in this thesis is my own except for quotations, excerpts, equations, summaries and references, which have been duly acknowledged.

30 August 2017

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ABSTRACT

Polymer composite is a versatile material used in making electrical insulating material to be used especially in high voltage system. Through this study, a new type of electrical insulating material is made by the addition of fillers. Application of this material, especially in high voltage, is in string insulation on transmission line, underground cables and transformer bushing cables. Besides that, this research adds value to natural rubber if it is used as electrical insulating material in the future. The purpose of this study is to characterise the performance of newly developed polymer composite, which is linear low density polyethylene-natural rubber with different type of fillers, namely palm oil fuel ash (POFA) and nanosilica, at different filler concentration of 0 to 6 parts per hundred (phr) to assess its use as a potential electrical insulating material. In this study, dielectric constant and tangent delta are investigated between 20Hz to 4000Hz. Corona discharge peak-to-peak voltage is investigated at electrical stress of 30kV. Other properties that are investigated are tensile strength, elongation at break, tensile modulus and water absorption. Fourier Transform Infrared (FTIR) spectroscopy is also done to analyse the changes in absorption bands, while Scanning Electron Microscopy (SEM) is performed to analyse the surface morphology. The results revealed that as frequency increased, the dielectric constant decreased for all samples. At 50 – 60 Hz, the dielectric constant for all type of composites increased. Corona discharge test showed that LLDPE-NR composites with nanosilica at 2phr decreased the PTP voltage by 16%, which were supported by Fourier Transform Infrared (FTIR) spectroscopy. FTIR revealed that the O-H bonds intensity of nanosilica at 3500-3200 cm^{-1} due to corona discharge test was less than POFA. From the tensile test, it was found that the tensile strength of LLDPE-NR composite increased the highest by 0.4% at 6phr of nanosilica. Elongation at break and tensile modulus also increased the most by 7% and 42%, respectively, at 6phr of nanosilica. Meanwhile, palm oil fuel ash (POFA) could increase the tensile modulus of the composite. The value of water absorption of LLDPE-NR with nanosilica was the lowest compared to composites with POFA. LLDPE-NR composite with nanosilica content of 2phr showed the best electrical properties, while LLDPE-NR composite with 6phr of nanosilica showed the best mechanical and physical properties. Nano-sized nanosilica particles have very large surface area as compared to micro-sized POFA particles. Because of that, nanosilica particles have better dispersion and large surface interaction with LLDPE-NR matrices which inhibited the motion of charge carriers within the composite. This resulted in increasing electrical insulating properties of LLDPE-NR composite. Better dispersion of nanosilica within the composite also resulted in stronger bonding with the polymer matrices. Nanosilica would act as temporary crosslinks between the polymer chains, which provided localized regions of enhanced strength, which would resist the growth of cracks or cavities in the LLDPE-NR composite. Meanwhile, micro-sized POFA particles have smaller surface area as compared to nanosilica. Because of that, micro-sized POFA particles have smaller surface interaction with LLDPE-NR composite which caused the charge carriers to be more mobile as compared to LLDPE-NR composite with nanosilica. This resulted in the decreasing of the electrical insulating properties of LLDPE-NR composite. POFA also have higher tendency to agglomerate together due to its large size as compared to nanosilica. Agglomeration of POFA, a type of defect within the composite, would weaken the mechanical strength of composite.

Overall, LLDPE-NR composites with nanosilica fared better than POFA-filled composites in terms of electrical, physical and mechanical properties.

ABSTRAK

KESAN SILIKA DAN HABUK KELAPA SAWIT KE ATAS SIFAT ELEKTRIKAL, FIZIKAL DAN MEKANIKAL POLIETILENA KETUMPATAN RENDAH LINEAR-GETAH ASLI

Komposit polimer merupakan bahan yang mempunyai ciri-ciri yang sesuai sebagai bahan penebat elektrik sistem voltan tinggi. Bahan penebat elektrik yang baru telah dicipta melalui penambahan bahan pengisi. Aplikasi penggunaan bahan penebat baru ini, terutama dalam sistem voltan tinggi, adalah sebagai penebat talian penghantaran elektrik (transmission line), *kabel bawah tanah dan penebat kabel pengubah* (transformer bushing cable). *Kajian ini berpotensi memberi nilai tambah kepada getah asli jika bahan ini digunakan sebagai penebat elektrik voltan tinggi pada masa hadapan.Tujuan kajian ini dijalankan adalah untuk mencirikan prestasi komposit polimer, polietilena ketumpatan rendah linear-getah asli* (LLDPE-NR) dengan *bahan pengisi berbeza, iaitu habuk kelapa sawit (POFA) dan nanosilika, pada konsentrasi 0 hingga 6* phr*, untuk menentukan kesesuaian bahan baru ini sebagai penebat elektrik. Pemalar dielektrik* (dielectric constant) *dan kehilangan dielektrik* (dielectric loss) *dikaji pada frekuensi 20 Hz ke 4000 Hz. Voltan puncak-ke-puncak* (PTP Voltage) *penyahcasan korona telah dikaji pada tekanan elektrik setinggi 30 kV. Ciri-ciri lain yang dikaji adalah sifat tegangan* (tensile properties) *dan nilai serapan air oleh komposit. Spektroskopi inframerah jelmaan Fourier* (FTIR) *turut dijalankan untuk menganalisis perubahan sifat kimia bahan komposit selepas ujian penyahcasan korona. Analisis mikroskopi electron penskanan* (SEM) *terhadap permukaan bahan komposit selepas ujian tegangan. Hasil kajian mendapati bahawa semakin tinggi frekuensi, semakin rendah pemalar dielektrik* (dielectric constant) *untuk semua jenis bahan komposit. Pada 50 Hz – 60 Hz, pemalar dielektrik meningkat untuk semua jenis bahan komposit dengan bahan pengisi. Ujian penyahcasan korona menunjukkan* LLDPE-NR *dengan nanosilica pada tahap 2phr dapat mengurangkan voltan PTP sebanyak 16%. Ini disokong oleh analisis FTIR yang menunjukkan keamatan atau intensiti ikatan O-H yang terendah pada spectrum 3500 – 3200* cm^{-1} *untuk bahan komposit dengan bahan pengisi nanosilika berbanding POFA. Daripada ujian tegangan yang dijalankan, didapati nanosilika pada 6phr meningkatkan kekuatan tegangan* (tensile strength) *sebanyak 0.4%, manakala modulus tegangan* (tensile modulus) *meningkat sebanyak 42% dan peratus pemanjangan ketika putus* (elongation at break) *meningkat sebanyak 7%. Di samping itu, nanosilika dapat mengurangkan nilai serapan air oleh komposit* LLDPE-NR*, manakala POFA pula meningkatkan nilai serapan air oleh komposit tersebut. Keseluruhannya, komposit* LLDPE-NR *dengan nanosilika pada 2phr mempunyai sifat elektrik terbaik, manakala nanosilika pada 6phr mempunyai sifat mekanikal dan fizikal yang terbaik. Partikel-partikel nanosilika yang bersaiz nano mempunyai luas permukaan yang lebih besar berbanding dengan partikel-partikel POFA yang bersaiz mikro. Disebabkan itu, partikel-particle nanosilika mempunyai keseragaman taburan (dispersion) yang lebih baik, dan interaksi permukaan* (surface interaction) *yang lebih luas dengan matriks LLDPE-NR dan mengehadkan pergerakan pembawa-pembawa cas* (charge carriers) *di dalam komposit, seterusnya menambahbaik sifat penebat elektrikal LLDPE-NR komposit. Keseragaman taburan nanosilika yang lebih baik berbanding POFA dalam komposit LLDPE-NR juga menguatkan ikatan antara partikel-partikel nanosilika dengan*

matriks polimer tersebut. Partikel-partikel nanosilika akan bertindak sebagai penghubung silang sementara (temporary crosslinks) *antara rantaian polimer* (polymer chains) *yang membawa kepada peningkatan kekuatan* (enhanced strength) *dalam kawasan-kawasan setempat* (localized regions) *komposit*, *seterusnya mengekang keretakan* (cracks) *atau pembentukan kerak* (cavities) *dalam komposit LLDPE-NR. Partikel-partikel POFA yang bersaiz mikro pula mempunyai luas permukaan yang lebih kecil berbanding nanosilika. Disebabkan itu, partikel-partikel POFA mempunyai interaksi permukaan* (surface interaction) *yang bersaiz lebih kecil dengan komposit LLDPE-NR, seterusnya menjadikan pergerakan pembawa-pembawa cas* (charge carriers) *lebih bebas berbanding komposit dengan nanosilika, dan akhirnya melemahkan sifat penebat elektrikal komposit LLDPE-NR. POFA juga mempunyai kecenderungan yang lebih tinggi untuk mengaglomerat* (agglomerate) *disebabkan saiz POFA yang lebih besar berbanding nanosilika. Aglomerasi* (agglomeration) POFA, *sejenis cacatan* (defect) *dalam komposit, akan melemahkan kekuatan mekanikal komposit LLDPE-NR. Secara keseluruhannya, nanosilika merupakan bahan pengisi terbaik untuk* LLDPE-NR *komposit berbanding POFA dari segi sifat elektrik, mekanikal dan fizikal.*

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LIST OF ABBREVIATIONS

A/D - Analogue to digital

AC - Alternating current

CD - Corona discharge

CNT - Carbon nanotube

EDX - Energy-dispersive X-ray spectroscopy

EPDM - Ethylene Propylene Diene Monomer

LLDPE - Linear low density polyethylene

LLDPE-g-MAH - Linear low-density polyethylene grafted with maleic anhydride

FTIR - Fourier Transform Infrared

HDPE - High density polyethylene

HPF - High pass filter

HV - High voltage

HTV - High-temperature vulcanized

kHz - KiloHertz

mm - Millimetre

MMT - Montmorillonite

MWCNT - Multi-walled carbon nanotube

MPa - Megapascal; N/mm^2

NBR - Acrylonitrile Butadiene Rubber

NiCoZN - Nickel-Cobalt-Zinc

NR - Natural rubber

pC - Picocoulomb

PC - Personal computer

PD	**-**	Partial discharge
PET	-	Polyethylene terephthalate
phr	**-**	Parts per hundred
POFA	**-**	Palm oil fuel ash
PTP	**-**	Peak-to-peak
RTV	-	Room temperature vulcanised
SAN	-	Styrene acrylonite
SEM	**-**	Scanning Electron Microscopy
SDS	-	Sodium dodecyl sulfate
SIR	-	Silicon rubber
TPNR	-	Thermoplastic natural rubber
UTM	-	Universal tensile machine

LIST OF SYMBOLS

%	-	Percentage
^{o}C	-	Degree celcius
ε_r	-	Relative permittivity
ε_r'	-	Real part of permittivity; dielectric constant
C	-	Carbon
H	-	Hydrogen
N	-	Newton
O	-	Oxygen
ρ_v	-	Volume resistivity
Si	-	Silicon
V	-	Voltage
W	-	Water absorption (%)
W1	-	Initial weight of sample before water absorption test
W2	-	Final weight of sample after water absorption test

LIST OF PHOTOS

LIST OF PUBLICATIONS

1. Junian, S., Sahari, J. and Makmud, M. Z. H. 2015. Natural Rubber as Electrical Insulator: A Review. *Journal of Advanced Review on Scientific Research*. **6**(1), 28-42.

2. Junian, S. S., Sahari, J., Makmud, M. Z. H., Arief, Y. Z. and Wahit, M. U. 2016. Tensile and Physical Properties of Linear Low Density Polyethylene-Natural Rubber Composite: Comparison between Size and Filler Types. *International Journal of Engineering Transactions C: Aspects*. **29**(9), 1239-1244.

CHAPTER 1

INTRODUCTION

1.1 Research Backgrounds

Electricity is very important in our daily life. The use of electricity ranges from powering homes, schools to large factories. Since the creation of alternating current (AC) power system by George Westinghouse, high voltage electricity transmission is made possible (Gibbs, 2002). As shown in Figure 1.1, the electricity is produced in a generating station and a transformer is used to “step up” the voltage as high as 500 kV which is transmitted over transmission lines before the voltage is “stepped down” as low as 120 V to be distributed to end users. This also allows electricity to be transmitted over long distance.

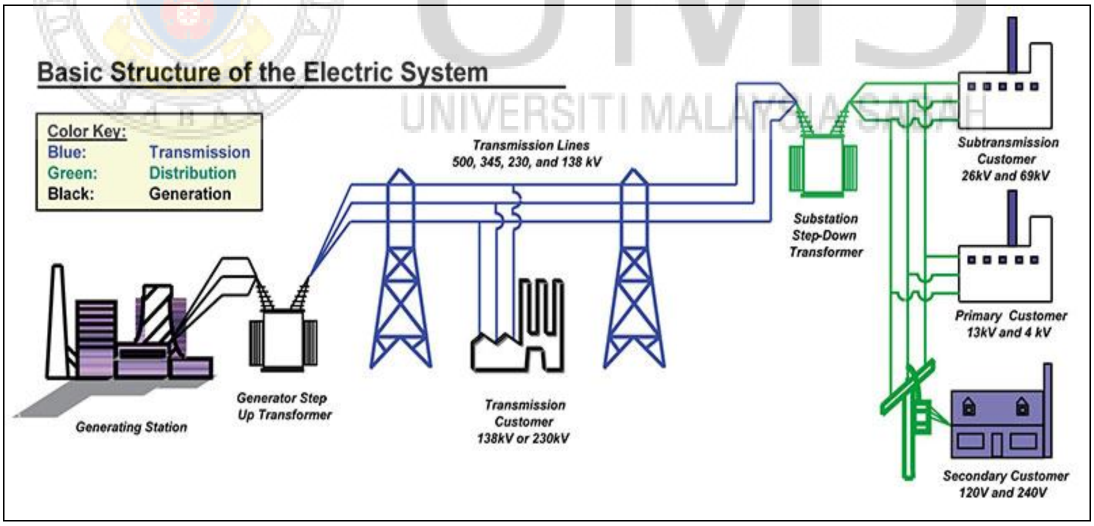

Figure 1.1: Basic structure of the Electric System

Source : http://www.ucsusa.org/

Since the 19th century, electrical insulation has played a major part in the transmission and distribution needs of telegraph, telephone and electrical systems (Zuidema, Kegerise, Fleming, Welker, and Boggs, 2011). In a power system, insulating material protects it from breaking down. Electrical insulator is defined as a poor conductor which has high resistivity towards an electric current's flow. Electrical insulators separate conductors from one another and to hold them in position, besides protecting the conductors from surrounding structures. They also keep the current's flow to wires or other conducting paths only. The protection of electrical circuits is a vital prerequisite for the effective operation of all electrical and electronic equipment which is made possible by using electrical insulators ("Insulator," 2012). Figure 2.1 shows the design of an electrical power cable with insulation.

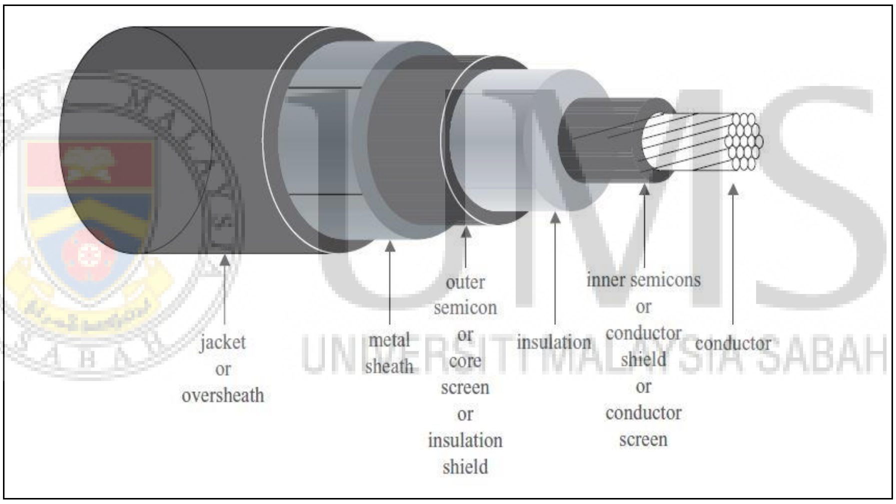

Figure 1.2: Power cable design

Source : Fothergill and Hampton (2007)

From the explanation above, installing a proper electrical insulation material is a must, more so in a power system that operates at a voltage as high as 500kV. Several ageing factors of an insulation material include electrical (Lee, 2006; Toriyama, Okamoto, Kanazashi, and Horii, 1967), mechanical (Densley, 2001) and environment e.g. exposure to water (Nikolajevic, 1999; Sigmond and Sigmond, 1991). Hence, a good electrical insulation material must have a strong electrical resistance, water resistance and high mechanical strength to support the weight of the conductor and to withstand wind load.

Cable technology is heavily dependent on electrical insulation technology. It involves the development of new insulation materials of low dielectric loss and high electrical strength, optimized stress control techniques in cable joints and terminations, and efficient heat dissipation methods (Yun and Chung, 1998). Glass, porcelain and polymer composite are commonly used as high voltage electrical insulators (Swift, 2007). Polymer composite is defined as a “macroscopically homogeneous mixture of two or more different species of polymer” to form a composite (Jenkins, Kratochvil, Stepto, and Suter, 1996). Current trend shows polymer composites, such as epoxy resin and silicon rubber (Iyer, Gorur, Richert, Krivda, and Schmidt, 2011; Manjang, Putera, Akil, and Kitta, 2015; Yao et al., 2016), as the subject of interest in the field of electrical insulating materials.

The uses of nano silicon dioxide (SiO_2), or nanosilica, as fillers have been reported to improve the characteristics of electrical insulating materials. At very low content below 10 weight percentage (wt%) of polymer composites, nanosilica can increase characteristics such as dielectric strength (Gao, Zhang, Wang, Li, and Li, 2013), partial discharge resistance (Sugumaran, 2013), and tensile strength (Cheng, Miao, Zhang, and Peng, 2011). LLDPE-NR has not yet been studied with nanosilica, which leaves a gap in the study of its characteristics as potential electrical insulating filler of LLDPE-NR.

There is a need to make use of excessive residue from palm oil industry, which could reach 44 million tonnes a year (Chiew and Shimada, 2013). Palm oil fuel ash (POFA), a type of palm oil residue has been utilised to replace commonly used concrete-making materials which can yield higher compressive strength (Al-mulali, Awang, Abdul Khalil, and Aljoumaily, 2015; Awal and Shehu, 2015; Bashar et al., 2016; Islam, Alengaram, Jumaat, and Bashar, 2014; Khankhaje et al., 2016; Munir et al., 2015; Ranjbar, Behnia, Alsubari, Birgani, and Jumaat, 2016). The study on palm oil fuel ash, or POFA, specifically in use as electrical insulating material is uncommon. However, the main component of POFA is silica

(Chindaprasirt, Chotetanorm, and Rukzon, 2011), which is used in many studies as filler in polymer insulators (Manjang et al., 2015).

1.2 Problem statement

The use of natural rubber-based insulations for electrical wire dates from the earliest days of the electrical industry. It was the only polymeric material used as a wire and cable dielectric up to the 1930s, when the first suitable synthetics became available (Zuidema et al., 2011). Natural rubber is chosen as a potential electrical insulating material in this study. One of the reasons is that it is one of the main natural resources in Malaysia (Sakdapipanich and Rojruthai, 2014). Besides that, natural rubber is abundant and, evidently, Malaysia is the world's sixth biggest producer of natural rubber ("The Industry," 2015).

Polyethylene has been used during the World War II as an insulation material for underwater cables and it is the most widely used plastic in the world (Demirors, 2011). Linear low-density polyethylene (LLDPE) was introduced by the Phillips Petroleum Company in 1968. LLDPE has similar properties to low-density polyethylene (LDPE), with melting point of approximately 110°C, and also competes for the same markets where the principle uses are in packaging film, grocery bags, wire and cable insulation ("polyethylene (PE)," 2015).

There are not many studies on the characterisation of natural rubber (NR)-linear low-density polyethylene (LLDPE) composite as electrical insulating material. LLDPE-NR composite has been studied with a few types of fillers, such as montmorillonite (MMT) (Makmud, Sayuti, Arief, and Wahit, 2011), titanium (IV) oxide (TiO_2) (Makmud, Arief, Aulia, and Wahit, 2012) and alumina trihydrate (ATH) (Piah, Darus, and Hassan, 2005).

However, the use of palm oil fuel ash (POFA) and nano silicon dioxide (nanosilica) fillers with LLDPE-NR have not yet been reported on whether these fillers can improve the characteristics of LLDPE-NR as electrical insulating materials. Hence, this study will investigate the characteristics of LLDPE-NR which use nanosilica and POFA as fillers.