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Natural Rubber as Electrical Insulator: A Review

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Abstract – *Natural rubber, which comes from Hevea trees, were introduced to Malaysia in the 1870s. It is almost the only commercial rubber from a natural source. 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. Natural rubber does not necessarily exhibit all the desired properties for use in the rubber industry. Thus, it is possible to obtain better mechanical and physical properties at a lower cost by blending natural rubber with synthetic rubbers.*

Keywords: Natural rubber; Electrical insulator; Insulating material**.**

1.0 INTRODUCTION

Hevea rubber, known as natural rubber (NR), is almost the only commercial rubber from a natural source, even though over 2,500 species of plants have also been found to produce cis-1,4-polyisoprene. Among the different plant species producing rubber, H. brasiliensis (Fig.1) has proved to be the best rubber producer at the present time, owing to the high productivity of the plant and excellent physical properties of the rubber product [1]. H. brasiliensis is a tall softwood tree indigenous to Brazil. However, the major producer of NR is the South-East Asia region. In the 1870s, the Hevea trees germinated from seeds originally from Brazil were introduced to Sri Lanka, Malaysia and Singapore and later found to thrive in surrounding countries, where the weather, terrain and soils were well suited for its growth [2]

Figure 1: Rubber tree [3]

Due to its outstanding properties, NR cannot be replaced by synthetic rubber in some specific applications such as heavy-duty tyres, gloves, *etc* [1]. NR could have a leading role in reducing the global carbon footprint in a low-carbon society [2]. NR is an interesting material made from a renewable source, while the synthetic rubber is obtained from non-renewable oil-based resources [1].

2.0 NATURAL RUBBER: GRADES AND STANDARDS

NR latex exuded from the Hevea trees is a colloidal suspension. The amount of latex obtained on each tapping depends on clones, ages, soil, climatic conditions and tapping frequency. The collected latex is usually treated with formic acid to coagulate the suspended rubber particles within the latex. After being pressed between rollers to form the rubber into 3-5 mm thick slabs or thin crepe sheets, the rubber is air- or smoke-dried at 50-60 ^{o}C for 2-3 days into a final thickness of 2-3 mm for shipment. These rubbers are known as air-dried sheet (ADS) and ribbed smoked sheet (RSS), respectively. RSS is visually subdivided into six grades based on colour, consistency and observed impurities, as described in the 'Green Book'. Other commercial types of NR, known as technically specified rubbers (TSRs) in block form, are systematically graded based on technical specifications specified by the International Standards Organisation (ISO), i.e. dirt content, ash content, nitrogen content, volatile matter content, *etc*. Based on these specifications, major rubber-producing countries introduced their respective standards as Standard Malaysian Rubber (SMR), Standard Thai Rubber (STR), Standard Indonesian Rubber (SIR) and Standard Vietnamese Rubber (SVR) [1].

3.0 FABRICATION OF NATURAL RUBBER BLENDS

Arayapranee [4] stated that natural rubber blends are difficult materials to process, because in both the raw and compounded state they have viscoelastic properties. The production sequence is mixing, forming and vulcanizing. Simple mixing ensures that the mixture has a uniform composition throughout its bulk. The solid rubber and the other additives have to be mixed. Rubber mixing mills create uniform blends of NR and synthetic rubber with other raw materials. A mill consists of two hollow metal cylinders rotating towards each other. The gap between the rolls, called a nip, allows the rubber to pass through to achieve mixing caused by the shearing action in the nip (Figure 2).

Mixing is always preceded by a masticating process, introduced in order to soften the rubber so that compounding ingredients can be added. The long polymer chains must be partially broken by mastication, mechanical shearing forces applied by repeatedly passing the dried rubber into the gap between the two mill rolls.

The entire mixing operation is performed through repeated charging and discharging of rubber. The additives are charged while the rubber is bound onto a roll (Figure 1).

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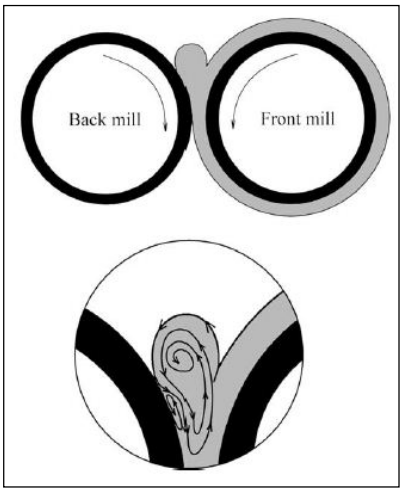

Figure 2: Conceptual view of two-roll mills [4]

Minnath et al. [5] studied the transport properties of thermoplastic polyurethane/natural rubber (TPU/NR) blends at different temperatures. The mixing was done on a two-roll mixing mill (150 x 300 mm^2) with a nip gap of 1.3 mm and at a friction ratio of 1:1.4 at room temperature. The temperature was maintained constant by circulating water through the rolls. The vulcanizing agent, dicumyl peroxide (DCP), was incorporated as per ASTM D15-627. TPU/NR blend membranes were prepared by the masterbatch technique. The blend compositions are $T_{10}N_0$, $T_{80}N_{20}$, $T_{60}N_{40}$, $T_{20}N_{80}$, and T_0N1_{00}, where T and N stand for thermoplastic polyurethane and NR, respectively and the subscripts denote the weight percentage of each component in the blend. In all the formulations, 4g of DCP has been added.

Nabil et al. [6] reported a work on thermal stability of natural rubber/recycled ethylene-propylene-diene-monomer blends. A fixed blend ratio of 70/30 (phr/phr) was produced using a laboratory-sized two-roll mixing mill (Model XK-160). N330 grade carbon black was also incorporated into the blends. The blended compounds were then compression molded at 150 ^oC with a force of 10MPa using a hot press according to the respective curing times (tc_{90}), determined by the MDR 2000. In addition to this study, conventional vulcanization (CV) was not selected because the purpose of blending of NR and recycled ethylene-propylene-diene-monomer is mainly to reduce compounding cost as well as eliminate the waste rubber.

Sabbagh [7] studied the compatibility of NR and ethylene-propylene diene rubber blends. The graft polymer (EPDM-g-MAH) was prepared using EPDM rubber as a backbone polymeric chain and maleic anhydride: γ-radiation was utilized as a polymerization initiator. Blending of the components was achieved by mastication on a two-roll mill for 5 min, then each blend was mixed in a Brabander Plasticorder at a rotor speed of 70 rpm for 5 min. The mixing temperature was 150 ^oC. The amount of the compatibilizer, BR, chlorinated rubber, chlorosulfonated polyethylene and PVC was 10 parts in each case. The compatibilizers were added subsequently to the blend composition and mixing was continued for another 5 min.

In a research by Pal et al. [8], natural rubber and high styrene rubber (HSR) with its various ingredients are mixed in a two-roll mill. A friction ratio of 1:2 followed standard mixing sequence.

Senna et al. [9] melted polypropylene (PP) and different ratios of epoxidized natural rubber (ENR) in Brabender Plastic-order PL2100 Mixed with a volume capacity of about 50 cm^3 at 180 ^oC and 60 rpm for 10 min to obtained different PP/ENR blend ratios. PP/ENR films were prepared by compression molding under hot press at 185 ^oC for 7 min. The time of press was controlled carefully to ensure that the obtained films were not exposed to thermal degradation. A hot press machine type Saspol made by Costuruzioni Meccaniche (Italy) was used. The conditions of hot press were kept at a pressure of 1800 kg/cm^2.

4.0 RUBBER AS ELECTRICAL INSULATING MATERIAL

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 [10]. The use of natural rubber-based insulations for electrical wire dates from the earliest days of the electrical industry. Early manufacturers found that rubber benefits from the admixture of various compounding ingredients, such as mineral fillers and process aids, in addition to the vulcanization agents discovered by Charles Goodyear. Often rubber for electrical insulation was mixed with a product made by heating certain vegetable oils with sulfur in a reaction analogous to Goodyear's vulcanization process and possibly spawned by that same idea. These materials are known variously as factice or pitch and are the source of the term "oil based rubber" often encountered as a description of old rubber compounds. Natural rubber-based electrical insulation was the only polymeric material used as a wire and cable dielectric up to the 1930s, when the first suitable synthetics became available [11].

The dielectric losses of purified rubber when dried are small and exhibit a broadened Debye type of characteristic superimposed on a constant background loss at low temperature and an ionic or conduction-loss type of characteristic at room temperature and above. The losses are increased slightly by exposure to a moist atmosphere. The dielectric losses of dry crude rubber are very similar to those of the purified material. However, in a moist atmosphere, the impurities normally present in crude raw rubber absorb sufficient moisture to raise the power factor to a marked extent [12].

Composite insulators have been increasingly used worldwide due to their many advantages over conventional insulators [10]. These have advantages over glass and porcelain insulators due to ease of installation, lightweight construction, and vandal resilience [13]. The Korea Electric power Corporation (KEPCO) has just started to install composite insulators in service. It is known that silicone rubber (SIR) composite insulators show excellent long-term performance because the SIR surface retains its hydrophobic properties even under polluted conditions. The Korea Electrotechnology Research Institute (KERI) has developed a 22.9 kV SIR composite insulator and obtained good results on the basis of test codes specified by KEPCO [10].

5.0 AGEING OF ELECTRICAL INSULATORS

5.1 Mechanisms

A comprehensive understanding of the insulators long-term performance is yet to be realized, due to the impact of different electrical and environmental aging mechanisms. Environmental aging factors include: ultra violet (UV) radiation from sunlight, organic growth, acid rain and ozone. Electrical aging factors consist primarily of corona, from water droplets or metallic fittings, and dry-band arcing [13].

Lutz et al. [14] analysed a failed 500 kV ac composite insulator by using different test methods. The insulator had fractured after 5 operation years within a lightly polluted environment. It showed no clear signs for brittle fracture (i.e. end-fitting seal undamaged, no smooth fracture surface) indicating different aging mechanisms involved. The mechanical strength of the core material was degraded by water induced aging process, i.e. ion exchange glass corrosion and hydrolysis.

5.2 Electrical Treeing

Treeing is the name given to that type of damage that progress through a dielectric section under electrical stress, so that, if visible, its path looks something like a tree. Treeing has been generally associated with alternating or impulse voltages and it has also been observed with high direct-voltage stresses in wet experimental conditions [15]. It is a failure mechanism, where by fine erosion channels grow from a region of high electric stress within the insulation owing to partial discharge activity and progress towards the opposite electrode [16]. An electrical tree is one that is caused by electrical stress alone [15].

The influence of mechanical forces on the degradation of electrical insulation by treeing has been known for many years. When the mechanical properties of an electrical insulating material are directly degraded, e.g. by the introduction of a plasticizing agent, a correlation can be found between the changes in mechanical properties that result and the changes in the initiation and growth of electrical trees. Thus a decrease in the tensile strength, modulus of elasticity and fracture toughness, as a result of plasticization, is mirrored in the decreasing resistance of the material to electrical tree propagation [17].

In a 1991 study, it is stated that ferrocene is an excellent additive which can enhance not only the treeing initiation voltage of polyethylene largely, but the breakdown strength in some degrees as well. However, it still has the disadvantage of separating from polyethylene with time. In order to modify this, the molecular with long chain of carbon-hydrogen had been bound to the cyclopentadiene ring of the ferrocene to form a ferrocene derivative [18].

6.0 NATURAL RUBBER BLENDS

NR does not generally exhibit all the desired properties for use in the rubber industry. Normally, natural rubber is deteriorated by ozone and thermal attacks due to its highly unsaturated backbone, and it also shows low oil and chemical resistances due to its non-polarity. Thus, it is possible to obtain better mechanical and physical properties at a lower cost by blending natural rubber with synthetic rubbers [19].

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6.1 Natural Rubber With Polymer

The electrical properties of chloroprene rubber (CR) and NR blends were evaluated in a 2005 study. Dielectric measurements were carried out by a Dielectric Spectrometer at room temperature and the frequency ranged from 10 to 100 kHz for measuring the dielectric constant and loss factor. The dielectric constant decreases with the increase of applied frequency. The reduction of the dielectric constant is directly related to the time lag for the orientation of the dipoles in the high frequency alternating electric filed. As content of CR increased, the dielectric constant increased. The dielectric constant is higher for CR-NR blend with ratio of 3:1 from a one-stage blend vulcanizates, compared to the two-stage process. For the physical properties, the modulus of CR and CR enriched blend vulcanizates is higher than NR. For the one-stage vulcanizates, the modulus value increased until around 1.2 MPa as CR content increased at 100 wt.% content. However, the tensile strength decreased to around 5 MPa as CR content increased to 100 wt.% content. The two-stage vulcanizates made the modulus values higher for higher CR content [20].

In a 2011 study, a 33kV transformer bushing insulator was developed from NR and High Density Polyethylene (HDPE). The HDPE is used for increasing weather resistance properties of NR. The compounding rubber consists of 60% NR and 40% HDPE ratio. NR-HDPE is more superior in terms of electrical resistance than NR-EPDM as shown in Table 1. The surface resistivity of NR-HDPE is about 70.7% less than NR-EPDM. The volume resistivity of NR-HDPE is about 92.5% more than NR-EPDM. For the physical properties, it is found that NR-HDPE is more superior to NR-EPDM in terms of hardness, but not for tensile strength and elongation at break [21].

Table 1: Materials properties of NR-EPDM and NR-HDPE [21]

Material Properties	NR-EPDM (60:40)	NR-HDPE (60:40)
Hardness (Shore A)	46.7±3	83.7±0.9
Tensile strength (MPa)	8.82±1.58	4.28±5.4
Elongation at break (%)	548±29	92±29
Volume resistivity (Ω-cm)	$1.6x10^{15}$	$2.14x10^{16}$
Surface resistivity (Ω/square)	$1.9x10^{17}$	$5.56x10^{16}$
Dielectric strength, kV/mm	17.9	19.7

The electrical properties of blends from NR and chitosan (CS) have been studied in the frequency range of 10^2-10^6 Hz in a 2011 study. Pure and vulcanized blends of NR-CS with different ratios were prepared. NR and CS has a volume resistivity (ρ_v) in the order of 10^9 Ω

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cm and 10^2 Ω cm respectively. As the concentration of chitosan increased, the ρ_v value of NR-CS blend decreased. This decrease in ρ_v was due to the introduction of low resistivity CS. The resistivity increased by dynamic vulcanization using dicumyl peroxide (DCP) on the blends of $NR_{85}CS_{15}$ $NR_{65}CS_{35}$. The AC conductivity showed that the vulcanized material sample became more insulative compared to pure and MA treated blends [22].

In a 2012 study, dielectric properties and AC conductivity studies of novel NR/polyvinyl alcohol (PVA) Full-interpenetrating polymer networks (IPN) were done. Samples with total weight of 10 g were prepared with different blend ratios up to 50% (wt/wt) PVA in NR. Addition of glutaraldehyde to PVA crosslinks the PVA chains for the use in the mechanism of formation of Full-IPN based NR/PVA. The capacitance and tan δ were measured at frequencies ranging from Hz to MHz at different temperatures. The dielectric constant and dielectric loss of NR, PVA and its IPNs have been carried out at frequencies ranging from 10^2 to 10^6 Hz. The values of dielectric constant for all the samples are found to decrease by increasing frequency at lower frequency followed by a leveling off at higher frequency. Dielectric values increased with increasing temperature at 1kHz. But compared to pure blends, the IPNs exhibited less effect on temperature. The conductivity values were found to decrease for all IPNs samples compared to pure blends. Pure NR and PVA showed the least values of dielectric loss, which were 6 each. For IPN blend, dielectric loss of $NR_{90}PVA_{10}$ is 5. $NR_{90}PVA_{10}$ (IPN) showed the lowest value of dielectric constant [23].

6.2 Natural Rubber With Filler

6.2.1 Organic Filler

Fiore et al [24] stated that there is a growing attention on the use of natural fibers as reinforcement of polymer composites due to their availability and properties, such as good acoustic insulation property and electrical resistance [25]. The utilization of wood or in general lignocellulosic material has become more attractive particularly for price driven/high volume applications [26]. Oil palm empty fruit bunch (EFB) studies have been recorded in many journals [26]–[32]. Southeast Asian countries like Malaysia and Indonesia are the major oil palm cultivating countries [33]. The solid oil palm residue is a biomass consisting of, by wet weight, 23.8 million tonnes (54%) of empty fruit bunch (EFB), 13.2 million tones (30%) of shell, and 7.9 million (18%) tonnes of fiber [34].

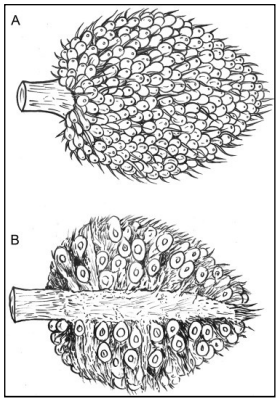

Figure 3: Sketch of (A) oil palm fresh fruit bunches (B) Cross section of EFB showing fiber arrangement [25]

The effect of oil palm empty fruit bunch (OPEFB) filler on the electrical tree propagation in silicone rubber was investigated in a 2012 study. OPEFB has high dielectric strength, high toughness, high drying shrinkage and high Young's Modulus constant. It is found that silicone rubber filled with OPEFB can help in inhibiting the electrical tree propagation. For the pure silicone rubber, the electrical tree grew in length ranging from 1.14 to 1.92 mm in 60 minutes of propagation time. For the filled silicon rubber, the tree length ranged from 0.77 to 0.91 mm. This filler helped to retard the growth of tree [28].

Starch is an abundant biopolymer, which is totally biodegradable. Starch nanocrystals obtained from starch have been used as fillers in polymeric matrices leading to desired reinforcing effect [35]. A 2011 study is done on the correlation morphology and electrical properties of NR rubber latex with potato starch nanocrystal nanocomposites as fillers. The value of the electrical resistivity for the surface is relatively constant (10^7-$10^{10}\Omega$/cm) up to a content of starch of 10% w/w. This electrical surface resistance increases drastically to $10^{22}\Omega$/cm for 15% w/w of starch and for higher content decreases to reach a similar value as obtained for the matrix alone. For the volume electrical resistivity, its value increases even if a small amount of starch is added, but the same threshold is observed for a composition close to 15% w/w of starch. This indicated that nanoparticles act as a barrier to the electrical charge movements. In morphological analysis, a TEM experiments performed on potato starch nanocrystals showed that no aggregate and no impurities are observed [36].

6.2.2 Inorganic Filler

A 2005 study researched the electrical tracking performance of NR blended with linear low-density polyethylene (LLDPE) added with and without filler, alumina trihydrate (ATH), with different formulations. A high voltage of 2.5 kV was applied on each sample. For all blend formulations, a range of 2-6 mA of leakage current (LC) is observed throughout the experiment. The same results are also found in the case of silicone rubber, polyolefin and polyethylene vinyl acetate. The low level of LC indicated a high surface resistance. It is found that all samples with filler concentration of 50 pph tend to suppress leakage current development. By using LC level with the carbon tracking rate as data, the normalized degradation index (*NDI*) for each sample was calculated. LLDPE/NR/ATH sample with ratio of 80:20:0 pph is the best compound formulation for resistance to tracking and erosion as it has the lowest *NDI*. It also received minimum damage compared to other compounds. For the compounds filled with ATH, the LLDPE/NR/ATH compounds with ratio 20:80:50 pph and 20:80:100 pph offer good tracking resistant properties [37].

A 2011 study found that nano montmorillonite (MMT) filled LLDPE-NR composites have less partial discharge numbers compared to LLDPE-NR specimens containing nano titanium (IV) Oxide (TiO_2) fillers after partial discharge (PD) test. The PD test was done according to CIGRE Method II at 7kVrms for 1 and 3 hour ageing time. The blend samples were as shown in Table 2. Total PD numbers for sample B2 (with nano-MMT as filler) is the lowest, followed by sample A5 (without filler) and sample C1 (with nano-TiO_2 as fillers) as shown in Figure 4. The tensile modulus of the composite without filler increased after MMT addition [38]. Using SEM test, it was found that nano-TiO_2 at 4phr shows eroded surface of composite specimen, but not a lot aggregates of fillers appeared compared to nano-MMT filled specimen. The movement of electron flow due to PD activities bridging two electrodes (anode and cathode) created micro-cavities [39].

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Table 2: Compound formulation and coding of LLDPE/NR blends [38]

Sample	Blend component				
	LLDPE	**NR**	**LLDPE-g-MAH**	**MMT***	**TiO_2**
A1	80	10	10	-	-
A2	70	20	10	-	-
A3	60	30	10	-	-
A4	50	40	10	-	-
A5	40	50	10	-	-
B1	70	20	10	2	-
B2	70	20	10	4	-
C1	70	20	10	-	2
C2	70	20	10	-	4

*phr (part per hundred) of LLDPE/NR weight

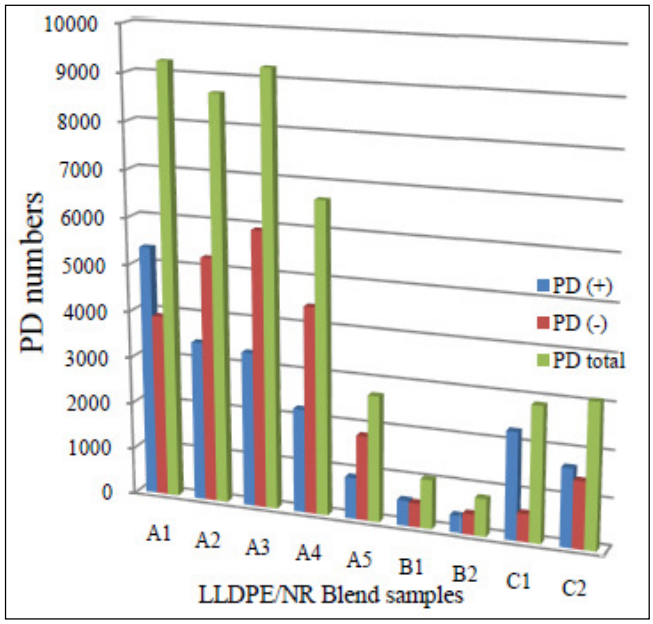

Figure 4: PD numbers for all LLDPE/NR blends formulation [38]

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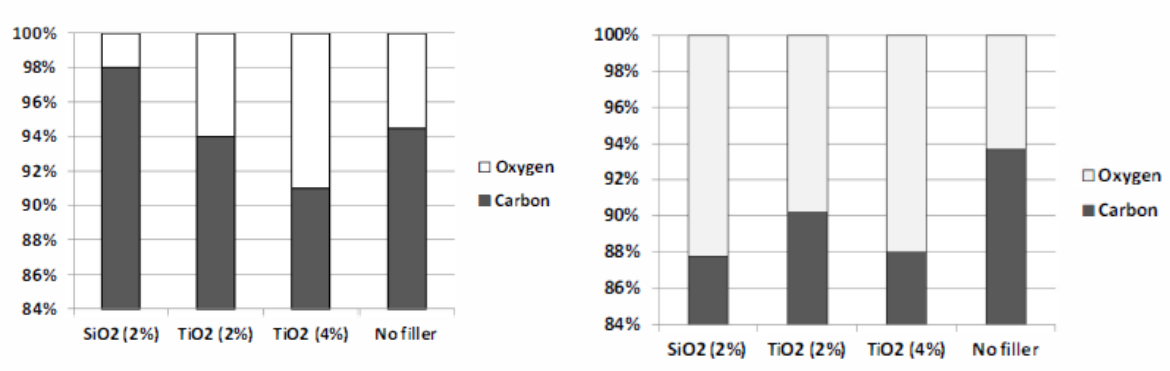
A study was done in 2014 where the sample with the highest percentage of nanofiller loading has the lowest PD pulse number when tested with PD. The research was about the effect of nanosilica and nanotitania on PD characteristics of NR-LLDPE blends as high voltage insulation material. Blend formulation were as shown in Table 3, where LLDPE, NR and LLDPE-g-MAH additive was blended in wt% of 80-20-10. The nanofillers were silicon dioxide or silica (SiO_2) and titanium dioxide or titania (TiO_2). Sample B2 has the lowest PD pulse number compared to sample without fillers. EDX was used to investigate composition elements of carbon and oxygen in every sample before and after PD test. The percentage of oxygen elementally increased after high voltage (HV) stress and vice versa for carbon element. Sample A1 with silica as filler showed the highest percentage of oxygen content after HV test. Oxidation may cause structural weakness which can further degrade the samples [40].

Table 3: Sample composition of investigated material [40]

Sample	Polymer	Nanofiller
A1	LLDPE (80wt%) + NR (20wt%) + LLDPE-g-MAH (10wt%)	SiO_2 (2phr)
B1		TiO_2 (2phr)
B2		TiO_2 (4phr)
C		0phr



(a) Before ageing process (b) After ageing process

Figure 5: Percentage of carbon and oxygen for all natural rubber blends LLDPE samples before and after PD test using EDX analysis [40]

In a 2006 study, the effect of Nickel-Cobalt-Zinc (NiCoZn) ferrite filler on the electrical and mechanical properties of thermoplastic NR (TPNR) composites was studied. The TPNR composites were NR, liquid natural rubber (LNR), and high-density polyethylene (HDPE) in the weight ratio of 5:1:4. The filler concentration was in the range of 0 wt.% to 30 wt.%. The

resistivity of the composites for low filler contents is nearly equal to that of pure polymer matrix. The electrical resistivity (ρ) decreases with increasing filler content. At 0 wt.% and 30 wt.%, the resistivity is around 7 W m and 6 W m respectively. The dielectric constant (ε_r') increases with increasing filler content, where 0wt.% and 30wt.% filler contents having dielectric constant of 30 and 72 respectively . After tensile test, the sample without filler is shown to have high elasticity compared to sample with high filler content. It is found that the Young's modulus increased with the increase of filler content up to 10wt.%. When the filler content is exceeding 10wt.%, there is a continuous decrease in Young's modulus. At 10wt.% and 30 wt.%, the Young's modulus is around 27 MPa and 13 MPa respectively. The elongation at break decreased sharply up to 5 wt.% filler and then decreased slightly with filler content above 5wt.%. The hardness of the composites decreased from 80 to 75 up to 15 wt.% when the ferrite filler is added, and then is almost constant [41].

A study in 2012 used iron oxide-filled carbon nanotubes (CNT) as fillers in NR. Several samples have been prepared containing different amounts of CNTs (from 0.01 to 10 wt.%). The samples will be referred as NR (pure natural rubber), NRA (composites prepared with acid-treated MWCNT dispersions) and NRP (composites prepared using pristine MWCNT/SDS dispersion). At CNT concentration above the percolation threshold (which is about 1-2 wt.% and 2-3.5 wt.% of CNTs for the NRP and NRA samples), a drastic reduction in the electrical resistivity is observed. The sample that contains 10 wt.% of CNTs exhibits a decrease in electrical resistivity of seven orders of magnitude compared to neat NR (from 10^9 Ω-cm to 10^2 Ω-cm) [42].

7. CONCLUSION

Electrical insulating properties of NR can be improved by blending with polymers and other fillers. There are not many studies that concentrate on the electrical characteristics of NR. Most of the researches are more inclined to the study of other types of synthetic rubbers. More studies need to be done to develop electrical insulating materials based on NR and with other blends and fillers.

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