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## Electrical properties of polymer blend composites based on silicone rubber/EPDM/clay hybrid for high voltage insulators

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### ABSTRACT

Silicone rubber (SR) and ethylene-propylene-diene monomer (EPDM) are widely-used polymers as housing for high voltage insulators. In this work, SR/EPDM/clay nanocomposites were obtained by two-roll mill mixing for outdoor polymeric insulators. Morphology, dielectric properties, dielectric breakdown strength (DBS), and surface and volume resistivity of different weight contents of nanoclay (Cloisite 15A) incorporated in SR, EPDM, and SR/EPDM hybrid nanocomposites were characterized. In addition, the distribution of breakdown voltages was fit to the distribution of Weibull and estimated the scale and shape parameters. The polar groups of the clay particles enhanced the polarization capability of the nanocomposites. Moreover, DBS results showed an enhancement of the dielectric strength proportional to clay content. Finally, the surface and volume resistance of all nanocomposites decreases but maintains very high electrical resistance. The experimental data presented in this study will be useful for designing and manufacturing the outdoor insulators.

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### 1. Introduction

In dry environments, fiberglass has a high mechanical and electrical performance. Nevertheless, under humid conditions, it retains moisture and serves as a weak insulator [1]. Because of its potential to have enhanced thermal, electrical, and mechanical characteristics and offering promising properties related to each component [2-6], polymer nanocomposites design is an exciting field of advanced science [7]. While employing polymer nanocomposites in the electrical insulating technology sector has started recently, due to their strong dielectric capabilities, lightweight, better pollution efficiency, low price, and easy production, polymeric materials have provided various benefits to external insulation systems [8].

Silicon rubber (SR), ethylene-propylene-diene monomer (EPDM), and ethylene-propylene rubber (EPR) are widely used polymeric components for electrical insulators. Notably, owing to affordable price, a high percent of elongation, being lightweight, thermal resilience, and improved wear resistance, the use of EPDM rubber materials has been expanded in tire sidewalls, wires, vibration, and noise isolation, cables, and outdoor insulation system products. It also exhibits superior tem-

perature, oxygen, and ozone resistance [9, 10]. However, because of its low rigidity, EPDM rubber cannot be employed for primary structural uses without including fillers such as carbon nanotubes (CNTs) or clay particles [2, 11]. Insulators made of SR exhibit excellent characteristics of hydrophobicity transference in action. As a result, SR insulators exhibit high electrical efficiency in damp and contaminated areas and they are commonly used in toxic settings. However, they struggle with cost, mechanical characteristics, and resistance to tracking [12, 13]. The electrical and mechanical characteristics of EPDM/SR blends in various proportions were analyzed by Raja Prabhu et al. [14]. In addition to dissipation factor, due to silicone's low freezing temperature, low surface tension, and high oxidation resistance, an improvement in electrical properties including tracking resistance, comparable tracking index, dielectric constant, and surface and volume resistivity was observed with increasing amounts of SR. Due to their promising mechanical and physical characteristics, rubber/layered silicate (such as montmorillonite (MMT)) nanocomposites have recently attracted much interest. The distribution of MMT clay particles could lead to either exfoliated and/or intercalated structures in the polymer matrix [15]. Rana et al. [9] studied the crystallographic, thermal, electrical, and mechanical behavior of out-

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door insulating EPDM/clay nanocomposites and found that these qualities are improved by incorporating clay into the composite. Vijayalekshmi [16] studied the mechanical, electrical, and thermal characteristics of organically modified MMT (OMMT) clay distributed in an EPDM/SR (50:50) blend nanocomposites with variable clay content (1, 3, 5 and 7 phr). Increasing OMMT loading to 5 phr was shown to improve electrical and mechanical properties, and a further increase in the filler content led to the reduced properties, which could be related to a low particle dispersion at high clay loadings. Although there are some studies on mechanical and electrical properties of EPDM/SR/clay nanocomposites, to the best of our knowledge, limited researches were made to characterize the electrical and dielectric properties of nanocomposites based on EPDM, SR, and hybrid SR/EPDM matrices [17, 18]. Therefore, the main purpose of the present study is to compare the electrical and dielectric characteristics of these blend composites for high-voltage insulators.

## 2. Materials and method

### 2.1. Materials

Vinyl methyl siloxane (HD-9170, Hongda Co., Ltd, China) was the employed silicone rubber. The hardness and density of SR were 70 (Shore A) and 1.18 g/cm<sup>3</sup>, respectively. EPDM (commercial name: KEP 270) was supplied from Kumho Polychem Co., Ltd., Korea; its Mooney viscosity (ML (1 + 4) at 125 °C) was 70 along with a density of 0.86 g/cm<sup>3</sup>, with a 4.5%ethylene norbornene content and ethylene content of 57.0%. Dicumyl peroxide (DCP; Hercules Inc., USA) with 98% purity was used as the curing agent and the nanoclay was Cloisite 15A fabricated by Southern Clay, USA.

### 2.2. Preparation of the samples

To prepare the samples, EPDM gum was first softened on a two-roll mill manufactured by Polymix 200L, Schwabenthan, Germany, for 3 min at 50 °C (rotor speed=15 rpm, friction ratio=1.6), followed by the SR addition and blending for a further 3 min. Subsequently, the addition of 3, 6, or 9 phr nanoclay and mixing for 3 min were done. Eventually, 1 phr of DCP was added for 2 min. In a 100-ton hydraulic hot press fabricated by Bucher, Switzerland, the mixture was molded by compression

**Table 1.**

Composition of the composites

Sample code*	EPDM	SR	Cloisite 15A
Q100C0	0	100	0
Q100C3	0	100	3
Q100C6	0	100	6
Q100C9	0	100	9
E100C0	100	0	0
E100C3	100	0	3
E100C6	100	0	6
E100C9	100	0	9
Q70E30C0	30	70	0
Q70E30C3	30	70	3
Q70E30C6	30	70	6
Q70E30C9	30	70	9

\*Curing of all the samples was performed with 1 phr of DCP

and cured for 15 min at 160 °C. Specimens were later post-cured at 150 °C for 2 hours in an oven. Prior to testing, the samples were allowed to rest for 24 h at 25°C. Table 1 presents the composition and coding of the nanocomposites.

### 2.3. Microstructural studies

Microstructural images were obtained from cross-sections of different samples fractured in liquid nitrogen by a VEGA TESCAN scanning electron microscope (SEM), operating at 10 kV. An ion-sputter device was employed to gold-coat the specimens in order to minimize charging and enhance electrical conductivity.

### 2.4. Surface and volume resistivity

The measurements of the surface and volume resistivity of the nanocomposites were performed by a high-insulation resistance meter device from Shanghai Precision & Scientific Instrument Co., Ltd, following the procedure indicated in the IEC 60093 standard. The determination of surface resistivity was done by placing the electrodes on the surface and volumetric resistivities were measured applying 500 V for 60 s to the opposite surfaces of the samples. Specific volume resistance was calculated by the following equation (Eq. 1):

$$\rho = R_x \frac{A}{h} \quad (1)$$

where  $R_x$ ,  $A$ , and  $h$  denote the measured resistance, surface area of the electrode, and thickness of the specimen, respectively.

### 2.5. Dielectric properties

Dielectric spectroscopy specifies the dielectric characteristics of materials, including permittivity ( $\epsilon'$ ) and loss tangent ( $\tan \delta$ ), based on the interaction of the applied field and the specimen's internal electric dipole moments. Dielectric properties were measured in an E4980A Precision LCR meter (Agilent Technologies, Inc.). The  $\epsilon'$  and  $\tan \delta$  of the nanocomposites were assessed at low frequency (50 Hz).

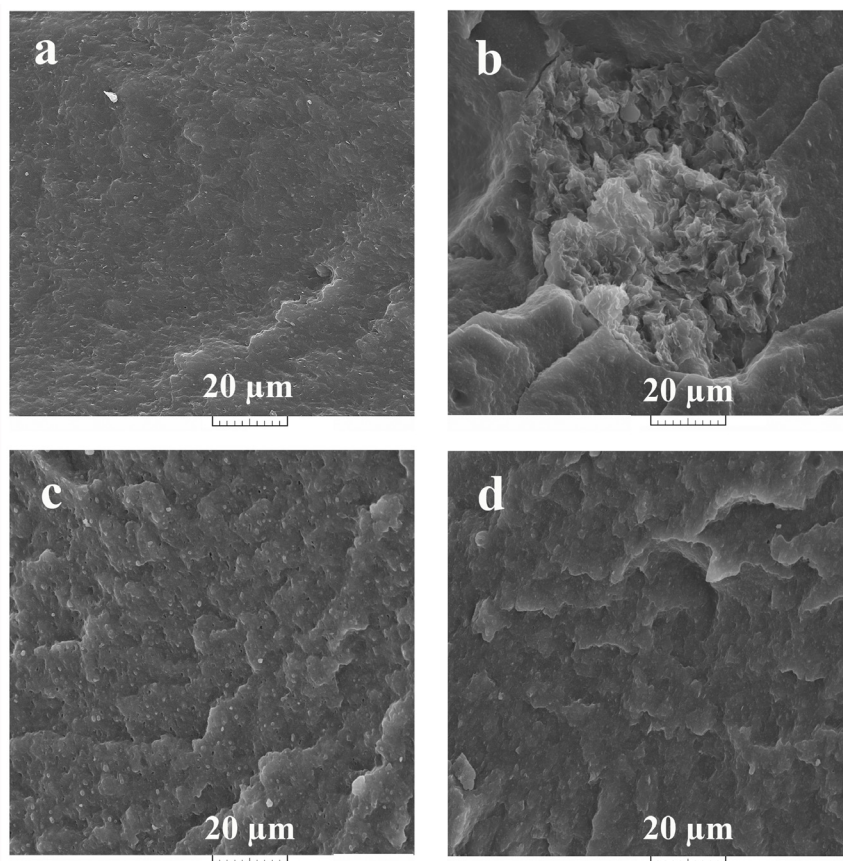
### 2.6. Dielectric breakdown strength

The characterization of the DBS was carried out according to IEC 60243-1 using a semiautomatic dielectric test set (DTS90, BAUR), equipped with copper parallel plates as electrodes. For the room temperature estimation, insulating oil was used as an embedding medium. Between the two electrodes, the specimen with a thickness of 1 mm was held, and an incremental alternating voltage of 3 kV/s was applied until the specimen could no longer withstand the flow of current, therefore the electrical breakdown takes place. The breakdown voltage was determined as the value at which the electric breakdown (represented by sparking) occurs. Notice that all of the reported values were the peak voltage ( $V_p$ ). For every sample, 20 different locations were measured.

## 3. Results and discussion

### 3.1. Microstructural studies

As shown in our previous report [19], nanoclay was efficiently distributed and due to good compatibility between EPDM rubber chains and Cloisite 15A surfactant (2M2HT1), an exfoliated morphology was obtained in the dispersed EPDM phase. Consequently, higher filler-rubber interaction leads to clay migration into the EPDM phase by blending, enhancing its elasticity. The polarity differences between Cloisite 15A and SR, on the other hand, caused nanoclay particles to become agglomerated. As shown in Fig. 1, In the Q70E30C0 sample that is a blend of SR and EPDM blend has a droplet-matrix morphology and good dis-



**Fig. 1.** SEM images of the samples, a) E100C9, b) Q100C9, c) Q70E30C0, d) Q70E30C9.

persion of EPDM is observed in the SR matrix. The incorporation of nanoclay filler into the SR/EPDM blend led to an enhanced distribution of the dispersed phase of EPDM in the SR matrix (Q70E30C9). The stabilization effect of the nanoclay in the blend could be inferred to be the result of a decrease in interfacial tension; moreover, the rigid layer of nanoclay prohibits the dispersed droplets from coalescing through steric hindrance [20, 21].

The change from spherical EPDM droplets to oval shapes could be described by the relationship of elasticity and viscosity of the phases. The morphology evolves towards a co-continuous configuration because of the nanoclay presence in the dispersed phase and matrix and the consequent drop in viscosity. Additionally, owing to their high interactions, the trapping of nanoclay in EPDM led to an increase in the elasticity of this phase, a decrease in shape relaxation associated with the droplets, and maintaining the oval-shaped droplets. Therefore, nanoclay showed a potential compatibilizing effect in these nanocomposites [22].

### 3.2. Surface and volume resistivity

Besides analyzing the electrical performance of different components to define industrial applications, the electrical characterization can be used as a suitable complement to study the molecular behavior of polymers, as well as their thermal and optical properties in combination with mechanical properties. The key electrical properties that determine the characteristics of insulating material are dielectric constant, dielectric strength, and surface and volumetric resistance.

As observed in Fig. 2, the surface and volume resistance of all samples were decreased with increasing filler content but were still maintaining a high value. No polymer is entirely free of conduction mechanism; therefore, all of them are able to have a low number of charge carriers. Conduction is often carried out by impurities that introduce charge carriers in the form of electrons or ions. Therefore, the addition of impurities such as clay, which can supply ions and electrons in the bulk of materials, reduces its electrical resistance. In addition, the electrical

resistance of EPDM is about 10 times that of SR, and adding it to the mixture increases its electrical resistance. Khodaparast et al. [23] reported that the reason for the decrease in electrical resistance of  $\text{TiO}_2/\text{PVDF}$  nanocomposite obtained from in situ method compared to commercial nanocomposite was the presence of ion and electron supply introduced from impurities left by the sol-gel method. By studying the electrical properties of EPDM and clay nanocomposites, Kang et al. [24] showed that the addition of impurities plays an important role in electrical conductivity.

Hidayah et al. [25] observed that the addition of nanofillers reduced the volume resistivity of low-density polyethylene (LLDPE)/SR nanocomposites, regardless of the kind of filler. The volume resistivity of all forms of nanocomposites also decreased significantly with increasing SR content. Nevertheless, the addition of a larger amount of nanofiller had a negligible influence on its volume resistivity. Rajini et al. [26] produced reinforced hybrid polyester composites using coconut sheath/clay. The findings revealed that resistivity values were reduced with the inclusion of MMT nanoclay. It was discussed that, owing to the conductive properties of crystalline clay materials, the inclusion of clay allows the movement of electrons in the clay/polyester blend.

### 3.3. Dielectric constant

According to Fig. 3, the dielectric constant of all nanocomposites has increased with the increase of nanoparticles due to the high number of polar groups and the increase of polarization capability. Applying an electric field to a polymer causes the redistribution of any charge on the polymer, which makes them mobile enough to respond within the period of the applied field. If some moving charges can penetrate through the sample and migrate through the interface of sample and electrode, a DC conduction is often created, and if the penetration of moving charges under the field is spatially limited, polymers can be polarized by the field and DC conduction is not observed. The dielectric constant ( $\epsilon'$ ) of a material is the relationship between the capacity of a capacitor in

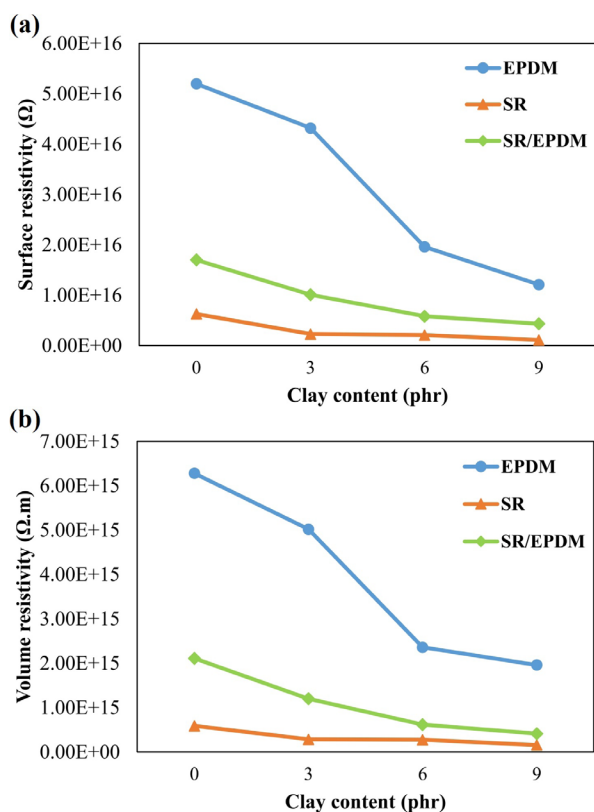


Fig. 2. The surface and volume resistance of samples.

the presence of that material and the same capacitor in the vacuum. The capacity of a capacitor measures its ability to store charges. For a non-polar material, the increase in capacity is related to the charges on the capacitor, which polarizes the molecules and absorbs the positive charges on the molecules at one end and the negative charges at the other end, resulting in an increase in charges on the surface. Therefore, the greater the polarization capacity of the molecules, the greater the dielectric constant of the material. The improvement in nanocomposite dielectric features attributed to the addition of nanoclays depends on the quality and proportion of the nanoclay [27]. The filler is polarized by the introduction of an external electric field, causing a charge disturbance in the nanoclays. In this situation, on the surface of the filler, the charge carriers could be redistributed, and hence the nanocomposite interface becomes polarized, resulting in interfacial polarization. The density of the interfacial polarized electrical charge relies on both the filler and matrix properties.

Khodaparast et al. [23] also observed that a better filler distribution and no agglomeration in the samples increased the effective interface between the nanoparticles and the polymer, and there is evidence that a greater interface formed between the filler and polymer chains leads to increased dielectric constant and it can trap charge carriers. Su et al. [28] reported the improvement of dielectric constant in EPDM composites filled with graphite flakes due to interfacial polarization. Razzaghi et al. [29] worked on SR/clay nanocomposites and showed an increase in the dielectric constant by the addition of nanoclay due to the interfacial polarization between clay and SR. Arya et al. [30] also reported the improvement of the dielectric constant by adding MMT to polymer nanocomposite films based on blend poly(ethylene oxide)-polyvinyl chloride (PEO-PVC).

Clay-filled SR composites were designed by Sarkarat et al. [31] and their electrical characterizations revealed that the neat SR dielectric constant was about 2.9 over the frequency spectrum implemented. The relaxation peak in the dielectric loss was detected along with an increase in

Table 2.  
Weibull parameters for the composites

Samples	Scale parameter $\alpha$ (V)	Shape parameter $\beta$	R <sup>2</sup>
Q100C0	2.87	9.33	0.97
Q100C9	3.00	8.09	0.87
E100C0	3.57	8.49	0.96
E100C9	3.63	19.14	0.95
Q70E30C0	3.08	10.98	0.97
Q70E30C9	3.20	14.35	0.95

dielectric permittivity at a frequency <100 Hz with the inclusion of 2.5 % of clay in the SR mixture. Rana et al. [9] also found that the conductivity values and dielectric constant of nanocomposites are higher than those of neat EPDM rubber. Thus, it has been shown that adding clay to rubber matrices improves the electrical characteristics.

### 3.4. Dielectric breakdown strength

Fig. 4 illustrates the dielectric strength (dielectric breakdown) of the composites. As mentioned above, in insulating materials, a low level of conduction can exist in different ways. This is usually due to the presence of impurities that introduce low charge carrier concentrations, such as ions or electrons, into materials. In strong electric fields, new charge carriers may also be injected into the polymer by the electrodes, hence, with the increase in voltage, the current will increase more rapidly compared to what is expected based on Ohm's law. In very intense fields, this phenomenon, besides other processes, often leads to surface conduction and hence, the complete polymer failure as a dielectric material (dielectric breakdown). The maximum voltage that a dielectric material is able to tolerate for a long time without failure is known as dielectric strength and can be defined as the voltage divided by the thickness of the material. In other words, dielectric strength is the maximum electric field that the material can withstand indefinitely. As Fig. 4 shows, increasing the amount of clay increases the dielectric strength of the nanocomposites. This can be explained by the fact that more obstacles and traps are created in the path of space charges. Spatial charge in a dielectric material means the accumulation of electric charges near its interface with the electrode. The accumulation of charge creates an internal electric field and reduces the current. Electrons can be trapped in a solid, therefore, trapping more charges in the material results in increasing the opposite electric field, and reducing the flow of electrons from the electrode to the material, thus increasing the dielectric strength [32, 33].

The scheme of the spatial charge phenomenon is shown in Fig. 5. Since EPDM has a higher dielectric strength than SR (almost double), its addition to SR has significantly enhanced the blend's dielectric strength and nanocomposite. Dong et al. [32] reported that increasing the amount of TiO<sub>2</sub> nanoparticles in silicon rubber reduced the electrical resistance due to the increase of the impurity content by introducing ions and electrons in the polymer matrix. Additionally, the dielectric strength increased due to the increase of obstacles in the way of electrons and, consequently, the increase of the space charge. Also, Hidayat et al. [25] reported that the DBS of LLDPE/SR loaded with Si, BN, and Zn nanofillers are better in all blended formulations relative to the related compositions without nanofillers. Dutta et al. [34] studied polyethylene octene elastomer/ethylene-vinyl acetate copolymer (POE/EVA) reinforced with organoclay particles. The findings revealed that the nanocomposite containing 0.5 wt% of organoclay showed an improvement in the breakdown voltage value from 6 kV to 10 kV. Besides, Shah et al. [35]



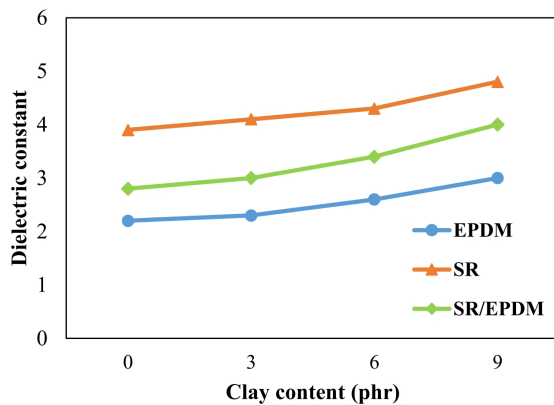


Fig. 3. The dielectric constant of the nanocomposites.

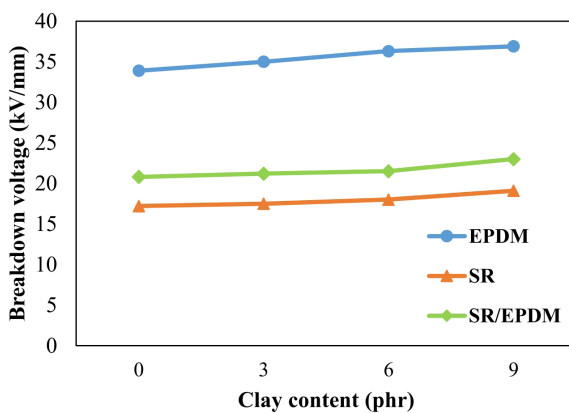


Fig. 4. The dielectric strength (dielectric breakdown) of the nanocomposites.

observed a noticeable increase in high-density polyethylene (HDPE) dielectric breakdown strength from 33 kV/mm to 68 kV/mm after processing organic clay with the mixture of titanate and silane coupling agents.

The distribution of breakdown voltages was fit to the distribution of Weibull. The value  $F$  is the Weibull cumulative distribution function, which [36, 37] is obtained from Eq. 2:

$$F(V_{BD}) = 1 - \exp\left[-\left(\frac{V_{BD}}{\alpha}\right)^\beta\right] \quad (2)$$

where the scale parameter of  $\alpha$  denotes the voltage at which 63% of all capacitors have broken down, and the Weibull module indicating the distribution width is represented by the shape parameter  $\beta$ . Usually, by taking two logarithms, the function of Weibull cumulative distribution is rearranged as:

$$\ln\ln[-\ln(1-F)] = \beta \ln(V_{BD}) - \beta \ln\alpha \quad (3)$$

Using Eq. 3, the y-intercept ( $-\beta \ln \alpha$ ) and slope ( $\beta$ ) are easily measured from a plot of  $\ln[-\ln(1-F)]$  versus  $\ln(V_{BD})$ . Results that match the distribution of Weibull would fall along a line (Fig. 6). As reported in Table 2, linear regression was utilized to evaluate alpha and  $\beta$  for the specimens. For low probability events (low  $V_{BD}$ ), the transformed data inspection indicates deviations from the Weibull distribution. This tail, which comprising below 10% of the results, could imply the presence of a separate low-voltage breakdown dominant mechanism or the presence of a threshold voltage under which the samples do not experience a breakdown.

The competitive advantage associated with nanocomposites may be

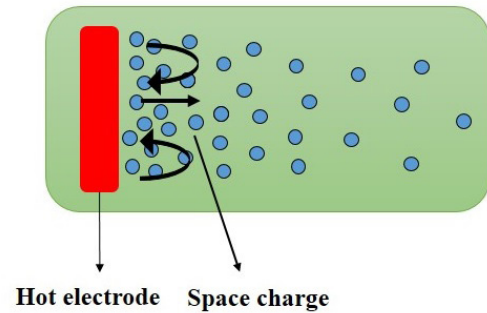


Fig. 5. Schematic of the spatial charge phenomenon.

described by the nanofiller effect, which was assumed to interfere with the consistency of the pathway given to the charge carriers, leading to increased breakdown strength. The significant number of small particles and the vast interfacial area could also serve as scattering sites for electrons, decreasing their momentum and raising the breakdown voltage [38]. The decrease in the inner bulk charge in the nanocomposite structure may also be a potential explanation for enhancing the DBS of SR/EPDM nanocomposite after nanofillers have been added. It was attributed to the addition of a regional conducting route by the overlap of nanometric double layers or localized charge flow. This could keep inner localized fields low and help to increase DBS [39].

The most remarkable point was that the Weibull shape variable,  $\beta$ , was increased for the nanocomposites, as shown in Table 2. The findings indicate that in nanocomposites rather than the unfilled SR/EPDM blend, a less disparate or relatively consistent dielectric breakdown process happened indicating an improvement in the uniformity of the sample (material with a lower weak point, typically composed of macrovoids). The existence of nanoclays could have pervaded the formed voids in the SR/EPDM. Li et al. [40] reported that the incorporation of aligned nanoplatelet fillers, in addition to any enhancements attributable to the crystal orientation, produced an extra and complementary boost in the dielectric breakdown strength. Thus, in the nanostructured matrix, with the coordinated fillers and arranged polymer crystals functioning as a macroscopic shield formed across the specimen, a significantly enhanced breakdown strength could be achieved. The findings of Hizioglu et al. [41] also showed that the breakdown strength of the natural polypropylene filled with nanoclay was greater than that of the unfilled polypropylene, and the optimal nanoclay density tended to be 2% by weight. A substantial variation in DBS value was found from the findings of the nanocomposite specimens produced by various blending processes. The DBS value has been found to be overly reliant on the relative dispersion of nanofillers in the composite mixture [14]. Furthermore, it is commonly assumed that new gaps or defects can be formed into polymers by introducing nano- or micro-particles [42]. Therefore, to boost DBS, it is crucial to monitor the distribution of nanofillers when they are introduced into the composite.

## 4. Conclusions

SR, EPDM, and SR/EPDM hybrid nanocomposites containing organically modified clay (Cloisite 15A) were developed. Thus, the specimen morphology, surface and volume resistivity, and dielectric properties of different SR/clay, EPDM/clay, and SR/EPDM/clay hybrid nanocomposites were studied. The findings were as follows:

The addition of nanoclay leads to a fine EPDM distribution in the SR matrix. The stabilization effect of the nanoclay is followed by an interfacial tension reduction; also, the rigid layer of nanoclay prohibits the dispersed droplets from coalescing and offers steric hindrance.

If the filler content increases, the volume and surface electrical resis-

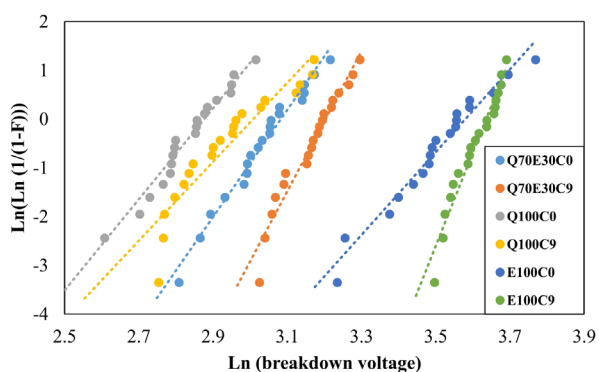


Fig. 6. Weibull distribution of breakdown voltages of the nanocomposites.

tance of all specimens are reduced, but they still maintain fairly strong electric resistance. The presence of impurities, including clay, which can provide the bulk of materials with electrons and ions, decreases their electrical resistance.

The increase in polar groups produced by a larger quantity of nanoparticles increases the dielectric constant of all nanocomposites.

An increase in the clay content improves the dielectric strength of the nanocomposite since the route of space charges develops more barriers and traps. In a dielectric substrate, spatial charging implies the storage of electrical charges along with its electrode interface.

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