

## Electrical treeing and partial discharge characteristics of silicone rubber filled with nitride and oxide based nanofillers

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### ABSTRACT

This article presents a study on electrical treeing performances with its associated partial discharge (PD) and the influence of filler concentration in silicone rubber (SiR) samples which are filled with silicon dioxide (SiO<sub>2</sub>) and silicon nitride (Si<sub>3</sub>N<sub>4</sub>) as nanofillers for electrical tree growth suppression. There are many researches on electrical treeing in SiR with SiO<sub>2</sub> nanofillers but none of the publication have reported on Si<sub>3</sub>N<sub>4</sub> nanofillers for suppression of the electrical tree growth. In this study, the treeing experiments were conducted by applying a fixed AC voltage of 10 kV and 12 kV at power frequency of 50 Hz on unfilled SiR, SiR/SiO<sub>2</sub>, and SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites with different filler concentrations by 1, 3, and 5 weight percentage (wt%) and the treeing parameters were observed with its correlated PD patterns. The outcome from this study found that the SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites were able to withstand the electrical treeing better than the pure SiR or SiR/SiO<sub>2</sub> nanocomposites. Furthermore, the increase in filler concentration improved the electrical tree performances of the nanocomposites. This finding suggests the Si<sub>3</sub>N<sub>4</sub> can be used as filler in polymeric insulating materials for electrical tree inhibition. Meanwhile, the PD activity shows increment when the tree progresses thereby indicating correlation in both parameters which can be as key parameter for monitoring unseen treeing in non-transparent samples.

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## 1. INTRODUCTION

Electrical treeing is a pre-breakdown phenomenon caused by endless electrical stress in polymeric insulations. Triggered by partial discharge (PD), electrical treeing occurs under electrical stress through tree like paths. The growth of an electrical tree consequences in the formation of a conductive path between the high-voltage and grounded parts of the insulation, therefore causing breakdown [1, 2]. Electrical tree activities have also been described to take place within the areas of weak points such as cable accessories due to electric field localization which is accompanied by PD. In application, cable accessories such as joints, terminations, and stress cones are usually made by silicone rubber (SiR). SiR has characteristics of both inorganic and organic materials. It also offers a number of benefits that not found in other organic rubber such as hydrophobic property, high temperature resistance, electrical insulation, good chemical stability, and flame retardants but it has poor solvent resistance and mechanical properties [3, 4]. In order to enhance the electrical tree resistance in the high-voltage cable accessories, different methods have been used by

manufactures of SiR, such as improving material treatment, adding treeing inhibitors or fillers, modifying the material, etc [5, 6]. Adding nanoparticles has lately drawn great attention with the potential of improving the physical, chemical, mechanical and electrical properties. Polymers such as Polyamide (PA), polyethylene (PE) and epoxy resin, etc. combined with nanoparticles including MgO, TiO<sub>2</sub>, and, SiO<sub>2</sub>, etc. have been extensively studied [7-14]. Jamil et al examined the electrical tree propagation of SiR filled with organo-montmorillonite (OMMT) and found that the nanoparticles were able to slow down the progression of the electrical treeing [15].

Hosier et al studied on low-density polyethylene (LDPE) polymer which used silicon dioxide (SiO<sub>2</sub>) as its filler and compared it with silicon nitride (Si<sub>3</sub>N<sub>4</sub>) as the filler. It stated Si<sub>3</sub>N<sub>4</sub> provided potential pluses over an oxide based system through a reduction of surface hydroxyl groups and leading potentially to a composite with dielectric properties that are considerably less influenced by absorbed water or environmental effect. They found that nitride composites shows improved breakdown strength over oxide composites in all conditions they tested which are dry, ambient and wet [16, 17]. In spite of that improvement shown by Si<sub>3</sub>N<sub>4</sub> as a filler, there are no researches have been carried out on the electrical tree performances associated with PD in SiR added with Si<sub>3</sub>N<sub>4</sub> nanoparticles. Therefore, this paper presents electrical treeing associated with PD studies in SiR added with Si<sub>3</sub>N<sub>4</sub> to suppress the growth of electrical treeing. In addition, the propagation length of the electrical trees was studied in order to investigate the performance of the SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites as well as the structure and the growth speed of trees and the electrical tree characteristics collectively discussed.

## 2. RESEARCH METHOD

### 2.1. Material used

The host polymer material is Sylgard 184 Silicone Elastomer which low in viscosity with dielectric strength, tensile strength and tears strength being 24 kV/mm, 6.2 MPa and 2.7kN/m respectively. The silicon dioxide is supplied by Sigma Aldrich which is was fumed silicon dioxide with an average size of 12 nm. Silicon nitride is supplied by NanoAmor with average size of 15-30 nm. Hardener used is dimethyl methylhydrogen siloxane (DMS) which mixed with polymer in the ratio of 10: 1 (silicone rubber: hardener) [18].

### 2.2. Preparation of silicone rubber nanocomposites

The silicone rubber (SiR) Sylgard 184 was used in form of leaf-like specimen. The needle-plane electrode geometry was engaged for initiation and propagation of electrical treeing. The gap distance between the needle tip and the plane electrode was adjusted to 1 mm. Silicon dioxide (SiO<sub>2</sub>) and silicon nitride (Si<sub>3</sub>N<sub>4</sub>) nanoparticles were dried at 100 °C for 24 hours using conventional vacuum oven to remove moisture. After preconditioning of nanofillers, it was mixed with the SiR varies by 1, 3 and 5 weight percentage (wt%) using magnetic stirrer at room temperature for 30 minutes at 60 rpm. Then, the nanofillers were dispersed using an ultrasonicator to obtain a homogeneous dispersion for 1 hour [19]. After that, the SiR nanocomposite compound were mixed with its hardener (ratio 10:1) for 30 minutes at 125 rpm using a magnetic stirrer at room temperature. The specimen was cured at 100 °C for 45 minutes. The specimen was prepared in form of leaf-like as shown in Figure 1.

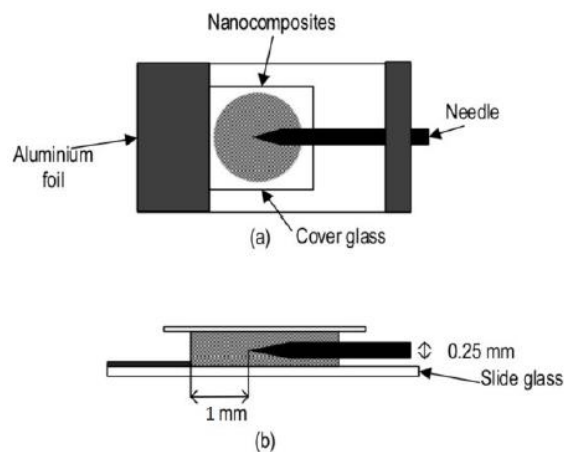


Figure 1. Configuration of leaf like specimen (a) top view (b) side view

### 2.3. Test sample preparation

To study the electrical treeing and its partial discharge (PD), online monitoring system was developed in this work which consists of a stereomicroscope, an oscilloscope, a personal computer, and a charge-coupled device (CCD) camera as shown in Figure 2. The system consisted of an Olympus SZX16 Research stereo microscope that equipped with auxiliary DP 26 Olympus CCD camera with 115x magnification capability which sufficient to capture magnified images of electrical tree propagation. Measuring impedance was used to detect the PD pulses together with the Coupling-capacitor which is the voltage divider so the voltage does not rise on the impedance of the PD [20]. The purpose of this procedure was to observe electrical treeing growth optically and its associated PD at room temperature.

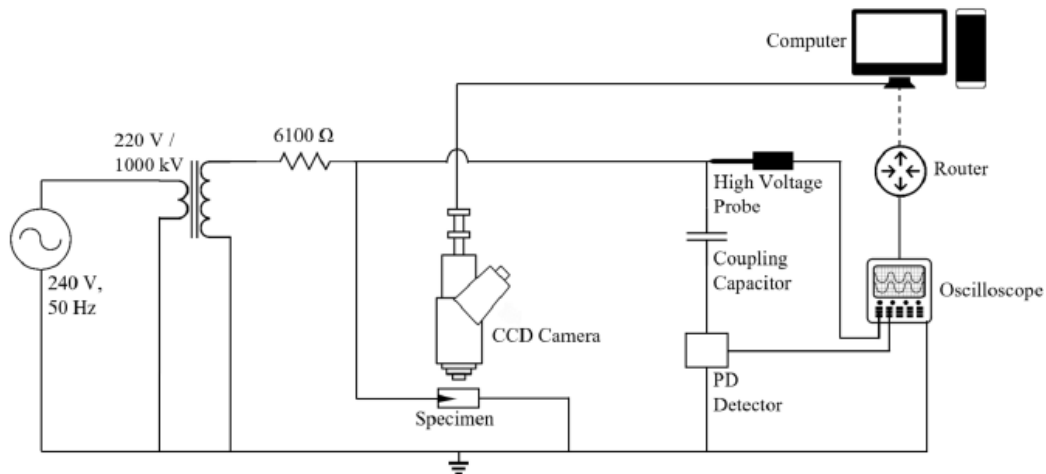


Figure 2. Schematic diagram of experimental setup for electrical treeing studies

The experiments were conducted to record the tree growth and PD by applying a constant AC voltage of 10 kV and 12 kV at 50 Hz which the voltage is monitored using oscilloscope. The samples were placed inside an acrylic cell containing silicone oil to avert surface flashover. Tree growth was constantly observed using the specially established method. The microscope and the CCD camera were interfaced through a computer. The images of electrical treeing at test voltage were captured and recorded. During electrical tree growth, the real time images of electrical tree were taken using CCD camera fixed at stereo microscope with the assist of CellSens digital imaging software. The PD reading was continually monitored by PD detector and oscilloscope which then recorded to the computer by LabVIEW programme.

### 3. RESULTS AND ANALYSIS

The tree initiation time, tree bridging time, tree propagation length and partial discharge (PD) of the investigated nanocomposite samples were analysed. Figure 3 shows the photograph of the electrical tree captured by the microscope at different stages.

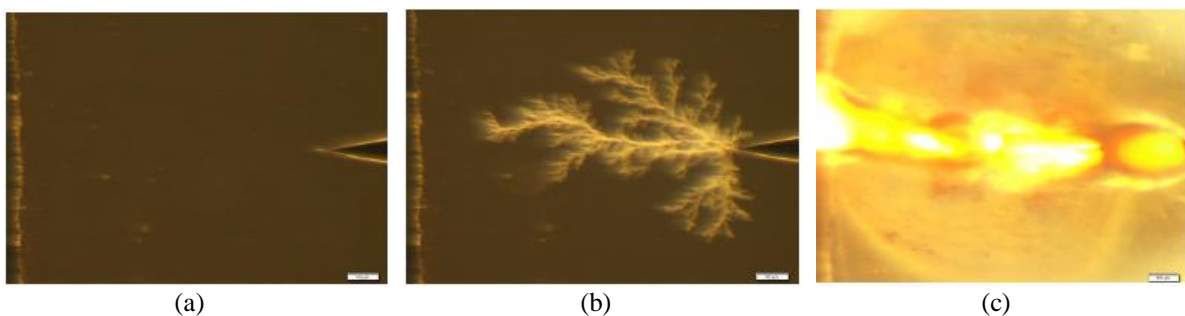


Figure 3. Electrical treeing (a) initiation stage (b) propagation stage and (c) bridging stage

### 3.1. Tree initiation time

The tree initiation time (Ti) of an electrical tree is the time at which small, observable trees initiate at the needle tip. Figures 4(a) & 4(b) show the Ti of the electrical trees of SiR, with nanofillers of SiO<sub>2</sub> and Si<sub>3</sub>N<sub>4</sub> that varies by 0, 1, 3, and 5 wt%. Under 10 kV injection, the average Ti for unfilled SiR was 7.34 s, while the average Ti for SiR/SiO<sub>2</sub> were 6.04 s, 5.68 s, and 8.99 s for 1, 3, and 5 wt%, respectively; the SiR/Si<sub>3</sub>N<sub>4</sub> showed a faster Ti of 5.28 s, 5.68 s, and 7.57 s. Meanwhile, with 12 kV injection, the average Ti for pure SiR was 6.86 s, while the average Ti for SiR/SiO<sub>2</sub> were 6.86 s, 6.75 s, and 10.65 s for 1, 3, and 5 wt%, respectively; the SiR/Si<sub>3</sub>N<sub>4</sub> showed a faster Ti of 6.93 s, 6.04 s, and 7.10 s respectively. Results from this statistical analysis show that for the Ti, only a small difference exists between the pure and two different nanofiller types. Most of the initiation times are only between 5 to 12 seconds.

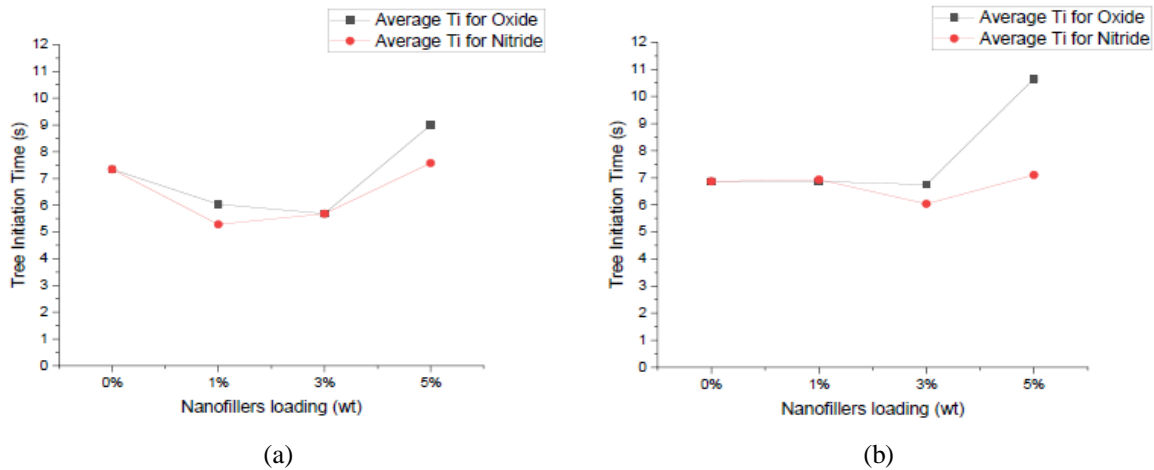


Figure 4. Tree initiation time for Pure, SiR/SiO<sub>2</sub>, and SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites with varies loading levels at voltage of (a) 10 kV and (b) 12 kV

The result shows that the tree initiation times of the nanocomposites varied based on the nanofillers type and filler loading used. There is an increment in the initiation time for the 5 wt% loading sample; however, the difference is too small. This result is different with the result from Jamil et al.'s work; which they showed that even small amounts of added nanofiller led to significant improvements in tree initiation times as the nanocomposites with nanofillers took a long time to initiate compared to unfilled SiR. This finding difference occur as the current study use a shorter distance of 1 mm between the high-voltage and ground compared to the previous study that use a longer distance which is 2 mm. Generally, the nanocomposites played a role in prolonging tree initiation times compared to unfilled polymer, because the trapped charges needed higher energy to be extracted from the polymer; more time was thus required for the tree to initiate within the nanocomposites [21]. However, if the distance between the high-voltage and ground is too short, energy needed for the trapped charges to be extracted from the polymer is lower; thus the tree is easier to initiate within the nanocomposites.

### 3.2. Tree propagation length

Figure 5 shows the result of electrical tree performance for the SiR nanocomposites with different nanofillers at 1, 3, and 5 wt% filler loading. The unfilled SiR showed the fastest propagation rate, followed by SiR/SiO<sub>2</sub> nanocomposites. SiR/Si<sub>3</sub>N<sub>4</sub> exhibited the slowest propagation time. Similar results were observed for the rest of the samples. It can be seen from Figure 5 that the tree propagation increased with an increase in nanofillers loading. These results are in line with other studies that have been conducted in which higher filler concentrations led to densely packed structures for resisting electrical tree progression. The tree channels needed to propagate through the interfaces of the nanofiller in the packed structures of the polymer, which resulted in longer propagation times [11, 22-24]. From Figure 5, it is suggested that Si<sub>3</sub>N<sub>4</sub> nanofillers has a strong ability to slow down the electrical tree growth. This result agrees with Hosier et al.'s work, in which Polyethylene/nitride nanocomposites exhibited higher breakdown voltage, thus indicating that nitride nanocomposites has ability in resistance against electrical treeing [16].

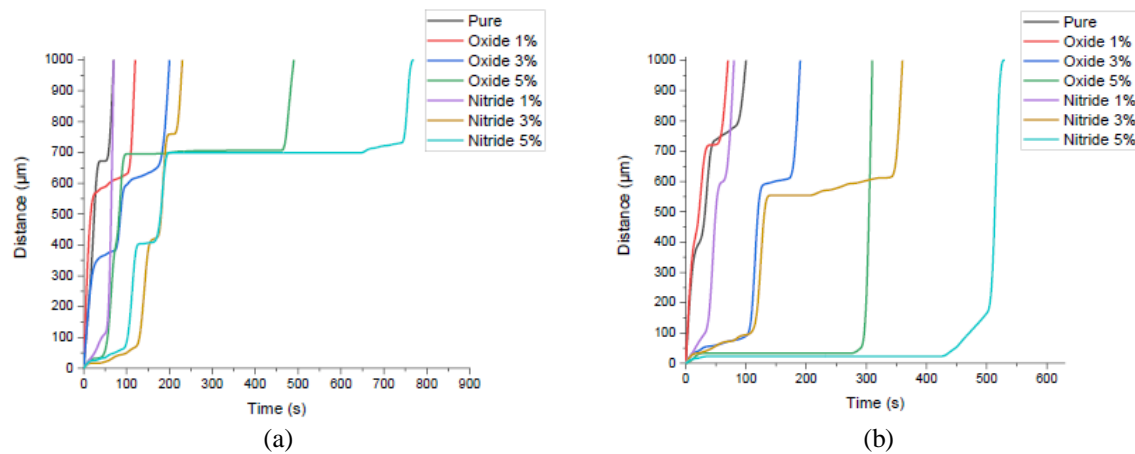


Figure 5. Tree propagation lengths across nanocomposites with varies loading levels at voltage of (a) 10 kV and (b) 12 kV

### 3.3. Tree bridging time

Figure 6 shows the average tree bridging time ( $T_b$ ) for Pure, SiR/SiO<sub>2</sub>, and SiR/Si<sub>3</sub>N<sub>4</sub> with different nanofiller loadings.  $T_b$  is the time taken for the tree to reach the ground electrode, which in our study was at 1000 µm from the high-voltage electrode. It is confirmed that both SiR/SiO<sub>2</sub> and SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites improved the tree bridging time compared to unfilled SiR.

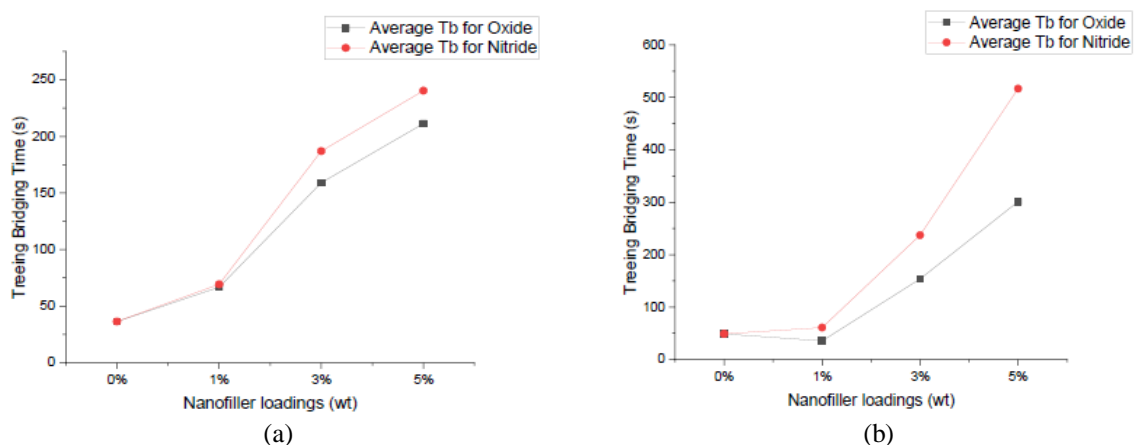


Figure 6. Tree bridging time for Pure, SiR/SiO<sub>2</sub>, and SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites with varies loading levels at voltage of (a) 10 kV and (b) 12 kV

From Figure 6 (a), the unfilled SiR recorded the shortest  $T_b$  result, followed by SiR/SiO<sub>2</sub> and SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites. The average  $T_b$  of the unfilled SiR was 36.45 s, while the  $T_b$  for SiR/SiO<sub>2</sub> nanocomposite were 66.74 s, 159.04 s, and 211.34 s for 1, 3, and 5 wt%, respectively. SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites showed the longest  $T_b$  of 69.30 s, 186.97 s, and 240.45 s for the same three wt%, respectively. From Figure 6 (b), the 12 kV injection show a same pattern of  $T_b$  as the 10 kV injection. The unfilled SiR recorded the shortest  $T_b$  result, followed by SiR/SiO<sub>2</sub> and SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites. The average  $T_b$  of the unfilled SiR was 48.57 s, while the  $T_b$  for SiR/SiO<sub>2</sub> nanocomposite were 35.26 s, 153.72 s, and 301.04 s for 1, 3, and 5 wt%, respectively. SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites showed the longest  $T_b$  of 60.83 s, 237.14 s, and 516.88 s for the same three wt%, respectively.

From these results, it is clear that the  $T_b$  of electrical treeing increased with an increase in the nanofiller loading. SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites showed the longest  $T_b$  when compared to pure SiR and SiR/SiO<sub>2</sub> nanocomposites. The result shows that the tree bridging time of the nanocomposites is also dependent on filler type and the nanofiller loading. Nanocomposites with nanofiller take a longer time to

break down compared to unfilled SiR. In addition, the Tb increased with an increase in nanofiller concentration. Overall, this result showed that SiR/Si3N4 nanocomposites has a longer breakdown time than the other nanocomposites. This result agrees with Xu et al.'s work, in which Polypropylene/nitride nanocomposites exhibited higher breakdown voltage and longer endurance under constant electrical stress [25].

**3.4. Partial discharge patterns**

In this study, it is discovered that when the tree structure grows longer and more compound, the intensity and frequency of partial discharge (PD) events are greater. Table 1 and Table 2 show phase resolved partial discharge (PRPD) patterns recorded along the electrical treeing growth with each dots representing single PD event. The PD activities occur generally only in the 1st quadrant (phases of 0° to 90°) and 3rd quadrant (phases of 180° to 270°), where the AC voltage rises to the positive or negative value respectively [26].

Table 1. PRPD patterns on three following AC cycles after its first PD event for a tree grown at 10 kV on the three type of samples

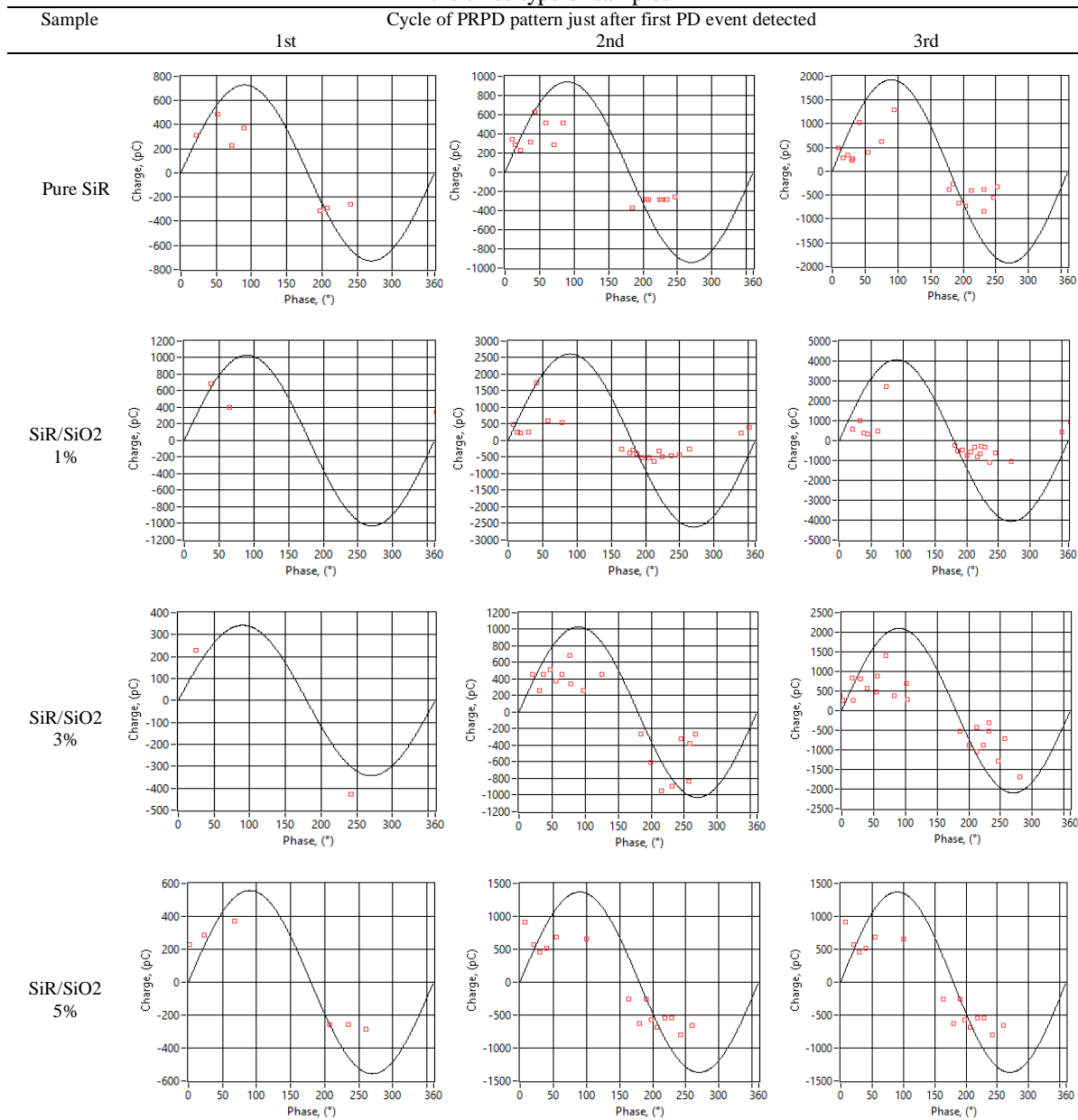




Table 1. PRPD patterns on three following AC cycles after its first PD event for a tree grown at 10 kV on the three type of samples (*continue*)

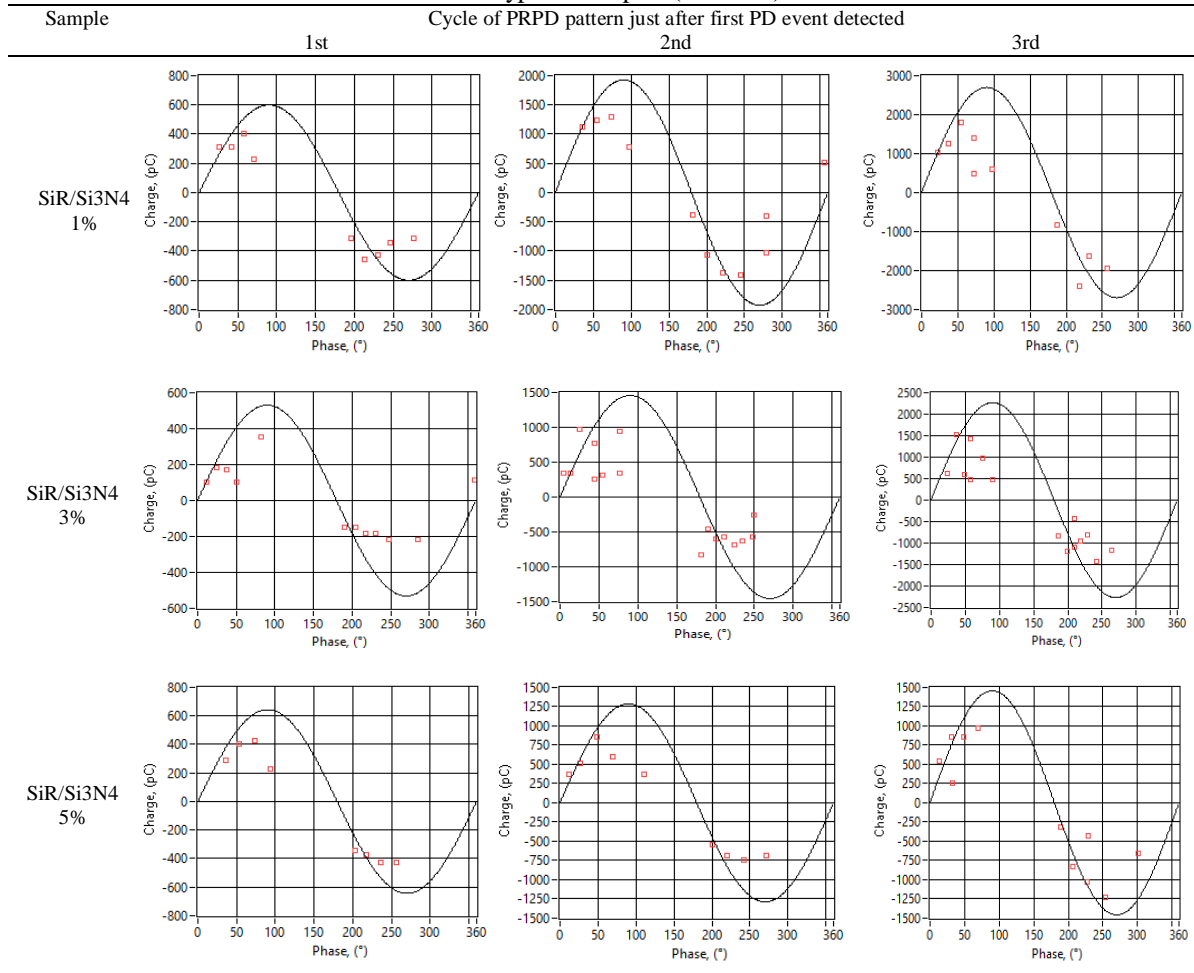


Table 2. PRPD patterns on three following AC cycles after its first PD event for a tree grown at 12 kV on the three type of samples

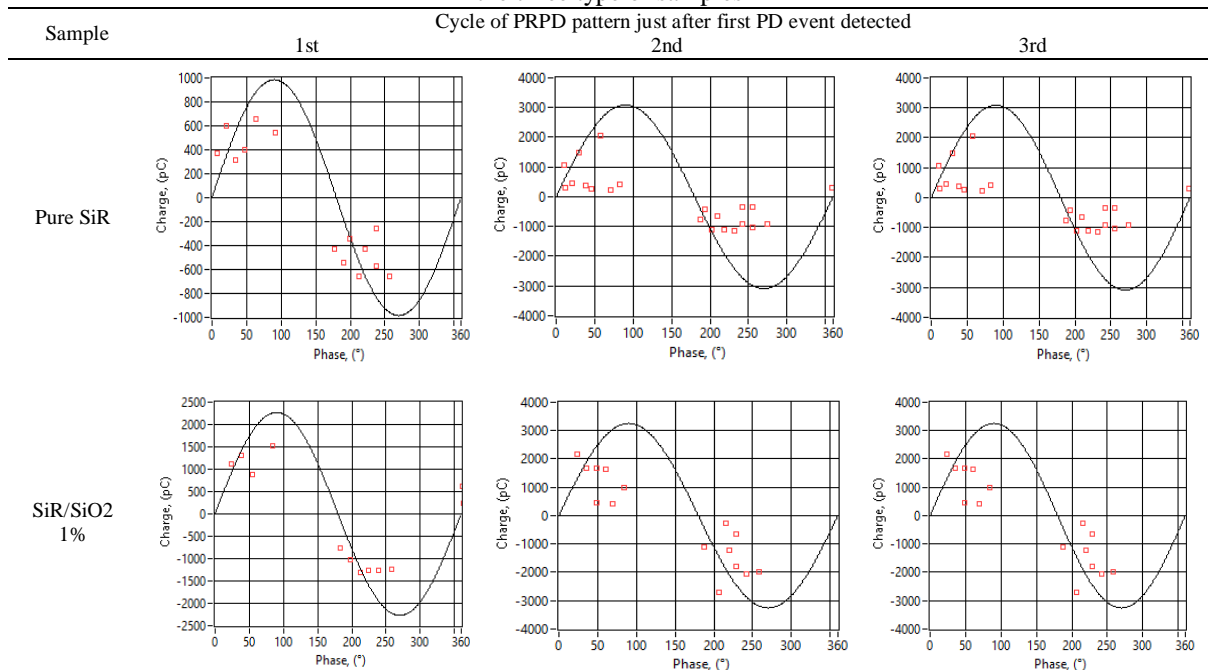
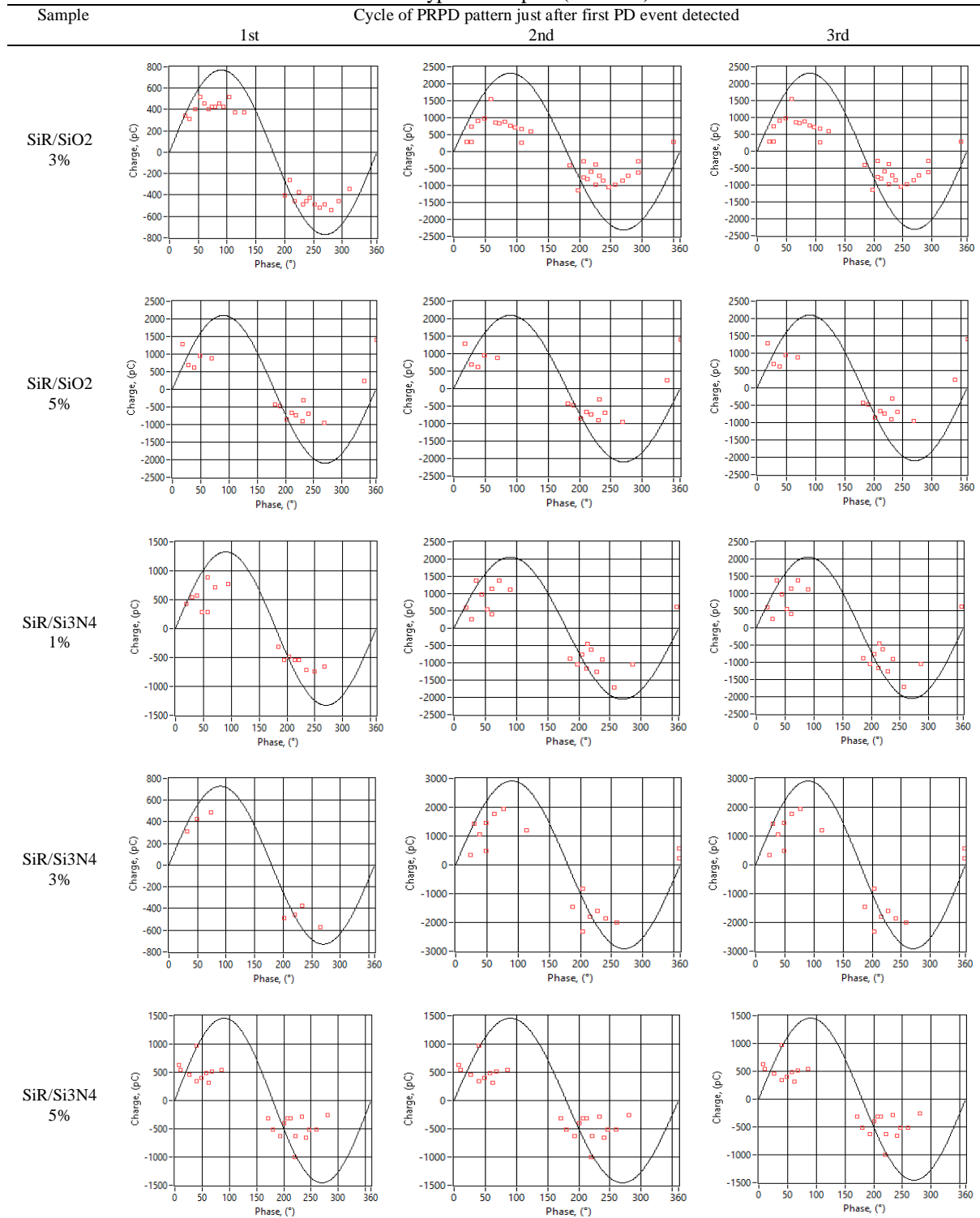


Table 2. PRPD patterns on three following AC cycles after its first PD event for a tree grown at 12 kV on the three type of samples (*continue*)



From the PRPD pattern in Table 1 and Table 2, it is noted that the intensity and frequency of PD events increased by cycle. For example, as shown in the Table 1 from the pure SiR sample, the 1st AC cycle shows only a few level discharge which is lower than 600 pC, then it builds up to greater than 600 pC in the 2nd AC cycle and finally up to more than 1000 pC in the 3rd AC cycle as the tree approaches the ground. Similar behaviour can also be seen in other samples of SiR, SiR/SiO<sub>2</sub> and SiR/Si<sub>3</sub>N<sub>4</sub> nanocomposites as shown in Table 1 and Table 2. From these patterns, it is certain that PD activity are related to the extent of



tree growth. Thus, the behaviour of tree growth in the later stage that can be shown by a high level of PD activity is potentially to be applied in plant use which is to identify the presence of damage of the insulation which is caused by the electrical treeing [26]. Moreover, the PD magnitude of 5wt% of silicone rubber nanocomposite with 5wt% showing similar pattern behaviours interestingly but the magnitude shows the lowest PD magnitude among other silicone rubber nanocomposites with 1wt% and 3wt%. This PD behaviours are linked with the electrical tree suppression behaviours as the  $\text{Si}_3\text{N}_4$  with the appropriate amount of 5wt% has contributed to the slowest propagation of treeing and the lowest PD magnitude. Therefore, this study has shown that the  $\text{Si}_3\text{N}_4$  nanofillers can be more effective than the  $\text{SiO}_2$  nanofillers in resisting the growth of electrical treeing in silicone rubber nanocomposites.

#### 4. CONCLUSION

Electrical treeing and partial discharge (PD) studies on silicon nitride ( $\text{Si}_3\text{N}_4$ ) and silicon dioxide ( $\text{SiO}_2$ ) nanofillers added to silicone rubber (SiR) focusing on electrical treeing investigation has been presented in this paper. As a result, the following conclusions were obtained; the presence of  $\text{SiO}_2$  and  $\text{Si}_3\text{N}_4$  nanofillers in SiR had slowed down the growth of electrical treeing thereby acting as electrical tree retardants. The increase in filler concentration has also resulted in better electrical treeing suppression performance. Furthermore, the improved performance of SiR/ 5 wt%  $\text{Si}_3\text{N}_4$  nanocomposite was observed in this study which that it has the best improvement on the electrical tree propagation time and tree growth rates compared to unfilled SiR and SiR/ $\text{SiO}_2$  nanocomposite. In light of this, it can be concluded that  $\text{Si}_3\text{N}_4$  nanofiller has a good potential to be used as an additive in polymeric insulating material to retard the electrical tree growth in insulation. Then, it is found that the associated PD activities intensify from the treeing initiation until the bridging. The relationship between PD activities and the electrical tree growth conclude that it is handy for diagnosis the unseen electrical trees on related high-voltage insulation material by just using PD detection method

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