Clarification of the optimum silica nanofiller amount for electrical treeing resistance

Z. Nawawi1, M. A. B. Sidik2, M. I. Jambak3, C. L. G. Pavan Kumar4, Aulia5, M. H. Ahmad6, A. A. Abd Jamil7

1,2,3Department of Electrical Engineering, Faculty of Engineering, Sriwijaya University, Palembang, West Sumatera, Indonesia
4,6Institute of High Voltage and High Current, School of Electrical Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, Johor Bahru, Johor, Malaysia
5Department of Electrical Engineering, Faculty of Engineering, Andalas University, Padang, West Sumatera, Indonesia
7School of Electrical & Electronic Engineering, Universiti Sains Malaysia, Nibong Tebal, Penang, Malaysia
*Corresponding author, e-mail: mohdhafizi@utm.my

Abstract

This paper aims to clarify the optimum amount of fumed silica (SiO2) nanofiller in resisting the initiation and propagation of electrical treeing in silicone rubber (SiR). Unlike other works, SiR/SiO2 nanocomposites containing seven different weight percentages of SiO2 nanofiller were prepared for this purpose. To achieve the objective, the electrical tree characteristics of the SiR/SiO2 nanocomposites were investigated by comparing the tree initiation voltage, tree breakdown time, tree propagation length and tree growth rate with its equivalent unfilled SiR. Moreover, the structural and morphological analyses were conducted on the SiR/SiO2 nanocomposite samples. The results showed that the SiR, when added with an appropriate amount of SiO2 nanofiller, could result in an improved electrical tree resistance. It implies that the 5 wt% of silica is the optimum amount to achieve the optimal electrical tree resistance such that above 5 wt%, the tree resistance performance has been abruptly reduced, subjected to the agglomeration issue.

Keywords: electrical treeing, nanocomposites, silica, silicone rubber

Copyright © 2019 Universitas Ahmad Dahlan. All rights reserved.

1. Introduction

Polymeric insulating materials have become an important part of high voltage apparatuses over two decades [1, 2]. The main function of polymeric insulating materials in high voltage apparatuses is to provide electrical insulation. However, degradation processes occur in polymeric materials, which subsequently jeopardize the insulation performance of the materials. Such degradation processes are normally initiated by small electrical sparks, also popularly known as partial discharges [3].

In light of the foregoing, electrical treeing is one of the partial discharge phenomena that commonly results in the degradation of polymeric insulating materials [4]. Therefore, electrical tree resistance of polymeric insulating materials needs to be improved. This can be achieved by mixing two or more different types of polymers to form polymer blend materials [5-7]. Promising improvements of polymeric insulating materials against partial discharge activities were also reported when certain amounts of inorganic/organic nanofillers were mixed with polymers [8-10].

Recently, polymer nanocomposite insulating materials are gaining popular interests from dielectric researchers in attempts to improve polymers’ resistance against electrical degradation processes with the addition of nanofillers [11-13]. In this regard, the improved resistance of nanocomposites against partial discharges is commonly attributed to the nanometer-sized nature of the fillers that contributes to the presence of large interfacial areas between the polymers and the nanofillers [14-16]. Consequently, the addition of nanofillers in polymers has led to an enhanced partial discharge resistance and electrical tree resistance of polymer nanocomposites [17]. Not only addition of nanofillers but the quantity of silica nanofiller influenced the initiation time of electrical treeing and the resistance towards electrical treeing in solid insulation [18-21].

In line with the development of nanocomposites in electrical insulation, researchers discovered that the performance of silicone rubber (SiR) could also be enhanced when
the material is added with nanofillers. SiR, a widely used polymeric material in industrial applications, has been used as a coating material for high voltage outdoor insulators especially for glasses and ceramics to help reducing maintenance costs and resist potential flashovers of the insulators [22-24]. Besides that, SiR has also been used as insulating materials for cable accessories such as to joint materials and stress cones [25, 26].

Since the use of SiR in high voltage insulation systems, many studies have been conducted to understand its role in improving insulation performance [27, 28]. However, the electrical properties, especially the electrical tree resistance of SiR, when added with SiO2 nanoparticles, are less explored. Also, the optimum formulation of SiO2 nanofillers that can effectively suppress the growth of electrical treeing in SiR has not been well studied and clarified. This article, therefore, reports on the characterization of the electrical tree characteristics of SiR based nanocomposites when added with a different weight ratio of SiO2 nanoparticles. The optimal SiR/SiO2 nanocomposite formulation towards electrical tree characteristics has been discussed accordingly.

2. Methodology
The sample preparation and experimental setup procedure are elucidated in this section.

2.1. Sample Preparation
Fumed or pyrogenic SiO2 nanofiller, obtained from Sigma Aldrich, with a specific particle size of 12 nm, was used as a nanofiller. The Sylgard 184 SiR elastomer, obtained from Dow Corning, was used as a polymer. The SiR is of the Polydimethylsiloxane (PDMS) type and belongs to a group of polymeric organosilicone compounds referred to as silicones. It is optically clear, inert, non-toxic and non-flammable.

The mixing process of SiR and fumed SiO2 nanofillers was performed with the aid of a magnetic stirrer. SiR and SiO2 nanofillers were mixed at a speed of 125 rpm for 1 hour. Ultrasonicator (Fisher, FB705BX) was used in the mixing process to ensure that the SiO2 nanoparticles were dispersed in SiR matrices. The dispersion energy, power operation, and amplitude were more than 100 kJ, between 100-150 W, and 70%, respectively.

A re-weighing process was carried out after the sonication process because some weight losses were expected during the redeployment process from one beaker to another at the mixing stage. After completing the re-weighing process, the SiR/SiO2 nanocomposites were mixed with a hardener based on 10:1 SiR to hardener ratio, for 15 minutes at 125 rpm. The SiR/SiO2 nanocomposites were further mixed using a magnetic stirrer. The next step was the degasification process to remove potential voids and bubbles formed in the SiR/SiO2 nanocomposites during the mixing process. The degasification process was performed by using a vacuum oven for 25 minutes at 27 °C. Afterwards, the SiR/SiO2 nanocomposite sample was cast onto the electrodes gap on a microscope slide glass. Then, the leaf-like specimen was covered with a microscope cover glass. The leaf-like specimen of SiR/SiO2 nanocomposites was cured at 100 °C for 45 minutes inside a vacuum oven.

Table 1 shows the compositions of SiR/SiO2 nanocomposites prepared for electrical treeing tests.

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>Nanocomposite Compositions</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Unfilled SiR</td>
</tr>
<tr>
<td>2</td>
<td>SiR+1 wt% SiO2</td>
</tr>
<tr>
<td>3</td>
<td>SiR+2 wt% SiO2</td>
</tr>
<tr>
<td>4</td>
<td>SiR+3 wt% SiO2</td>
</tr>
<tr>
<td>5</td>
<td>SiR+4 wt% SiO2</td>
</tr>
<tr>
<td>6</td>
<td>SiR+5 wt% SiO2</td>
</tr>
<tr>
<td>7</td>
<td>SiR+6 wt% SiO2</td>
</tr>
<tr>
<td>8</td>
<td>SiR+7 wt% SiO2</td>
</tr>
</tbody>
</table>

2.2. Experimental Setup
Images of electrical treeing in unfilled SiR and SiR/SiO2 nanocomposites were captured with the aid of an online monitoring system. The online monitoring system consisted of three main parts, i.e., a personal computer, a stereo microscope and a charge-coupled device (CCD) camera—the Olympus SZX16 Research Stereomicroscope equipped with an auxiliary DP-26.
Olympus CCD camera was used. The camera has 72 times magnification capability and it was interfaced to a computer manufactured by Olympus Corporation. Each test specimen was placed in a container filled with silicone oil to avoid potential flashover during the experiment.

Figure 1 shows the experimental setup for the electrical treeing study. The needle electrode of each leaf-like specimen was connected to an AC high voltage source while the plane electrode was connected to the ground. A voltage source of 220 V with a frequency of 50 Hz was used as a supply to the voltage regulator. The voltage regulator regulated the HV transformer from 0 to 30 kV. The apparent power of the HV transformer was 1.38 kVA with a maximum output current of 14 mA. A 6100 Ω, 60 W high voltage resistor was connected in series with the secondary side of HV transformer and the needle electrode. The purpose of the HV resistor was to limit the current flow when a short circuit occurred. A high-resolution field emission electron microscope (FESEM) and a Fourier transform infrared (FTIR) spectroscope was later used to study the morphology of the samples.

Figure 1. The equivalent circuit of the experimental setup

3. Results and Discussion

Treeing analysis and chemical and physical bonding study were explained in detail in this section.

3.1. Treeing Analysis

The tree inception voltage, tree breakdown time, tree propagation length and tree growth rate of the investigated nanocomposite samples were analysed.

3.1.1. Tree Inception Voltage

The dependence of the tree inception voltage of SiR on nanofiller loading levels is shown in Figure 2. The tree inception voltage was higher for all SiR/SiO\textsubscript{2} nanocomposite samples compared with the unfilled SiR. The tree inception voltage for the unfilled SiR was 9.4 kV. The tree inception voltage increased with increasing nanofiller loading levels, with the highest voltage at 19.4 kV for SiR/SiO\textsubscript{2} nanocomposites containing 7 wt% of SiO\textsubscript{2}.

3.1.2. Tree Breakdown Time

Figure 3 shows the tree breakdown time for unfilled SiR and SiR/SiO\textsubscript{2} nanocomposites with 1 wt%, 2 wt%, 3 wt%, 4 wt%, 5 wt%, 6 wt% and 7 wt% of SiO\textsubscript{2} nanofiller. The unfilled SiR recorded the earliest tree breakdown time compared with the SiR/SiO\textsubscript{2} nanocomposites. The tree breakdown time of the unfilled SiR was 151 s. For the SiR/SiO\textsubscript{2} nanocomposites, the tree breakdown time increased with increasing amounts of SiO\textsubscript{2} up to 5 wt%; the tree breakdown time for samples containing 1 wt%, 2 wt%, 3 wt%, 4 wt% and 5 wt% of SiO\textsubscript{2} loadings in SiR were 316 s, 621 s, 745 s, 1082 s and 1362 s, respectively. At 6 wt% and 7 wt% of SiO\textsubscript{2} loading levels, tree breakdown time reduced dramatically—the tree breakdown time is, however, longer than the unfilled SiR; the tree breakdown time for samples containing 6 wt% and 7 wt% SiO\textsubscript{2} became 234 s and 213 s respectively.
3.1.3. Tree Propagation Length

For the SiR/SiO$_2$ nanocomposites, the measurement of the electrical tree propagation length was done in the axial direction between the tip of the needle and the plane electrode. Based on Figure 4, the trees in nanocomposite samples with 1 wt%, 2 wt% and 3 wt% of SiO$_2$ bridged the gap of the needle and plane electrodes (2000 µm) at 400 s, 700 s and 900 s, respectively. For samples containing 4 wt% and 5 wt% of SiO$_2$ nanofiller, the electrical trees did not bridge the gap of the needle and plane electrode – these samples recorded electrical tree propagation lengths of 1877 µm and 1631 µm, respectively, after the electrical trees propagated for 1000 s. Meanwhile, the electrical trees in the samples with 6 wt% and 7 wt% of SiO$_2$ propagated rapidly until breakdown occurred. It is noteworthy that these samples were chosen as representatives of the overall electrical tree propagation length for each sample type [1].
3.1.4. Tree Growth Rate
The growth rate of electrical treeing for unfilled SiR and SiR/SiO$_2$ nanocomposites with 1 wt%, 2 wt%, 3 wt%, 4 wt%, 5 wt%, 6 wt% and 7 wt% of SiO$_2$ nanofiller loading levels is shown in Figure 5. The growth rate of electrical treeing in SiR with 0 wt%, 1 wt%, 2 wt%, 3 wt%, 4 wt%, 5 wt%, 6 wt% and 7 wt% were 11.99 µm/s, 8.55 µm/s, 4.57 µm/s, 3.86 µm/s, 3.25 µm/s, 2.58 µm/s, 68.66 µm/s and 89.21 µm/s, respectively.

![Figure 5. The growth rate of electrical treeing of unfilled SiR and SiR/SiO$_2$ samples filled with different SiO$_2$ contents](image)

3.1.5. Electrical Treeing Channel
The electrical tree channels for all investigated samples were analyzed based on the propagation of electrical trees upon the initiation of treeing. The electrical tree images for nanocomposite samples containing 1 wt%, 2 wt%, 3 wt%, 4 wt%, 5 wt%, 6 wt% and 7 wt% of SiO$_2$ and pure SiR are shown in Figures 6 (a-h), respectively. Of note, the electrical treeing for all nanocomposite samples were of the branch type.

![Figure 6. Electrical tree images for samples containing](image)

(a) pure SiR (b) 1 wt%, (c) 2 wt%, (d) 3 wt%, (e) 4 wt%, (f) 5 wt%, (g) 6 wt% and (h) 7 wt% of SiO$_2$; the measurement scale is 200 µm

3.2. Chemical and Physical Bonding Study
The chemical and physical bondings were analysed using field emission scanning electron microscopy and Fourier transform infrared spectroscopy.

---

*Clarification of the optimum silica nanofiller amount for electrical treeing resistance (Z. Nawawi)*
3.2.1. Field Emission Scanning Electron Microscopy

The dispersion state of the SiO$_2$ nanofiller in SiR was analyzed using Field Emission Scanning Electron Microscopy (FESEM). Figures 7(a–e) show SEM micrographs for nanocomposite samples containing 1 wt%, 3 wt%, 5 wt%, 6 wt% and 7 wt% of SiO$_2$, respectively. SiO$_2$ nanofillers have larger specific surface areas than the conventional particulate filled system. SiO$_2$ nanofillers are closely located with their neighboring nanofillers. Due to the small size of nanoparticles, large interfacial areas exist between the SiO$_2$ nanofillers in the SiR, which is considered to be one of the key roles leading to the unique properties of nanocomposites. This interfacial area can be considered as an interaction zone or interphase. The interaction zone at the surface of SiO$_2$ nanofillers could cause the nanofillers to be attached physically in the SiR matrices. This attachment results in the structure of SiR becoming densely packed and robust. From the micrographs, fine-sized SiO$_2$ particles can be observed in all the nanocomposite samples. Agglomerated SiO$_2$ particles can also be observed in part of the samples. As the amount of SiO$_2$ increased to 6 wt% and 7 wt%, increased nanoparticle agglomerations can clearly be observed, as in Figures 7(d) and 7(e) for the nanocomposite samples containing 6 wt% and 7 wt% of SiO$_2$, respectively.

Figure 7. SEM micrographs of nanocomposite samples containing (a) 1 wt%, (b) 3 wt%, (c) 5 wt%, (d) 6 wt%, and (e) 7 wt% of SiO$_2$
3.2.2. Fourier Transform Infrared Spectroscopy

Figure 8 shows the Fourier Transform Infrared (FTIR) spectra for the SiO$_2$ nanofiller and the nanocomposite samples containing 0 wt%, 1 wt%, 3 wt%, 5 wt%, 6 wt% and 7 wt% of SiO$_2$. The characteristic transmittance band of SiO$_2$ nanofiller can be observed at 1110 cm$^{-1}$. The spectrum corresponds to the silicone (Si-O) bonding.

![FTIR spectra](image)

Figure 8. FTIR spectra of SiO$_2$ powder and nanocomposite samples containing 1 wt%, 3 wt%, 5 wt%, 6 wt% and 7 wt% of SiO$_2$ and pure SiR

4. Discussion

The SEM micrographs showed that fine-sized SiO$_2$ particles could be observed in all the nanocomposite samples. Increasing the amount of SiO$_2$ nanofiller, however, resulted in increasing amounts of SiO$_2$ particle agglomerations. Meanwhile, the FTIR spectra indicated that the SiO$_2$ nanofiller had strong physical interactions with the SiR. The additional polymer chains that were inserted into the SiO$_2$ confined the polymer nanoscopically. The interaction between the SiO$_2$ and SiR resulted in the formation of a highly ordered structure due to the presence of neighbouring chains that surrounded mobile SiR chains. Hence, molecular orientation of the chains became higher, and further additional crosslinks in the system were formed [29].

From the electrical tree images, a densely packed electrical tree structure was observed when the amount of SiO$_2$ in SiR increased. The formation of the densely packed tree structure could cause the electrical trees to be difficult to initiate in the SiR matrices. Therefore, the tree inception voltages for all the SiR/SiO$_2$ nanocomposites were higher compared with the unfilled SiR.

In addition, SiO$_2$ nanofiller itself could also be the key factor contributing to the higher tree inception voltages and longer tree breakdown times of the SiR/SiO$_2$ nanocomposites (up to 5 wt% of SiO$_2$ loading levels) than the unfilled SiR. The presence of SiO$_2$ nanofiller could slow down the electrical tree propagation process since the nanofiller could serve as physical obstacles to curb the growth of the electrical treeing. In this regard, the electrical trees are expected to grow along the surface of the SiO$_2$ nanofiller.

Meanwhile, the presence of SiO$_2$ nanofiller in SiR could create trapping sites. SiO$_2$ nanofillers could trap charge carriers from the needle electrodes after the high voltage was applied. The trapped charge carriers required high energy to move from one trap to another trap. So, the electrical charge mobility was reduced due to the presence of the trapping sites. The reduction of electrical charge carrier mobility along the nanofiller and SiR interfaces could be a major factor in improving the dielectric properties and causing the growth of electrical treeing to become slower for the nanocomposite samples containing up to 5 wt% of SiO$_2$ nanofiller [30].
Among the nanocomposite samples, the sample with 5 wt% of SiO$_2$ showed the highest resistance to electrical treeing. This shows that an appropriate loading of SiO$_2$ nanoparticles can effectively suppress the growth of an electrical tree. For the nanocomposite samples containing 6 wt% and 7 wt% of SiO$_2$, however, much shorter electrical tree breakdown times and much higher growth rates were recorded. This could be due to the agglomerated structure of SiO$_2$ nanoparticles within the nanocomposite samples. Severe nanofiller agglomations, as depicted in Figures 7 (d) and 7 (e), could result in a much shorter path for a tree to propagate through the agglomerated SiO$_2$ surfaces.

5. Conclusion
In the investigated SiR/SiO$_2$ nanocomposites up to 5 wt% of SiO$_2$ loading levels, the electrical tree growth was slower compared with the unfilled SiR. The addition of SiO$_2$ nanofiller in SiR could, therefore, act as an inhibitor to retard the growth of electrical treeing. FTIR results showed that the SiO$_2$ nanofiller could have good physical interactions with the structure of the SiR. From the results of the electrical tree inception voltage, tree breakdown time, tree propagation length and tree growth rate, SiR/SiO$_2$ nanocomposites (up to 5 wt% of SiO$_2$ loading levels) showed advantageous electrical tree characteristics compared with the unfilled SiR.

From the current work, 5 wt% of SiO$_2$ loading was identified as the optimum amount for the SiR/SiO$_2$ nanocomposites. This nanocomposite formulation resulted in a desirable electrical tree resistance performance compared with other investigated samples. Therefore, it is generally envisaged that SiR/SiO$_2$ nanocomposites with 5 wt% of SiO$_2$ amount level can be utilized as electrical insulating materials for improving the discharge resistance performance and increasing the insulation lifetime of the silicone rubber insulating material.

Acknowledgement
Authors would like to express gratitude to Universiti Teknologi Malaysia and Sriwijaya University for funding this work under research grants (4B377, 4B340, 4B342, 07G05, and 04G81).

References

Clarification of the optimum silica nanofiller amount for electrical treeing resistance (Z. Nawawi)