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Effects of water on breakdown characteristics of polyethylene composites

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Abstract

The current work investigated the effects of oxide- and nitride-based silicon nanoparticles on the water absorption and dielectric breakdown characteristics of low density polyethylene (LDPE). The results showed that hydrophobic LDPE, when added with silicon dioxide nanoparticles, became prone to water absorption. In contrast, LDPE, when added with silicon nitride nanoparticles, attracted much less water. These water absorption behaviors consequently affected the breakdown characteristics of the materials, where LDPE with silicon dioxide nanoparticles showed lower breakdown strength than LDPE with silicon nitride nanoparticles. These differences are discussed based on the surface chemistries of the nanoparticles.

Keywords: Breakdown; Water; Polyethylene; Silicon dioxide; Silicon nitride; Nanoparticles

1.0 Introduction

Dielectric nanocomposites have been reported to possess favorable dielectric properties that the dielectric community has long been waiting for, such as improved partial discharge resistance, treeing propagation, space charge build-up and dielectric breakdown performance [1-10]. Due to the effect of nanostructuration, extensive particle surface areas are anticipated to be present around nanometer-sized fillers. Consequently, properly engineered surface states of nanoparticles, which commonly serve as the interfacial region between the nanoparticles and the polymer, would allow nanocomposites to function as unique high voltage electrical insulation materials [11-21].

Since the dielectric properties of nanocomposites are closely associated with the mechanisms at the nanoparticles' surface, several interfacial models have been suggested in attempts to explain the electrochemical features (i.e., chemical processes that result in changes in electrical properties) of nanocomposites. These include the diffuse electrical double layer model [13, 22, 23], the multi-core model [14], the multi-region structure model [24] and the dual layer model [25]. Although each of the models has different viewpoints in relation to the interfacial region of nanocomposites, they exhibit a common element: the interfacial layer most adjacent to the nanoparticles is the key to controlling the dielectric behavior of nanocomposites. This highlights the importance of controlling the surface state of nanoparticles in engineering the dielectric properties of nanocomposites.

The mechanisms at the interfacial region of nanocomposites, largely affected by the nanoparticles' surface state are, however, far from being understood. Although the presence of the interfacial region has often been regarded as the main factor leading to the distinct dielectric properties of nanocomposites, it may also act as a preferred location for the aggregation of water molecules. For example, Zhang and Stevens [26] studied the dielectric behavior of nanoalumina-filled polyethylene upon water absorption and reported that dielectric loss peaks associated with absorbed water in unfilled and nanoalumina-filled polyethylene appeared at different frequencies and had different dynamics. Similar observations were reported for the case of nanoalumina-filled epoxy in comparison with unfilled epoxy. Zhang and Stevens [26] therefore suggested that water absorption behaviors in nanocomposites were different from unfilled polymers. Specifically, the interfacial region in nanocomposites, which was not found in unfilled materials, was likely to be the site for the aggregation of water molecules.

Zou et al. [27] also discovered that epoxy/silica nanocomposites absorbed significantly more water than unfilled epoxy when exposed to humid environmental conditions. The extra water was found to be located around the surface of silica nanoparticles, resulting in the presence of water shells surrounding the nanoparticles. Conversely, filler particles that have surfaces that were functionalized to be hydrophobic reduced considerably the amount of absorbed water in nanocomposites under the same conditions of humidity. Similar observations were reported by Huang et al. [28] for the case of polyethylene/silica nanocomposites.

Meanwhile, Fabiani et al. [29] investigated the effect of nanoparticle drying on water contents of nanocomposites and reported increased permittivity and loss in samples added with wet nanoparticles compared to samples added with dried nanoparticles. These adverse dielectric effects were thought to be more pronounced in nanocomposites containing high aspect ratio fillers than in nanocomposites containing low aspect ratio fillers. Therefore, the presence of the nanoparticle/polymer interphase may negatively affect the overall dielectric performance of nanocomposites.

The recent work of Lau et al. [30, 31] demonstrated that polyethylene/silica nanocomposites absorbed significantly more water than unfilled polyethylene. Consequently, the permittivity and loss tangent of the nanocomposites increased with increasing duration of water immersion. Although the addition of nanosilica to polyethylene resulted in significant water uptake (presumably due to the presence of the interphase) compared to unfilled polyethylene, surface functionalization of nanosilica using trimethoxy(propyl)silane coupling agent was found to improve matters, with less water uptake, lower permittivity and lower loss tangent. Similar findings were reported by Hosier et al. [32] with the use of silicon nitride nanoparticles in place of functionalized silica nanoparticles. These findings lead to the suggestion that the surface states (or more precisely, the surface chemistries) of nanoparticles are vital in establishing the interfacial region of nanocomposites, which subsequently determine the materials' dielectric behaviors.

To date, published literature on the effect of water absorption in relation to the breakdown characteristics of polyethylene nanocomposites is relatively scarce. Although polyethylene itself is hydrophobic, the addition of nanoparticles could alter the water absorption behavior of the resulting nanocomposites due to the presence of the interfacial region between the nanoparticles and polymer that could act as a preferred site for the aggregation of water molecules [33]. Consequently, the presence of water can have detrimental effects on the otherwise improved electrical properties of dielectric materials [34-37]. In the current work, the tendency for hydrophobic low density polyethylene (LDPE) to absorb water upon nanoparticle addition was investigated. Two types of nanoparticles with different surface chemistries, i.e., silicon dioxide (SiO_2) and silicon nitride (Si_3N_4), were added respectively to LDPE to explore the interfacial mechanisms with regard to water absorption and breakdown characteristics of the resulting nanocomposites.

2.0 Experimental

2.1 Materials and sample preparation

The polymer used in the experimental work was LDPE (Titanlene LDF200YZ, (Lotte Chemical Titan). The nanoparticles used were silicon dioxide (SiO_2) nanopowder and silicon nitride (Si_3N_4) nanopowder, obtained from NanoAmor. The Si_3N_4 nanoparticles had a manufacturer-quoted average particle size of 15-30 nm. For the SiO_2 nanoparticles, two manufacturer-quoted average particle sizes were considered, i.e., 20 nm and 80 nm – these SiO_2 nanoparticles were referred to as $20SiO_2$ and $80SiO_2$, respectively, hereafter. Figure 1 illustrates that SiO_2 nanoparticles typically contains Si-O-Si bonds [38-41] while Si_3N_4 nanoparticles typically contains Si-N-Si bonds [42-44]; this will be further discussed later.

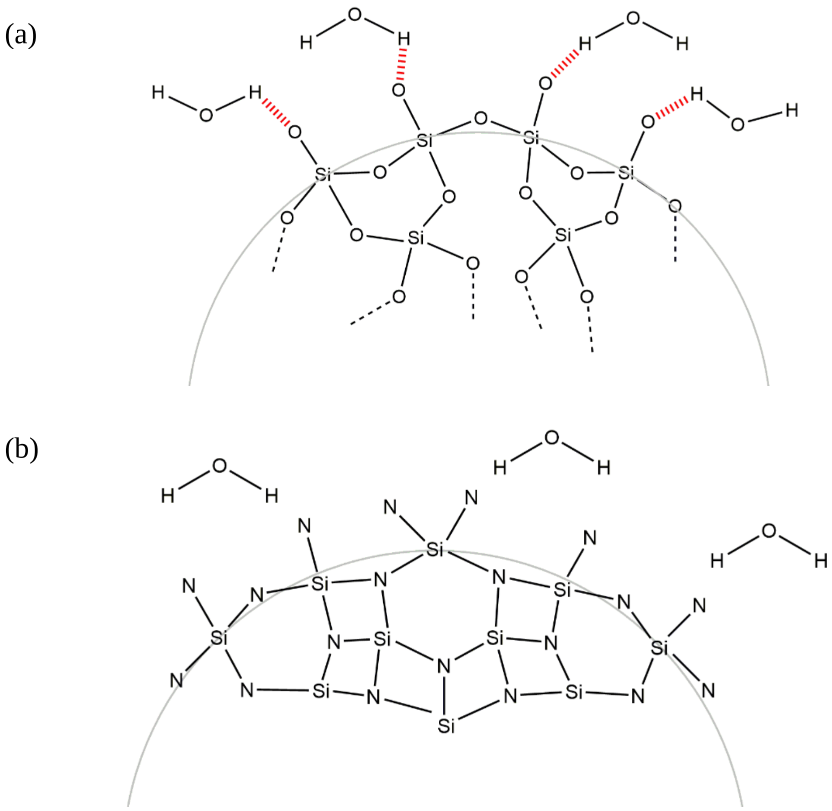

Figure 1. Surface chemistries of (a) SiO_2, (b) Si_3N_4.

Nanocomposite samples were prepared using a mechanical mixing method. The desired amount of $20SiO_2$, $80SiO_2$ or Si_3N_4 nanoparticles was mixed with LDPE in a laboratory two-roll mill at a temperature of 140 ºC for 15 min. Samples for water immersion tests and breakdown measurements were then prepared using a hydraulic laboratory press at a temperature of 160 ºC and a load of 3 ton. The thickness of the prepared samples was 100 µm.

2.2 Characterization

The prepared samples were immersed in distilled water and, at different time periods, were removed and dried with tissues, followed by sample weighing and DC breakdown testing. The sample weights (typically 0.5 g) were monitored using a 4-digit analytical balance (OHAUS PA214C); repeated measurements indicate an uncertainty in all measurements of about ±0.04%. The percentage increase in mass for each sample was calculated using the following equation:

$$\% \, Mass = \left| \frac{M_w - M_a}{M_a} \right| \times 100\% \tag{1}$$

where M_w is the weight of a water absorbed sample while M_a is the weight of the sample prior to water immersion.

DC breakdown testing was conducted by placing a test sample between two opposing 6.35 mm diameter steel ball-bearing electrodes immersed in Hyrax Hypertrans transformer oil to prevent surface flashover. A DC voltage with a step voltage of 2 kV every 20 s was applied until the sample experienced breakdown. Fifteen breakdown tests were performed on each type of material. The voltage obtained from each measurement was divided by the sample thickness at the breakdown point in order to obtain the breakdown field. The resulting dielectric breakdown data were statistically analyzed assuming two-parameter Weibull statistics [45, 46].

3.0 Results

3.1 Water uptake characteristics

3.1.1 Changes in absorbed water over water immersion period

Figure 2 shows the water uptake characteristics of LDPE with 20SiO$_2$, 80SiO$_2$ and Si$_3$N$_4$. For referencing purposes, unfilled LDPE showed negligible water absorption effects. However, adding 20SiO_2 to LDPE resulted in an increase in the material's mass upon water immersion. After three days of water immersion, the mass of LDPE with 3 wt% of 20SiO_2 increased as much as 0.93%. The mass of the sample increased to 1.73% after immersing in water for 15 days. By adding 10 wt% of 20SiO_2 to LDPE, the water absorption effects became more apparent than the 3 wt% equivalent counterparts – the water uptake percentage increased to 1.61% and 2.01% after three and fifteen days of water immersion, respectively. The addition of 80SiO_2 to LDPE also resulted in an increase in the material's mass upon water immersion. Nevertheless, the water uptake characteristics of LDPE with 80SiO_2 and LDPE with 20SiO_2 were similar.

Although the addition of Si_3N_4 to LDPE resulted in an increase in the material's mass upon water immersion, the increase is much less compared to LDPE with 20SiO_2 or 80SiO_2. After three days of water immersion, the mass increase for LDPE with 1 wt% and 3 wt% of Si_3N_4 was negligibly small (~0.08%). Generally, the water absorption effects of LDPE with Si_3N_4 became more apparent with an increasing amount of Si_3N_4. After 20 days of water immersion, LDPE with 5 wt% of Si_3N_4 showed an increase of 0.68% water absorption, much less than the water absorption effects of LDPE with 20SiO_2 and 80SiO_2.

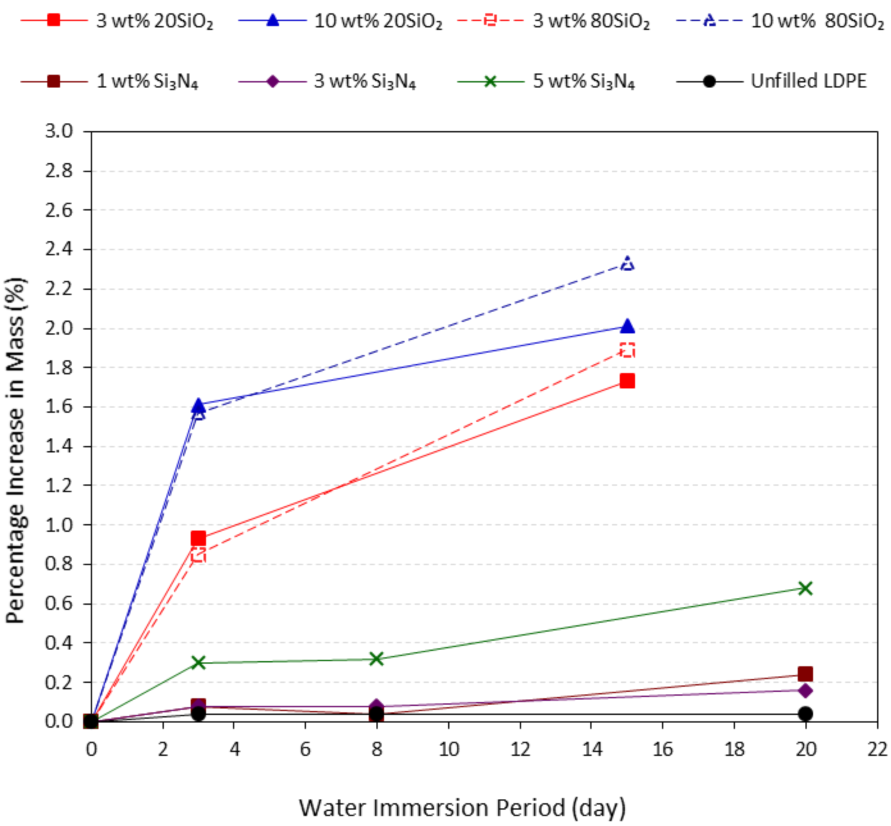

Figure 2. Percentage increase in mass for LDPE with $20SiO_2$, $80SiO_2$ and Si_3N_4, with unfilled LDPE as a reference

3.1.2 Changes in absorbed water over nanoparticle loading levels

Figure 3 shows the plot of the mass increase (or water absorption) for LDPE with $20SiO_2$, $80SiO_2$ andSi_3N_4 over different nanoparticle loading levels after immersing in water for different time periods. Apparently, the water uptake capability of LDPE with Si_3N_4 was different from that of LDPE with $20SiO_2$ and $80SiO_2$. The mass of LDPE with Si_3N_4 did not increase much for the amount of nanoparticles up to 3 wt%. Meanwhile, the mass of LDPE with $20SiO_2$ and $80SiO_2$ increased up to 1% for nanoparticle loading level of 3 wt%. Nevertheless, the mass increase became less steep between 3 wt% and 10 wt% of nanoparticle loading level. Similar trends of water absorption over nanoparticle loading levels were observed after the samples were immersed in water for fifteen or twenty days.

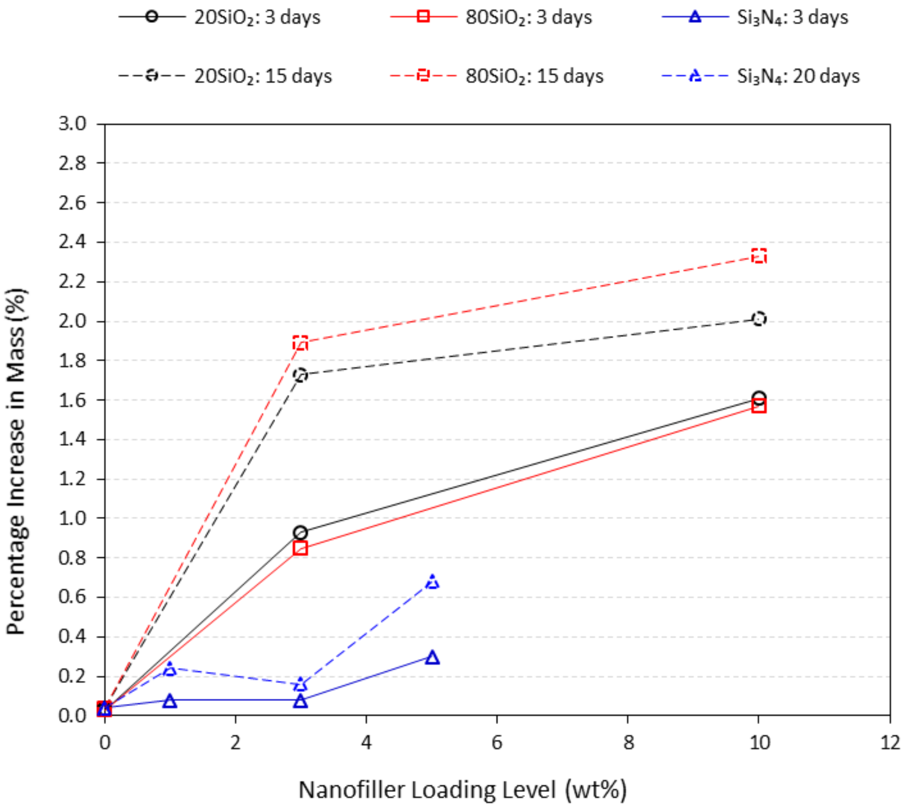

Figure 3. Percentage increase in mass for LDPE with 20SiO_2, 80SiO_2 and Si_3N_4 over different nanoparticle loading levels for different water immersion periods

3.2 Breakdown characteristics

3.2.1 Breakdown strength of unfilled LDPE

Figure 4a shows the breakdown strength of unfilled LDPE upon water immersion for 0, 3 and 15 days (the Weibull parameters are shown in Table 1). Considering the uncertainties in Weibull analysis, the breakdown strength of unfilled LDPE was not affected upon water immersion. The unaltered breakdown strength of unfilled LDPE was further confirmed by another batch of similar LDPE sample immersed in water for 0, 3, 8 and 20 days (see Figure 4b).

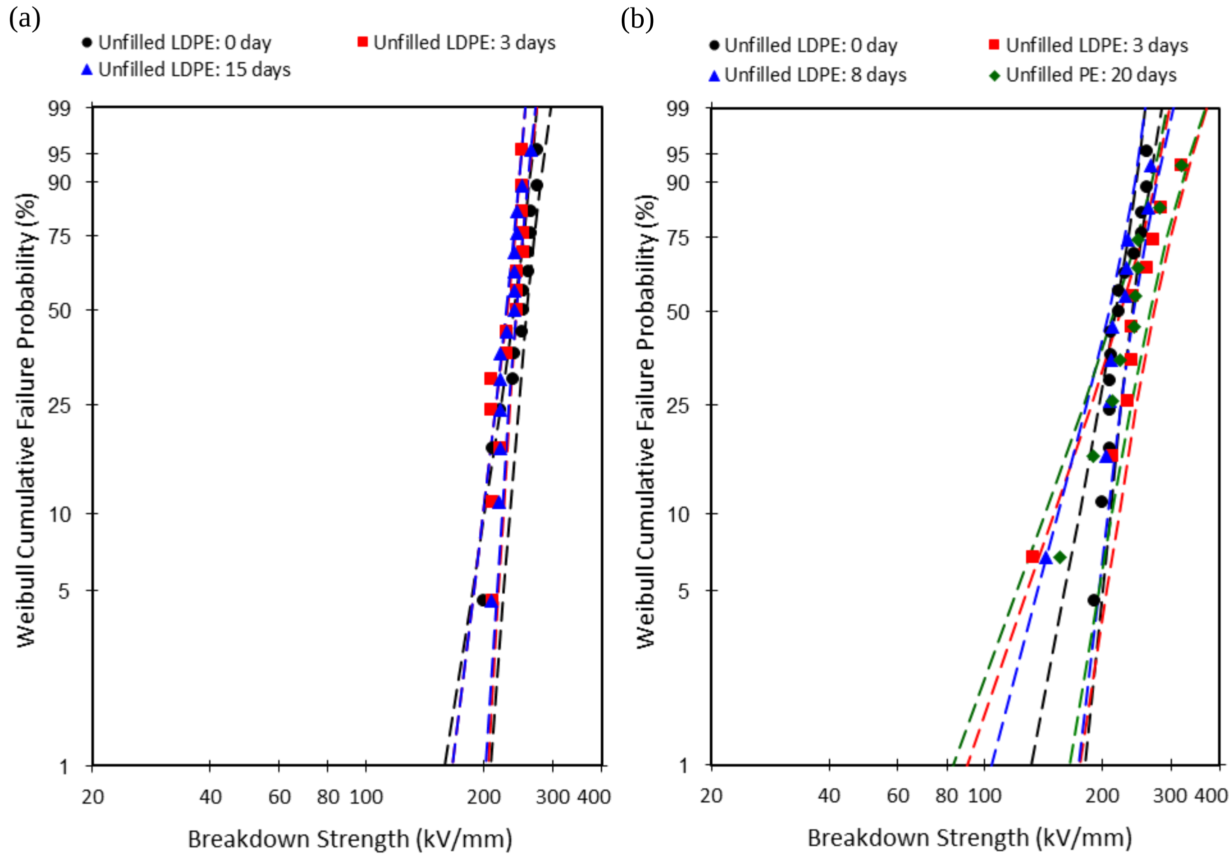

Figure 4. Breakdown strength of (a) unfilled LDPE samples upon water immersion for 0, 3 and 5 days, (b) another batch of similar LDPE samples upon water immersion for 0, 3, 8 and 20 days

Table 1. Weibull parameters for unfilled LDPE upon different water immersion periods

Sample	α (kV/mm)	β
Unfilled LDPE: 0 day	254 ± 9	14 ± 5
Unfilled LDPE: 0 day	235 ± 10	11 ± 4
Unfilled LDPE: 3 days	241 ± 6	18 ± 7
Unfilled LDPE: 3 days	261 ± 23	6 ± 3
Unfilled LDPE: 8 days	234 ± 16	8 ± 5
Unfilled LDPE: 15 days	240 ± 6	17 ± 6
Unfilled LDPE: 20 days	254 ± 25	6 ± 3

3.2.2 Breakdown strength of LDPE with $20SiO_2$

Figure 5a shows the breakdown strength of LDPE with 3 wt% of $20SiO_2$ (the breakdown strength of unfilled LDPE from Figure 4a are re-shown as grey colored background). The breakdown strength of LDPE with 3 wt% of 20SiO$_2$ was not as high as that of unfilled LDPE. In addition, the breakdown strength of LDPE with 3 wt% of 20SiO$_2$ reduced with increasing water immersion periods. A similar breakdown trend was also observed for LDPE with 10 wt% of 20SiO$_2$ (see Figure 5b; the breakdown strength of unfilled LDPE from Figure 4a and LDPE with 3 wt% of 20SiO$_2$ from Figure 5a are re-shown as grey colored background). Furthermore, the reduction of breakdown strength was more apparent for LDPE with 10 wt% of 20SiO$_2$ compared to LDPE with 3 wt% of 20SiO$_2$. The Weibull parameters for LDPE with 20SiO$_2$ are shown in Table 2.

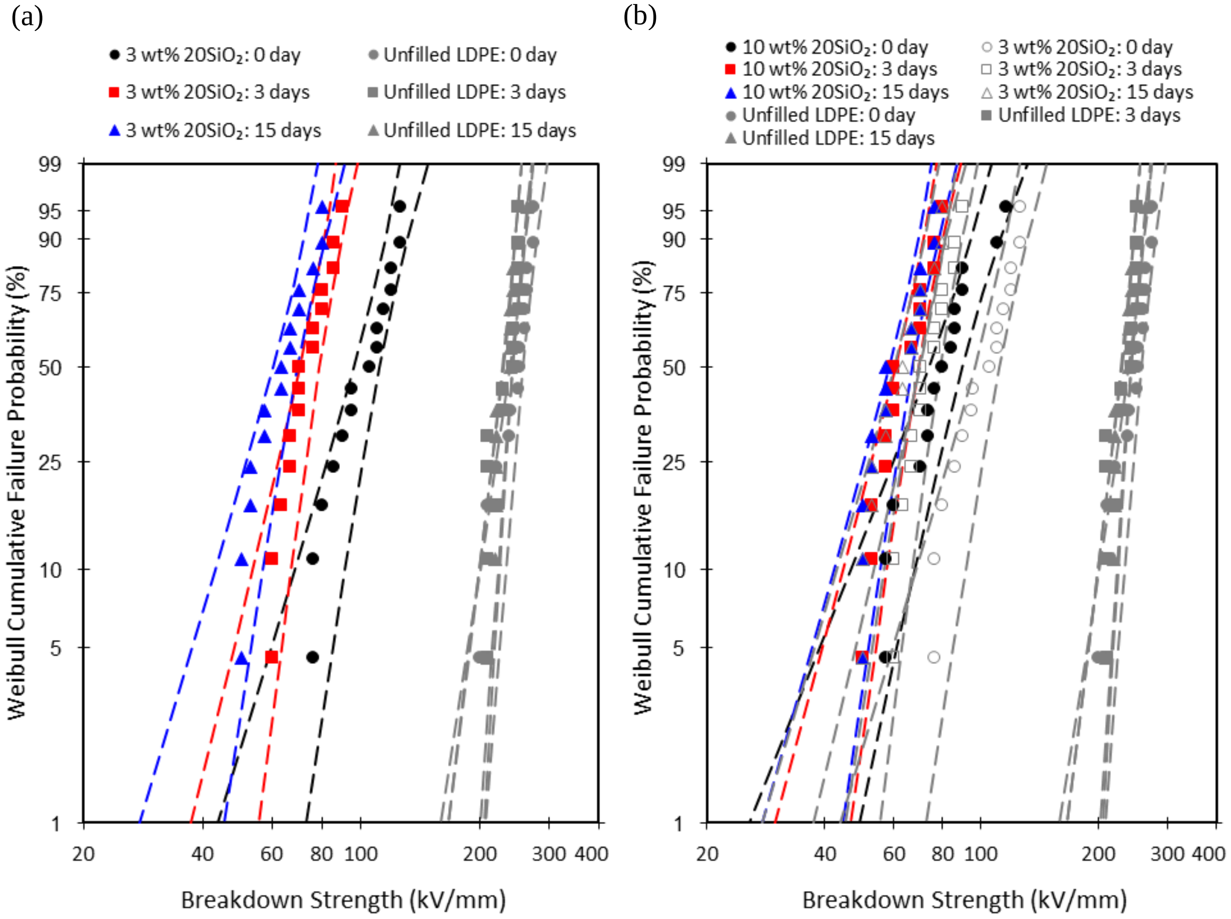

Figure 5. Breakdown strength of LDPE with (a) 3 wt% of 20SiO$_2$ upon water immersion for 0, 3 and 15 days (unfilled LDPE from Figure 4a are re-shown as grey colored background), (b) 10 wt% of 20SiO$_2$ upon water immersion for 0, 3 and 15 days (unfilled LDPE from Figure 4a and LDPE with 3 wt% of 20 nm SiO$_2$ from Figure 5a are re-shown as grey colored background)

Table 2. Weibull parameters for LDPE with 3 wt% and 10 wt% of 20SiO$_2$ upon water immersion for 0, 3 and 15 days

Sample	α (kV/mm)	β
3 wt% 20SiO$_2$: 0 day	109 ± 8	7 ± 3
3 wt% 20SiO$_2$: 3 days	77 ± 5	9 ± 3
3 wt% 20SiO$_2$: 15 days	68 ± 5	7 ± 3
10 wt% 20SiO$_2$: 0 day	87 ± 9	5 ± 2
10 wt% 20SiO$_2$: 3 days	68 ± 4	8 ± 3
10 wt% 20SiO$_2$: 15 days	66 ± 4	7 ± 3

3.2.3 Breakdown strength of LDPE with 80SiO$_2$

Figure 6a shows the breakdown strength of LDPE with 3 wt% of 80SiO$_2$. Again, the breakdown strength of the samples reduced with increasing water immersion periods. A similar trend was also observed for LDPE with 10 wt% of 80 SiO$_2$ (see Figure 6b). Again, the reduction of breakdown strength was more apparent for LDPE with 10 wt% of 80SiO$_2$ compared to LDPE with 3 wt% of 80SiO$_2$. The Weibull parameters for LDPE with 80SiO$_2$ are shown in Table 3.

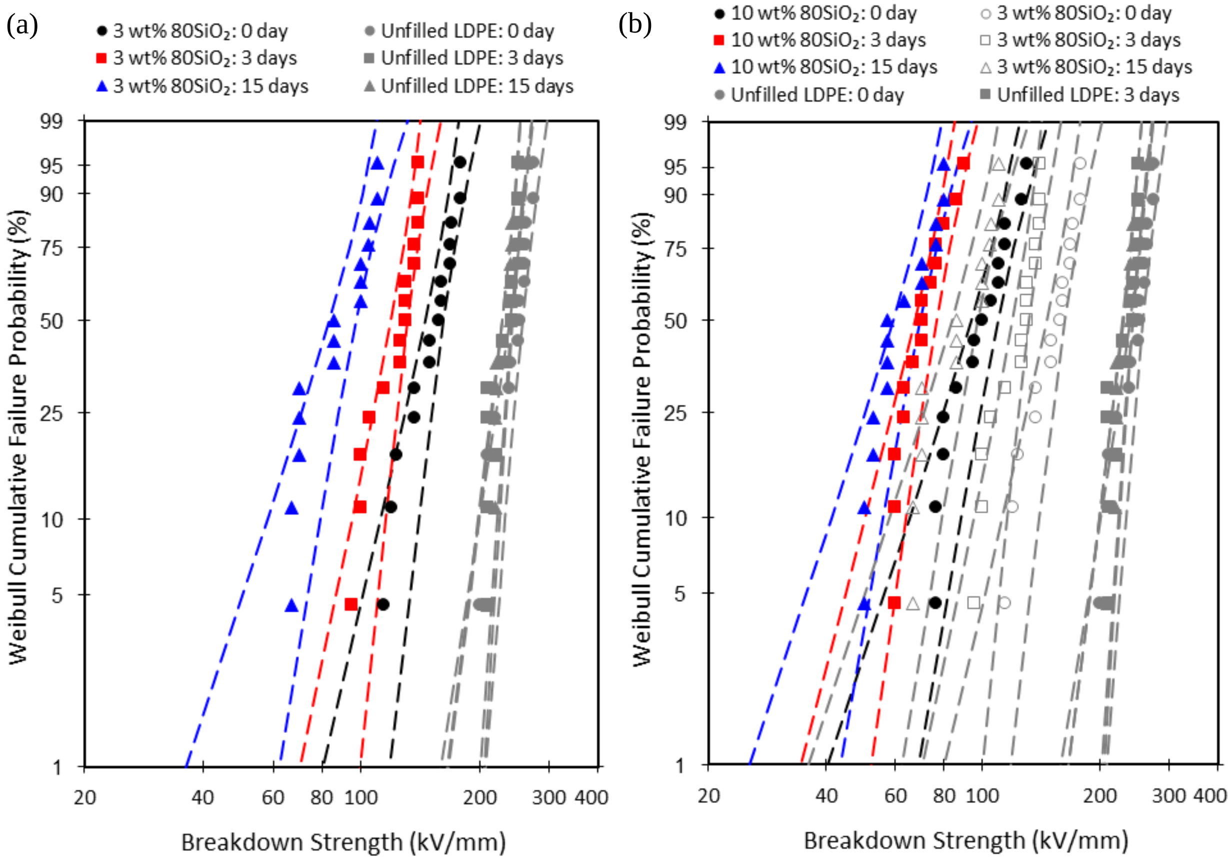

Figure 6. Breakdown strength of LDPE with (a) 3 wt% of 80SiO$_2$ upon water immersion for 0, 3 and 15 days (unfilled LDPE from Figure 4a are re-shown as grey colored background), (b) 10 wt% of 80SiO$_2$ upon water immersion for 0, 3 and 15 days (unfilled LDPE from Figure 4a and LDPE with 3 wt% of 80 nm SiO$_2$ from Figure 6a are re-shown as grey colored background)

Table 3. Weibull parameters for LDPE with 3 wt% and 10 wt% of 80SiO$_2$ upon water immersion for 0, 3 and 15 days

Sample	α (kV/mm)	β
3 wt% 80 nm SiO$_2$: 0 day	160 ± 8	9 ± 4
3 wt% 80 nm SiO$_2$: 3 days	130 ± 6	11 ± 4
3 wt% 80 nm SiO$_2$: 15 days	95 ± 7	7 ± 2
10 wt% 80 nm SiO$_2$: 0 day	107 ± 8	6 ± 3
10 wt% 80 nm SiO$_2$: 3 days	75 ± 4	8 ± 3
10 wt% 80 nm SiO$_2$: 15 days	68 ± 3	6 ± 3

3.2.4 Breakdown strength of LDPE with Si$_3$N$_4$

Figure 7a shows the breakdown characteristics of LDPE with 1 wt% of Si$_3$N$_4$. Before subjected to water immersion, the breakdown strength of LDPE with 1 wt% of Si$_3$N$_4$ was 244 kV/mm, similar to the breakdown strength of unfilled LDPE. After immersing in water for 3 days, the breakdown strength of LDPE with 1 wt% of Si_3N_4 reduced drastically to 130 kV/mm. Further reductions of the breakdown strength were observed after LDPE with 1 wt% of Si_3N_4 was immersed in water for 8 and 20 days.

Figure 7b shows that the breakdown trend for LDPE with 3 wt% of Si_3N_4 is similar to that of LDPE with 1 wt% of Si_3N_4. However, the addition of 5 wt% of Si_3N_4 to LDPE significantly reduced the breakdown strength of the material to 180 kV/mm (see Figure 7c). Again, the breakdown strength of the samples reduced with increasing water immersion periods. The Weibull parameters for LDPE with Si_3N_4 are shown in Table 4.

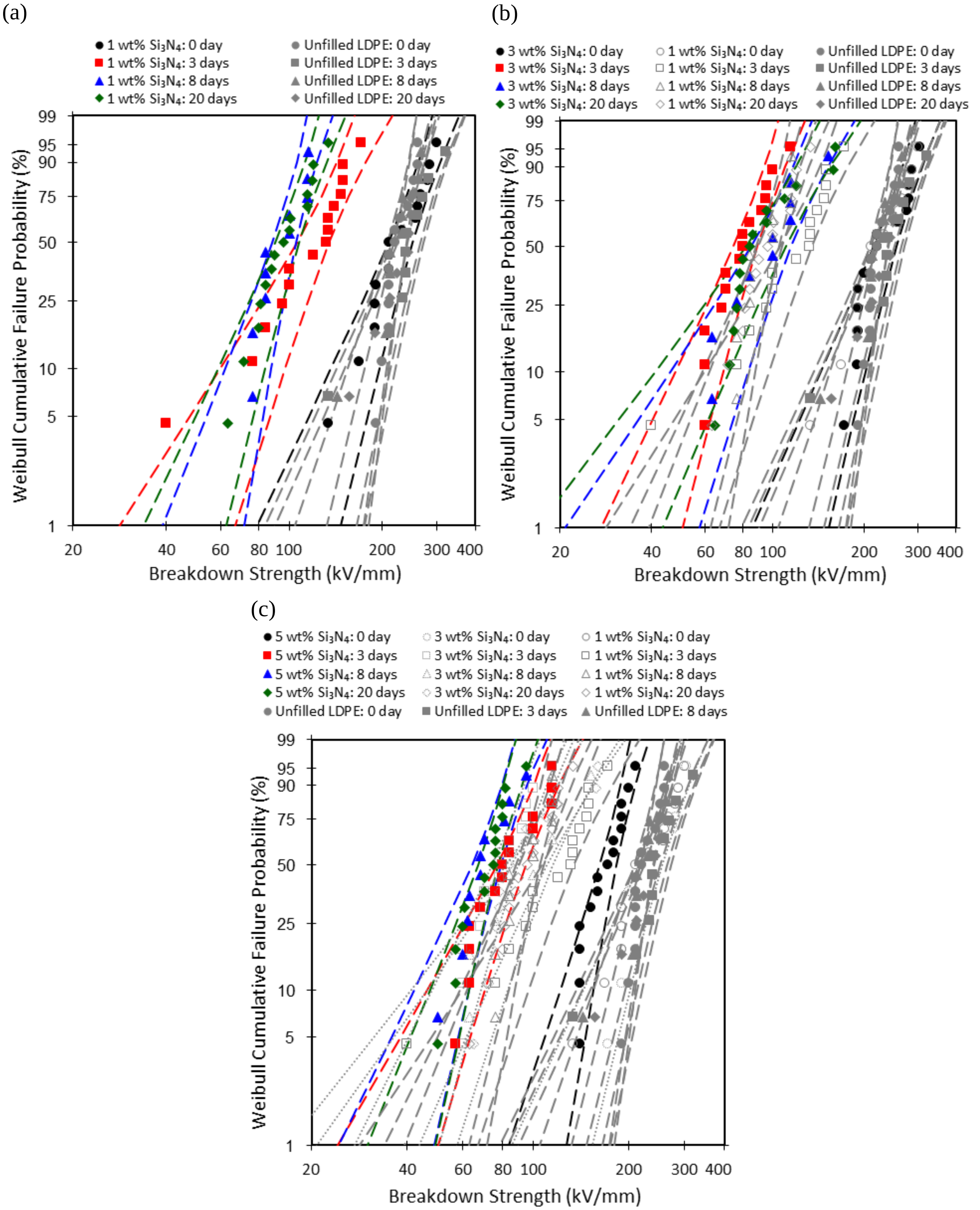

Figure 7. Breakdown strength of LDPE with (a) 1 wt% of Si_3N_4 upon water immersion for 0, 3, 8 and 20 days (unfilled LDPE from Figure 4a are re-shown as grey colored background), (b) 3 wt% of Si_3N_4 upon water immersion for 0, 3, 8 and 20 days (unfilled LDPE from Figure 4a and LDPE with 3 wt% of Si_3N_4 from Figure 7a are re-shown as grey colored background), (c) 5 wt% of Si_3N_4 upon water immersion for 0, 3, 8 and 20 days (unfilled LDPE from Figure 4a, LDPE with 1 wt% of Si_3N_4 from Figure 7a and LDPE with 3 wt% of Si_3N_4 from Figure 7b are re-shown as grey colored background)

Table 4. Weibull parameters for LDPE with 1 wt%, 3 wt% and 5 wt% of Si_3N_4 upon water immersion for 0, 3, 8 and 20 days

Sample	**α (kV/mm)**	**β**
1 wt% Si_3N_4: 0 day	244 ± 20	6 ± 2
1 wt% Si_3N_4: 3 days	130 ± 15	4 ± 2
1 wt% Si_3N_4: 8 days	101 ± 9	7 ± 4
1 wt% Si_3N_4: 20 days	105 ± 9	6 ± 2
3 wt% Si_3N_4: 0 day	249 ± 20	6 ± 2
3 wt% Si_3N_4: 3 days	87 ± 8	5 ± 2
3 wt% Si_3N_4: 8 days	108 ± 15	4 ± 4
3 wt% Si_3N_4: 20 days	106 ± 15	3 ± 2
5 wt% Si_3N_4: 0 day	180 ± 10	8 ± 4
5 wt% Si_3N_4: 3 days	92 ± 9	5 ± 2
5 wt% Si_3N_4: 8 days	76 ± 7	6 ± 3
5 wt% Si_3N_4: 20 days	76 ± 5	7 ± 2

4.0 Discussion

The behavior at the interphase of nanocomposites depends significantly on the interfacial layer most adjacent to nanoparticles, which in turn is closely related to the surface chemistry of the nanoparticles [13, 14, 22-25]. In the current work, two types of silicon-based nanoparticles with different chemical compounds, i.e., oxide (SiO_2) and nitride (Si_3N_4), were investigated to understand the key phenomena associated with changes in nanoparticle surface chemistries. In Figure 1a, it was illustrated that SiO_2 nanoparticles typically contains Si-O-Si bonds. Due to their electronegativity, the oxygen (O) atoms on the SiO_2 surface can form stable chemical bonds with almost all elements to give the corresponding oxides. Naturally, the O atoms have a high tendency to form hydrogen bonding with water molecules. Consequently, SiO_2 is prone to water adsorption, where surface hydroxyl (OH) groups tend to be present on the surface of SiO_2 when exposed to air or water surroundings. Indeed, available literature [32, 61] showed that SiO_2 adsorbed significant amount of water even when stored under ambient conditions, suggesting that water can readily be partitioned to polar SiO_2 surfaces.

Meanwhile, Si_3N_4 nanoparticles typically contains Si-N-Si bonds, as illustrated in Figure 1b. According to the Pauline scale [47], the electronegativity of an N atom is 3.04, less than that of an O atom (3.44). Therefore, the affinity of the N atoms on the surface of Si_3N_4 to attract water molecules is lower, albeit that Si_3N_4 exposed to air can form an oxide film on its surface, where the oxidation film can thus react with water [42, 48]. This was also reported in previous findings [32, 61] where, under ambient conditions, Si_3N_4 adsorbed little water compared to SiO2. Therefore, the water adsorption effects were much less pronounced for Si_3N_4 compared to SiO_2. The results from the current water immersion test were, indeed, in line with the above illustrated surface chemistry changes. LDPE is hydrophobic in nature, so it repels rather than absorbs water [49]. Consequently, no appreciable changes in mass were observed for unfilled LDPE. Since unfilled LDPE was not affected by its surrounding water, the breakdown strength of unfilled LDPE remained similar upon water immersion.

Meanwhile, most nanoparticles are hydrophilic in nature. The addition of hydrophilic nanoparticles to hydrophobic LDPE would render the resulting nanocomposite material to become hygroscopic – having tendency to absorb or adsorb water from surroundings. Based on the current experimental work on LDPE with $20SiO_2$ or $80SiO_2$, an increase in the materials' mass was recorded upon water immersion. The increase in mass could be attributed to the presence of water within LDPE/SiO_2 interphase, since OH groups tend to be formed on the surface of SiO_2. With increasing water immersion periods, more water molecules could reside within the interphase, and this negatively affected the breakdown strength.

The presence of water within nanocomposites are not unusual and was anticipated by Zou et al. [27] and Fabiani et al. [29] through their proposed water shell models, where layers of water were expected to surround nanoparticles (at the interphase). Rowe [50] suggested the "interphase aging scenario", whereby water that diffused into composites would reach the particle/polymer interphase where it accumulated, thus forming a network of semi inter-connected water pathway at the interphase. Percolation of water through the interphase would then occur, and an electrical path would appear through the sample, initiating conduction current and a runaway mechanism that would lead to electrical breakdown. Indeed, Hosier et al. [51, 52] reported increased electrical conductivity, dielectric loss and permittivity, thus reduced breakdown strength, as a consequence of increased water absorption in polyethylene/silica nanocomposites. A progressive reduction in breakdown strength with increasing moisture content was also observed by many other researchers [53-55], in agreement with the current findings.

The use of nanometer-size fillers commonly leads to the presence of more extensive interfacial areas [12], hence higher amounts of absorbed water. While using SiO_2 with a larger manufacturer-quoted diameter, i.e., $80SiO_2$, was expected to reduce the water absorption effect due to its smaller overall interfacial areas compared to $20SiO_2$, this was not apparent in the current work. This imply two possibilities. First, the water absorption characteristics were similar in LDPE with $20SiO_2$ and $80SiO_2$, indicating that they absorbed water in a similar way. Second, the water absorption characteristics were different in LDPE with $20SiO_2$ and $80SiO_2$, but these characteristic change was difficult to be measured directly from the current experiment. The latter is thought to be more relevant, since the breakdown strength of LDPE with $80SiO_2$ was higher than that of LDPE with $20SiO_2$ counterparts, thus indicating less water molecules within the LDPE with $80SiO_2$.

Significantly, the addition of Si_3N_4 to LDPE resulted in much less water absorption when compared to the addition of $20SiO_2$ or $80SiO_2$ to LDPE. In this regard, different surface chemistries governing of Si_3N_4 and SiO_2 is important. Since the affinity of N atoms on the surface of Si_3N_4 to attract water molecules is lower than O atoms on the surface of SiO_2, water is less attractive to the surface of Si_3N_4. Nevertheless, the presence of the interphase region in LDPE with Si_3N_4 could still serve as a preferable site for the aggregation of water molecules. Since this region would nevertheless contain water upon water immersion, the breakdown strength of LDPE with Si_3N_4 would be negatively affected. It is noteworthy that the breakdown strength of LDPE with Si_3N_4 significantly decreased although very few water was absorbed. This could be a consequence of the formation of a percolating water network that enhanced electrical conductivity in well dispersed Si_3N_4-based nanocomposites; in SiO_2-based nanocomposites, a percolating network was not formed due to particle aggregation [32]. Consequently, small changes in water content could manifest themselves as major changes in electrical performance.

The breakdown strength of LDPE reduced with increasing amounts of SiO_2 or Si_3N_4 loading levels. This has often been associated with nanoparticles aggregation [56, 57]. Nevertheless, the current breakdown results indicated that the same sample, when immersed in water, could result in much lower breakdown strength. Therefore, although the aggregation of nanoparticles could negatively affect the breakdown strength of nanocomposites, the presence of water could also be detrimental to the breakdown strength of the materials. Furthermore, an apparent increase in mass could be noticed for LDPE with 10 wt% of SiO_2 than 3 wt% of SiO_2. This could be related to increased interphase volumes (and therefore increased water contents) as a consequence of increased nanoparticles amounts. The breakdown strength of LDPE with higher SiO_2 loading levels was consequently further reduced.

In the previous work [58, 59], changes in the permittivity values of nanoparticles were demonstrated to affect the electric field distribution within nanocomposites, thus the breakdown performance. Generally, nanoparticles with permittivity values higher than polymers showed distorted electric field intensity within the resulting nanocomposites, and the distortion effect amplifies if the permittivity of nanoparticles further increases. So, the breakdown strength of a material is commonly reported to be inversely proportional to the permittivity of the material [60]. In the current work, however, the permittivity of Si_3N_4 (7.5) was higher than SiO_2 (3.9), but adding

Si_3N_4 to LDPE resulted in higher breakdown strength than adding SiO_2 to LDPE even before the water immersion test. The current results are, nevertheless, consistent with the work of Hosier et al. [61] in comparing the breakdown strength of polyethylene containing Si_3N_4 and SiO_2. Hosier et al. [32, 61] suggested that as-received Si_3N_4 nanoparticles were generally drier than as-received SiO_2 nanoparticles under ambient conditions, so the overall permittivity of polyethylene/Si_3N_4 nanocomposites was lower than polyethylene/SiO_2 nanocomposites. Consequently, the effects of nanofillers' permittivity on the breakdown strength of nanocomposites were overshadowed by the effects of water surrounding the nanofillers. It is therefore important to note that, while addressing permittivity mismatches between the nanoparticles and the polymer is crucial in nanocomposites, efforts to eliminate water on nanoparticles' surface or within the nanocomposite interphase are of equal importance. This is because water could readily be adsorbed onto the surface of nanoparticles, creating a volume of water within the interphase, thus jeopardizing the dielectric properties of nanocomposites.

5.0 Conclusions

The current work demonstrated that the addition of oxide- and nitride-based silicon nanoparticles to LDPE could alter the material's water absorption behavior. While LDPE itself is hydrophobic, LDPE when added with SiO_2 nanoparticles, became prone to water absorption. In contrast, LDPE with Si_3N_4 nanoparticles attracted much less water. These water absorption behaviors consequently affected the breakdown properties of the materials, where LDPE with SiO_2 nanoparticles showed lower breakdown strength than LDPE with Si_3N_4 nanoparticles. The observed changes in the water absorption and breakdown characteristics of the nanocomposites were attributed to the interfacial mechanisms of the nanocomposites, in particular, in relation to surface chemistries of SiO_2 and Si_3N_4. Significantly, the results from the breakdown tests showed that understanding nanoparticles' surface chemistry and hence the nanocomposite interphase, in relation to its water absorption (or adsorption) behavior is particularly important, since the presence of water in any dielectrics is strongly undesirable.

Acknowledgments

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