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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.

1. 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 resistances, treeing propagations, space charge build-ups and dielectric breakdown performances [1–10]. Due to the effect of nanostructuring, 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

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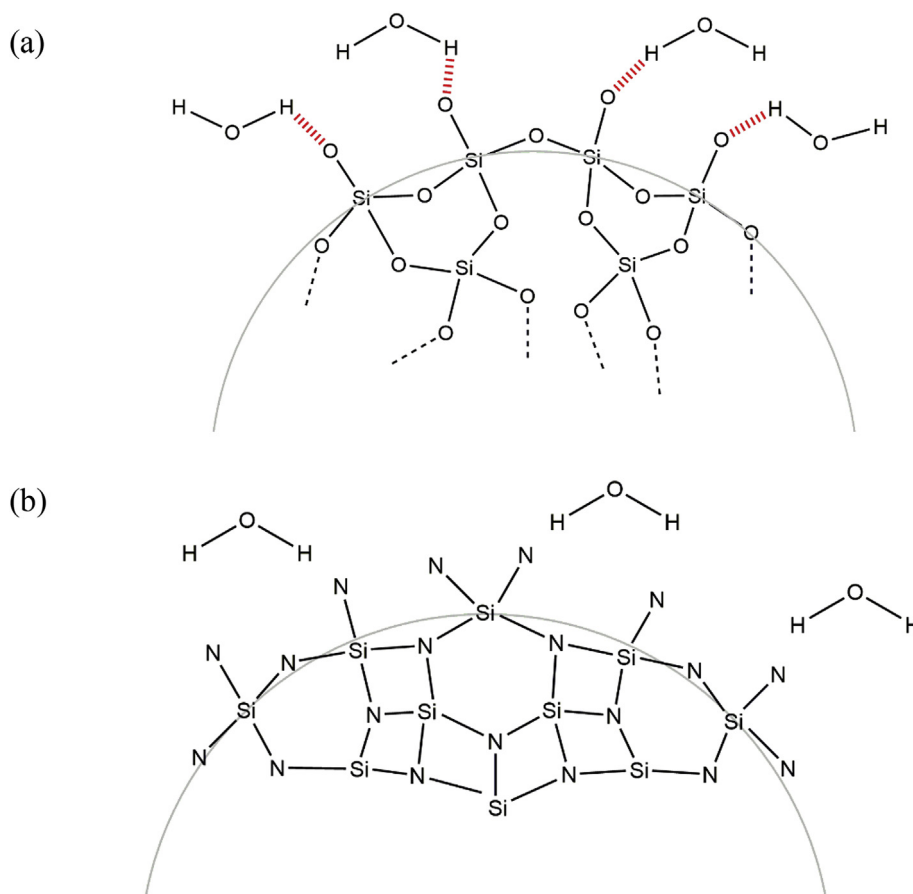


Fig. 1. Surface chemistries of (a) SiO₂, (b) Si₃N₄.

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 performances 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 (SiO₂) and silicon nitride (Si₃N₄), were added respectively to LDPE to explore the interfacial mechanisms with regard to water absorption and breakdown characteristics of the resulting nanocomposites.

2. 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 (SiO₂) nanopowder and silicon nitride (Si₃N₄) nanopowder, obtained from NanoAmor. The Si₃N₄ nanoparticles had a manufacturer-quoted average particle size of 15–30 nm. For the SiO₂ nanoparticles, two manufacturer-quoted average particle sizes were considered, i.e., 20 nm and 80 nm – these SiO₂ nanoparticles were referred to as 20SiO₂ and 80SiO₂, respectively, hereafter. Fig. 1 illustrates that SiO₂ nanoparticles typically contains Si-O-Si bonds [38–41] while Si₃N₄ nanoparticles typically contains Si-N-Si bonds [42–44]; this will be further discussed later.

Nanocomposite samples were prepared using a mechanical mixing method. The desired amount of 20SiO₂, 80SiO₂ or Si₃N₄ nanoparticles was mixed with LDPE in a laboratory two-roll mill at a temperature of 140 °C for 15 min. Samples for water immersion tests and breakdown measurements were then prepared using a hydraulic laboratory press at a temperature of 160 °C and a load of 3 ton. The thickness of the prepared samples was 100 μm.

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