

## PREFACE

Electrical insulation constitutes the backbone for all electrical equipment. At the same time, they are considered as the weakest link. Electrical insulation's capacity to endure degradation under various operating stresses (e.g., electrical, mechanical, thermal, and environmental) is a key factor in determining the reliability of electrical components and systems. Polymeric materials are widely used insulating materials in electrical generators, transformers, bushings, line insulators, circuit breakers, underground cables, GIS, surge arresters, and electrical machines. These materials enjoy widespread acceptance among electrical insulation design engineers due to many desirable attributes (e.g., good dielectric properties, high strength to weight ratio, and ease of molding). However, charge accumulation at the high dc field, poor discharge resistance, low thermal conductivity, limited-service temperature range, and inadequate stiffness have proven to be severe obstructions to the far-reaching utilization of these materials. Often, inorganic oxide fillers are added to the host polymer in order to increase its thermo-mechanical properties. Unfortunately, the synthesis of polymer composites using conventional micro-sized fillers demands a large filler loading; as a result, better thermo-mechanical properties are obtained at the expense of deteriorated electrical properties. An idea that originated under the name of polymer nanocomposites (PNCs) is supposed to offer a potential solution to the aforementioned problems. The combination of nanoscale reinforcement and a polymer matrix has the potential to produce exceptional material characteristics. Recent literature is a testimonial of the enormous research on synthesis and characterization of PNCs for various engineering applications. Polymer nanocomposites offer unprecedented material properties, which are widely attributed to a high proportion of interphase generated between the matrix and filler material. Thus, interphase characterization is critical for tailoring these novel

materials to a broad range of engineering and scientific applications. The literature striving to make interphase perceivable is indeed exiguous. In the recent past, few theoretical models have been proposed to corroborate interphase and its affiliation material properties. However, current research falls far short of providing a mechanistic description of the interphase formation and its experimental manifestation. Furthermore, a quantitative analysis of interphase, as well as qualitative analysis, is critical for obtaining tailored material properties for a variety of applications and making PNCs commercially viable. This work unravels and sheds light on the interphase formation and its affiliation with electro-thermal properties in epoxy alumina nanocomposites. Furthermore, a comprehensive quantitative analysis of interphase provides an intellectual foundation for evaluating nanodielectrics for future applications in the electrical power industry. Epoxy resin (LY556 Bisphenol-A, density 1.17 g/cm<sup>3</sup> at 25°C) supplied from Huntsman was used as a host material. Utilities have extensive experience using traditional micro-sized aluminum oxide (Al<sub>2</sub>O<sub>3</sub>) fillers to improve mechanical strength in composites. Thus, nanometer-sized aluminum oxide/or alumina particles are used as filler material in the present study.

One of the most difficult aspects of nanocomposites synthesis is achieving homogeneous nanofiller dispersion in the polymer matrix. Nanoparticles tend to agglomerate due to their high specific surface area and hydrophilic nature. Additionally, inorganic oxide fillers are incompatible with organic polymers due to their chemical composition. Thus, the first part of the study establishes an experimental protocol for the fabrication of nanocomposites with well-dispersed nanofillers. Additionally, inorganic fillers have their surfaces functionalized with a suitable surfactant to improve their dispersion and bonding to the host polymer. FTIR spectroscopy is used to verify the surface functionalization of nanofillers, and FESEM is used to qualitatively examine

filler dispersion in the polymer matrix. Furthermore, a numerical method for quantifying nanofiller dispersion in the polymer matrix is developed.

The second part of the thesis explores the formation of interphases in epoxy alumina nanocomposites. To investigate interphase and its relationship to dielectric properties, three types of samples are examined: neat epoxy, nanocomposites (incorporated with surface-treated nanofillers), and nanocomposites (formed with untreated nanofillers). Synthesized samples are subjected to dielectric properties measurements i.e., measurement of ac dielectric strength, measurements of complex permittivity, and measurement of dc conductivity. Further, the thermal characteristics of nanocomposites are analyzed by thermogravimetric analysis (TGA). Characteristics information received from thermogravimetric analysis and dielectric properties measurements insinuates a change in chemistry and molecular mobility at the filler matrix interfaces. Using the chemical structure of different constituent phases and FTIR spectroscopic analysis, a chemical interactive model is presented to elicit interphase formation in composites. The impact of interfacial interaction on the long-term performance of the nanodielectrics is examined by conducting endurance tests under divergent ac stress. Nanocomposites exhibit a clear superior erosion resistance over the neat polymer. Under the application of cyclic non-uniform ac stress, progressive erosion is likely to initiate and grow from high-stress region. Nanofillers may act as an obstacle and force the eroded channels to move through a zig-zag path. Additionally, surface-treated nanofillers owing to their strong chemical bonding with polymer matrix expected to retard the damage process by alleviating fatigue and distributing electromechanical stress between filler and polymer matrix.

The interphase is anticipated to have material properties distinct from those of the matrix and filler materials. To obtain tailored material properties for a variety of

applications, quantitative interphase analysis in conjunction with qualitative analysis is critical. As a result, the third part of the thesis describes a methodology for quantifying interphase's permittivity and thickness. Epoxy-based nanocomposites samples (synthesized using surface-treated nanoalumina) are subjected to dielectric spectroscopic measurements over a frequency range of  $10^{-2}$  to  $10^7$  Hz. A finite element-based numerical model is developed to estimate the effective permittivity of composites for different values of assumed interphase parameters (i.e. thickness and permittivity). A bisection method-based algorithm is devised to assign actual thickness and permittivity to interphase based on the best fit of experimental and simulated results. It is observed that interphase in epoxy alumina nanocomposites may extend up to 100 nm, and the relative permittivity of interphase is slightly lower than the effective permittivity of composites.

Low electrical conductivity combined with high thermal conductivity has become an increasingly desirable characteristic for polymeric insulating materials operating at elevated voltage levels. Conduction loss is the primary source of heat generation within dielectric materials in a high voltage direct current (HVDC) system, and conduction-based heat transfer is governed by the dielectric material's thermal conductivity. In recent years, polymer nanocomposites (PNCs) have gained prominence as promising candidates for addressing these two conflicting requirements. As evidenced by a growing body of literature, interphase appears to be critical in enhancing the properties of PNCs. On the other hand, the thermoelectric properties of interphase are unknown and are assumed to be significantly different from those of the filler and base polymer. Because electrical conduction losses generate heat, thermal analysis under electrical stress requires knowledge of dc conductivity. Thus, the fourth section of the thesis examines the interphase's electrical and thermal conductivity in epoxy

alumina nanocomposites. To begin, we determine the dc conductivity of epoxy alumina nanocomposite samples using the three-electrode method and a Keithley 6715B electrometer. The effective dc conductivity of nanocomposites is estimated using a numerical model based on finite elements. Following that algorithm described in the previous section is used to determine the electrical conductivity of interphase using experimental results and numerical modeling in conjunction. Subsequently, the thermal conductivity of epoxy alumina nanocomposite samples synthesized in the laboratory is measured using TPS-500. A finite element method (FEM) based numerical model is developed to estimate the effective thermal conductivity of epoxy alumina nanocomposites for a range of assumed interphase thermal conductivity values. By using simulation and experiment results in conjunction, the thermal conductivity of interphase is estimated by using the algorithms described above. A detailed analysis of experimental and numerical modeling data suggests that the thermal conductivity of the interphase in epoxy alumina nanocomposites is significantly higher than that of the host polymer. On the other hand, dc conductivity of the interphase found to be lower than that of neat epoxy. The aligned polymer chain at filler matrix interfaces may be responsible for the high thermal conductivity of the interphase. The alignment of the polymer chain lowers phonon scattering and hence improves heat transmission efficiency. Furthermore, filler matrix interaction at the interface may promote crystallinity, which reduces electrical conductivity. As a result, nanocomposites made with surface-treated nanofillers are successful in increasing thermal conductivity while decreasing electrical conductivity.

Polymers are frequently filled with nanosized inorganic fillers that are electrically insulating but conduct heat more efficiently than the base polymer. It is critical to determine the filler concentrations that improve thermal conductivity without

impairing electrical properties. Additionally, as demonstrated in the preceding chapters, the interphase surrounding nanofillers plays a critical role in the change of properties in polymer nanocomposites. Thus, the fifth part of the work investigates the effect of interphase on optimizing the nanofiller content for improved electro-thermal properties. It has been observed that that thermal property is governed by nanofiller concentrations, whereas dielectric properties are governed by interphase volume fraction. The thermal conductivity increases monotonically as filler concentrations are increased, provided that filler dispersion is adequate. At high filler content, thermal conductivity is expected to increase dramatically due to the formation of a conducting network. As a result, the optimal filler concentrations for enhanced dielectric and thermal properties are those with the highest interphase volume fraction. Beyond these optimal filler concentrations, increased thermal conductivity is possible at the expense of degraded dielectric properties.

In summary, the work constitutes an extensive study of interphase and associated thermo-electrical properties in epoxy-alumina nanocomposites. Furthermore, the thesis presents a comprehensive numerical modeling strategy that will be critical for assessing the electro-thermal behavior of various polymer-based nanocomposites.