Engineered Interfaces in Extruded Polyphenylsulfone-Boron Nitride Composite Insulation

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***Abstract*- Improving matrix-filler interactions is critical for optimizing dielectric performance in composite insulation; however, the technique used to introduce inorganic fillers into an organic matrix varies in its ability to satisfactorily reduce the size of cavities and interfaces. This study reports effects from changing the filler incorporation strategy on the rheology, dielectric performance and thermal conductivity of extruded polyphenylsulfone (PPSU) – hexagonal boron nitride (BN) composite insulation. Depending on the technique used to introduce BN into the host matrix, the viscosity, torque, and melt viscosity temperature of the polymer were reduced, enabling better mixing, heat transfer, and smaller voids. This corresponded to an increase in dielectric strength compared to other formulations at either a similar or lower filler loading. On the other hand, the thermal conductivity in these specimens shifted further away from the thermal conductivity target of 1 W/m·K, which was an indication of a larger separation distance between the particles in samples with smaller interfaces. A trade-off between dielectric strength and thermal conductivity may exist when maximizing thermal conduction without sacrificing dielectric strength.**

I. Introduction

Next-generation power electronics will require thermally conductive electrical insulation to achieve superior dielectric performance with effective thermal management characteristics. Dielectric failure in solid composite insulators is more likely to initiate at the interface, through either delamination or charge buildup at the organic-inorganic boundary layers. Modifying the processing method provides a means to enhance interfacial interactions either between layers in multilayer insulation or between the polymer and filler in particulate-filled polymer matrix composites. In the case of particulate filled composites, it is common practice to introduce fillers to the polymer using high shear mixing methods to disperse and distribute particles; however, this method alone is not always sufficient to improve the interfaces and corresponding properties of composite dielectrics.

Optimal dielectric performance and thermal conduction in composite insulation are influenced by many factors, including mixture viscosity, preferred polymer chain and particle orientation, filler concentration, geometry, and size, as well as affinity between the polymer and filler. For example, higher dielectric breakdown strength of polyimide composites has been demonstrated using BN nanoplatelets as a filler, compared to virgin polyimide [1]. On the other hand, it has also been observed that significant improvements to thermal conductivity is often achieved when larger micronized particles are used either alone or in combination with nanoscale particles [2,3]. On the other hand, to create greater resistance against partial discharge (PD), BN nanoplatelets have demonstrated effectiveness at reducing PD and the degradation area [4]. Polyamide-silica nanocomposites have also demonstrated improvements to PD resistance compared to microcomposites [5]. In addition to filler size, partial discharge inception voltage (PDIV) in composite insulation can also be influenced by the quality of the interfaces [5,6]. Since PD occurrence is often a precursor to more significant breakdown events, more thorough studies into interfacial engineering of composite insulators can be beneficial.

Few papers have offered insight into how altering the insulation processing conditions affects the interfaces, and consequently, a composite insulator's dielectric strength and PDIV. The objective of this work is to investigate how the method used to introduce inorganic fillers into a matrix affects the quality of the interfaces and thus, dielectric performance and thermal conductivity of composite insulation.

Polyphenylsulfone (PPSU) was selected for its high impact strength, chemical and oxidation resistance, and flame retardancy. It is also reported that PPSU has potential to demonstrate resistance to electrical treeing [7]. Boron nitride is known for its in-plane and thru-plane thermal conductivity, as well as its electrically insulating properties. The effects of BN filler concentration, composition, and incorporation method on the dielectric breakdown strength, PD, and thermal conductivity of PPSU are reported.

II. Experimental Methods and Procedures

A. Extrusion

Films were fabricated using twin-screw extrusion. Virgin PPSU pellets were extruded with either micronized or nanoscale BN. Virgin and modified PPSU pellets were melted and compounded with either the micronized BN or a mixture of micronized + nanoBN. Modified PPSU consisted of an additive applied to the surface of the raw PPSU pellets. Samples were identified as PPSU A, PPSU B and PPSU C. PPSU A consisted of unmodified PPSU and a blended filler mixture of nano and micronized BN. PPSU B included unmodified PPSU with micronized BN filler. PPSU C consisted of modified PPSU with micronized BN. The thermal

profile for the eight-zone extruder was established by rheology and the vendor's recommendations.

B. Methods

Rheology was carried out to identify the low melt viscosity temperature and flow behavior changes between the virgin and modified PPSU pellets, which aided in establishing the extrusion parameters. Experiments were performed using a parallel plate rheometer. A temperature ramp was carried out from ambient temperature to 445 °C. Steady state shear experiments were carried out at 5% strain at ~380 °C.

Dielectric breakdown testing was carried out using a dielectric test rig conforming to the ASTM D149 Standard Test Method. Samples were placed in the dielectric test rig and the dielectric breakdown voltage was measured in air. Round, ~6.35 mm diameter electrodes were used for each test. Repetitive pulse PD measurements were carried out at the High Voltage and Power Electronics Laboratory at The Ohio State University. An overview of the test can be found in [8]. The effect of hBN filler loading on the thermal conductivity in PPSU and PPSU composite films was investigated using laser flash analysis. Energy dispersive spectrometry-scanning electron microscopy (EDS-SEM) was used to examine microstructure of the composites. An acceleration voltage of 10 kV was used.

III. Results and Discussion

The rheological behavior of the virgin PPSU and modified PPSU pellets are observed in Fig. 1. Fig. 1 (a) represents a temperature-dependent viscosity profile between ambient temperature to ~445 °C. The ambient temperature viscosity of virgin PPSU pellets began around 6630 Pa-s, whereas the starting viscosity of modified PPSU was ~34,000 Pa-s. As the temperature increased beyond 200 °C, the viscosity of the modified PPSU had a lower viscosity than virgin PPSU, reaching its lowest viscosity of ~48 Pa-s near 305 °C. On the other hand, the viscosity of the virgin PPSU dropped to its lowest value of ~447 Pa-s at a higher temperature than the modified PPSU, at approximately 415 °C. The differences may be attributed to wall slip and/or the coating on the PPSU pellets. If wall slip was not a factor, then the lower melt viscosity temperature in modified PPSU could infer that more efficient compounding occurs with the additive applied to the PPSU pellet surface. Although ~415 °C is the low melt viscosity temperature, the maximum extrusion temperature is recommended near 388 °C according to the vendor.

The torque vs time profile from the steady state shear experiments is shown in Fig. 1(b). The maximum torque was lower for modified PPSU than the virgin PPSU pellets; however, its curve was broader for the modified PPSU compared to the neat. The behavior may be attributed to the more structured thermoplastic mixture delaying reach to the steady state, which is often observed in filled polymers.

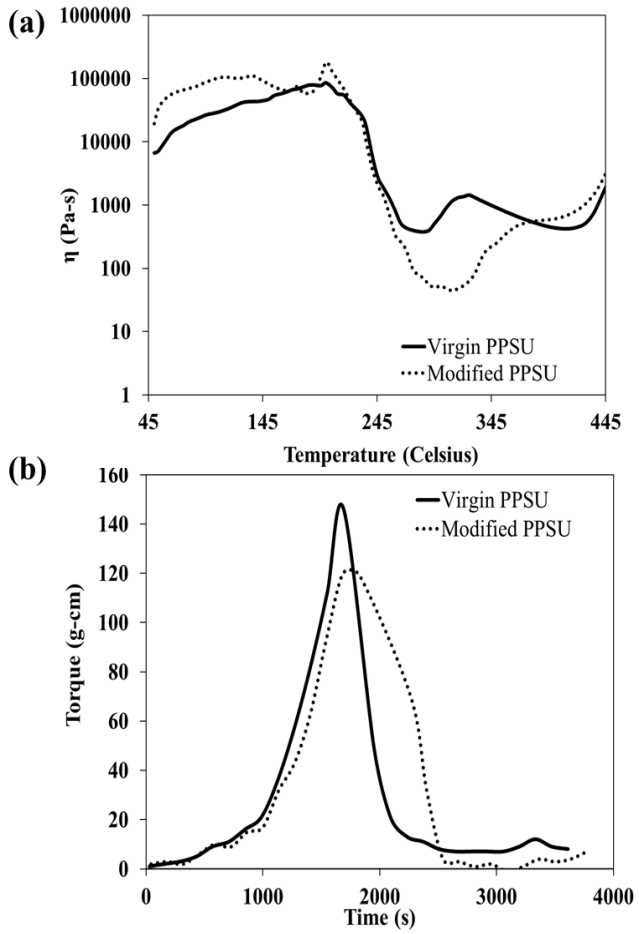

Fig. 1 (a). Temperature-dependent viscosity for virgin polyphenylsulfone (PPSU) and modified PPSU. (b). Steady state shear torque vs time for PPSU and modified PPSU at 380 °C.

Partial discharge measurements of PPSU-BN film samples are represented in Fig. 2. The sample sets varied by BN filler size (micronized and/ or nano), filler incorporation strategy and average thickness. The effect of sample thickness on PDIV is clear, with the PPSU A batch having the highest thickness and demonstrating the highest PDIV out of all the samples. It also appears that higher concentrations of the blended micronized + nanoBN in PPSU A showed the most noticeable change in PD and illustrated that higher concentrations of the micronized + nanoBN mixture adversely affected PDIV. As found in other work, it is clear that nanofillers have a more prominent effect over interfacial interactions with the host polymer compared to microcomposites, which are believed to be attributed to stronger ionic interactions and smaller interfiller spacing [5]. On the other hand, the relatively unchanged PD resistance for the PPSU A composite at ~7.7 wt% compared to the unfilled specimen, and the lower PD resistance for the ~15.1 wt% PPSU A micronized + nanoBN composites could suggest that large cavities or agglomerates existed in the microstructure. The PPSU B samples showed a higher average PDIV in samples near ~14.3 wt% BN compared to its virgin PPSU

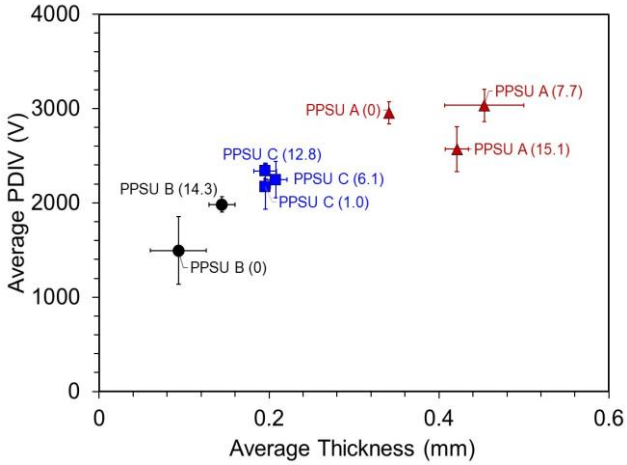

Fig. 2. Effect of sample thickness on partial discharge inception voltage (PDIV) as a function of processing method and boron nitride (BN) concentration. Values in parenthesis specify BN concentration.

counterpart.

The extent of the PDIV increase that was attributed to sample thickness is unclear. If PDIV increased with thickness for all specimens, then it suggests that PDIV is not strongly dependent on filler concentration when micronized BN platelets are used. In the case of the PPSU C samples, which were extruded with modified PPSU and micronized BN as the predominant filler, the results revealed that there was a small increase in PDIV with increasing micronized BN concentration.

The effects of BN concentration on dielectric strength and thermal conductivity are observed in Fig. 3. Fig. 3 (a) showed that the dielectric strength for PPSU A samples decreased as the blended micronized + nanoBN filler concentration increased. Although Fig. 3 (a) showed that the optimal BN filler concentration was between 6 and 12 wt% for the PPSU B and C batches, PPSU C, which consisted of modified PPSU, showed the highest dielectric strength at the lowest BN concentration.

Fig. 3 (b) illustrated that thermal conductivity values for the PPSU B sample were highest overall, whereas the PPSU C sample had the lowest thermal conductivity with increasing BN concentration. Thermal conductivity observed in the PPSU C sample more than likely correlated with the rheological behavior of the modified PPSU material shown in Fig. 1. In addition to the additive on the PPSU pellets leading to a decrease in low melt viscosity temperature, viscosity, and torque, the coating appeared to have a lubrication effect on the polymer and increased the separation distance between neighboring micronized BN platelets. The spacing distance between particles was more obvious in the EDS images in Fig. 4.

Energy dispersive spectrometry depicted differences in BN dispersion and distribution in the PPSU-BN composite tapes, even though the BN concentrations were similar. Fig. 4(a) showed the extruded PPSU-BN composite tape with approximately 7.7 wt% micronized + nanoBN, whereas Fig. 4(b) showed the composite tape that was extruded with the

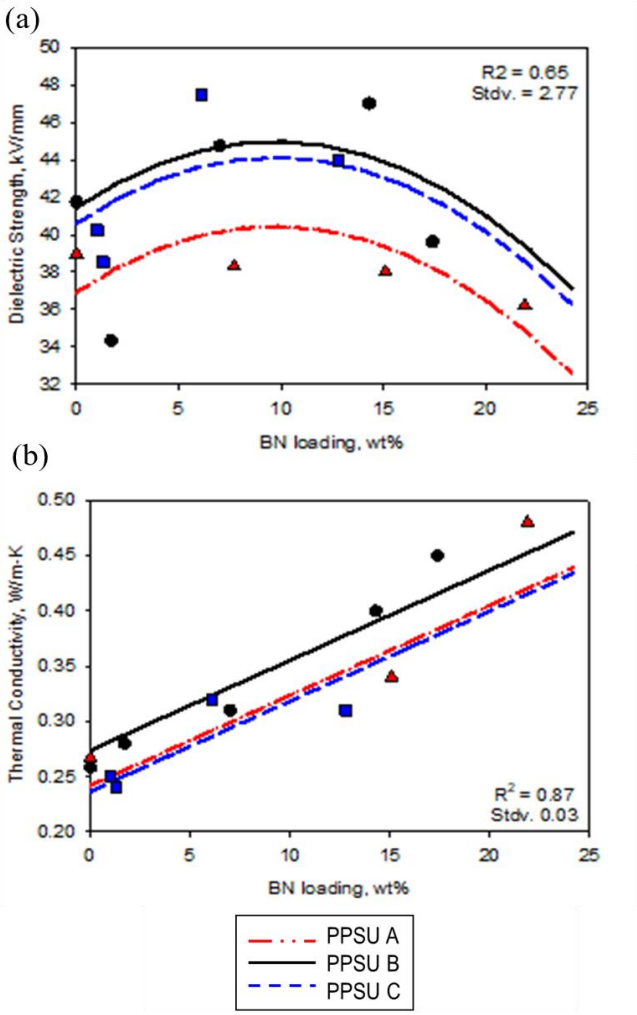

Fig. 3. Effect of boron nitride content on (a) dielectric strength and (b) thermal conductivity for PPSU A, -B, and -C samples. Dielectric breakdown tests were conducted in air.

modified PPSU and micronized BN. The filler distribution and dispersion appeared more uniform in the PPSU C films. It should be noted that EDS images only show a very small area of the sample, so more images will have to be collected to offer a more accurate representation of filler dispersion and distribution in the composite films.

To better understand how the BN incorporation strategy affected the insulating performance caused by microstructural changes, Fig. 4(c) showed the current leakage of the extruded PPSU-BN composite tapes as a function of BN filler concentration and sample preparation method. Fig. 4(c) shows how all PPSU samples displayed an exponential increase in current with increasing voltage. PPSU B samples exhibited the sharpest rise in current with increasing voltage, with the virgin PPSU B film slightly surpassing the 14.3 wt% PPSU-BN tape. The PPSU A samples displayed the slowest rise in current leakage with increasing voltage, which more than likely attributed to the PPSU A samples having the highest thickest out of all samples.

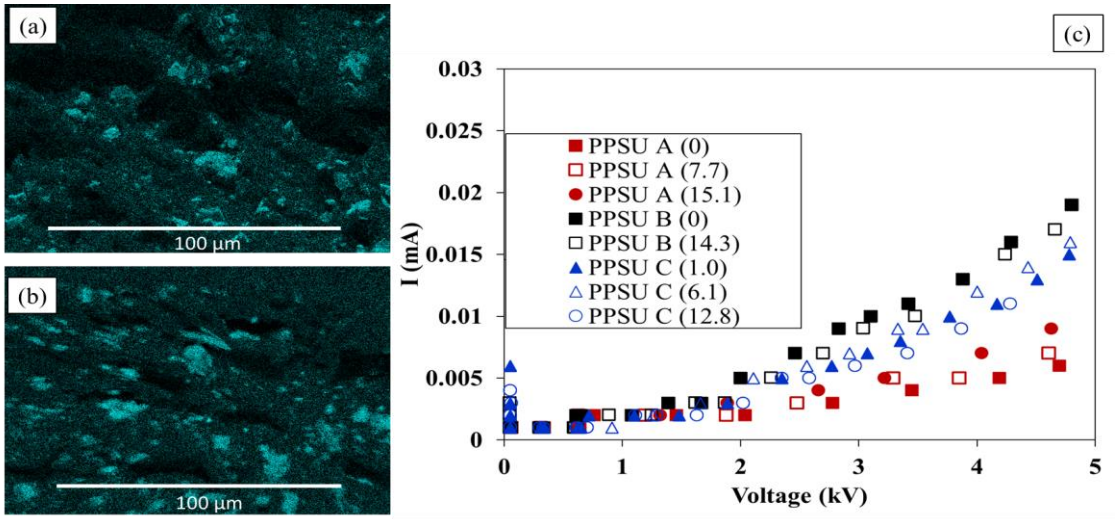

Fig. 4 (a) Energy dispersive spectrometry (EDS) of PPSU A (7.7 wt%) with blended micronized/nano boron nitride fillers; (b) EDS of PPSU C (6.1 wt%) extruded using PPSU C; (c) effect of sample preparation on leakage current in extruded PPSU-hBN tapes

Similar to the PDIV data in Fig. 2, higher concentrations of the blended micronized + nanoBN filler mixture did not correspond to lower current as the voltage increased. The remaining PPSU B and C samples had similar thickness, and it was observed that PPSU C composite tapes made with the modified polymer showed slightly more resistance to current leakage at lower concentrations compared to the unmodified PPSU B films.

IV. Conclusion

In this work we discussed how the rheological, dielectric, and thermal properties varied by the method selected to introduce fillers into extruded thermoplastic composite films. A decrease in torque and melt viscosity was observed when the raw PPSU thermoplastic pellets were modified compared to the as-received pellets, which were favorable for mixing and compounding. The modified polymer pellets may have shown evidence of wall slip; therefore, rheology will be repeated using roughened or textured parallel plates to eliminate this effect.

Partial discharge measurements appeared mostly thickness dependent, and more analysis is needed to understand the effects of processing parameters and microstructure on the PDIV. The PPSU C sample with the modified polymer demonstrated the highest average dielectric strength at the lowest BN loading, near 6.1 wt%, followed by the PPSU B with ~14.3 wt% BN. On the other hand, the PPSU C sample had the lowest thermal conductivity values. A higher concentration of BN would have to be introduced to bring the volume fraction of this material closer to its percolation threshold in order to significantly reduce phonon scattering effects. Leakage current correlated with the PDIV data but observing the rate change in current with increasing voltage offered a clearer illustration of how changes in microstructure caused by processing variations affected dielectric properties.

Acknowledgment

The authors acknowledge funding from the NASA Advanced Air Transport Technologies program. The authors also acknowledge Dr. Jin Wang from The Ohio State University for partial discharge measurements, Linda McCorkle for rheology and Wayne Jennings for EDS.

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