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An Investigation of Mechanical Properties and Breakdown Strength of Polypropylene/Ultra-High Molecular Weight Polyethylene Nanocomposites

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***Abstract*- This work studies the effects of surface-modified magnesium oxide (MgO) nanofiller on the mechanical properties and AC breakdown strength of polypropylene (PP) and ultra-high molecular weight polyethylene (UHMWPE) composites. The inclusion of nanoparticles results in improved interfacial interactions as a consequence of the transition from separate crystallization to co-crystallization. Thus, nano-MgO enhances the breakdown strength of PP/UHMWPE by acting as a compatibilizer between the PP/UHMWPE. UHMWPE decreases Young's modulus and ultimate tensile strength while increasing elongation at yield point in the PP matrix. PP/UHMWPE has increased elasticity due to weak interfacial adhesion. The addition of nano-MgO, however, promotes stronger bonding between PP and UHMWPE phases, providing stiffer composites than those without MgO. There are no apparent differences between PP/UHMWPE and PP/UHMWPE/MgO regarding ultimate tensile strength. It is obvious that the dielectric breakdown strength and elastic enhancement are significantly influenced by the interfacial adhesion between the polymers.**

I. Introduction

Polypropylene (PP) with its good dielectric properties and high working temperature has the potential to replace cross-linked polyethylene for high voltage cable insulation materials. However, brittleness at room temperature and poor thermal conductivity are the drawbacks of PP as a cable insulation material.

Generally, thermoplastic elastomers are used to improve the elasticity of PP, for example polyolefin elastomer (POE) [1] [2]. However, the increased thermal conductivity of heterogeneous polymer-polymer composites is restricted by the relatively low thermal conductivity of the polymer fillers.

Blending PP with a filler that would improve the elasticity and has high thermal conductivity could be a potential solution to this problem. Ultra-high molecular weight polyethylene (UHMWPE) has been mainly reported to enhance toughening of PP composites [3] [4]. Some investigations showed PP/UHMWPE systems had improved elasticity [5]. They claimed that the decline in compound crystallinity is the reason for the decrease in Young's modulus and tensile strength. In addition, previous study showed that, compared to neat PP, the combination of UHMWPE and PP increased thermal conductivity by 35% [6]. Unfortunately, the AC breakdown strength of PP/UHMWPE slightly dropped.

Polypropylene nanocomposites have demonstrated improved dielectric properties, such as increased dielectric breakdown strength and space charge suppression [7]. Typically, a surface-modified nanofiller is required to enhance nanofiller dispersion and its interaction with the polymer matrix. For example, the dielectric breakdown strength of PP was improved by adding magnesium oxide (MgO) nanoparticles surface-modified by 3-aminopropyl triethoxy (EA) [8]. Chi et al. demonstrated that surface-modified silica (SiO_2) in PP/POE improved tensile strength and breakdown strength due to the strong interaction of polymer chains [9].

In this work, tensile strength and dielectric breakdown strength of PP/UHMWPE with surface-modified MgO nanoparticles are investigated. X-ray diffraction (XRD) and differential scanning calorimetry (DSC) are used to evaluate the crystal structure and thermal behavior of the composites.

II. Materials and Characterization

A. Materials and Compounding

Isotactic PP and UHMWPE were obtained from Sigma Aldrich. MgO nanoparticles with mean particle size of 10–30 nm were purchased from SkySpring Nanomaterial, Inc. Silane coupling agent of EA was obtained from Aladdin industrial Inc.

An anhydrous method was used to produce surface-modified nanoparticles. The full explanation of surface modification was published in [8]. Here, a solution blending method was used to mix PP with 2.5 wt.% surface modified MgO in four different UHMWPE contents. This wt.% nano-MgO was chosen based on previous work [8], which demonstrated that the dielectric breakdown strength of PP increased by up to16%.

Samples were prepared using a solution blending method in which UHMWPE and MgO powder were sonicated in xylene for 30 min before pouring into a boiling PP solvent. The solid was then precipitated by adding the mixture to methanol. The compound was dried, first, for 24 h in a fume cupboard and, then for 3 d in a vacuum oven. The dry solid material was pressed at 180 °C to produce thin, which were subsequently cooled in distilled water at room temperature. Prior to any measurements, the films were stored in a vacuum vessel to minimize the impact of ambient humidity.

B. Characterization Techniques

XRD was used to investigate the crystalline structure using a Bruker Power X-ray Diffraction, D2 Phaser. Diffraction patterns were recorded from 2θ equals 10° to 80° with λ=1.54184 A° and a step size of 0.0202° increment.

A Mettler Toledo DSC-820 was used to examine melting and crystallization behavior. The melting behavior was determined by heating a 5 mg sample from 20 °C to 200 °C, holding it for 5 min, and then cooling it from 200 °C to 20 °C to determine the crystallization behavior. All measurements were performed in nitrogen atmosphere at a heating/cooling rate of 10 °C/min.

AC breakdown was carried out using a method based on ASTM D149. Samples with a thickness of 100±10 μm were sandwiched between two spherical electrodes with a diameter of 6.3 mm. To avoid flashover, the test cell was submerged in silicone oil. The samples were subjected to an AC voltage ramp at a rate of 1.5 kV/s until breakdown. Weibull statistics via OriginLab were used to analyze twenty data points per sample.

A Tinius Olsen H25KS Tensometer was used to investigate stress-strain behavior according to ASTM D638-14. The crosshead was moved at a constant 500 mm/min speed until the sample broke in the gauge section. At least five measurements were made using dumbbell-shaped samples with a thickness of 1.0±0.1 mm.

III. Results and Discussion

A. Thermal and Crystal Evaluation

Fig. 1 shows the XRD curves of different materials. The neat PP diffractogram has typical peaks at 2θ of 14.1°, 17°, 18.6°, 21.3°, 21.8°, 25.6° and 28.7°, which correspond to the 110, 040, 130, 111, 131, 041, 060 and 220 crystal planes of the α-crystal structure [10]. UHMWPE diffractogram has peaks at 2θ of 21.6°, 24° and 36.3°, which relate to the 110, 200, and 020 crystal planes of orthorhombic PE crystals [5].

Compared to neat PP, there are no obvious peaks at 2θ of 16° and 20°, which represent the β crystal form of PP, for the PP/UHMWPE and PP/UHMWPE/MgO compounds. This indicates that UHMWPE and MgO have no effects on the PP crystal forms. However, the 110 reflection of UHMWPE is combined with the PP reflections of 111, 131, and 041 in the blends. XRD measurement cannot be used to determine the changes in UHMWPE and PP crystallinity in the compounds.

DSC measurements were performed, and the melting and crystallization behavior are shown in Fig. 2. DSC results obtained from neat PP, neat UHMWPE and PP/UHMWPE were reported in [6] which, here, were used to compare with PP/UHMWPE/MgO composites. DSC scan shows a clear melting peak for PP and UHMWPE, while a small peak at 146 °C represents the melting point of β-PP crystal. This β-PP melting peak cannot be observed in PP/UHMWPE/MgO composites. This change would be related to the MgO-induced modifications at the PP/UHMWPE interfaces. However, XRD measurements cannot record the small β-PP crystal in PP/UHMWPE. This can be attributed to the sensitivity of the method may not be sufficient to confirm the β crystal structure.

Total crystallinity was calculated from the XRD and DSC measurements. XRD crystallinity was determined by dividing the total area of the crystalline peaks by the corresponding area of all peaks, while the details of the DSC crystallinity calculation were explained in [6] [8]. The overall crystallinity from both DSC and XRD experiments is compared in Fig. 3. Neat PP and UHMWPE show XRD total crystallinity of 55.3% and 54.4%, respectively, which are ~12% higher compared to the DSC measurement. Krishnakumar et al. [11] performed crystallinity of PP with three different methods: XRD, density measurement and DSC. The crystallinities obtained by XRD and density measurement were similar, while those from DSC were slightly different, especially at high crystallinity. They claimed that the lower crystallinity could be the result of the recrystallization during DSC measurement.

In both measurements, the crystallinity of composites is around 20% higher than in neat samples. Adding MgO particles provides a ~3% increase in crystallinity, which is insignificantly higher than for those without MgO. More than 50% overlap between error bounds demonstrate that the MgO nanoparticles have no significant impact on the crystallinity of the PP and UHMWPE phases.

Although introducing MgO particles has little effect on crystallinity, it has a significant impact on crystallization behavior. A single crystallization peak at 118 °C in the PP/UHMWPE/MgO instead of a bimodal behavior indicates that the polymer composites have better interfacial interaction. MgO nanoparticles act as heterogeneous nucleating agents, accelerating the crystallization process. The crystal growth acceleration due to nanoparticle may cause the lack of β-PP melting peak. The formation of β-PP crystals in PP/UHMWPE is due to long relaxation times from long UHMWPE chains [12].

B. AC Breakdown Strength

Fig. 4 shows Weibull AC breakdown strength (BDS) plots of PP/UHMWPE/MgO composites compared to neat PP. The graph includes scale (α) and shape (β) parameters. Fig. 5 demonstrates BDS between PP/UHMWPE and PP/UHMWPE/MgO with 95% confidence interval. A summary of the PP/UHMWPE data has been discussed in [6], and here, the study is expanded and used as a reference to examine the effects of nanoparticles. The BDS of PP/UHMWPE blends is improved by adding 2.5 wt.% nano-MgO. For example, at 20 wt.% UHMWPE, the BDS for PP/UHMWPE/MgO is 9% higher than PP/UHMWPE and 6.2% higher than neat PP.

Increasing BDS would be supported by better two-phase polymer mixing. As can be seen from the DSC crystallization in Fig. 2, the presence of MgO nanoparticles causes PP/UHMWPE interfaces to co-crystallize instead of crystallizing separately. This suggests that nanoparticles act as a compatibilizer between two immiscible polymer blends [13], which result in improved interfacial interaction at the interfaces. The BDS in nanocomposites increases with increasing UHMWPE. The small drop at 30 wt.% compared to 20 wt.% is insignificant considering measurement uncertainties.

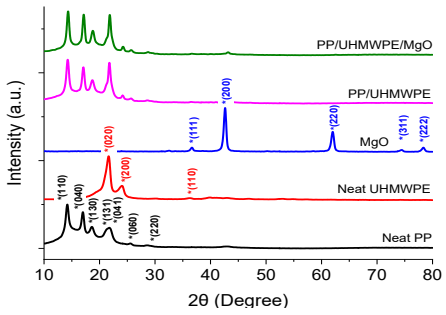

Fig. 1. XRD curves of (80/20) PP/UHMWPE and PP/UHMWPE/MgO composites.

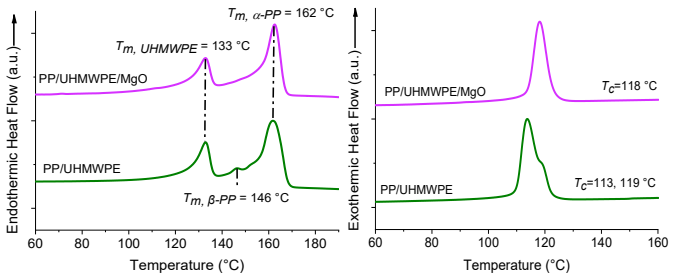

Fig. 2. DSC curves of melting (left) and crystallization (right) temperatures of (80/20) PP/UHMWPE and PP/UHMWPE/MgO composites.

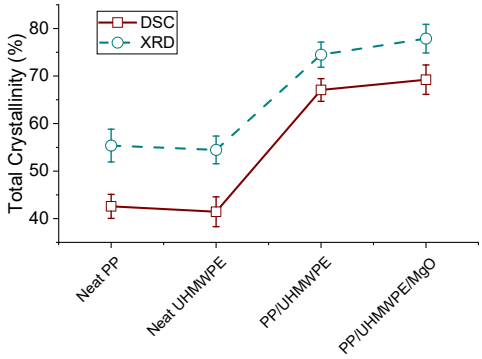

Fig. 3. Crystallinity of PP/UHMWPE blends from XRD and DSC.

C. *Mechanical Properties*

Fig. 6 shows graphs of the stress-strain curves of PP/UHMWPE composites at various compositions. With an increase in UHMWPE content, the ultimate tensile strength decreases about 15% at 30 wt.%. Young's modulus and elongation at yield point of PP/UHMWPE composites as a function of UHMWPE content are shown in Fig. 7. Young's modulus is calculated from the slope in linear region of the strain-stress curve. Young's modulus decreased monotonically with increasing UHMWPE content compared to neat PP. It decreased by 24% when the UHMWPE content is increased up to 30 wt.%. A material with a low Young's modulus is more likely to deform under stress than one with a high Young's modulus. The amount of stress a material can withstand before permanently deforming is measured by elongation at yield point, which monotonically increases with UHMWPE contents and reaches 12% higher than neat PP at 30 wt.% UHMWPE.

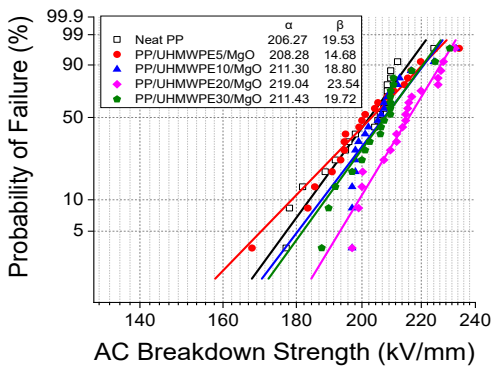

Fig. 4. Weibull probability plots of PP/UHMWPE/MgO blends.

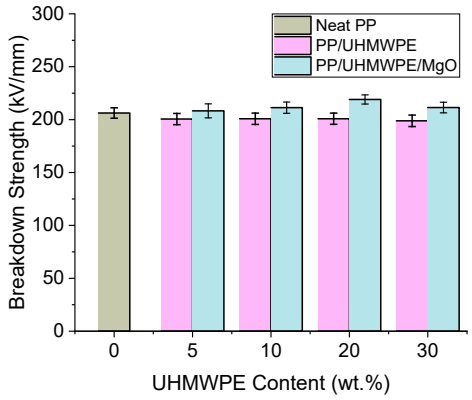

Fig. 5. The comparison of breakdown strength with 95% confidence interval between PP/UHMWPE and PP/UHMWPE/MgO blends.

To investigate the effects of UHMWPE on mechanical properties, we first consider the nature of the binary PP/UHMWPE system. PP/UHMWPE is a rigid-brittle system, with UHMWPE being a rigid phase mixed into a brittle PP matrix. This system is generally used to enhance the toughness and stiffness of the brittle matrix [3]. Debonding or cavitation processes take place at the interface when the applied stress is higher than the interfacial adhesion strength. This results in the formation of microscopic voids, which can increase tensile strength and toughness. The deformation and absorption of strain energy are caused by rigid phase instead of the brittle matrix if two polymer phases have strong interfacial adhesion and good dispersion [4].

However, in this study, the weak bonding strength at the interfaces due to the incompatibility between PP and UHMWPE (bimodal behavior) result in lower tensile strength. This is consistent with the findings in [3], which showed that the PP/UHMWPE prepared by a twin-screw extruder had a immiscibility and poorer dispersion, resulted in lower tensile strength, than for the composites produced by a four-screw extruder.

Although an elastomeric material generally contributes to increased elasticity in the brittle PP matrix, rigid-brittle PP/UHMWPE composites exhibit increased elasticity. This could be attributed to poor interfacial bonding between the two materials. According to the findings in [14], PP/LDPE blends

had a lower Young's modulus than PP/HDPE blends. This is due to PP/LDPE having weaker interfacial interaction, as demonstrated by bimodal behavior in DSC scan.

The addition of nano-MgO particles into PP/UHMWPE blends promotes co-crystallization at PP/UHMWPE interfaces (a single crystallization peak). This suggests improved interfacial interaction of the two polymers, resulting in the increased Young's modulus and decreased elongation at yield point in PP/UHMWPE blends as shown in Fig. 7. This is in line with the results in [9], when SiO_2 nanoparticles were added to PP/POE. The authors claimed that the introduction of nanoparticles formed a strong interaction with the polymer chains, increasing the stiffness of materials. However, as shown in Fig 8, the enhanced interfacial interaction due to MgO has only a small impact on the ultimate tensile strength in PP/UHMWPE.

IV. Conclusion

In this work, the mechanical properties and AC breakdown strength of PP/UHMWPE and PP/UHMWPE/MgO composites are investigated. The breakdown strength of PP/UHMWPE can be increased by adding nano-MgO. MgO acts as a compatibilizer at the PP/UHMWPE interface. UHMWPE added to a PP matrix increases yield point elongation while decreasing ultimate tensile strength and Young's modulus. Due to the poor interfacial adhesion at the interfaces, UHMWPE can improve the elasticity of PP. Conversely, a stiffer PP composite is produced after adding nano-MgO, due to stronger bonding between the two polymer phases. When comparing PP/UHMWPE with and without MgO, there are no

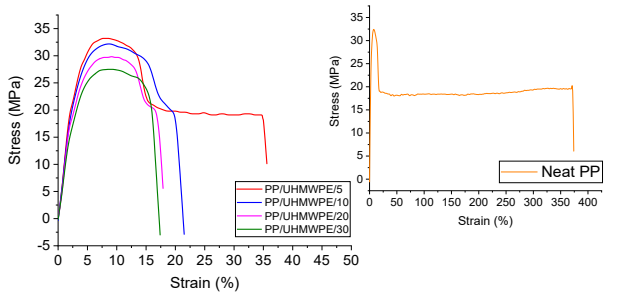

Fig. 6. Stress-strain curves of neat PP and PP/UHMWPE composites.

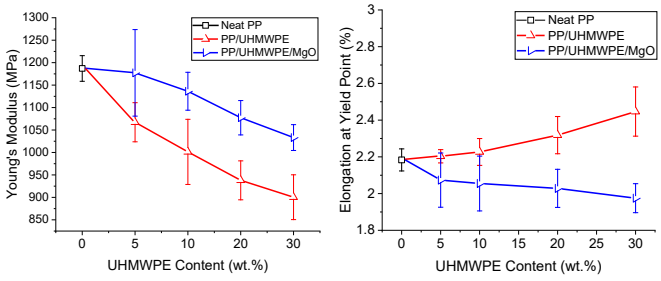

Fig.7. Young's Modulus (left) and elongation at yield point (right) of different PP/UHMWPE compounds.

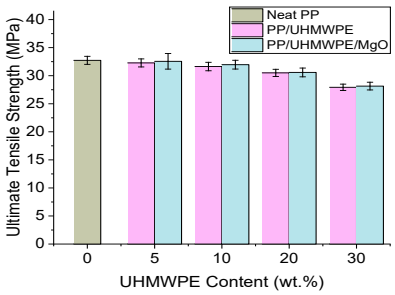

Fig.8. Ultimate tensile strength at various PP/UHMWPE composites.

significant differences in ultimate tensile strength. Therefore, it can be concluded that the interfacial adhesion between the polymer phases is directly related to breakdown strength and elasticity in PP/UHMWPE blends.

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