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Dispersion and interfacial management for designing polypropylene nanocomposites

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Along with the worldwide trend toward more energy-efficient structure and reduced emissions in industry and transportation, there is an increasing demand for developing lightweight, high-performance, and low-cost materials. Plastics are among the material of choices, which not only provide the light-weight structure, but also require less energy manufacturing processes into the final products as compared to other materials such as metals and glass. Currently, nanofillers are widely used to enhance mechanical and thermomechanical performances of polymers. However, the addition of nanofillers often results in a considerable loss in the toughness and elongation at break due to the rigidity of the embedded nanoparticles as well as loose interfaces. Thus, it is essential to explore a way to alleviate these problems in order to devise high-performant polymer nanocomposites. In this dissertation, I exploited different strategies, with the aim to fabricate polymer nanocomposites bearing desired properties without sacrificing the toughness. The main research results are as follows:

In **Chapter 2**, organically modified SiO_2 was used to prepare graft-type polypropylene (PP) nanocomposites, where PP containing less than one functional group per chain prepared by catalyzed copolymerization between propylene and 7-octenyltrimethoxysilane (PP-OTMS) was used as a reactive matrix. It was found that surface modification of SiO_2 with silane coupling agents enhanced the hydrophobicity of SiO_2 surfaces to improve the dispersion of SiO_2, which in turn promoted the in-situ grafting of PP-OTMS onto filler surfaces via hydrolysis/condensation. The modification with long alkyl chains led to efficient grafting to strengthen the interfacial interaction, and recovered the deterioration in the elongation at break from its plasticizing ability. This provided an opportunity to balance the reinforcement and the toughness of the materials, which is hardly achieved by the in-situ grafting or surface modification alone.

In **Chapter 3**, a novel PP matrix bearing methoxy-phenyl side-functional groups (PP-ADMB) was synthesized by copolymerization of propylene with 4-allyl-1,2-dimethoxybenzene. The introduction of methoxy-phenyl side-functional groups even at a trace amount not only improved the compatibility between the matrix and fillers to promote the dispersion of SiO_2, but also softened the polymer, which dramatically improved the elongation at break and the toughness without deteriorating the reinforcement of PP nanocomposites. These functions of PP-ADMB make it advantageous over the homo PP in balancing the mechanical properties of nanocomposites.

In **Chapter 4**, reactor granule technology (RGT), where TiO_2 nanoparticles were in-situ formed in the porous structure of PP granules, was used to fabricate biaxially oriented polypropylene (BOPP) nanocomposites. Benefitted from highly dispersed TiO_2 nanoparticles, biaxial stretching without breakage was achieved. The permittivity of nanocomposites was greatly enhanced beyond the classical mixing rule by a small amount of TiO_2. This suggested the critical role of the interphase around nanoparticles.

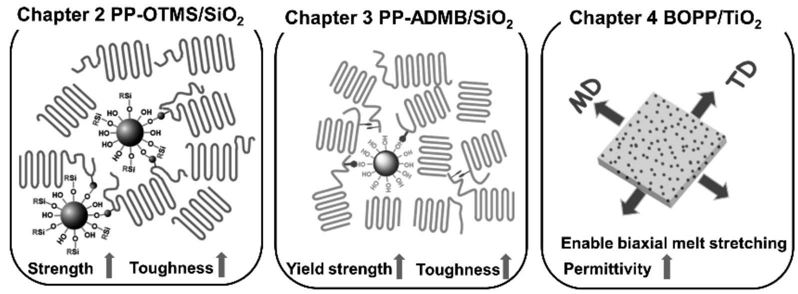

Fig. 1. Materials design implemented in this thesis.

In conclusion, three different strategies were successfully implemented to fabricate PP nanocomposites (Fig. 1), which facilitated desired properties without sacrificing the ductility of the material. These developed strategies are believed to be promising for designing high-performance polymer nanocomposites.

Keywords: Polymer nanocomposites; Mechanical properties; Reactor granule technology; Biaxially oriented polypropylene; Dielectric properties