Division of Materials Chemistry
– Chemistry of Polymer Materials –

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Scope of Research

We perform kinetic and mechanistic analyses toward understanding chemical and physicochemical reactions occurring in polymerization systems and better routes for synthesis of well-defined polymers. In particular, new well-defined polymers or polymer assemblies are prepared by living polymerization techniques, and their structure–properties relationships are precisely analyzed. Projects in progress include: 1) kinetics and mechanisms of living radical polymerization (LRP); 2) synthesis of new polymeric materials by living polymerizations and their structure/properties studies; and 3) synthesis, properties, and applications of concentrated polymer brushes (CPB).

KEYWORDS

Precision Polymerization
Living Radical Polymerization
Polymer Brush
Hybrid Materials
Biointerface

Selected Publications

Tsujii, Y.; Nomura, Y.; Okayasu, K.; Gao, W.; Ohno, K.; Fukuda, T., AFM Studies on Microtribology of Concentrated Polymer Brushes in Solvents, *J. Phys.: Conf. Ser.*, **184,** 012031 (2009).

Arita, T.; Kayama, Y.; Ohno, K.; Tsujii, Y.; Fukuda, T., High-Pressure Atom Transfer Radical Polymerization of Methyl Methacrylate for Well-Defined Ultrahigh Molecular-Weight Polymers, *Polymer*, **49,** 2426-2429 (2008).

Tsujii, Y.; Ohno, K.; Yamamoto, S.; Goto, A.; Fukuda, T., Structure and Properties of High-Density Polymer Brushes Prepared by Surface-Initiated Living Radical Polymerization, *Adv. Polym. Sci.*, **197,** 1-45 (2006).

Ohno, K.; Morinaga, T.; Takeno, S.; Tsujii, Y.; Fukuda, T., Suspension of Silica Particles Grafted with Concentrated Polymer Brush: Effects of Graft Chain Length on Brush Layer Thickness and Colloidal Crystallization, *Macromolecules*, **40,** 9143-9150 (2007).

Ohno, K.; Morinaga, T.; Koh, K.; Tsujii, Y.; Fukuda, T., Synthesis of Monodisperse Silica Particles Coated with Well-Defined, High-Density Polymer Brushes by Surface-Initiated Atom Transfer Radical Polymerization, *Macromolecules*, **38,** 2137-2147 (2005).

Structure Analysis of Composite Membranes of Polymer-brush-afforded Nanoparticles and Ionic Liquid

Previously, we succeeded in fabricating a solid membrane of high ionic conductivity using self-assembled nanoparticles with CPBs in an ionic liquid (IL) and in developing a novel bipolar-type lithium-ion rechargeable batteries, where CPB represents the concentrated polymer brush, successfully and systematically synthesized by living radical polymerization techniques. This success was caused by the CPB effects (including super lubrication) originating from the highly stretched-chain conformation and high-dense segmental density in a solvent. In this study, we achieved in analyzing the higher-order structure of the composite membranes using concentrated ionic liquid-type polymer brush-modified nanoparticles (PSiP) and IL by ultra-small angle X-ray scattering (USAXS). The results showed that the higher-order structure of PSiP/IL composites changed from disorder to random hexagonal-closed-packing (rhcp), and face-centered-cubic (fcc) as the concentration of PSiP increased. As concentrated polymer brushes have unique properties such as high elasticity in a good solvent, it can be concluded that these results are applied to the model for particle assemblies with high-elasticity.

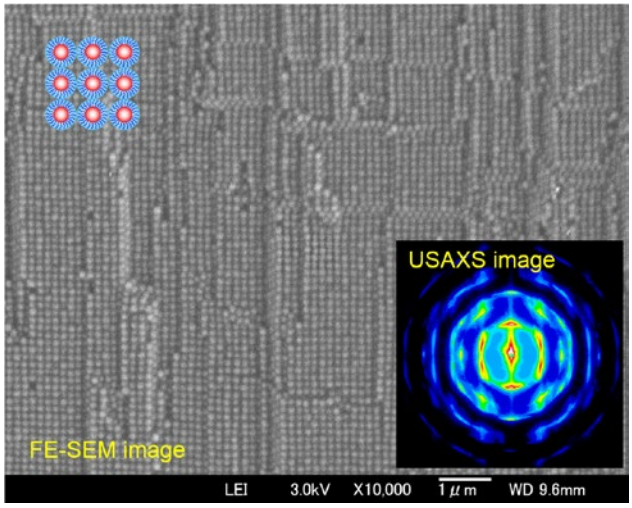

Figure 1. USAXS and FE-SEM images of composite membranes of concentrated ionic liquid-type polymer brush-modified nanoparticles and ionic liquid.

Surface Engineering of Cellulose Nanofiber by Adsorption of Diblock Copolymer Dispersant for Green Nanocomposite Materials

An effective approach for the dispersion of hydrophilic cellulose nanofiber (CNF) in hydrophobic high-density polyethylene (HDPE) is presented using adsorption of a diblock copolymer dispersant. The dispersant consists of both resin compatible poly(lauryl methacrylate) (PLMA) and cellulose interactive poly(2-hydroxyethyl methacrylate) blocks. The PLMA-adsorbed CNFs are characterized by FT-IR and contact angle measurement, revealing successful hydrophobization. X-ray CT imaging shows there are apparently less CNF aggregates in the nanocomposites if adding amount of the dispersant was enough. The good dispersion results in a high mechanical reinforcement, corresponding to 140 % higher Young's modulus and 84 % higher tensile strength than the neat HDPE. This study represents a potential approach to mediate CNF-CNF and CNF-resin interactions and to influence the CNF dispersion significantly without chemical functionalization, which may be compatible with a low-cost and environmentally green process and therefore an industrial application. Based on this concept, simultaneous nano-fibrillation compounding (SFC) process has been developed.

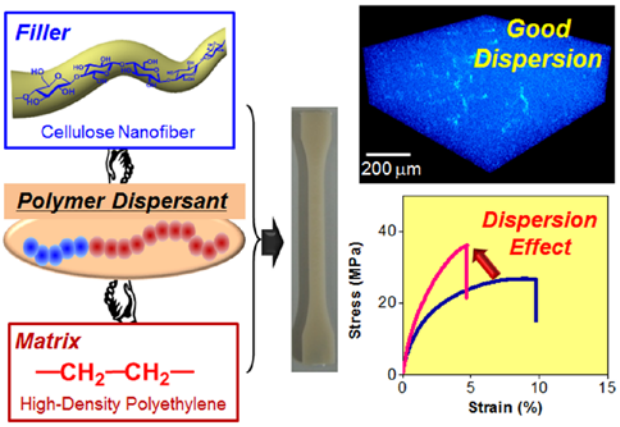

Figure 2. X-ray CT image and strain-stress curve of CNF-reinforced HDPE nanocomposite materials.