Frontier Chemistry Center
フロンティア化学教育研究センター

演　題：**Well-Defined Polyethylene-Based Block Copolymers and Molecular Brushes by Combining Anionic or Ring Opening Metathesis Polymerization with Polyhomologation**

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場　所：工学部材料・化学棟大会議室（MC102）

要　旨：Access to model (high degree of structural, molecular weight and compositional homogeneity) polyethylenes (PEs) and PE-based block copolymers is necessary in order to elucidate the structure-properties relationships, which are very important for polymer performance. The synthesis of such polymers requires a truly living process. These approaches usually require high vacuum techniques, which are demanding, time consuming and leads to a small quantities of products. Nevertheless, this is a small price to pay given the tremendous potential of model macromolecules for selecting the appropriate structures needed for specific applications. Our group is using the following two methodologies to synthesize PEs and PE-based block copolymers:

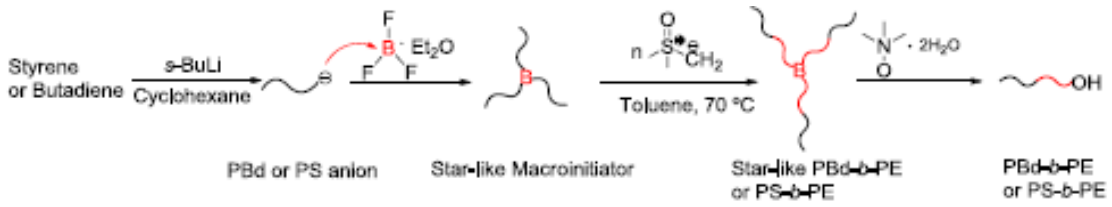
a) Anionic polymerization and Hydrogenation

Hydrogenation of 1,4-polybutadiene with various architectures (star, comb, dendritic, etc.) synthesized by anionic polymerization high vacuum techniques and appropriate post polymerization chemistry [1,2]. A few examples will be given, showing the importance of access to a variety of well-defined structures for a deeper understanding of polyethylene performance.

b). Combination of Anionic polymerization and Polyhomologation

Recently, Shea developed a novel polymerization methodology leading to perfectly linear PEs [2]. The general reaction scheme involves the formation of an organoboron zwitterionic complex between a methylide (monomer) and a trialkylborane Lewis acid (initiator) which breaks down by the intramolecular 1,2-migration. As a consequence, the methylene group of methylide is randomly inserted one by one into the three branches of the trialkylborane leading to a 3-arm PE star. The resulting star is subsequently oxidized/hydrolysed to give perfectly OH-end-capped linear PEs.

By combining anionic polymerization and polyhomologation, through a “bridge” molecule (BF3OEt2), a novel one-pot methodology was developed [3] for the synthesis of PE-based block copolymers (Scheme 1).



Scheme 1. Synthesis of PE-based diblock copolymers by combining anionic polymerization and polyhomologation

Furthermore, by combining polyomologation and ROM polymerization a new strategy was developed for the synthesis of well-defined PE-based molecular brushes [5].

References

*[1] N. Hadjichristidis et al., Macromolecules **2000**, 33, 2424-2436; [2] J. Luo, K. J. Shea. Acc. Chem.Res. **2010**, 43, 1420-1433; [3] H. Zhang, N. Alkayal, Y. Gnanou, N. Hadjichristidis, Macromol. Rapid Commun. **2014**, 35, 378-390; [4] H. Zhang, N. Alkayal, Y. Gnanou, N. Hadjichristidis, Chem. Commun. **2013**, 49, 8952-8954; [5] H. Zhang, Y. Gnanou, N. Hadjichristidis, Polym. Chem., ASAP.*

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