Division of Materials Chemistry
– Polymer Controlled Synthesis –

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Scope of Research

Our research focuses on creation of new organic molecules with potential as key reagents and materials for future science and technologies. Furthermore, we have been developing new organic and polymeric materials based on our tailor-made molecules. For example, we are developing a new living radical polymerization method using heavier heteroatom compounds as controlling agents. Another topic is the synthesis of cycloparaphenylenes, hoop-shaped π-conjugated molecules, based on new synthetic strategies. We also study various condensed states of polymers by both static and dynamic methods to understand the relationship between structure and physical properties.

KEYWORDS

Organic Synthesis
Polymer Synthesis
Living Radical Polymerization
Polymer Properties
Curved π-Conjugated Molecules

Selected Publications

Hashimoto, S.; Iwamoto, T.; Kurachi, D.; Kayahara, E.; Yamago, S., Shortest Double-Walled Carbon Nanotubes Composed of Cycloparaphenylenes, *Chempluschem*, **82,** 1015-1020 (2017).

Lu, Y.; Nemoto, T.; Tosaka, M.; Yamago, S., Synthesis of Structurally Controlled Hyperbranched Polymers Using a Monomer Having Hierarchical Reactivity, *Nat. Commun.*, **8,** 1863 (2017).

Shortest Double-Walled Carbon Nanotubes Composed of Cycloparaphenylenes

Multiwalled carbon nanotubes (CNTs) consist of multiple rolled layers of graphene. This type of materials have interesting electrical, optical, mechanical, and thermal properties. Because variety of applications are expected for these materials, it is important to better understand CNTs on the molecular level. Cycloparaphenylenes (CPPs) are good compounds as a model for the basic studies on CNTs because these nanorings are the smallest possible structures corresponding to the sidewalls of CNTs. It had been predicted that CPPs of different sizes would form host–guest complexes stabilized by attractive van der Waals interactions.

Our group did prove that CPPs form host–guest complexes (example pictured) selectively: when [6]CPP is mixed with equimolar amounts of [8]–[12]CPPs, the only complex that forms is that in which [6]CPP is nested inside [11]CPP, denoted as [11]CPP ⊃ [6]CPP (Figure 1, left). Our work indicated that in other favored combinations, the inner and outer rings differ in size by five phenylene units. The ternary complex [15]CPP ⊃ [10]CPP ⊃ C_{60} formed a "planetary orbit" structure (Figure 1, right). In all of these complexes, the difference in the diameters of the nesting aromatic hydrocarbons was found to be 0.035 nm which corresponds to the distance separating the curved graphene layers in CNTs.

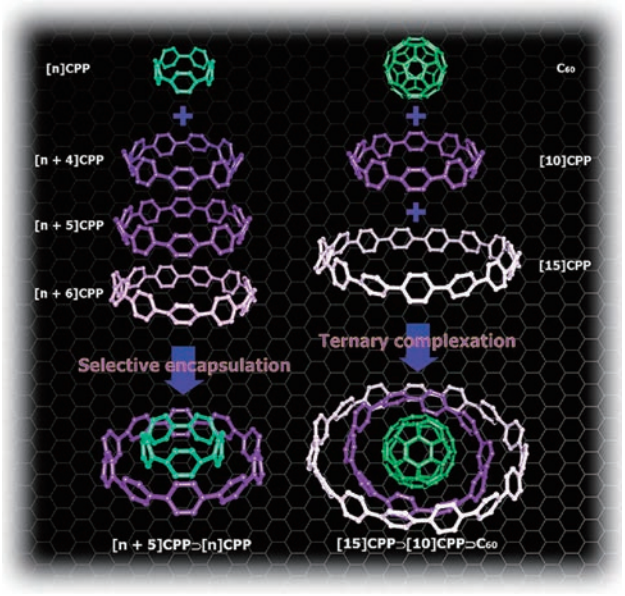

Figure 1. Selective encapsulation of CPPs and/or C_{60} into the larger CPP.

Synthesis of Structurally Controlled Hyperbranched Polymers Using a Monomer Having Hierarchical Reactivity

Hyperbranched polymers (HBPs) have attracted significant attention because of their characteristic topological structure associated with their unique physical properties compared with those of the corresponding linear polymers. Dendrimers are the most structurally controlled HBPs. However, the necessity of a stepwise synthesis have significantly limited their applications in materials science. Several methods have been developed to synthesize HBPs by a one-step procedure, as exemplified by the use of AB_2 monomers and AB' inimers under condensation and self-condensing vinyl polymerization conditions. However, none of these methods provides structurally controlled HBPs over the three-dimensional (3D) structure, i.e., molecular weight, dispersity, number of branching points, branching density, and chain-end functionalities, except under special conditions. We introduced a monomer design concept involving two functional groups with hierarchical reactivity and demonstrated the controlled synthesis of dendritic HBPs over the 3D structure by the copolymerization of the designed monomer and acrylates under living radical polymerization conditions.

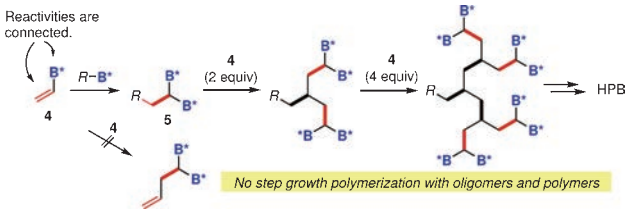

Figure 2. Scheme for the synthesis of structurally controlled hyperbranched polymer.

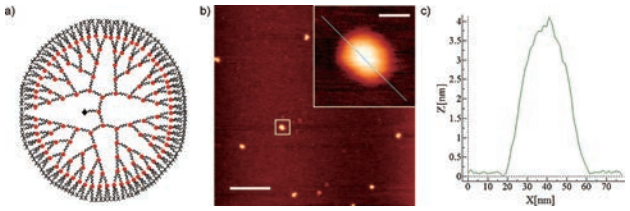

Figure 3. Structure and AFM image of the dendritic hyperbranched polymer.