

演題: Discontinuities in high generation dendronized polymers

Frontier Chemistry Center

フロンティア化学教育研究センター

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講演会

- 場所:工学部材料・化学棟中会議室(MC102)
- 要旨: Dendronized polymers (DPs) belong to a class of comb polymers to which a regularly branched unit (dendron) is attached to each main chain repeat unit.[1] They are usually characterized by their cylindrical shape, rigidified backbone and tuneable thickness. So far the most effective method to synthesize DPs of high generations is the graft-from (or divergent) approach, which means to graft dendrons in a stepwise fashion onto a preformed backbone. By this means, the largest structure-defined linear polymer has been successfully synthesized whose size and shape are comparable to that of tobacco mosaic virus.[2] However, when being applied to higher generations, this approach has encountered an unexpected chain scission during the acidic deprotection of the 5^{th} generation (PG5), which is interesting by itself but detrimental to the synthetic journey towards higher generations.[3] We therefore modified the conventional divergent approach by grafting a second generation dendron (DG2) directly onto the deprotected PG4 core ("n+2" approach), so as to avoid the problematic intermediate at the fifth generation.[4] By this approach, PG6 has been synthesized efficiently with a surprisingly high coverage (~88%) and satisfying yield (~60%). Somewhat unexpectedly, deprotection of PG6 was not accompanied by any chain scission, so the resulting deprotected polymer was dendronized by "n+1" and n"+2" approaches to yield PG7 and PG8 respectively. It should be noted that these unprecedentedly high generations are already beyond the constraint-induced maximum generation $(g_{max} \sim 6.1)$, which sets up the upper limit for structurally perfect DPs.[5] Above g_{max}, considerable amount of defects would appear, which would lead to significantly lower molar masses comparing to the ideal values for perfect DPs that increase exponentially with generation. These changes would further influence other DP behaviours, so they would be in a discontinuous manner at high generations. Herein we report the synthesis of DPs up to PG8 and the observed discontinuities with respect to main chain stability, atom mobility and cross-section size. This should shed some light on the nature of those unusually "thick" cylindrical polymers.

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