

Exploring the Dynamics of Ring-Closing Metathesis of Dendrimers

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Dendrimers represent an attractive class of polymers for studying chemical and structural relationships as a result of their monodispersity, compact globular structure and amenability to incorporation of functional groups. In this group we have constructed monomolecularly imprinted dendrimers employing the ring-closing metathesis (RCM) reaction and were interested in studying this reaction in more detail. Prior cross-linking studies by this group have used dendrimers to analyze the rate of cross-linking and to examine the extent of inter-dendron versus intra-dendron cross-linking. These studies led to questions regarding the dynamics of the cross-linking process and whether mechanical intertwining (catenation) of the dendrons was occurring.

The dynamic nature of the RCM reaction was studied by comparing the results of cross-linking dendrimers with 1st and 2nd generation Grubbs catalyst. To examine the two catalysts in detail, three dendrimers were synthesized. Dendrimer 1 contained a pre-cross-linked portion and a portion with monosubstituted alkenes. Dendrimer 2 contained two pre-crosslinked portions with only disubstituted alkenes. To test whether catenation was occurring, dendrimer 3 was constructed which contained a hydrogenated portion and a portion with monosubstituted alkenes. After cross-linking, the dendrimers were hydrolyzed and the products analyzed by analytical SEC and MALDI-TOF MS to determine whether the dendrimers remained intact or fragmented.

