Efficient Solution-Phase Synthesis of Sequence-Defined Oligourethanes with Precise Chirality Control

Autorzy

Anuj Sharma

Tapendu Samanta

Joanna Cybińska

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Nature relies on precisely defined macromolecules for complex biochemical processes with exceptional specificity and efficiency. To replicate these properties in synthetic systems, chemists have developed sequence-defined macromolecules—polymers with absolute control over monomer sequence and structure, enabling tailored functions. However, their exploration in material science remains limited due to the challenges of synthesis, which is often low-yielding and time-consuming. To address this, we designed and synthesized Fmoc-assisted stereo-controlled sequence-defined oligourethanes (SDOUs) in the solution phase. Our step-economical synthesis employs a two-step, one-pot strategy, eliminating intermediate purification and achieving an average yield of >85% per step. The optimized protocol, using six modified chiral monomers, enables precise stereochemical and sequence control. Thermal analysis revealed that stereochemistry significantly influences thermal transitions, including glass transition, crystallization, and melting behaviors. Tandem mass spectrometry provided in-depth sequencing analysis. We also demonstrated the post-synthetic modification of the SDOUs with dansyl chloride and explored their photophysical properties, such as solvatochromism and aggregation. Circular dichroism analysis highlighted their unique structural and conformational features. This approach establishes scalable, efficient synthetic routes for stereochemically controlled sequence-defined oligourethanes with diverse functional groups.

Słowa kluczowe

chiral oligourethanes, one-pot synthesis, sequence-defined oligomers, sequence folding, structure-property relationship

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