Self-Healing Supramolecular Polymer Glass Composed of Only Macrocycles

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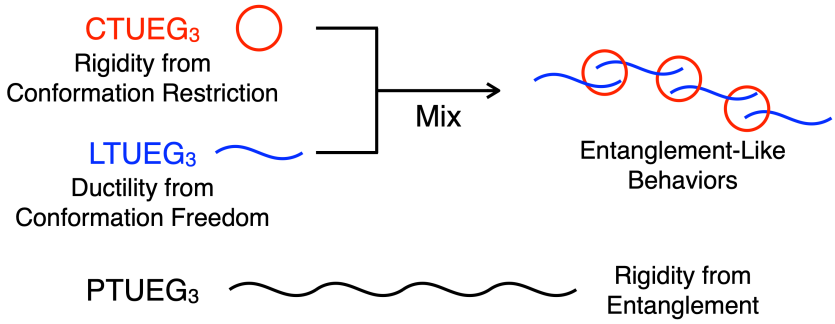
The precise architectural control of polymers has been a key focus in synthetic polymer chemistry due to the potential for novel polymer topologies to unlock unique properties. Cyclic polymer architectures, which lack end groups, have garnered particular interest for their ability to exhibit distinct material properties. However, efficient and practical methods for synthesizing cyclic polymers remain a significant challenge.[1] Therefore, the potential of cyclic molecules as materials has not been deeply explored.

People believe that polymeric materials show high mechanical properties because they have complicated chain entanglement. Here, we serendipitously discovered the formation of cyclic oligomers ($CTUEG_3$), analogous to mechanically robust poly(ether thiourea) ($PTUEG_3$),[2] exhibited rigid properties despite the absence of chain entanglement. We optimized the synthesis route and succeeded in obtaining a specific ring size of $CTUEG_3$ on a gram scale, enabling detailed investigations including mechanical and rheological measurements. Comparisons with the linear oligomer $LTUEG_3$ and $PTUEG_3$ highlighted the rigidity of $CTUEG_3$ due to its conformational restrictions and clarified its unique properties. Furthermore, we also investigated the properties of the mixture of $CTUEG_3$ and $LTUEG_3$. The obtained results from rheological measurements cannot be explained by simple summation of $CTUEG_3$ and $LTUEG_3$ properties, indicating the formation of rotaxane structures. We will focus on the fundamental interest in $CTUEG_3$ from a viewpoint of materials science.



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