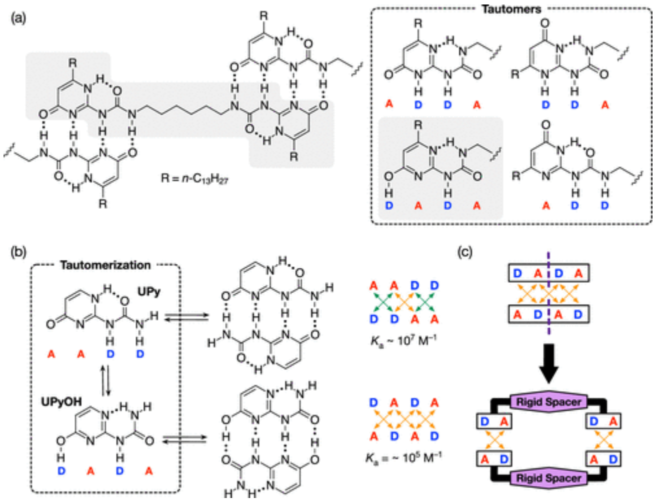
Article #1: A Molecular Replication Process Drives Supramolecular Polymerization

Feng, Y.; Philp, D. A Molecular Replication Process Drives Supramolecular Polymerization. *Journal of the American Chemical Society* **2021**, *143* (41), 17029–17039. https://doi.org/10.1021/jacs.1c06404.

Figures Mentioned: Figure 1



Part (a): Shows how a monomer (based on a molecule called UPy) forms strong bonds with other monomers through four hydrogen bonds arranged in a specific pattern (AADD). This pattern allows the monomers to stick together efficiently. Other versions of UPy exist, but only the AADD pattern is good at forming these stable connections.

Part (b): Explains how UPy can change into a slightly different form (UPyOH), which doesn't bind as strongly because the pattern of its hydrogen bonds (DADA) creates weaker interactions.

Part (c): Proposes a way to improve the weak DADA arrangement. The weaker interactions are eliminated by splitting it into two separate units (DA) and connecting them with a rigid spacer, and the binding becomes much stronger. This new design is simpler to make and can help create better materials.

Synopsis

The article "A Molecular Replication Process Drives Supramolecular Polymerization" by Yuanning Feng and Douglas Philp, published in the Journal of the American Chemical Society, introduces an innovative approach to supramolecular polymerization by integrating molecular replication processes. Non-covalent interactions distinguish supramolecular polymers between monomers and confer unique properties such as self-healing and responsiveness to environmental stimuli.

The authors designed a bifunctional monomer composed of two self-replicating templates connected by a rigid spacer. This design enables the monomer to undergo self-assembly into a polymer through a process driven by molecular replication. The self-replication mechanism is facilitated by the high binding affinity between the self-complementary replicating templates, promoting efficient polymerization in solution.

The key figure in the article, Figure 1, illustrates the molecular structure of the bifunctional monomer and the self-assembly process leading to supramolecular polymerization. The figure depicts the two replicating templates (represented as complementary shapes) connected by a rigid spacer. The self-assembly process is shown as the monomers align and interact through non-covalent bonds, forming a polymer chain. This visual representation underscores the central concept of the study: the coupling of molecular replication with supramolecular polymerization to create advanced materials with self-repairing capabilities.

The study employed various analytical techniques to characterize the polymerization process and the resulting materials. Diffusion-ordered NMR spectroscopy (DOSY NMR) was used to investigate the assembly of the polymer in solution, confirming the formation of high-molecular-weight species. Additionally, microcrystalline and thin films of the polymeric material were prepared and analyzed using powder X-ray diffraction and scanning electron microscopy, providing insights into the structural organization and morphology of the supramolecular polymers.

This research demonstrates a novel strategy for constructing supramolecular polymers by leveraging molecular replication processes. The findings suggest potential extensions of this method to systems where the rigid spacer between replicating templates is replaced by one carrying additional functions, paving the way for the development of more complex and functional supramolecular materials.

In summary, the article presents a significant advancement in supramolecular chemistry by introducing a molecular replication-driven approach to polymerization. The integration of self-replicating templates into the design of bifunctional monomers offers a promising pathway for the development of self-healing materials with potential applications in various technological domains.