

Hyperbranched, supramolecular polymer adhesives exhibiting high toughness and stiffness

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Abstract

In a supramolecular polymer, monomeric units are assembled via directed, secondary interactions such as hydrogen bonding or electrostatic interactions.[1] The dynamic, reversible nature of these non-covalent bonds renders these materials susceptible to a range of stimuli so that depending on the nature of the bonds, the equilibrium of the assembled state can be tuned by triggers such as temperature or light. While the temporary disassembly of the polymer can, for example, result in improved melt processability, the development of supramolecular materials exhibiting superior mechanical properties remains a challenge and generally limits their industrial applicability. A supramolecular material of high mechanical strength and good melt processability has yet to be developed.

Our group has recently reported a trifunctional monomer unit carrying three quadruple hydrogen-bonding ureido-4-pyrimidinone (UPy) groups that assembles into a disordered glass of high stiffness and promising adhesive properties.[2] However, limited toughness and high brittleness were identified as the limiting material properties. Building on this work, this contribution will discuss the synthesis of a hyperbranched macromonomer displaying UPy end groups and its solid-state assembly into a supramolecular network through UPy-UPy interactions. The hyperbranched macromonomer was synthesized via branching of trifunctional amine-terminated polypropylene oxide (PPO) units with molecular weights of ca. 440 g/mol and 3000 g/mol with hexamethylene diisocyanate and by end capping of the remaining amine groups with UPy and 2-ethylhexyl groups. The one-pot synthesis results in materials exhibiting two coexisting, amorphous domains: a low glass transition (T_g) domain offering high toughness and a high T_g domain providing stiffness. The combination of these characteristics and the low melt viscosity induced through the supramolecular motif afford a promising material of unprecedented mechanical performance. In addition, promising adhesive properties were observed and the temperature-induced bonding and debonding on demand present an interesting opportunity for a broad range of applications.

References

- [1] Yang, L.; Tan, X.; Wang, Z.; Zhang, X., *Chemical Reviews*, **115** (2015) 7196–7239.
[2] Balkenende, D. W. R.; Monnier, C. A.; Fiore, G. L.; Weder, C., *Nat. Commun.*, **7** (2016) 1–9.

Figures

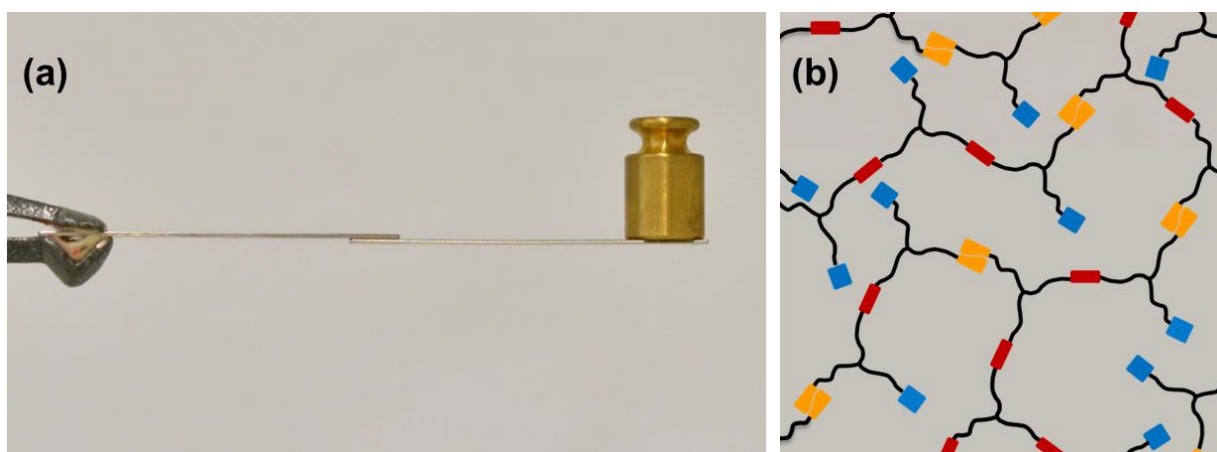


Figure 1. (a) Picture of a stainless steel lap joint bonded with the supramolecular adhesive. (b) Schematic representation of the hyperbranched, supramolecular network.