Hydrogels Based on Dynamic Covalently Linking and Crosslinking

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End-modified PEG and PLA chains: As basic modules for forming dynamic covalent amphiphlic di-, tri- and multi-block copolymers, hydrophilic PEG (A1, A2, and A3) and hydrophobic PLA (B1 and B2) chains with terminal units capable of sequence-specific association that is mediated by both H-bonding and reversible covalent (disulfide) bonding interactions have been prepared.

Dynamic covalent block copolymers are being prepared based on the highly efficient ligation of these polymeric modules. The ligation reaction is mediated by the nearly quantitative crosslinking, I.e., H-bond-mediated disulfide bond formation, between terminal units X and Y that bear complementary H-bonding patterns.

H-bond-mediated disulfide bond formation

Treating modules A with modules B in the presence of I₂ has led to the formation of different block copolymers. We have now prepared a diblock (A1+B1), and a triblock (B1 + A2 +B1) copolymers under mild conditions in very high efficiency. These block copolymers formed very stable polymer micelles with high capacities of loading hydrophobic drugs. Besides, adding free thiols such as GSH and DTT was found to cleave the disulfide bonds, leading to the decomposition of the micelles. We are systematically characterizing block the copolymers, their micelles and solubilizing of various and transport hydrophobic molecules in aqueous media.