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Photoresponsive supramolecular copolymers from diarylethene–perylene bisimide hydrogen bonded complexes

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ABSTRACT

Photoresponsive molecular assembly based on diarylethene (DAE) is an interesting class of supramolecular system as it can allow remarkable morphological transition via small structural changes upon photoisomerization. Herein, we report coassemblies of diarylethene and perylene bisimide through complementary multiple hydrogen-bonding. The open isomer of the diarylethene and the perylene bisimide coassemble to afford photoresponsive helical supramolecular polymer nanofibers. The ring-closure photoreaction of the diarylethene unit enables transformation of helical nanofibers to non-helical fibers. The non-helical fibers can only be obtained through the photoisomerization, and are never achievable by thermal coaggregation of the close isomer and the perylene bisimide. This study illustrates that the proper incorporation of photochromic units into a non-equilibrated molecular assembly system achieved by strong intermolecular interaction enables us to design new self-assembled products in terms of morphology and functions, which are not obtainable through the self-assembly of ground state molecules under either kinetic or thermodynamic control.

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1. Introduction

Control over the structure and property of supramolecular aggregates of organic dyes is an important task for the advancement of their materials applications [1–3]. As supramolecular aggregates are formed by weak noncovalent interactions, they are usually in equilibrium with monomers by slow exchange in solution, which leads to the formation of thermodynamically stable structure. Molecules with strong noncovalent functionality have a propensity to form kinetically trapped aggregates, which may lead to non-equilibrated yet stimuli-responsive supramolecular systems [3–5]. Recently, such non-equilibrated molecular assemblies, especially supramolecular polymers, have attracted significant attention because a small change in their assembly pathways could potentially make diverse structures and properties [6–11]. It can be envisaged that the introduction of stimuli-responsive unit in non-equilibrated system may further diversify aggregate structures yet it has not been studied well.

Light is one of the most attractive stimuli for the manipulation of supramolecular polymers because of its noninvasive, clean as well as remote-controlling properties [12–17]. The non-equilibrated supramolecular assemblies, coupled with reversible photochromic molecules, may offer a variety of intriguing photoresponsive supramolecular assemblies showing morphological transitions, because photochromic molecules can make different energy states by changing their molecular structure/conformation. In this regard, diarylethenes (DAEs) have particularly attracted significant attention among various photo-switching molecules because of their reversible ring-closure/opening reactions upon irradiation with UV/visible-light with high photosensitivity and photofatigue resistance, which is accompanied by large changes in their optical as well as electronic properties [18–22]. However, compared to azobenzene [12–17], DAE has not been well explored for the design of supramolecular polymer exhibiting well-defined photoinduced morphological transition likely due to rather small structural changes upon photoisomerization [23–30].

We previously reported a supramolecular copolymer system composed of melamine-functionalized DAE [31,32] and core-substituted perylene bisimide (PBI) dye by exploiting unsubstituted imide groups of the PBI as multiple hydrogen-bonding sites [33]. The 1:2 complexation of the tetratopic DAE with the ditopic PBI afforded photoresponsive supramolecular copolymer that

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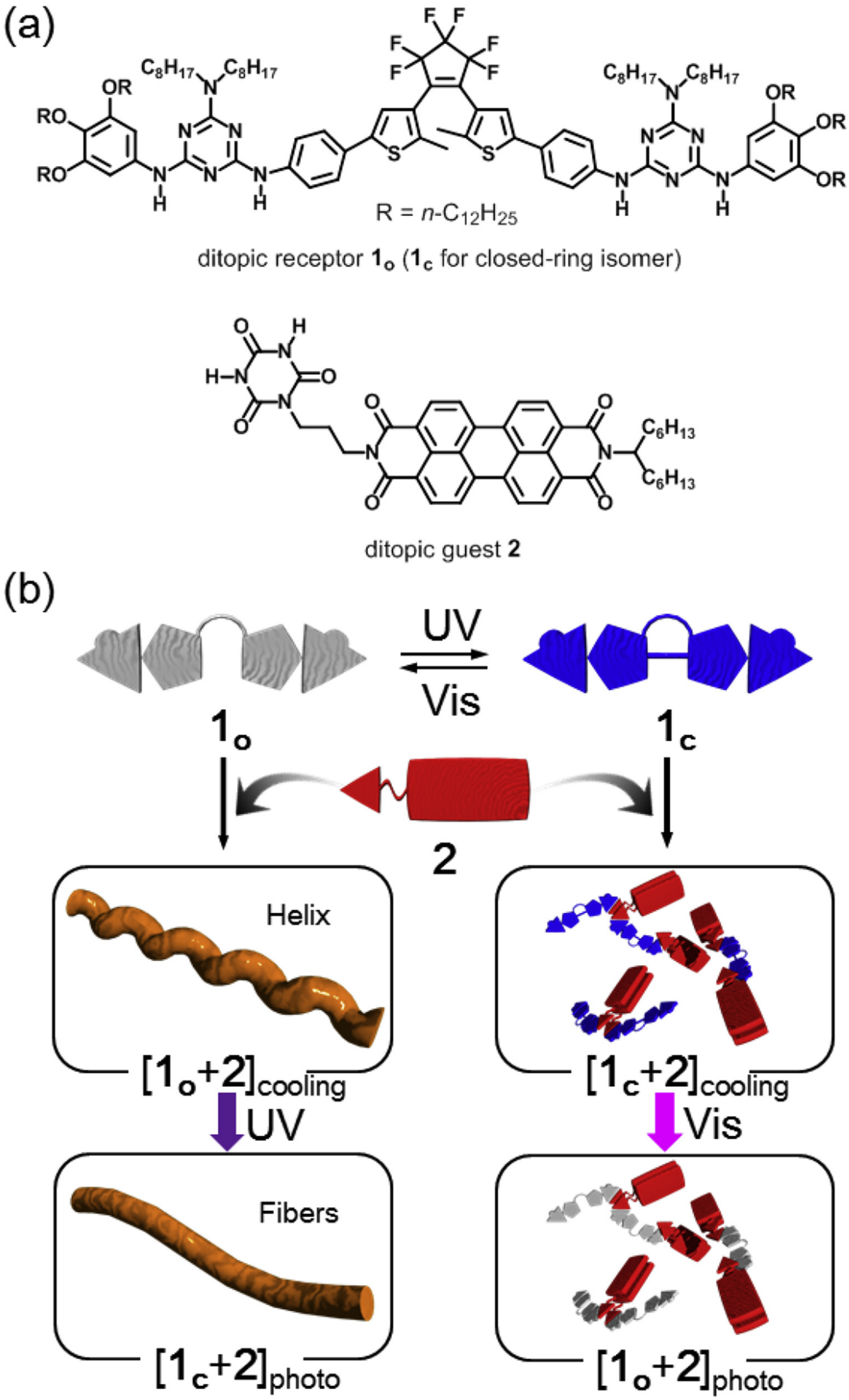

Fig. 1. a) Molecular structures of diarylethene **1** and cyanuric acid (CA) functionalized PBI **2**. b) Schematic presentation for the formation of supramolecular structures between DAE $\mathbf{1_o}$/$\mathbf{1_c}$ ($\mathbf{1_o}$ and $\mathbf{1_c}$ correspond to open and closed form respectively) and PBI **2** by different pathways including both thermal and photo.

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