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Review

Supramolecular polymers fabricated by orthogonal self-assembly based on multiple hydrogen bonding and macrocyclic host–guest interactions☆

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ABSTRACT

Supramolecular polymers constructed by orthogonal self–assembly based on multiple hydrogen bonding and macrocyclic host–guest interactions have received increasing attention due to their elegant structures, outstanding properties, and potential applications. Hydrogen bonding endows these supramolecular polymers with good adaptability and reversibility, while macrocyclic host–guest interactions give them good selectivity and versatile stimuli-responsiveness. Therefore, functional supramolecular polymers fabricated by these two highly specific, noninterfering interactions in an orthogonal way have shown wide applications in the fields of molecular machines, electronics, soft materials, *etc.* In this review, we discuss the recent advances of functional supramolecular polymers fabricated by orthogonal self-assembly based on multiple hydrogen bonding and host–guest interactions. In particular, we focus on crown ether- and pillar[*n*]arene-based supramolecular polymers due to their compatibility with multiple hydrogen bonds in organic solution. The fabrication strategies, interesting properties, and potential applications of these advanced supramolecular materials are mainly concerned.

1. Introduction

Supramolecular polymers (SPs), which combine the advantages of traditional polymer science and supramolecular chemistry, are advanced materials constructed by reversible non-covalent interactions [1–6]. In recent years, SPs have been developed rapidly due to their fascinating properties and wide applications. Compared with SPs constructed only by one type of non-covalent interaction, such as hydrogen bonding, host–guest interactions, metal–ligand coordination, or π–π interactions, functional SPs self-assembled by the combination of multiple non-covalent interactions in an orthogonal way can lead to some excellent properties [7–10]. The fabrication strategy can also be learned from nature. For example, for DNA, multiple non-covalent interactions including hydrogen bonding, π–π interaction, and hydrophobic interaction contribute to its double helix structure and properties [11–13]. In artificial systems, the orthogonal self-assembly means the fact that “different kinds of non-covalent interactions within one supramolecular complex do not interfere with each other” or “two or more non-covalent interactions occur in system with no crosstalk” [14,15]. In 1997, Reinhoudt reported the first discrete nanostructure assembled by hydrogen bonding and metal–ligand coordination in an orthogonal way [16]. Multiple non-covalent interactions endow the complexes with strong ability to undergo reorganization and dynamic exchange processes by means of the reversible assembly procedures.

Among different types of non-covalent interactions employed in the construction of functional SPs, hydrogen bonding and host–

☆ Dedicated to Professor Yu Liu on the occasion of his 65th birthday.

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guest interactions are two main types of supramolecular interactions and have been researched extensively. Hydrogen bonding plays an important role in biological systems and is also a powerful tool to construct dynamic supramolecular architectures. Hydrogen bonding is very attractive for their high directionality and fidelity. Although individual hydrogen bonding is weak, multiple hydrogen bonding array may offer high stability and show much greater binding strength [17]. The most well-known multiple hydrogen bonding ureidopyrimidinone (UPy) motif, first reported by Meijer *et al.*, showed extremely strong dimerization constant ($K_{\text{ass}} > 10^7$ L/mol in $CHCl_3$) *via* quadruple hydrogen bonding [18,19]. In recent years, a large number of functional SPs have been constructed from UPy motif. For example, Xu, Zhang *et al.* developed supramolecular interfacial polymerization by using UPy groups [20]. Tang *et al.* reported an aggregation-induced emission SP from a ditopic UPy monomer [21]. Moreover, Yang *et al.* constructed photores-ponsive SPs and fluorescent nanoparticles based on the UPy motif [22,23].

In addition, macrocyclic host–guest interactions have played a critical role in supramolecular chemistry and developed rapidly due to the diversity of the macrocycles. Recently, Liu *et al.* reviewed water soluble calix[*n*]arenes and its biological applications [24]. Tian *et al.* systematically summarized photoresponsive host–guest functional systems [25]. Wang *et al.* reviewed the applications of cucurbit[*n*]urils in pharmaceutical sciences [26]. T. Xiao, L. Wang *et al.* carefully summarized dynamic hydrogels and artificial light-harvesting systems constructed by host–guest interactions [27,28]. In recent years, macrocyclic host–guest interactions have also been extensively employed to construct SPs, especially by crown ethers or pillar[*n*]arene macrocycles. For example, Huang *et al.* summarized cryptand-based host–guest system and crown ether-based SPs [29,30]. Li, Tian *et al.* reviewed SPs fabricated from pillar[*n*]arenes [31]. Yin *et al.* fabricated a series of fluorescent SPs driven by orthogonal self-assembly based on host–guest interactions and metal–ligand coordination [32–37]. In short, host–guest interactions endow SPs with inherent adaptability, leading their responsiveness to multiple external stimuli and therefore gaining more interesting functions [38,39].

In this review, we aim to describe the recent developments in functional SPs driven by orthogonal multiple hydrogen bonding and host–guest interactions, with an emphasis on those having applications in the fields of dynamic materials, molecular machines, and soft matter. Herein, we describe the SPs depending on the types of the macrocycles that were employed. Therefore, the content of this review is organized as follows: (a) SPs constructed by multiple hydrogen bonding and crown ether-based host–guest interactions, (b) SPs constructed by multiple hydrogen bonding and pillararene-based host–guest interactions, (c) SPs constructed by multiple non-covalent interactions more than hydrogen bonding and host–guest recognition. We will discuss those SPs constructed from orthogonal self-assembly in detail, covering their fabrication, properties, and potential applications. Finally, current challenges and future perspectives for SPs constructed by orthogonal self-assembly are also illustrated.

2. SPs constructed by multiple hydrogen bonding and crown ether-based host–guest interactions

In 2002, Tokunaga *et al.* reported a pseudorotaxane dimer driven by crown ether-based host–guest interactions and quadruple hydrogen bonding [40]. Next year, Li *et al.* constructed well-defined supramolecular heterodimers with unique pseudo[2]rotaxane structures which were also stabilized by the host–guest interactions and quadruple hydrogen bonding [41]. Following the above studies, Wang *et al.* have also employed these two supramolecular orthogonal interactions to construct various supramolecular complexes that possessed dynamic behaviors with responses to external stimuli. The electron deficient macrocycles, such as cyclobis(paraquat-*p*-phenylene)cyclophane (known as “blue box” or $CBPQT^{4+}$), have attracted much attention due to their interesting properties [42,43]. Wang *et al.* prepared a novel “blue box”-based dynamic [2]catenane, in which the $CBPQT^{4+}$ was interlocked by the quadruple hydrogen bonding UPy motif [44]. In this work, a ditopic UPy derivative involving 1,5-dioxynaphthalene (DNP) unit was firstly synthesized, which could undergo a concentration-dependent ring-opening polymerization to form linear SPs. However, in the presence of $CBPQT^{4+}$, a dynamic donor–acceptor [2]catenane interlocked by dimerized UPy could be obtained selectively. Interestingly, the dynamic properties of the [2]catenane were further characterized *via* a ring-exchange experiment by employing a similar [2]catenane which was formed by $CBPQT^{4+}$ and a 1,4-dioxyphenylene bridged ditopic UPy compound. In the ring-exchange experiment, the color of the solution changed gradually from magenta to purple, due to the different strengths of host–guest interactions. This is a new type of dynamic catenane interlocked by hydrogen bonding and it represents a new kind of functional supramolecular complex constructed by quadruple hydrogen bonding and host–guest interactions *via* orthogonal self-assembly.

In 2011, Wang *et al.* first constructed a polymeric supramolecular complex based on two small-molecular-weight heteroditopic monomers by using UPy-based quadruple hydrogen bonding and crown ether-based host–guest recognition [45]. Dynamic light scattering (DLS) experiments showed that the equimolar solution of two monomers (40 mmol/L for each monomer) had a hydrodynamic radius of 31 nm in a mixed solvent of chloroform and acetonitrile (1/1, v/v). When the concentration increased to 433 mmol/L, polymerization degree of 38.5 was determined according to ^1H NMR by using the Carothers equation, which corresponds to a molecular weight of 65.9 kDa. The polymeric supramolecular complex constructed by orthogonal self-assembly was further confirmed by viscosity measurements, scanning electronic microscopy (SEM) and concentration- dependent ^1H NMR experiments. In a follow-up study, they further fabricated a novel kind of crown ether–paraquat supramolecular polypseudorotaxanes with the hydrogen bonded SP as the main chain (Fig. 1) [46]. This polypseudorotaxane network was based on two homoditopic monomers: One is the bifunctional UPys with a bis(*p*-phenylene)-34-crown-10 (BPP34C10) located at the center of the monomer (**1**) and the other is the ditopic paraquat molecule (**2**). Monomer **1** can self-assemble into linear SPs *via* quadruple hydrogen bonding. Upon addition of monomer **2**, the paraquat group can thread into the crown ether cavity to achieve polypseudorotaxane networks. It is worth noting that the molar ratios of the bisparaquat are critical for the solution viscosity due to the different cross-linking degrees. The DOSY NMR also demonstrated that larger aggregates were formed after the addition of 0.5 equiv. of **2**.

To develop responsive SPs, Wang *et al.* further explored the construction of metal-ion-responsive dynamic SPs driven by orthogonal quadruple hydrogen bonding and benzo-21-crown-7 (B21C7)-based host–guest interactions [47]. They first synthesized two types of monomers: the homoditopic building block **3**, which bears two dialkylammonium units, and the heteroditopic building block **4**, which is comprised of one B21C7 motif and one UPy group (Fig. 2). As we can see, a supramolecular dimer from **4** was initially obtained, followed by polymerization upon addition of **3**. The disassembly/assembly of this SP could be reversibly switched by addition/removal of K^+ without interfering with UPy dimerization due to the stronger binding of K^+ with B21C7 moiety than dialkylammonium moiety.

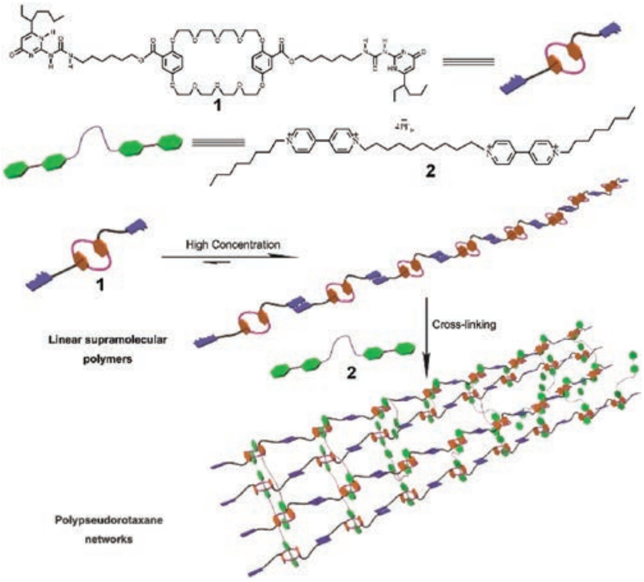

Fig. 1. Graphical representation of the construction of polypseudorotaxane networks from monomers **1** and **2**. Reproduced with permission [46]. Copyright 2011, the Royal Society of Chemistry.

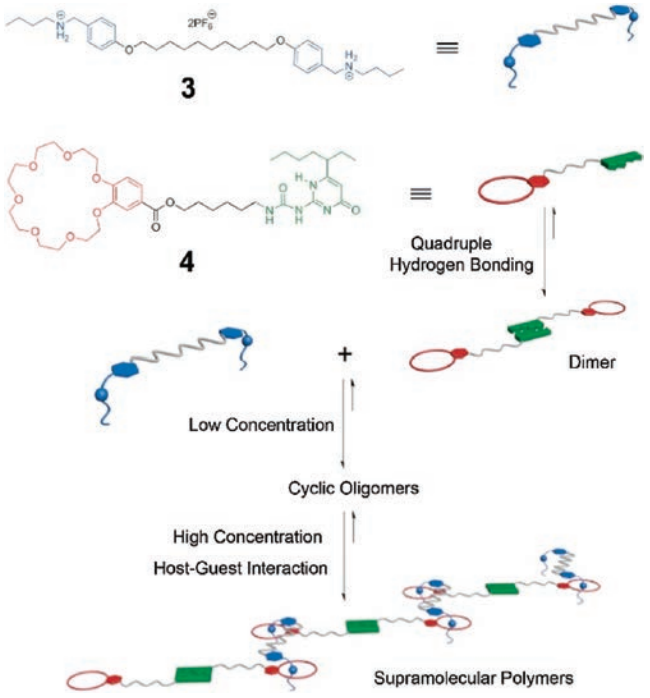

Fig. 2. Graphical representation of the SPs constructed from monomers **3** and **4** by orthogonal self-assembly. Reproduced with permission [47]. Copyright 2013, the Royal Society of Chemistry.

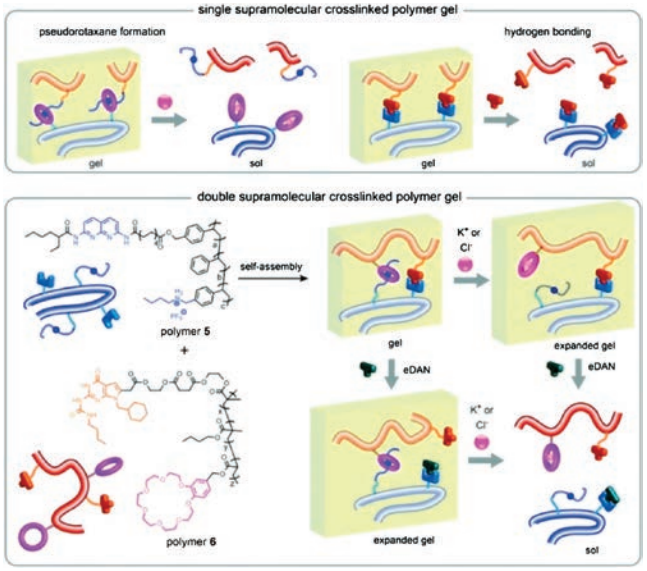

Fig. 3. Cartoon representations of polymers **5** and **6** and the illustration of the advantage of doubly *vs.* singly supramolecular crosslinked polymer gels. Reproduced with permission [48]. Copyright 2015, the Royal Society of Chemistry.

In 2015, Zimmerman, Huang *et al.* reported a double supramolecular crosslinked polymer gel which showed macroscale expansion and contraction behavior and multistimuli responsiveness [48]. As we know, most of the supramolecular polymeric gels contain only one type of non-covalent interaction. In this example, they constructed a novel supramolecular polymeric gel possessing two types of supramolecular crosslinks. Two types of non-covalent interactions are orthogonal: diamido-1,8-naphthyridine (DAN)-based quadruple hydrogen bonding and B21C7-based host–guest interactions. As shown from Fig. 3, polymer **5** is polystyrene with pendant DAN groups and dialkylammonium salt units, and polymer **6** is poly(butyl methacrylate) bearing deazaguanosine (DeUG) groups and B21C7 units. Notably, DAN can bind DeUG to form stable quadruple hydrogen bonding complexes, in which the hydrogen bonding interaction is highly orthogonal to B21C7-based host–guest interactions. Thus polymers **5** and **6** could be crosslinked by these two orthogonal non-covalent interactions, resulting in the formation of a supramolecular polymeric gel. This double crosslinked gel could undergo macroscale expansion and contraction behavior *via* the introduction of competitive compounds or ions, such as the DAN monomer (eDAN), K$^+$ or Cl$^-$. This example demonstrated that the macroscopic property of SPs can be induced by controlling the orthogonal self-assembly on a molecular scale.

In 2016, Qu *et al.* reported a quadruple hydrogen-bonded SP with a [c2]daisy chain structure in its backbone [49]. This SP was constructed by phototriggered supramolecular polymerization of the [c2]daisy chain monomer (Fig. 4). The monomer **7** is a daisy chain rotaxane containing two coumarin protected UPy terminals at two ends. The supramolecular polymerization was driven by light irradiation that can remove the coumarin substituents and yield **8** with two terminal UPy groups. Then, linear SPs can be self-assembled by **8** through quadruple hydrogen bonding. This strategy of phototriggered supramolecular polymerization represents a good supplement for the fabrication of stimuli-responsive SPs.

Molecular machines have attracted much research interest in recent years due to their ability to produce mechanical work at the nanoscale and may create new functions by energy transduction. When running in an integrated system, this kind of molecular machines can amplify their motion to a macro scale. In 2012, Giuseppone *et al.* synthesized muscle-like SPs by using bistable [c2]daisy chain and terpyridine-based metal–ligand coordination [50]. In the next work, they constructed an acid–base switchable [c2]daisy chain rotaxane terminated with two 2,6-diacetylamino pyridine units, which could self-assemble with a bis(uracil) linker through complementary hydrogen bonding [51]. As a result, unidimensional SPs associated in bundles of muscle-like fibers could be obtained. In 2017, they further reported the integration of bistable [c2]daisy

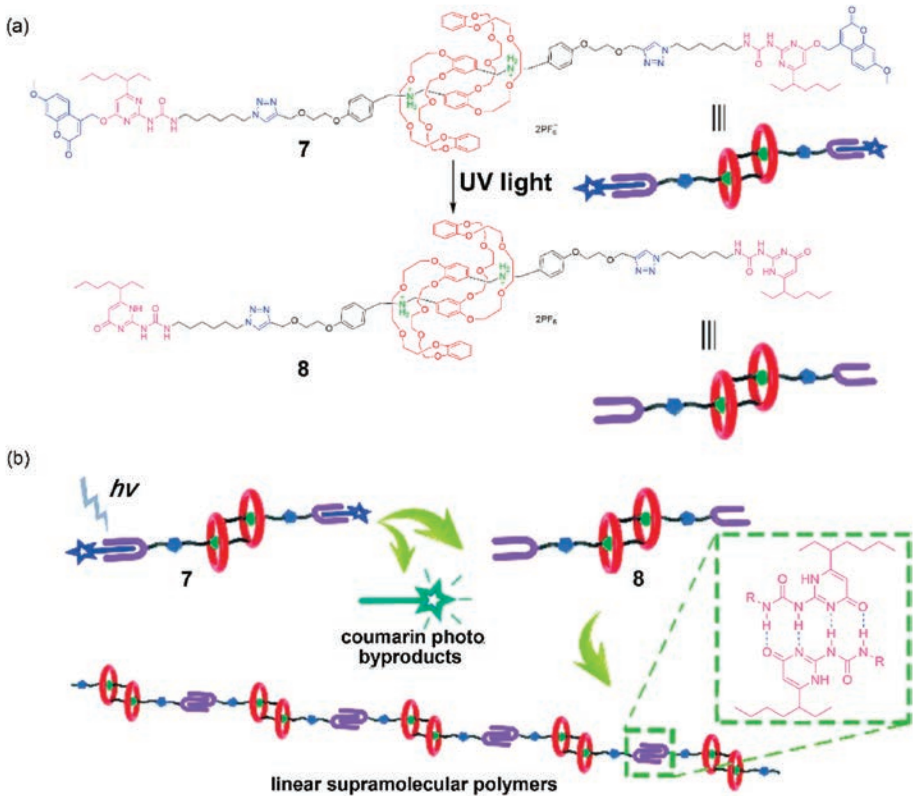

Fig. 4. (a) Chemical structures of **7** and **8**; (b) Graphical representation for the phototriggered supramolecular polymerization of the [c2]daisy chain rotaxane **7**. Reproduced with permission [49]. Copyright 2016, the Royal Society of Chemistry.

chain rotaxanes by employing UPy-based quadruple hydrogen bonding SP [52]. The monomer exhibits a chemical structure similar to the previous example reported by Qu *et al.* However, the big difference in this case is that the [c2]daisy chain is a bistable rotaxane. Therefore, the monomer can form "extended" and "contracted" SPs by controlling the pH of the system. In toluene, the "extended" SP forms a gel while the "contracted" one remains as a solution. The sol–gel transition could be changed *in situ* by adjusting the bistable [c2]daisy chains. Therefore, the macroscopic physical state of the material could be controlled by changing the mechanical bond at nanoscale. This example demonstrates the possibility of integrating molecular machines into novel smart materials.

In recent years, Dong *et al.* prepared many novel supramolecular polymers based on host–guest interactions [53–58]. Very recently, they constructed a cross-linked SP network based on the B21C7/secondary ammonium salt host–guest recognition and 1,3,5-benzenetricarboxamide (BTA) hydrogen bonding motif [59]. The BTA motif is a well-known building block for supramolecular polymerization, which could be stabilized by threefold H-bonding [60]. They firstly designed and synthesized the BTA derivative with three B21C7 units (**9**) (Fig. 5). Monomer **9** can self-assemble into one-dimensional linear SPs through the aggregation of BTA cores *via* hydrogen bonding. Then, cross-linked SPs could be formed by adding guest molecule **11**, which possesses two secondary ammonium salts and plays the role of a linker to connect the linear polymers formed by **9** together. The formation of the cross-linked polymers was evidenced by ^{1}H NMR, DOSY, and viscosity measurements. Interestingly, soft and long viscous fibers could be pulled out from a concentrated solution of the cross-linked polymers. Notably, the cross-linked supramolecular (**9** at 160 mmol/L and **11** at 240 mmol/L) existed in the gel-like state below 45 °C and converted into a viscous solution above 50 °C. The SPs constructed in this work provide new understanding for supramolecular polymerization.

3. SPs constructed by multiple hydrogen bonding and pillar[*n*]arene-based host–guest interactions

Pillar[*n*]arenes are a relatively new type of macrocyclic host in supramolecular chemistry and have displayed versatile host–guest behavior with various kinds of guests [61–66]. The synthetic route of pillar[*n*]arenes is easy and pillar[*n*]arenes have unique shape, versatile solubility, and functionality. The symmetrical pillar architecture and easy derivatization endow pillar[*n*]arenes with a good ability to bind different kinds of guests, such as cationic guests, anionic guests, neutral guests, flexible guests, and rigid guests. In recent years, pillar[*n*]arenes have been employed to construct various kinds of functional supramolecular materials, such as gels [67–77], hydrogels [78–83], mechanically interlocked systems [84–86], supramolecular vesicles/nanocarriers [87–113], functional nanoparticles [114,115], sensors [116–120], crystal materials [121–130], stimuli-responsive materials [131–138], and SPs [139–148]. In this section, we focus on SPs constructed orthogonally by hydrogen bonding and pillar[*n*]arene-based host–guest interactions.

In recent years, Wang *et al.* fabricated a series of SPs orthogonally by employing pillar[*n*]arene-based host–guest interactions and UPy-based quadruple hydrogen bonding. For example, they synthesized a UPy functionalized pillar[5]arene (**12**) and a bisparaquat derivative (**13**), which can orthogonally self-assemble into polymeric architectures (Fig. 6) [149]. The binding constant was calculated to be 4.46×10^3 L/mol *via* fluorescent titration by using a model compound. The complexation of **12** with **13** was

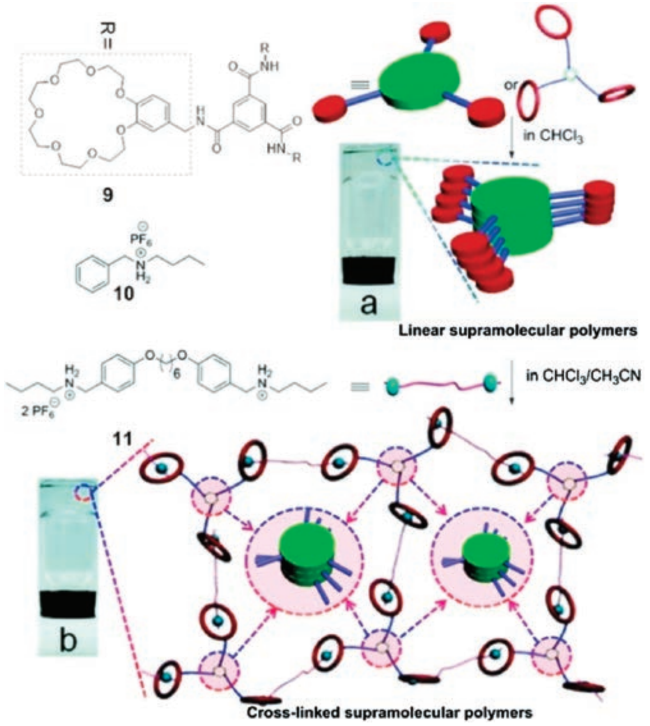

Fig. 5. Cartoon representation for the construction of one dimensional SPs and cross-linked SPs. Reproduced with permission [59]. Copyright 2018, the Royal Society of Chemistry.

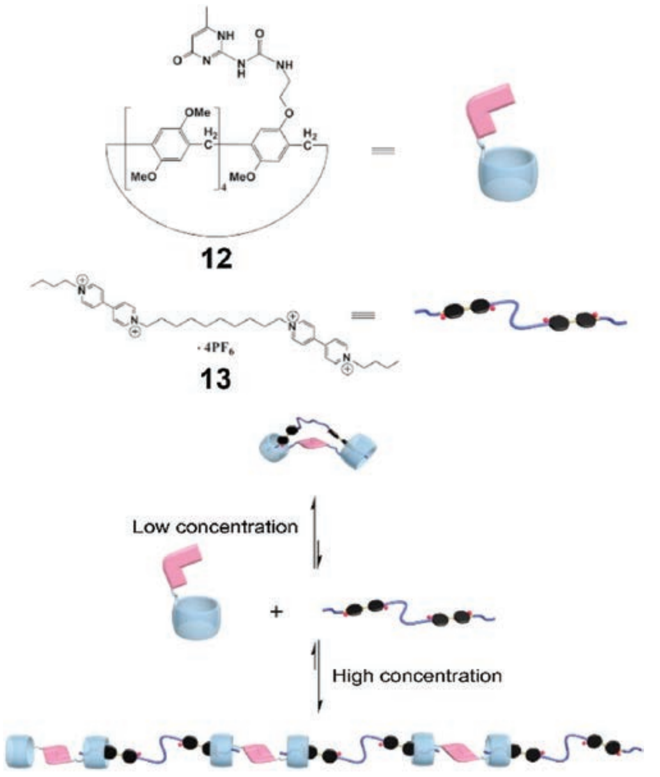

Fig. 6. Illustration of the formation of the SP from **12** and **13**. Reproduced with permission [149]. Copyright 2012, the Royal Society of Chemistry.

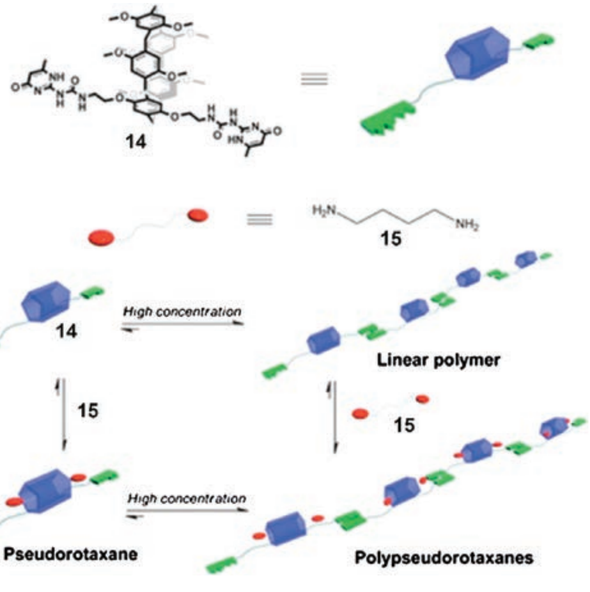

Fig. 7. Graphical representation of the construction of polypseudorotaxane polymers from **14** and **15**. Reproduced with permission [150]. Copyright 2012, the Royal Society of Chemistry.

confirmed by ^{1}H NMR titration and electrospray ionization mass spectrometry (ESI-MS). The formation of SPs at high concentration driven by two orthogonal interactions, quadruple hydrogen bonding and host–guest interactions, was proven by concentration-dependent ^{1}H NMR, 2D NOESY NMR, DOSY, viscosity measurements, and cyclic voltammetry. This study opened a new window to construct orthogonal SPs by employing pillar[*n*] arenes.

As we can see from the above examples, the combination of different supramolecular interactions is a very easy and efficient way for the construction of complicated supramolecular structures. In another examples, Wang *et al.* fabricated a pillar[5]arene-based supramolecular polypseudorotaxane [150]. In this study, they first synthesized a ditopic UPy-functionalized pillar[5]arene **14** (Fig. 7). Monomer **14** can form linear SPs at high concentration. After that, when the guest molecule 1,4-butanediamine (**15**) was added, a polypseudorotaxane could be obtained in the concentrated solution. This study affords the first example of a linear supramolecular polypseudorotaxane driven by efficient quadruple hydrogen bonding interactions and pillar[*n*]arene based host–guest recognition, and it also enriches the applications of the host–guest chemistry of pillar[*n*]arene. In the follow-up study, they further constructed polypseudorotaxane-based polymer networks by using **14** and a bisparaquat derivative, in which the backbones were constructed from quadruple hydrogen bonding [151].

Immediately after the above work, they further constructed polyrotaxanes by employing the method of "diamine threading followed by end-capping with UPy units" to access the solvent-responsive SPs [152]. The obtained dynamic polyrotaxanes were fabricated from pillar[5]arene derivatives **16** and diamines **17** or **17′** (Fig. 8). The polyrotaxanes were interlocked by orthogonal quadruple hydrogen bonding and host–guest interactions. The UPy groups played a dual role, acting not only as an end-capping stopper but also as a hydrogen bonded linking group to regulate the conformation and dynamic properties of the supramolecular polyrotaxanes. Notably, the reversibility of main-chain backbones and the sliding process of the axle could be achieved by changing the polarity of the solvent.

Very recently, a dual-responsive hyperbranched SP was constructed by Zhang, Guo *et al.* [153]. This hyperbranched SP was driven by cooperative hydrogen bonding interaction and host–guest recognition (Fig. 9). They first prepared a homotritopic

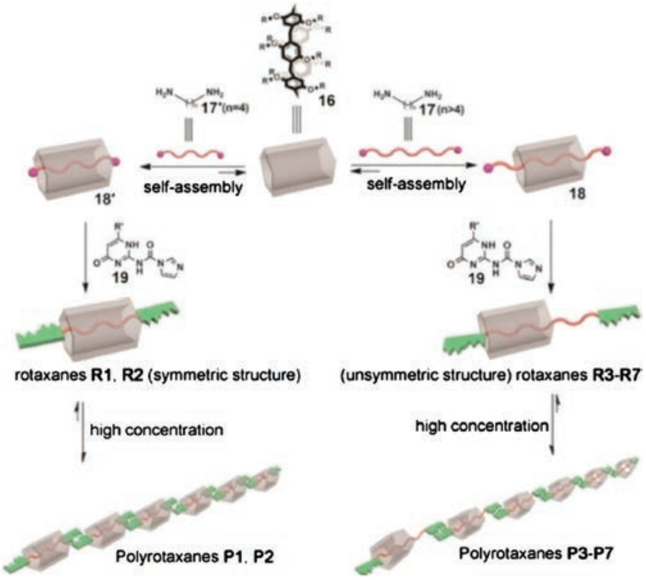

Fig. 8. Graphical representation of the construction of dynamic polyrotaxanes. Reproduced with permission [152]. Copyright 2012, American Chemical Society.

pillar[5]arene containing adenine groups (**21**). Then they synthesized a uracil derivative (6-(2,4-dioxo-3,4-dihydropyrimidin-1(2*H*)-yl)hexanenitrile, **22**). ^{1}H NMR suggested that **22** could thread into the cavity of the model compound **20** to form a [2] pseudorotaxane and the binding stoichiometry was determined to be 1:1. The hyperbranched SP could be formed from **21** and **22** at a low concentration due to the strong cooperative interactions. The supramolecular polymerization was proved by the combination of experiments including ^{1}H NMR, viscosity measurements, and DOSY experiments at various concentrations. This material displayed dual-responsiveness to heating and cooling, or the addition of acid/base.

4. SPs constructed by multiple non-covalent interactions involving hydrogen bonding and host–guest recognition

Nature has provided lots of examples which display the ability to construct exquisite and complex biological systems from relatively simple building blocks *via* multiple types of non-covalent interactions. It is also noteworthy that hierarchical self-assembly strategy is an effective way to fabricate well-organized structures. Inspired by these facts, chemists began to adopt multiple orthogonal non-covalent interactions to fabricate functional SPs hierarchically. In an integrated system, multiple orthogonal non-covalent interactions will endow the SPs with more responsivenesses upon external stimuli and improve the dynamic properties of the material. For example, Li *et al.* first constructed a three-component dynamic [2]catenane by employing hydrogen bonding, metal–ligand coordination, and host–guest interaction in 2007 [154]. In this section, we will give some typical examples to illustrate the diversity of the construction of SPs by employing multiple orthogonal non-covalent interactions involving multiple hydrogen bonding and host–guest interactions.

Besides the traditional covalent polymerization method, mechanically connected polyrotaxanes with unique topological structures can also be realized *via* the non-covalent methods. In recent years, Wang *et al.* constructed a series of supramolecular materials with various topological structures [155–160]. In 2015, they modified the [2]rotaxane backbone with two orthogonal recognition motifs and successfully realized the poly[2]rotaxanes by mechanical linkage through the hierarchical self-assembly method [161]. The authors firstly synthesized the [2]rotaxane **24**, which was based on the B21C7/secondary ammonium salt recognition unit (Fig. 10). On one hand, terpyridine group was connected on the axle of the [2]rotxane, which could be coordinated with Zn^{2+} to form dimeric [3]rotaxane **26**. On the other hand, a Hamilton receptor was connected to the ring of the [2]rotaxane, which can drive the [2]rotaxane to form hydrogen bonded complex **25** with cyanuric acid functionalized polycaprolactone (PCL) **23**. Finally, when mixing all of three components together, the poly[2]rotaxanes **27** could be successfully obtained. Notably, self-standing films could be fabricated by solution casting of 1:2:1 mixture of monomers **23**, **24** and $Zn(OTf)_2$ in concentrated solution. This novel type of mechanically linked polyrotaxanes stand for a special SP driven by orthogonal self-assembly based on multiple non-covalent interactions.

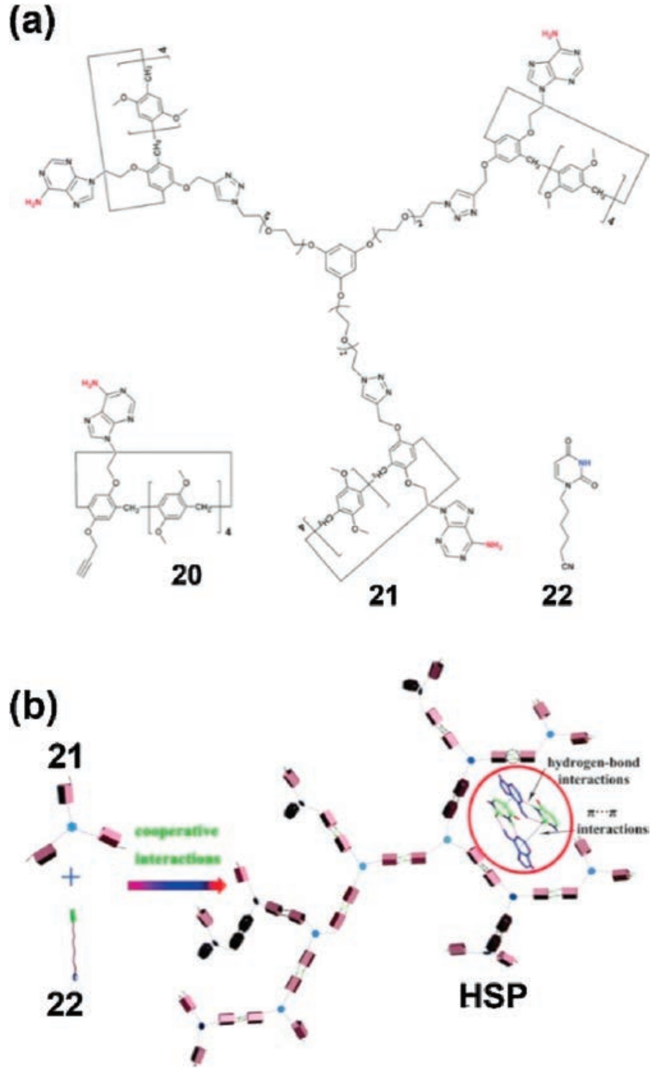

Fig. 9. (a) Chemical structures of **20**, **21** and **22**. (b) The illustration of the construction of hyperbranched SP **HSP** from **21** and **22** through cooperative host–guest recognition and hydrogen-bond interactions. Reproduced with permission [153]. Copyright 2018, the Royal Society of Chemistry.

As we can see from the above example, the employment of multiple orthogonal interactions to construct supramolecular complex provides advantages over traditional methods that use only one kind of non-covalent bond. In next example, we introduce a special SP constructed by triply orthogonal non-covalent interactions, which was reported by Yan, Stang and co-workers [162]. As shown from Fig. 11, supramolecular polymeric network

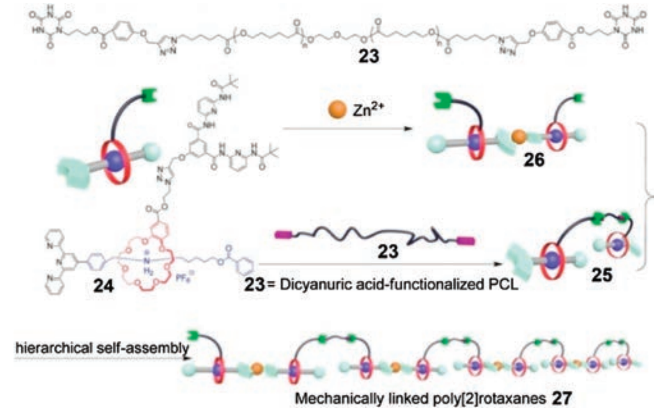

Fig. 10. Schematic representation for hierarchical construction of mechanically linked poly[2]rotaxanes **27**. Reproduced with permission [161]. Copyright 2015, American Chemical Society.

28 was fabricated from a 120° B21C7-tethered acceptor **29** and UPy-decorated 120° donor **30** through metal–ligand bond. The B21C7 moieties within the network offers a platform for further modification *via* host–guest interaction. For example, tetraphenylethylene (TPE)-functionalized dialkylammonium salt **31b** were incorporated into network **28** to get supramolecular networks with aggregation induced emission property. The last step was highly modular and can only occur when the substrate does not interfere with the metal–ligand coordination or quadruple hydrogen bonds. The orthogonal strategy employed in this research based on non-covalent bonds afforded advantages over the covalent method for achieving complicated polymeric structures in a highly modular way.

The π–π stacking interaction is also prevalent in biological systems, such as the stacking of the base pairs in DNA. In recent years, an increasing number of supramolecular complexes have been constructed by employing π–π stacking interaction and other non-covalent interactions at the same time. For example, Wang *et al.* prepared a highly stable supramolecular cyclic monomer from a ditopic UPy molecule, which was stabilized by the π–π interaction between the aromatic group in the molecule and the dimeric UPy plane [163,164]. In 2017, Q. Wang *et al.* prepared a UPy-based SP network by photo-cross-linking between the coumarin groups [165]. Later, they further constructed a neutral SP by three kinds of non-covalent interactions including UPy-based quadruple hydrogen bonding, pillar[5]arene-based molecular recognition, and π–π donor–acceptor interaction [166]. Firstly, they designed and synthesized an unsymmetrical guest **34** bearing a UPy motif and a carbamate group, which can dimerize through quadruple hydrogen bonding to form a homoditopic AA-type supramonomer (Fig. 12). Secondly, they prepared a pillar[5]arene derivative **35**, which can bind with **36** to generate a 2:1 exo-wall complex *via* π–π interaction, leading to the formation of a homoditopic BB-type supramonomer. Finally, the neutral linear SPs could be successfully fabricated in concentrated solution. Interestingly, the degradation of the SP backbone could be effectively regulated by introducing the competitive adiponitrile guest. In follow-up study, they reported a pillar[5]arene-fused cryptand-based SP, in which three orthogonal supramolecular interactions, pillar[5]arene-based host–guest recognition, cryptand-based host–guest interaction, and quadruple hydrogen bonding were combined together [167]. Moreover, the obtained SPs could be effectively degraded with a change in fluorescence by the addition of a competitive cryptand host.

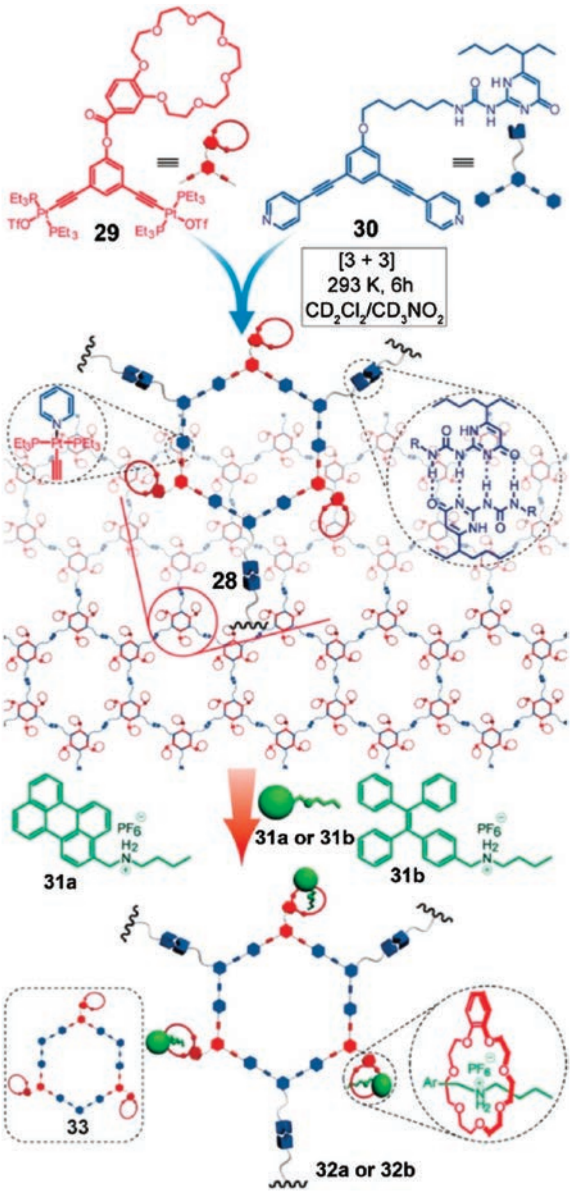

Fig. 11. Cartoon representations of the formation of a supramolecular polymeric network from monomer **29**, **30** and **31** *via* triply orthogonal non-covalent interactions. Reproduced with permission [162]. Copyright 2016, American Chemical Society.

5. Conclusions and perspectives

In conclusion, recent developments in the construction of SPs based on orthogonal host–guest interactions and multiple hydrogen bonding are reviewed. The orthogonal construction strategy is versatile by employing multiple hydrogen bonding and host–guest interactions due to the versatility of macrocyclic hosts, such as crown ethers and pillar[*n*]arenes. In addition, SPs constructed by multiple non-covalent interactions involving hydrogen bonding and host–guest interactions are also concerned. The potential to introduce stimuli-responsive motifs into the architectures and the dynamic property of the supramolecular interactions endow these SPs with multiple stimuli-responsive properties. With these characters, many of these species can be made to be used as mechanically interlocked molecules or molecular machines,

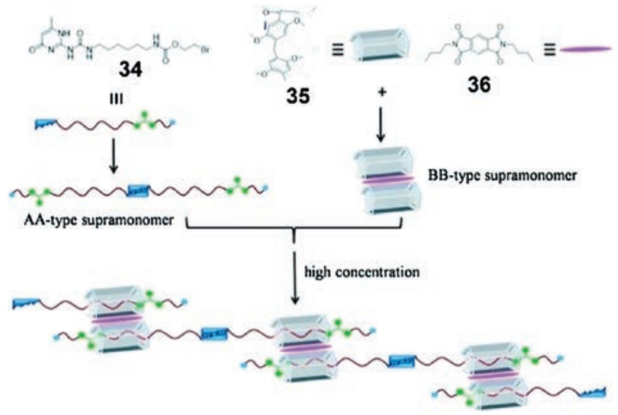

Fig. 12. Graphical representation of SPs constructed by three orthogonal interactions. Reproduced with permission [166]. Copyright 2017, the Royal Society of Chemistry.

adaptive SPs, functional gels, nanofiber and self-standing film materials.

Compared with the complex biological systems, SPs constructed by orthogonal self-assembly are still in the initial stage of research. Therefore, the orthogonal self-assembly is still a powerful tool for scientists to understand and mimic the biological systems and also a necessary way to prepare dynamic materials. The current research in this field is still immature and still requires continued exploration. Before getting more practical applications, we are still facing some basic challenges. Herein, we prefer to describe some future directions that may have significant effects in this field. Firstly, the mechanism of orthogonal self-assembly process needs further in-depth study to gain clear insight into the relationship between building blocks and macroscopic functions. Secondly, more water-soluble supramolecular complexes fabricated by orthogonal self-assembly need to be developed. Particularly, the construction of biocompatible supramolecular complexes, which can be incorporated into biological systems, can provide a versatile platform to understand, study, and replicate biological processes. Thirdly, how to promote dynamic SPs to real functional materials is another challenge. For example, more attention needs to be placed on the synthetic simplicity, economic suitability, mechanical strength, and environmental friendliness of such materials. Nevertheless, we believe that the examples described in this review will motivate scientists to further develop multifunctional SPs, which will enrich nanoscience, materials science, and biological technology.

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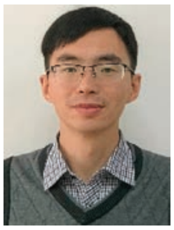
Biography of the first author



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