

Self-healing materials based on host-guest interaction

Self-healing materials are able to repair the cracks or other damage occurring upon usage without human intervention. Such microcracks especially those deep within the structure can fatally deteriorate the function reducing the lifetime. The development of materials able to repair themselves is a relatively new field of intelligent materials.

One main type of self-healing polymers are intrinsic self-healing ones able to heal cracks by themselves upon heating or thermo-mechanical effect. For instance, the friction generates heat to induce local melting and ability to repair the damage via chain interdiffusion and entanglements. Another possibility is thermally reversible crosslinking without additional catalysts or monomers. In the other main type of such materials the healing agents (catalysts, monomers) are *a priori* encapsulated or embedded in fragile microcapsules or capillaries [1]. In this review we try to summarize the self-healing materials based on cyclodextrins.

Controlled release of corrosion inhibitor

The corrosion inhibitor can be encapsulated by CDs and incorporated in the coating this way ensuring the slow release of the inhibitor and self-healing of the corrosion defects. For instance, mercaptobenzothiazole and mercaptobenzimidazole (MBT and MBI) complexed by BCD in the coating provided long-term protection for high strength aluminum alloys against atmospheric corrosion [2]. No corrosion products can be seen on the scratched sample doped with inhibitor/CD complex while the sample with the non-complexed inhibitor was covered by white corrosion products above the scratch. The encapsulation of the corrosion inhibitor with CDs results in bulkier complex with lower diffusion within the coating, slower leaching and longer lasting effect [3,4].

Intelligent anticorrosion coating was developed by embedding aniline/ACD complex and bisammonium/cucurbit[6]uril (C6B) complex into hollow mesoporous silica nanoparticles as acid- and alkaline responsive elements of the pH responsive intelligent coating [5]. The release of the corrosion inhibitor (benzotriazol) was controlled by the encapsulated pH-responsive compounds. The corrosion inhibitor was released only upon the change of pH in the corroded area.

Poly(o-phenylenediamine) (PoPD) nanotubes were fabricated through chemical oxidative polymerization of o-phenylenediamine in cetyl trimethyl ammonium bromide (CTAB)

microemulsion polymerization using BCD and iron (III) chloride (FeCl_3). Improved resistance against corrosion of mild steel by epoxy coating containing synthesized PoPD nanotubes was observed [6].

Self-healing materials based on host-guest interaction

The molecular recognition of polymers by CDs is utilized. The polyrotaxanes comprising CDs threaded on the main chain of a polymer have no such stimuli responsive properties. Among the supramolecular polymers the side chain complexes are suitable for the preparation of self-healing materials [7,8]. The polymers modified with CD (host polymers) form gels with polymers modified with guests (guest polymers) in water via self-assembly (Figure 1). The host polymer can be a cross-linked CD polymer, too [9]. These gels show self-healing properties. The stimuli responsiveness depends on the properties of the guest moieties.

Redox-responsive self-healing materials

Redox stimuli of poly(acrylic acid) (PAA) possessing BCD as host polymer and PAA having ferrocene moieties as guest polymer induce sol-gel transition and readhesion of the cut surfaces [10]. Treating the gel with an oxidizing agent ferrocenium cation is formed, which does not form complex with BCD and the gel is converted to sol. The gel is restored by treating the sol with a reducing agent.

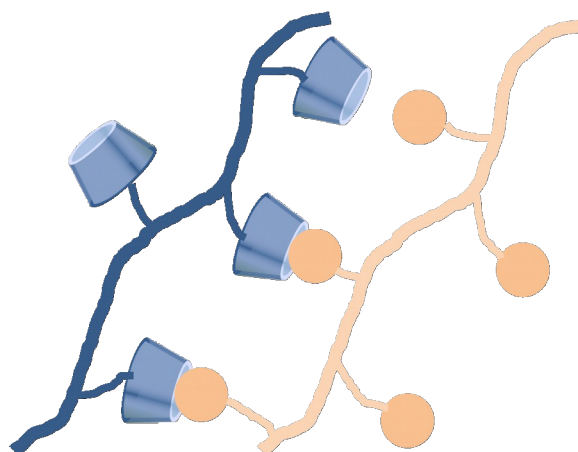


Figure 1 Self-assembly of host polymers and guest polymers on the effect of external stimuli

The polymers can be prepared also by self-assembly of host dimers and guest dimers. Ferrocene- and cyclodextrin-terminated monomers form water-soluble AA-BB-type supramolecular polymers on the basis of inclusion complex formation of ferrocene with BCD resulting in one-dimensional supramolecular nanofibers (Figure 2). Changing the redox potential the nanofibers fall apart or assemble reversibly providing unique self-degrading and -healing properties [11].

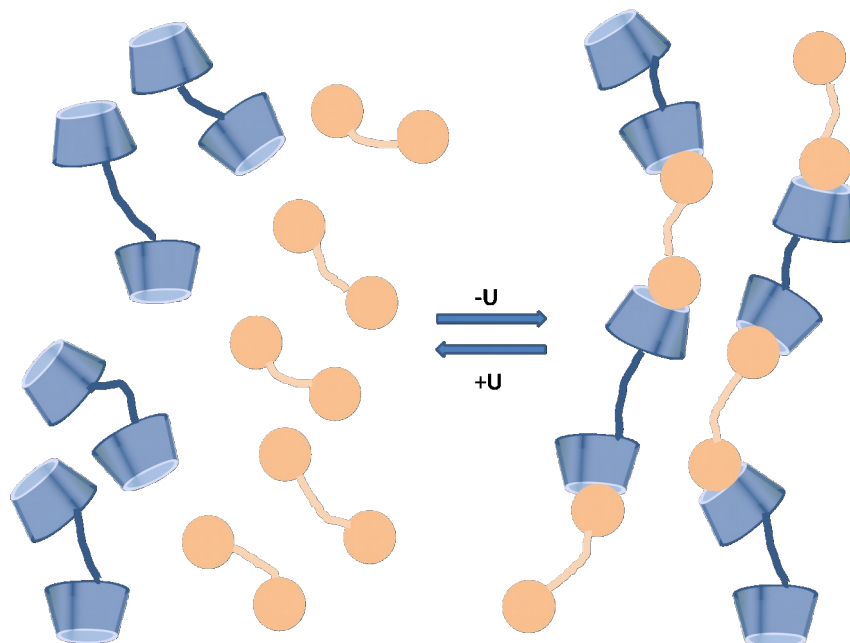


Figure 2 Supramolecular polymers formed by interaction of dimer hosts and dimer guests with redox stimuli responsiveness

A mixture of the above two systems is shown in Figure 3. A multifunctional ferrocene-modified poly(glycidyl methacrylate) (PGMA-Fc) and a BCD dimer was prepared for the construction of electrically driven removable and self-healing polymeric materials based on the complexation reaction between ferrocene and BCD groups. The self-healing performance could be enhanced with wetting the sample to increase the electrical conductivity. The material is a promising self-healing agent for commercial painting products [12].

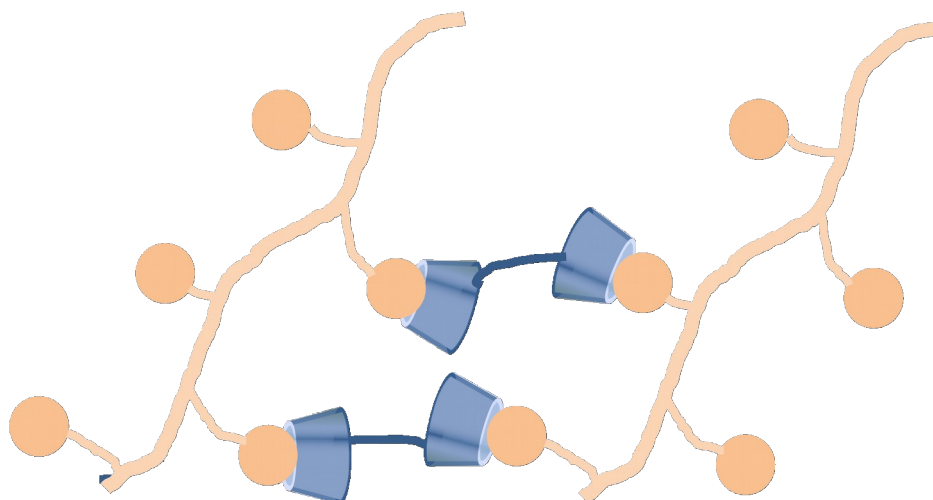


Figure 3 Self-assembly of host dimer and guest polymers

Light responsive self-healing materials

When azobenzene was used as a guest, the gel showed sol-gel transition by photoirradiation. While ACD forms complex selectively with *trans*-azobenzene, BCD forms complex selectively with the *cis* isomer. Upon irradiation with UV light *trans*-azobenzene is transformed into the *cis*

isomer, which is readily transformed back to the *trans* isomer upon irradiation with visible light. The viscosity of a hydrogel consisting of poly(sodium acrylate) modified with dodecyl side chain was decreased upon addition of ACD, while increased again when diazobenzoic acid was added as a competitive guest. With this photoisomerization controlled association and dissociation of the dodecyl chain with ACD was achieved [13].

Reversible adhesion and dissociation of the host gel with CD moieties from the guest gel with azobenzene moieties may be controlled by photoirradiation. The differential affinities of ACD and BCD for the *trans*-azobenzene and *cis*-azobenzene are employed in the construction of a photoswitchable gel assembly system [14].

Self-assembly controlled by pH, temperature or solvent

Materials responsive to various external stimuli such as pH, temperature and solvent can be prepared using the host-guest interaction with CDs. Harada's group has elaborated several examples.

BCD forms complex with dansyl groups at neutral pH but not at lower pH. This phenomenon was utilized for the development of pH responsive gel assembly between CD gels and the gels with dansyl moieties [15].

Poly(acryl amide) (PAAm) gel modified with benzyl moiety forms assemblies with BCD gel at room temperature, with ACD gel at 15 °C and GCD gel at 5 °C providing a tool for the development of temperature controlled systems [16].

The aggregation state of pyrene is different in water and in DMSO. The aggregates present in water fall apart at a certain DMSO concentration in the solution. The aggregate forms complex with GCD, while the monomeric form associates with BCD. Thus, the gel containing pyrene moiety can differentiate between gels modified with ACD, BCD and GCD, selectively, based on the solvent composition [17].

Self-healing materials responsive to magnetic effects

Multiwall carbon nanotubes (MWCNTs) were covalently modified with BCD via grafting with maleic anhydride and then esterified with BCD. Then Fe²⁺ and Fe³⁺ ions were precipitated on the surface of the BCD-functionalized MWCNTs. Composites were made by incorporating Fe₃O₄@BCD-MWCNTs reinforced nanoparticles into poly(caprolactone) matrix. Then nanofibers were fabricated from the composite by electrospinning. It was found that the Fe₃O₄ particles were aligned along the nanofiber axis (Figure 4). The composite nanofibers showed an excellent shape memory effect triggered by an alternating magnetic field [18].

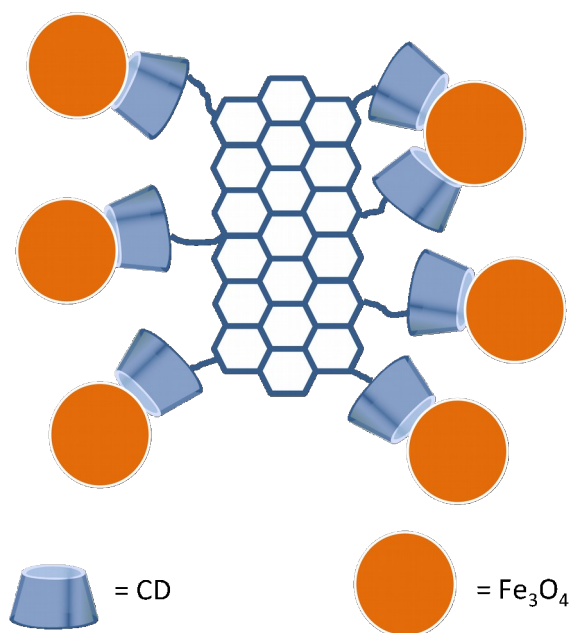


Figure 4 Schematic structure of $\text{Fe}_3\text{O}_4@CD\text{-MWCNT}$ composite nanoparticles (redrawn after ref. 18)

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β -CD, Stability, Orange arome

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One-pot synthesis, Multicomponent condensation, Solvent-free conditions, Recyclable catalyst

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