

Reconfigurable soft matter

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This special issue focuses on soft, reconfigurable materials. A distinctive feature of these systems is their ability to sense and respond to external stimuli or changes in the environment by readjusting their morphology, chemistry and/or functionality. Examples abound in biology since this adaptive behaviour is vital for survival. Consider, for example, the ability of the mimic octopus and cuttlefish to change their colour and texture and thus, camouflage their identity to evade their predators. The ability of plants to bend or unravel their leaves in the presence of sunlight is another example of adaptive behavior in biology that is crucial for survival. Herein, we highlight research focused on imbuing synthetic materials with analogous abilities to undergo remarkable reconfiguration that leads to changes in materials properties and performance. Advances in this area could enable new multi-functional systems, which display one structure and function in one environment but display another shape, surface property and function under different conditions with applications in robotics, self-assembly, actuated materials, stimuli sensing, controlled

transport and release systems, and programmable shape control.

In order to undergo significant structural reconfiguration, artificial systems typically encompass a coupling between mechanical and chemical, electrical, optical or thermal behaviour. Geryak and Tsukruk (DOI: 10.1039/c3sm51768c) reviewed recent advances in synthetic and fabrication approaches to reconfigurable soft materials, with a focus on the use of either long-range fields (*i.e.* electrical and magnetic) or changes in local thermodynamic parameters (*e.g.* solvent quality) in order to elicit a precise dimensional and controlled response. Hydrogels considered by Yoshida *et al.* (DOI: 10.1039/c3sm51537k) and Vaia *et al.* (DOI: 10.1039/c3sm51650d) involve an inherent coupling between chemical and mechanical energy. In particular, both these studies involve the self-oscillating Belousov–Zhabotinsky (BZ) reaction, enabling gels to undergo autonomous pulsations, so that they appear to beat spontaneously like a heart. In studies presented here, Vaia developed new modes of inserting the catalyst critical to the reaction into a number of different gels in order to broaden the range of systems that can exhibit these remarkable, periodic expansions and contractions, which can be harnessed to perform mechanical work. Yoshida, who was the first scientist to synthesize these BZ gels, now reports on a new catalyst for the BZ reaction, showing how this

catalyst could lead to dramatic viscosity oscillations in the host solution as the system undergoes a periodic sol–gel transition.

One of the desired properties of reconfigurable materials is their ability to undergo or induce movement. Dickey *et al.* (DOI: 10.1039/c3sm51921j) exploited the coupling between chemical, electrical and mechanical energy in polyelectrolyte hydrogels, introducing an external electric field to create gel “legs” that can be driven to “walk”. Notably, the ability to undergo directed motion is a hallmark of biological systems, with simple cells showing a remarkable sensitivity to the compliance of the underlying substrate. Intrigued by this behavior, Aranson *et al.* (DOI: 10.1039/c3sm51597d) devised a theoretical model to simulate the “crawling” of cells on surfaces with different mechanical properties and uncovered a scenario where the cell actually exhibits a “bipedal” motion. Directed propulsion was also the focus of work by Christianen *et al.* (DOI: 10.1039/c3sm52294f), who reviewed research on harnessing magnetic fields to steer nano- and micro-scale synthetic objects, including anisotropic Janus particles. The dynamic reconfigurations of artificial cilia, which undulate due to the motion of the surrounding fluid, can set in motion and expel the particles from the underlying surface. These findings by Balazs *et al.* (DOI: 10.1039/c3sm52156g) suggest that such

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“passive” cilia (which simply move in response to local flows) can be used for anti-fouling applications.

The ability of materials to reconfigure and subsequently change their function through self-assembly and replication became the theme of several papers in this special issue. Lewis *et al.* (DOI: 10.1039/c3sm52418c) demonstrated the electric field-induced assembly of patchy Janus rods into reconfigurable chains, whose structure could be tailored by varying the features of the rods and the field. Balazs *et al.* (DOI: 10.1039/c3sm52300d) as well focused on assemblies of solid particles, modeling polymer grafted nanoparticles (PGNs) that are cross-linked into extensive networks through high-strength and weaker bonds. The latter studies revealed that these cross-linked PGNs can undergo significant structural rearrangement in response to mechanical deformation and thus, are exceptionally strong and tough. In effect, these networks mimic some of the desirable features of natural dermal armor found on various animals such as the armadillo. Olvera de la Cruz *et al.* (DOI: 10.1039/c3sm52501e) proposed a scheme for self-replication where the fine-tuning of asymmetric interactions between anisotropic particles leads to highly accurate self-replication. The kinetic Monte Carlo algorithm further showed that with the introduction of energy cycling to enable the periodic assembly/disassembly of the system's components, the replicator population can grow exponentially.

Liquid crystalline materials formed another class of reconfigurable substances examined by researchers in this issue. In particular, dramatic changes in shape were demonstrated in work by Verduzco *et al.* (DOI: 10.1039/c3sm51654g) who constructed a bilayer material from liquid crystalline elastomers (LCE) and polystyrene (PS) and showed that the “petals” formed from these LCE-PS systems could be made to open and close in response to variations in temperature. As described in a review article by Lavrentovich (DOI: 10.1039/c3sm51628h), liquid crystals can be harnessed to manipulate the motion of colloidal particles and can even lead to the levitation of these particles. Vaia *et al.* (DOI: 10.1039/c3sm51865e) showed that the light-driven

flexural-torsional response of azobenzene-functionalized liquid crystal polymer networks can be controlled by specifying the material alignment and actuating the material with polarized light. These studies lay the groundwork for efforts to combine polarization-controlled flexural-torsional deformations with complex geometry and loading conditions.

The use of azobenzene chemistry, molecular reconfiguration and surface hydrophobicity in designing novel dynamic materials is further demonstrated by Sukhorukov *et al.* (DOI: 10.1039/c3sm51648b), who showed that UV-responsive microcapsules containing azobenzene could be ruptured and broken apart through the application of light. These studies can lead to new means of modulating the release of species encapsulated in these carriers. Burgess *et al.* (DOI: 10.1039/c3sm52684d) demonstrated that inverse opal films, whose surfaces are functionalized with a polymer doped with azobenzene chromophores, undergo

continuous changes in material hydrophobicity and wettability upon light exposure. The presence or absence of the liquid infiltration and the associated switching of the opalescence can enable the use of such dynamic opals as colorimetric indicators in anti-tampering applications. Going from synthetic opals to marbles, Zang *et al.* (DOI: 10.1039/c3sm51885j) showed that liquid water marbles coated with fumed silica nanoparticles exhibit various recoverable shape transformations upon freezing, which correlate with the nanoparticles' hydrophobicity.

While flexural-torsional molecular deformations are responsible for the structural changes of stimuli-responsive and shape-memory polymers, self-shaping capabilities of the material can arise from materials microstructure and reconfiguration that takes place at the microscopic scale. The review article by Studart and Erb (DOI: 10.1039/c3sm51883c) discusses recent attempts to replicate in synthetic materials such shape-changing behavior of selected natural materials displaying deliberately tuned fibrous, one-dimensional architectures. Simple processing methods such as drawing, spinning or casting under magnetic fields are shown to be effective in mimicking the orientation and spatial distribution of reinforcing fibers of natural materials, thus enabling unique shape-changing features in synthetic systems. Yang *et al.* (DOI: 10.1039/c3sm51640g) investigated the reconfiguration of two-dimensional pH and temperature dual-responsive hydrogel membranes that were perforated with micron-scale pores. Swelling-induced instabilities were shown to induce four distinctive morphologies of the hydrogel membrane and reversible switching among the chiral twisted state, buckled state, and circular state *via* changing the temperature or pH. Lyon *et al.* (DOI: 10.1039/c3sm52518j) demonstrated that microgel multilayers have unique viscoelastic properties that can be controlled to induce morphological changes in biological cells cultured on the film.

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