

Collective Assemblies

Stronger together

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Considering responsive materials as transient collective assemblies rather than individual shape-changing objects allows for emergent functionalities that cannot be derived from the properties of single objects but are driven by interactions between them.

The whole is greater than the sum of its parts. Those famous words, attributed to Aristotle, are often used to popularize one of the most vital concepts of biological systems – emergent behaviour. It describes the synergistic, collective actions appearing in complex systems that stem from communication and interactions between their constituents: emergent behaviour cannot be derived from the properties of the individual components of the system. The concept of emergence is pertinent to a wealth of phenomena, ranging from cellular functions to adaptive motions observed in flocks, schools, and swarms¹ and problem-solving in groups of ants². In the context of materials science, there is a quest for developing synthetic materials that strive to mimic the intelligence of biological systems: materials that can interact with and adapt to their surroundings^{3,4}. But how can the collective actions observed in large biological populations be mimicked in synthetic responsive materials? This is the question Mustafa Abdelrahman and coworkers address in their recent contribution in *Nature Materials*⁵.

The bioinspiration source for Abdelrahman and colleagues stems from transient, mechanically interlocked structures formed by species like fire ants and California blackworms^{6,7}. Such “living aggregates” accomplish several functions related to protection, thermal management, rafting, and/or crossing obstacles⁸ (Fig. 1a, left), and they disassemble at will when the task has been executed or threat removed. Herein, mechanical interlocking is demonstrated with ribbons of thermoresponsive shape-changing polymers (Fig. 1a, right). The resulting polymer aggregates behave as transient viscoelastic solids with mechanical properties that are markedly different than those of the constituent polymer ribbons. The aggregates exhibit large volume contraction compared to dispersions of individual ribbons, which is accompanied with up to 12-fold increase in yield stress and 350-fold increase in normal force upon penetration tests. Together they are stronger!

The building blocks for the mechanically interlocked aggregates are liquid crystal elastomer (LCE) ribbons. LCEs are thermoresponsive polymers that combine the elasticity of loosely crosslinked polymer networks and controlled, anisotropic molecular alignment of liquid crystals. By distorting the molecular alignment with external stimuli such as temperature or light, LCEs can undergo programmed reversible shape changes, causing initially straight ribbons to bend, twist, or contract, as dictated by molecular alignment programming during the polymerization⁹. Abdelrahman and colleagues utilized twisted LCE ribbons where the molecular alignment rotates by 90° through the thickness of the ribbon (Fig. 1b). In such ribbons, the deformation mode depends on the direction in which the ribbons are cut with respect to the molecular alignment, ranging from pure bending (0°) to pure twisting (45°). The mechanical interlocking is shown to be driven by bending, as LCE ribbons that exhibit pure twisting do not aggregate. Interestingly, a combination of bending and twisting is needed for reversible control over the assembly–disassembly process: ribbons that interlock via pure bending form irreversible entanglements.

Perhaps the biggest drawback of the LCE-based mechanically interlocked assemblies is the high temperature required to drive the aggregate formation, typically 175 °C. The experiments were conducted in a heated silicon oil bath, under conditions that are sufficient to prove the phenomenon but are hardly suitable for further practical utilization. To address this deficiency and demonstrate the generality of the concept, the authors replaced the LCE ribbons with hydrogel bilayer ribbons, which deform and assemble in biologically relevant temperatures (37 °C). Utilizing the versatile and programmable shape changes of LCEs, they also

fabricated semicrystalline LCE ribbons that bend and form the transient aggregates at room temperature, while high temperatures are required to redisperse the ribbons.

Considering shape-morphing materials as transient collective assemblies rather than individual deforming ribbons brings about a wealth of possibilities. Coating the LCE ribbons with liquid metal renders them light-responsive, allowing for remote – and, if needed, localized – control over the assembly–disassembly process. At the collective level, the contact points formed by the mechanical interlocks enable the reversible transition from an electrically insulating (dispersed ribbons) to a conductive (collective assembly) state. The conduction pathways can be distorted by cutting the assembly in two parts, breaking the electrical circuit. Intriguingly, cyclic light illumination triggers the formation of new interlocks to “heal” the damaged part, which closes the circuit and allows the lost functionality – in this case electric conduction – to be regained (Fig. 1c). The damage repair and electric conductivity are examples of emergent functionalities that require the ribbons to interact and entangle. They are attributable only to the whole, making it greater than the sum of its parts.

The concept proposed by Abdelrahman and coworkers opens up several future prospects. Can the collective assemblies be formed in response to stimuli other than global or photothermal heating? Can multi-stimuli-responsive aggregates be devised where, depending on the stimulus used, different functionalities can be obtained? Can gradients be programmed into the mechanically interlocked assemblies? In terms of potential biomedical applications, dynamic cell scaffolds with controllable and tunable mechanical properties or programmable mechanical gradients can be envisaged for the hydrogel aggregates. Injectable biomaterials with controlled porosity, based on collective assemblies that form only after reaching the target, is another unique opportunity provided by the whole of carefully designed, stimuli-responsive building blocks. Time will tell what emerges next – the journey is only at its beginning.

Competing Interests

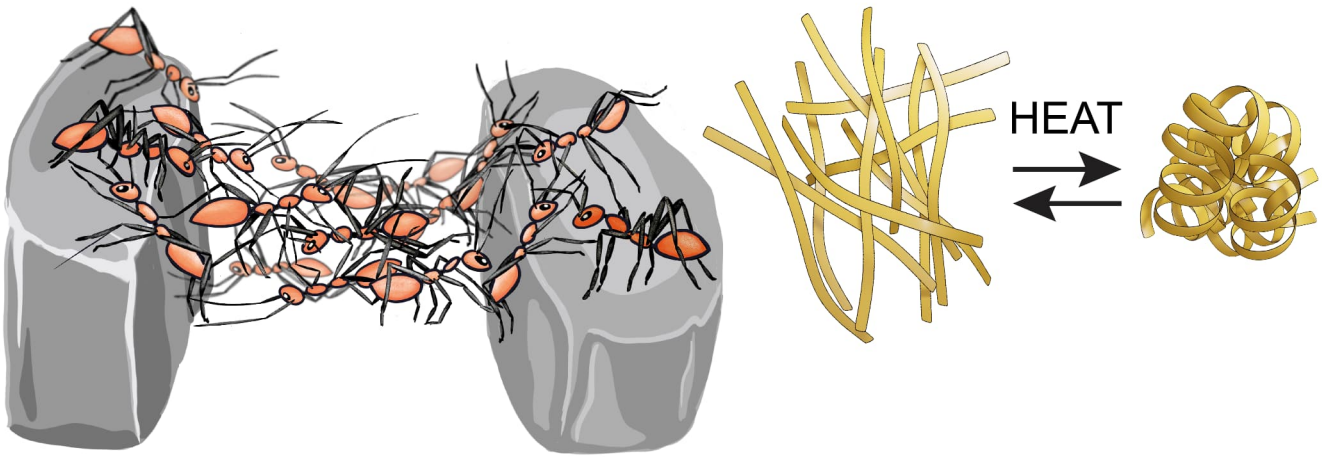
The author declares no competing interests.

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Figure 1 | Collective shape-changing assemblies with emergent functionalities. a, Some biological species such as fire ants form transient aggregates in order to accomplish tasks they are not able to execute alone. Using liquid crystal elastomers (LCEs) and other shape-changing materials, this concept was extended to synthetic aggregates. b, LCEs were polymerized in twisted nematic configuration, and ribbons were cut in a specific angle to yield both bending and twisting deformations in order to obtain aggregates that reversibly assemble and disassemble. c, Synthetic assemblies display emergent functionalities driven by the interactions between them, such as damage repair and electric conductivity which do not occur in dispersed assemblies of LCE ribbons.

a Biological aggregates Synthetic aggregates



b Liquid Crystal Elastomer

