Designing two-dimensional materials that spring rapidly into three-dimensional shapes

By Chunhong Ye and Vladimir V. Tsukruk

One of the hallmarks of living organisms, including humans, is the ability to actively respond and adapt to stress. Even plants, which lack a nervous system, react to different stimuli by changing their shape. Some well-known examples are the wrinkled edges of leaves as a response to compression during growth, wrinkles of skin to accommodate flexing and bending movements or age-drying stresses, or the mimosa’s rapid folding of its leaves via propagating waves of osmotic pressure when touched. Similar phenomena have been used in engineered artificial two-dimensional (2D) materials that can respond to external stimuli, but these applications typically are simple, slow, and work only along one dimension. On page 154 of this issue, Xu et al. (1) demonstrate a new paradigm of designing functional materials that can quickly snap into complex 3D architectures via localized buckling.

Buckling phenomena have been widely explored as a means for measuring elastic properties of ultrathin polymer films (2), as periodic templates for organized nanoparticle assemblies (3), as a complex patterning approach for metal-polymERIC sandwiched films (4), for bistable patterning of nanoparticle-polymer multilayers (5), and for chiral patterning of periodic porous materials (6). The underlying physics of wrinkle formation is well understood and arises from the mechanical instability that develops in compressed thin films (7). However, all of these phenomena have an essentially 2D nature, with “penetration” into the third dimension being extremely limited. Furthermore, attempts to overstress the materials result in irregular large-scale folding and crumpling.

Thus, efforts to break through into the realm of 3D shapes now exploit controlled interfacial stresses. Among them are multistep folding of polymer bilayer films into 3D pyramids and “flowers” (8), chemomechanical mediation of a gel-embedded high-aspect ratio nanorod array (9), self-rolling of helical microtubes from laterally patterned hydrogel sheets (10), and self-rolling of bilayer gel sheets with different shapes into helical and smooth tubules (11) (see the figure, panels A and B). Complex shapes can also be induced by thermal control of out-of-plane transformation of gels with halftone-patterned cross-linking (12). These transformations and the resulting 3D shapes are predicted by either analytical considerations for simple shapes (7, 8) or finite-element analysis for complex shapes (9).

In the case of nanoscale materials, more sophisticated computer modeling is required to predict corresponding transformations (see the figure, panel C). Moreover, tuning composition, thickness, and mechanical properties with fine resolution can be a very challenging feat of materials engineering. Even if some astonishing examples of shape transformations have been demonstrated, these materials are usually slow to respond (on a time scale of minutes), frequently require controlled environment (e.g., swollen hydrogels need liquid water or humidity),

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and are often limited to fairly simple shapes and transformation paths.

New design principles for materials with fast-snapping microstructures suggested by Xu et al. cleverly combine conventional uniform compressive stresses with multiple preprogrammed “weak” points that allow buckling to occur in a hierarchical fashion. Compressive stresses initiate a cascade of buckling events that spring the initial surface-confined planar architectures into 3D complex structures. In this way, various metal and silicon serpines and ribbons are converted into a collection of microscopic tents, tables, baskets, flowers, boxes, stars, and many other shapes. An amazing variety of conveniently separated “first floor” and “second floor” buckled microstructures are demonstrated as well.

Although the prospective applications of this technique are astounding in breadth and impact, there are more intriguing questions to address. How can the extremely tedious planar processing of large-scale microfabrication be improved or replaced by more facile approaches, such as directed assembly, with similar outcomes? Can these architectures be “unbuckled”—for example, can an open box be closed? Can these structures eventually be sustainable in their 3D shape as stand-alone structures after release from supporting substrates? How does splaying into a 3D architecture affect the global functions of nanostructured constructs, such as electronic, optical, or magnetic properties? Can structures that mimic the mimosa, in which a highly localized stimulus with a pinpoint compression results in a rapid cascade of 3D shape transformations, be made on much larger scale? And finally, what are the limits of scale? Perhaps one day, if you order a house complete with furnishings and a white picket fence, you may just receive a box with a label that reads, “Compress to unfold.”

REFERENCES AND NOTES

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MATERIALS SCIENCE
Using all energy in a battery
Controlled electrode structure improves energy utilization

By Nancy J. Dudney and Juchuan Li

It is not easy to pull all the energy from a battery. For a battery to discharge, electrons and ions have to reach the same place in the active electrode material at the same moment. To reach the entire volume of the battery and maximize energy use, internal pathways for both electrons and ions must be low-resistance and continuous, connecting all regions of the battery electrode. Traditional batteries consist of a randomly distributed mixture of conductive phases within the active battery material. In these materials, bottlenecks and poor contacts may impede effective access to parts of the battery. On page 149 of this issue, Kirshenbaum et al. (1) explore a different approach, in which silver electronic pathways form on internal surfaces as the battery is discharged. The electronic pathways are well distributed throughout the electrode, improving battery performance.

Commercial battery electrodes (anodes and cathodes) are typically created by casting a porous powder composite (a mixture of the active material, a small amount of polymer binder, and a conductive additive such as carbon) onto a metal foil current collector. Electrons are conducted via chains of particles through the composite to the current collector. In contrast, ions move through the liquid or solid electrolyte that fills the pores of the composite. Optimization of both pathways is critical for battery performance. Although this slurry-cast electrode structure works very well, better control of the three-dimensional (3D) architecture would enhance the energy per unit mass and volume of the electrode. Cobb and Blanco recently reported an important step in this direction by creating a cathode consisting of alternating low- and high-density stripes. The low-density stripes provide higher porosity and better access for ions traveling through the liquid electrolyte into the cathode (2).

Kirshenbaum et al. now report the fabrication and performance of a silver vanadium phosphate cathode with a well-defined 3D architecture. The cathode consists of relatively dense thick pellets without binder or conductive additives. When Li+ ions and electrons move into the silver vanadium phosphate particles, V5+ is reduced to V3+ and Ag+ is reduced to metallic silver; the latter remains at the surface of the active material particles as small silver particles, presumably electrically connected by a thin layer of Ag (see the image for a micrograph following such a reaction). Under the right conditions, the silver forms an effective electronic path throughout the electrode, enhancing the insertion of Li+ into the cathode lattice and hence increasing the amount of accessible energy in the battery. Such reduction displacement reactions, also known as conversion reactions, occur upon Li+ reaction with a wide range of binary and bimetallic oxide, fluoride, and sulfide compounds (3). These materials have potentially very high energy densities that may yield rechargeable and low-cost battery materials. The biggest challenge for practical use of such reversible conversion electrodes is the voltage penalty, where the voltage of the battery during discharge (conversion) is much less than the voltage needed for recharge of the battery (reconversion). It remains unclear how much of this voltage penalty is intrinsic and how much of it is a result of kinetic limitations that could in principle be minimized (4, 5).

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