

fields experimentally should thus open exciting avenues, from the fundamental to the applied.

The knotted colloids of Smalyukh and colleagues bring the prospect of creating Kelvin-like knots in liquid crystals one step closer to reality. On the one hand, and unlike the recent realization of Kelvin vortex knots in simple fluids<sup>6</sup>, the liquid-crystal knots are statically pinned to the fabricated tubular structure — made possible by the structured nature of the fluid — and makes them more amenable to practical applications. Also, the knotted defect loops should give rise to directional interactions between the knotted tubes to which they are linked,

potentially offering enticing prospects for materials engineering<sup>3</sup>. On the other hand, the techniques employed by Smalyukh and co-authors may prove useful to further explore the interaction of liquid-crystal order with more complex topological obstructions. Could liquid-crystal knots be created in the absence of templating solid knots? Perhaps knot-shaped optical fields will provide an answer<sup>7–9</sup>. If liquid-crystal knots could be templated by optical fields, might their structure shed light on the connection between knot theory and physical fields? Although the problems ahead are likely to be knotty, the prospects are enticing indeed. □

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## GLUING GELS

# A nanoparticle solution

Synthetic polymer gels with certain surface chemistries can be glued together by a simple and inexpensive method that uses commercially available silica nanoparticles. Biological tissues can also be joined by this nanotechnological route, eliminating the need for sutures, additional adhesives or chemical reactions.

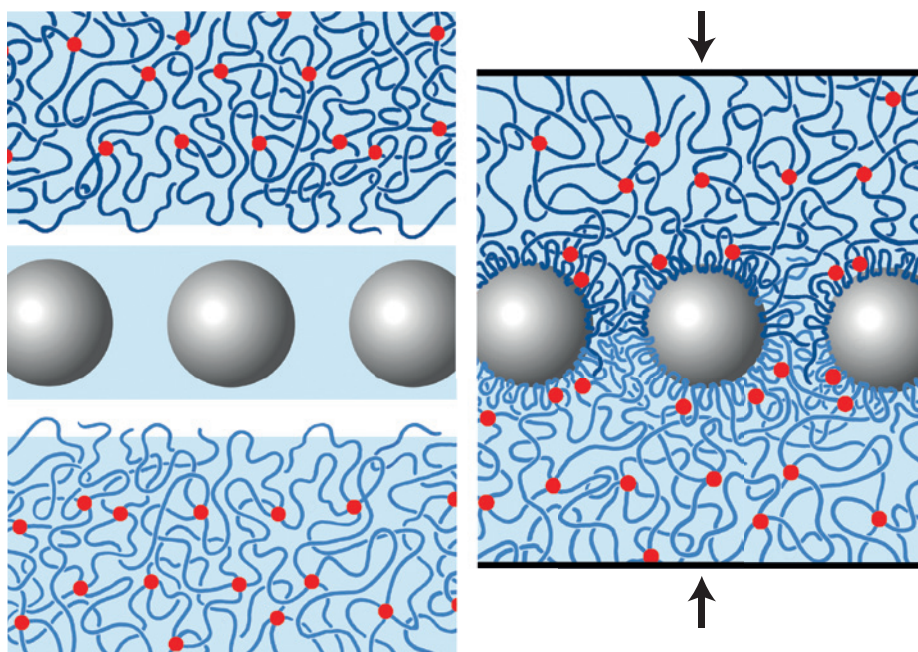
Eric A. Appel and Oren A. Scherman

For over 200,000 years humans have been sticking things together<sup>1</sup>, yet when it comes to gluing soft, squishy and slippery materials, such as hydrogels, strong adhesion is notoriously difficult to accomplish. Many adhesives are made of polymers because, unlike other materials, they ensure good contact between surfaces and they can dissipate energy under stress through rearrangement of their chains<sup>2</sup>. Yet, polymer-based glues are often not water soluble or require *in situ* polymerization that can lead to unwanted side reactions and, as a consequence, significant alteration of the materials' properties. To circumvent these problems, scientists have exploited water-based molecular recognition motifs (for example, cyclodextrins<sup>3</sup> and cucurbiturils<sup>4</sup>) or have taken hints from nature, developing mussel-inspired catechol-based adhesives<sup>5,6</sup>.

Now, writing in *Nature*, Leibler and co-workers report a simple yet elegant method for 'gluing' hydrogels together that takes advantage of the unique adhesion properties of nanoparticles and stress dissipation properties of polymer chains (Fig. 1)<sup>7</sup>. The method builds on previous studies of self-healing rubbers<sup>8</sup> and malleable thermosets<sup>9</sup>, and results in strong and rapid adhesion between two hydrogels at room temperature. It uses

inexpensive nanoparticles and works for a diverse range of polymeric hydrogels, making the process accessible to researchers

of different backgrounds and providing the potential for rapid translation to commercial applications.

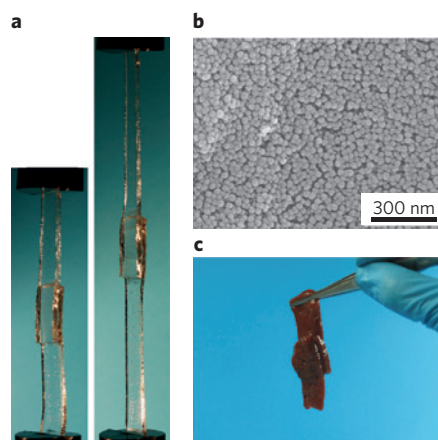


**Figure 1** | Gluing gels with nanoparticles. A schematic illustration showing strong, yet dynamic and reversible gluing of gels by exploiting adsorption of polymer chains to the surface of the nanoparticles when pressure is applied (black arrows)<sup>7</sup>. The strongest adhesion is expected when the nanoparticle diameter is comparable to the distance between crosslinks (denoted by the red dots).

Leibler and co-workers show that by spreading commercially available silica nanoparticles in suspension onto the surface of gels composed of poly(dimethylacrylamide) (PDMA) robust monolayers of the nanoparticles can be formed (Fig. 2b). When a second piece of PDMA gel is brought into contact with the nanoparticle-functionalized surface and the two are pressed together for a few seconds, a strong adhesive bond is readily formed that can sustain large deformation (Fig. 2a). The silica nanoparticles act as linking units between the two surfaces of the PDMA gels and the strongest adhesion is expected when the diameter of the nanoparticles is comparable to the mesh size of the gel network (Fig. 1). Adhesion occurs only when the surface chemistry of the nanoparticles is matched appropriately with the polymeric gels; it does not occur when the surface chemistry of the nanoparticles and polymers, for example polyacrylamide, is incompatible.

The PDMA gels form dynamic and multivalent attachments to the nanoparticles with constant exchange between adsorbed and desorbed states, providing strong adhesion, but also allowing for dissipation of energy under stress through the rearrangement of the polymer chains. When an adhesive junction is strained, adsorbed polymer chains can detach from the particle surface to relax the tension, leaving room on the surface that can be filled by a neighbouring polymer strand, and thus avoiding failure. Indeed, this process can be so efficient that the hydrogels themselves tear in lap-shear tests before failure at the adhesive junction occurs. Moreover, the dynamic rearrangement of polymer adsorption to the nanoparticles permits re-positioning, and thus self-healing, of adhesive joints after failure. It is shown that glued gels can fully recover their initial strength after failure of the adhesive joint by simply pressing the two pieces together again, without the need for further application of nanoparticles.

Probing the concept further, Leibler and co-workers demonstrate that these nanoparticles can solve the difficult problem of gluing gels of different compositions: more specifically, PDMA gels can be glued to gelatin gels, because both gels can be sufficiently adsorbed onto the nanoparticles. It is also possible to glue gels together that have different rigidities, providing they have similar equilibrium swelling. The strength of the adhesion between two gel pieces was found to increase with particle size as more chains could adsorb onto each nanoparticle,



**Figure 2** | Strong adhesion between diverse surfaces<sup>7</sup>. **a**, Two gel pieces are glued together after spreading a drop of silica nanoparticle suspension between them (left). The glued joint is able to sustain large deformations (right). **b**, Scanning electron micrograph of a monolayer of silica nanoparticles on a PDMA gel surface. **c**, Two pieces cut from a calf liver (with no special treatment) are glued together with silica nanoparticles. After pressing the two sections together for 30 seconds, the joint holds well and the tissue is manipulable.

constituting stronger crosslinking between hydrogel pieces. The optimal nanoparticle size, however, is expected to be comparable to the mesh size of the gel network. Further investigation of the effect of nanoparticle size is necessary to develop prescriptive models — in particular, very large particles, which may exhibit sub-optimal adhesion properties, are yet to be explored. Leibler and co-workers also demonstrate that the concept works with other nanomaterials including cellulose nanocrystals and carbon nanotubes, again providing they are modified to have the appropriate surface chemistry. From these collective studies, the researchers conclude that it is possible to tune the adhesion strength between two gels by both altering the surface chemistry of the particles and by changing the particle size. One limitation of the reported process is that the use of tightly crosslinked or highly swollen materials, where polymer strands are more constrained, could weaken the association to the nanoparticles. It may be possible, however, to bypass this limitation, in a practical sense, by increasing the polymer association to the nanoparticles by altering the nanoparticles' surface chemistry.

Beyond relatively simple and homogeneous synthetic polymer hydrogels, Leibler and co-workers apply

their system to complex soft biological tissues. Using the same commercially available nanoparticle suspension, it is possible to glue together two ribbons of freshly cut calf liver without any special treatment or drying procedures (Fig. 2c). Of course, much of the extracellular matrix composing such tissues is made of natural polymers that show robust adsorption to the nanoparticles. Yet biological tissue is extremely diverse in composition and it will certainly be advantageous for researchers to investigate just how far these concepts can be used to glue together other tissues. It may be possible to tune the nanoparticle size and surface chemistry appropriately to glue any number of biological tissues. The concept of suture-less wound healing<sup>10</sup> is surely one of great interest within the biomedical community. One can envision a multitude of biomedical applications including tissue engineering (for example, an assembly of complex, large or multilayered tissue *in vitro*), surgical procedures and drug delivery (for example, the immobilization of site-specific drug-eluting implants).

The discovery of simple, efficient, cost-effective and safe methods for gluing complex materials is extremely powerful and many will certainly be rushing to apply these systems in various ways. One of the legacies of this work may indeed be its influence on the way scientists and engineers think about adhesion in both industrial and biological settings, opening doors for building functional materials. □

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