Self-healing hydrogels ease into production

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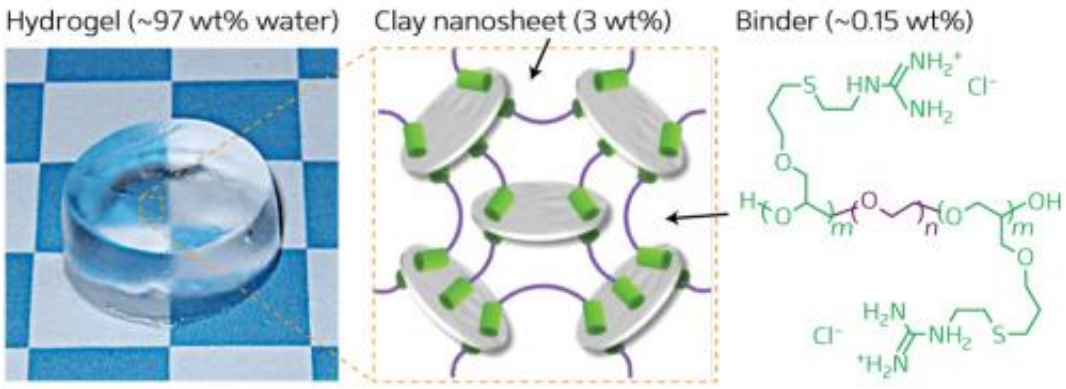

Figure 1: Free-standing hydrogels formed by the interactions between clay nanosheets and polymer binding agents might find future use in biomedical applications. Credit: American Chemical Society

Hydrogels are semi-solid materials formed by polymer chains that trap water molecules into three-dimensional gels. They are used in a variety of applications, including soft contact lenses, but the fragile nature of the materials means that their utility has remained limited. Yasuhiro Ishida, Takuzo Aida and colleagues at the RIKEN Center for Emergent Matter Science are challenging this limitation by developing strong hydrogels, or 'aqua materials', that could outperform even conventional plastics. As part of an international collaboration, the team has now improved the production of a robust, moldable hydrogel that heals itself rapidly after being sliced open.

The RIKEN team previously developed a hydrogel containing three

primary ingredients: tiny flakes or 'nanosheets' of anionic clay, an exfoliating chemical that keeps the nanosheets from agglomerating and a polymer binder containing positively charged guanidinium cations. Mixing small amounts of these substances into a beaker containing water causes a self-standing gel to form within seconds due to cross-linking interactions between the clay nanosheets and the polymer binder (Fig. 1). Since the hydrogel is held together by hydrogen bonding and electrostatic forces instead of permanent chemical bonds, if cut open it can be repaired by simply pressing the gel back together.

Developing practical applications for this hydrogel proved difficult, however, because the polymer binder contains dendritic units—multiply branched, star-shaped molecular chains that can only be synthesized through time-consuming procedures. Unfortunately, binding agents made from more traditional acrylic polymers severely affect the performance of the self-healing aqua materials.

To devise a solution, the RIKEN team collaborated with Craig Hawker and colleagues from the University of California, Santa Barbara, in the United States to investigate the possibility of using advanced polymers known as 'ABA triblock copolyethers' that link adhesive ionic end-units (A block) and a flexible poly(ethylene oxide) core (B block) into a linear chain. This type of polymer mimics the essential attributes of dendritic binding agents but can also be easily synthesized.

Experiments demonstrated that the ABA triblock copolyethers cross-linked with the clay nanosheets as well as the original dendritic polymer binder. After optimizing the chain lengths of each ABA triblock segment, their new polymer binder rapidly generated a hydrogel with comparable mechanical strength and self-mending capabilities. The hydrogel also displayed an intriguing 'shape memory' behavior that enabled it to retain its structure after drying and re-wetting with water and organic or ionic liquids. "With these advantageous features, the

hydrogel could find applications in biomedical treatments and surgical operations, including use as anti-adhesive materials," notes Ishida.

More information: Tamesue, S., et al. Linear versus dendritic molecular binders for hydrogel network formation with clay nanosheets: Studies with ABA triblock copolyethers carrying guanidinium ion pendants, *Journal of the American Chemical Society* 135, 15650–15655 (2013). dx.doi.org/10.1021/ja408547g

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