Editorial corner – a personal view

Repeated self-healing of wider cracks in polymers

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Self-healing polymeric materials have received increasing research interest in recent years, as the technology is believed to significantly improve the reliability and durability of products. Before these smart materials are commercialized, however, a series of challenges have to be tackled.

The rehabilitation of wider cracks, for example, remains a big problem. Many reports in the literature showed that healing efficiency becomes rather low when crack width is larger than 100 micrometers.

In principle, the combination of the shape memory effect and the self-healing function, as proposed by Professor Karger-Kocsis (<https://doi.org/10.1007/s10853-007-2176-7>), would be a solution. Namely, the shape memory effect may help close the wide cracks first, which creates the pre-requisite for the subsequent self-healing reaction across the interface. Although there have been some researches in this aspect, *e.g.* extrinsic self-healing assisted by shape memory alloy wires (<https://doi.org/10.1016/j.compscitech.2018.03.040>), the available methods are not suitable for the repeated self-healing of wider cracks, due to the limitations of the one-way shape memory effect or extrinsic self-healing.

The ideal solution will probably be a combination of the reversible shape memory effect and reversible self-healability. When reversible intrinsic self-healing ability is combined with the two-way shape memory effect in a single polymer, larger cracks can be repeatedly closed autonomously and then rebonded whenever they appear. In this case, no programming is needed for the polymer to regain the shape memory effect after each healing event. As a proof-of-concept, a reversible C–ON (alkoxylamine) bond

is introduced into the main chain of crosslinked polyurethane (PU) which contains an elastomeric dispersed phase (styrene–butadiene–styrene block copolymer, SBS). After programming, the temperature-induced reversible, opposite directional deformabilities of the crystalline phase of PU and the compressed SBS phase, in association with the melting and orientated re-crystallization of the crystallites, produce a two-way shape memory effect. As a result, the thermal retractility offered by the shape memory effect coupled with the reversible C–ON bonds can successively close and repair wider cracks (~250 μm in width) multiple times.

More recently, we have developed a polymer hydrogel that combines temperature responsiveness with intrinsic self-healing ability through host-guest orthogonal self-assembly (<https://doi.org/10.1021/ac-sami.0c03523>). It is capable of self-repairing millimeter-scale cracks (up to ~3.5 mm in width), fast and repeatedly.

The above clearly demonstrates the importance of integrated innovation, which can lead to a breakthrough when a single technology cannot.

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