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Self-healing thermoreversible covalent networks – from material design to soft robotics

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The thermoreversible Diels-Alder reaction between furan (diene) and maleimide (dienophile) groups is increasingly used for developing self-healing (SH) applications, ranging from SH coatings to SH soft robotic actuators [1-3]. The principle of these materials is quite straightforward: at low temperatures cycloadducts links the network together, while at higher temperatures the reaction equilibrium shifts towards the disconnected state. Nevertheless, the application and processing of these thermoreversible networks requires detailed insight in their reaction kinetics and chemorheology.

A comprehensive study of the kinetics provided a model that can be used to simulate the cross-linking of the polymer network upon cooling, facilitating the optimization of temperature schedules for materials having a specific network composition. A complicating factor is the existence of two stereoisomeric cycloadducts: one being more thermodynamically stable, while the other one forms faster [4]. Nevertheless, by controlling the composition and the temperature programs, the network formation kinetics and the healing quality can be steered.

The reversible gel conversion, at which the material switches between a liquid and elastic state, is crucial for the design of healing protocols during which the shape of the object must be retained. Tuning the chemorheology, involves the careful selection of building blocks. The achievable SH materials range from reversible elastomers and thermosets to networks with variable crosslink densities.

A judicious material design, combined with detailed insight in the material behaviour, facilitated the development filament extrusion and the 3D printing of SH actuators for robotics applications [3,5].

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