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# Studying reversible reactions and chemorheological behaviour for the additive manufacturing of self-healing actuators

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Over the past decade, the development of self-healing or remendable polymers markedly increased. Our work in this field is focussed on thermoreversible polymer networks based on the thermoreversible Diels-Alder reaction between furan (diene) and maleimide (dienophile) groups. Detailed knowledge about the reversible reaction kinetics and the connected chemorheological changes, gained through extensive use of thermal analysis and dynamic rheometry, helped us to develop these materials and their processing for application in self-healing (SH) soft robotic actuators [1-3].

The principle of these materials is quite straightforward: at high temperatures, the Diels-Alder/retro Diels-Alder reaction equilibrium is shifted towards the disconnected state, while at low temperatures, the cycloadducts are formed through the Diels-Alder reaction, linking the network together. A complicating factor is the existence of two stereoisomeric cycloadducts: one being more thermodynamically stable, while the other one forms faster [4]. As the network formation occurs at a lower temperature, e.g., in ambient conditions, it is generally quite slow, so the knowledge of the thermoreversible reaction kinetics is crucially important for optimizing the temperature program used during processing or healing. Moreover, combining this kinetic information with detailed insight in the conversion and temperature dependent visco-elastic behaviour of the materials facilitated the adaptation of the network composition to tune the material behaviour during processing and healing.

A comprehensive study of the thermoreversible reaction kinetics, using DSC, microcalorimetry, and FTIR and NMR spectroscopy, provided a reaction kinetics model that can be used to simulate the changes in the cross-linking of the polymer network upon heating and cooling, facilitating the optimization of temperature schedules for a specific material, as well as the optimization of the network composition to ease the processing or improve the healing kinetics.

The reversible gel conversion, the reaction conversion at which the material switches between a liquid and elastic state (and vice versa), is crucial for the design of healing protocols, during which the shape of the object must be retained. Of course, the chemorheology can be tuned by a careful selection of the building blocks used to construct the network, which was a key aspect for the development of filament extrusion and filament deposition modelling of these thermoreversible polymer networks [3,5].

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