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Magnetothermal-Triggered Response and Self-Healing of Recyclable and Reprocessable Nanocomposites for Actuation Systems

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The field of stimuli-responsive soft materials is rapidly evolving, particularly with recent advances in the design and fabrication of magnetic soft materials. By incorporating magnetic nanoparticles (MNPs) into an elastic polymer matrix, materials with rapid, fully reversible mechanical responses to magnetic fields can be created, making them highly suitable for developing flexible, remotely controlled soft robots. However, conventional crosslinked elastomer materials often lack reprocessability and recyclability, limiting their lifespans, especially under repeated mechanical stress. These limitations can be overcome by using vitrimers as the elastic matrix. Vitrimers are dynamically crosslinked polymers that can be reshaped at high temperatures and offer valuable features such as self-healing and shape memory. The dynamic mechanical properties of the material can be tailored for specific applications.

In this work, we present a magnetic elastomer containing $Co_xFe_{3-x}O_4$ MNPs ($x = 0.42$–0.49) of different sizes (Figure 1a). The dynamic nature of the materials is provided by the vinylogous urethane bonds formed upon mixing SxAcAc (with x denoting the mean number of acetoacetate groups) with the commercially available trifunctional amine Jeffamine T403. This allows network reshuffling without a catalyst at temperatures above 40 °C. The resulting composites can be molded into the desired shape and directly used. More complex structures can be fabricated by welding the pure matrix material and the nanocomposite, yielding defect-free structures with enhanced deformation behavior. Moreover, the magnetic properties and thermoreversibility can be combined using alternating magnetic fields (AMF) to induce localized heating and trigger self-healing or reshaping of the material (Figure 1b). For example, a composite containing 8 nm MNPs exposed to an AMF (36 mT, 142.70 kHz) reaches temperatures up to 112 °C (Figures 1c,d). The recyclability of the material was also investigated, demonstrating full mechanical reprocessability without magnetic particle segregation via heat compression. Overall, we demonstrate that magnetic vitrimer elastomers combine rapid magnetic actuation with shape memory, self-healing, and recyclability, all remotely activated by AMF-induced heating.

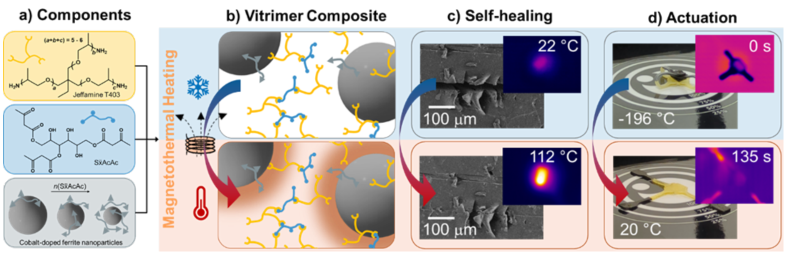

Figure 1: (a) Schematic of the individual system components and (b) the resulting magnetic vitrimer nanocomposite, whose dynamic nature is activated magnetothermally by heating above the glass-transition temperature using an alternating magnetic field. The temperature increase of up to 112 °C (36 mT, 142.70 kHz) enables (c) self-healing and (d) reshaping of the composites. Reshaping is achieved approximately three times faster than without AMF.

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