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Self-healing polymers for sustainable soft robots

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Abstract— **Soft robots, inspired by biological systems, often face vulnerability to damage. To address this, our Brubotics group has pioneered the integration of self-healing polymers, enabling soft robots to autonomously recover from cuts, punctures, and tears multiple times. These polymers, based on reversible Diels-Alder reactions, have evolved from coatings to structural components with enhanced mechanical properties. Our research highlights a trade-off between mechanical strength and the intensity or duration of required healing stimuli, influencing design choices for soft robots capable of autonomous recovery. Innovations include self-healing robotic structures like a bionic hand capable of ambient healing and grippers with integrated stimulus (heating) systems. Additionally, our work extends to multi-material interfaces fortified by strong covalent Diels-Alder bonds, enhancing robustness without the need for adhesives, and to self-healing stretchable sensors and heaters directly embedded in soft robots. These advancements, coupled with novel additive manufacturing techniques and advanced modeling capabilities, underscore a transformative shift towards more sustainable and economically viable soft robotics.**

I. AUTONOMOUS SELF-HEALING IN SOFT ROBOTS

Advancements over the past decade in self-healing polymers, particularly elastomers, have led to simultaneous improvements in mechanical and healing properties. These have enabled the transition from coatings to applications where self-healing polymers are now used as structural components. In this context, our Brubotics group has pioneered the use of self-healing polymers in soft robots [1], where all soft body parts can fully recover from sustained damage multiple times. This provides an answer to the vulnerability of these systems toward external (cuts, punctures and tears) and internal (overloading, delamination and fatigue) damages. The materials are reversible polymers, which self-healing relies on reversible covalent Diels-Alder reaction between maleimide and furan. In the following years, a wide variety of other self-healing polymers were introduced into soft robotic parts and systems. In our review paper [2], we collected these, along with other promising self-healing polymer candidates with exceptional potential for use in soft robotics. In this review, we critically assessed and compared self-healing technologies using newly defined performance parameters, including mechanical strength, stimulus, healing time, and healing efficiency. We also evaluated their integration into robotics and mechatronics systems based on criteria such as the ability to repeatedly heal macroscopic damages, recovery of properties, high-strength elastomers, reprocessability, and recyclability. From this, it can be concluded that a general trade-off exists between the mechanical strength and stability of the elastomeric network and the intensity or duration of the healing stimulus required. This leads to soft robots capable of autonomous healing but with restricted force and power output due to their hyper-elastic bodies as well as those with superior mechanical performance that rely on thermal or light stimuli for healing. These clear limitations pushed our material level research, striving towards superior self-healing polymers. By strategically designing polymer network structures, especially by incorporating an excess of furan moieties, we enhanced the reaction rates of the self-healing process [3]. This led to an autonomous healing in a soft bionic hand [4] at ambient conditions without any stimulus, while combining mechanical strength and highly efficient healing. The complex 3D robotic structures were able to fully restore their functional performance even after being cut completely in two parts. Alternatively, we have integrated stimulus-providing systems, e.g. heaters, into a self-healing soft gripper based on granular jamming. Hence, autonomous recovery is achieved by utilizing non-autonomous self-healing polymers that offer higher stiffness and strength, while enhancing the control over the healing process. To autonomously heal large gaping damages, we embedded shape memory alloy wires that simultaneously provide heat to accelerate healing as well as contraction that leads to self-closing of large damages [5].

II. ROBUST MULTI-MATERIAL INTERFACES

Via extensive research on structure-property relationships of Diels-Alder polymers, we have achieved individual control over their mechanical, processing and healing characteristics via defined parameters (functionality, stoichiometry, Diels-Alder concentration) allowing us to design these polymers to meet requirements, imposed by their application and manufacturing [6]. By varying concentration, stiff polymers with moduli in the GPa range can be achieved as well as hyperflexible polymers with moduli in the 100 kPa range [6]. Regardless of their properties, all these Diels-Alder polymers can be covalently bind together through a heat-cool cycle, creating strong multi-material interfaces without the need of adhesives [7]. This property, unique for reversible covalent polymers, is of particular interest in soft robotics in which performance is often enhanced by multi-material design but comes with a loss in robustness as delamination occurs at their multi-material interfaces that are traditionally bonded with adhesives, relying on weak secondary interactions. Therefore, the utilization of Diels-Alder polymers not only extends

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lifetime of soft robots [7] and suction cups [8] via healing but also enhances their overall robustness.

III. SELF-HEALING STRETCHABLE ELECTRONICS

Through compounding our Diels-Alder polymers with fillers, we created self-healing composites with additional functionalities. By adding a hybrid filler of carbon black and clay, the self-healing capability was combined with an electrical conductivity [9]. This composite was explored by fabricating it into a self-healing heater which was embedded in a self-healing actuator [10]. When damaged, a current flow generate heat locally at the damaged site, via an intrinsic physical intelligence, accelerating the self-healing which allows to recover the full performance of both the actuator and the heater. In many soft robotics, for proprioceptive (strain, deformation) and exteroceptive (force, contact) sensing is achieved via embedded sensors. However, as these need to be flexible and stretchable, in order to not compromise the soft robotics functionality, they are susceptible to damage. Therefore, our self-healing composite was leveraged to create self-healing stretchable sensors with various sensing capabilities (e.g. piezoresistive strain and capacitive), embedded in self-healing soft robots [11]. Aside from recovering their force or deformation sensing after multiple damage-healing cycles, the sensors allow to detect damage as well. However, creating analytical and numerical models for these sensors is challenging due to their highly non-linear and time dependent behaviour that results from the visco-elasticity of the Diels-Alder polymer. Nevertheless, we have demonstrated that machine learning can be used to calibrate stretchable electronic skins containing multiple self-healing sensors, enabling both touch and damage localization [12].

IV. MODELING OF SELF-HEALING POLYMERS IN SOFT ROBOTS

Advances in modelling is required over the complete value chain, from material and product design over processing to robotic applications. Via reaction kinetics modelling the composition of the self-healing polymers and the resulting mechanical, rheological and healing properties can be simulated as function of time and temperature using an in-house VUB-software called MATKIN. By fitting constitutive models on the mechanical properties of our Diels-Alder polymers, the kinematic and dynamic performance of our self-healing soft robots can be modelled via finite element method (FEM) in SOFA [15]. This software holds potential for real-time modelling of soft robots for control purposes.

V. EXPLOITING SELF-HEALING IN MANUFACTURING

In our review paper "Processing of self-healing polymers for soft robotics" [13], we investigated and validated the potential of processing techniques present in the literature for manufacturing self-healing soft robots. To go beyond the state of art on the manufacturing level and to respond to different industrial needs, we developed dedicated processing techniques to make healable soft robots, including additive manufacturing/3D printing [14], compression moulding [15], casting, laser cutting and welding and novel assembly & binding techniques [1]. These processes are generally not accessible to traditional irreversibly crosslinked polymers, and certainly not for their reprocessing. The self-healing polymers can be reprocessed thanks to the reversible nature of their covalent crosslinks. Material-extrusion additive manufacturing was used to fabricate complex self-healing robotics structures [14]. Whereas traditional filaments result in high anisotropy due to insufficient inter-layer bonding during printing, FFF and FGF with our self-healing polymers leads to exceptional isotropy due to inter-layer covalent binding via strong Diels-Alder bonds and due to their exceptional low viscosity in their liquid phase [14]. In addition, these polymers are completely reprocessable, and can therefore be recycled for multiple times [1].

VI. CONCLUSION

Our advancements in self-healing polymers, their composites, and additive manufacturing have enabled us to create soft robots with embedded sensors that can fully recover from damage multiple times. Additionally, the integration of damage detection, stimuli, and healing assistant systems has achieved full autonomy in the healing process, making this approach not only ecologically sustainable but also economically viable. Consequently, the development of these advanced functional materials has significantly disrupted the soft robotics research field. Their successful implementation has synergistically propelled innovation in robotics, self-healing materials, and their processing and manufacturing.

Supplementary Video: https://youtu.be/fQ4t5GcS0_g

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