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DOCTOR OF ENGINEERING SCIENCES

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The public defense will take place on **Friday 29th November 2024 at 4pm** in room **I.0.02** (Building I, VUB Main Campus)

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MORPHOLOGICAL INVESTIGATION OF SELF-HEALING POLYMER COMPOSITES AND BLENDS FOR DEFORMATION AND DAMAGE SENSING APPLICATIONS

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Abstract of the PhD research

The stretchable electronics used in soft robotics represent exciting frontiers offering significant advantages across various applications. These innovative technologies are designed to be flexible, adaptable, and resilient, which allows for integration into environments and devices that require dynamic movement and interaction. However, the susceptibility of the soft substrates to damage is detrimental and their failure has catastrophic consequences. Therefore, the reliability of products can significantly be improved by utilizing self-healing materials that repair damages before they grow into larger issues. This research focuses on the use of self-healing polymers based on a thermo-reversible Diels-Alder reaction between furan and maleimide reacting groups. These dynamic covalent bonds create reversibly crosslinked networks where thermal reversibility not only enables the reprocessing and recycling of the network but also allows the polymer network structure to recover its properties favoring sustainable manufacturing. This work aims to develop self-healing formulations for deformation and damage-sensing applications that revolve around reversible Diels-Alder polymers. With the central point of structure-property-application relationships and with emphasis on the influence of the network design parameters Diels-Alder-based polymer composites or polymer blends were synthesized. As a first attempt by meticulously integrating hybrid fillers including carbon black particles and nanoclay platelets into the Diels-Alder network, self-healing electrically conductive composites were developed that synergistically benefit from enhanced electrical and self-healing properties. Hence, the self-healing conductive composite was used as a conducting component to fabricate self-healing strain sensors with decent electro-mechanical performance. These sensors could recover their properties after damage-healing cycles. To explore other potentials of these Diels-Alder networks and to benefit from high electrical conductivity combined with superior stretchability, in the second attempt a liquid metal was chosen as the conducting component of the strain sensor. In this case, the challenge was to preserve the shape of the liquid metal and avoid its oxidation. The flowability of the liquid metal, which serves as an inherent self-healing behavior combined with the stretchable and self-healing encapsulant provided a route to the fabrication of self-healing liquid metal-based strain sensors. Since soft polymers suffer from poor barrier properties against water and oxygen a blending procedure was considered as a solution. Two dynamic covalent networks based on the Diels-Alder reaction were blended to exploit the properties of the immiscible polymer backbones. By relying on the hydrophobicity of two chemistries and with choosing the right blend/network design parameters phase-separated morphologies evolved that compromised the trade-off between the mechanical flexibility and barrier properties of the polymer encapsulant. This compromise could successfully pave the way to fabricate liquid metal-based strain sensors that show decent sensitivity and linearity in the electro-mechanical response. Moreover, owing to the special design of the sensor its electrical response with almost no hysteresis was independent of the polymer network dynamics. This addressed the hysteresis of the sensor arising from the viscoelasticity of the polymers. These findings indicated that the Diels-Alder-based self-healing polymers can be effectively engineered to serve as substrates for conductive materials, offering promising potential for the future of wearable technologies, stretchable electronic devices, and soft robotic systems.