VRIJE UNIVERSITEIT BRUSSEL

The faculty of Engineering of the Vrije Universiteit Brussel invites you to attend the public defense leading to the degree of

DOCTOR OF ENGINEERING SCIENCES

of **Aleix Costa Cornellà**

The public defense will take place on **Thursday 3rd October 2024 at 4:00 pm** in room **D.2.01** (Building D, VUB Main Campus)

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SUSTAINABILITY OF DYNAMIC POLYMER NETWORKS FROM A HOLISTIC POINT OF VIEW: FROM GREEN CHEMISTRY TO SELF-HEALING SOFT ROBOTS AND SENSORS

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Abstract of the PhD research

Polymer networks featuring dynamic bonds offer a promising solution to address key challenges in polymer materials, especially in terms of sustainability. The dynamic covalent chemistries enhance processability, enable recycling of the reversibly crosslinked thermosets and elastomers, and offer repairability. Over the last 10 years, the maturity of these networks has exponentially increased. However, they still haven't made it into the mass consumer market. To stand a chance in the competitive polymer industry, dynamic polymer networks need to further exploit their advantageous properties and enhanced performance in combination with a focus on sustainability. This overarching objective has been divided in three stages, where the results of each stage were sequentially added to the next one. The first main focus is sustainability in all its aspects. Dynamic covalent polymer networks with self-healing properties were developed using the 12 principles of green chemistry as the designing driving force. The sustainability was tackled from a holistic point of view, taking into account the whole life cycle of the material (circular economy). The developed self-healing elastomers account for the sustainability of (1) the raw materials and (2) solvents, (3) the reusability, (4) the (re)processability, and (5) the end of life of the material. They are synthesized by a simple one-pot, solventless synthesis from commercially available reagents, they can be reprocessed and recycled, they autonomously heal at room temperature and they can be hydrolytically degraded at the end of their service life. The objective of the second stage was to enhance the performance of dynamic covalent networks.

Starting from the materials and the synthesis processes developed in the first stage, the trade-off between self-healing/reprocessability and creep resistance was minimized. Solving this trade-off has been a priority in the field, and it is currently the main performance challenge of dynamic polymer networks. This was achieved by combining a dissociative and an associative dynamic covalent chemistry in the same polymer network. This combination enables the fabrication of materials where the timescale at which they relax can be designed independently from its mechanical properties and tailored for the application and lifetime of the material. Finally, these materials were used in several applications where the dynamics bonds bring a performance advantage over traditional polymers. The materials were used in soft robotics, smart textiles, electronic skin, and 3D printing, proving its applicability in key technologies for the future without giving up on sustainability.