

# **Design, synthesis and characterization of novel depolymerizable polydienes with intrinsically-weakened carbon-carbon bonds**

## **Abstract:**

The widespread use of plastics has brought both convenience and environmental challenges, particularly due to the accumulation of single-use plastics. Traditional recycling methods are often inefficient, expensive, and energy-intensive, leading to low recovery rates and poor selectivity of virgin materials. To address this, new strategies focus on designing recyclable polymers that can support closed-loop systems. However, practical chemical recycling solutions remain limited by the lack of cost-effective and scalable recycling methods and material designs.

In the first study, a circularly recyclable polymers based on poly(muconate) (polyME) synthesized via free-radical polymerization is developed. These materials exhibit tunable mechanical properties and depolymerize rapidly at 258 °C due to weakened carbon-carbon backbones. By varying side-chain functionalities and copolymer compositions, the mechanical properties of these polymers can be tuned to match commercial plastics. A comprehensive techno-economic and life cycle analysis highlights their environmental and economic advantages compared with commercial thermoplastics. When recycled content is reused, both cost and ecological impact are significantly reduced.

The second study addresses the more difficult challenge of recycling crosslinked rubbers, which are typically non-recyclable due to permanent covalent bonds formed during vulcanization. Using bio-based polyME, a new class of recyclable thermosets is developed. These materials undergo traditional polymerization followed by side-chain modification, introducing terminal vinyl groups that enable vulcanization while maintaining the core polymer structure. This design allows for thermal depolymerization of the crosslinked network, enabling recovery of both monomers and oligomers. The resulting thermosets, known as PolyME-BuPe, exhibit strong mechanical performance, compatibility with industrial fillers, and processability with existing rubber manufacturing techniques. This work demonstrates a scalable, modular pathway to integrate recyclable thermosets into current production systems and marks a substantial step toward achieving a circular economy for rubber and polymer materials.