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ABSTRACT:

High-Performance Next-Generation Thermoset Matrices: From Bio-Based Systems to 3R Materials (Recyclability, Repairability, and Reprocessability)

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The development of sustainable thermosetting polymers remains a major scientific and technological challenge due to their intrinsic crosslinked structure, which traditionally prevents recyclability and reprocessability. In this work, a comprehensive strategy toward next-generation thermoset matrices is presented, combining high-performance bio-based chemistry with circularity concepts, namely recyclability, repairability, and reprocessability (3R). Fist stage consisted of developing bio-based epoxy thermosets derived from epoxidized resveratrol (RESEP), a trifunctional aromatic monomer. These materials exhibit outstanding thermomechanical performance, reaching ultra-high glass transition temperatures above 300 °C, surpassing conventional petroleum-based epoxy systems. In some cases, the materials approach thermal degradation before achieving a classical rubbery plateau, highlighting their exceptional crosslink density. Additionally, they display excellent thermal stability, high residual char formation, and remarkable electrical insulation properties, together with tunable optical behavior.

The second stage focuses on the development of high-performance structural composites by incorporating inorganic reinforcements such as glass and carbon fibers and multifunctional capabilities by the addition of discontinuous conductive fillers, such as carbon nanotubes and recycled short carbon fibers.

Finally, a paradigm shift toward circular thermosets is achieved through the introduction of covalent adaptable networks (CANs) based on dynamic imine bonds. These reversible linkages enable bond exchange reactions, allowing the materials to be repaired, reprocessed, and recycled without significant loss of performance. In parallel, the incorporation of mechanically recycled short carbon fibers introduces multifunctionality, including electrical conductivity, Joule heating capability, and structural health monitoring. The resulting hybrid systems combine high thermal and mechanical performance with adaptive and functional behavior