Catalyst-free PLG based covalent adaptable networks

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Plastic waste recycling remains a significant challenge for modern societies. While thermoplastics can be recycled relatively easily, thermoset materials cannot. These crosslinked materials are insoluble and unmeltable, making their recycling possible only through methods such as mechanical crushing to obtain filler materials, incineration for energy recovery, pyrolysis to recover crude hydrocarbon mixtures, or disposal in landfills. However, these processes have major drawbacks, including significant environmental pollution such as high CO_2 emissions, as well as being highly energy- and cost-intensive.
To address this issue, a new class of polymers has emerged, aiming to combine the reprocessability of thermoplastics with the superior properties and stability of thermosets. These are referred to as covalent adaptable networks (CANs).[1] In general, bond exchange reactions during processing of thermosets require an appropriate catalysts, often metal-organic compounds, sometimes leading to the problem of leaching and their general toxicity.[2]
Here, we present catalyst-free poly(*DL*-lactide-*co*-glycolide) (PLG)-based covalent adaptable networks that can be easily processed using compression molding.[3] Once processed, the networks are insoluble and exhibit a high gel content. The resulting material demonstrates a high breaking force during tensile testing and can be reshaped easily when heated above the glass transition temperature. Reversibility is introduced via the linker pyromellitic dianhydride, which exists in two temperature-dependent states: open (diester) and closed (bisanhydride). Furthermore, the polymer network is rapidly hydrolyzable under basic conditions at room temperature, enabling accessible chemical recycling or degradation into lactic acid and glycolic acid.

Keywords: Covalent Adaptable Networks, Vitrimers, Crosslinked Polymers, Thermoset Recycling

References

[1] Kloxin, C.J.; Bowman, C.N.; Covalent adaptable networks: smart, reconfigurable and responsive network systems. *Chem. Soc. Rev.* **2013**, 42, 7161.
[2] Schwarzer, L.; Agarwal, S.; Adaptable Polyurethane Networks Containing Tertiary Amines as Intrinsic Bond Exchange Catalyst. *Macromol. Chem. Phys*. **2024**, 2400072.
[3] Eger, E.M.; Agarwal, S.; Poly(lactic-*co*-glycolic acid) Networks with Dynamic Covalent Bonds: Synthesis and Characterization. *ACS Apl. Polym. Mater.* **2023**, 5, 5141-5148.