A RETROBIOSYNTHETIC APPROACH TO GENERATE TERPENE-DERIVED POLYMERS

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With an increasing global population and an enhanced awareness of the impact of human activities on our environment and climate, innovative concepts for resource efficiency are urgently needed.[1] This is especially relevant for synthetic polymers, that today are mainly generated from petroleum. In fact, at present only 1% of all man-made synthetic materials are generated from renewable resources, which is not aligned with the global vision of a circular bioeconomy. Upgrading bio-based molecules from wood and other sustainable raw material into activated monomers to generate biopolymers is thus a contemporary research area.[1-3]

Herein, a novel retrobiosynthetic approach[4] to enable biopolymer generation by chemoenzymatic catalysis is presented (Figure 1). By employing a retrobiosynthetic analysis applied to polyesters (Figure 1A), and capitalizing on synthetic biology in concert with polymer chemistry, my research group has recently successfully generated unprecedented biopolymers with interesting thermal properties (Figure 1B).[4,5]

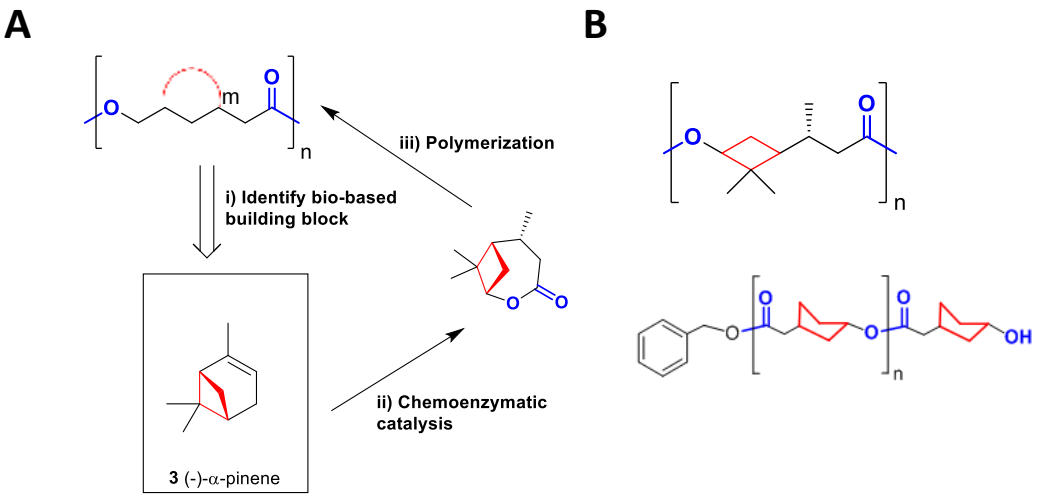

Figure 1. A) Retrobiosynthetic analysis for polyesters harboring rings as backbone motifs enabled biocatalytic formation of green caprolactone-analogues and corresponding terpene-derived polyesters.[4] B) Examples of novel materials generated in my group by polymer retrobiosynthesis (Stamm et al.[4] and Farhat et al.[5]).

The forefront in sustainability science lays in the development of biocatalytic processes based on engineered enzymes, to enable mild generation of biofuels, biomaterials and fine chemical synthons from renewable resources.[2] In particular, terpenoids and other multicyclic scaffolds represent a rich and untapped source of chiral biochemicals and biopolymers.[3] However, the inert backbones towards controlled polymerization found in many terpene-based building blocks hamper their full potential in material science. By merging biocatalysis, enzyme engineering and synthetic biology mild valorization of renewable synthons into desired monomer structure can be achieved with high-precision. Recent results from my research group on design and engineering of oxidoreductases in concert with polymer chemistry to enable access to advanced green functionalized terpene-derived materials by polymer retrobiosynthesis will be discussed.

References

[1] X. Zhang, M. Fevre, G. O. Jones, R. M. Waymouth, *Chem. Rev.* 2018, *118*, 839-885.
[2] M. Hoenig, P. Sondermann, N. J. Turner, E. M. Carreira, *Angew. Chem., Int. Ed.* 2017, *56*, 8942-8973.
[3] Y. Zhu, C. Romain, C. K. Williams, *Nature* 2016, *540*, 354-362.
[4] A. Stamm, A. Biundo, B. Schmidt, J. Brücher, S. Lundmark, P. Olsén, L. Fogelström, E. Malmström, U. T. Bornscheuer, P.-O. Syrén, *ChemBioChem* 2019, DOI: 10.1002/cbic.201900046.
[5] W. Farhat, A. Stamm, M. Robert-Monpate, A. Biundo, P.-O. Syrén. *Z. Naturforsch. C.* 2019, *74*, 91–100.