Inaugural Open Plastic Lecture

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Recent Progress in the Enzymatic Degradation of Plastics and Especially PET

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This lecture will cover recent achievements in the protein engineering of enzymes for the degradation of polyethylene terephthalate and other chemical polymers. These are important strategies to cope with the environmental pollution caused by PET as well as other polymers as summarized in our recent reviews [1].

The first enzymes able to hydrolyze the polymer PET were described more than 15 years ago, which include lipases, but also cutinases. These have been the scaffolds serving as starting points for numerous enzyme engineering studies resulting finally in improved biocatalysts suitable for large-scale processing of PET as demonstrated by a team of French scientists [2]. Another milestone has been the discovery of the natural microorganism *Ideonella sakaiensis* by a team of Japanese researchers [3] who identified this strain at a plastic waste disposal site. Interestingly, this bacterium secretes a PETase to hydrolyse the polymer into small units, which are taken up by the cell to be finally degraded by an intracellular MHETase, which releases terephthalic acid and ethylene glycol. *I. sakaiensis* then can grow on these monomers. Keeping in mind that PET was first synthesized in 1941 by organic chemists, this is an interesting example how natural evolution can create enzymes able to degrade this man-made non-natural polymer [4]. We determined the first structure of the MHETase in complex with a substrate analogue [5] and also provided important adjustments of a published PETase structure [6]. We also used various methods of protein engineering [7] including rational design and directed evolution to improve several PET-hydrolases for improved activity and thermostability [8].

In addition to PET, we also investigate the enzymatic conversion of much more challenging polymers such as polyethylene where full degradation requires a cocktail of enzymes from various classes to convert these highly recalcitrant compounds into valuable building blocks.

[1] Wei, R. et al. (2022), *ACS Catal*., **12**, 3382-3396 (2022); Tiso, T. et al. *Metabol. Eng*.**, 71**, 77-98 (2022); Ballerstedt, H. et al., *Environ. Sci. Eur*., **33**, 99 (2021); Jönsson, C. et al. *ChemSusChem*, **14**, 4028-4040 (2021); Wei, R. et al., *Nature Catal.*, **3**, 867-871 (2020).
[2] Tournier, V. et al., *Nature* **580**, 216-219 (2020)
[3] Yoshida, S. et al., *Science* **351**, 1196-1199 (2016)
[4] Bornscheuer, U.T., *Science*, **351**, 1155-1156 (2016)
[5] Palm, G.J. et al., *Nature Commun*., 10, 1717 (2019)
[6] Wei, R. et al., *Nature Commun*., **10**, 558 (2019)
[7] Bornscheuer, U.T. et al., *Nature*, **485**, 185-194 (2012)
[8] Shuai, L. et al., *Biochem. Biophys. Res. Commun*., **626**, 100-106 (2022); Pfaff, L. et al., *ACS Catal.*, **12**, 9790-9800 (2022); Brott, S. et al., *Eng. Life Sci.*, **22**, 192-203 (2022)