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Grant Title: Synthetic Glycan-Processing Catalysts for Modern Glycomics

Abstract

Carbohydrates are involved in numerous biological processes and all major human diseases. They form ~75% of entire biomass and provide a carbon-neutral and potentially sustainable source of energy. The main objective of this grant is to develop a new class of biomimetic catalysts to process carbohydrates through selective hydrolysis. The catalysts are inspired by natural glycosidase and constructed through molecular imprinting in doubly cross-linked surfactant micelles. The catalytic groups are introduced through functional monomers used during the imprinting process or via postfunctionalization of the imprinted sites by chemical reactions. The resulting molecularly imprinted nanoparticles (MINPs) resemble enzymes in size, a hydrophilic exterior, a hydrophobic interior, and custom-tailored active sites in the micellar core. Both natural and unnatural catalytic motifs can be introduced. MINP catalysts have been shown to catalyze the cleavage of alkyl glycosides and cellulose under mildly acidic or neutral conditions. An acid–base-functionalized catalyst in 60 °C water outperformed some natural enzymes (e.g., digestive β-glucosidase GH1 from *Spodoptera frugiperda* at 30 °C in pH 6 buffer) in cellobiose hydrolysis, with a rate acceleration of 10^{10}. The catalytic proficiency reached 10^{15} M^{-1}, corresponding to 24.5 kcal/mol of affinity for the transition state.