

Phosphorylase-catalyzed bottom-up synthesis of cellooligosaccharides and property-tunable materials

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Cellulose-based materials are produced industrially via top-down processing from natural lignocellulose biomass, while challenges involved in this approach has promoted the development of bottom-up synthesis. Building up a cellulose chain via bottom-up approach is promising since it offers higher synthetic precision than the top-down processing.[1]

Glycoside phosphorylases (GPs) are carbohydrate-active enzymes involved in the formation/cleavage of glycosidic bond. The primarily relevant GP for biosynthesis of cellulose is cellodextrin phosphorylase (CdP, EC 2.4.1.49). It catalyses iterative β -1,4-glycosylation from α -D-glucose 1-phosphate to elongate a diversity of acceptors including cellobiose and a range of synthetic glycosides with non-sugar aglycons.1 Due to iterative glycosylation leading to different degrees of polymerization (DP), soluble cello-oligosaccharides (COS) or insoluble cellulosic materials are formed.[2] Here, we present an approach using CdP as bio-catalyst for the bottom-up synthesis of functional cellulose materials. The synthetized soluble COS (DP \leq 6) exhibited a selective prebiotic effect.[3] The synthetic cellulose with chains DP \geq 9 form as insoluble materials in a sheet-like crystalline structures of cellulose allomorph II. Reducing-end functionalized (e.g., thiol-) materials thus obtained can expand cellulose applications towards the fields that are difficult to access via top-down approach.[4,5] With solvent condition set, bottom-up synthesis also enables the possibility to generate composite materials which can be useful as functional hydrogels.[6]





Glycosylation and oligosaccharide synthesis / Biosynthesis and Carbohydrate Active Enzymes / Enzymatic synthesis and biocatalysis