24th World Chemistry & Systems Biology Conference

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4th International Conference on **Biochemistry & Metabolomics**

October 03-04, 2018 | Los Angeles, USA

Preparation of bio-based supramolecular materials by helical assemblies from enzymatically synthesized amylose

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Polysaccharides are well-known biopolymers, which form controlled higher-order assemblies by non-covalent linkages such as hydrogen bonds. Accordingly, the construction of hierarchically assembled structures, so-called supramolecules, from polysaccharides has attracted much attention to obtain new polysaccharide-based functional materials. Amylose, which is a linear polysaccharide linked through α(1γ4)-glycosidic linkages and well-known as a component of starch, forms regularly controlled assemblies, that is, inclusion complex and double helix, depending on whether guest compounds are present or not. Amylose with well-defined structure is synthesized by phosphorylase-catalyzed enzymatic polymerization using α-D-glucose 1-phosphate (G-1-P) and α(1γ4)-oligoglucan (maltooligosaccharide) as monomer and primer, respectively. As the polymerization is initiated from the non-reducing end of the maltooligosaccharide primer, the enzymatic polymerization can be conducted using primers covalently linked to other polymeric materials (immobilized primers) at the reducing end, giving rise to amylose-grafted bio-based polymeric materials. By means of the property of spontaneously double helix formation from the enzymatically synthesized amylose, the phosphorylase-catalyzed enzymatic polymerization using the immobilized primers produces supramolecular structures comprising the double helix cross-linking points. For example, the phosphorylase-catalyzed enzymatic polymerization using the immobilized primers on chitin nanofibers was investigated to produce amylose-grafted chitin nanofiber hydrogels.

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