

Design and Synthesis of Polymers for Corona Phase Molecular Recognition (CoPhMoRe) of Carbohydrates

by

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ABSTRACT

The molecular recognition of carbohydrates is difficult to realize synthetically due to their relatively low affinity for a wide range of substrates, yet this recognition is the underpinning of human immunity, cell signaling, and glycobiology. For the past decade, significant effort has been made in this field to create new technologies to profile glycans and carbohydrates.

Corona Phase Molecular Recognition (CoPhMoRe), the concept introduced from Strano group, generates a nanoparticle coupled polymer phase capable of recognizing a specific molecule with high affinity and selectivity. CoPhMoRe has been successfully demonstrated using polymer wrapped single walled carbon nanotubes, resulting in molecular recognition complexes, to date, for dopamine, estradiol, riboflavin, L-thyroxine, and the protein fibrinogen, utilizing combinatorial library screening. As an alternative to this empirical, library screening, we first solve the mathematical formulation that we introduce as the CoPhMoRe inverse problem to provide a theoretical basis for understanding certain types of CoPhMoRe recognition.

In addition, we demonstrate that a polymer or surfactant corona phase surrounding a single walled carbon nanotube can substantially modify the selectivity of various pre-adsorbed phenylboronic acids (PBA) for mono-, di- and polysaccharides. Based on these findings, a simple and robust RAFT polymerization process is employed to produce novel and distinct classes of water-soluble PBA-based polymers. These polymers in SWNT corona phases demonstrate enhanced selectivity towards specific sugar alcohols, which differ only in the orientation of the hydroxyl groups. By changing the polymer backbone structure, highly selective D-Arabinose sensor is developed and used to differentiate D-Arabinose from L-Arabinose for the first time. Finally we develop a glucose sensor that can measure glucose concentration instantaneously by detecting changes in local refractive index.

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