

Biosensing and Supramolecular Bioconjugation in Single Conical Polymer Nanochannels. Facile Incorporation of Biorecognition Elements into Nanoconfined Geometries

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Journal of the American Chemistry Society, 130 (48) (2008) 16351-16357

There is a growing quest for tailorable nanochannels or nanopores having dimensions comparable to the size of biological molecules and mimicking the function of biological ion channels. This interest is based on the use of nanochannels as extremely sensitive single molecule biosensors. The biosensing capabilities of these nanochannels depend sensitively on the surface characteristics of their inner walls to achieve the desired functionality of the biomimetic system. Nanoscale control over the surface properties of the nanochannel plays a crucial role in the biosensing performance due to the chemical groups incorporated on the inner channel walls that act as binding sites for different analytes and interact with molecules passing through the channel. Here we report a new approach to incorporate biosensing elements into polymer nanochannels by using electrostatic self-assembly. We describe a facile strategy based on the use of bifunctional macromolecular ligands to electrostatically assemble biorecognition sites into the nanochannel wall, which can then be used as recognition elements for constructing a nanobiosensor. The experimental results demonstrate that the ligand-functionalized nanochannels are very stable and the biorecognition event (protein conjugation) does not promote the removal of the ligands from the channel surface. In addition, control experiments indicated that the electrostatically assembled nanochannel surface displays good biospecificity and nonfouling properties. Then, we demonstrate that this approach also enables the creation of supramolecular multilayered structures inside the nanopore that are stabilized by strong ligand-receptor interactions. We envision that the formation of multilayered supramolecular assemblies inside solid-state nanochannels will play a key role in the further expansion of the toolbox called "soft nanotechnology", as well as in the construction of new multifunctional biomimetic systems.