Surface self-assembled hybrid nanocomposites with electroactive nanoparticles and enzymes confined in a polymer matrix for controlled electrocatalysis

A three-dimensional network of highly branched poly(ethyleneimine) (PEI) is designed and synthesized on gold electrode surfaces. A self-assembled monolayer (SAM) of dithiobis(succinimidyl propionate) (DTSP) on a gold electrode was first prepared, which is confirmed by the reductive desorption of Au-S units. The PEI polymer was then covalently immobilized onto the DTSP layer, leaving free primary amine groups acting as a 3D skeleton for high loading of electroactive enzyme-size Prussian blue nanoparticles (PBNPs, 6 nm) via electrostatic trapping. Atomic force microscopy was used to disclose the microscopic structures of the different layers during the surface architecture formation. The resulting surface-bound nanostructured composite shows high electrochemical activity arising from confined PBNPs, and acts as an efficient electrocatalyst towards H₂O₂ reduction. Facile electron communication is achieved as reflected by a large electron transfer (ET) rate constant (kₛ) of 200 s⁻¹, and the possible electron propagation mechanisms in the polymer network are discussed. This surface/interfacial nanocomposite can be further used in the accommodation of enzymes for electrochemical bio-catalysis. Glucose oxidase (GOD) was used towards this end, in a proof-of-concept study. This enzyme can be co-trapped in the PEI matrix and is interconnected with PBNPs, leading to highly efficient electrocatalytic oxidation and detection of glucose.

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