Gold nanoparticle assisted assembly of a heme protein for enhancement of long-range interfacial electron transfer - DTU Orbit (30/11/2018)

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Interfacial electron transfer (ET) of biological macromolecules such as metalloproteins is the key process in bioelectrochemistry, enzymatic electrocatalysis, artificial ET chains, single-molecule electronic amplification and rectification, and other phenomena associated with the area of bioelectronics. A key challenge in molecular bioelectronics is to improve the efficiency of long-range charge transfer. The present work shows that this can be achieved by nanoparticle (NP) assisted assembly of cytochrome c (cyt c) on macroscopic single-crystalline electrode surfaces. We present the synthesis and characterization of water-soluble gold nanoparticles (AuNPs) with core diameter 3-4 nm and their application for the enhancement of long-range interfacial ET of a heme protein. Gold nanoparticles were electrostatically conjugated with cyt c to form nanoparticle-protein hybrid ET systems with well-defined stoichiometry. The systems were investigated in homogeneous solution and at liquid/solid interface. Conjugation of cyt c results in a small but consistent broadening of the nanoparticle plasmon band. This phenomenon can be explained in terms of long-range electronic interactions between the gold nanoparticle and the protein molecule. When the nanoparticle-protein conjugates are assembled on Au(111) surfaces, long-range interfacial ET across a physical distance of over 50 Å via the nanoparticle becomes feasible. Moreover, significant enhancement of the interfacial ET rate by more than an order of magnitude compared with that of cyt c in the absence of AuNPs is observed. AuNPs appear to serve as excellent ET relays, most likely by facilitating the electronic coupling between the protein redox center and the electrode surface.

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