
Molecularly resolved Bioelectrochemistry & Probe Directed Molecular Coupling

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I will talk about two of the research groups recent projects; both grounded in the coupling of functional surfaces with molecular-scale imaging.

Biological electron transfer -redox linked interfacial optics

The generation of functionally-active biomolecular monolayers is important in both analytical science and biophysical analyses. Our ability to monitor the redox-active state of immobilised proteins or enzymes at a molecular level, from which stochastic and surface-induced variations would be apparent, is impeded by comparatively slow electron transfer kinetics and associated signal:noise difficulties. We demonstrate herein that, by covalently tethering an appropriate dye to the copper protein azurin, a highly oxidation-state sensitive FRET process can be established which enables redox switching to be optically monitored at protein levels down to the zeptomolar limit. The surface-potential induced cycling of emission enables the redox potential of clusters of a few hundred molecules to be determined.

Catalytic nanolithography

The generation of patterned surfaces has attracted considerable attention during the past decade, due to significant and varied potential application in optoelectronics, sensory technology, and the electronics industry. The initiation and control of chemical coupling has the potential to offer much within the context of “bottom up” nanofabrication. In recent work we have reported the use of a palladium-modified, catalytically active, AFM probe to initiate and spatially control surface-confined Suzuki and Heck carbon-carbon coupling reactions. These “chemically written reactions”, detectable by lateral force and chemically specific optical and topographic labeling, were patterned with line widths down to 15 nm or ~20 molecules. Catalyzed organometallic coupling was, in this way, carried out at subzeptomolar levels. By varying the catalyst substrate interaction times, turnover numbers of $(0.6-1.2) \times 10^4$ and $(3.0-5.0) \times 10^4$ molecules s^{-1} were resolved for Suzuki and Heck reactions, respectively.

Jason Davis (b. 1971) studied Chemistry at Kings College London, where he was awarded The Victor Gold Prize for Chemistry in 1991, The Ivor John Prize for Organic Chemistry in 1992, and The Robert Wakeford Memorial Prize in Chemistry and a first class honours degree in 1993. He moved to the Inorganic Chemistry Laboratory at the University of Oxford in 1994. After obtaining a PhD and postdoctoral research on carbon nanotubes, electroanalysis and scanning probe microscopy he was elected to an Extraordinary Junior Research Fellowship at The Queens College in 1998, a Ramsey Fellowship and a Royal Society University Research Fellowship in 1999 and a Lectureship in Chemistry at Jesus College, Oxford, in 2001. He was made a University Lecturer and Official Student and Tutor in Chemistry at Christ Church in 2003. His work has focused on the molecular and nanometre-scale construction and analysis of bioinorganic, sensory, electronic and optical systems. He has published over 60 papers on carbon nanotubes, nanoparticles, biological and molecular sensing, biomolecular electronics and nanotechnology.