



Scanning Electrochemical Microscopy of DNA Monolayers Modified with Nile Blue

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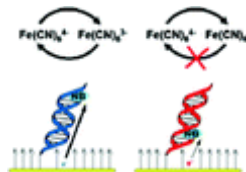
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Abstract



Scanning electrochemical microscopy (SECM) is used to probe long-range charge transport (CT) through DNA monolayers containing the redox-active Nile Blue (NB) intercalator covalently affixed at a specific location in the DNA film. At substrate potentials negative of the formal potential of covalently attached NB, the electrocatalytic reduction of $\text{Fe}(\text{CN})_6^{3-}$ generated at the SECM tip is observed only when NB is located at the DNA/solution interface; for DNA films

containing NB in close proximity to the DNA/electrode interface, the electrocatalytic effect is absent. This behavior is consistent with both rapid DNA-mediated CT between the NB intercalator and the gold electrode as well as a rate-limiting electron transfer between NB and the solution phase $\text{Fe}(\text{CN})_6^{3-}$. The DNA-mediated nature of the catalytic cycle is confirmed through sequence-specific and localized detection of attomoles of TATA-binding protein, a transcription factor that severely distorts DNA upon binding. Importantly, the strategy outlined here is general and allows for the local investigation of the surface characteristics of DNA monolayers both in the absence and in the presence of DNA binding proteins. These experiments highlight the utility of DNA-modified electrodes as versatile platforms for SECM detection schemes that take advantage of CT mediated by the DNA base pair stack.