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Simultaneous Spectroscopic and Topographic Imaging of Single-Molecule Interfacial Electron Transfer Reactivity and Local Nanoscale Environment[1] YUFAN HE, VISHAL RAO, JIN CAO, PETER LU, Bowling Green State University, LU GROUP TEAM — The fundamental information related to the energy flow between molecules and substrate surfaces as a function of surface site geometry and molecular structure are critical for understanding interfacial electron transfer (ET) dynamics. The inhomogeneous nanoscale molecule-surface and molecule-molecule interactions are presumably the origins of the complexity in interfacial ET dynamics, thus, identifying the environment of molecules at nanoscale is crucial. We have developed AFM correlated single-molecule fluorescence intensity/lifetime imaging microscopy (AFM-SMFLIM) capable of identifying and characterizing individual molecules distributed across the heterogeneous surface at nanometer length scale. Using the combined AFM-SMFLIM imaging, we are able to obtain nanoscale morphology and interfacial ET dynamics at single-molecule level. Moreover, the correlated information about the fluorescence blinking behavior of each individual dye molecule and its lifetime along with the local nano scale topography explains the inhomogeneity of coupling strength of each dye with TiO2 NPs and its effect on interfacial electron transfer reactivity intermittency. The molecular-level understanding of the interfacial ET reactivity, derived from our study, sheds light on the intrinsic fluctuating and inhomogeneous interfacial ET dynamics, which may, for example, help on the development of solar energy conversion science and photocatalysis. (J. Phys. Chem. Lett., 7,2221-2227(2016))

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