Electron transfer patterns of the di-heme protein cytochrome c(4) from Pseudomonas stutzeri - DTU Orbit (19/12/2018)

**Electron transfer patterns of the di-heme protein cytochrome c(4) from Pseudomonas stutzeri**

We report kinetic data for the two-step electron transfer (ET) oxidation and reduction of the two-domain di-heme redox protein Pseudomonas stutzeri cytochrome (cyt) c(4) by [Co(bipy)(3)](2- 3-) (bipy = 2,2'-bipyridine). Following earlier reports, the data accord with both bi- and tri-exponential kinetics. A complete kinetic scheme includes both "cooperative" intermolecular ET between each heme group and the external reaction partner, and intramolecular ET between the two heme groups. A new data analysis scheme shows unequivocally that two-ET oxidation and reduction of P. stutzeri cyt c(4) is entirely dominated by intermolecular ET between the heme groups and the external reaction partner in the ms time range, with virtually no contribution from intramolecular interheme ET in this time range. This is in striking contrast to two-ET electrochemical oxidation or reduction of P. stutzeri cyt c(4) for which fast, ms to sub-ms intramolecular interheme ET is a crucial step. The rate constant dependence on the solvent viscosity has disclosed strong coupling to both a (set of) frictionally damped solvent/protein nuclear modes and intramolecular friction-less "ballistic" modes, indicative of notable protein structural mobility in the overall two-ET process. We suggest that conformational protein mobility blocks intramolecular interheme ET in bulk homogeneous solution but triggers opening of this gated ET channel in the electrochemical environment or in the membrane environment of natural respiratory cyt c(4) function.

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