Abstract Submitted
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Effect of DNA binding on geminate CO recombination kinetics in CooA[1] ABDELKRIM BENABBAS, VENUGOPAL KARUNAKARAN, Northeastern University, HWAN YOUN, California State University, THOMAS POULOS, University of California, Irvine, PAUL CHAMPION, Northeastern University — CooA proteins are heme-based CO-sensing transcription factors. Here we study the ultrafast dynamics of geminate CO rebinding to RrCooA. The effects of DNA binding and the truncation of the DNA binding domain on the CO geminate recombination kinetics were investigated. The CO rebinding kinetics in these CooA complexes takes place on ultrafast timescales but remains non-exponential over many decades in time. We show that this non-exponential kinetic response is due to a quenched enthalpic barrier distribution resulting from a distribution of heme geometries that is frozen or slowly evolving on the timescale of CO rebinding. We also show that, upon CO binding, the distal pocket of the heme in RrCooA relaxes to form a very efficient hydrophobic trap for CO. DNA binding further tightens the narrow distal pocket and slightly weakens the iron-proximal histidine bond. Analysis of our data reveals that the uncomplexed and inherently flexible DNA binding domain adds additional structural heterogeneity to the heme doming coordinate. When CooA forms a complex with DNA, the flexibility of the DNA-binding domain decreases and the distribution of the conformations available in the heme domain becomes restricted.

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