

Abstract Submitted
for the MAR13 Meeting of
The American Physical Society

Multistage Enzymatic Pathways of the Copper-containing Nitrite Reductase (CuNiR) YAN LI, MIROSLAV HODAK, JERRY BERNHOLC, North Carolina State University, CENTER OF HIGH PERFORMANCE SIMULATION TEAM — Copper-containing nitrite reductase (CuNiR) catalyzes the reduction of NO_2^- to NO in the global nitrogen cycle. Experimental X-ray data have provided good insight into the overall function of CuNiR. However, many important questions, such as the relevance of the conformational change of Asp⁹⁸ as well as the transformation from the O-coordination of the substrate to the N-coordination of the product remain unanswered. We present a computational study of the enzymatic mechanism of CuNiR based on density functional theory. The climbing-image nudged elastic band (CI-NEB) method is used to find the minimum energy pathways and the activation energy barriers of the reaction. Furthermore, the effects of hybrid functionals and solvent on the activation barriers are investigated. A critical residue Asp⁹⁸ is found to control the access to the binding site and to stabilize a previously reported “side-on” coordination of the nitrosyl intermediate, although this geometry does not occur during the reaction. We also find that the transformation of the O- to N-attachment is achieved by an electron transfer from Type I copper.

Yan Li
North Carolina State University

Date submitted: 12 Dec 2012

Electronic form version 1.4