

COMPUTATIONAL STUDY OF THE PROTON TRANSFER PATHWAY IN HYDROGENASE

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The photobiological generation of H₂ by the green alga offers a promising method for renewable H₂ production. The photochemical energy absorbed by the algae is converted into electrochemical energy as low potential electrons which are further transferred to the [FeFe] hydrogenase via protein ferredoxin. At the catalytic center (H-cluster) of hydrogenase, these electrons are used to reduce protons and generate H₂ gas. A potential proton transfer pathway toward the hydrogenase H-cluster has been identified, and was found to be highly conserved in [FeFe] hydrogenases. However, the details of the proton transfer process along the pathway are still little known. In this research, based on the crystal structure of hydrogenase CpI, we used classic MD and QM/MM MD simulation methods to study the proton transfer pathway in hydrogenase. We found three key residues important to the proton transfer process along the pathway. They are GLU278, CYS299, and SER318. Residue CYS299 is close to the H-cluster and functions as the final relay of the proton. Residue GLU278 can adopt two different conformations, which are stabilized by forming hydrogen bond with residue SER318. The free energy profiles of this conformation change were then obtained using “umbrella sampling” method. Our calculation results suggest that the conformation change of GLU278 is important to the function of the proton transfer pathway.