Chemical reaction processes in biological systems studied by combined ab initio quantum mechanical/molecular mechanical simulations

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Chemical reactions such as bond dissociation/formation and excited state processes are involved in many biological processes. In spite of their significance in biological functions, the electronic and atomic details have been little known due to difficulty in describing the electronic wave functions of reactive molecules in complex biological systems. In the present study, we have investigated chemical reactions in retinal proteins and F_1-ATP synthase through combined ab initio quantum mechanical/molecular mechanical (QM/MM) simulations. The methods permit one to explicitly describe the chemical reaction energetics and dynamics under electrostatic and steric influence of the protein matrix. We have carried out for the first time a multi-electronic state ab initio QM/MM molecular dynamics simulation for the chromophore's photo-isomerization dynamics in bacteriorhodopsin (bR), revealing the essential role of the protein for the characteristic kinetics and high selectivity of the photoisomerization. We have then predicted an early intermediate state structure of bR's photocycle, which has been confirmed by recent x-ray crystallography, and elucidated molecular mechanism of its photo-energy storage. We have also applied the QM/MM method to a hydrolysis reaction of ATP in F_1-ATP synthase and found a new multi-center reaction path of the hydrolysis.