Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

QTES-DFTB dynamics study on the effect of substrate motion on quantum proton transfer in soybean lipoxygenase-1[1] JAMES MAZZUCA, SOPHYA GARASHCHUK, Department of Chemistry and Biochemistry, University of South Carolina, Columbia, South Carolina 29208, JACEK JAKOWSKI, National Institute for Computational Sciences, University of Tennessee, Oak Ridge, Tennessee 37831 — It has been shown that the proton transfer in the enzymatic active site of soybean lipoxygenase-1 (SLO-1) occurs largely by a quantum tunneling mechanism. This study examined the role of local substrate vibrations on this proton tunneling reaction. We employ an approximate quantum trajectory (QT) dynamics method with linear quantum force. The electronic structure (ES) was calculated on-the-fly with a density functional tight binding (DFTB) method. This QTES-DFTB method scales linearly with number of trajectories, and the calculation of the quantum force is a small addition to the overall cost of trajectory dynamics. The active site was represented as a 44-atom system. Quantum effects were included only for the transferring proton, and substrate nuclei were treated classically. The effect of substrate vibrations was evaluated by freezing or relaxing the substrate nuclei. Trajectory calculations were performed at several temperatures ranging from 250-350 K, and rate constants were calculated through the quantum mechanical flux operator which depends on time-dependent correlation functions. It was found that the substrate motion reliably increases the rate constants, as well as the P/D kinetic isotope effect, by approximately 10% across all temperatures examined.

[1]NSF Grant No. CHE-1056188, APRA-NSF-EPS-0919436, and CHE-1048629, NICS Teragrid/Xsede TG-DMR110037

James Mazzuca
Dept of Chemistry and Biochemistry, University of South Carolina,
Columbia, South Carolina 29208

Date submitted: 13 Nov 2013 Electronic form version 1.4