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U.S. DEPARTMENT OF ENERGY | Office of Science

Catalytic Mechanism for Single-Site Water Oxidation Process: A Theoretical Study

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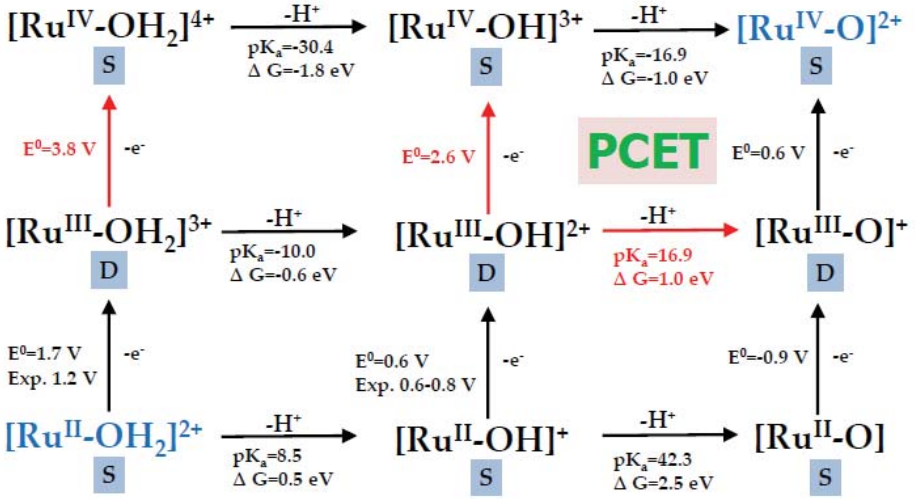
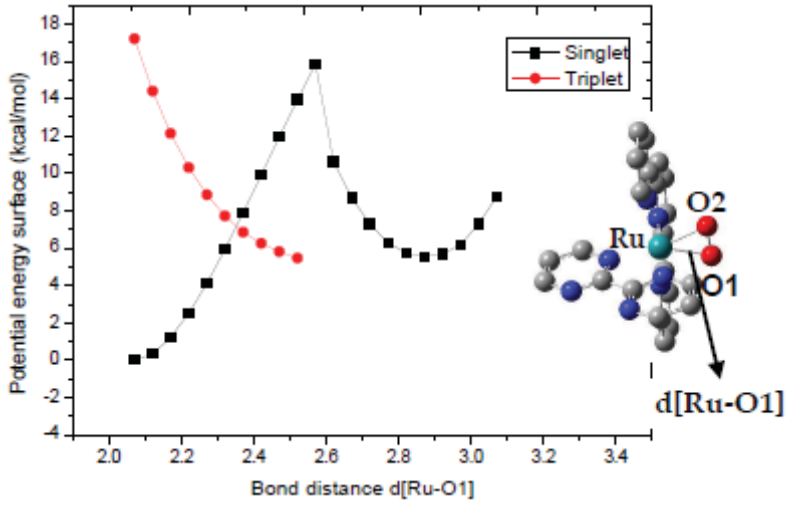


Significance:

- How water oxidation process can be catalyzed by single-site ruthenium complexes are important in solar fuels.
- Using several theoretical tools including our new QM/MM-MFEP approaches, we can characterize the entire catalytic cycle with atomistic details.
- Our studies should be helpful to design new catalysts based on other earth-abundant metals for solar fuels.

Achievement:

- The electronic spin states of ruthenium intermediates during the catalytic cycle are identified as well as the corresponding optimal geometries.
- Our pK_a and redox calculations for first two PCETs and following ET process from $[Ru^{II}\text{-}OH_2]^{2+}$ to $[Ru^{IV}{=}O]^{2+}$ and then to $[Ru^{V}{=}O]^{3+}$ suggest that these processes can proceed readily in weak base or acidic conditions.
- To release O_2, the Ru compounds must achieve high spin state by spin-orbital couplings.



SOLAR FUELS & NEXT GENERATION PHOTOVOLTAICS

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