

A Photo-Induced $1e^-$, $2H^+$ Transfer Reaction

Scientific Achievement

A one-electron two proton transfer reaction was quantified in a chromophore-catalyst molecule competent for water oxidation.

Significance and Impact

Proton coupled electron transfer is of central importance to solar fuel production. The demonstration of a single photon initiated $1e^-$ $2H^+$ transfer represents new chemistry.

Research Details

- The chromophore-catalyst molecule shown was synthesized and anchored to oxide surfaces.
- The Pourbaix diagram reveals that oxidation of $Ru(III)-OH_2$ yields $Ru(IV)=O$ and $2H^+$ at pH 1.
- The chromophore-catalyst in the formal oxidation states $Ru(II)-Ru(III)$ was illuminated at pH 1 on TiO_2 which initiated the sequence of reactions shown schematically.
- The $1e^-$, $2H^+$ transfer reaction was found to occur with an observed rate constant $k_{obs} = 0.03\ s^{-1}$.

Ke Hu, Yusuke Tamaki, Seth Marquard, Gerald J. Meyer, and Thomas J. Meyer
In preparation.

Work was performed at the University of North Carolina at Chapel Hill

