Photocatalytic cycle of water splitting with small manganese oxides and the roles of water cluster as a direct resource of oxygen molecules

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Photoinduced water oxidation summarized as

$$2H_2O + 4h\nu \rightarrow 4H^+ + 4e^- + O_2$$

is a very fundamental process not only in biological systems but for the technology of water splitting. To comprehend this simple looking but very complicated reaction, the mechanisms of at least three crucial phenomena, among others, need to be elucidated: (i) Charge separation as $H^+ + e^-$, (ii) basic scheme of catalytic cycle, and (iii) generation of molecular oxygen in spin triplet. We have clarified the photodynamical mechanism of charge separation to consider the item (i), which is referred to as coupled proton electron-wavepacket transfer (CPEWT). [1]

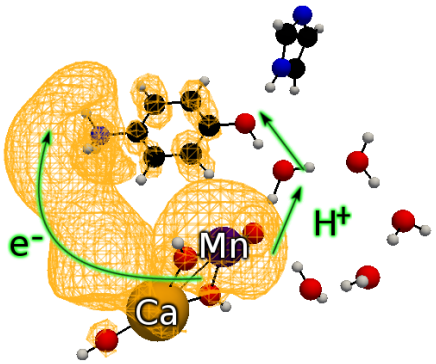

Fig. 1 Selected snapshot of the CPEWT.

In the present poster, we show chemical principles behind photodynamics of water-splitting based on the CPEWT to address all the three items above. The molecular system of the previous study [1] is extended to include a water cluster as a resource of oxygen molecules. The CPEWT also works in the extended system as shown in Fig. 1. We assume that the CPEWT similarly occurs four times, each of which triggers individual series of chemical reactions. As a result of our semi-quantitative analysis, it is found that triplet O_2 is formed from the water cluster within a range of given photon energy of 3.0 eV. [2]

References

[1] K. Yamamoto and K. Takatsuka, ChemPhysChem **18**, 537 (2017).

[2] K. Yamamoto and K. Takatsuka, Phys. Chem. Chem. Phys. (accepted for publication).