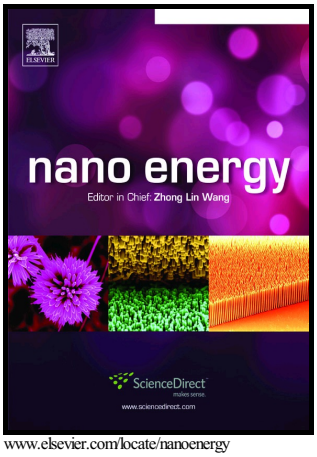

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Efficient design principle for interfacial charge separation in hydrogen-intercalated nonstoichiometric oxides

Zhenao Gu[a,e], Le Zhang[d], Bo Wen[d], Xiaoqiang An[a,b*], Huachun Lan[c], Li-Min Liu[c*], Tao Chen[f], Jing Zhang[f], Xingzhong Cao[g], Junwang Tang[h], Huijuan Liu[c], Jiuhui Qu[a,e]

[a] Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China.

[b] Center for Water and Ecology, State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China

[c] School of Physics, Beihang University, 100191, China

[d] Beijing Computational Science Research Center, Beijing 100094, China.

[e] University of Chinese Academy of Sciences, Beijing 100049, China.

[f] Beijing Synchrotron Radiation Facility (BSRF), Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100049, China.

[g] Multi-discipline Research Division, Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100049, China.

[h] Department of Chemical Engineering, University College London, Torrington Place, London, WC1E 7JE, UK.

E-mail: xqan@rcees.ac.cn

E-mail: limin.liu@csrc.ac.cn

Abstract: Establishing effective strategies to boost the separation of interfacial charge carriers is necessary to address the limiting bottlenecks of photocatalysis. Although oxygen vacancy modulation has become the prevalent strategy to improve the photoactivity, controversy persists regarding the real role of these defects in charge separation. To exert the great potential of nonstoichiometric semiconductors, one needs not only to establish a full atomistic picture of oxygen vacancies, but also to deliberate their possible interactions with other interfacial structures (represented by the ubiquitous intercalated hydrogen). Herein, WO_3 was used as a typical model to demonstrate the principle of hydrogen-intercalated nonstoichiometric oxides for photoelectrochemical water splitting. Both experimental characterizations and theoretical calculations evidenced the synergetic interactions between oxygen vacancies and intercalated hydrogen. The sequential formation of subsurface defect clusters and surface O-H bonds contributed significantly to the spatial separation of charge carriers and the impressive stability of nonstoichiometric photoanodes. Profiting from this principle, an unprecedented photocurrent of 2.94 mA•cm^{-2} at 1.23 V vs. RHE was achieved, apart from a 100 mV