Supporting Information

Investigating the behavior of various cocatalysts on LaTaON$_2$ photoanode for visible light water splitting

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Fig. S1 Kubelka−Munk absorbance for LaTaON$_2$, NiO$_x$−LaTaON$_2$, Ni$_{0.7}$Fe$_{0.3}$O$_x$−LaTaON$_2$, and CoO$_x$−LaTaON$_2$ as well as post−loaded IrO$_x$−LaTaON$_2$.

In comparison with the bare LaTaON$_2$, only slight changes occur in the absorbance at 405 nm (light source for PEC measurement) for the cocatalysts loaded LaTaON$_2$. This is understandable if considering quite low nominal loading amount (2 wt%) of these cocatalysts. In addition, the absorption edge is also slightly blue shifted after loading cocatalysts, which could be due to the shading effect of cocatalysts.
**Fig. S2** Potentiodynamic measurements in 0.5 M NaOH (pH=13.0) for NiO$_x$–LaTaON$_2$ after treatment in air and under NH$_3$.

**Fig. S3** Potentiodynamic measurements in 0.5 M NaOH (pH=13.0) for sputtered Ni–LaTaON$_2$ and CuO$_x$–LaTaON$_2$ in comparison to as prepared–LaTaON$_2$.

**H$_2$O$_2$ as hole scavenger**

H$_2$O$_2$/O$_2$ has a relatively negative oxidation potential (0.682 V$_{RHE}$) and its rate constant is 10 to 100 times higher than that of water.$^1$ Thus, H$_2$O$_2$ is an efficient hole scavenger in electrolyte to suppress surface recombination. Holes that come to the surface will be immediately captured and participate in the oxidation of H$_2$O$_2$. Assuming that all samples have the same bulk recombination, any difference seen in the photocurrent of H$_2$O$_2$ oxidation should therefore be due to the recombination at the interfaces between LaTaON$_2$ and the cocatalysts as well as the electrolyte.

As shown in Fig. S3, the photocurrent of NiO$_x$–LaTaON$_2$ is the highest followed by that of Ni$_{0.7}$Fe$_{0.3}$O$_x$–LaTaON$_2$, both higher than that of bare LaTaON$_2$. The improvement in the photocurrent indicates a reduced recombination and good passivation at the interface of LaTaON$_2$ and the cocatalysts, which is consistent with our conclusions.

CoO$_x$– and IrO$_x$–loaded LaTaON$_2$ exhibit huge background cathodic and anodic currents, which makes the chopped photocurrent characteristics undetectable. This is probably due to their high catalytic activity for H$_2$O$_2$ oxidation and the photocurrent–doubling phenomena, which causes the oxidized form of H$_2$O$_2$ to be oxidized again through injection of an electron into the conduction band of the semiconductor.$^{1-4}$ The oxidation procedures can be described as follows: H$_2$O$_2$ + h$^+ \rightarrow 2H^+$ + O$_2$ and O$_2$ + e$^- \rightarrow O_2$ + e$^-$. In comparison to NiO$_x$– and Ni$_{0.7}$Fe$_{0.3}$O$_x$–LaTaON$_2$, CoO$_x$– and IrO$_x$–loaded LaTaON$_2$ have higher background currents, which is also consistent with the PEC measurement without H$_2$O$_2$, indicating the more dependency of the system’s photocurrent on the catalytic effect of CoO$_x$ and IrO$_x$. 

1−4
However, the presence of a hole scavenger and cocatalyst still did not significantly enhance the photocurrent up to the mA cm$^{-2}$ range, implying that the PEC performance is not limited by surface or interface recombination, but rather by the recombination or low electronic conductivity in the bulk of LaTaON$_2$.

![PEC performance](image)

**Fig. S4** PEC performance under chopped light in 0.5 M NaOH with the presence of 0.1 M H$_2$O$_2$ for pre-loaded NiO$_x$-LaTaON$_2$, Ni$_{0.7}$Fe$_{0.3}$O$_x$-LaTaON$_2$, and CoO$_x$-LaTaON$_2$ as well as post-loaded IrO$_x$-LaTaON$_2$. (a). Zoomed-in view of PEC performance. (b). PEC performance between 0.6–1.8 V$_{RHE}$, calculated based on the illuminated area of 3 mm$^2$.

**References**