Supporting Information (SI)

Effect of Layers on the Photocatalytic Hydrogen Evolution in Dion-Jacobson Layered-Tantalum Perovskites

Peng Wang[a,b,#], Wenqian Chen[a,b,#,*], Zihan Wang[a,b], Ya Tang[c,*], Wenyan Shi[a,b], Liang Tang[a,b*]

a. Key Laboratory of Organic Compound Pollution Control Engineering, Ministry of Education, Shanghai 200444, PR China.

b. School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, PR China.

c. Department of Chemistry, School of Science, Shanghai University, No. 99, Shangda Roda, Baoshan District, Shanghai, China

Correspondence and requests for materials should be addressed to Wenqian Chen (email: wenqianchen@shu.edu.cn, tangya0709@shu.edu.cn, tang1liang@shu.edu.cn).

Table S1. Atomic composition of all samples was determined by SEM-EDS.

Samples	Atomic ratios		
	K/Ta	La/Ta	Ca/Ta
$LaTaO_4$	-	0.98(±0.05)	-
$KLaTa_2O_7$	0.43(±0.01)	0.59(±0.05)	-
$KCa_2Ta_3O_{10}$	0.34(±0.01)	-	0.67(±0.04)

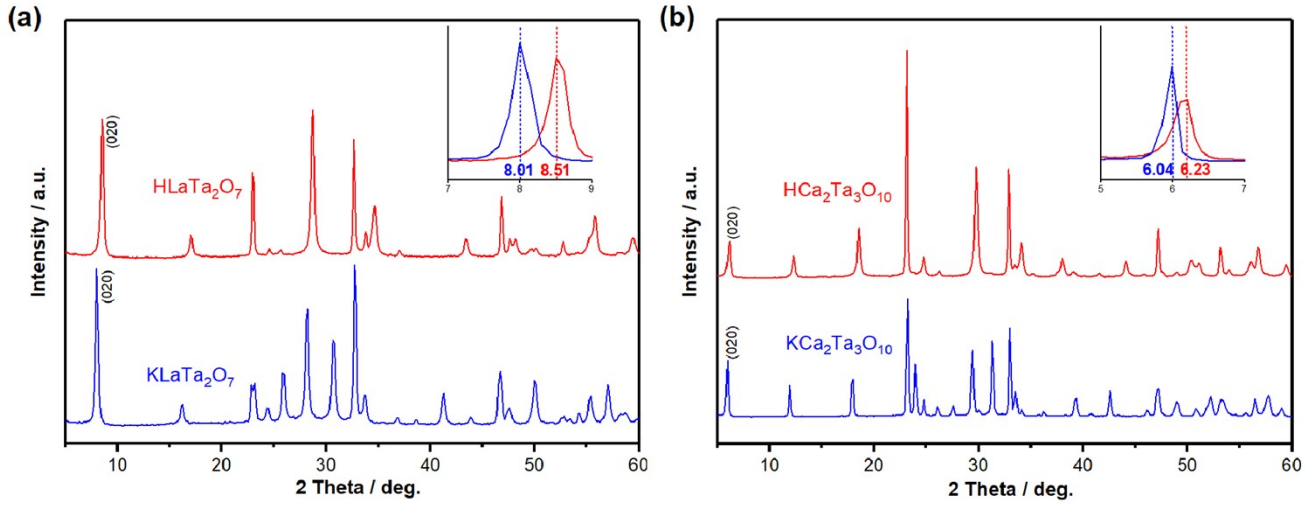

Fig. S1 XRD patterns of (a) $HLaTa_2O_7$ and (b) $HCa_2Ta_3O_{10}$ protonated perovskites. For comparison, XRD patterns of $KLaTa_2O_7$ and $KCa_2Ta_3O_{10}$ were also given, and in brackets are given (hkl) Miller index.

Table S2. The hydrogen evolution yield and quantum efficiency of the samples.

Samples	H_2 evolution amount in 3 h (μmol)	Quantum efficiency (%)
$LaTaO_4$	12.89	0.006
$KLaTa_2O_7$	506.97	0.26
$KCa_2Ta_3O_{10}$	298.62	0.15

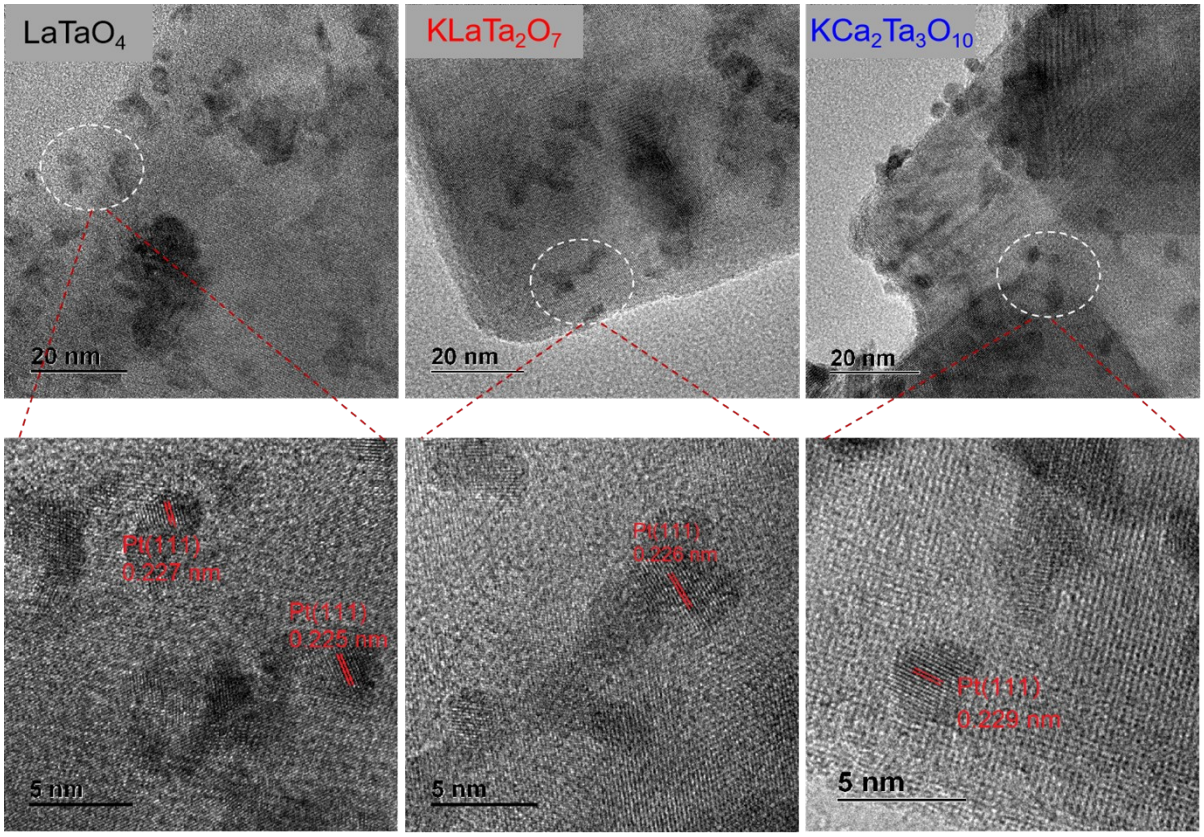

Fig. S2 TEM images of $Pt/LaTaO_4$, $Pt/KLaTa_2O_7$, and $Pt/KCa_2Ta_3O_{10}$ photocatalysts.

Table S3. Atomic percentage of Pt-loaded samples before and after H_2 evolution reaction was determined by SEM-EDS.

Samples	Atomic percentage		
	K	La or Ca	Ta
$Pt/KLaTa_2O_7$	10.83	9.78	22.75
$Pt/KLaTa_2O_7$-after	2.57	11.15	20.79
$Pt/KCa_2Ta_3O_{10}$	6.13	13.84	22.56
$Pt/KCa_2Ta_3O_{10}$-after	2.06	13.50	22.51

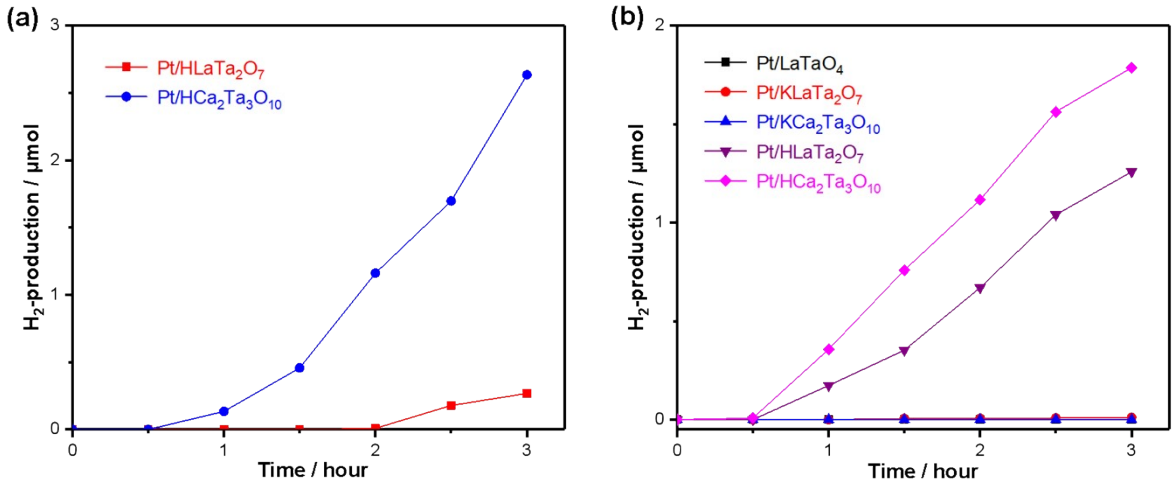

Fig. S3 (a) Time courses of photocatalytic H_2 evolution over $Pt/HLaTa_2O_7$, and $Pt/HCa_2Ta_3O_{10}$. Reaction conditions: fresh catalyst, 50 mg; reaction solution, 20 vol % aqueous methanol solution (pH ≈ 2.5, 100 mL); light source, 300 W Xe lamp (λ ≥ 350 nm). (b) Time courses of H_2 evolution over $Pt/LaTaO_4$, $Pt/KLaTa_2O_7$, $Pt/KCa_2Ta_3O_{10}$, $Pt/HLaTa_2O_7$, and $Pt/HCa_2Ta_3O_{10}$. Reaction conditions: catalyst, 50 mg; reaction solution, 5 mmol NaI solution (pH ≈ 2.5, 100 mL); light source, 300 W Xe lamp (λ ≥ 350 nm).

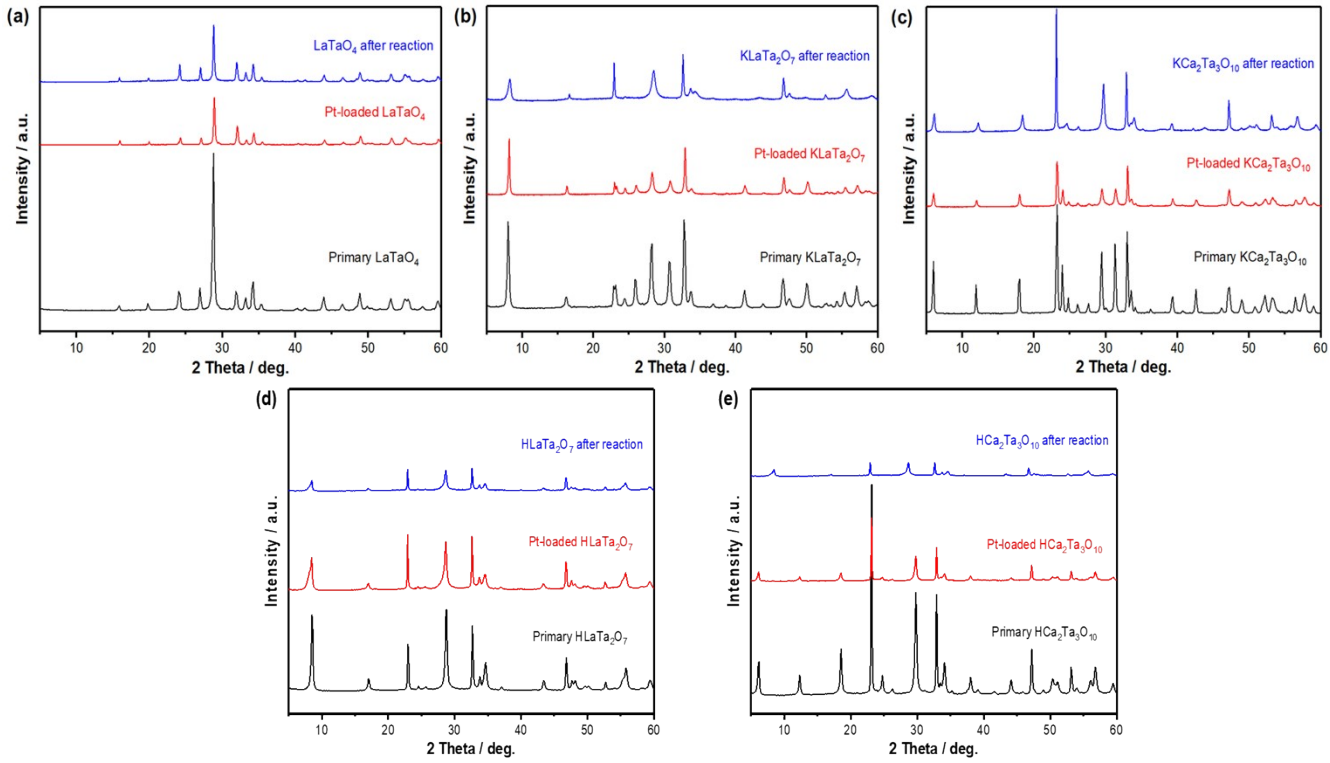

Fig. S4 XRD patterns of the Pt-loaded samples, along with patterns of samples before and after H_2 evolution reactions in 20 vol % aqueous methanol solution. (a) $LaTaO_4$, (b) $KLaTa_2O_7$, (c) $KCa_2Ta_3O_{10}$, (d) $HLaTa_2O_7$, (e) $HCa_2Ta_3O_{10}$.

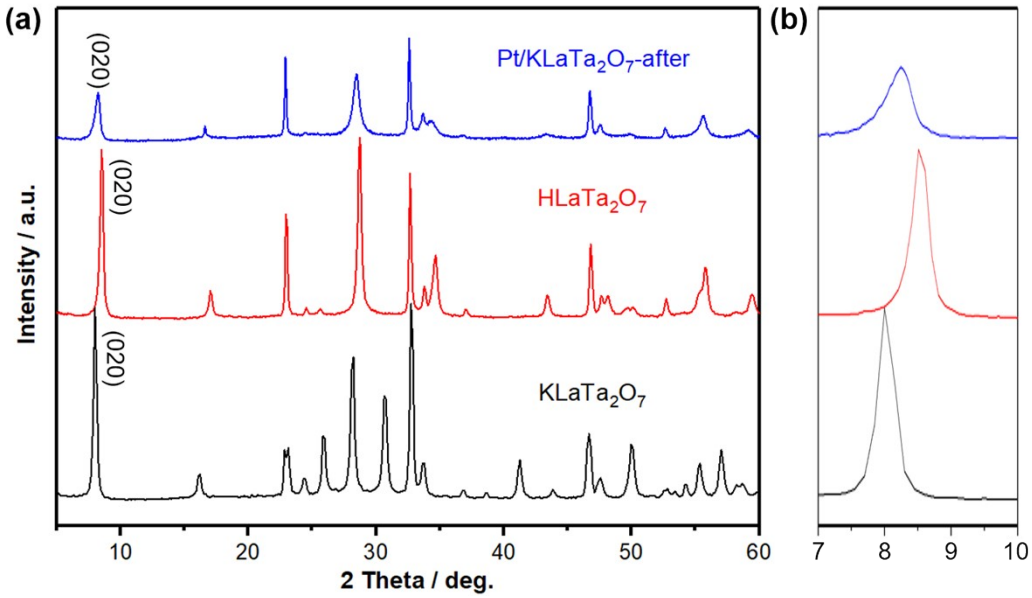

Fig. S5 (a) XRD patterns of $KLaTa_2O_7$, $HLaTa_2O_7$, along with pattern of $Pt/KLaTa_2O_7$ after H_2 evolution reaction in 20 vol % aqueous methanol solution (pH ≈ 2.5). (b) Enlarged XRD patterns at 2θ = 7-10°.

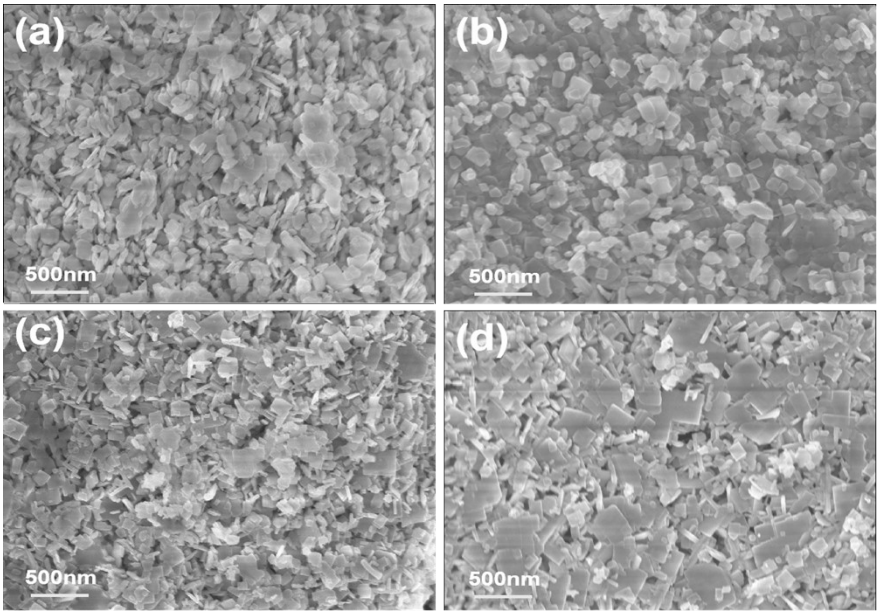

Fig. S6 SEM images of (a) $Pt/KLaTa_2O_7$, (b) $Pt/KLaTa_2O_7$ after H_2 evolution reaction, (c) $Pt/KCa_2Ta_3O_{10}$, (d) $Pt/KCa_2Ta_3O_{10}$ after H_2 evolution reaction.

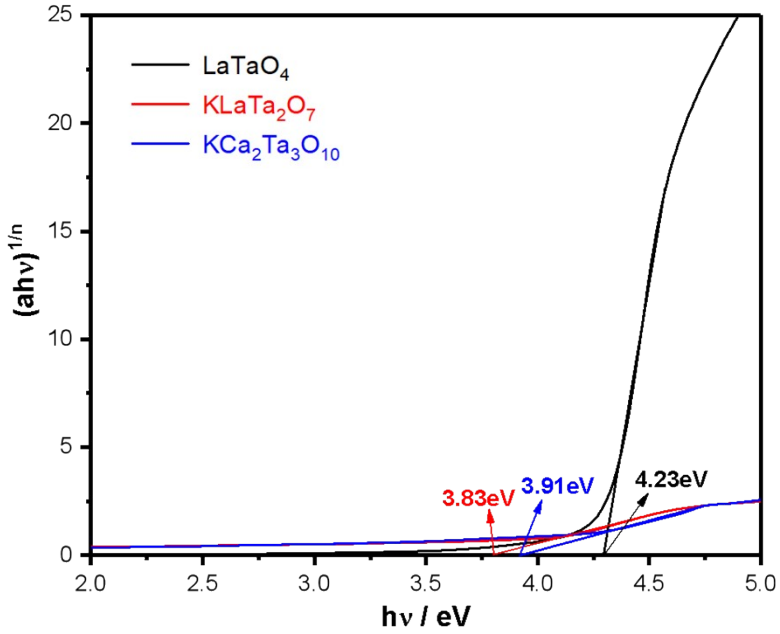

Fig. S7 Tauc plot of direct-band gap semiconductor $LaTaO_4$ and indirect-band gap semiconductors $KLaTa_2O_7$, $KCa_2Ta_3O_{10}$.

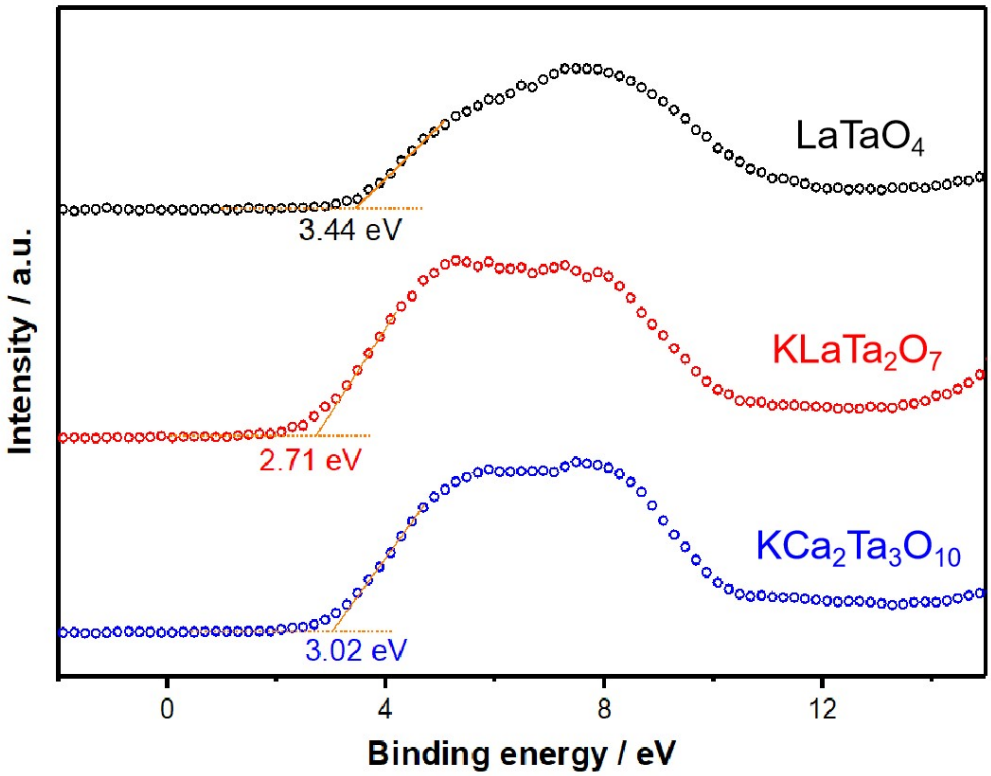

Fig. S8 VB-XPS spectra of $LaTaO_4$, $KLaTa_2O_7$, and $KCa_2Ta_3O_{10}$.