Frustrated lewis pair chemistry in 2D CeO_2 for efficient alkaline hydrogen evolution

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Supplementary materials:

1. Fig. S1 to S7

2. Table S1 to S3

1. Fig. S1 to S7

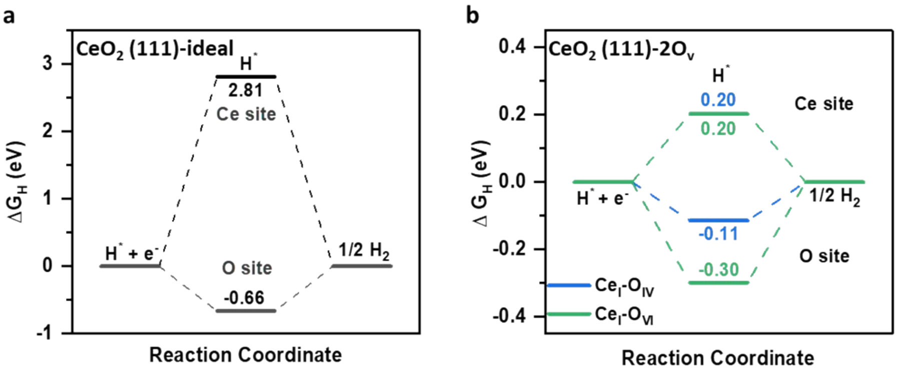


Fig. S1. ΔG_H of the HER on (a) CeO_2 (111)-ideal facet and on the Ce_I-O_{IV} and Ce_I-O_{VI} FLPs on the CeO_2 (111)-2O_v facet.

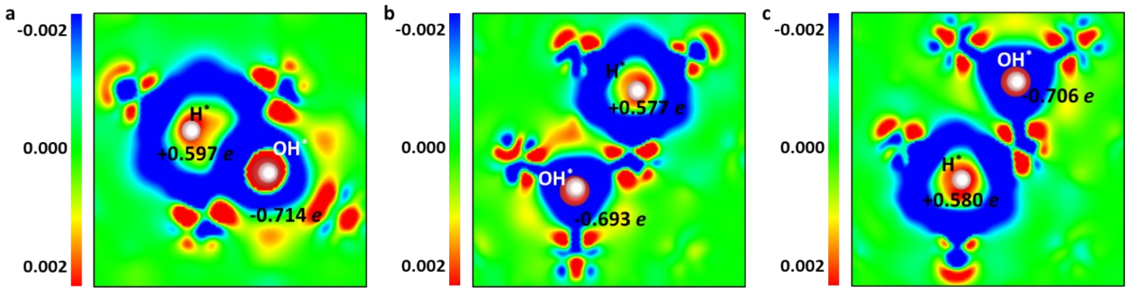


Fig. S2. Charge density difference maps of water dissociation on (a) Ce_I-O_V FLP sites of CeO_2 (111)-O_v facet and (b) Ce_I-O_{IV} and (c) Ce_I-O_{VI} of CeO_2 (111)-2O_v facet.

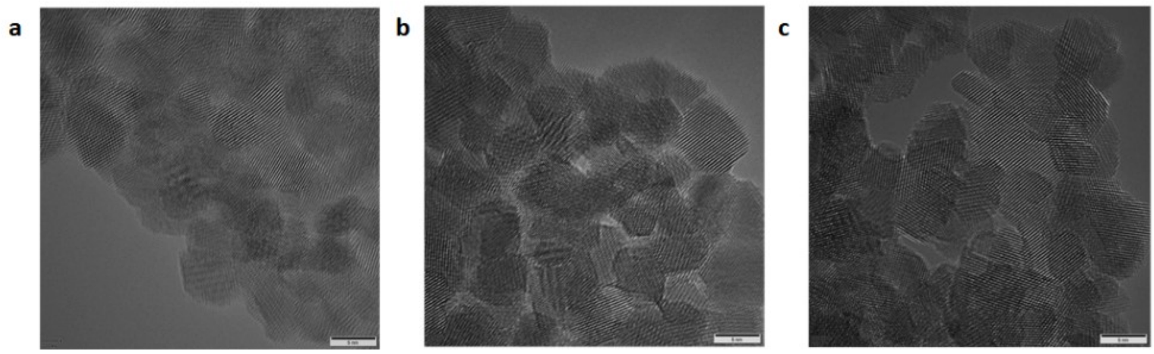


Fig. S3. HRTEM images of (a) CeO_2-I, (b) CeO_2-II and (c) CeO_2-III. (d)The SEM images of CeO_2-I after testing electrocatalytic properties.

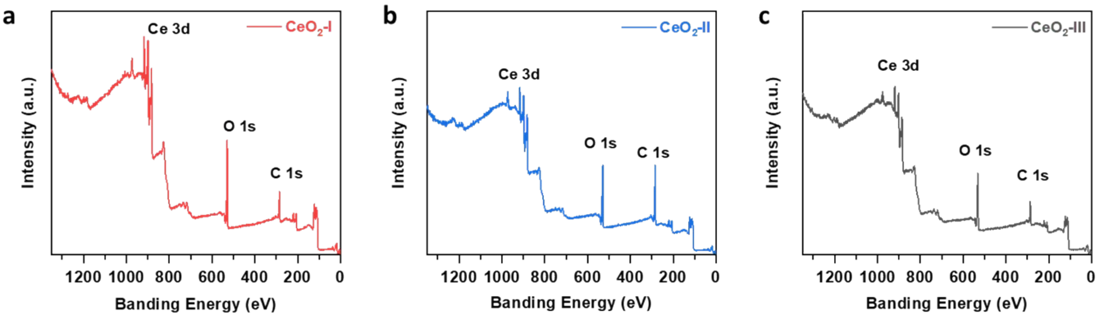


Fig. S4. Full spectra of (a) CeO_2-I, (b) CeO_2-II and (c) CeO_2-III.

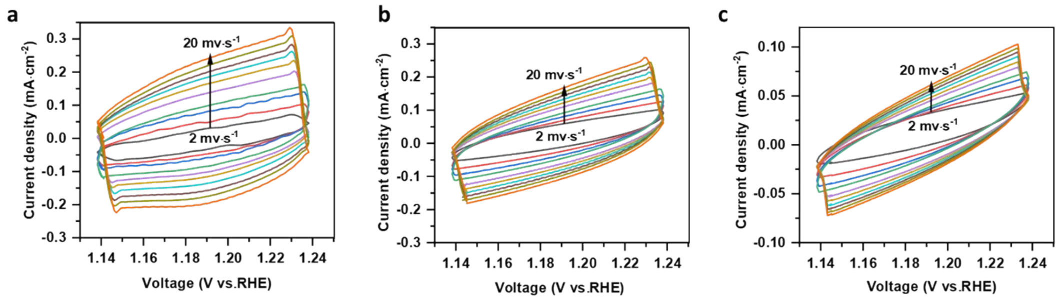


Fig. S5. CV measurements in a non-faradic current region (1.14-1.24 V) in 1.0 M KOH solution at scan rates of 2 to 20 mV s^{-1} of (a) CeO_2-I, (b) CeO_2-II and (c) CeO_2-III.

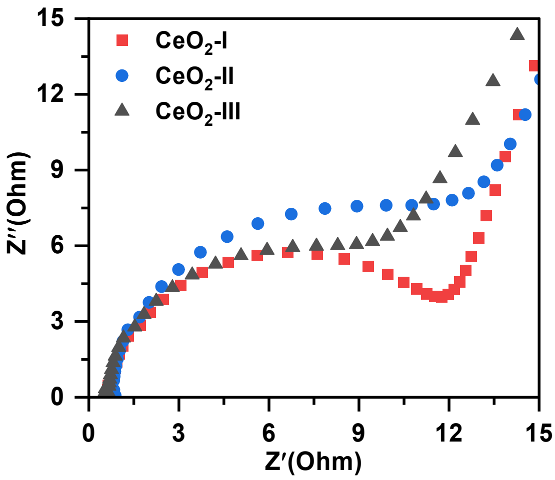


Fig. S6. Comparison of Nyquist plots for CeO_2-I, CeO_2-II, and CeO_2-III.

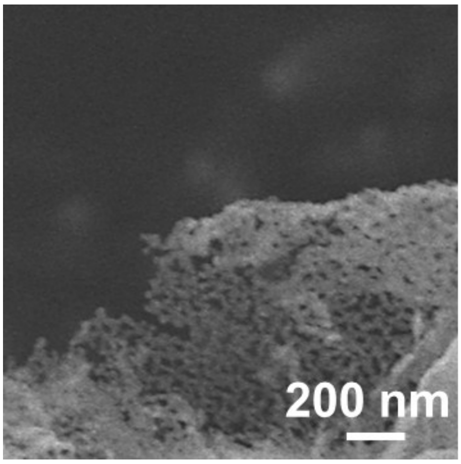
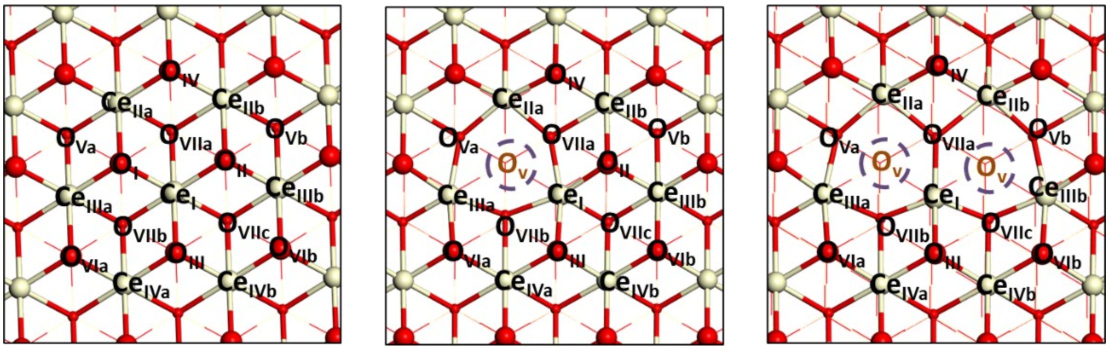


Fig. S7. The SEM images of CeO_2-I after 15 hours of chronopotentiometric measurements.

2. Table S1 to S3

Table S1. Bader charge population of the atoms on the surface of CeO_2 (111).



Atom	**Bader charge population / *e***		
	Ideal CeO_2(111)	CeO_2(111) with one oxygen vacancy	CeO_2(111) with two oxygen vacancy
Ce_{I}	**+2.246**	**+2.206**	**+2.160**
Ce_{IIIb}	+2.246	+2.205	+2.199
Ce_{IIb}	+2.246	+2.259	+2.199
Ce_{IIIa}	+2.246	+2.202	+2.192
Ce_{IIIb}	+2.246	+2.241	+2.192
Ce_{IVa}	+2.246	+2.256	+2.253
Ce_{IVb}	+2.246	+2.244	+2.251
O_{I}	-1.132	/	/
O_{II}	-1.132	-1.147	/
O_{III}	-1.132	-1.147	-1.159
O_{IV}	-1.132	-1.147	-1.148
O_{Va}	-1.137	-1.178	-1.185
O_{Vb}	-1.137	-1.183	-1.186
O_{VIa}	-1.132	-1.151	-1.155
O_{VIb}	-1.132	-1.142	-1.155
O_{VIIa}	-1.137	-1.183	-1.251
O_{VIIb}	-1.137	-1.178	-1.175
O_{VIIc}	-1.137	-1.123	-1.172

Table S2. Top views of structures related to water dissciation on the CLP of CeO_2 (111)-ideal, and the FLPs of CeO_2 (111)-O_v and CeO_2 (111)-2O_v.

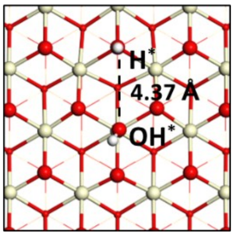
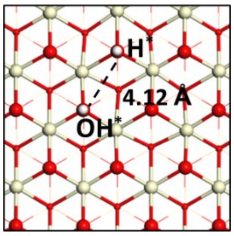
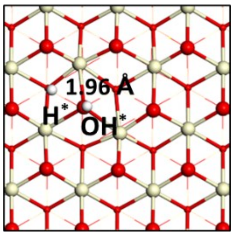
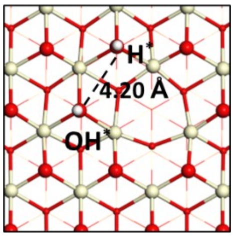
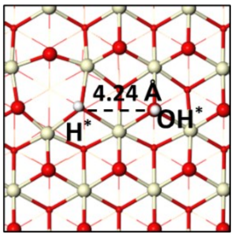
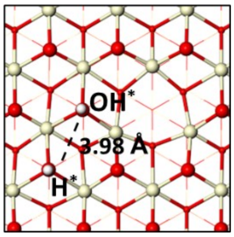
Surface	**Ce_I-O_{IV}**	**Ce_I-O_V**	**Ce_I-O_{VI}**
CeO_2 (111)-ideal			
CeO_2 (111)-O_v			
CeO_2 (111)-2O_v			

Table S3. Bader charge population of surface atoms after water dissociation by CeO_2 (111).

Atom	Bader charge population / *e*		
	Ideal CeO_2(111)	CeO_2(111) with one oxygen vacancy	CeO_2(111) with two oxygen vacancy
Ce_I	**+2.351**	**+2.262**	**+2.251**
Ce_{IIIb}	+2.244	+2.243	+2.269
Ce_{IIb}	+2.245	+2.228	+2.274
Ce_{IIIa}	+2.245	+2.234	+2.225
Ce_{IIIb}	+2.242	+2.233	+2.203
Ce_{IVa}	+2.238	+2.285	+2.274
Ce_{IVb}	+2.239	+2.233	+2.269
O_I	-1.085	/	/
O_{II}	-1.094	-1.149	/
O_{III}	-1.109	-1.145	-1.153
O_{IV}	-1.268	-1.270	-1.158
O_{Va}	-1.109	-1.169	-1.305
O_{Vb}	-1.116	-1.145	-1.133
O_{VIa}	-1.135	-1.144	-1.153
O_{VIb}	-1.139	-1.140	-1.150
O_{VIIa}	-1.139	-1.145	-1.181
O_{VIIb}	-1.115	-1.127	-1.144
O_{VIIc}	-1.116	-1.146	-1.168