Electronic Supplementary Information

Computational Insights into the Strain Effect in Electrocatalytic Reduction of CO$_2$ to CO on Pd Surfaces

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Table S1. Slope (*a*) and intercept (*b*) of the linear relations fitted in Figure 1.

		Pd(111)		Pd(100)		Pd(110)		Pd(211)	
		a	*b*	*a*	*b*	*a*	*b*	*a*	*b*
-2%	CO	3.71	-2.83	1.57	-1.97	0.57	-1.83	0.65	-1.94
	COOH	4.01	-1.18	4.49	-1.87	1.77	-1.18	1.02	-0.91
-1%	CO	3.49	-2.79	1.53	-2.04	0.53	-1.81	0.72	-2.01
	COOH	3.86	-1.19	4.26	-1.85	1.57	-1.13	1.02	-0.92
1%	CO	3.00	-2.66	1.36	-2.00	0.44	-1.74	0.97	-2.20
	COOH	3.52	-1.19	3.79	-1.74	1.68	-1.18	1.26	-1.03
2%	CO	2.78	-2.60	1.27	-1.97	0.53	-1.79	0.99	-2.20
	COOH	3.30	-1.18	3.51	-1.67	1.17	-1.00	1.18	-1.00

Table S2. Average net charge at C and O atom of the CO molecules on the surface of Pd(111), (100), (110) and (211) with varying strains.

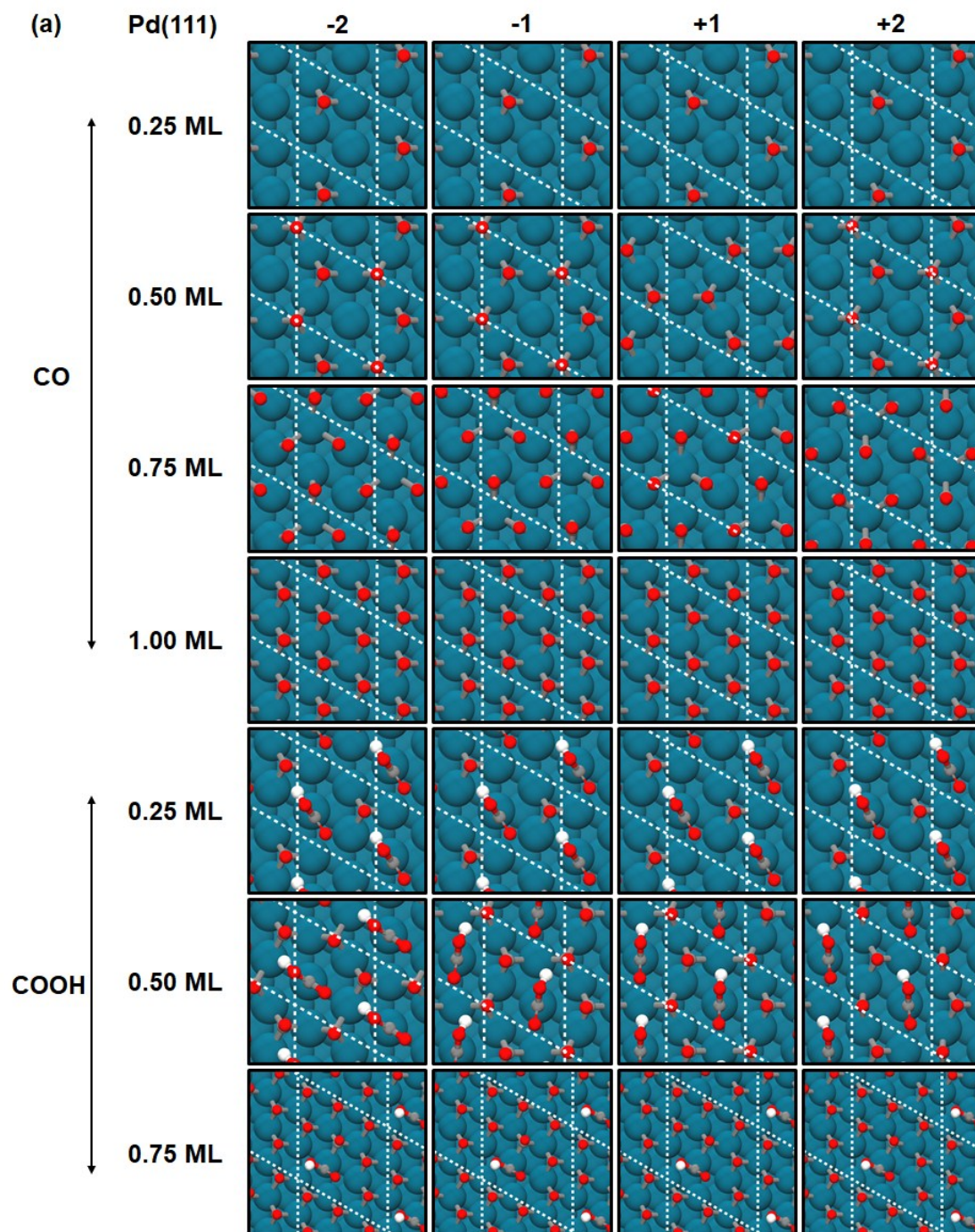|	111		100		110		211	
	C	O	C	O	C	O	C	O
-2	0.859	-0.985	0.859	-0.974	0.919	-1.036	0.868	-1.026
-1	0.839	-0.983	0.844	-0.964	0.915	-1.044	0.863	-1.021
0	0.828	-0.963	0.886	-0.997	0.916	-1.035	0.868	-1.030
1	0.850	-0.989	0.889	-1.003	0.906	-1.038	0.868	-1.030
2	0.835	-0.973	0.848	-0.981	0.917	-1.040	0.865	-1.031
Average	0.842	-0.979	0.865	-0.984	0.915	-1.038	0.866	-1.028

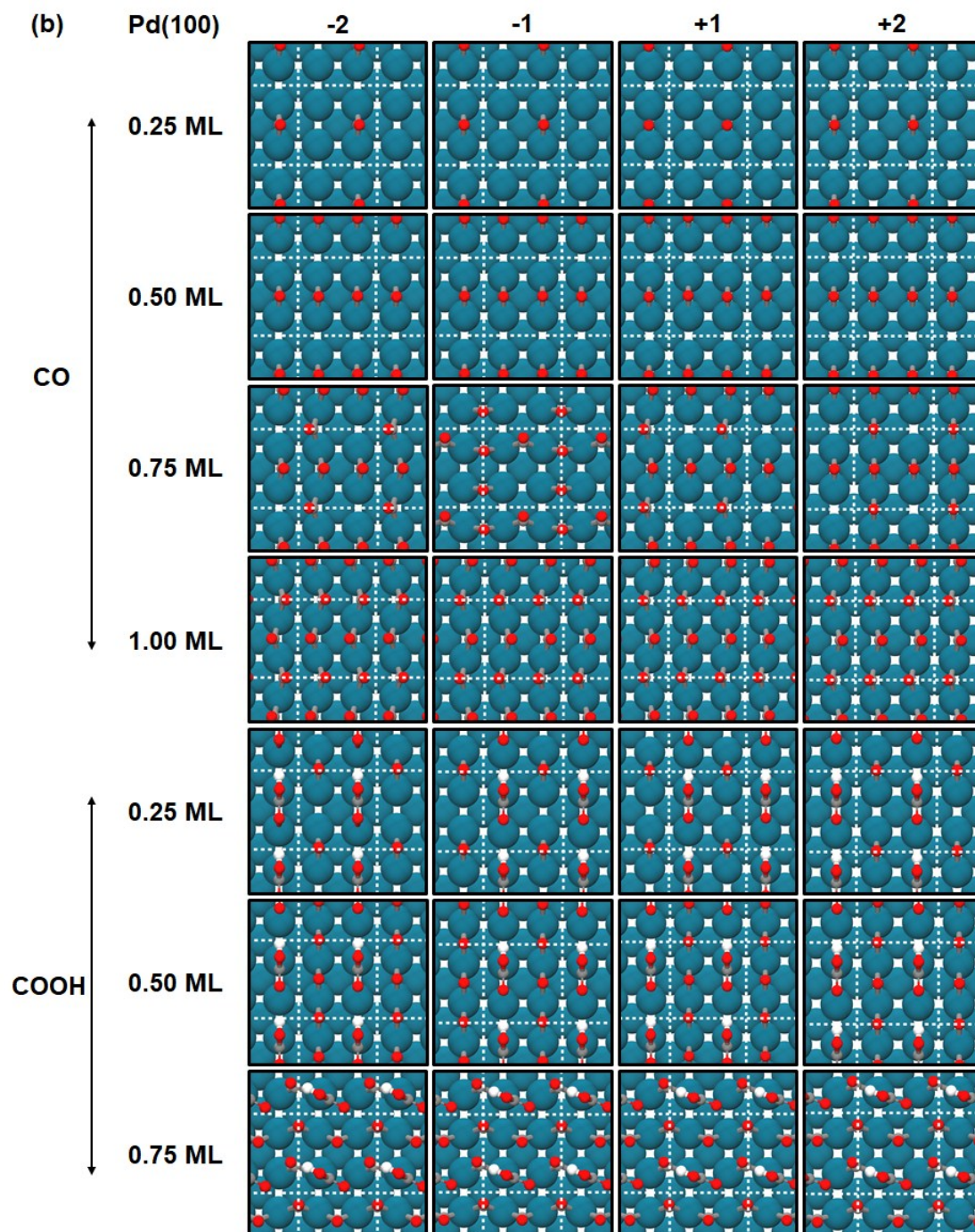**Table S3.** Steady-state coverage of CO (in ML) while assuming reaction R1 or R3 as the rate-determining step.

Pd(111)	-2%		-1%		1%		2%	
	COOH	CO	COOH	CO	COOH	CO	COOH	CO
-0.6	0.551	0.904	0.576	0.946	0.625	0.990	0.649	0.990
-0.7	0.551	0.952	0.576	0.986	0.625	0.990	0.649	0.990
-0.8	0.551	0.987	0.576	0.990	0.625	0.990	0.649	0.990
-0.9	0.551	0.990	0.576	0.990	0.625	0.990	0.649	0.990
-1	0.551	0.990	0.576	0.990	0.625	0.990	0.649	0.990
-1.1	0.551	0.990	0.576	0.990	0.625	0.990	0.649	0.990
-1.2	0.551	0.990	0.576	0.990	0.625	0.990	0.649	0.990

Pd(100)	-2%		-1%		1%		2%	
	COOH	CO	COOH	CO	COOH	CO	COOH	CO
-0.6	0.720	0.990	0.781	0.990	0.840	0.990	0.872	0.990
-0.7	0.720	0.990	0.781	0.990	0.840	0.990	0.872	0.990
-0.8	0.720	0.990	0.781	0.990	0.840	0.990	0.872	0.990
-0.9	0.720	0.990	0.781	0.990	0.840	0.990	0.872	0.990
-1	0.720	0.990	0.781	0.990	0.840	0.990	0.872	0.990
-1.1	0.720	0.990	0.781	0.990	0.840	0.990	0.872	0.990
-1.2	0.720	0.990	0.781	0.990	0.840	0.990	0.872	0.990

Pd(110)	-2%		-1%		1%		2%	
	COOH	CO	COOH	CO	COOH	CO	COOH	CO
-0.6	0.990	-0.600	0.990	0.990	0.990	0.990	0.990	0.990
-0.7	0.990	-0.700	0.990	0.990	0.990	0.990	0.990	0.990
-0.8	0.990	-0.800	0.990	0.990	0.990	0.990	0.990	0.990
-0.9	0.990	-0.900	0.990	0.990	0.990	0.990	0.990	0.990
-1	0.990	-1.000	0.990	0.990	0.990	0.990	0.990	0.990
-1.1	0.990	-1.100	0.990	0.990	0.990	0.990	0.990	0.990
-1.2	0.990	-1.200	0.990	0.990	0.990	0.990	0.990	0.990

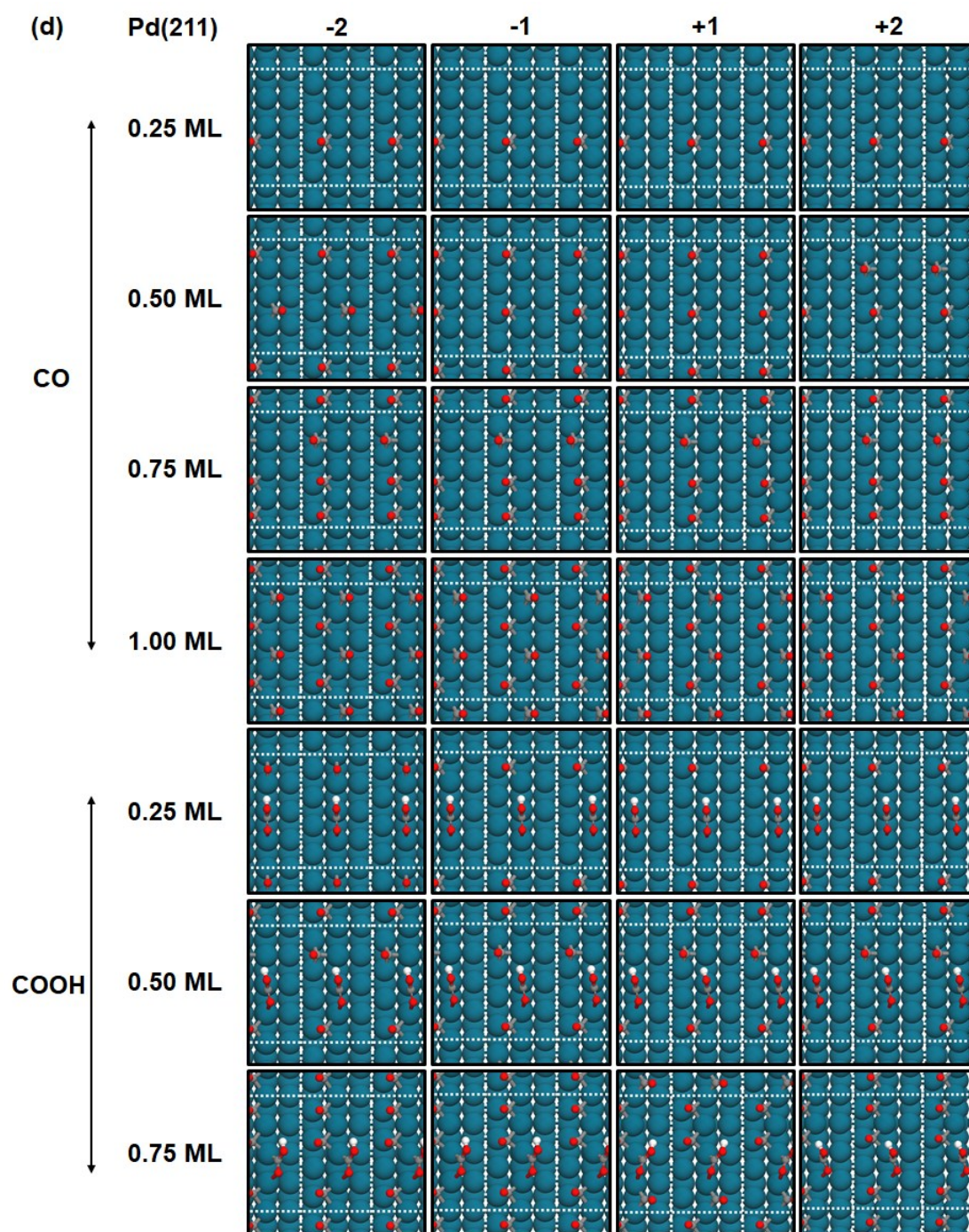| Pd(211)	-2%		-1%		1%		2%	
	COOH	CO	COOH	CO	COOH	CO	COOH	CO
-0.6	0.990	-0.600	0.990	0.990	0.990	0.990	0.990	0.990
-0.7	0.990	-0.700	0.990	0.990	0.990	0.990	0.990	0.990
-0.8	0.990	-0.800	0.990	0.990	0.990	0.990	0.990	0.990
-0.9	0.990	-0.900	0.990	0.990	0.990	0.990	0.990	0.990
-1	0.990	-1.000	0.990	0.990	0.990	0.990	0.990	0.990
-1.1	0.990	-1.100	0.990	0.990	0.990	0.990	0.990	0.990
-1.2	0.990	-1.200	0.990	0.990	0.990	0.990	0.990	0.990

(a) Pd(111)

Figure S1. Adsorption configurations of CO and COOH at different CO coverages on Pd(111), (100), (110) and (211) with varying strains.
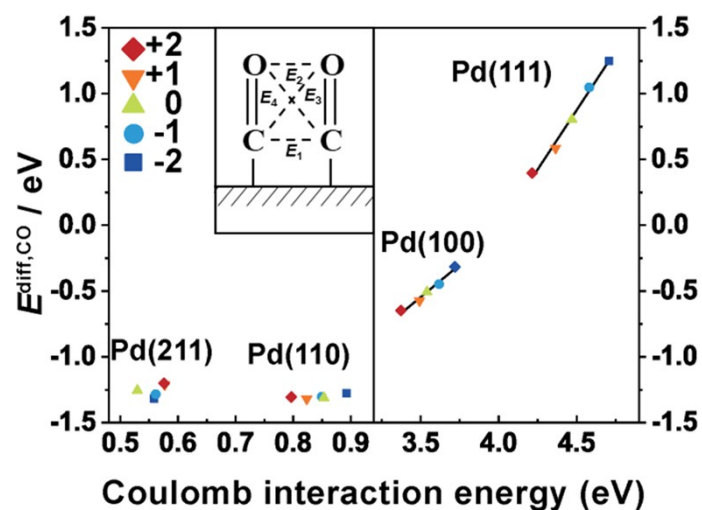
Figure S2. The correlation between differential adsorption energy of CO* at the highest coverage and Coulomb interaction energies between CO* molecules when the lattice parameters are changed. The illustration is a schematic diagram of the Coulomb interaction between two neighboring CO* on the surface. The Coulomb interaction energies were calculated with the averaged net charge value for C and O on the same miller-index surface with different strains, the values of which are also listed in **Table S2**.