Exploration of structure sensitivity of gold nanoparticles in low-temperature CO oxidation

Lei Ying,[ab] Yu Han,[ab] Beien Zhu,*[ac] and Yi Gao*[acd]

[a.] Key Laboratory of Interfacial Physics and Technology, Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, China.

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

[c.] Phonon Science Research Center for Carbon Dioxide, Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201210, China.

[d.] Key Laboratory of Low-Carbon Conversion Science & Engineering, Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201210, China

*Corresponding author. Email: gaoyi@sari.ac.cn (Y.G.); zhube@sari.ac.cn (B.Z.).

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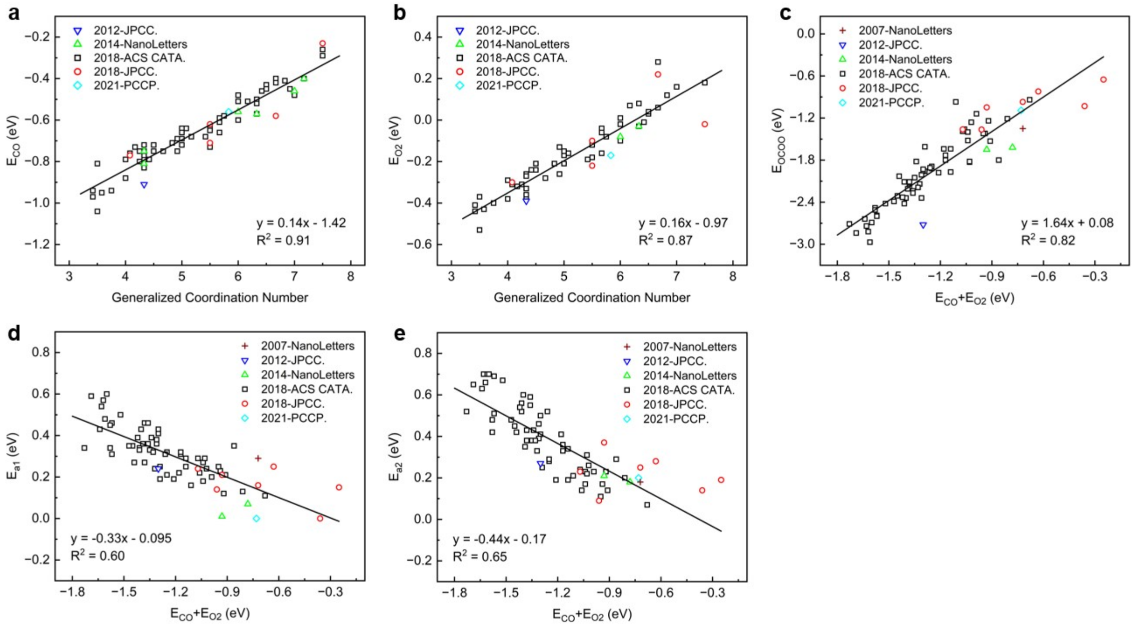

Fig. S1 Adsorption energies versus generalized coordination number for CO (a) and O_2 (b). The scaling of the OCOO adsorption energy (c), activation energy for [$CO^* + O_2^* \rightleftharpoons OCOO^*$] reaction (d), activation energy for [$OCOO^* \rightarrow CO_2(g) + O^* + *$] reaction (e) with the sum of the CO and O adsorption energies. The data are collected from Refs. [1–6].

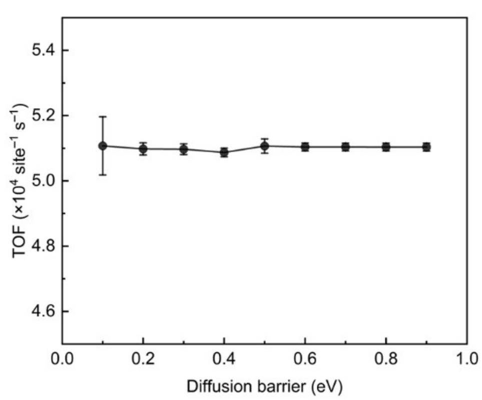

Fig. S2 TOFs with different diffusion barrier at 273 K with P_{O2}/P_{CO} = 1. Error bars are the standard deviation of 10 identically simulations.

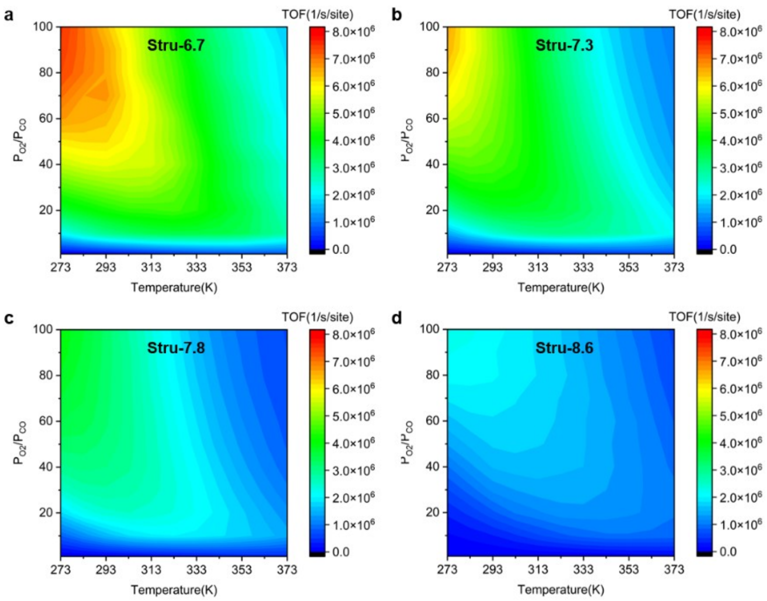

Fig. S3 Contour plot of TOFs for 5 nm Au NPs with $\bar{CN}$ = 6.7 (a), 7.3 (b), 7.8 (c), and 8.6 (d) changing with the temperature and partial pressure ratio (P_{total} = 1 bar).

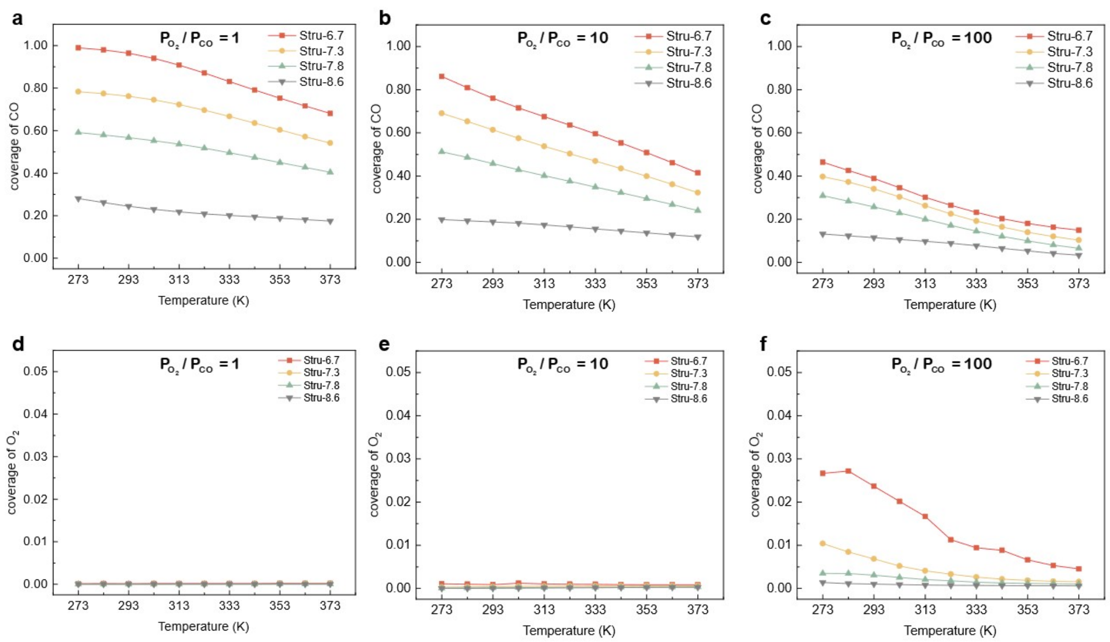

Fig. S4 Coverage of CO and O_2 as a function of temperature at P_{total} = 1 bar with ${P_{O_2}}/{P_{CO}}$ = 1 (a, d), 10 (b, e), and 100 (c, f) for Au NPs with different $\bar{CN}$

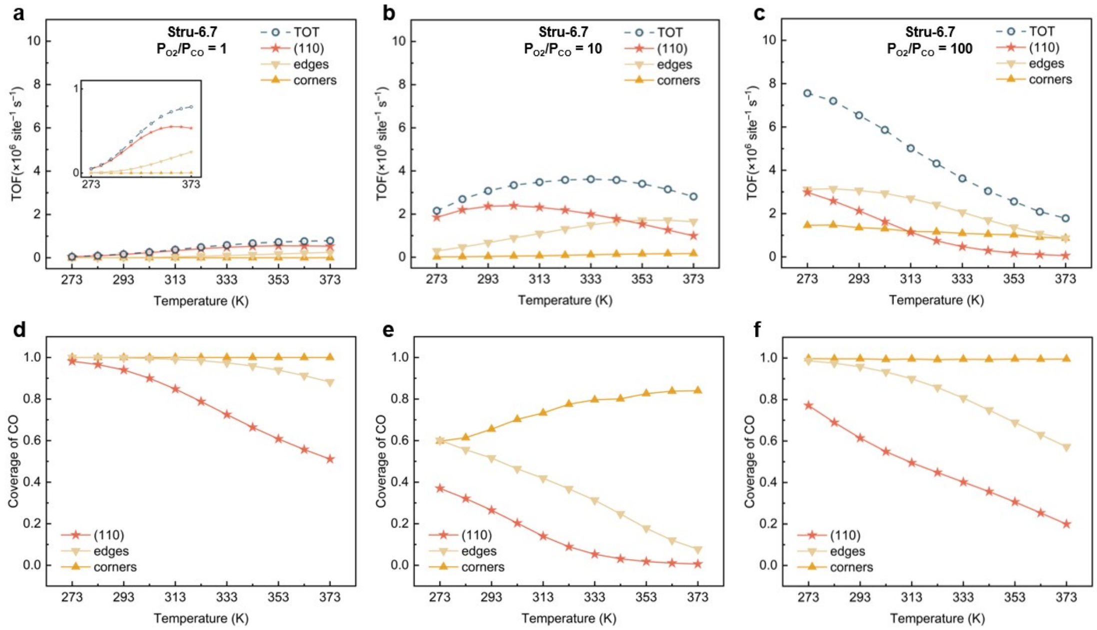

Fig. S5 site-specific contributions to TOFs and coverages of CO for Au NPs with Stru-6.7 under P_{O_2}/P_{CO} = 1 (a, d), 10 (b, e), and 100 (c, f).

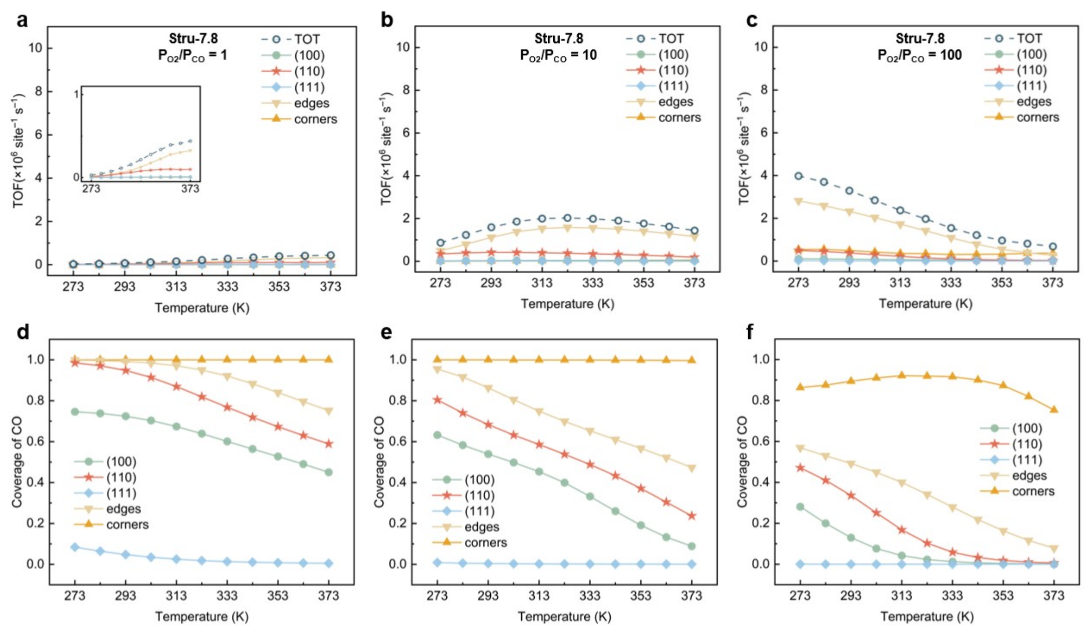

Fig. S6 site-specific contributions to TOFs and coverages of CO for Au NPs with Stru-7.8 under P_{O_2}/P_{CO} = 1 (a, d), 10 (b, e), and 100 (c, f).

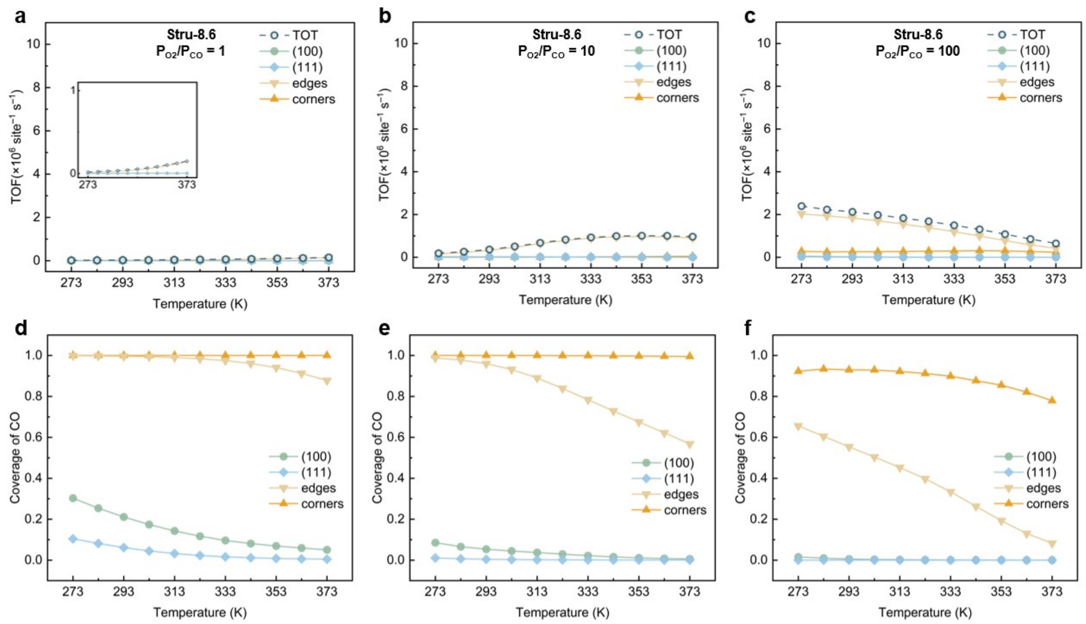

Fig. S7 site-specific contributions to TOFs and coverages of CO for Au NPs with Stru-8.6 under P_{O_2}/P_{CO} = 1 (a, d), 10 (b, e), and 100 (c, f).

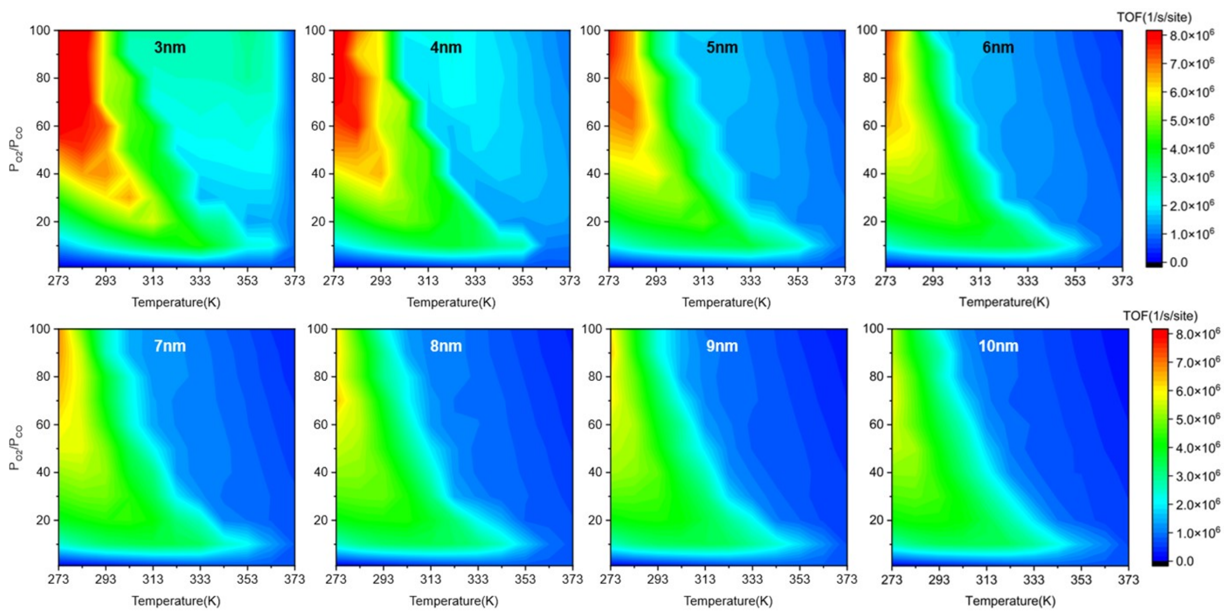

Fig. S8 Contour plot of TOFs for Au NPs changing with the temperature and partial pressure ratio (P_{total} = 1 bar) in the size range of 3-10 nm.

Table S1. Surface tension under vacuum (Units in eV/Å^2) and adsorption energies (Units in eV)

	γ_{hkl}	E_{CO}	E_{O2}	E_O	E_{OCOO}
Au(100)	0.055	-0.48	-0.02	0.06	-0.76
Au(110)	0.056	-0.55	-0.08	0.03	-0.91
Au(111)	0.043	-0.22	-0.01	0.00	-0.77

Table S2. Lateral interaction between A* and B* (Units in eV)$^{\alpha}$

A*	B*	Au(100)	Au(110)	Au(111)	Average
CO	CO	-0.12	-0.08	-0.08	-0.09
	O_2	-0.02	-0.02	-0.00	-0.01
	O	-0.12	-0.18	-0.08	-0.13
	$OCOO_C$	-0.02	0.01	-0.00	-0.00
	$OCOO_O$	-0.02	0.04	-0.00	0.01
O_2	O_2	-0.03	-0.04	-0.03	-0.03
	O	-0.16	-0.26	-0.12	-0.18
	$OCOO_C$	-0.00	-0.02	0.00	-0.00
	$OCOO_O$	-0.00	-0.03	-0.19	-0.07
O	O	-0.18	-0.14	-0.21	-0.18
	$OCOO_C$	-0.06	-0.06	-0.11	-0.08
	$OCOO_O$	-0.07	-0.05	-0.07	-0.06

$^{\alpha}$ $OCOO_C$ means the O-C- side of OCOO*, and $OCOO_O$ means O-O- side.

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