Supplementary Information (SI) for Reaction Chemistry & Engineering.

A simple redox model of low-T NO + CO adsorption onto Pd-CHA as effective Passive NOx Adsorbers

Umberto Iacobone[1], Andrea Gjetja[1], Nicola Usberti[1],

Isabella Nova[1], Enrico Tronconi[1,*]

[1]Laboratory of Catalysis and Catalytic Processes, Dipartimento di Energia,

Politecnico di Milano, Via La Masa 34, I-20156, Milano, Italy

Djamela Bounechada[2], Roberta Villamaina[2], Maria Pia Ruggeri[2], Andrew P. E. York[2],

Loredana Mantarosie[2], Jillian Collier[2]

[2]Johnson Matthey Technology Centre, Blounts Court Road,

Sonning Common, Reading RG4 9NH, UK

Supporting Information

* Corresponding author: Tel.: +39 02 23993264

E-mail address: enrico.tronconi@polimi.it

Apparent rate constants

0.84% Pd	T = 100°C	T = 120°C	T = 150°C	T = 200°C	
k_1	0.004	0.004	0.004	0.004	$[s^{-1}]$
k_2	0.080	0.080	0.080	0.080	$[s^{-1}]$
k_3	0.030	0.030	0.030	0.030	$[s^{-1}]$
k_4^{ads}	0.035	0.035	0.065	0.085	$[s^{-1}]$
k_4^{des}	0.005	0.008	0.050	0.100	$[s^{-1}]$
k_5^{dry}	0.003	0.005	0.020	0.035	$[s^{-1}]$
k_6^{dry}	0.7 E-04*	2.0 E-04*	9.5 E-04*	72.3 E-04*	$[s^{-1}]$
k_7	3.0 E-04	7.0 E-04	35.0 E-04	200.0 E-04	$[s^{-1}]$
k_5^{wet}	0.003	0.005	0.050	0.120	$[s^{-1}]$
k_6^{wet}	0.0 E-04	1.0 E-04	4.0 E-04	6.0 E-04	$[s^{-1}]$

Table SI.1 Apparent rate constants for the 0.84% Pd sample. The estimates for k_5 and k_6 in wet conditions are reported at the bottom of the table. *Extrapolated from the Arrhenius equation.

0.50% Pd	T = 100°C	
k_1	0.00252	$[s^{-1}]$
k_2	0.080	$[s^{-1}]$
k_3	0.030	$[s^{-1}]$
k_4^{ads}	0.035	$[s^{-1}]$
k_4^{des}	0.005	$[s^{-1}]$
k_5^{dry}	0.0018	$[s^{-1}]$
k_6^{dry}	1.19 E-04	$[s^{-1}]$
k_7	1.79 E-04	$[s^{-1}]$
k_5^{wet}	0.0018	$[s^{-1}]$
k_6^{wet}	0.0 E-04	$[s^{-1}]$

Table SI.2 Apparent rate constants for the 0.50% Pd sample. The estimates for k_5 and k_6 in wet conditions are reported at the bottom of the table.

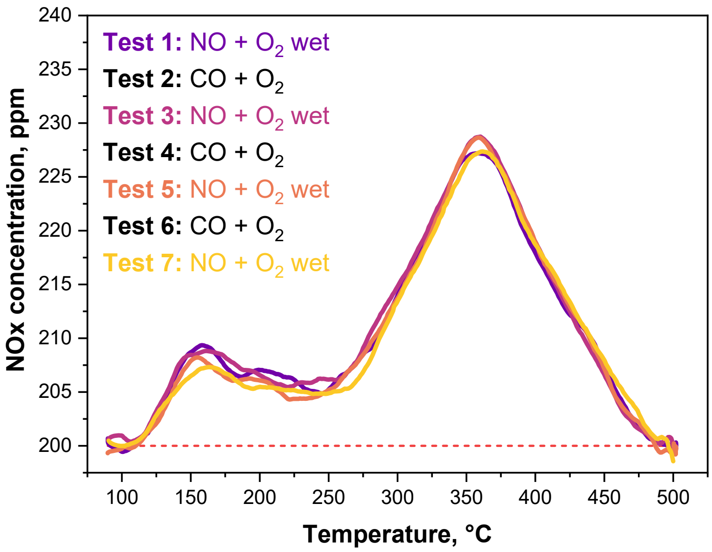

Fig. SI.1 Replicated Temperature Programmed Surface Reaction (TPSR) runs (tests 1, 3, 5, 7) in a NO + O_2 + H_2O atmosphere on the 0.84% Pd-CHA sample: nominal NO feed concentration (dashed line), measured outlet NO concentration (solid lines). T = 100-500 °C at 15 °C/min, W_{cat} = 40 mg, GHSV = 300,000 cm^3/h/gcat (STP). Gas feed: NO = 200 ppm, CO = 0 ppm, O_2 = 10 % v/v, H_2O = 5 % v/v. The figure shows that exposing the catalyst repeatedly to CO + O_2 in tests 2, 4 and 6 (not shown) did not deactivate the catalyst.

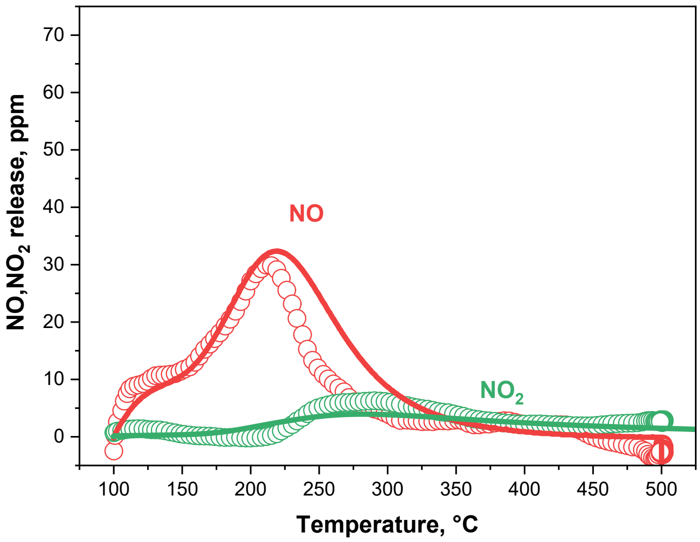

Fig. SI.2 Temperature Programmed Surface Reaction (TPSR) run in a NO + O_2 dry atmosphere on the 0.84% Pd-CHA sample: measured NO release (red dots, difference between NO detected and NO fed), NO_2 trace (green dots), kinetic fit of NO (red solid line) and NO_2 (green line). T = 100-500 °C at 15 °C/min, W_{cat} = 40 mg, GHSV = 300,000 cm^3/h/gcat (STP). Feed: NO = 200 ppm, CO = 0 ppm, O_2 = 10 % v/v, H_2O = 0 % v/v.

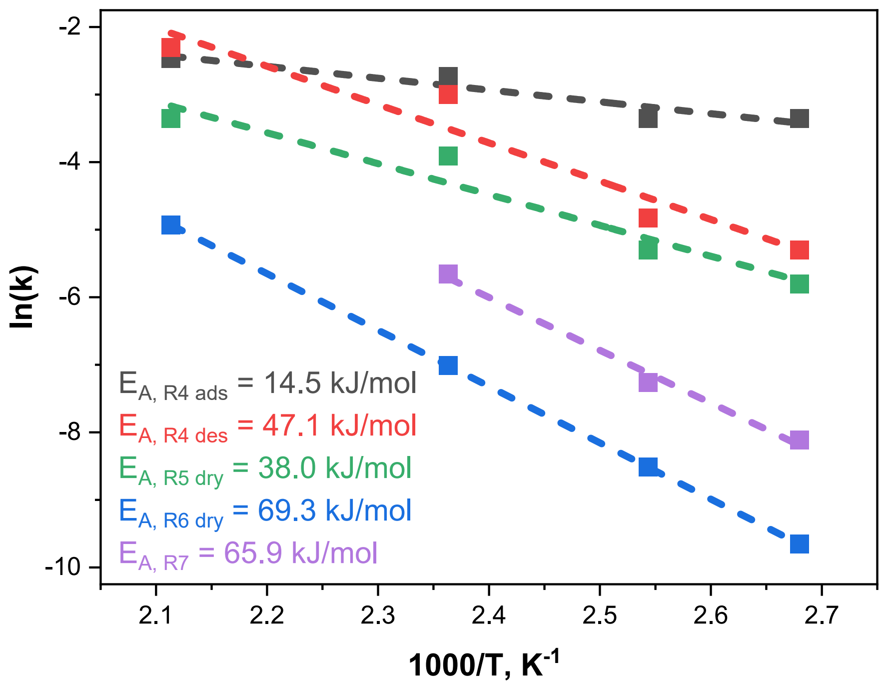

Fig. SI.3 Arrhenius plots of the rate constants for R4ads, R4des, R5 and R6 (dry-gas) over the 0.84% Pd-CHA sample. T-range = 100–200 °C.

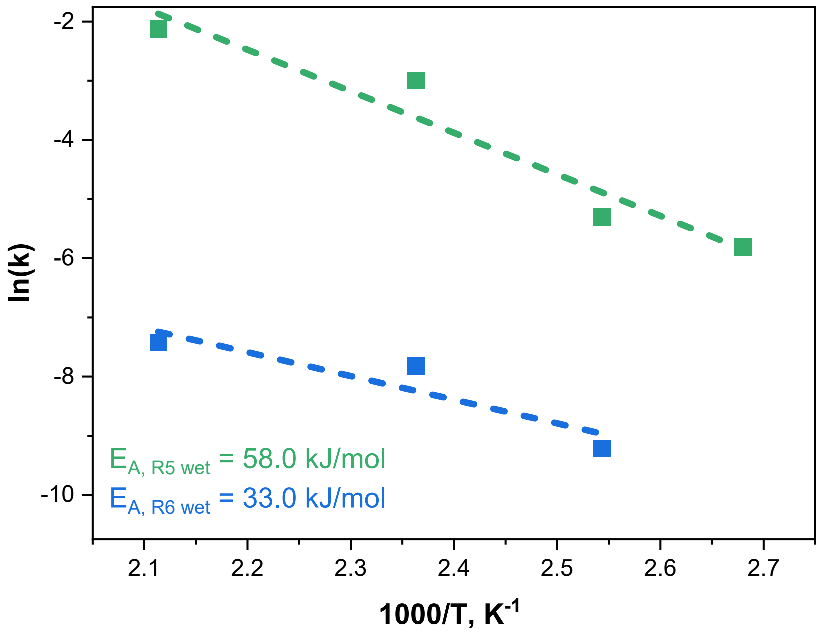

Fig. SI.4 Arrhenius plots of the rate constants for R5 and R6 (wet feed) over the 0.84% Pd-CHA sample. T-range = 100–200 °C.

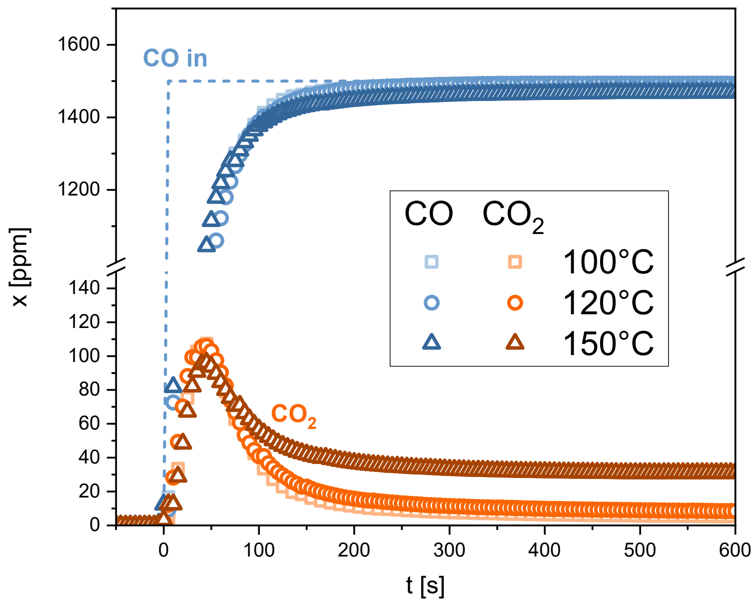

Fig. SI.5 CO + O_2 feed mixture: experimental results over 0.84% Pd sample at 100 (squares), 120 (circles) and 150 °C (triangles). W_{cat} = 40 mg, GHSV = 300,000 $cm^3/h/g_{cat}$ (STP). Gas feed: CO = 1500 ppm, O_2 = 10% v/v, H_2O = 0% v/v.

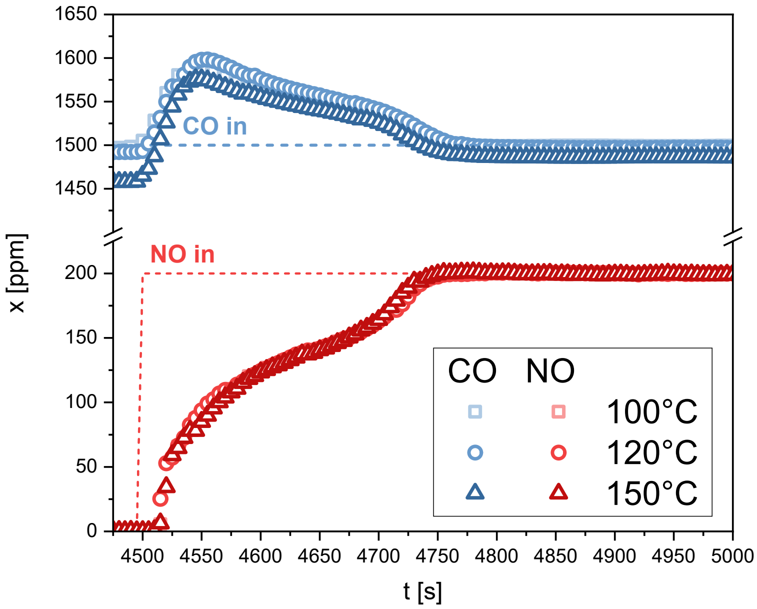

Fig. SI.6 CO + O_2 + delayed NO feed mixture: experimental results over 0.84% Pd sample at 100 (squares), 120 (circles) and 150 °C (triangles). W_{cat} = 40 mg, GHSV = 300,000 $cm^3/h/g_{cat}$ (STP). Gas feed: CO = 1500 ppm, NO = 200 ppm, O_2 = 10% v/v, H_2O = 0% v/v.

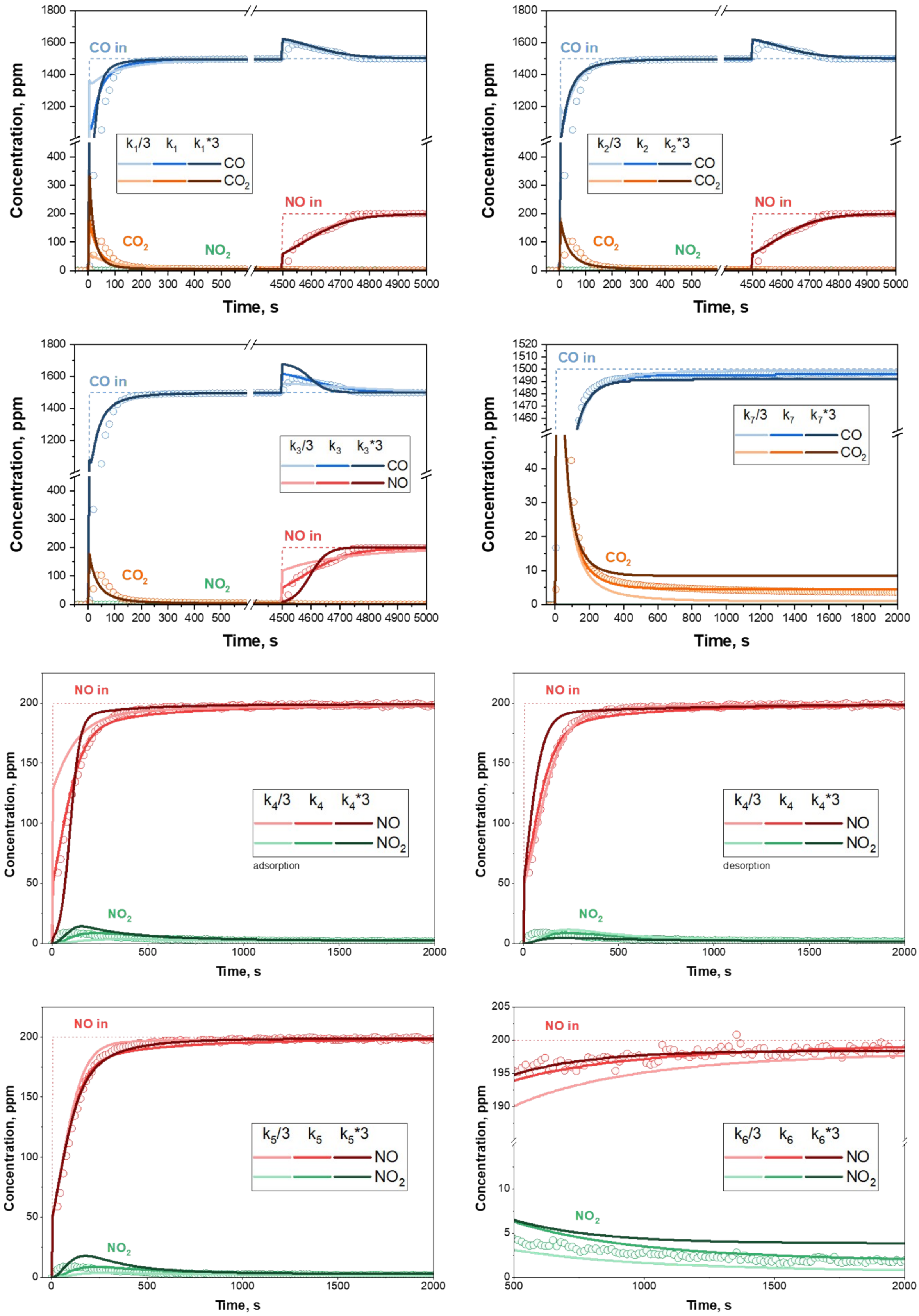

Fig. SI.7 Sensitivity analysis for the different kinetic parameters on the test that was used for the fit. Light colors are for k/3, regular colors for the fitted k and dark colors for k*3.