Supporting Information

Evolution of Pd Species for the Conversion of Methane under Operation Conditions

Jianjun Chen,[a] Yang Wu,[a] Wei Hu,[b, c] Pengfei Qu,[b] Guochen Zhang,[b] Yi Jiao, [a] Lin Zhong [b *], and Yaoqiang Chen [a, b, d *]

[a] Institute of New Energy and Low-carbon Technology, Sichuan University, Chengdu 610064, China

[b] College of Chemical Engineering, Sichuan University, Chengdu 610064, China

[c] Institute of Atmospheric Environment, Chongqing Academy of Environmental Science, Chongqing 401147, China

[d] College of Chemistry, Sichuan University, Chengdu 610064, China

Corresponding author: Lin Zhong (Email: zhonglin@scu.edu.cn; Tel/Fax: +86-28-85418451)

Corresponding author: Yaoqiang Chen (Email: nic7501@scu.edu.cn; Tel/Fax: +86-28-85418451)

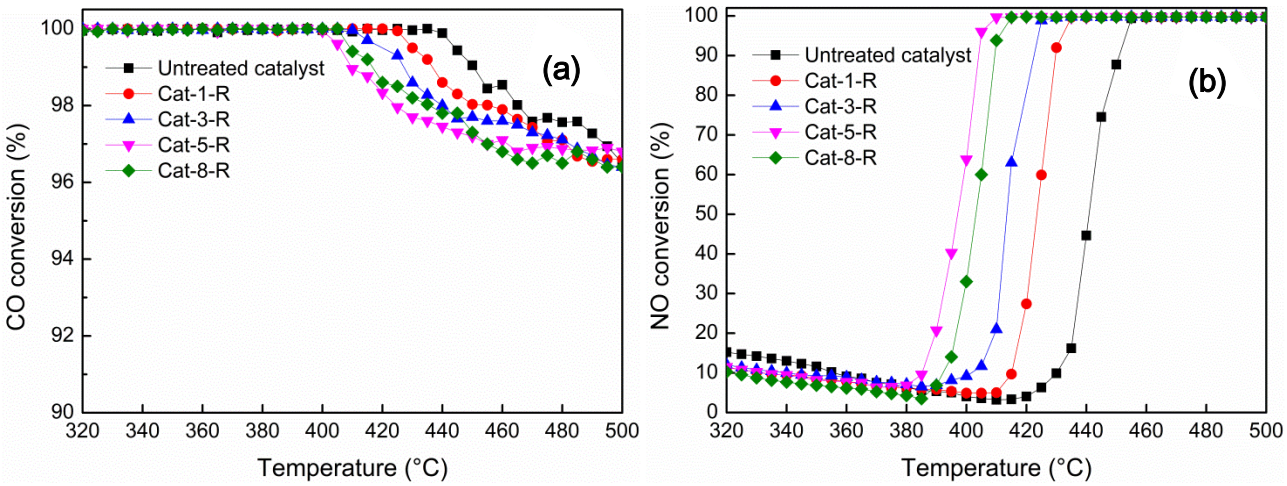

Figure S1. (a) CO and (b) NO light-off curves as temperature was raised from 300 to 500 °C in simulated stoichiometric NGVs exhaust gas for the Pd model catalyst (50 g/ft^3) after pretreated in reaction gas. Conditions: 1000 ppm CH_4 + 5000 ppm CO + 930 ppm NO + 4035 ppm O_2 + 10vol. %CO_2 + 10vol. %H_2O balanced with N_2 (lambda=1); Gas hourly space velocity (GHSV) = 50,400 h^{-1}.

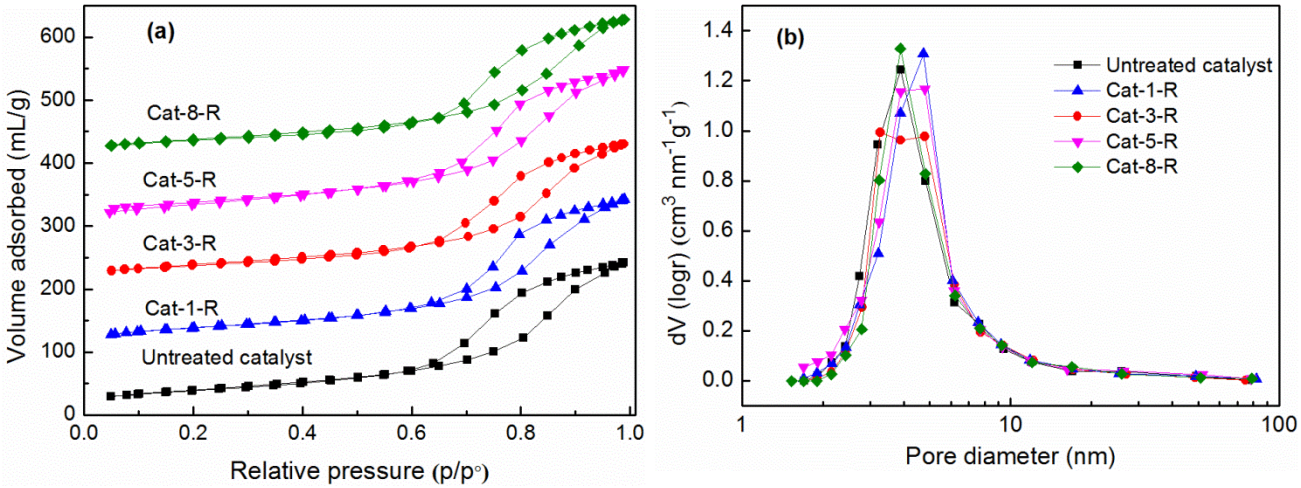

Figure S2. (a) Nitrogen adsorption-desorption isotherms and (b) Barrett-Joyner-Halenda pore size distribution of the Pd model catalysts after pretreatment in reaction gas.

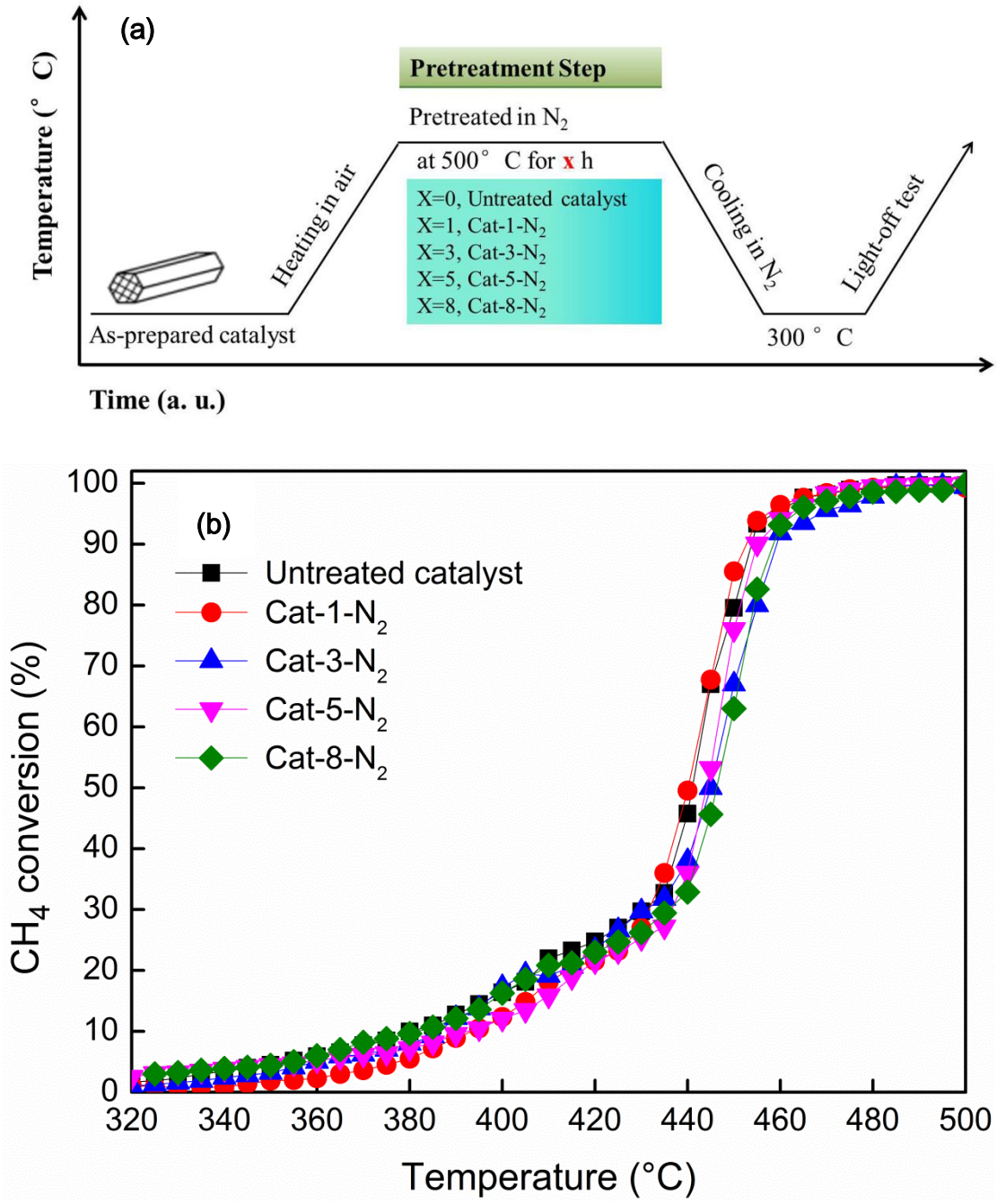

Figure S3. (a) Description of the pretreatment procedures under flowing N_2; (b) CH_4 light-off test for Pd model catalyst (50 g/ft^3) after the above pretreatment.

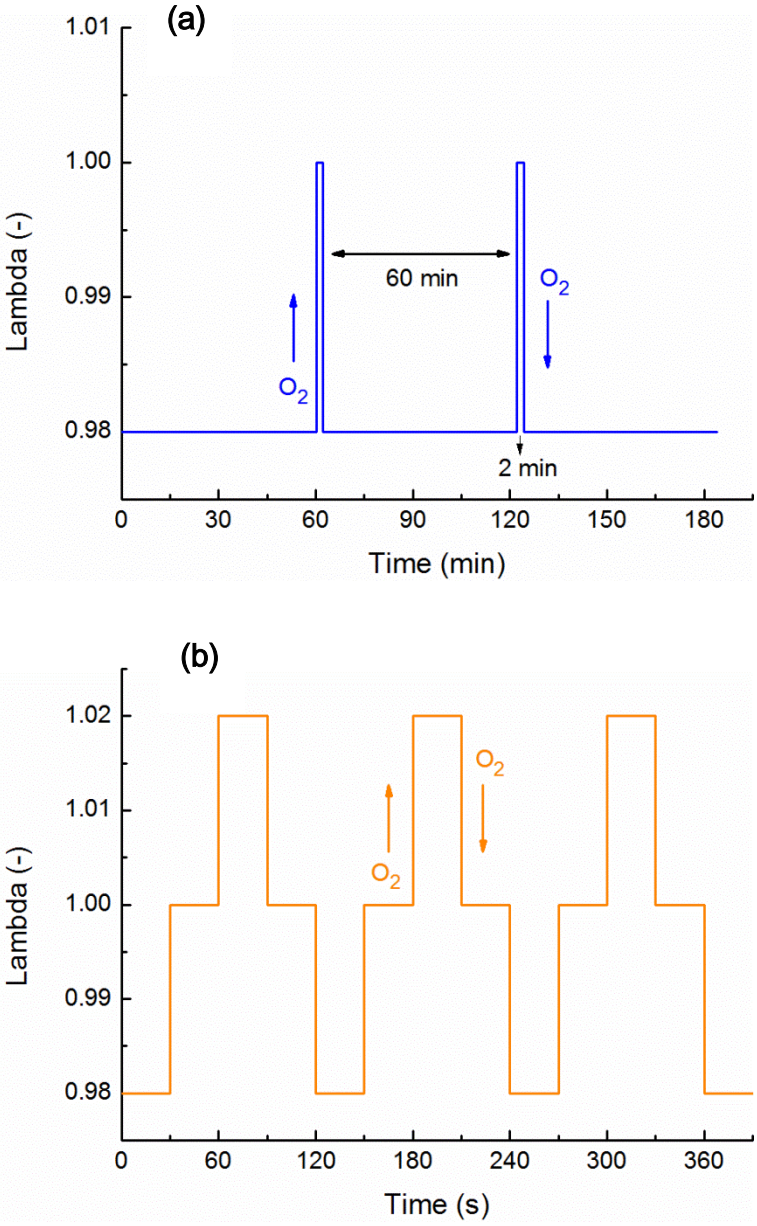

Figure S4. (a) Periodic pulse experiments were conducted by performing every 2 min long stoichiometric pulses (lambda=1.00) per 60 min on a stationary rich reaction gas mixture (lambda=0.98); (b) Lambda oscillating operating tests: lambda oscillation value from 0.98 to 1.02 at constant cycle period of 120 s.

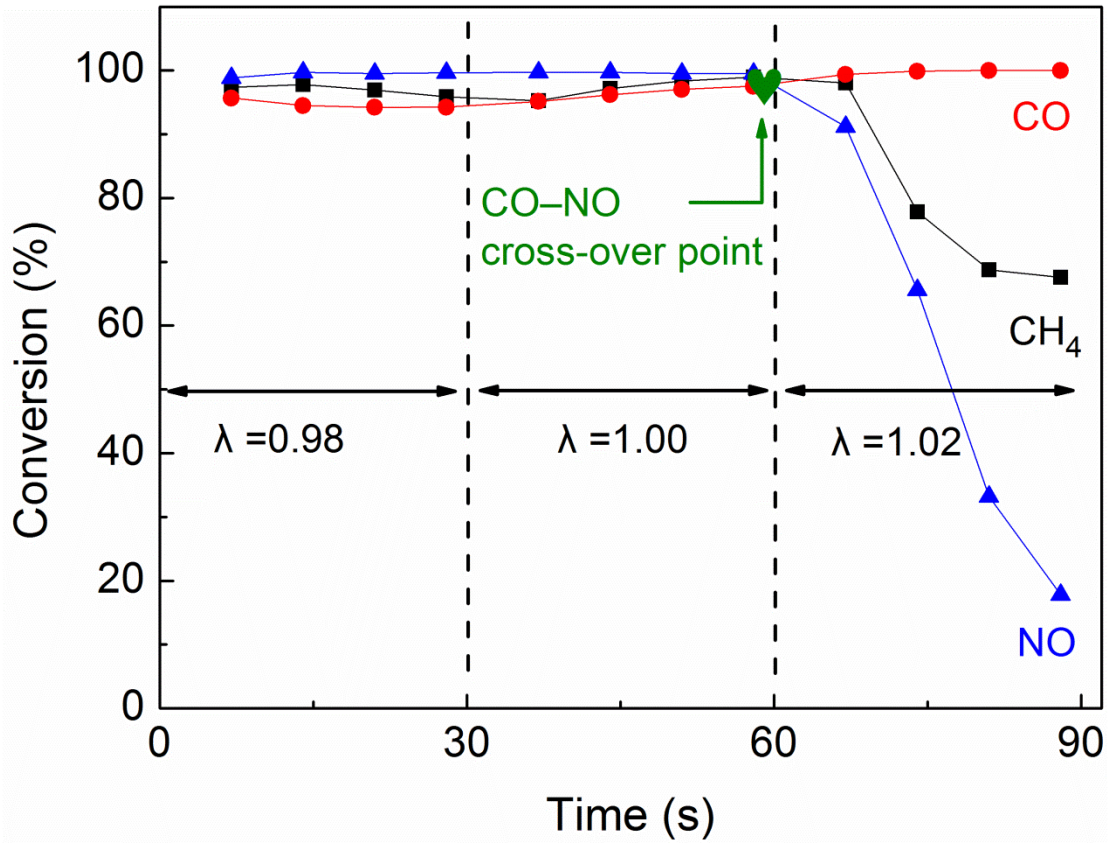

Figure S5. The changes in the pollutants conversion with lambda over the pretreated catalyst (T_{oven} = 450 °C). The time shown in the Figureure is corresponding to that in Figure. 11. The green heart-shaped point indicates the CO–NO crossover point, also indicated by a green arrow.

Table S1. Texture properties of the Pd model catalysts after pretreatment in reaction gas.

Samples	BET area (m^2/g)	Pore volume (mL/g)	Mean pore radii (nm)
Untreated catalyst	142.8	0.37	5.3
Cat-1-R	138.7	0.37	5.4
Cat-3-R	139.1	0.36	5.1
Cat-5-R	137.6	0.38	5.6
Cat-8-R	134.9	0.35	5.2