Supplementary Information

H_2-SCR at high water concentrations with *in situ* generated NH_3 for efficient removal of NO_x from H_2 engines

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Experimental

Synthesis of Na-SSZ-13

Firstly, a synthesis gel was prepared with the molar composition of 0.1 Na_2O : 1 SiO_2 : 0.025 Al_2O_3 : 0.2 N,N,N-trimethyl-1-adamantylammonium hydroxide (TMAdOH) : 44 H_2O.[1] Sodium hydroxide (VMR Chemicals), aluminium hydroxide hydrate (Sigma-Aldrich), and fumed silica (Sigma-Aldrich) were used as sodium, aluminium and silica sources, respectively. TMAdOH (TCI) was used as a template to promote the formation of the Chabazite (CHA) framework. After the mixture was stirred and aged overnight, a uniform gel was obtained. The gel was then transferred to a Teflon-lined stainless-steel autoclave and crystallized at 160 ℃ for 6 days in a rotating oven. The resulting solid product was recovered by filtration, washed several times with Milli-Q water, and dried at 80 ℃ for 12 h. The dried material was subsequently ground into a powder and calcined at 600 ℃ for 8 h to remove the organic template. The Na-form SSZ-13 was obtained after calcination.

Results and Discussions

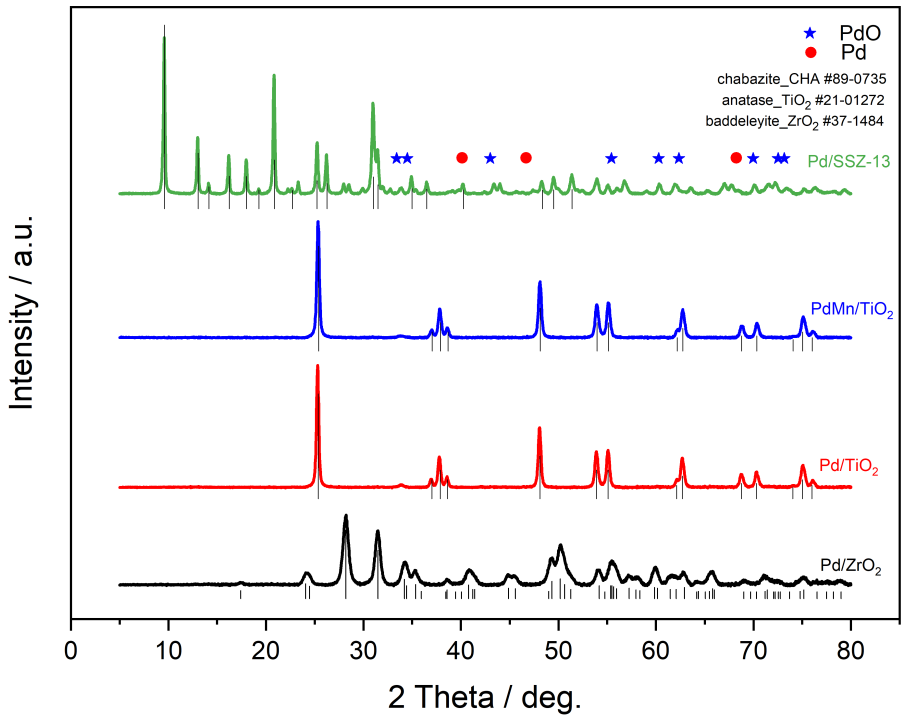

Fig. S1. XRD patterns of pre-treated 1 wt% Pd catalysts.

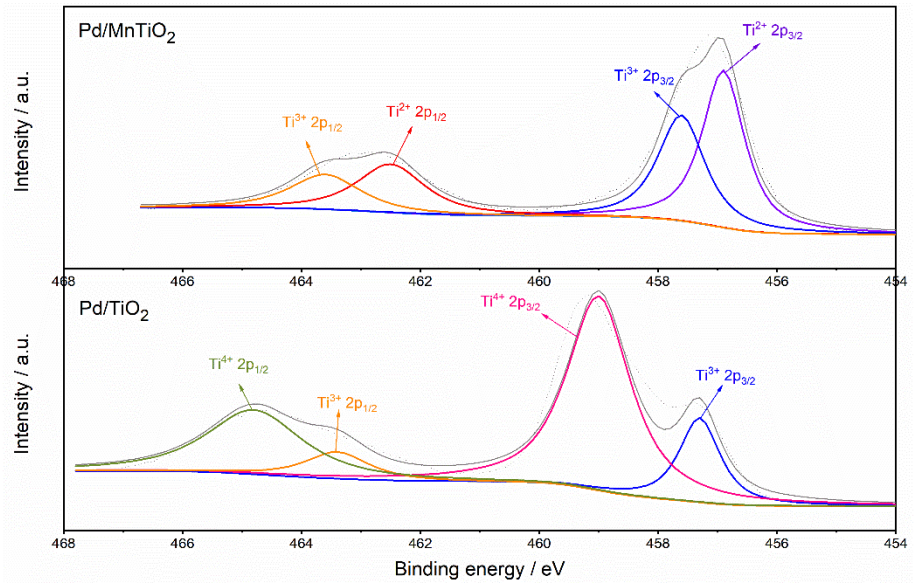

Fig. S2. XPS of Ti2p for pre-treated Pd/TiO_2 and $Pd/MnTiO_2$ catalysts.

Table S1. X-ray photoelectron spectroscopy § the TiO_2 supported catalysts.

Sample	Ti^{2+}		Ti^{3+}		Ti^{4+}	
	BE/eV ($Ti2p_{3/2}$/$Ti2p_{1/2}$)	Fraction/%	BE/eV ($Ti2p_{3/2}$/$Ti2p_{1/2}$)	Fraction/ %	BE/eV ($Ti2p_{3/2}$/$Ti2p_{1/2}$)	Fraction/ %
Pd/TiO_2			457.3/463.4	21.9	459.0/464.8	78.1
$Pd/MnTiO_2$	456.8/462.5	57.3	457.6/463.6	42.7		

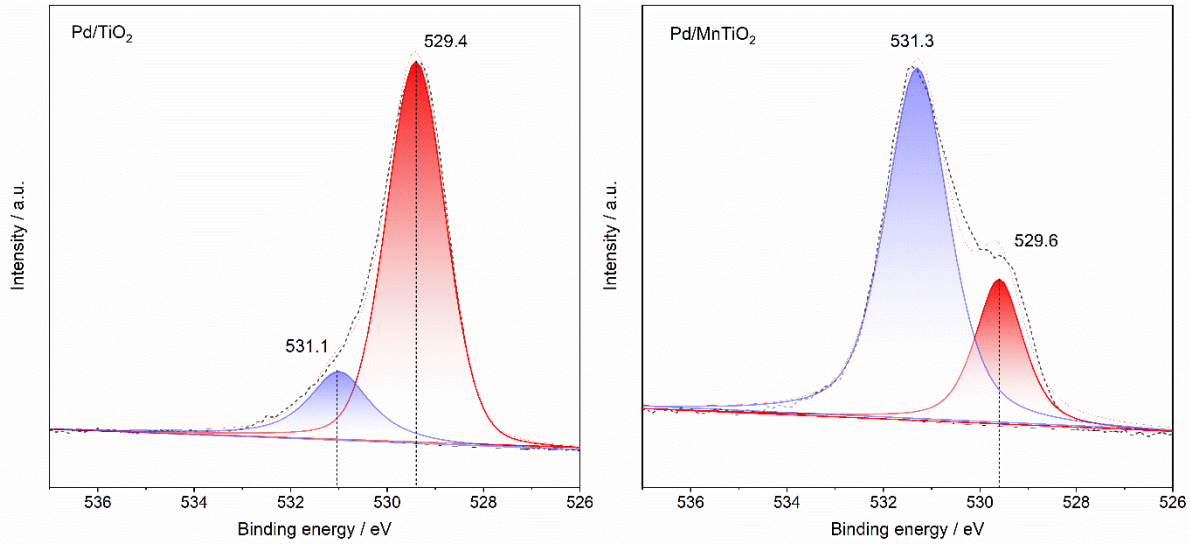

Fig. S3. XPS of O1s in pre-treated Pd/TiO_2 and $Pd/MnTiO_2$ catalysts.

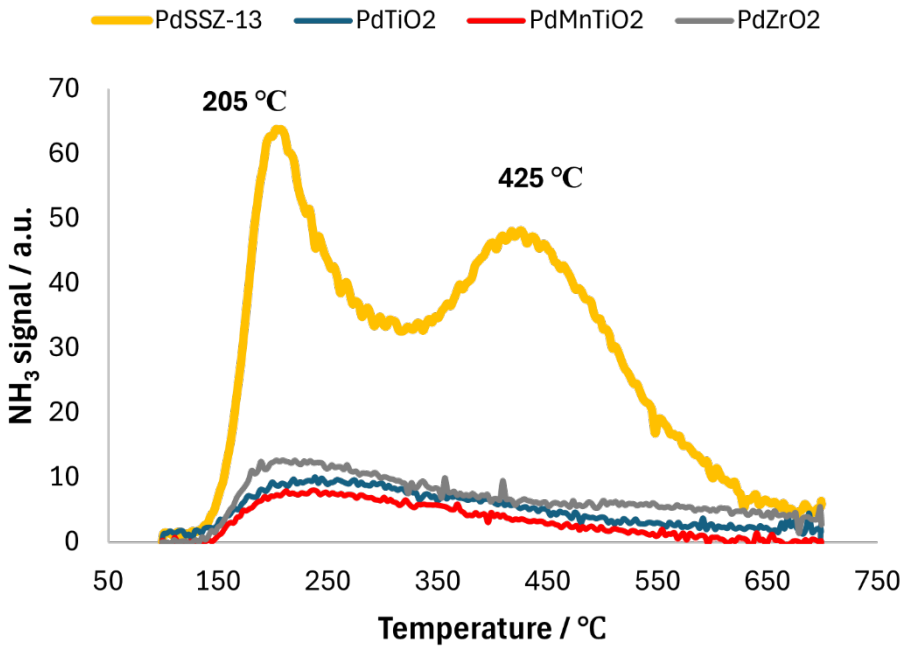

Fig. S4. NH_3-Temperature Programmed Desorption (TPD) profiles for the Pd catalysts.

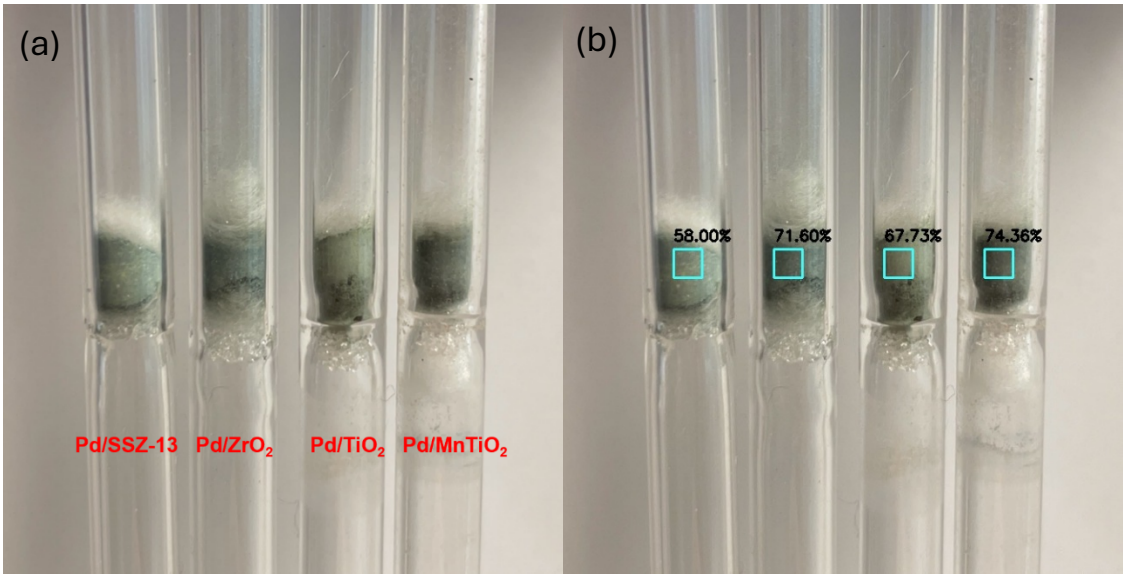

Fig. S5 Images of the sample mixtures (weight ratio: catalyst/WO_x=1/8) after H_2 treatment at 200 ℃ for 1h. (a) original (b) The RGB color components were extracted from the image, and the average proportion of the blue channel are shown. For info, the WO_x has a yellow color.

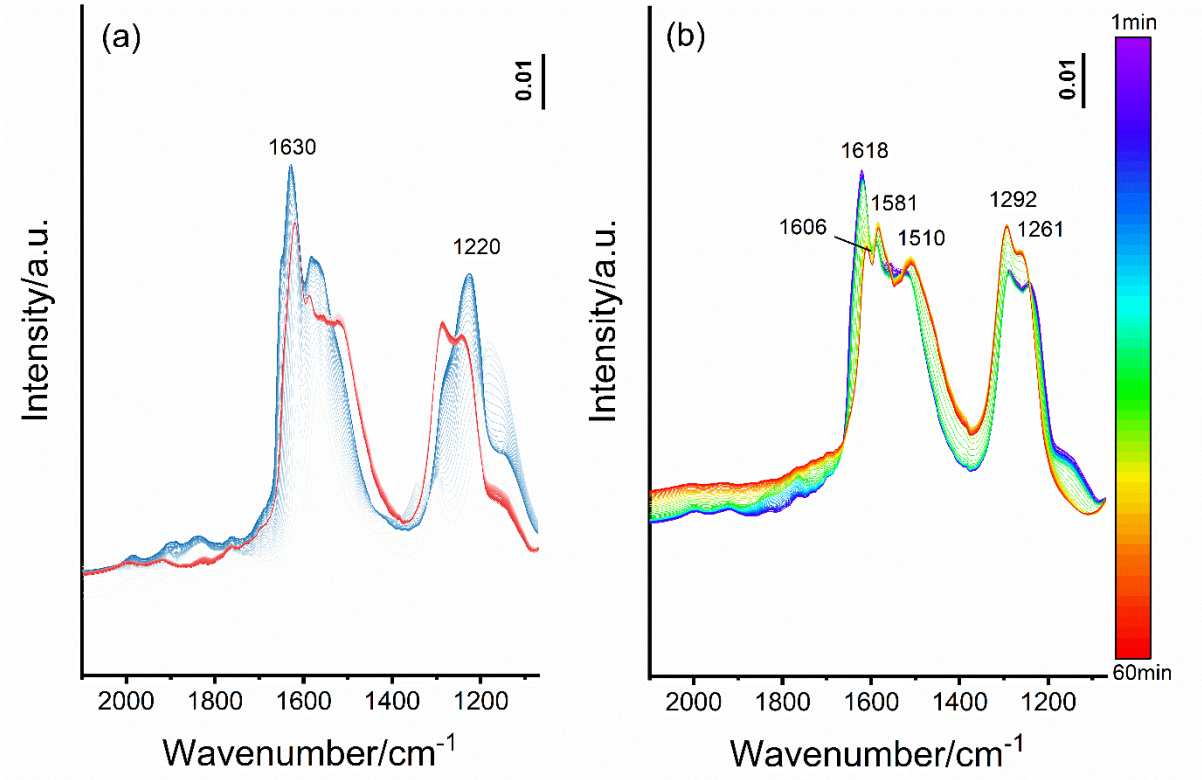

Fig. S6. In situ DRIFTS spectra at 80 ℃ of (a) 500 ppm NO for 0.5h (blue) and then 500 ppm NO + 1% H_2O for 1h(red); and (b) 2000 ppm H_2 exposure for 1h after flowing 500 ppm NO + 1% H_2O.

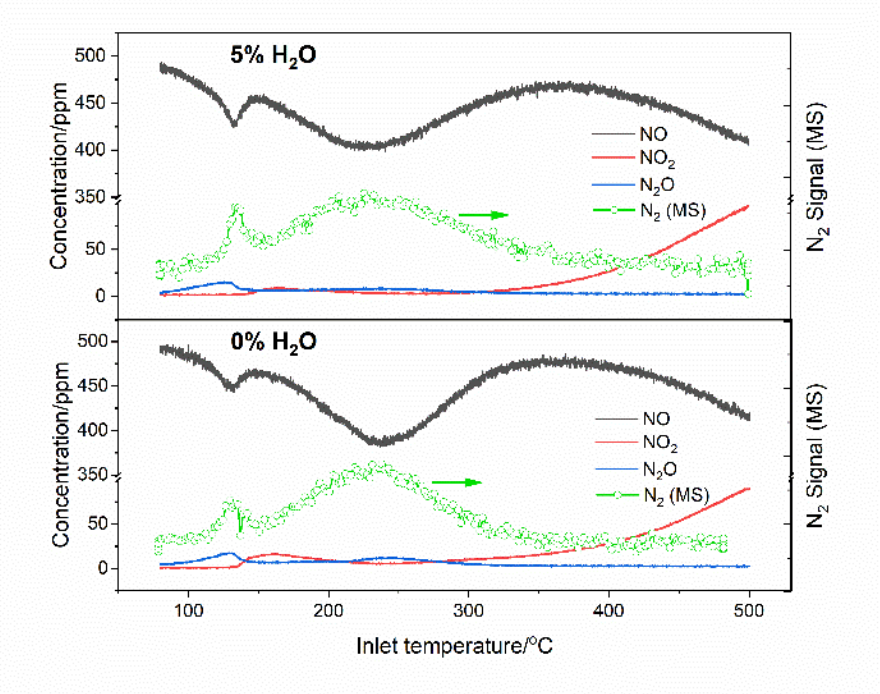

Fig. S7. Comparison of $NO/NO_2/N_2O$ concentrations from IR and N_2 signal from MS for Pd/SSZ-13 catalyst with and without water.

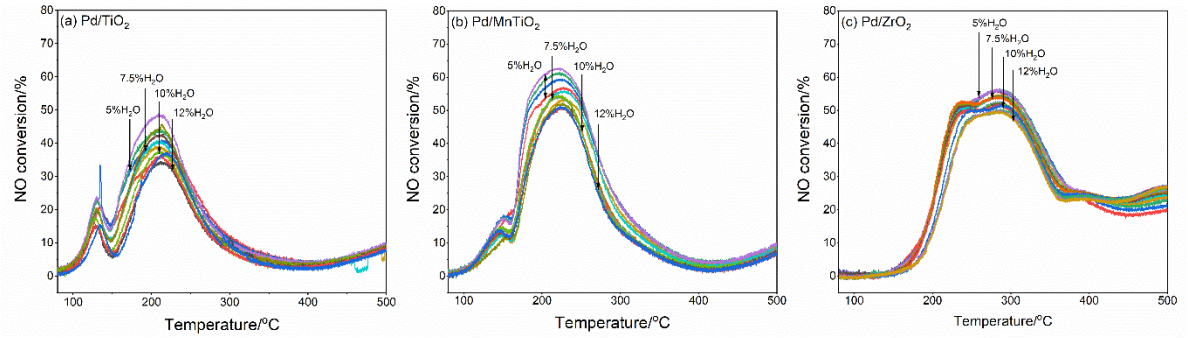

Fig. S8. NO conversion profiles for all cycles with varying water content (a) 1 wt% Pd/TiO_2, (b) 1 wt% $Pd/MnTiO_2$, (c) 1 wt% Pd/ZrO_2 (GHSV=20,000 h^{-1}(STP); gas inlet: 500 ppm NO, 5000 ppm H_2, 10% O_2, and 5/7.5/10/12% H_2O balanced in Ar; 80-500 ℃ with heating rate of 5 ℃ min^{-1}).

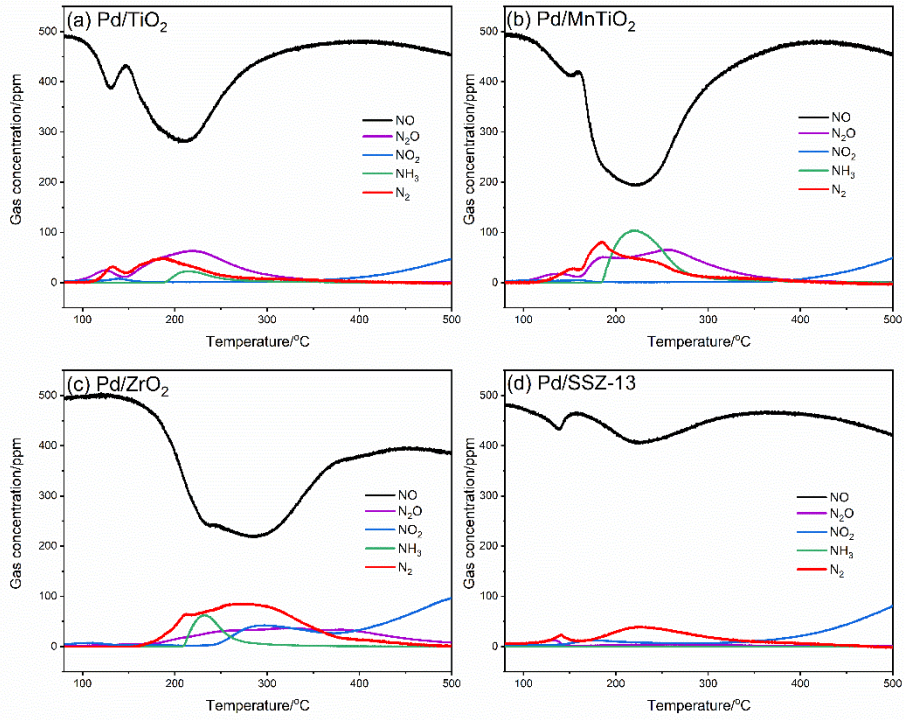

Fig. S9. The effluent gas concentration profiles of Pd catalysts with 5% water content from 80-500 °C for (a) Pd/TiO_2, (b) $Pd/MnTiO_2$, (c) Pd/ZrO_2 and (d) Pd/SSZ-13.

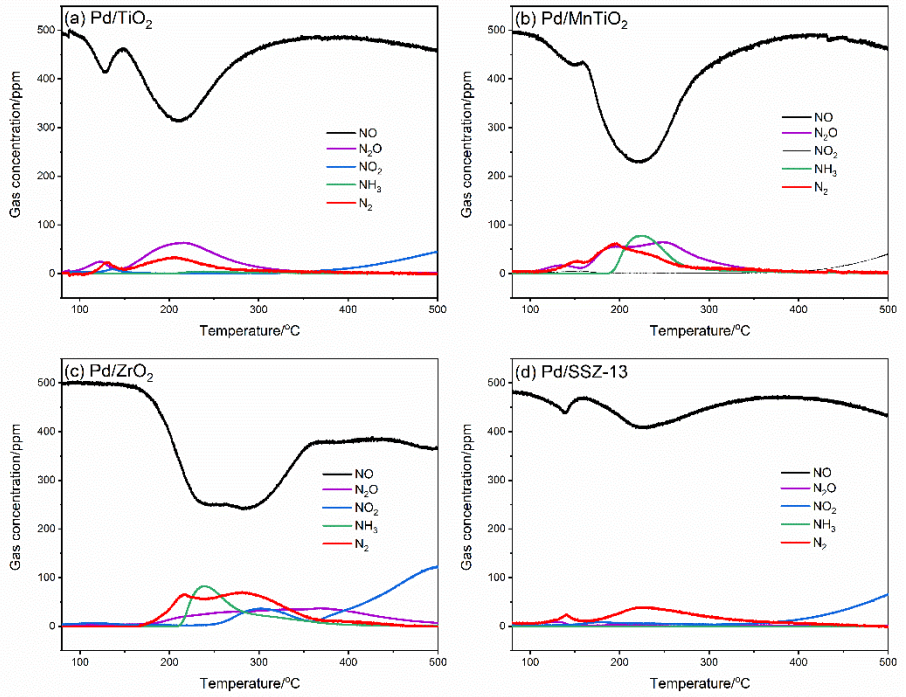

Fig. S10. The effluent gas concentration profiles of Pd catalysts with 10% water content during temperature ramp from 80-500 °C; (a) Pd/TiO_2, (b) $Pd/MnTiO_2$, (c) Pd/ZrO_2 catalysts, and (d) Pd/SSZ-13 catalyst.

References

1. Z. Li, M. T. Navarro, J. Martínez-Triguero, J. Yu and A. Corma, Catal. Sci. Technol., 2016, 6, 5856-5863.