Electronic Supplementary Material (ESI) for Catalysis Science & Technology.

Supplementary Information for

Understanding the multiple interactions in vanadium-based SCR catalysts during simultaneous NO_x and soot abatement

Lei Zheng,[1] Maria Casapu,[1] Jan-Dierk Grunwaldt,[1,2,]*

[1] Institute for Chemical Technology and Polymer Chemistry (ITCP), Karlsruhe Institute of Technology (KIT), Engesserstraße 20, 76131 Karlsruhe, Germany

[2] Institute of Catalysis Research and Technology (IKFT), Karlsruhe Institute of Technology (KIT), Hermann-von-Helmholtz Platz 1, 76344, Eggenstein-Leopoldshafen, Germany

[*]Corresponding author: Prof. Dr. Jan-Dierk Grunwaldt

Email: grunwaldt@kit.edu

Content

1. Experimental section

1.1 Testing protocol

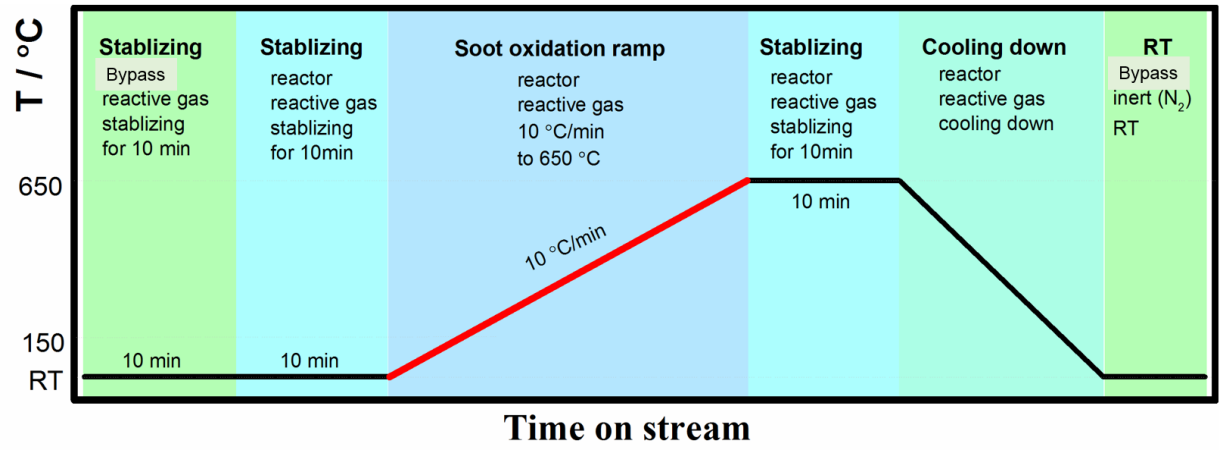

Figure S1. Testing protocol to probe gas-soot-catalyst interactions applied in the present study.

2. Further / additional results

2.1 Catalyst characterization

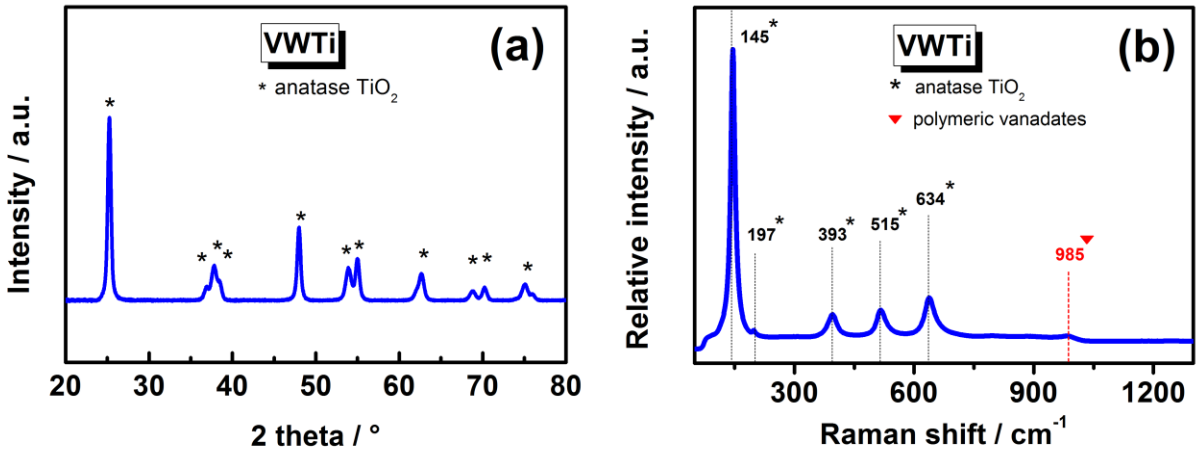

Figure S2. (a) X-ray diffraction (XRD) pattern of investigated VWTi catalyst: XRD pattern was recorded using a Bruker Advance D8 diffractometer with Ni-filtered Cu Kα radiation (1.54060 Å) in a range of 2θ = 20-80° and a step size of 0.0150°. (b) Raman spectra of investigated VWTi catalyst: Raman spectroscopy measurement was conducted on a Renishaw inVia confocal Raman microscope. A Nd-YAG laser with a wavelength of 532 nm (100 mW) and a 2400 l/mm grating was used. Spectra were taken with 0.5% laser power and ca. 40–50 spectra of a small line area were measured which were averaged after cosmic ray removal using Renishaw WiRE™ software.

Table S1. The specific surface area and chemical composition of the investigated VWTi catalyst.

Sample	Specific surface area S_{BET} [m^2/g] [1]	Elemental composition [wt.%] [2]		
		V	W	Ti
VWTi	64	1.66	7.49	50.8

[1] The specific surface area was measured by N_2 physisorption at -196°C using multipoint measurements on a BELSORP-mini instrument (MicrotracBEL, Osaka, Japan). Prior to the measurement, all samples were degassed in vacuum at 300 °C for 2 h.
[2] Chemical composition of the catalyst is determined using inductively coupled plasma optical emission spectroscopy (ICP-OES) on an OPTIMA 4300 DV spectrometer (PerkinElmer). The standard deviation is below 0.4 for Ti, and below 0.1 for other elements. The V_2O_5 and WO_3 concentrations are 2.96 wt.% and 10.01 wt.%, respectively, according to ICP-OES results, which is consistent with our target composition of 3 wt.% V_2O_5-10 wt.% WO_3/TiO_2.

2.2 Summary of $NTCO_x$ results

Table S2. List of the normalized total CO_x formation ($NTCO_x$) when feeding various SCR-related gas mixtures to different soot-catalyst contact types including soot-only, soot-catalyst in both loose and tight contact. $NTCO_x$ is a ratio that in each condition the obtained $CO+CO_2$ formation was normalized to the value obtained from O_2-soot reaction (benchmark). The gas mixture mixtures are listed in Table 1.

Gas mixture	Main factor	$NTCO_x$ (normalized by non-catalytic soot oxidation in $10\%O_2/N_2$)		
		gas-soot	gas-soot-catalyst (loose)	gas-soot-catalyst (tight)
Gas-1	inert (N_2)	0.5	0.9	1.7
Gas-2	baseline (O_2)	1.0	1.8	12.9
Gas-3	NO_2	12.7	11.6	25.9
Gas-4	NO	1.3	5.2	20.4
Gas-5	NH_3	0.4	3.2	18.0
Gas-6	H_2O	1.6	8.4	26.7
Gas-7	NO in H_2O	1.7	11.8	31.0
Gas-8	NO_2 in H_2O	10.5	20.2	33.6
Gas-9	dry standard SCR	0.6	4.9	20.6
Gas-10	dry fast SCR	2.9	6.6	22.9
Gas-11	wet standard SCR	1.2	12.6	31.2
Gas-12	wet fast SCR	5.7	13.3	34.0

2.3 Summary of T_o results

Table S3. List of the temperatures corresponding to the soot oxidation onset (T_o), and the corresponding CO_x formation at T_o for all gas-soot-catalyst reactions. The gas mixture mixtures are listed in Table 1.

Gas mixture	Main factor	T_o (°C) and the corresponding $CO+CO_2$ emission (ppm)		
		gas-soot	gas-soot-catalyst (loose)	gas-soot-catalyst (tight)
Gas-1	inert (N_2)	- (-)	452 °C (22.7 ppm)	202 °C (6.4 ppm) and 312 °C (19 ppm)
Gas-2	baseline (O_2)	346 °C (13 ppm)	329 °C (28.5 ppm)	203 °C (13.6 ppm) and 329 °C (28 ppm)
Gas-3	NO_2	344 °C (12 ppm)	315 °C (30.7 ppm)	204 °C (11.3 ppm) and 314 °C (32.5 ppm)
Gas-4	NO	320 °C (18 ppm)	312 °C (37 ppm)	212 °C (20 ppm) and 300 °C (40.5 ppm)
Gas-5	NH_3	348 °C (7.8 ppm)	336 °C (16 ppm)	224 °C (9.5 ppm) and 337 °C (23.3 ppm)
Gas-6	H_2O	338 °C (13 ppm)	310 °C (23.1 ppm)	203 °C (12.9 ppm) and 334 °C (32.5 ppm)
Gas-7	NO in H_2O	341 °C (11.2 ppm)	331 °C (25.9 ppm)	217 °C (13 ppm) and 316 °C (27.1 ppm)
Gas-8	NO_2 in H_2O	313 °C (20.3 ppm)	327 °C (40.2 ppm)	- (-)
Gas-9	dry standard SCR	347 °C (9.7 ppm)	335 °C (18 ppm)	300 °C (20.5 ppm)
Gas-10	dry fast SCR	312 °C (13.7 ppm)	333 °C (19.7 ppm)	300 °C (29.2 ppm)
Gas-11	wet standard SCR	345 °C (10.5 ppm)	329 °C (23.9 ppm)	325 °C (24.3 ppm)
Gas-12	wet fast SCR	310 °C (17.8 ppm)	332 °C (32 ppm)	203 °C (21.7 ppm) and 289 °C (28.7 ppm)

2.4 Summary of T_{50p} results

Table S4. List of the temperatures at $CO+CO_2$ formation of 50 ppm (T_{50p}, °C) for all investigated gas-soot-catalyst reactions. The gas mixture mixtures were listed in Table 1.

Gas mixture	Main factor	T_{50p} (°C)		
		gas-soot	gas-soot-catalyst (loose)	gas-soot-catalyst (tight)
Gas-1	inert (N_2)	-	-	617
Gas-2	baseline (O_2)	629	605	515
Gas-3	NO_2	468	460	377
Gas-4	NO	609	550	474
Gas-5	NH_3	649	573	496
Gas-6	H_2O	602	558	470
Gas-7	NO in H_2O	595	508	453
Gas-8	NO_2 in H_2O	449	391	349
Gas-9	dry standard SCR	648	559	467
Gas-10	dry fast SCR	522	548	468
Gas-11	wet standard SCR	604	532	465
Gas-12	wet fast SCR	477	532	459

For Gas-10 and Gas-12, even lower T_{50p} was observed in gas-soot systems compared to gas-soot-catalyst (loose) systems, the integrated CO_x formation was significantly lower, as shown in Figure 8 and Figure S5, respectively.

2.5 Influence of NO+H_2O gas mixture on soot oxidation

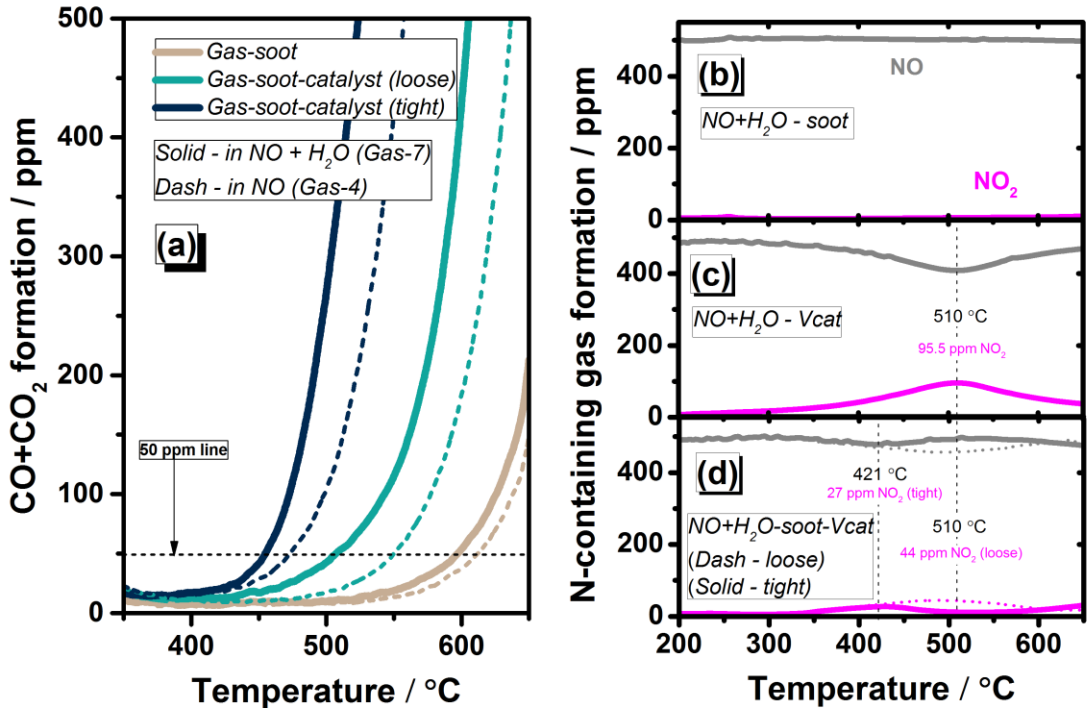

Figure S3. Comparison of gas evolution for NO+H_2O-containing systems: (a) CO+CO_2 formation, the N-containing gases formation from (b) gas-soot system, (c) gas-catalyst system and (d) gas-soot-catalyst systems (loose and tight). Gas-7: 500 ppm NO, 5% H_2O, 10% O_2 in N_2 with total gas flow of 300 mL/min. 5 mg soot with 245 mg catalyst (or inert quartz sand).

2.6 Influence of NO_2+H_2O gas mixture on soot oxidation

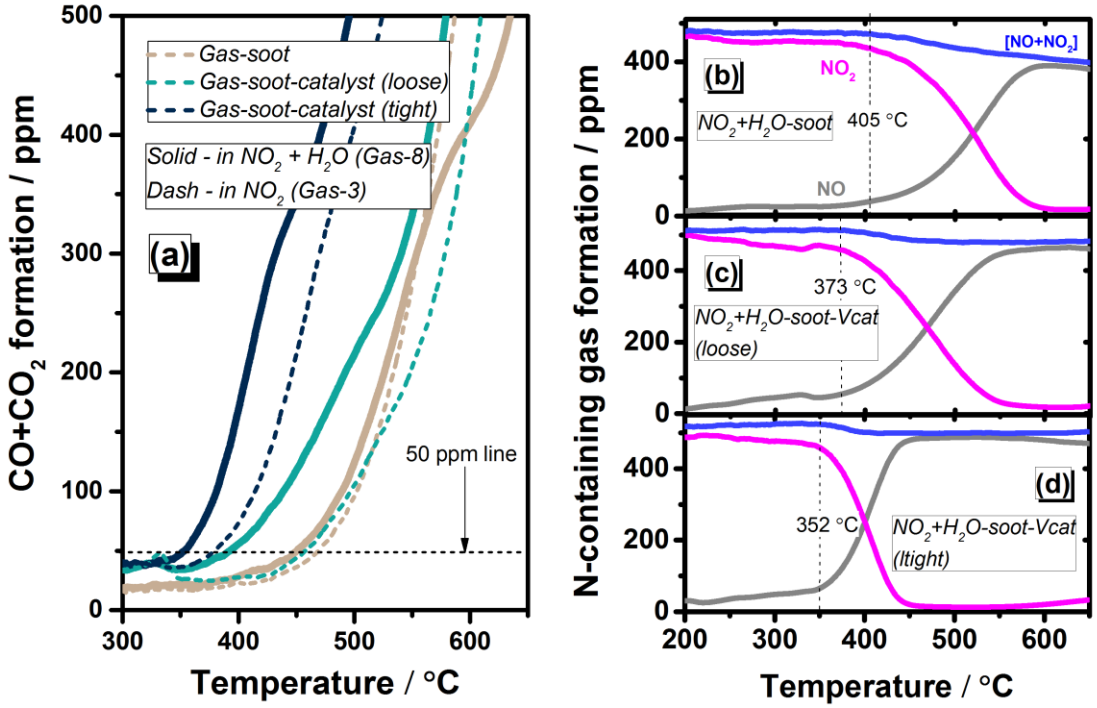

Figure S4. Comparison of gas evolution for NO_2+H_2O-containing systems: (a) CO+CO_2 formation, the N-containing gases formation from (b) gas-soot system, (c) gas-catalyst system and (d) gas-soot-catalyst systems (loose and tight). Gas-8: 500 ppm NO_2, 5% H_2O, 10% O_2 in N_2 with total gas flow of 300 mL/min. 5 mg soot with 245 mg catalyst (or inert quartz sand).

2.7 Effect of wet standard SCR gas mixture on soot oxidation

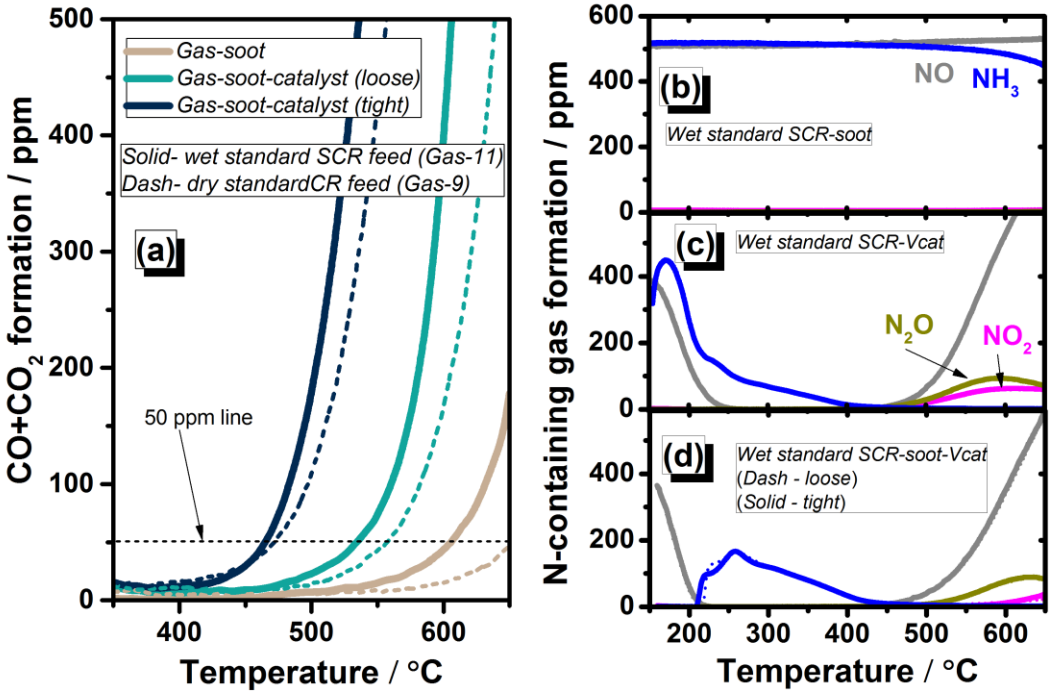

Figure S5. Comparison of gas evolution for wet standard SCR gas-containing systems: (a) $CO+CO_2$ formation, N-containing gases formation from (b) gas-soot system, (c) gas-catalyst system and (d) gas-soot-catalyst systems (loose and tight). Gas-11: 500 ppm NO, 500 ppm NH_3, 5% H_2O, 10% O_2 in N_2 with total gas flow of 300 mL/min. 5 mg soot with 245 mg catalyst (or inert quartz sand).

2.8 Effect of wet fast SCR gas mixture on soot oxidation

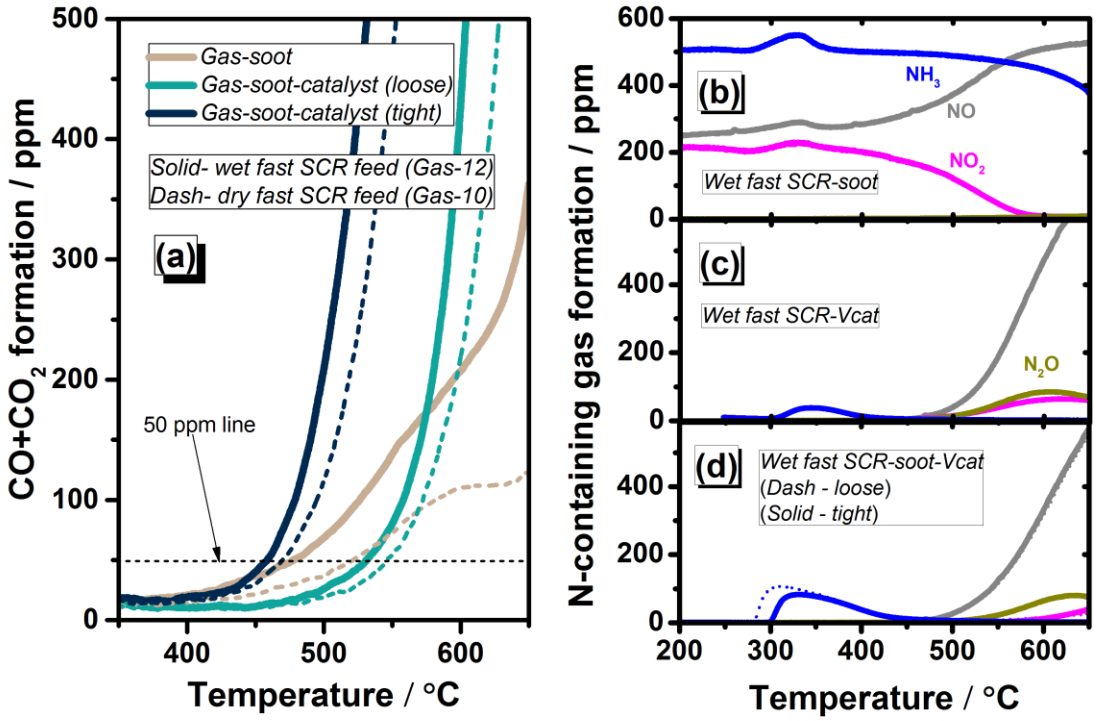

Figure S6. Comparison of gas evolution for wet fast SCR gas-containing systems: (a) $CO+CO_2$ formation, the N-containing gases formation from (b) gas-soot system, (c) gas-catalyst system and (d) gas-soot-catalyst systems (loose and tight). Gas-12: 250 ppm NO, 250 ppm NO_2, 500 ppm NH_3, 5% H_2O, 10% O_2 in N_2 with total gas flow of 300 mL/min. 5 mg soot with 245 mg catalyst (or inert quartz sand).

2.9 Effect of soot on SCR of NO_x with NH_3

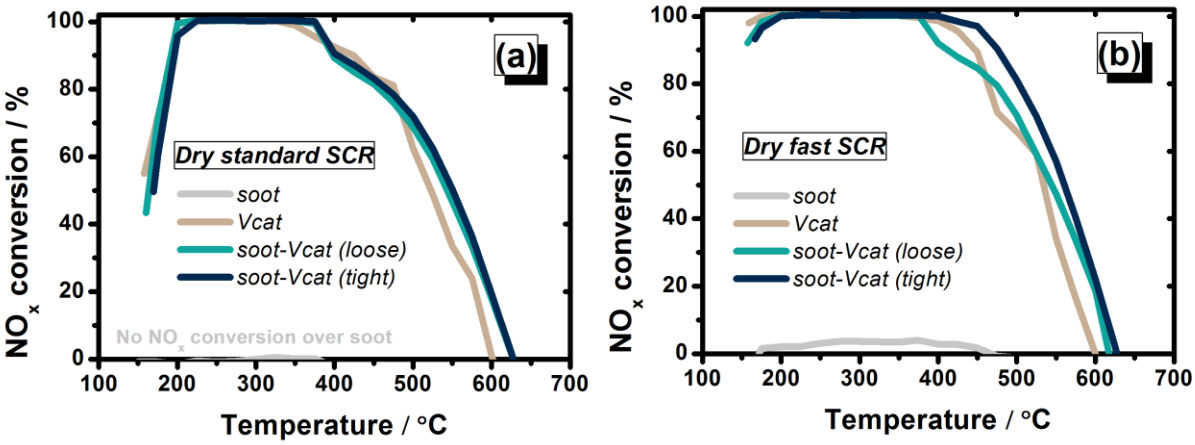

Figure S7. Comparison of NO_x conversion from (a) dry standard SCR and (b) dry fast SCR over different gas-soot-catalyst reactions. Standard SCR: 500 ppm NO, 500 ppm NH_3, 10% O_2 in N_2 with total gas flow of 300 mL/min; Fast SCR: 250 ppm NO, 250 ppm NO_2, 500 ppm NH_3, 10% O_2 in N_2 with total gas flow of 300 mL/min. 5 mg soot with 245 mg catalyst (or inert quartz sand).