1) Geometric parameters of Cu(I) sites before and after single NO molecule adsorption in high silica model.

PBE/TZVP results. Distances in Å, angles in degrees.

Cu(I) site	Cu(I)-FAU[a]			NO-Cu(I)-FAU				
	*n*Al-O(H)[b]	CN_{Cu}[c]	R_{Cu-O}[d]	CN_{Cu}	R_{Cu-O}	R_{Cu-N}	R_{N-O}	θ_{Cu-N-O}
site II	1Al	3(3)	*1.98*, 2.06, 2.19 (O2)	2(1)	*2.05* (O2), *2.05* (O4)	1.795	1.175	143.6
						1.793	1.174	145.8*[e]
	2Al-*meta*-1[f]	3(3)	*1.98, 2.08*, 2.13 (O2)	2(1)	*2.03* (O2), *2.08* (O4)	1.795	1.177	143.0
						1.792	1.176	145.7*
	2Al-*para*-1	3(3)	*2.02, 2.06*, 2.11 (O2)	2(1)	*2.04* (O2), *2.08* (O4)	1.798	1.177	142.3
						1.794	1.176	145.8*
	3Al-1,1	3(3)	*2.00, 2.06, 2.08* (O2)	2(1)	*2.04* (O2), *2.11* (O4)	1.798	1.180	142.2
						1.794	1.178	145.4*
				3(3)	*2.04, 2.16, 2.47* (O2)	1.837	1.180	137.3
						1.837	1.181	136.4*
site III	1Al	2(1)	*2.02* (O1), *2.04* (O4)	2(1)	*2.01* (O1), *2.03* (O4)	1.785	1.176	145.8
	3Al-3,3	2(2)	*2.01, 2.02* (O1)	2(2)	*2.04, 2.05* (O1)	1.797	1.175	144.8
	4Al-1,1,3	2(1)	*1.96* (O1), *2.16* (O4)	2(1)	*1.95* (O1), *2.14* (O4)	1.785	1.175	145.8

[a] Data from ref. 65. [b] *n*Al-*m*: *n* denotes number of Al atoms within QM cluster, *m* are crystallographic types of protonated O atoms. [c] Cu coordination number to framework O atoms. *m(n)*: *m* denotes total Cu-O coordination number, *n* number of distinct coordinated TO_4 units. [d] Only Cu-O distances closer than 2.5 Å considered. Italics depict distances between Cu and O atoms belonging to AlO_4 units. Crystallographic types of O atoms in parenthesis. [e] Star depicts Cu-N-O species bent toward the plane of 6T ring. [f] *Meta* and *para* denote mutual Al atoms positions in 6T ring.

2) Geometric parameters of Cu(I) sites before and after single NO molecule adsorption in Y models.

PBE/TZVP results. Distances in Å, angles in degrees.

model	Cu(I) site	Cu(I)-FAU[a]			NO-Cu(I)-FAU				
		*n*Al[b]	CN_{Cu}[c]	R_{Cu-O}[d]	CN_{Cu}	R_{Cu-O}	R_{Cu-N}	R_{N-O}	θ_{Cu-N-O}
Y1[e]	site II	1Al (2)	3(3)	*2.10*, 2.06, 2.19 (O2)	2(1)	*2.05* (O2), *2.07* (O4)	1.801 1.796	1.174 1.178	142.6 146.8*[f]
					2(2)	*2.05*,2.20 (O2)	1.848 1.843	1.173 1.174	137.2 137.9*
		2Al-*meta*[g] (2)	3(3)	*1.98*, *2.06*, 2.32 (O2)	2(1)	*2.01*(O2), *2.07* (O4)	1.800 1.793	1.178 1.177	141.4 146.1*
					2(2)	*2.00*, *2.27*(O2)	1.838 1.833	1.176 1.178	135.9 137.8*
		2Al-*para* (1)	3(3)	*2.04*, *2.09*, 2.14 (O2)	2(2)	*2.03*, *2.12* (O2)	1.834 1.835	1.175 1.172	139.9 139.6*
	site III	2Al	2(1)	*2.01*(O4), *2.04*(O1)	2(1)	*2.04*(O1), *2.01* (O4)	1.790	1.175	145.1
Y2	site II	1Al (1)	3(3)	*2.02*, 2.09, 2.15 (O2)	2(1)	*2.03*(O2), *2.11*(O4)	1.804 1.798	1.174 1.172	142.0 146.5*
					2(2)	*2.03*, 2.29 (O2)	1.851 1.846	1.173 1.172	136.8 138.5*
		2Al-*meta* (1)	3(3)	*2.02*, *2.09*, 2.13 (O2)	2(2)	*2.04*, *2.16* (O2)	1.854 1.845	1.177 1.173	130.3 138.3*
		2Al-*para* (1)	3(3)	*2.06*, *2.10*, 2.16 (O2)	2(1)	*2.05*(O2), *2.10* (O4)	1.817 1.804	1.178 1.177	138.1 145.2*
					2(2)	*2.07*, *2.29*(O2)	1.862 1.860	1.176 1.179	135.1 134.7*
X	site II	3Al (2)	3(3)	*2.02*, *2.06*, *2.10* (O2)	3(3)	*2.06*, *2.20*, *2.35* (O2)	1.852 1.855	1.180 1.183	135.6 133.8*
					2(1)	*2.04*(O2), *2.07*(O4)	1.801 1.793	1.180 1.178	141.4 147.0*

[a] Data for Y1 model from ref. 65. [b] *n*Al: for site II clusters denotes number of Al atoms within 6T ring for site II, value in parenthesis refers to number of Al atoms in the remaining part of QM cluster. For site III cluster is total number of Al atoms in QM cluster. [c] Cu coordination number to framework O atoms. *m(n)*: *m* denotes total Cu-O coordination number, *n* number of distinct coordinated TO_4 units. [d] Only Cu-O distances closer than 2.5 Å considered. Italics depict distances between Cu and O atoms belonging to AlO_4 units. Crystallographic types of O atoms in parenthesis. [e] See text for detailed description of various 'Y lattice' models. [f] Star depicts Cu-N-O species bent toward the plane of 6T ring. [g] *Meta* and *para* denote mutual Al atoms positions in 6T ring.

3) Properties of the dinitrosyl species calculated for high-silica FAU model.

Singlet (S)and selected triplet(T) states: structural parameters (distances in Å, angles in degrees), interaction energies (kJ/mol) and NO stretching bonds harmonic frequencies (cm^{-1}).

Cu(I) site	*n*Al-O(H)[a]	spin	CN_{Cu}[b]	R_{Cu-O}[c]	R_{Cu-N}	R_{N-O}	R_{O-O}[d]	R_{N-N}	θ_{Cu-N-O}	E_{ads} (E_{def})[e]	ΔH_{ads}[f]	ω_{NO}
site II	1Al	S	2(1)	*2.08*(O2) *2.11* (O4)	1.889 1.956	1.168 1.164	2.37	2.72	127.5 125.1	-77 (71)	39	1691 1814
		T	2(1)	*2.14* (O2) *2.11* (O4)	1.845 1.929	1.168 1.166	2.88	2.77	137.9 133.4	-56 (71)	27	1689 1829
	2Al-*meta*-1[f]	S	2(1)	*2.09* (O2) *2.10* (O4)	1.888 1.952	1.170 1.166	2.37	2.72	127.3 125.2	-75 (83)	38	1682 1805
		T	2(1)	*2.09* (O2) *2.16* (O4)	1.874 1.913	1.168 1.169	2.84	2.79	134.5 132.6	-55 (84)	26	1674 1814
	2Al-*para*-1	S	2(1)	*2.12* (O2) *2.12* (O4)	1.931 1.934	1.168 1.167	2.39	2.76	124.8 124.9	-72 (84)	36	1682 1802
	3Al-1,1	S	2(1)	*2.10*(O2) *2.12*(O4)	1.872 1.975	1.174 1.164	2.38	2.73	127.7 124.4	-73 (98)	37	1673 1802
		S	3(3)	*2.13* (O2) *2.36* (O2) *2.37* (O2)	1.972 1.975	1.174 1.164	2.36	2.70	123.3 123.3	-65 (69)	32	1663 1791
site III	1Al	S	2(1)	*2.05* (O1) *2.15* (O4)	1.902 1.915	1.169 1.167	2.39	2.73	126.3 125.9	-87 (3)	45	1686 1807
	3Al-3,3	S	2(1)	*2.08* (O4) *2.20* (O4)	1.891 2.032	1.174 1.163	2.44	2.80	123.2 119.3	-73 (23)	37	1670(1700)[g] 1789(1825)
	4Al-1,1,3	S	2(1)	*1.97* (O1) *2.22* (O4)	1.900 1.920	1.171 1.166	2.40	2.73	125.6 125.0	-89 (2)	46	1681(1698) 1785(1817)
		T	1(1)	*1.96* (O1)	1.854 1.895	1.174 1.167	2.86	2.78	135.1 132.8	-71 (8)	36	1672 1789

[a] *n*Al-*m*: *n* denotes number of Al atoms within QM cluster, *m* are crystallographic types of protonated O atoms. [b] Cu coordination number to framework O atoms. *m(n)*: *m* denotes total Cu-O coordination number, *n* number of distinct coordinated TO_4 units. [c] Only Cu-O distances closer than 2.5 Å considered. Italics depict distances between Cu and O atoms belonging to AlO_4 units. Crystallographic types of O atoms in parenthesis. [d] Distances between atoms belonging to NO molecules. [e] See text for definition. [f] Calculated according to the equation 3 with f. taken form MP2 and CASPT2 calculations. [f] *Meta* and *para* denote mutual Al atoms positions in 6T ring. [g] Values in parenthesis obtained from 22T QM clusters.